Dissertation for the degree of Doctor of Philosophy

System Analyses
on Advanced Nuclear Fuel Cycle and Waste Management

Myeongguk Cheon
Department of Nuclear Engineering
University of California, Berkeley

May 2005
This work was performed as part of the project supported by
the United States Department of Energy
and
the Japan Nuclear Cycle Development Institute

Comments and notification of any errors in this report would be appreciated.

Joonhong Ahn
Principal investigator of the project
Department of Nuclear Engineering
University of California
Berkeley, CA 94720
USA

ahn@nuc.berkeley.edu
Abstract

System Analyses on Advanced Nuclear Fuel Cycle and Waste Management

By

Myeongguk Cheon
Doctor of Philosophy in Engineering-Nuclear Engineering
University of California, Berkeley
Professor Joonhong Ahn, Chair

To evaluate the impacts of accelerator-driven transmutation of waste (ATW) fuel cycle on a geological repository, two mathematical models are developed: a reactor system analysis model and a high-level waste (HLW) conditioning model. With the former, fission products and residual trans-uranium (TRU) contained in HLW generated from a reference ATW plant operations are quantified and the reduction of TRU inventory included in commercial spent-nuclear fuel (CSNF) is evaluated. With the latter, an optimized waste loading and composition in solidification of HLW are determined and the volume reduction of waste packages associated with CSNF is evaluated.

WACOM, a reactor system analysis code developed in this study for burnup calculation, is validated by ORIGEN2.1 and MCNP. WACOM is used to perform multi-cycle analysis for the reference lead-bismuth eutectic (LBE) cooled transmuter. By applying the results of this analysis to the reference ATW deployment scenario considered in the ATW roadmap, the HLW generated from the ATW fuel cycle is quantified and the reduction of TRU inventory contained in CSNF is evaluated.

A linear programming (LP) model has been developed for determination of an optimized waste loading and composition in solidification of HLW. The model has been applied to a US-defense HLW. The optimum waste loading evaluated by the LP model was compared with that estimated by the Defense Waste Processing Facility (DWPF) in the US and a good agreement was observed. The LP model was then applied to the volume reduction of waste packages associated with CSNF. Based on the obtained reduction factors, the expansion of Yucca Mountain Repository (YMR) capacity is evaluated.

It is found that with the reference ATW system, the TRU contained in CSNF could be reduced by a factor of ~170 in terms of inventory and by a factor of ~40 in terms of toxicity under the assumed scenario. The number of waste packages related to CSNF could be reduced by a factor of ~8 in terms of volume and by factor of ~10 on the basis of electricity generation when a sufficient cooling time for discharged spent fuel and zero process chemicals in HLW are assumed. The expansion factor of Yucca Mountain Repository capacity is estimated to be a factor of 2.4, much smaller than the reduction factor of CSNF waste packages, due to the existence of DOE-owned spent fuel and HLW. The YMR, however, could support 10 times greater electricity generation as long as the statutory capacity of DOE-owned SNF and HLW remains unchanged.

This study also showed that the reduction of the number of waste packages could strongly be subject to the heat generation rate of HLW and the amount of process chemicals contained in HLW. For a greater reduction of the number of waste packages, a sufficient
cooling time for discharged fuel and efforts to minimize the amount of process chemicals contained in HLW are crucial.
# Table of Contents

1 Introduction and Summary ............................................................................ 1  
   1.1 Backgrounds/Motivations ....................................................................... 1  
   1.2 State of the Arts ...................................................................................... 3  
      1.2.1 Reviews of ATW Systems .................................................................. 3  
      1.2.1.1 Reference ATW Plant in the Roadmap .......................................... 3  
      1.2.1.2 Reference ATW Implementation Scenario .................................. 5  
      1.2.2 Reviews of Available Codes .............................................................. 5  
      1.2.3 Disposal Form of High-Level Waste .............................................. 6  
   1.3 Objectives .................................................................................................. 6  
   1.4 Plans of This Study ................................................................................... 6  
   1.5 Scopes and Limits ..................................................................................... 8  

2 Reactor Model .................................................................................................. 9  
   2.1 Transmutation Chain of Actinides .......................................................... 9  
   2.2 Multiplication Factor Formula ................................................................ 13  
   2.3 Numerical Scheme ................................................................................... 14  
   2.4 Verification of Numerical Scheme for 2-Nuclide Chain ....................... 15  

3 Fuel Cycle Model ............................................................................................ 17  
   3.1 Fuel Discharge and Partitioning .............................................................. 17  
   3.2 Determination of Makeup Fuel ............................................................... 18  
   3.3 Flow Chart for Code Programming ......................................................... 20  

4 Computation for a Single Cycle Case ............................................................ 22  
   4.1 Reference LBE-Cooled Transmuter ......................................................... 22  
   4.2 Neutronics of Start-up Core of the LBE-Cooled Transmuter ............. 24  
   4.3 Fuel Inventory Evolution in the LBE-Transmuter for a Single Cycle .... 27  
   4.4 Benchmarking of WACOM against MOCUP for a Single Cycle ....... 28  
   4.5 Summary and Conclusions .................................................................... 31  

5 Application of WACOM for Multi Cycle Calculation ................................ 32  
   5.1 Introduction ............................................................................................. 32  
   5.2 Accounting for Neutronics Variation ....................................................... 32  
      5.2.1 Neutron Spectrum Variation ............................................................ 32  
      5.2.2 Revised Flow Chart of WACOM Code ........................................... 34  
      5.2.3 Validation of Interpolating Scheme for Estimation of Neutronics ... 36  
   5.3 Analysis of the Reference LBE-Cooled Transmuter ............................ 37  
   5.4 Effects of Difference in the Branching Ratio of $^{241}$Am ................... 45  
   5.5 Summary and Conclusions ................................................................. 47  

6 Reduction of TRU Inventory Contained in LWR-Spent Fuel .................. 49  
   6.1 Introduction ............................................................................................ 49  
   6.2 Characteristics of the LBE-Cooled Transmuter ................................. 49  
   6.3 Reduction of TRU Inventory Contained in LWR-SNF ........................ 50  
   6.4 Discussions ............................................................................................ 57  
   6.5 Conclusions ............................................................................................ 58  

7 Model for Conditioning of High-Level Waste ........................................... 59  
   7.1 Introduction ............................................................................................ 59  
   7.2 Solidification of High-Level Waste (HLW) ............................................ 60
7.2.1 Solidification Process of HLW ................................................................. 60
7.2.2 Requirements of Solidified Waste Products ........................................... 61
  7.2.2.1 General Considerations in HLW Solidification ............................... 61
  7.2.2.2 Specific Requirements for US-Defense HLW Solidification ............ 61
7.3 Mathematical Formulation of Waste Consistency Tests (PCT) Limits ........................... 63
  7.3.1 Formulation of Product Consistency Tests (PCT) Limits ....................... 63
    7.3.1.1 Product Consistency Tests (PCT) .............................................. 63
    7.3.1.2 Analytic Model of PCT Method A ............................................ 63
      7.3.1.2.1 Abstracted Model for Defense HLW Glass Degradation .......... 63
      7.3.1.2.2 Application of Abstracted Model for PCT-A ......................... 66
  7.3.2 Formulation of Temperature Limit .................................................... 67
    7.3.2.1 Temperature Distribution in a Cylinder Block with Heat Sources .. 67
    7.3.2.2 Temperature Distribution in Canister Wall without Heat Sources ... 68
7.3.3 Formulation of Solidified Generation Limit ........................................... 71
7.3.4 Formulation of Solidified Waste Glass Volume Limit ............................. 71
7.3.5 Formulation of Filled Canister Weight Limit ......................................... 72
7.3.6 Formulation of Plutonium Concentration Limit ...................................... 72
7.4 Linear Programming Model for HLW Conditioning ....................................... 73
7.5 Solution Methods for Linear Programming Problem ...................................... 75
  7.5.1 Graphical Solution Method .............................................................. 75
  7.5.2 The Simplex Method ..................................................................... 75
7.6 Implementation of LP Model to a US-Defense HLW ..................................... 78
  7.6.1 Compositions of HLW and Borosilicate Glass ..................................... 78
  7.6.2 Construction of LP Problem and Graphical Solution ............................ 79
  7.6.3 Constraints of the Defense Waste Processing Facility (DWPF) ............. 82
7.7 Discussions and Conclusions .................................................................. 88
8 Solidification of HLW Generated from ATW Fuel Cycle .................................... 89
  8.1 Introduction ....................................................................................... 89
  8.2 HLW Generated from ATW Fuel Cycle ................................................... 89
  8.3 Determination of Optimum Waste Loadings ........................................... 91
    8.3.1 Empirical Formula for Density of Solidified HLW ......................... 91
    8.3.2 LP Problem for HLW Generated from Separation Process of LWR-SNF93
    8.3.3 LP Problem for HLW Generated from Separation Process of ATW-SNF95
  8.4 Effects of Composition Variation on the Optimum Waste Loading .............. 96
    8.4.1 Feasible Solution Space Bounded by Composition-Independent Constraints .......................................................... 96
    8.4.2 Composition Dependency of PCT-Limit Constraints ....................... 97
    8.4.3 Composition Dependency of Homogeneity Constraint ...................... 98
    8.4.4 Composition Dependencies of Constraints on Pu-concentration, Heat Generation Rate, and Temperature Limits ........................................ 99
    8.4.5 Necessary and Favorable Conditions of Composition-Dependent Constraints ........................................................................ 100
    8.4.6 Effects of Cooling Time on Heat Generation Rate .......................... 101
    8.4.7 Effects of Process Chemicals and Corrosion Products on Heat Generation Rate ........................................................................ 102
  8.5 Discussions and Conclusions ................................................................ 103
9 Expansion of the Yucca Mountain Repository Capacity by ATW Fuel Cycle .......... 105
  9.1 Introduction ................................................................................................................. 105
  9.2 Statutory YMR Capacity ............................................................................................. 105
  9.3 Waste Packages Generated from HLW Solidification ................................................. 105
      9.3.1 Determination of Mass of HLW for Solidification ................................................. 107
      9.3.2 Optimum Waste Loadings and Waste Packages .................................................. 109
      9.3.3 Reduction of the Number of Waste Packages ..................................................... 110
  9.4 Expansion of the YMR Capacity by ATW Fuel Cycle .................................................. 111
  9.5 Discussions .................................................................................................................... 111
10 Summary and Conclusions ............................................................................................... 113
11 Future Works .................................................................................................................... 116
REFERENCES ...................................................................................................................... 117
Appendix A: Reference ATW Plant Parameters in the Roadmap ........................................... 121
Appendix B: Input Data Card for MCNP Calculation for Startup-Core ................................. 122
Appendix C: Neutronics Used for Linear Fits and Formulations .......................................... 125
Appendix D: MOCUP Input Data Cards for Evaluation of End of Equilibrium Cycle
    Composition in ATW Transmuter ..................................................................................... 127
    D.1 Input Data Card for MCNP .......................................................................................... 127
    D.2 Input Data Card for ORIGEN2.1 ................................................................................ 134
Appendix E: Heat Generation Rate of HLW from LWR- and ATW-SNF .............................. 135
Appendix F: Application of LP Model for Solidification of HLW from PWR ....................... 137
    F.1 Introduction .................................................................................................................. 137
    F.2 Formulation of LP Problem ........................................................................................ 137
        F.2.1 Constraints for JNC-HLW Conditioning .............................................................. 137
        F.2.2 Composition Vectors for HLW and Glass Frit ..................................................... 138
    F.3 Results ......................................................................................................................... 138
List of Figures

Figure 1.1 Components of an ATW system [DOE/RW-0519, 1999] .......................... 3
Figure 1.2 Reference ATW plant sized to process 10,155 MT of spent fuel [Hill, et al., 1999] .......................................................... 4
Figure 1.3 Reference ATW plant implementation to address the 86,317 MT of spent fuel [Hill, et al., 1999] .......................................................... 5
Figure 2.1 Simplified ATW fuel cycle .......................................................... 9
Figure 2.2 The 18-nuclide chain considered for this study .................................. 10
Figure 2.3 Results of the numerical and analytical solutions for 1-year irradiation time .............................................................................. 16
Figure 3.1 Flow chart for a code, WACOM .................................................. 20
Figure 4.1 Proposed LBE-Cooled blanket configuration (192 fuel assemblies) [AAA-RPO-SYS-01-0008, 2001] .................................................. 22
Figure 4.2 Simplified LBE-Cooled point design geometry used for the MCNP simulation .............................................................................. 24
Figure 5.1 Fuel composition evolution with cycle number, calculated by using the effective one-group (n,γ) cross sections of the start-up core ...................... 32
Figure 5.2 Variations of effective one-group (n,γ) cross sections of U-isotopes with respect to the mass fraction of actinide in fuel .................................. 34
Figure 5.3 Revised flow chart of WACOM code ........................................... 35
Figure 5.4 Fuel composition variation with cycle number predicted by using interpolated neutronics .............................................................. 36
Figure 5.5 Cycle duration of the LBE-cooled ATW system ............................... 38
Figure 5.6 Cycle dependent in-core inventory of fuel isotopes in the LBE-cooled ATW system ............................................................................. 39
Figure 5.7 Fractional transmutation of TRU isotope with cycle number ......... 41
Figure 5.8 Cumulative mass reduction ratio of TRU with cycle number ......... 42
Figure 5.9 Cumulative mass fraction of TRU isotope in waste with cycle number .......................................................... 43
Figure 5.10 Cumulative toxicity reduction ratio of TRU with cycle number ...... 44
Figure 5.11 Cumulative toxicity fraction of TRU isotope in waste with cycle number .................................................................................. 45
Figure 5.12 Transmutation chain of Am-isotopes used in WACOM .................. 46
Figure 6.1 Mass fraction of TRU isotope in 900 MT TRU (before ATW treatment) and 5.291 MT TRU (after ATW treatment) ........................................... 54
Figure 6.2 Toxicity fraction of TRU isotope in 900 MT TRU and 5.291 MT TRU... 54
Figure 6.3 Variation of toxicity reduction factor with time after waste generation (not taken into account the actinides decay during time it takes to accumulate) ........................................................................ 55
Figure 6.4 Toxicity variation of TRU isotope with time after waste generation .... 56
Figure 7.1 An ATW fuel cycle and HLW mass flow ....................................... 59
Figure 7.2 A simplified HLW process flow of the DWPF at the SRTC ........... 60
Figure 7.3 The graphical solution of the linear programming problem for a US-defense HLW .................................................................................. 81
Figure 7.4 The graphical solution for the DWPF-HLW conditioning ............... 86
Figure 7.5 Revised graphical solution for the DWPF-HLW conditioning incorporating constraints of both EM-WAPS and the DWPF.................. 87
Figure 8.1 HLW streams of the ATW fuel cycle ....................................................... 90
Figure 8.2 Quadratic equalities and linear approximates for the filled waste volume constraint.......................................................... 92
Figure 8.3 The graphical solution of the linear programming problem for HLW generated from separation process of LWR-SNF................................. 94
Figure 8.4 The graphical solution of the linear programming problem for HLW generated from separation process of ATW-SNF................................. 95
Figure 8.5 The feasible solution space bounded by the composition-independent constraints only for solidification of US-defense HLW ......................... 97
Figure 8.6 The variation of the inequality of boron concentration limit with the variation of fractional boron in HLW and glass frit ......................... 98
Figure 8.7 The variation of the inequality lines for plutonium concentration limit, heat generation limit, and temperature limit .................................. 99
Figure 8.8 Dependency of heat generation rate on cooling time of discharged spent fuel (HLW generated from separation process of LWR-spent fuel with a burnup of 33,000 MWd/MT; process chemicals and corrosion products based on [PNC SN8410 90-061, 1990])........................................ 101
Figure 8.9 Dependency of heat generation rate on content of process chemicals and corrosion products (HLW generated from separation process of LWR-SNF with a burnup of 33,000 MWd/MT and a 25-year cooling time) ............ 102
Figure 9.1 Overview of waste-package generation paths from the ATW fuel cycle 106
Figure 9.2 Heat generation rate of HLW generated from reprocessing LWR-spent fuel with cooling time of discharged LWR-spent fuel......................... 109
Figure 9.3 Heat generation rate of HLW generated from reprocessing ATW-spent fuel with cooling time of discharged ATW-spent fuel ..................... 110
Figure F.1 Optimum waste loading estimated by the graphical solution method for JNC-HLW conditioning.................................................. 138
List of Tables

Table 2.1  Input Data for Comparison with Analytical Solution ........................................... 16
Table 4.1  Design Parameters for the Proposed LBE-Cooled Blanket Point Design [AAA-RPO-SYS-01-0008, 2001] ................................................................. 22
Table 4.2  Fuel Isotope Properties [KAERI, 2000] and PWR-TRU Composition .... 25
Table 4.3  Composition and Properties of LBE-Coolant [Spencer, 2000; Liquid Material Handbook, 1952] ................................................................. 25
Table 4.4  Composition and Properties of Structure Material HT-9 [IAEA-TECDOC-1039, 1997] ........................................................................................................... 26
Table 4.5  Neutronics of the Start-up Core Calculated by MCNP (TRU-31.8 wt%, Zr-68.2 wt%) ........................................................................................................ 27
Table 4.6  Fractional Mass Changes of TRU, Fission Product, and Zirconium for the Start-up Cycle Calculated by WACOM Using the Neutronics of the Start-up Core ........................................................................................................ 28
Table 4.7  Isotopic Composition Change of TRU for the Start-up Cycle Calculated by WACOM Using the Neutronics of the Start-up Core ........................................................................................................ 28
Table 4.8  Fractional Changes of Fuel Isotopes for Irradiation Time of a Year Calculated with WACOM and MOCUP Using the Neutronics of the Start-up Core ........................................................................................................ 29
Table 5.1  Effective One-Group (n,γ) Cross Sections of Several Fuel Compositions Plotted in Figure 5.1 (Estimated by MCNP) ................................................................................ 33
Table 5.2  k_{eff} -Values of 10^{th}, 20^{th}, and 100^{th} Fuel Composition in Figure 5.3 Estimated by MCNP ........................................................................................................ 37
Table 5.3  Comparison of Interpolated Capture Cross Sections with MCNP-Estimated Capture Cross Sections for the BOC Fuel Composition at the 100^{th} cycle ........................................................................................................ 37
Table 5.4  Isotopic Composition of TRU in the Start-up and the Equilibrium Core. 40
Table 5.5  Capture to Fission Ratios of TRU Isotopes in the Start-up and the Equilibrium Core ........................................................................................................ 40
Table 5.6  TRU Compositions in Fuel after a Single Cycle Irradiation ......................... 46
Table 6.1  Performance Characteristics of the Reference LBE-Cooled Transmuter Estimated by WACOM ........................................................................................................ 50
Table 6.2  Isotopic Compositions of 0.935 MT, 2.380 MT, and 1.976 MT TRU . 51
Table 6.3  Mass Reduction Factor of Each TRU-Isotope by the Reference ATW Plant ........................................................................................................ 52
Table 6.4  Toxicity Reduction Factor of Each TRU-Isotope by the Reference ATW Plant ........................................................................................................ 53
Table 7.2  Composition of Nominal MB3 sludge Used for Initial Model Assessments and Frit Development Activities [Peeler, et al., 2001] .................................................. 78
Table 7.3  Oxide Composition of Glass Frit 320 [Peeler, et al., 2001] ......................... 78
Table 7.4  Concentration, Decay Constant, and Average Decay Energy of Several Radionuclides in the HLW Sludge in DWPF MB3 [Bibler, et al., 2002] 79
Table 7.5  Constraints Applicable to DWPF Acceptability Decisions [Brown, 2002] ................................................................. 83
Table 8.1  Composition of HLW Generated from Separation Process of LWR-SNF with a Burnup of 33,000 MWd/MT and a 25-year cooling period ............ 93
Table 8.2  Composition of the Glass Frit, PF798, Developed by JNC [Yamashita, et al., 1995] .............................................................................. 94
Table 8.3  Composition of HLW Generated from Separation Process of ATW-SNF with a Burnup of 27.2 wt% and a 25-year cooling period ....................... 95
Table 8.4  Necessary Conditions of Composition-Dependent Constraints for HLW Solidification ........................................................................ 100
Table 8.5  Favorable Conditions of Composition-Dependent Constraints for Redundancy .................................................................................. 101
Table 9.1  Constituents and Mass of HLW for Table 8.2 and Table 8.3, and Waste Packages Generated: Per 1 MT of SNF, 22.3 kg-Na, 0.393 kg-P, 4.2 kg-Fe, 1.1 kg-Ni, and 1.09 kg-Cr Contained in HLW, 25-Year Cooling after Discharge before Reprocessing Applied ............................................................... 106
Table 9.3  Constituents and Mass of HLW for Case (2): 22.3 kg Na/MT-SNF and 33 kg Gd/MT-SNF Contained in HLW, 25-Year Cooling after Discharge before Reprocessing Applied ................................................................. 108
Table A. 1  Reference ATW Plant Parameters for the Separation Facility .......... 121
Table A. 2  Reference ATW Plant Parameters for the Accelerator ....................... 121
Table A. 3  Reference ATW Plant Parameters for the Transmuter Systems ........ 121
Table C. 1  Effective One-Group Cross Sections Used for Linear Fits ...................... 125
Table C. 2  Formulations Implemented in WACOM Code for Interpolating Neutronics ......................................................................................... 126
Table E. 1  Heat Generation Rate per kg-HLW from LWR-SNF (25-year cooling) 135
Table E. 2  Heat Generation Rate per kg-HLW from ATW-SNF (25-year cooling) 136
ACKNOWLEDGEMENT

I would like to express my sincere gratitude to Professor Joonhong Ahn and Professor Paul L. Chambré, who guided this work and helped me whenever I was in need. They gave me stimulating discussions and good suggestions related to this work. They also showed me clearly what the logical argument should be. If this dissertation is illogical, the responsibilities are all mine. Without their support, this work could not have been completed. I think their presence at UC-Berkeley was the best thing that could have happen to me and my dissertation.

I also wish to express my deep appreciation to Professor Ehud Greenspan, who provided me with many helpful and insightful comments. I appreciate Professor Stanley A. Berger for reviewing my thesis with a great passion.

I would like to thank the fellow graduate students in Nuclear Waste Lab for their assistance: Doo-Hyun Lim, Byung-Hyun Park, Daisuke Kawasaki, and Bob Borrelli.

I would like to acknowledge financial supports from the Korean Army, the United States Department of Energy, and the Japan Nuclear Cycle Development Institute.

Lastly, I would like to thank my wife, daughter, and parents for their continuous encouragement, support, and love. I would like to dedicate this thesis to them with my greatest thanks.

Myeongguk Cheon
Berkeley, California
May 2005
1 Introduction and Summary

1.1 Backgrounds/Motivations

Since the first nuclear power plant began its operation, spent nuclear fuel (SNF) has been accumulating steadily in the U.S. as in other countries having nuclear power plants [EIA, 2002]. With respect to the disposition of SNF, in the U.S., according to the Nuclear Waste Policy Act of 1982 [US DOE, 1982], spent nuclear fuel from commercial power reactors is required to be packaged and buried in a geological repository as well as DOE-spent fuel and defense high-level waste (HLW). For a geologic repository for commercial spent nuclear fuel (CSNF) and DOE-owned SNF and HLW, a project has been ongoing to develop the site at Yucca Mountain, Nevada [US DOE, 1998; US DOE, 2002].

SNF from light-water reactors (LWRs) contains uranium, plutonium, other actinides, and fission products. Of the species in LWR-spent fuel, some actinide and fission product isotopes have relatively long half-lives, and challenge the repository performance because it is difficult to predict performance hundreds of thousands of years into the future [Benedict, et al., 1981]. The treatment of spent fuel could simplify some of the technical difficulties of geologic disposal and such treatment can be considered as a technology option to enhance repository development [Pigford, et al., 1983; Croff, et al., 1990].

A concept developed recently for reducing the amount and toxicity of this spent fuel is Accelerator-Driven Transmutation Technology (ADTT). Transmutation refers to a nuclear transformation that converts one isotope into another by nuclear reactions. In terms of dealing with the long-lived constituents of SNF (roughly 1%), this involves fissioning the transuranic (TRU) isotopes and converting the long-lived fission products (FP) into short-lived or stable isotopes [Van Tuyle, et al., 1999].

One of the most effective methods to achieve nuclear transmutation is through exposure of material to neutrons, either in a critical nuclear reactor or in an accelerator-driven subcritical nuclear system. In the latter, additional neutrons result from a beam of high-energy particles that collide with a dense, high atomic number target [Sailor, et al., 1994]. The leading ADTT concept in the U.S. is the Accelerator-Driven Transmutation of Waste (ATW) system [Van Tuyle, et al., 1999].

In 1999, the US DOE has prepared a roadmap [DOE/RW-0519, 1999] for developing ATW technology and several important issues related to ATW development were assessed. The conclusion of the roadmap associated with impacts of ATW technology on the civilian spent fuel program was the following: “Applying an ATW system to commercial civilian spent fuel as described in this report, would reduce its contribution to the dose predicted in the total system performance analysis for the Yucca Mountain repository project... The inventory of fissionable materials from commercial spent fuel in the repository could be reduced by a factor of 1,000. The volume of waste packages associated with commercial spent fuel would be slightly reduced.”

These results are based on several key assumptions such that (1) only commercial spent fuel is processed using ATW technology and (2) the ATW process removes 99.9% of the TRU and 95% of 99Tc and 129I from the CSNF. This indicates that long-lived radioactivity, which could be generated from reprocessing ATW-spent fuel, from the fabrication process of ATW fuel, and from decommissioning of an ATW system, was neglected. With respect to long-lived radioactivity, thus, the roadmap identified the quantification of total long-lived radioactivity generated from the ATW system based on
more realistic considerations as one of key R&D issues to be addressed for ATW development.

With respect to separation processes, pyrochemical processes for spent fuel processing and ATW fuel-processing operations are assumed. For waste forms from separation processes, two types of HLW forms (ceramic and metal) are considered in the roadmap. For waste forms and separation processes, the roadmap identified several key technical issues to be addressed for ATW development. The key technical issues of the ceramic form are (1) the definition of the waste stream composition, (2) the development of full-scale processing methods, and (3) the development of waste minimization and salt recycling technology. The key technical issues for the metal waste form are (1) the definition of the waste stream composition, (2) the characterization and qualification of zirconium-8 stainless steel waste form, (3) the development of full-scale processing methods for salt removal and casting, and (4) the definition of process residuals that must go to the waste form. Particularly in the case of the ceramic waste form, the roadmap emphasizes on means to minimize the volume of HLW produced.

From the conclusions and the key technical issues on long-lived radioactivity and waste forms identified in the roadmap, followings could be questioned:

- Based on more realistic considerations in ATW waste streams, to what extent the inventory of TRU could be reduced?
- By the quantification of waste stream composition and the development of waste minimization technology, to what extent the volume of waste packages related to CSNF could be reduced?

The inventory of TRU being included in ATW waste streams is subject to several elements such as a type of transmuter, transmuter operating mode, recovery yields of separation processes for TRU isotopes, and so on. The volume of waste packages being produced from reprocessing of both LWR-SNF and ATW-SNF depends largely on the design of a separation process related to process chemicals [Benedict, et al., 1981] in addition to those to which the inventory of TRU subjects.

In this study, thus, to perform analyses of ATW system to answer above questions:

- A deployment scenario of ATW system in the roadmap is defined.
- As a transmuter of ATW system, a lead-bismuth eutectic (LBE) cooled transmuter proposed in [AAA-RPO-SYS-01-0008, 2001] as an optimized transmuter with respect to transmutation performance is considered.
- For an operating mode of a transmuter, similar parameter values that are assumed in the ATW deployment scenario are used. For more reliable analyses, however, a cycle-by-cycle calculation for fuel inventory evolution in a transmuter core is performed instead of an equilibrium cycle analysis\(^1\).
- For TRU loss to waste streams, same recovery yields that are assumed for the ATW system are used but TRU loss also occurs in separation of ATW-spent fuel as well as in separation of LWR-spent fuel.
- For the design data of a separation process related to process chemicals, to quantify waste stream composition, design data of a PUREX plant are referred due to unavailability of design data of pyrochemical processes.

\(^1\) The needs for a cycle-by-cycle calculation are issued in [AAA-RPO-SYS-01-0008, 2001; Yang, et al., 2001]
• As an effort to minimize the volume of waste packages, an optimization approach is applied to HLW solidification process.

1.2 State of the Arts

1.2.1 Reviews of ATW Systems

The ATW system consists of three major technologies: (1) the chemistry processes that allow the spent fuel components to be separated, (2) the accelerator technology needed to provide a high power beam of charged particles, and (3) the target/blanket technology needed to transmute the long-lived hazards into stable or short-lived materials. Figure 1.1 shows components of an ATW system.

![Figure 1.1 Components of an ATW system](DOE/RW-0519, 1999)

1.2.1.1 Reference ATW Plant in the Roadmap

A reference ATW plant was specified in a Roadmap for Developing ATW Technology [Hill, et al., 1999] to process 86,317 metric tons (MT) of initial heavy metal of spent nuclear fuel. This quantity will accumulate by year 2036 from existing U.S. nuclear power plants without new license and license extensions. The ATW plant consists of several subcritical transmuters driven by accelerators, a separation facility, and a fuel fabrication facility. The reference ATW plant is designed to produce electricity by fissioning the transuranics (TRU) taken from LWR-SNF during its life-time. It is also supposed to transmute long-lived fission products such as technetium and iodine into short-lived or stable isotopes as well as TRU. Figure 1.2 shows the reference ATW plant sized to process 10,155 MT of spent fuel in 60 years.
The separations process in Figure 1.2 includes three steps. The uranium is first removed via the UREX process and then by an oxide-reduction process, transuranics (TRU) and fission products (FP) are converted from oxide to metallic form. Finally the transuranic components are separated and converted into ATW fuel. 99.9% of the TRU and 95% of Tc and I in LWR-SNF are supposed to be separated from uranium and other fission products.

The reference ATW plant produces a net power of 6,720 MWt by fissioning 1.766 MT TRU per year. It has eight subcritical transmuters and thus each transmuter will process 1/8 of 1.766 MT TRU per year. During the 60-year plant life time, an ATW plant is supposed to consume ~106 MT of TRU.

Reference ATW plant parameters used in the roadmap for the separation facility, the accelerator, and the transmuter systems are listed in Appendix A.

---

**Spent Nuclear Fuel**

10,155 MT (169 MT/yr for 60 years)

**Separations process for spent fuel from current light-water reactors**

- 2.3 MT Iodine
- 8.1 MT Technetium
- 106 MT TRU

**ATW fuel processing**

Fresh fuel: 5.87 MT/yr

Used fuel: 30% burned

**Separation Waste**

355 MT stable or short-lived FPs
- 0.43 MT Tc-99
- 0.12 MT I-127 & 129
- 0.11 MT TRU

**Uranium**

9,684 MT
(Recycling or Class C low-level waste)

**ATW Separation Waste**

Stable or short-lived FPs: ~116 MT

**Subcritical ATW Systems**

- 45 MW proton beam
- 8 targets/blankets, 840 MWt each
- 2 Accelerators and proton beams

---

**Figure 1.2 Reference ATW plant sized to process 10,155 MT of spent fuel [Hill, et al., 1999]**
1.2.1.2 Reference ATW Implementation Scenario

Figure 1.3 shows the reference ATW plant implementation scenario. Around 2030, an ATW power plant which is a prototype and has four subcritical transmuters would begin operating and will process about 5,000 MT of LWR-SNF. The full deployment of an eight-unit ATW system could then proceed.

![Diagram showing the reference ATW plant implementation scenario.](image)

**Figure 1.3 Reference ATW plant implementation to address the 86,317 MT of spent fuel [Hill, et al., 1999]**

1.2.2 Reviews of Available Codes

To quantify the composition of waste stream generated from reprocessing LWR-SNF or ATW-SNF, various reactor analysis codes can be used. For the analysis of LWR system, ORIGEN code [*RSICC Code Package CCC-371, 1999*], a point depletion burnup code using matrix exponential method, is widely used. It utilizes its own effective one-group cross-section and decay libraries for calculation of fuel inventory evolution in a reactor core. For the analysis of ATW system, for which effective cross-section libraries are not available, reactor analysis code systems that has a transport code coupled with a burnup code can be used. The fuel cycle analysis of ATW can be performed rigorously using Monte Carlo calculations coupled with detailed depletion calculation such as MOCUP [*Moore, et al., 1995*] or MONTEBURNS [*Poston, et al., 1999*]. However, applying these codes to study the time evolution of the fuel constituents in the ATW system requires significant effort and computer time [*Yang et al., 1999*]. The effort is particularly demanding if the analysis is to include re-designing the core each cycle so as to get a given value of $k_{eff}$ for the beginning of cycle (BOC) and another given value of $k_{eff}$ at the end of cycle (EOC). Some other available code systems, such as REBUS-3 [*Toppel, 1983*], can automatically find the fuel composition of a reactor system in the equilibrium cycle without a significant effort. But, for the analysis of a reactor system with recycle of the discharged fuel in transition rather than in equilibrium condition,
required efforts are appreciable. Thus, for the cycle-by-cycle analysis of ATW system, a more efficient but also reliable fuel cycle analysis model is required.

1.2.3 Disposal Form of High-Level Waste

HLW contained in waste streams from reprocessing LWR-SNF and ATW-SNF is solidified with matrix materials such as glass or ceramic and then packaged in a waste package for final disposal [US DOE, 1982]. For solidified HLW products to be accepted into the Civilian Radioactive Waste Management System (CRWMS), those should satisfy regulations and guidelines specified in [DOE/EM-0093, 1996]. If we could achieve a maximum waste loading into a waste package while satisfying constraints of [DOE/EM-0093, 1996], a minimized number (or volume) of waste packages containing HLW could be expected from ATW fuel cycle. For this, an optimization scheme would be useful.

By comparing the inventory of TRU before ATW processing with that after ATW processing, the reduction of TRU contained in LWR-spent fuel will be quantitatively estimated.

By comparing the number of waste packages before ATW processing with that after ATW processing, the impacts of ATW fuel cycle on a repository capacity will be quantitatively evaluated.

1.3 Objectives

The objectives of this dissertation are:

- To evaluate the reduction of TRU inventory contained in CSNF by the reference ATW system based on more realistic considerations than done in the roadmap. For this, a fuel cycle analysis tool for multi-cycle burnup analyses of ATW system will be developed. It should be able to calculate a cycle-by-cycle fuel inventory evolution of a transmuter core in transition from startup to equilibrium efficiently and reliably. For efficiency, computing time of the model developed will be checked and compared. For reliability, it will be benchmarked with other available tools.
- To evaluate the volume reduction of waste packages related to CSNF quantitatively based on waste stream compositions identified by the fuel cycle analysis tool and on an optimized HLW solidification. For this, a model for HLW solidification will be developed, using a linear programming approach.
- To evaluate the expansion of the Yucca Mountain Repository capacity by the reference ATW system quantitatively. The evaluation will be performed based on reference cases which could reflect the effects of waste minimization technology.

1.4 Plans of This Study

In the later part of this chapter, an overview of ATW system proposed by Los Alamos National Lab (LANL) is provided. Major components of the ATW system, system parameters, key technologies, and system deployment scenario are summarized.

The mathematical model for the analysis of ATW fuel cycle is described in Chapter 2 & 3. In Chapter 2, a reactor model for the calculation of fuel inventory evolution by nuclear reactions in a reactor core is developed. A transmutation chain for 18 actinide
isotopes is modeled, for which a set of coupled differential equation system is developed and a numerical solution method is implemented. To verify the numerical solution method implemented in the code WACOM, an analytic solution for two nuclide chain is obtained and it is compared with the numerical solution. In Chapter 3, a mass flow model for external fuel cycle is provided. The model describes fractional fuel discharge from transmuter core, separation of TRU and zirconium from the discharged fuel, determination of FPs and residual TRU in waste stream, and determination of makeup materials based on system constraints.

In Chapter 4, calculation procedures for a single cycle analysis of a reference LBE-cooled transmuter and a computer code are developed and benchmarked. For neutronics input, MCNP code [RSICC Code Package CCC-700, 2000] is applied to the startup core composition of the transmuter. With neutronics obtained, the computer code developed calculates fuel inventory evolution of the transmuter for a-year irradiation time and the results are benchmarked against MOCUP code [Moore, et al., 1995].

Multi-cycle analysis of the LBE-cooled transmuter using the code is described in Chapter 5. To take into account neutronics variation with fuel composition change, a linear interpolating scheme is implemented in the model developed in Chapter 4 based on two sets of MCNP-generated effective one-group cross sections2. For several sets of BOC- and EOC-fuel compositions evaluated by the implemented code (named “WACOM”) in multi-cycle calculations, their $k_{eff}$ values are checked by MCNP to validate the linear interpolating scheme implemented for revising the effective one-group cross sections with respect to the variation of fractional TRU mass in a transmuter core. Performance characteristics of the LBE-cooled transmuter in transition from startup to equilibrium are investigated.

In Chapter 6, the reduction of TRU contained in CSNF by the ATW system is discussed. Overall TRU waste being contained in waste streams of ATW fuel cycle is quantified based on the ATW deployment scenario described in [Hill, et al., 1999]. Then, the quantified TRU is compared with that contained in CSNF in terms of mass and toxicity.

In Chapter 7, a model for solidification of HLW being contained in waste streams of the ATW fuel cycle is developed. For this, US-defense HLW solidification processes are investigated and modeled based on related regulations/guidelines set by US DOE or a waste processing facility. To minimize the volume of waste packages being produced from solidification processes, an optimization scheme is implemented. The model developed is applied to the solidification of a US-defense HLW and the resulting optimum values are compared with those estimated by the Defense Waste Processing Facility (DWPF) managed by Savannah River Technology Center (SRTC) in the US. Assuming that the same regulations/guidelines are applied to the HLW generated from ATW fuel cycle, the developed model is applied for the determination of optimized waste loading and composition of solidified HLW from ATW fuel cycle in Chapter 8.

In Chapter 9, the expansion of the Yucca Mountain Repository (YMR) capacity by the ATW fuel cycle is evaluated quantitatively based on two reference cases: (1) a case that no process chemicals are included in HLW and thus only FPs and residual TRU are included in HLW, and (2) a case that a significant amount of process chemicals such as

---

2 One for the startup core, the other for pseudo-equilibrium core estimated using startup-core neutronics.
sodium and gadolinium are included in the HLW in addition to FPs and residual TRU, and thus resulting in significant amount of HLW for final solidification.

Summary and conclusions of this study are summarized in Chapter 10. Chapter 11 describes future works.

1.5 Scopes and Limits

In this study, most of the analyses are performed based on the ATW system deployment scenario specified in [Hill, et al., 1999]. For a transmuter, however, an optimized LBE-cooled transmuter design [AAA-RPO-SYS-01-0008, 2001] is employed and its life-time performance characteristics (rather than equilibrium performance characteristics) evaluated by WACOM are used for ATW fuel cycle analysis.

For long-lived radionuclides, 18 actinide isotopes are taken into account in a transmutation chain. Transmutation of long-lived fission products such as $^{99}$Tc and $^{129}$I is not treated here even though a key feature of the ATW system is the capability of transmuting long-lived fission products as well as TRU$^3$ [Van Tuyle, et al., 1999]. This simplification does not affect on the evaluation of TRU inventory or waste package reduction.

As waste streams of ATW fuel cycle, we consider liquid HLW streams only generated from separation processes of LWR-spent fuel and ATW-spent fuel. 100% of zirconium cladding generated from reprocessing LWR-spent fuel is assumed to be recycled as ATW fuel matrix materials, and zirconium in ATW fuel is also assumed to be separated with a recovery yield of 100% and recycled many times. Solid HLW such as cladding hulls generated from ATW-spent fuel chopping is not taken into account as the volume of waste packages. Assumptions on zirconium recycling and solid HLW would result in an underestimated volume of waste packages from ATW fuel cycle. Investigations on the effects of zirconium recycling and solid HLW on the volume of waste packages are beyond the current scope. TRU loss from ATW fuel fabrication process is assumed to be zero.

$^3$ Actinide transmuting by itself would make only a negligible reduction in the risk to the public from contaminated water from the repository, because the species most likely to migrate from the repository are the water-soluble long-lived fission products, especially $^{99}$Tc, $^{135}$Cs, and $^{129}$I [Pigford, 1991].
2 Reactor Model

We consider an ATW fuel cycle, shown in Figure 2.1. This ATW fuel cycle consists of an ATW-reactor, a partitioning plant, a fuel-fabrication facility, a makeup material storage, and a repository. Actinides are assumed to be recovered from the discharged fuel by the partitioning plant and passed through the fabrication plant before recycling. A cycle starts at the beginning of irradiation in the reactor. At the end of one irradiation period, the fraction, $f$, of the fuel in the core is discharged and partitioned. Recovered actinides are mixed with makeup materials (actinides and zirconium) and fabricated into fuel, which is returned to the reactor core. The initial composition of the fuel in the core for the next irradiation is determined. This is the point where one cycle ends. For sufficient transmutation of toxic actinides, this cycle needs to be repeated many times, as the numerical results from this study show.

The model is based on the following assumptions: (1) The reactor core is homogenized. (2) The recharged fuel is mixed instantaneously and homogeneously with the fuel that stays in the core. (3) The reactor is operated at a constant power. (4) The fission products (FP) effect on the neutron multiplication factor $k_{eff}$ is accounted for by considering a fictitious FP pair. (5) Effective one-group cross sections are used for reaction rate calculations. These are generated by MCNP [RSICC Code Package CCC-700, 2000] for the representative core composition and assumed to be constant within a cycle. (6) The total fuel volume in the reactor is constant. (7) The actinides-to-Zr ratio is adjusted at each beginning of the cycle (BOC), so that the BOC $k_{eff}$ will be 0.98. (8) The cycle terminates when $k_{eff}$ drops to 0.92.

The present model can be divided into two parts: (1) determination of the fuel composition by nuclear reactions and decays in the reactor core and (2) determination of fuel composition after partitioning, fabrication, and reloading. In this chapter, we develop mathematical formulation for a mass flow model for a certain cycle.

![Figure 2.1 Simplified ATW fuel cycle](image)

2.1 Transmutation Chain of Actinides

We develop a model for $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, $^{237}$Np, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{241}$Am, $^{242m}$Am, $^{243}$Am, $^{242}$Cm, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm, and $^{246}$Cm. These nuclides are indexed by $i$ from 1 to 18 in this order.

Figure 2.2 shows the 18-nuclide chain considered in this work. The number in the parenthesis of each box shows the nuclide index, $i$. Three types of neutron reactions and four types of decay modes are considered to account for the mass change of a nuclide. If a nuclide captures a neutron, it turns into either a nuclide with the mass number greater...
by one, emitting a photon, or a nuclide with the mass number smaller by one, emitting two neutrons. These are represented by vertical arrows in Figure 2.2.

Upon neutron absorption, fission may also occur. The fission reaction is represented by letter “f” in Figure 2.2. A downward diagonal arrow represents an alpha decay, while a horizontal arrow indicates a beta (or EC) decay.

Figure 2.2 The 18-nuclide chain considered for this study

The short-lived nuclides, $^{237}$U, $^{239}$U, $^{238}$Np, $^{239}$Np, $^{237}$Pu, $^{243}$Pu, $^{240}$Am, $^{242}$Am, $^{244}$Am, and $^{241}$Cm (6.75 day, 23.45 minute, 2.12 day, 2.36 day, 45.2 day, 4.96 hour, 50.8 hour, 16.0 hour, 10.1 hour, and 32.8 day [KAERI, 2000], respectively), boxed by dotted lines in
Figure 2.2, are not included in the present model. It is assumed that they decay into their
daughter nuclides immediately.

Knowing the values, $\tilde{N}^o$, at the beginning of the irradiation, the change of the
numbers of atoms of nuclides, $i = 1, 2, \ldots, 18$, in the fuel in the reactor core by the
nuclear reactions and the radioactive decays shown in Figure 2.2 is expressed by

$$\frac{d\tilde{N}(t)}{dt} = D\tilde{N}(t), \quad 0 < t \leq T,$$

subject to $\tilde{N}(0) = \tilde{N}^o$, \hspace{1cm} (2.1)

where $\tilde{N}(t)$ is the vector whose elements express the number of atoms of each nuclide
shown in Figure 2.2 during the irradiation period. The time $t = 0$ is the beginning of the
irradiation time in the cycle, while $t = T$ is the end of the irradiation period in the same
cycle. The value of the irradiation time length $T$ is to be determined by numerically
checking when the value of the neutron multiplication factor for the reactor core becomes
equal to the specified lower-bound value (in this study it is assumed to be 0.92).

The matrix $D$ accounts for the rate constants of the transmutation reactions and for
the radioactive decays, which are shown in Figure 2.2, and is formulated as

$$D = (\Gamma_p - \Gamma_d)\phi(t) + (A_p - A_d), \hspace{1cm} (2.2)$$

where

$$\Gamma_d = \text{diag}(\sigma_{a1}, \sigma_{a2}, \ldots, \sigma_{a18}), \hspace{1cm} (2.3)$$

$$A_d = \text{diag}(\lambda_1, \lambda_2, \ldots, \lambda_{18}). \hspace{1cm} (2.4)$$

\[
\begin{bmatrix}
0 & \sigma_{20,2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\sigma_{2,3} & 0 & \sigma_{20,3} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \sigma_{2,2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \sigma_{3,3} & \sigma_{20,4} & 0 & \sigma_{20,6} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \sigma_{3,3} & 0 & \sigma_{20,5} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \sigma_{4,4} & 0 & \sigma_{20,5} & 0 & \sigma_{20,8} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \sigma_{3,5} & \sigma_{20,6} & 0 & \sigma_{20,9} & 0 & \sigma_{20,11} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & \sigma_{5,5} & \sigma_{20,8} & 0 & \sigma_{20,10} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{6,6} & 0 & \sigma_{20,9} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,7} & 0 & \sigma_{20,10} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{8,8} & 0 & \sigma_{20,11} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,9} & 0 & 0.173\delta\sigma_{7,11} & 0 & 0.173\sigma_{20,13} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{20,12} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & (1-\delta)\sigma_{7,11} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,10} & 0 & \sigma_{20,12} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0.827\delta\sigma_{7,11} & 0 & 0.827\sigma_{20,13} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,14} & 0 & \sigma_{20,16} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,13} & 0 & \sigma_{20,15} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,15} & 0 & \sigma_{20,16} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,17} & 0 & \sigma_{20,17} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,18} & 0 & \sigma_{20,18} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,19} & 0 & \sigma_{20,19} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sigma_{7,20} & 0 & \sigma_{20,20} & 0 & 0 & 0 \\
\end{bmatrix}
\]

and
\[
\sigma_{a,i} = \sigma_{f,i} + \sigma_{2n,i} + \lambda_{\gamma,i},
\]

\(\lambda_i\) is the radioactive decay constant for nuclide \(i\), and is equal to the sum of those for alpha and other emissions. The decay constant for alpha emission is denoted specifically as \(\lambda_{a,i}\) for nuclide \(i\). In the nuclide-chain model shown in Figure 2.2, only the beta decay of \(^{241}\text{Pu}\) is included; other EC, \(\beta^+\), or \(\gamma\) decays expressed by dashed horizontal arrows in Figure 2.2 have so short half-lives that they are combined with previous reactions.

The quantity \(\delta\) is the branching ratio for the capture reaction in \(^{241}\text{Am}\). With the probability of \(\delta\), an atom of \(^{241}\text{Am}\) that has absorbed a neutron would be transmuted to an \(^{242}\text{Am}\) atom in the ground state, whereas with \((1-\delta)\) probability it would be transmuted to an \(^{242m}\text{Am}\) atom in the meta-stable state. An \(^{242}\text{Am}\) atom then decays to a \(^{242}\text{Cm}\) atom with the probability of 0.827, while it decays to \(^{242}\text{Pu}\) with the probability of 0.173.

The fuel consists of actinide nuclides as well as zirconium, which is included as the base matrix. It is assumed that the number of atoms of zirconium does not change by the neutron reaction. The rate of fission-product-pair generation by the fission of actinide atoms is expressed by

\[
\frac{dN_F(t)}{dt} = \sum_i \sigma_{f,i} \phi(t) N_i(t), \quad 0 < t \leq T,
\]

where \(N_F(t)\) is the number of fission-product pairs at time \(t\) in the reactor, \(\phi(t)\) is the time-dependent neutron flux. The time-dependent neutron flux is calculated from the expression,
\[
\phi(t) = \frac{6.242 \times 10^{18} P}{\sum_i \gamma_i \sigma_{f,i} N_i(t)},
\]

(2.7)

where \( P \) is the power [MWt] of the ATW reactor, which is assumed to be constant, and \( N_i(t) \) is a component of vector \( \vec{N}(t) \) in (2.1), representing the number of atoms of nuclide \( i \) at time \( t \) in the reactor. \( \gamma_i \) is the recoverable energy of nuclide \( i \) per fission, and is given by [Croff, 1983]

\[
\gamma_i = 1.29927 \times 10^{-3} Z^2 A^{1/2} + 33.12 \text{ [MeV]}.
\]

(2.8)

### 2.2 Multiplication Factor Formula

To simulate the fuel burnup in the reactor, the reactivity loss of fuel in a cycle is constrained in this model. The reactivity change of a reactor core can be expressed by the multiplication factor.

The multiplication factor, \( k \), characterizing the chain reaction is defined as [Duderstadt, et al., 1976]

\[
k = \frac{\text{Number of fission neutrons in one generation}}{\text{Number of fission neutrons in preceding generation}}.
\]

(2.9)

The multiplication factor characterizing a finite system is usually referred to as the effective multiplication factor denoted by \( k_{\text{eff}} \). In a finite system, neutrons produced from the preceding generation by fission reactions are absorbed in the system or leak out of the system. The neutrons absorbed in the system cause fission or capture reactions. Therefore, for a finite system, (2.9) can be modified as

\[
k_{\text{eff}} = \frac{\text{Number of fission neutrons in one generation}}{\text{Leakage + Number of neutrons absorbed in the system in preceding generation}}.
\]

(2.10)

In MCNP [RSICC Code Package CCC-700, 2000], calculating \( k_{\text{eff}} \) consists of estimating the mean number of fission neutrons produced in one generation per fission neutron started. A generation is the life of a neutron from birth in fission to death by escape, parasitic capture, or absorption leading to fission. The computational equivalent of a fission generation in MCNP is a \( k_{\text{eff}} \) cycle; i.e., a cycle is a computed estimate of an actual fission generation. Processes such as \((n,2n)\) and \((n,3n)\) reactions are considered internal to a cycle and do not act as termination. Therefore, \( k_{\text{eff}} \) is formulated as

\[
k_{\text{eff}} = \frac{\int_V \int_0^\infty \int_E v \Sigma_f \Phi(r,t,E,\Omega) dV dt dE d\Omega}{\int_V \int_0^\infty \int_E \nabla \cdot J dV dt dE d\Omega + \int_V \int_0^\infty \int_E \left( \Sigma_c + \Sigma_f + \Sigma_m \right) \Phi(r,t,E,\Omega) dV dt dE d\Omega},
\]

(2.11)

where the phase-space variables are \( t, E, \) and \( \Omega \) for time, energy, direction, and implicitly \( r \) for position with incremental volume \( dV \) around \( r \). The denominator is the loss rate, which is the sum of leakage, capture, fission, and multiplicity \((n,xn)\) terms. The multiplicity term is:
\[
\frac{1}{V} \int_{E}^{O} \int_{E}^{O} \int_{E}^{O} \int_{E}^{O} \int_{E}^{O} \sum_{m} \Phi dV dt dE d\Omega = \sum_{i}^{18} \eta_{i} N_{i} \bar{\sigma}_{a,i} \bar{\phi} \]

(2.12)

where

\[\eta_{i} = \frac{\nu_{i} \sigma_{i,f}}{\sigma_{i,a}}, \quad i=1,2,...,18,\]

\[\nu_{i} = \text{the average number of neutrons produced per neutron-induced fission in nuclide } i,\]

\[\bar{\sigma}_{a,F} = \text{the effective one-group absorption cross section of a fictitious fission-product pair},\]

\[\bar{\sigma}_{2n,F} = \text{the effective one-group (n,2n) cross-section of a fictitious fission-product pair},\]

\[\bar{\sigma}_{a,Zr} = \text{the effective one-group absorption cross-section of zirconium},\]

\[\bar{\sigma}_{2n,Zr} = \text{the effective one-group (n,2n) cross-section of zirconium},\]

\[\bar{\sigma}_{a,p} = \text{the effective one-group absorption cross-section of an element, } p, \text{ in the structure or the coolant of the reactor},\]

\[\bar{\sigma}_{2n,p} = \text{the effective one-group (n,2n) cross-section of an element, } p, \text{ in the structure or the coolant of the reactor},\]

\[\bar{\phi} = \text{the space, solid angle, time, and energy averaged neutron flux}.
\]

The numerator in (2.13) represents the number of neutrons generated by neutron-induced fission, while the denominator denotes the number of neutrons lost by absorption reactions and leakage. In the denominator, the neutron multiplicity term such as (n,2n) or (n,3n) reaction is not treated as a termination as in MCNP but (n,3n) reaction rate is neglected due to its very small fraction compared to (n,2n) reaction rate. The leakage is the number of neutrons that leak out of the homogenized core. It is estimated from the MCNP calculated neutron balance.

2.3 Numerical Scheme

To solve (2.1) numerically, a predictor-corrector method was used [Chapra, et al., 1998]. This method uses two derivatives to improve the estimate of the slope for a certain interval—one at initial point and another at the end point. The two derivatives are averaged to obtain an improved estimate of the slope for the entire interval. To extrapolate linearly
from the value at the beginning of an interval to the value at the end of the interval, the
slope at the beginning point is used. However, the obtained value from the slope at the
previous point is not the final answer, but an intermediate prediction. This intermediate
value is called a predictor. It allows the calculation of an estimated slope at the end of the
interval. Thus, the two slopes can be combined to obtain an average slope for the interval.
This average slope is then used to extrapolate linearly from the previous value to the next
value. The newly obtained value at the end point of the interval is called a corrector. If
we apply this method to (2.1) for a discrete time step, \([t_j, t_{j+1}]\), (2.1) can be expressed as
\[
\frac{\tilde{N}_n^{(j+1)} - \tilde{N}_n^{(j)}}{\Delta t_n^{(j)}} = D_n^{(j)} N_n^{(j)}, \quad \Delta t_n^{(j)} = t_{j+1} - t_j. \tag{2.15}
\]
From this, a predictor distinguished with a superscript \(\text{prime}\) is obtained by
\[
\tilde{N}_n^{(j+1)'} = \tilde{N}_n^{(j)} + D_n^{(j)} \tilde{N}_n^{(j)} \Delta t_n^{(j)}, \tag{2.16}
\]
and then a corrector is expressed as
\[
\tilde{N}_n^{(j+1)} = \tilde{N}_n^{(j)} + \frac{1}{2} [D_n^{(j)} \tilde{N}_n^{(j)} + D_n^{(j+1)} \tilde{N}_n^{(j+1)'}] \Delta t_n^{(j)}. \tag{2.17}
\]
The percent relative error, \(\varepsilon\), is defined as
\[
\varepsilon = \frac{\text{current approximation} - \text{previous approximation}}{\text{current approximation}} \times 100\%. \tag{2.18}
\]
The size of time interval can be determined based on that the absolute value of the
relative error becomes less than a prescribed value for a certain time step. For I-nuclides,
we obtain I-different estimates for the size of time interval. The smallest one is the choice
for the next step.

### 2.4 Verification of Numerical Scheme for 2-Nuclide Chain

To check the numerical scheme developed for the calculation of the fuel composition
change in the reactor, analytical solutions for a simplified two-nuclide chain consisting of
\(^{235}\text{U}\) and \(^{236}\text{U}\) are obtained under the condition of a constant neutron flux. If we neglect
the effect of \(\alpha\) decay in two nuclides, the rate equation for \(^{235}\text{U}\) (\(N_1\)) is
\[
\frac{dN_1}{dt} = -\bar{\sigma}_{1,a} \phi N_1 + \bar{\sigma}_{2,\gamma} \phi N_2, \quad N_1(t = 0) = N_1^0, \tag{2.19}
\]
and the rate equation for \(^{236}\text{U}\) (\(N_2\)) is
\[
\frac{dN_2}{dt} = -\bar{\sigma}_{2,a} \phi N_2 + \bar{\sigma}_{1,\gamma} \phi N_1, \quad N_2(t = 0) = N_2^0. \tag{2.20}
\]

Using the Laplace transform method, we can get the solutions of (2.19) and (2.20) as follows:
\[
N_1(t) = \frac{\bar{\sigma}_{2,a} N_1^0 + \bar{\sigma}_{2,\gamma} N_2^0}{\alpha - \beta} \phi (e^{\alpha t} - e^{\beta t}) + \frac{N_1^0}{\alpha - \beta} (\alpha e^{\alpha t} - \beta e^{\beta t}), \tag{2.21}
\]
\[ N_2(t) = \frac{\bar{\sigma}_{1,a} N_1^0 + \bar{\sigma}_{1,a} N_2^0}{\alpha - \beta} \phi(e^{\alpha t} - e^{\beta t}) + \frac{N_2^0}{\alpha - \beta} (\alpha e^{\alpha t} - \beta e^{\beta t}), \]  

(2.22)

where

\[ \alpha = \frac{-\phi(\bar{\sigma}_{1,a} + \bar{\sigma}_{2,a}) + \sqrt{(\bar{\sigma}_{1,a} - \bar{\sigma}_{2,a})^2 + 4\bar{\sigma}_{1,a} \bar{\sigma}_{2,2n}}}{2}, \]  

(2.23)

\[ \beta = \frac{-\phi(\bar{\sigma}_{1,a} + \bar{\sigma}_{2,a}) - \sqrt{(\bar{\sigma}_{1,a} - \bar{\sigma}_{2,a})^2 + 4\bar{\sigma}_{1,a} \bar{\sigma}_{2,2n}}}{2}. \]  

(2.24)

Table 2.1 shows the input data used for this comparison. To compare under the same condition, a constant value of the neutron flux was used for both. The percent relative error to determine the size of time step in the developed model was applied as 0.01%.

Figure 2.3 shows the results of the numerical and analytical solutions for 1-year irradiation time. Two results show a very good agreement with each other.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>( \sigma_0 ) [b]</th>
<th>( \sigma_1 ) [b]</th>
<th>( \sigma_{2n} ) [b]</th>
<th>( N(t=0) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{235}\text{U} )</td>
<td>5.5846E-1</td>
<td>5.58E-1</td>
<td>4.61E-4</td>
<td>2.2223E+27</td>
</tr>
<tr>
<td>( ^{236}\text{U} )</td>
<td>4.8633E-1</td>
<td>4.86E-1</td>
<td>3.25E-4</td>
<td>2.2223E+27</td>
</tr>
</tbody>
</table>

Figure 2.3  Results of the numerical and analytical solutions for 1-year irradiation time
3 Fuel Cycle Model

3.1 Fuel Discharge and Partitioning

After irradiation, the fraction $f$ of the irradiated fuel is discharged. To describe the mass flow outside the reactor core, several vectors with different notations are introduced. The number of atoms of a nuclide discharged from the reactor core at the end of the $n$-th cycle is expressed by the vector, $\vec{N}_{D,n}$; the number of atoms of a nuclide recovered from the discharged fuel by the vector, $\vec{N}_{R,n}$; the number of atoms of a nuclide ending up in HLW by the vector, $\vec{N}_{W,n}$; the number of atoms of a nuclide in makeup material by the vector, $\vec{N}_{M,n}$; and the number of atoms of a nuclide in re-charged fuel by the vector, $\vec{N}_{C,n}$.

$\vec{N}_{D,n}$ can be related to the vector, $\vec{N}(T_n^o + T_n)$ whose elements are the number of atoms of each nuclide in the reactor core at the end of the $n$-th cycle as

$$\vec{N}_{D,n} = f\vec{N}(T_n^o + T_n),$$

where $T_n^o$ is the time at the beginning of the $n$-th cycle, $T_n$ is the irradiation time of the $n$-th cycle.

The volume of the discharged fuel, $V_D$, is expressed as

$$V_D = fV(T_n),$$

where $V(T_n)$ is the volume of the fuel in the reactor at the end of irradiation in the $n$-th cycle.

For modeling of external fuel cycle, it is assumed that partitioning of the discharged fuel, fuel fabrication, and fuel recharge are done instantaneously. So, the vectors introduced in this section are treated time-independent.

In the partitioning stage, actinides and zirconium in the discharged fuel are recovered for the next cycle. Any residual actinides and zirconium that are not recovered, together with all fission products in the discharged fuel are solidified and sent to the repository as HLW. The recovery fraction of element $e$, $R_e$, in the partitioning is defined as the ratio of the number of atoms of element $e$ recovered from the discharged fuel to that in the discharged fuel before the partitioning occurs. Index $e$ is used for numbering column in Figure 2.2. For example, all U isotopes are in column 1, all Np isotopes are in column 2, and so on. By mass balance,

$$\vec{N}_{D,n} = \vec{N}_{R,n} + \vec{N}_{W,n}.$$  \hspace{1cm} (3.3)

We can write $\vec{N}_{R,n}$ and $\vec{N}_{W,n}$ as

$$\vec{N}_{R,n} = R\vec{N}_{D,n} = Rf\vec{N}(T_n^o + T_n),$$  \hspace{1cm} (3.4)

$$\vec{N}_{W,n} = (I - R)\vec{N}_{D,n},$$  \hspace{1cm} (3.5)

where $I$ is the unit matrix and $R$ is the recovery fraction matrix. Elements of matrix $R$ are all zero except for the diagonal elements. The diagonal elements are equal to the recovery fractions of corresponding elements. For example, the fifth diagonal element is equal to
$R_{Np}$, which is the recovery fraction of $^{237}$Np at the partitioning. It is assumed that the isotopes of the same element have the same recovery fraction. For example, the first to the fourth diagonal elements correspond to the U isotopes in Figure 2.2. Therefore, these diagonal elements have the same value, $R_U$. The recovery fraction for element $e$ must be in the range $0 \leq R_e < 1$.

### 3.2 Determination of Makeup Fuel

In the fuel-fabrication facility, the recovered material inventory represented by the vector $\vec{N}_{R,n}$ is mixed with the makeup material inventory which is described by the vector $\vec{N}_{M,n}$. A fuel with $\vec{N}_{C,n}$ is fabricated and loaded into the reactor core. By mass balance,

$$\vec{N}_{C,n} = \vec{N}_{R,n} + \vec{N}_{M,n}. \quad (3.6)$$

By the volume constraint of this system, the volume, $V_o$, of the homogenized fuel at the beginning of each cycle should be kept constant through all cycles and thus, the volume, $V_{C,n}$, of the recharged fuel at the $n$-th cycle is determined as

$$V_{C,n} = V_o - V(T_n) + V_{D,n}. \quad (3.7)$$

$V_{C,n}$ can also be expressed in terms of the recovered material volume and the makeup material volume as

$$V_{C,n} = V_{R,n} + V_{M,n}, \quad (3.8)$$

where $V_{R,n}$ is the volume of the recovered material, and $V_{M,n}$ is that of the makeup material. $V_{R,n}$ is calculated from

$$V_{R,n} = \vec{\mu}^T \cdot \vec{N}_{R,n}, \quad (3.9)$$

where the elements of the vector, $\vec{\mu}$, are the reciprocals of the atomic density of nuclide $i$, and $\vec{\mu}^T$ is expressed as $\vec{\mu}^T = \{\rho_1^{-1}, \rho_2^{-1}, \rho_3^{-1}, \ldots, \rho_{18}^{-1}\}$.

The volume of makeup material, $V_{M,n}$, is the sum of the volume of actinides and that of zirconium. We set the volume fraction of actinides in the makeup material as $\omega$. Then, the volume of actinides in the makeup material is $\omega V_{M,n}$. We determine $\omega$ in such a way that the recharged fuel has a sufficiently high actinide content so that the whole core at the beginning of the next irradiation period has a $k_{eff}$ value of 0.98. The makeup material consists of actinides and zirconium taken from LWR-spent fuel. The atomic fraction, $x_i$, of actinide nuclide $i$ in the makeup material is assumed to be constant. Then,

$$\omega V_{M,n} = N_{tot,M,n} \sum_i \frac{x_i}{\rho_i}, \quad 0 \leq \omega \leq 1. \quad (3.10)$$

The total number of atoms, $N_{tot,M,n}$, of actinides in the makeup material at the $n$-th cycle and the number, $N_{i,M,n}$, of atoms of an actinide nuclide $i$ in the makeup material are related by

$$N_{tot,M,n} = \sum_i N_{i,M,n} \quad \text{and} \quad N_{i,M,n} = x_i N_{tot,M,n}. \quad (3.11)$$
The volume of zirconium in the makeup material, \((1-\omega)V_{M,n}\), can be written as

\[
(1-\omega)V_{M,n} = \frac{N_{Zr,M,n}}{\rho_{Zr}},
\]

where \(\rho_{Zr}\) is the atomic density of zirconium.

The number of fuel atoms at the beginning of the \((n+1)\)-st cycle is determined by summing the fuel remaining in the reactor core and the recharged fuel and thus

\[
\tilde{N}_{n+1}^{o} = (1-f)\tilde{N}_{n} (T_{n}^{o} + T_{n}) + fR\tilde{N}_{n} (T_{n}^{o} + T_{n}) + \tilde{N}_{M,n}
\]

obtained by substituting (3.4) into (3.6). The vector \(\tilde{N}_{M,n}\) is yet to be determined. For this, the aforementioned \(k_{eff}\) constraints and the characteristics of actinide taken from the LWR-spent fuel are used. The value of \(k_{eff}\) of the reactor at the beginning of each irradiation period is set to be 0.98. Then, at the beginning of the \((n+1)\)-st cycle, (2.13) must satisfy

\[
\sum_{i} \eta_{i} N_{i,n+1}^{o} (\sigma_{i,a} - 2\sigma_{i,2a}) + N_{F,n+1}^{o} (\sigma_{F,a} - 2\sigma_{F,2a}) + N_{Zr,n+1}^{o} (\sigma_{Zr,a} - 2\sigma_{Zr,2a}) + \sum_{p} N_{p} (\sigma_{p,a} - 2\sigma_{p,2a}) = 0.98
\]

The left side expresses the \(k_{eff}\) at the beginning of the \((n+1)\)-st cycle. By (3.13) and (3.11), the number of atoms of an actinide \(i\) in the reactor at the \((n+1)\)-st cycle is written as

\[
N_{i,n+1}^{o} = (1-f)N_{i,n} (T_{n}^{o} + T_{n}) + fR_{e}N_{i,n} (T_{n}^{o} + T_{n}) + x_{i}N_{tot,M,n}.
\]

The number of atoms of fission-product pairs in the reactor at the \((n+1)\)-st cycle is written as

\[
N_{F,n+1}^{o} = (1-f)N_{F,n} (T_{n}^{o} + T_{n}).
\]

Using (3.10) and (3.12), the number of atoms of zirconium in the reactor at the beginning of the \((n+1)\)-st cycle is given by

\[
N_{Zr,n+1}^{o} = (1-f)N_{Zr,n} (T_{n}^{o} + T_{n}) + N_{Zr,M,n}
\]

\[
= (1-f)N_{Zr,n} (T_{n}^{o} + T_{n}) + \rho_{Zr} V_{M,n} - \rho_{Zr} N_{tot,M,n} \sum_{i} \frac{X_{i}}{\rho_{i}}.
\]

Substituting (3.15), (3.16), and (3.17) into (3.14) yields an equation for \(N_{tot,M,n}\). The value of \(N_{tot,M,n}\) is obtained by solving that equation. By (3.10) and (3.11), the volume fraction, \(\omega\), of actinides in the makeup material at the \(n\)-th cycle and the number, \(N_{i,M,n}\), of atoms of an actinide nuclide \(i\) can be determined. Once \(N_{i,M,n}\) is known, \(\tilde{N}_{M,n}\) is determined also. The number, \(N_{Zr,M,n}\), of atoms of zirconium in the makeup material at the \(n\)-th cycle is obtained by (3.12).
3.3 Flow Chart for Code Programming

Figure 3.1 Flow chart for a code, WACOM
To carry out ATW fuel cycle simulation on a computer, a computational procedure based on Chapters 2 and 3 was developed and implemented in the code called WACOM. Figure 3.1 shows a flow chart for WACOM code. This algorithm starts with fuel burnup calculation by reading input data such as material properties and neutronics of a transmuter core. Neutronics of a transmuter core can be obtained by running a neutron transport code such as MCNP [RSICC Code Package CCC-700, 2000]. As the numerical solution for fuel burnup calculation, a predictor-corrector method is implemented. After fuel burnup calculation for a time step, the multiplication factor, $k_{\text{eff}}$, is estimated at the end of the time step. Then, the estimated $k_{\text{eff}}$ is compared to the lower bound of its value, here 0.92. If the estimated $k_{\text{eff}}$ is greater than 0.92, fuel burnup calculation proceeds for the next time step until it satisfies the condition, $k_{\text{eff}} \leq 0.92$. When $k_{\text{eff}}$ satisfies the condition, burnup calculation stops and the composition vector of discharged fuel is determined from the composition vector of the irradiated fuel multiplied by the discharge fraction ($f$). Next, by operating the recovery yield matrix ($R$) on the composition vector of the discharged fuel, the composition vector of the recovered fuel is obtained and the composition vector of waste is subsequently determined. The mass and the composition of makeup material are determined such that $k_{\text{eff}}$ of a newly charged transmuter core at BOC gives the upper bound of $k_{\text{eff}}$, here 0.98. The summation of the composition vectors of the recovered and the makeup materials results in the composition vector of a recharged fuel. By determining the composition vector of the newly charged core (2nd BOC fuel in Figure 3.1) the calculation of the first cycle ends. For multi-cycles, the whole procedure is repeated.
4 Computation for a Single Cycle Case

4.1 Reference LBE-Cooled Transmuter

In this chapter, to carry out the analyses of the ATW system using WACOM, a reference transmuter design is selected.

![Diagram of Proposed LBE-Cooled blanker configuration (192 fuel assemblies)]

As a transmuter of ATW system, several types are under considerations: sodium-cooled (Na), molten salt (MS), lead bismuth eutectic (LBE), gas-cooled, etc. Among those, LBE-cooled fast-spectrum transmuter is a transmuter concept that received extensive review in the ATW roadmap due to following features [Hill, et al., 1999]:

- Target (1)
- Buffer (18)
- Low TRU driver (42)
- Medium TRU driver (48)
- High TRU driver (102)
- Reflector (114)
- Shield (66)
- Lead and bismuth are excellent spallation target materials, with spallation neutron production rate on par with the best sub-actinide elements.
- The eutectic has a relatively low melting point (125 °C) offering the option of operating the system at low temperatures. Further, its high boiling point (1670 °C) offers a margin of safety.
- LBE is not reactive with water, so direct heat exchange with water is straightforward.
- The high atomic masses of lead and bismuth preserve a very hard neutron spectrum, allowing for large coolant channels in the transmuter.
- Both lead and bismuth have low neutron absorption cross sections, offering good neutron economy.

Several LBE-cooled transmuter designs were assessed as an effort to achieve efficient transmutation of the TRU separated from LWR-spent fuel and an optimized transmuter design was proposed based on nuclear transmutation performance [AAA-RPO-SYS-01-0008, 2001]. The blanket of the proposed LBE-cooled transmuter is shown in Figure 4.1. It consists of 19 hexagonal lattice positions containing the lead-bismuth eutectic (LBE) target/buffer and 192 fuel assemblies. The blanket is surrounded by two hexagonal rows of reflector assemblies and one row of B₄C shield assemblies. The principal design parameters of the proposed design are summarized in Table 4.1. A semi-annual 6-batch fuel management scheme is employed with the capacity factor of 75%.

### Table 4.1 Design Parameters for the Proposed LBE-Cooled Blanket Point Design [AAA-RPO-SYS-01-0008, 2001]

<table>
<thead>
<tr>
<th>Parameter (at operating temperature)</th>
<th>LBE target/buffer</th>
<th>Fuel</th>
<th>Structure</th>
<th>Coolant</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton Energy (GeV)</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Target Material</td>
<td>LBE</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel Material</td>
<td>(TRU-10Zr)-Zr</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pin Diameter (cm)</td>
<td>0.635</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cladding Thickness (cm)</td>
<td>0.056</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pitch-to-Diameter Ratio</td>
<td>1.727</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of Pins per Assembly</td>
<td>217</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel Smear Density (%)</td>
<td>75</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Volume Fraction</td>
<td>Fuel</td>
<td>0.140</td>
<td>Structure</td>
<td>0.103</td>
</tr>
<tr>
<td>Hexagonal Assembly Pitch (cm)</td>
<td>16.142</td>
<td></td>
<td>Coolant</td>
<td>0.695</td>
</tr>
<tr>
<td>Number of Assemblies</td>
<td>LBE target/buffer</td>
<td>19</td>
<td>Fuel</td>
<td>192</td>
</tr>
<tr>
<td></td>
<td>LBE Reflector</td>
<td>114</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Shield</td>
<td>66</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRU Fraction Split Factor</td>
<td>1.45/1.28/1.00</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Active Fuel Height (cm)</td>
<td>106.68</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Equivalent Fuel Region Diameter (cm)</td>
<td>246.21</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Maximum Blanket Diameter (cm)</td>
<td>357.07</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of Fuel Batches</td>
<td>Inner zone</td>
<td>5</td>
<td>Middle and Outer Zones</td>
<td>6</td>
</tr>
</tbody>
</table>

23
4.2 Neutronics of Start-up Core of the LBE-Cooled Transmuter

For ATW system analysis, WACOM requires neutronics of the ATW transmuter as part of input data and to obtain neutronics of the transmuter MCNP [RSICC Code Package CCC-700, 2000] code is used in this study.

Figure 4.2 shows the simplified LBE-cooled point design geometry used for the MCNP calculation. It is assumed that the reactor has a cylindrical geometry with three regions: the target region, the fuel region, and the reflector region. The inner region is a spallation target with a radius of 36.9 cm, the middle region is the homogenized fuel region with a radius of 123.1 cm, and the outer region is the reflector region with a radius of 152.8 cm. The shield is not considered for simplicity. The active fuel height is 106.7 cm and the height of the reactor is assumed to be 500 cm. The homogenized fuel region is composed of fuel, coolant, and structure material and their volume fractions are assumed to be 14.9%, 74.1%, and 11.0% instead of 14.0%, 69.5%, and 10.3% respectively of Table 4.1 to make the total homogenized volume fraction be unity.

Figure 4.2 Simplified LBE-Cooled point design geometry used for the MCNP simulation

The properties and composition of the TRU taken from PWR-spent fuel and the properties of zirconium in the homogenized fuel region are given in Table 4.2. These data are used as part of input for both MCNP and WACOM.

The proposed LBE-cooled reactor has a lead-bismuth eutectic (LBE) as its coolant. The composition and properties of LBE-coolant are shown in Table 4.3. The density of the LBE-coolant is assumed 10.151 g/cm$^3$ at the temperature of 475 °C.

Table 4.4 shows the properties of HT-9, the structural material assumed for the proposed LBE-transmuter. The density of HT-9 is assumed to be 7.66 g/cm$^3$ at 475 °C.
### Table 4.2  Fuel Isotope Properties [KAERI, 2000] and PWR-TRU Composition

<table>
<thead>
<tr>
<th>i</th>
<th>Isotope</th>
<th>Atomic mass [amu]</th>
<th>Nominal density [g/cm$^3$]</th>
<th>Isotopic density [g/cm$^3$]</th>
<th>$t_{1/2}$ [s]</th>
<th>ALI$^4$ for ingestion [Ci/l]</th>
<th>Weight fraction in TRU$^5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>U234</td>
<td>234.041</td>
<td></td>
<td>18.631</td>
<td>3E-10</td>
<td>0.0000</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>U235</td>
<td>235.044</td>
<td></td>
<td>18.811</td>
<td>3E-10</td>
<td>4.0000E-05</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>U236</td>
<td>236.046</td>
<td>18.95</td>
<td>18.891</td>
<td>3E-10</td>
<td>2.0000E-05</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>U238</td>
<td>238.051</td>
<td></td>
<td>19.052</td>
<td>3E-10</td>
<td>4.7800E-03</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>Np237</td>
<td>237.048</td>
<td>20.25</td>
<td>20.450</td>
<td>2E-11</td>
<td>5.0230E-02</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Pu238</td>
<td>238.050</td>
<td></td>
<td>19.332</td>
<td>2E-11</td>
<td>1.2720E-02</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Pu239</td>
<td>239.052</td>
<td></td>
<td>19.413</td>
<td>2E-11</td>
<td>5.3200E-01</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>Pu240</td>
<td>240.054</td>
<td>19.84</td>
<td>19.494</td>
<td>2E-11</td>
<td>2.1534E-01</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>Pu241</td>
<td>241.057</td>
<td></td>
<td>19.576</td>
<td>1E-09</td>
<td>3.7800E-02</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>Pu242</td>
<td>242.059</td>
<td></td>
<td>19.657</td>
<td>2E-11</td>
<td>4.6850E-02</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>Cm242</td>
<td>242.059</td>
<td>13.236</td>
<td>1.4083E+07</td>
<td>7E-10</td>
<td>0.0000</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>Cm243</td>
<td>243.061</td>
<td>13.291</td>
<td>9.4608E+08</td>
<td>3E-11</td>
<td>2.0000E-05</td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>Cm244</td>
<td>244.063</td>
<td>13.67</td>
<td>13.346</td>
<td>3E-11</td>
<td>1.0400E-03</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>Cm245</td>
<td>245.066</td>
<td></td>
<td>13.400</td>
<td>2E-11</td>
<td>9.0000E-05</td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>Cm246</td>
<td>246.067</td>
<td></td>
<td>13.455</td>
<td>2E-11</td>
<td>1.0000E-05</td>
<td></td>
</tr>
<tr>
<td>Zr</td>
<td></td>
<td>91.22</td>
<td>6.49</td>
<td>6.49</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table 4.3  Composition and Properties of LBE-Coolant [Spencer, 2000; Liquid Material Handbook, 1952]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Nominal density [g/cm$^3$]</th>
<th>Weight fraction in coolant</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>11.34</td>
<td>4.45E-01</td>
</tr>
<tr>
<td>Bi-209</td>
<td>9.80</td>
<td>5.55E-01</td>
</tr>
</tbody>
</table>

The fuel composition at the beginning of the each cycle is determined such that the beginning of cycle (BOC) $k_{eff}$ of the homogenized reactor core is 0.98. The cycle

4 “Appendix B-Annual limits on intake (ALIs) and derived air concentrations of radionuclides for occupational exposure; effluent concentrations; concentration for release to sewerage,” Title 10, Part 20, Code of Federal Regulations (1999)

5 Corresponding to a 25 year cooled PWR-TRU assuming that 99.995% of the uranium is removed in the UREX process
proceeds until \( k_{\text{eff}} \) drops to 0.92. The value of the fuel composition at BOC is controlled by adjusting the ratio of TRU to zirconium.

Table 4.4 Composition and Properties of Structure Material HT-9 [IAEA-TECDOC-1039, 1997]

<table>
<thead>
<tr>
<th>Element</th>
<th>Nominal density [g/cm(^3)]</th>
<th>Weight fraction in structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>7.87</td>
<td>8.5E-01</td>
</tr>
<tr>
<td>Cr</td>
<td>7.19</td>
<td>1.15E-01</td>
</tr>
<tr>
<td>Ni</td>
<td>8.90</td>
<td>5.0E-03</td>
</tr>
<tr>
<td>Mo</td>
<td>10.20</td>
<td>1.0E-02</td>
</tr>
<tr>
<td>Mn</td>
<td>7.43</td>
<td>6.0E-03</td>
</tr>
<tr>
<td>C</td>
<td>1.60</td>
<td>2.0E-03</td>
</tr>
<tr>
<td>Si</td>
<td>2.33</td>
<td>4.0E-03</td>
</tr>
<tr>
<td>W</td>
<td>19.20</td>
<td>5.0E-03</td>
</tr>
<tr>
<td>V</td>
<td>6.10</td>
<td>3.0E-03</td>
</tr>
</tbody>
</table>

Figure 4.3 Variation of \( k_{\text{eff}} \) with TRU weight fraction in start-up core fuel

To find out the ratio of TRU to zirconium which gives \( k_{\text{eff}} \) of 0.98 for the start-up core, we investigated \( k_{\text{eff}} \) for several fuel compositions with different ratios of TRU to zirconium by MCNP. From MCNP calculations, it is observed that \( k_{\text{eff}} \) varies almost linearly with the fraction of TRU in fuel and thus two sets of MCNP calculations give an estimation of the start-up core fuel composition. Figure 4.3 shows how two sets of MCNP calculations enable us to get an estimate of the start-up fuel composition which satisfies the \( k_{\text{eff}} \) constraint. Interpolating between two \( k_{\text{eff}} \) values corresponding to 30 wt% TRU
and 45 wt% TRU, it is found that the start-up core satisfying the $k_{\text{eff}}$ constraint has 31.8 wt% TRU and 68.2 wt% zirconium.

Table 4.5  Neutronics of the Start-up Core Calculated by MCNP (TRU-31.8 wt%, Zr-68.2 wt%)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(n,γ) [b]</th>
<th>(n,2n) [b]</th>
<th>(n,f) [b]</th>
<th>Average fission energy [MeV]</th>
<th>Average neutrons produced per fission</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>6.926E-01</td>
<td>5.980E-05</td>
<td>3.362E-01</td>
<td>201.7</td>
<td>2.405</td>
</tr>
<tr>
<td>U235</td>
<td>5.428E-01</td>
<td>4.337E-04</td>
<td>1.928E+00</td>
<td>201.7</td>
<td>2.471</td>
</tr>
<tr>
<td>U236</td>
<td>4.723E-01</td>
<td>3.065E-04</td>
<td>8.383E-02</td>
<td>201.7</td>
<td>2.370</td>
</tr>
<tr>
<td>U238</td>
<td>3.709E-01</td>
<td>5.409E-04</td>
<td>2.748E-02</td>
<td>201.7</td>
<td>2.523</td>
</tr>
<tr>
<td>Np237</td>
<td>1.597E+00</td>
<td>1.213E-04</td>
<td>3.189E-01</td>
<td>206.1</td>
<td>2.697</td>
</tr>
<tr>
<td>Pu238</td>
<td>7.624E-01</td>
<td>1.706E-04</td>
<td>1.145E+00</td>
<td>210.2</td>
<td>2.955</td>
</tr>
<tr>
<td>Pu239</td>
<td>5.213E-01</td>
<td>2.309E-04</td>
<td>1.827E+00</td>
<td>210.6</td>
<td>2.944</td>
</tr>
<tr>
<td>Pu240</td>
<td>5.314E-01</td>
<td>9.162E-05</td>
<td>3.189E-01</td>
<td>211.0</td>
<td>2.865</td>
</tr>
<tr>
<td>Pu241</td>
<td>4.565E-01</td>
<td>7.841E-04</td>
<td>2.566E+00</td>
<td>211.3</td>
<td>2.973</td>
</tr>
<tr>
<td>Pu242</td>
<td>4.741E-01</td>
<td>2.408E-04</td>
<td>2.570E-01</td>
<td>211.7</td>
<td>2.874</td>
</tr>
<tr>
<td>Am241</td>
<td>1.731E+00</td>
<td>2.627E-05</td>
<td>2.451E-01</td>
<td>215.2</td>
<td>3.294</td>
</tr>
<tr>
<td>Am242m</td>
<td>4.254E-01</td>
<td>4.409E-04</td>
<td>4.166E+00</td>
<td>215.5</td>
<td>3.334</td>
</tr>
<tr>
<td>Am243</td>
<td>1.536E+00</td>
<td>2.981E-05</td>
<td>1.906E-01</td>
<td>215.9</td>
<td>3.346</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.558E-01</td>
<td>6.844E-06</td>
<td>1.279E-01</td>
<td>219.4</td>
<td>3.510</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.718E-01</td>
<td>4.210E-04</td>
<td>2.834E+00</td>
<td>219.8</td>
<td>3.503</td>
</tr>
<tr>
<td>Cm244</td>
<td>8.806E-01</td>
<td>1.434E-04</td>
<td>4.223E-01</td>
<td>220.2</td>
<td>3.535</td>
</tr>
<tr>
<td>Cm245</td>
<td>3.183E-01</td>
<td>7.587E-04</td>
<td>2.508E+00</td>
<td>220.5</td>
<td>3.684</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.509E-01</td>
<td>1.916E-04</td>
<td>2.454E-01</td>
<td>220.9</td>
<td>3.560</td>
</tr>
<tr>
<td>Zr</td>
<td>2.887E-02</td>
<td>6.348E-05</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The neutronics of the start-up core as calculated by MCNP are listed in Table 4.5. The effective one-group cross sections for three types of neutron reactions, (n,γ), (n,f), and (n,2n), average fission energy released from the fission of an isotope, and average neutrons generated from the fission of an isotope are shown.

Input data for the MCNP calculation are listed in Appendix B.

4.3 Fuel Inventory Evolution in the LBE-Transmuter for a Single Cycle

Using the cross section data listed in Table 4.5, fuel inventory evolution in the reference LBE-cooled transmuter for the start-up cycle was calculated. Assumptions used for this calculation include the following: (1) The reactor power is 840 MWt. (2) The irradiation time is determined by the constraint 0.92 ≤ $k_{\text{eff}}$ ≤ 0.98. (3) The cross section values used are constant during the irradiation. (4) The mass of zirconium contained in fuel is not affected by neutron irradiation.

Table 4.6 shows that fractional mass changes of TRU, fission product, and zirconium for the start-up cycle. It is observed that the mass fraction of TRU in the core decreases from 31.8% at the beginning of the start-up cycle to 29.6% at the end of the start-up cycle. This could be interpreted that 6.9% TRU was transmuted into fission products during the
irradiation period. The irradiation period that takes for the value of $k_{eff}$ to reach to 0.92 was $\sim$108 days for the start-up cycle.

Table 4.6 Fractional Mass Changes of TRU, Fission Product, and Zirconium for the Start-up Cycle Calculated by WACOM Using the Neutronics of the Start-up Core

<table>
<thead>
<tr>
<th></th>
<th>TRU</th>
<th>Fission Products</th>
<th>Zirconium</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC (t=0)</td>
<td>31.8%</td>
<td>0</td>
<td>68.2%</td>
</tr>
<tr>
<td>EOC (t=108days)</td>
<td>29.6%</td>
<td>2.2%</td>
<td>68.2%</td>
</tr>
</tbody>
</table>

The isotopic composition change of TRU for the start-up cycle is listed in Table 4.7 in terms of number of atoms. In the last column of Table 4.7, positive or negative sign represents an increase or a decrease in the amount of an isotope during the cycle, respectively. Some isotopes such as $^{237}$Np, $^{239}$Pu, and $^{241}$Am show a relatively large reduction in their amount while others such as $^{234}$U, $^{236}$U, $^{238}$Pu, and $^{242m}$Am, and all Cm-isotopes show a significant increase in a cycle by neutron absorption and decay.

Table 4.7 Isotopic Composition Change of TRU for the Start-up Cycle Calculated by WACOM Using the Neutronics of the Start-up Core

<table>
<thead>
<tr>
<th>Isotope</th>
<th>BOC (t=0)</th>
<th>EOC (t=108days)</th>
<th>Fractional change</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>1.0000E+1</td>
<td>1.1238E+23</td>
<td>+1.124E+23</td>
</tr>
<tr>
<td>U235</td>
<td>1.3776E+23</td>
<td>1.3445E+23</td>
<td>-2.403E-02</td>
</tr>
<tr>
<td>U236</td>
<td>6.8586E+22</td>
<td>9.3094E+22</td>
<td>+3.573E-01</td>
</tr>
<tr>
<td>U238</td>
<td>1.6254E+25</td>
<td>1.5875E+25</td>
<td>-2.332E-02</td>
</tr>
<tr>
<td>Np237</td>
<td>1.7152E+26</td>
<td>1.5327E+26</td>
<td>-1.064E-01</td>
</tr>
<tr>
<td>Pu238</td>
<td>4.3253E+25</td>
<td>5.6684E+25</td>
<td>+3.105E-01</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.8013E+27</td>
<td>1.5699E+27</td>
<td>-1.285E-01</td>
</tr>
<tr>
<td>Pu240</td>
<td>7.2614E+26</td>
<td>7.3885E+26</td>
<td>+1.750E-02</td>
</tr>
<tr>
<td>Pu241</td>
<td>1.2700E+26</td>
<td>1.2553E+26</td>
<td>-1.157E-02</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.5670E+26</td>
<td>1.5735E+26</td>
<td>+4.148E-03</td>
</tr>
<tr>
<td>Am241</td>
<td>3.0111E+26</td>
<td>2.6959E+26</td>
<td>-1.047E-01</td>
</tr>
<tr>
<td>Am242m</td>
<td>4.6818E+23</td>
<td>5.4560E+24</td>
<td>+1.065E+01</td>
</tr>
<tr>
<td>Am243</td>
<td>3.0839E+25</td>
<td>3.2107E+25</td>
<td>+4.112E-02</td>
</tr>
<tr>
<td>Cm242</td>
<td>1.0000E+1</td>
<td>1.5294E+25</td>
<td>+1.529E+25</td>
</tr>
<tr>
<td>Cm243</td>
<td>6.6607E+22</td>
<td>2.2101E+23</td>
<td>+2.318E+00</td>
</tr>
<tr>
<td>Cm244</td>
<td>3.4493E+24</td>
<td>5.8992E+24</td>
<td>+7.103E-01</td>
</tr>
<tr>
<td>Cm245</td>
<td>2.9728E+23</td>
<td>4.7786E+23</td>
<td>+6.074E-01</td>
</tr>
<tr>
<td>Cm246</td>
<td>3.2896E+22</td>
<td>3.8996E+22</td>
<td>+1.854E-01</td>
</tr>
</tbody>
</table>

4.4 Benchmarking of WACOM against MOCUP for a Single Cycle

To check the isotope depletion module of the developed model, WACOM, it was compared to the depletion module of MOCUP [Moore, et al., 1995], a code package that couples MCNP for particle transport calculations and ORIGEN 2.1 for isotope depletion calculations. The proposed LBE-cooled transmuter point design described in Section 4.1 has been used for benchmarking.
Table 4.8 compares the fractional changes of the fuel isotopes in the reactor, which are calculated with WACOM and MOCUP using the neutronics of the start-up core. The fractional change is obtained by dividing the number of atoms of each isotope at the end of irradiation by its number of atoms at the beginning of irradiation. A very good agreement is found between WACOM and MOCUP.

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>MOCUP</th>
<th>WACOM</th>
<th>Relative difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>2.4779E+23</td>
<td>2.4782E+23</td>
<td>0.01%</td>
</tr>
<tr>
<td>U235</td>
<td>1.1303E+00</td>
<td>1.1304E+00</td>
<td>0.01%</td>
</tr>
<tr>
<td>U236</td>
<td>2.1522E+00</td>
<td>2.1447E+00</td>
<td>0.35%</td>
</tr>
<tr>
<td>U238</td>
<td>9.5655E-01</td>
<td>9.5647E-01</td>
<td>0.01%</td>
</tr>
<tr>
<td>Np237</td>
<td>7.9779E-01</td>
<td>7.9777E-01</td>
<td>0.00%</td>
</tr>
<tr>
<td>Pu238</td>
<td>1.7643E+00</td>
<td>1.7699E+00</td>
<td>0.32%</td>
</tr>
<tr>
<td>Pu239</td>
<td>7.4899E-01</td>
<td>7.4900E-01</td>
<td>0.00%</td>
</tr>
<tr>
<td>Pu240</td>
<td>1.0006E+00</td>
<td>1.0005E+00</td>
<td>0.01%</td>
</tr>
<tr>
<td>Pu241</td>
<td>9.3182E-01</td>
<td>9.2832E-01</td>
<td>0.38%</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.0033E+00</td>
<td>1.0033E+00</td>
<td>0.00%</td>
</tr>
<tr>
<td>Am241</td>
<td>7.7279E-01</td>
<td>7.7430E-01</td>
<td>0.20%</td>
</tr>
<tr>
<td>Am242m</td>
<td>2.0488E+01</td>
<td>2.0501E+01</td>
<td>0.06%</td>
</tr>
<tr>
<td>Am243</td>
<td>9.5276E-01</td>
<td>9.5288E-01</td>
<td>0.01%</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.3372E+25</td>
<td>3.3481E+25</td>
<td>0.33%</td>
</tr>
<tr>
<td>Cm243</td>
<td>6.9254E+00</td>
<td>6.9373E+00</td>
<td>0.17%</td>
</tr>
<tr>
<td>Cm244</td>
<td>3.1975E+00</td>
<td>3.1994E+00</td>
<td>0.06%</td>
</tr>
<tr>
<td>Cm245</td>
<td>2.8720E+00</td>
<td>2.8717E+00</td>
<td>0.01%</td>
</tr>
<tr>
<td>Cm246</td>
<td>1.5125E+00</td>
<td>1.5120E+00</td>
<td>0.03%</td>
</tr>
</tbody>
</table>

To better understand this comparison, however, it is helpful to know the characteristics of MOCUP and differences between MOCUP and WACOM.

MOCUP is a series of utility and data manipulation programs to solve time- and space-dependent coupled neutronics/isotopics problems. The neutronics calculation is performed by the Los Alamos National Laboratory code system, MCNP version 4a or later (CCC-200 or CCC-660), and the depletion and isotopics calculation are performed by CCC-371/ORIGEN2.1 developed at Oak Ridge National Laboratory. MOCUP consists of three utility programs (mcnpPRO, origenPRO, compPRO) to, respectively, search the MCNP output and tally files for relevant cell and tally parameters, prepare ORIGEN2.1 input files and execute the ORIGEN2.1 runs and search ORIGEN2.1 punch files for relevant isotope concentrations and produce new MCNP input files.

MCNP Post-processor (mcnpPRO) provides the cell fluxes and the radiative capture, alpha production, fission, (n,2n), and (n,3n) cross sections for the ORIGEN2.1 calculation through the MOCUP intermediate file, which is referred to as the MCNP processor output file. The MCNP fluxes are adjusted by the flux multiplier table. The ORIGEN2.1 Pre-processor (origenPRO) takes the flux and cross section information from the MCNP processor output files and merges it into the skeletal ORIGEN2.1 input files to create
modified ORIGEN2.1 input files for each MCNP cell to be depleted. ORIGEN2.1 Preprocessor (origenPRO) can also execute ORIGEN2.1 for each input file and prepare the newly generated composition data for the next time sequence.

While the reaction types which MCNP Post-processor recognizes are limited to \((n,\gamma)\) reaction, \((n,2n)\) reaction, \((n,3n)\) reaction, fission reaction, and alpha production, the skeletal file for ORIGEN2.1 requires a complete cross section/yield specification for each time-dependent isotope. \((n,\gamma)^6\) and \((n,2n)^7\) cross section data are not provided by MCNP and thus those are copied from the ORIGEN2.1 cross section library.

However, WACOM, the model developed here, is supposed to use MCNP-generated cross section data and does not have any cross section library like ORIGEN2.1 cross section libraries. Therefore, in order to compare this model with MOCUP using the similar input data, \((n,\gamma)^6\) and \((n,2n)^7\) cross section data of ORIGEN2.1 cross section library are manually added to the effective one group \((n,\gamma)\) and \((n,2n)\) cross sections generated by MCNP, respectively. The branching ratio of \((n,\gamma)\) and \((n,\gamma^*)\) cross section and that of \((n,2n)\) and \(2n^*\) are used to simulate the neutron-induced reaction of an isotope which has both ground state cross section and excitation cross section. For example, letting \(\delta\) as the ratio of \((n,\gamma)\) cross section to the combined cross section of \((n,\gamma)\) and \((n,\gamma^*)\) cross section, \((1-\delta)\) represents the ratio of \((n,\gamma^*)\) cross section to the combined cross section of \((n,\gamma)\) and \((n,\gamma^*)\) cross section. \(^{241}\)Am and \(^{243}\)Am have both \((n,\gamma)\) and \((n,\gamma^*)\) cross sections. \(^{241}\)Am leads to \(^{242}\)Am and \(^{242m}\)Am by \((n,\gamma)\) and \((n,\gamma^*)\) reactions, respectively. \(^{243}\)Am leads to \(^{244}\)Am and \(^{244m}\)Am by \((n,\gamma)\) and \((n,\gamma^*)\) reaction, respectively.

The effective one group cross-sections of the LBE-cooled transmuter have been calculated with MCNP for its start-up core that has a 31.8 weight % of TRU and a 68.2 weight % of zirconium. Table 4.5 shows the obtained effective one-group cross sections. For the fission product (FP), a pseudo cross section has been estimated under the assumption that the estimated neutron absorption rate by fission products in MOCUP is equal to the neutron absorption rate by fission products calculated from this model.

To estimate the effective cross sections of isotopes in homogenized fuel, target, and reflector regions, several cross section data sources such as ENDF/B-IV [BNL, 1974] and ENDF/B-V [BNL, 1979] were used by MCNP. The operating temperatures of the proposed LBE-cooled reactor are assumed to be \(-1000\ \text{°K}\) in fuel and target, \(-750\ \text{°K}\) in structure and \(-700\ \text{°K}\) in reflector. Due to the unavailability of cross section data sources at operating temperature levels, the effective cross sections of some isotopes such as Np-isotopes, Am-isotopes, Cm-isotopes, HT-9 constituents, and LBE constituents were estimated at the room temperature rather than their operating temperatures. Thus, the estimated effective cross sections of those materials could be deviated to some extent from those at their operating temperatures.

In addition to the unavailability of excitation cross section and insufficient cross section libraries at reactor operating temperatures in MCNP, the cross section data source of \(^{242}\)Am is not available in MCNP cross section libraries. However, ORIGEN2.1 has the cross section data of it in its own cross section library and uses them. From the facts described so far, it is obvious that MOCUP uses two different kinds of cross section data.

---

\(^6\) \((n,\gamma)\) reaction that results in a meta-stable nuclide.

\(^7\) \((n,2n)\) reaction that results in a meta-stable nuclide.
one provided by MCNP and the other from the cross section library of ORIGEN2.1 to calculate isotope depletion and generation. Except for benchmarking purpose, WACOM uses MCNP-generated cross section and does not consider excitation cross sections. This could make significant difference in the concentrations of americium and curium isotopes when comparing to the MOCUP calculation or other tools taking into account excitation reactions in their isotope depletion module. The cross section data sets in Table 4.5 were used for the benchmarking against MOCUP but the capture cross sections of $^{241}$Am and $^{243}$Am have been modified such that they include both ground state cross section and excitation cross section. The modified capture cross section of $^{241}$Am is 1.7279 [b] and that of $^{243}$Am is 2.1780 [b].

4.5 Summary and Conclusions

As a subcritical transmuter for the ATW system, an optimized LBE-cooled transmuter was selected. With MCNP code, the composition and the neutronics of the startup core were evaluated and the neutronics was provided as an input of WACOM.

For a single cycle, transmutation performance of the LBE-cooled transmuter was estimated by WACOM and the results showed that the cycle duration for the startup cycle is 108 days, during this period 6.9 wt% of the startup-core TRU is converted into fission products, and fissioned-TRU isotopes are mainly $^{237}$Np, $^{239}$Pu, and $^{241}$Am.

Benchmarking WACOM against MOCUP for a-year irradiation time showed that the composition of TRU calculated by WACOM at the end of irradiation is in good agreement with that obtained by MOCUP. This indicates that the nuclide chain model and the numerical solution implemented for burnup calculation in WACOM are well functional.

However, the neutronics that was used as input data in WACOM varies with fuel composition evolution in an actual transmuter core. Thus, before applying the newly developed tool to actual analysis of ATW systems it is necessary to work out an algorithm that will adjust the effective one-group cross sections to the variation in the TRU loading and composition.
5 Application of WACOM for Multi Cycle Calculation

5.1 Introduction

A mathematical model and computer code, WACOM, for the analysis of ATW fuel cycle was developed and benchmarked by MOCUP.

The purpose of this chapter is to develop a computational scheme for taking into account the neutronics variation for multi-cycle analyses of ATW system. The conventional approach in fuel cycle analysis is to re-calculate the reactor spectrum and effective one group cross sections at least once and often several times in one cycle, i.e., between refuelings. For multi-cycle analyses this approach requires significant effort and computational time, especially when using a Monte-Carlo based code, such as MCNP, for the neutronics calculations.

An alternative approach using interpolation of pre-prepared effective one-group cross sections is investigated in this work. Using the interpolation method developed, the effect of difference in reported values of the branching ratio of $^{241}$Am on the results of multi-cycle analyses is also investigated.

5.2 Accounting for Neutronics Variation

5.2.1 Neutron Spectrum Variation

The fuel inventory evolution in the LBE-cooled transmuter was calculated for 100 cycles by using the constant neutronics for the startup core, and is plotted in Figure 5.1. TRU and FP in fuel show a zigzag pattern. For TRU, a peak and a valley are corresponding to the beginning of a cycle (BOC) and the end of a cycle (EOC), respectively. For FP, a valley is for BOC and a peak is for EOC.

![Figure 5.1 Fuel composition evolution with cycle number, calculated by using the effective one-group cross sections of the start-up core](image-url)
In order to check if the fuel composition evolution calculated by WACOM satisfies the $k_{\text{eff}}$ constraint well enough, the $k_{\text{eff}}$ values of several BOC- and EOC-fuel compositions have been estimated by MCNP. Significant discrepancies were found between the $k_{\text{eff}}$ constraint and estimated $k_{\text{eff}}$ values. For example, the $k_{\text{eff}}$ value of the 100th BOC fuel is estimated to be 1.022 by MCNP, which should be 0.98. Because the nuclide chain model and the numerical solution of WACOM were validated by MOCUP, the reason for the discrepancies is likely due to the use of constant neutronics in WACOM calculation.

The effect of neutron spectrum variation in the transmuter core is investigated by considering the BOC-fuel compositions of several of the cycles plotted in Figure 5.1. Table 5.1 shows the effective one-group $(n,\gamma)$ cross sections for the selected cycles estimated by MCNP. The variation pattern of the $(n,\gamma)$ cross sections of the uranium isotopes are plotted in Figure 5.2 with respect to the mass fraction of TRU in the fuel. It is noticed that the $(n,\gamma)$ cross sections of the uranium isotopes vary almost linearly with the mass fraction of the TRU in the fuel. A similar behavior is observed for the other isotopes and also for the $(n,f)$ cross sections. It is concluded that it is sufficient to calculate with MCNP one set of effective one-group cross sections for two different fuel loadings. The effective one-group cross sections for any composition can then be estimated by interpolation and extrapolation. The two fuel compositions chosen as the basis for the interpolation are the compositions at the beginning of the first cycle and at the beginning of the equilibrium cycle (here, 100th cycle) plotted in Figure 5.1.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>1st BOC (31.8wt% TRU)</th>
<th>10th BOC (37.2wt% TRU)</th>
<th>20th BOC (40.0wt% TRU)</th>
<th>100th BOC (43.0wt% TRU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>6.926E-1</td>
<td>5.748E-1</td>
<td>5.550E-1</td>
<td>5.240E-1</td>
</tr>
<tr>
<td>U235</td>
<td>5.428E-1</td>
<td>4.867E-1</td>
<td>4.767E-1</td>
<td>4.520E-1</td>
</tr>
<tr>
<td>U236</td>
<td>4.723E-1</td>
<td>4.026E-1</td>
<td>3.930E-1</td>
<td>3.730E-1</td>
</tr>
<tr>
<td>U238</td>
<td>3.709E-1</td>
<td>3.137E-1</td>
<td>3.056E-1</td>
<td>2.870E-1</td>
</tr>
<tr>
<td>Np237</td>
<td>1.597E+0</td>
<td>1.423E+0</td>
<td>1.390E+0</td>
<td>1.320E+0</td>
</tr>
<tr>
<td>Pu238</td>
<td>7.624E-1</td>
<td>6.780E-1</td>
<td>6.641E-1</td>
<td>6.300E-1</td>
</tr>
<tr>
<td>Pu239</td>
<td>5.213E-1</td>
<td>4.459E-1</td>
<td>4.313E-1</td>
<td>3.990E-1</td>
</tr>
<tr>
<td>Pu240</td>
<td>5.314E-1</td>
<td>4.609E-1</td>
<td>4.478E-1</td>
<td>4.230E-1</td>
</tr>
<tr>
<td>Pu241</td>
<td>4.565E-1</td>
<td>4.051E-1</td>
<td>3.948E-1</td>
<td>3.740E-1</td>
</tr>
<tr>
<td>Pu242</td>
<td>4.741E-1</td>
<td>4.114E-1</td>
<td>3.987E-1</td>
<td>3.740E-1</td>
</tr>
<tr>
<td>Am241</td>
<td>1.731E+0</td>
<td>1.540E+0</td>
<td>1.508E+0</td>
<td>1.430E+0</td>
</tr>
<tr>
<td>Am242m</td>
<td>4.254E-1</td>
<td>3.561E-1</td>
<td>3.427E-1</td>
<td>3.140E-1</td>
</tr>
<tr>
<td>Am243</td>
<td>1.536E+0</td>
<td>1.341E+0</td>
<td>1.308E+0</td>
<td>1.230E+0</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.558E-1</td>
<td>2.941E-1</td>
<td>2.806E-1</td>
<td>2.560E-1</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.718E-1</td>
<td>2.278E-1</td>
<td>2.198E-1</td>
<td>2.020E-1</td>
</tr>
<tr>
<td>Cm244</td>
<td>8.806E-1</td>
<td>7.824E-1</td>
<td>7.660E-1</td>
<td>7.240E-1</td>
</tr>
<tr>
<td>Cm245</td>
<td>3.183E-1</td>
<td>2.887E-1</td>
<td>2.838E-1</td>
<td>2.710E-1</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.509E-1</td>
<td>2.159E-1</td>
<td>2.074E-1</td>
<td>1.950E-1</td>
</tr>
<tr>
<td>Zr</td>
<td>2.887E-2</td>
<td>2.232E-2</td>
<td>2.206E-2</td>
<td>2.133E-2</td>
</tr>
<tr>
<td>FP</td>
<td>2.662E-1</td>
<td>2.604E-1</td>
<td>2.523E-1</td>
<td>2.364E-1</td>
</tr>
</tbody>
</table>
The results from these two MCNP calculations are used for developing two linear fits for each isotope; one for \((n,\gamma)\) and the other for \((n,f)\) cross sections, with respect to the mass fraction change of TRU in the fuel\(^8\). The \((n,2n)\) cross sections are assumed to be constant through all cycles due to their small contribution to the isotopic transmutation.

Figure 5.2 Variations of effective one-group \((n,\gamma)\) cross sections of U-isotopes with respect to the mass fraction of actinide in fuel

5.2.2 Revised Flow Chart of WACOM Code

Figure 5.3 shows the revised flow chart of WACOM code which takes into account the variation of effective one-group cross sections due to fuel inventory evolution in the transmuting reactor core. Linear interpolating scheme to account for the variation of effective one-group cross sections is implemented in the module of fuel burnup calculation and in the process of determining makeup fuel and BOC fuel composition.

For the fuel burnup calculation, the neutronics of the start-up core \((\sigma_i)\) provided as input data are used only for the first time step calculation and thereafter those are updated at the end of every time step based on TRU mass fraction calculated at that time step. Updated effective one-group cross sections are expressed as \(\sigma_f\). At the end of fuel burnup, this represents the neutronics of the irradiated core just before discharge occurs.

For makeup fuel calculation, while the version of WACOM explained in Chapter 3 uses the effective one-group cross sections of the start-up core \((\sigma_i)\), in the revised version, the averaged effective one-group cross sections \(\sigma^* = (\sigma_i + \sigma_f)/2\) are used. The required makeup material is determined such that \(k_{\text{eff}}\) of each BOC fuel is of 0.98. \(k_{\text{eff}}\) formula in (3.14) requires the effective one-group cross sections of each BOC fuel to determine the mass and the composition of makeup material in each cycle. However, at this time the effective one-group cross sections of the BOC fuel are unavailable yet because each BOC fuel composition is determined by makeup material. Thus, an iteration scheme is employed to obtain a converging neutronics of the BOC fuel with \(k_{\text{eff}}\) of 0.98. Using the

---

\(^8\) Neutronics used for linear fits and formulations implemented in WACOM code are listed in Appendix C.
averaged effective one-group cross sections ($\sigma^*$) to get the first estimate of required makeup material is a starting point of the iteration.

Figure 5.3 Revised flow chart of WACOM code
Charged fuel which consists of the first estimate of makeup and the recovered material from the separation process is combined with the remaining fuel in the core, and then the first estimate of BOC fuel composition for the next cycle is obtained. To check if the first estimate of BOC fuel is acceptable, its neutronics ($\sigma^{**}$) are evaluated by the implemented linear interpolating scheme and then compared with the neutronics ($\sigma^*$) used for determining makeup material. If both are same or within a prescribed relative difference (here, 0.1%), the first estimate of BOC fuel is considered acceptable, otherwise, iterations continue until both are within a prescribed relative difference. The next iteration starts with the averaged neutronics of $\sigma^*$ and $\sigma^{**}$ for determining the second estimate of makeup material.

Through a number of iterations, the final estimate of BOC fuel satisfying the prescribed criterion is determined and its neutronics is used for burnup calculation for the first time step of the next cycle.

5.2.3 Validation of Interpolating Scheme for Estimation of Neutronics

Figure 5.4 shows the fuel composition variation with cycle number calculated by revised WACOM with an interpolating scheme to account for neutronic variations. This figure shows profiles similar to those in Figure 5.1, but has a smaller concentration of actinide and a larger concentration of zirconium at the equilibrium cycles. The mass fraction of actinides increases from ~32% at start-up to ~41% rather than ~43% in Figure 5.1 at the beginning of the equilibrium cycle.

![Figure 5.4](image)

**Figure 5.4** Fuel composition variation with cycle number predicted by using interpolated neutronics

To check whether the fuel compositions calculated by WACOM (Figure 5.3) satisfy the $k_{eff}$ constraint, several fuel compositions at beginning of cycle and end of cycle are
chosen arbitrarily and their $k_{\text{eff}}$ and effective one-group cross section values are calculated by MCNP. Table 5.2 shows that the $k_{\text{eff}}$ values calculated by MCNP are within ~$0.5\%$ of the $k_{\text{eff}}$ constraints. This confirms that the profile plotted in Figure 5.4 represents adequately the cycle-dependent fuel composition variation of the ATW system.

**Table 5.2** $k_{\text{eff}}$-Values of 10th, 20th, and 100th Fuel Composition in Figure 5.3 Estimated by MCNP

<table>
<thead>
<tr>
<th>Cycle</th>
<th>10th cycle</th>
<th>20th cycle</th>
<th>100th cycle</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOC-$k_{\text{eff}}$</td>
<td>0.984 (±0.001)</td>
<td>0.982 (±0.001)</td>
<td>0.979 (±0.001)</td>
</tr>
<tr>
<td>EOC-$k_{\text{eff}}$</td>
<td>0.925 (±0.001)</td>
<td>0.925 (±0.001)</td>
<td>0.922 (±0.001)</td>
</tr>
</tbody>
</table>

**Table 5.3** Comparison of Interpolated Capture Cross Sections with MCNP-Estimated Capture Cross Sections for the BOC Fuel Composition at the 100th cycle

<table>
<thead>
<tr>
<th>Isotope</th>
<th>MCNP [b]</th>
<th>Interpolated [b]</th>
<th>Relative difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>5.546E-01</td>
<td>5.536E-01</td>
<td>0.18%</td>
</tr>
<tr>
<td>U235</td>
<td>4.735E-01</td>
<td>4.680E-01</td>
<td>1.16%</td>
</tr>
<tr>
<td>U236</td>
<td>3.922E-01</td>
<td>3.904E-01</td>
<td>0.46%</td>
</tr>
<tr>
<td>U238</td>
<td>3.020E-01</td>
<td>3.017E-01</td>
<td>0.10%</td>
</tr>
<tr>
<td>Np237</td>
<td>1.382E+00</td>
<td>1.369E+00</td>
<td>0.94%</td>
</tr>
<tr>
<td>Pu238</td>
<td>6.592E-01</td>
<td>6.533E-01</td>
<td>0.90%</td>
</tr>
<tr>
<td>Pu239</td>
<td>4.275E-01</td>
<td>4.204E-01</td>
<td>1.66%</td>
</tr>
<tr>
<td>Pu240</td>
<td>4.444E-01</td>
<td>4.421E-01</td>
<td>0.52%</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.916E-01</td>
<td>3.885E-01</td>
<td>0.79%</td>
</tr>
<tr>
<td>Pu242</td>
<td>3.917E-01</td>
<td>3.916E-01</td>
<td>0.03%</td>
</tr>
<tr>
<td>Am241</td>
<td>1.498E+00</td>
<td>1.483E+00</td>
<td>1.00%</td>
</tr>
<tr>
<td>Am242m</td>
<td>3.390E-01</td>
<td>3.336E-01</td>
<td>1.59%</td>
</tr>
<tr>
<td>Am243</td>
<td>1.301E+00</td>
<td>1.284E+00</td>
<td>1.31%</td>
</tr>
<tr>
<td>Cm242</td>
<td>2.765E-01</td>
<td>2.736E-01</td>
<td>1.05%</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.175E-01</td>
<td>2.143E-01</td>
<td>1.47%</td>
</tr>
<tr>
<td>Cm244</td>
<td>7.578E-01</td>
<td>7.515E-01</td>
<td>0.83%</td>
</tr>
<tr>
<td>Cm245</td>
<td>2.821E-01</td>
<td>2.793E-01</td>
<td>0.99%</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.073E-01</td>
<td>2.048E-01</td>
<td>1.21%</td>
</tr>
</tbody>
</table>

Table 5.3 compares the interpolated cross sections at the beginning of the 100th cycle with the effective one-group cross sections calculated by MCNP for the WACOM calculated composition. The interpolated cross sections are in good agreement with those estimated by MCNP. This indicates that the interpolation scheme used to update the effective one-group cross sections is satisfactory and can be used for the ATW fuel cycle analyses.

### 5.3 Analysis of the Reference LBE-Cooled Transmuter

The performance of the reference LBE-cooled transmuter was assessed by revised WACOM described in Section 5.2. Used input data are listed in Table 4.2 and 4.5 and
following operating conditions are assumed: (1) the cycle duration is controlled by bounding the range of $k_{\text{eff}}$ variation from 0.98 to 0.92, (2) the recovery fraction of each TRU element by partitioning is 0.999 and recovered TRU is free from fission products, (3) the fuel discharge fraction is 1/6 although the burnup is assumed to be uniform across the blanket, (4) the reactor power is 840 MWt, (5) no cooling time for discharged fuel is assumed, (6) TRU composition of feed material is constant at that of SNF from PWR after 25-year cooling and 99.995% uranium removal, (7) no mass loss from fuel fabrication process is assumed, and (8) except fuel irradiation in reactor core, all processes are performed instantaneously. For (8), current version of WACOM model does not take into account the cooling time of discharged fuel before and during separation process, the cooling time of accumulating TRU waste from the separation process, and the time required for refueling in each cycle.

The evolution of the core inventory in the LBE-cooled transmuter is given in Figure 5.3. Figure 5.5 shows the corresponding evolution of the cycle duration. It is estimated that cycle duration of LBE-cooled transmuter will increase from ~86 days for the start-up cycle to ~144 days for the equilibrium cycle while the TRU mass fraction increases from ~32% to ~41%. The cycle duration for the equilibrium is well matched to that, 145 days, evaluated by REBUS3 code [Toppel, 1983] in [AAA-RPO-SYS-01-0008, 2001].

It takes nearly 60 cycles to approach the equilibrium cycle, with most of the variations taking place during the first 20 cycles or so.

![Figure 5.5](image)

**Figure 5.5  Cycle duration of the LBE-cooled ATW system**

Figure 5.6 shows the cycle dependent in-core inventory of fuel isotopes in the transmuter. Most nuclides exhibit a zigzag pattern which is due to the transmutation followed by fuel makeup. The difference between the top and the bottom points in the zigzag represents the amount of a nuclide transmuted in the given cycle. $^{237}\text{Np}$, $^{239}\text{Pu}$, and $^{241}\text{Am}$ have relatively high fractional transmutation per cycle. The concentration of most of nuclide except zirconium increases with cycle number until it reaches a steady state.
although some Cm isotopes such as $^{244}$Cm, $^{245}$Cm, and $^{246}$Cm do not reach a steady-state even in 100 cycles.

The increase in the mass fraction of actinides in the fuel compensates the reactivity loss due to fission products buildup and the change in the actinide composition. For example, the atomic fraction of $^{239}$Pu that has a large fission-to-capture cross section ratio decreases from ~53% to ~24% while that of $^{240}$Pu that has a small fission-to-capture cross section increases from ~22% to ~35%.

Table 5.4 shows the isotopic composition of TRU in the start-up core and in the equilibrium core. The capture-to-fission ratios of TRU isotopes in the start-up core and in the equilibrium core are listed in Table 5.5. All isotopes show decreased capture-to-fission ratios in the equilibrium core compared to the start-up core.

![Cycle dependent in-core inventory of fuel isotopes in the LBE-cooled ATW system](image)

**Figure 5.6** Cycle dependent in-core inventory of fuel isotopes in the LBE-cooled ATW system
Table 5.4  Isotopic Composition of TRU in the Start-up and the Equilibrium Core

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Start-up (at%)</th>
<th>Equilibrium (at%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>0.000</td>
<td>0.251</td>
</tr>
<tr>
<td>U235</td>
<td>0.004</td>
<td>0.067</td>
</tr>
<tr>
<td>U236</td>
<td>0.002</td>
<td>0.101</td>
</tr>
<tr>
<td>U238</td>
<td>0.481</td>
<td>1.124</td>
</tr>
<tr>
<td>Np237</td>
<td>5.077</td>
<td>2.674</td>
</tr>
<tr>
<td>Pu238</td>
<td>1.280</td>
<td>5.240</td>
</tr>
<tr>
<td>Pu239</td>
<td>53.314</td>
<td>23.936</td>
</tr>
<tr>
<td>Pu240</td>
<td>21.492</td>
<td>34.581</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.759</td>
<td>6.164</td>
</tr>
<tr>
<td>Pu242</td>
<td>4.638</td>
<td>11.093</td>
</tr>
<tr>
<td>Am241</td>
<td>8.912</td>
<td>5.490</td>
</tr>
<tr>
<td>Am242m</td>
<td>0.014</td>
<td>0.386</td>
</tr>
<tr>
<td>Am243</td>
<td>0.913</td>
<td>3.509</td>
</tr>
<tr>
<td>Cm242</td>
<td>0.000</td>
<td>0.572</td>
</tr>
<tr>
<td>Cm243</td>
<td>0.002</td>
<td>0.057</td>
</tr>
<tr>
<td>Cm244</td>
<td>0.102</td>
<td>3.270</td>
</tr>
<tr>
<td>Cm245</td>
<td>0.009</td>
<td>0.980</td>
</tr>
<tr>
<td>Cm246</td>
<td>0.001</td>
<td>0.503</td>
</tr>
</tbody>
</table>

Table 5.5  Capture to Fission Ratios of TRU Isotopes in the Start-up and the Equilibrium Core

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Start-up</th>
<th>Equilibrium</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>2.06</td>
<td>1.61</td>
</tr>
<tr>
<td>U235</td>
<td>0.28</td>
<td>0.27</td>
</tr>
<tr>
<td>U236</td>
<td>5.63</td>
<td>4.56</td>
</tr>
<tr>
<td>U238</td>
<td>13.50</td>
<td>10.43</td>
</tr>
<tr>
<td>Np237</td>
<td>5.01</td>
<td>4.21</td>
</tr>
<tr>
<td>Pu238</td>
<td>0.67</td>
<td>0.59</td>
</tr>
<tr>
<td>Pu239</td>
<td>0.29</td>
<td>0.25</td>
</tr>
<tr>
<td>Pu240</td>
<td>1.43</td>
<td>1.17</td>
</tr>
<tr>
<td>Pu241</td>
<td>0.18</td>
<td>0.17</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.84</td>
<td>1.48</td>
</tr>
<tr>
<td>Am241</td>
<td>7.06</td>
<td>5.93</td>
</tr>
<tr>
<td>Am242m</td>
<td>0.10</td>
<td>0.09</td>
</tr>
<tr>
<td>Am243</td>
<td>8.06</td>
<td>6.61</td>
</tr>
<tr>
<td>Cm242</td>
<td>2.78</td>
<td>2.07</td>
</tr>
<tr>
<td>Cm243</td>
<td>0.10</td>
<td>0.09</td>
</tr>
<tr>
<td>Cm244</td>
<td>2.09</td>
<td>1.76</td>
</tr>
<tr>
<td>Cm245</td>
<td>0.13</td>
<td>0.13</td>
</tr>
<tr>
<td>Cm246</td>
<td>1.02</td>
<td>0.82</td>
</tr>
</tbody>
</table>
Figure 5.7 Fractional transmutation of TRU isotope with cycle number

Figure 5.7 shows fractional transmutation of TRU isotope with cycle number. Fractional transmutation of TRU isotope is defined as the ratio of the difference between the amount of an isotope in the fuel at BOC and that at EOC to that at BOC within a cycle.

Each TRU isotope in the reactor core experiences increase or decrease in its concentration during irradiation. In Figure 5.7, positive value means that the amount of an isotope decreases during the cycle and negative value implies that it increases during the cycle. As observed in the figure, we can see that $^{237}$Np, $^{239}$Pu, and $^{241}$Am have relatively large positive fractional transmutations (more than ~10%), whereas other Pu isotopes, $^{238}$U, and $^{243}$Am have very small positive fractional transmutations (less than
~3%). Most of the uranium and curium isotopes show buildup in the reactor at early cycles and then they approach to a steady-state, which means no depletion or accumulation of those nuclides in the fuel by recycling.

Figure 5.8 shows the cumulative mass reduction ratio of TRU with the cycle number. The cumulative mass reduction ratio is defined as the cumulative mass of an isotope up to the n-th cycle in the waste stream divided by the cumulative mass of the same isotope up to the n-th cycle fed to the ATW system in the makeup stream. The ratio means to what extent TRU inventory can be reduced by the ATW system after a certain number of cycles. A small reduction ratio is desirable since it means more TRU is transmuted in the transmuter. Figure 5.8 represents the case in which the TRU in the start-up core or in the core to be decommissioned is not considered as makeup or waste. The cumulative inventory of TRU in both waste and makeup stream increases almost linearly with a similar rate except in early cycles. In early cycle the mass accumulation rate in waste stream is a little faster than that in makeup stream but as cycle number increases the mass accumulation rate in both streams becomes closer. Therefore, the cumulative mass reduction ratio increases with the cycle number but it converges to a certain value (here, 0.0025) as cycle number increases. Figure 5.8 shows the inventory of TRU taken from LWR spent fuel is reduced by a factor of ~400 at the end of the 100th cycle. In other word, ~12.6 MT of TRU is consumed as feed, while ~0.03 MT of TRU is generated as waste by the LBE-cooled transmuting system during the 100 cycle operations.

![Cumulative mass reduction ratio of TRU with cycle number](image)

**Figure 5.8  Cumulative mass reduction ratio of TRU with cycle number**

Figure 5.9 shows the cumulative mass fraction of TRU isotope in waste with cycle number. It shows that $^{239}$Pu is the most abundant TRU isotope in the waste from early cycles (50.5 wt% at the end of the 1st cycle) but as cycle number increases its fraction decreases gradually (24.9 wt% at the end of the 100th cycle). On the other hand, the fraction of $^{240}$Pu increases from 23.1 wt% (1st cycle) to 35.1 wt% (100th cycle). This is
because even though $^{239}\text{Pu}$-riched TRU is supplied to the transmuter for makeup, through multi-cycle operations $^{239}\text{Pu}$ is well depleted out by fission and capture reactions while $^{240}\text{Pu}$ builds up due to the neutron capture reaction of $^{239}\text{Pu}$. It implies that in order to make the waste less attractive from the aspect of proliferation, multi-recycling of TRU is beneficial.

\[ \text{Figure 5.9 Cumulative mass fraction of TRU isotope in waste with cycle number} \]

Figure 5.10 shows the cumulative toxicity reduction ratio of TRU with cycle. The toxicity is defined as the volume of air or water with which the mixture of radionuclides must be diluted so that breathing the air or drinking the water will result in accumulation of radiation dose at a rate no greater than 0.5 rem/year [Benedict, et al., 1981]. For a TRU isotope, here, the toxicity is calculated by dividing the radioactivity of the isotope by ALI of that isotope listed in Table 4.2. The cumulative toxicity reduction ratio is defined as the toxicity of all TRU isotopes up to n-th cycle included in the waste stream divided by that of all TRU isotopes up to n-th cycle included in the makeup material. This is a key measure for the transmuting reactor performance in terms of reducing the hazard of TRU. Figure 5.10 shows that the cumulative toxicity of TRU taken from LWR spent fuel is reduced roughly by a factor of 100 at the end of 100 cycle operations.

Figure 5.11 shows the cumulative toxicity fraction of TRU isotope in waste with the cycle number. In early cycles, $^{238}\text{Pu}$, $^{241}\text{Am}$, and $^{242}\text{Cm}$ are main sources of toxicity in waste but as cycle number increases the toxicity fraction of $^{241}\text{Am}$ gets smaller and smaller instead that of $^{244}\text{Cm}$ increases rapidly and becomes dominant in later cycles. It is noticeable that even though the combined mass fraction of $^{238}\text{Pu}$, $^{241}\text{Am}$, $^{242}\text{Cm}$, and
$^{244}$Cm remains small in the waste throughout the entire transmutation campaign, most of toxicity is attributed to those isotopes. As shown Figure 5.9 the combined mass fraction of those isotopes is 11.0% at the end of the 1st cycle and 14.7% at the end of the 100th cycle, while the combined toxicity fraction of those is 84.7% at the end of the 1st cycle and 91.2% at the end of the 100th cycle (Figure 5.11). This is because of their short half-lives and/or relatively small ALI values as shown in Table 4.2.

![Figure 5.10 Cumulative toxicity reduction ratio of TRU with cycle number](image)

The radioactivity of $^{242}$Cm per unit mass is very high compared to other actinides considered in this model due to its very short half-life (163 days). So, even a small mass fraction of $^{242}$Cm isotope can cause a large toxicity fraction in waste. In Figure 5.11, the toxicity fraction of $^{242}$Cm isotope in waste at the end of the 1st cycle is 30.8% in spite that its mass fraction is merely 0.4%. A similar observation is made for $^{244}$Cm isotope with a half-life of 18.1 years and an ALI value of $3 \times 10^{-11}$ [Ci/l]. Its cumulative toxicity fraction changes from 7.4% to 41.0% as its cumulative mass fraction increases from 0.2% to 2.7% and thus at the end of the 100th cycle its toxicity becomes most dominant. The combined toxicity fraction of $^{242}$Cm and $^{244}$Cm increases from 38.2% to 58.8% through 100 cycle operations while the combined mass fraction increases from 0.6% to 3.4%. These observations imply that in order to reduce the toxicity in TRU waste further, it is important to effectively transmute isotopes that have short half-lives and/or small ALI values and their precursors.

Fractional mass of $^{241}$Am decreases slowly through entire cycle (from 8.7% to 5.7%) but its fractional toxicity goes down relatively fast (from 23.7% to 5.5%). Fractional mass of $^{238}$Pu increases from 1.7% to 5.6% through 100 cycles while its fractional toxicity goes up from 22.8% to 26.9%. The fractional toxicity of $^{242}$Cm even decreases from 30.8% to 17.8% while its mass fraction increases a little (from 0.4% to 0.7%). These observations
are due to the effect of a rapid increase in the toxicity of $^{244}$Cm, which is most dominant at the end of the 100th cycle.

$^{239}$Pu and $^{240}$Pu, which are two major contributors to the mass of the TRU waste, pose no major concerns in terms of toxicity due to their small toxicity fractions, 0.4% and 2.3% at the end of the 100th cycle, respectively.

![Figure 5.11 Cumulative toxicity fraction of TRU isotope in waste with cycle number](image)

### 5.4 Effects of Difference in the Branching Ratio of $^{241}$Am

In an international benchmark exercise for an accelerator-driven system led by the OECD Nuclear Energy Agency in 1999, noticeable differences have been observed between the calculation results contributed by different organizations [NEA/NSC/DOC(2001)13, 2001]. There are many possible reasons for the differences. Among these is the use of different branching ratios for the conversion of $^{241}$Am into the two isomers of $^{242}$Am. WACOM code was applied to study the effect of the uncertainty in the branching ratio of $^{242}$Am:$^{242m}$Am on the evolution of the TRU inventory in multi-cycling in the ATW system.

Two sets of branching ratios are considered; one taken from the REBUS3 code system [Toppel, 1983] that used the branching ratio of 0.80 to 0.20 for $^{242}$Am to $^{242m}$Am whereas the other from ORIHET3 [Atchison, et al., 2001], an adaptation of the ORIGEN code, that used 0.85 to 0.15 for the branching ratio. Figure 5.12 shows the simplified transmutation chain of Am isotopes used in WACOM. Neutron reactions of $^{242}$Am and $^{244}$Am which have very short half-lives (16h, 10h) are neglected and they are assumed to decay into their daughters instantaneously.
Table 5.6 shows the WACOM results for the TRU composition in the fuel after a single irradiation cycle. The relative difference in concentration caused by use of different branching ratios is the largest for $^{242m}\text{Am}$ followed by $^{242}\text{Cm}$ and $^{245}\text{Cm}$. The branching ratio difference has only a very small effect on the concentration of the other isotopes. However, this effect increases with the cycle number, as shown in Table 5.7.

Table 5.6  TRU Compositions in Fuel after a Single Cycle Irradiation

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{242}\text{Am};^{242m}\text{Am}$ (0.8:0.2)</th>
<th>$^{242m}\text{Am};^{242}\text{Am}$ (0.85:0.15)</th>
<th>Relative Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>8.781E+22</td>
<td>8.795E+22</td>
<td>-0.16%</td>
</tr>
<tr>
<td>U235</td>
<td>1.349E+23</td>
<td>1.349E+23</td>
<td>0.01%</td>
</tr>
<tr>
<td>U236</td>
<td>8.821E+22</td>
<td>8.822E+22</td>
<td>-0.01%</td>
</tr>
<tr>
<td>U238</td>
<td>1.595E+25</td>
<td>1.595E+25</td>
<td>0.00%</td>
</tr>
<tr>
<td>Np237</td>
<td>1.568E+26</td>
<td>1.568E+26</td>
<td>0.01%</td>
</tr>
<tr>
<td>Pu238</td>
<td>5.377E+25</td>
<td>5.393E+25</td>
<td>-0.30%</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.615E+27</td>
<td>1.615E+27</td>
<td>0.01%</td>
</tr>
<tr>
<td>Pu240</td>
<td>7.373E+26</td>
<td>7.373E+26</td>
<td>0.01%</td>
</tr>
<tr>
<td>Pu241</td>
<td>1.259E+26</td>
<td>1.259E+26</td>
<td>0.00%</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.573E+26</td>
<td>1.575E+26</td>
<td>-0.13%</td>
</tr>
<tr>
<td>Am241</td>
<td>2.757E+26</td>
<td>2.757E+26</td>
<td>0.01%</td>
</tr>
<tr>
<td>Am242m</td>
<td>4.610E+24</td>
<td>3.555E+24</td>
<td>22.90%</td>
</tr>
<tr>
<td>Am243</td>
<td>3.186E+25</td>
<td>3.186E+25</td>
<td>0.02%</td>
</tr>
<tr>
<td>Cm242</td>
<td>1.294E+25</td>
<td>1.376E+25</td>
<td>-6.35%</td>
</tr>
<tr>
<td>Cm243</td>
<td>1.700E+23</td>
<td>1.773E+23</td>
<td>-4.28%</td>
</tr>
<tr>
<td>Cm244</td>
<td>5.421E+24</td>
<td>5.423E+24</td>
<td>-0.03%</td>
</tr>
<tr>
<td>Cm245</td>
<td>4.342E+23</td>
<td>4.343E+23</td>
<td>-0.04%</td>
</tr>
<tr>
<td>Cm246</td>
<td>3.749E+22</td>
<td>3.750E+22</td>
<td>-0.02%</td>
</tr>
</tbody>
</table>
Table 5.7  TRU Compositions in Fuel after 100 Cycle Irradiation

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{242}\text{Am}:^{242m}\text{Am}$ (0.8:0.2)</th>
<th>$^{242}\text{Am}:^{242m}\text{Am}$ (0.85:0.15)</th>
<th>Relative Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>1.322E+25</td>
<td>1.370E+25</td>
<td>-3.63%</td>
</tr>
<tr>
<td>U235</td>
<td>3.497E+24</td>
<td>3.611E+24</td>
<td>-3.28%</td>
</tr>
<tr>
<td>U236</td>
<td>5.329E+24</td>
<td>5.433E+24</td>
<td>-1.95%</td>
</tr>
<tr>
<td>U238</td>
<td>5.772E+25</td>
<td>5.788E+25</td>
<td>-0.28%</td>
</tr>
<tr>
<td>Np237</td>
<td>1.249E+26</td>
<td>1.252E+26</td>
<td>-0.30%</td>
</tr>
<tr>
<td>Pu238</td>
<td>2.716E+26</td>
<td>2.808E+26</td>
<td>-3.39%</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.093E+27</td>
<td>1.098E+27</td>
<td>-0.46%</td>
</tr>
<tr>
<td>Pu240</td>
<td>1.752E+27</td>
<td>1.756E+27</td>
<td>-0.21%</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.125E+26</td>
<td>3.127E+26</td>
<td>-0.08%</td>
</tr>
<tr>
<td>Pu242</td>
<td>5.692E+26</td>
<td>5.752E+26</td>
<td>-1.06%</td>
</tr>
<tr>
<td>Am241</td>
<td>2.610E+26</td>
<td>2.618E+26</td>
<td>-0.30%</td>
</tr>
<tr>
<td>Am242m</td>
<td>2.028E+25</td>
<td>1.528E+25</td>
<td>24.66%</td>
</tr>
<tr>
<td>Am243</td>
<td>1.818E+26</td>
<td>1.822E+26</td>
<td>-0.22%</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.007E+25</td>
<td>3.193E+25</td>
<td>-6.19%</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.980E+24</td>
<td>3.155E+24</td>
<td>-5.86%</td>
</tr>
<tr>
<td>Cm244</td>
<td>1.718E+26</td>
<td>1.719E+26</td>
<td>-0.04%</td>
</tr>
<tr>
<td>Cm245</td>
<td>5.159E+25</td>
<td>5.157E+25</td>
<td>0.04%</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.662E+25</td>
<td>2.652E+25</td>
<td>0.38%</td>
</tr>
</tbody>
</table>

5.5 Summary and Conclusions

WACOM code has been developed for efficient simulation of the cycle-by-cycle fuel inventory evolution in ATW systems. To account for the variation in the neutron spectrum of a transmuting reactor core, a method has been implemented, which involves interpolation of effective one-group cross sections generated with MCNP for the start up core composition and an equilibrium core composition. A linear interpolation scheme was found to be adequate; it enables prediction of the fuel composition evolution while satisfying the $k_{eff}$ constraint using only two MCNP calculations.

With the revised WACOM, the performance of the LBE-cooled transmuter in transition was assessed. It is estimated that the fractional mass of TRU of BOC-fuel in the LBE-cooled transmuter increases from ~32% to ~41%, if the $k_{eff}$ control is achieved by adjusting the TRU-to-zirconium volume ratio. The corresponding cycle duration increases from ~86 days to ~144 days. The variations in the TRU fraction and in the cycle duration are particularly pronounced during the first 20 cycles and taper off after ~60 cycles. Fractional transmutations of $^{237}\text{Np}$, $^{239}\text{Pu}$, and $^{241}\text{Am}$ are relatively high compared to other TRU isotopes in the LBE-cooled transmuter. Difference in isotopic compositions of TRUs in the start-up core and in the equilibrium core is noticeable. $^{239}\text{Pu}$ is the most abundant isotope in the start-up core (53.3%) but $^{240}\text{Pu}$ in the equilibrium core (34.6%). Calculation of cumulative mass and toxicity reduction ratios indicates that the TRU waste generated from the separation process of ATW-spent fuel are made by several isotopes with relatively short half-lives and small ALI values. About 91.2% of toxicity of TRU waste is attributed to $^{238}\text{Pu}$, $^{241}\text{Am}$, $^{242}\text{Cm}$, and $^{244}\text{Cm}$, whose combined mass
fraction is of 14.7%. This implies that in order to reduce toxicity in TRU waste further, such isotopes and their precursors need to be transmuted more effectively. Considering a practical cooling time of discharged fuel from transmuting core would also result in a reduced toxicity of the TRU waste.

Effects of difference in the branching ratio of $^{241}$Am on results of multi-cycle analyses for the ATW reactor have also been investigated. It is found that a 5% difference in the branching ratios of $^{241}$Am makes a significant difference in the $^{242m}$Am concentration and some differences in the $^{242}$Cm and $^{243}$Cm concentrations after a single irradiation cycle. After 100 cycles the discrepancy propagates to other radionuclides such as U and Pu isotopes. These findings indicate that accurate estimation of the branching ratio of $^{241}$Am is important for reliable evaluations of actinide transmutation performance.

The performances of the LBE-cooled transmuter assessed in this chapter are based on a single transmuter independent of the ATW deployment scenario [Hill, et al., 1999]. In the next chapter, we evaluate the reduction of TRU inventory contained in LWR-SNF based on the ATW deployment scenario prepared for processing a specific amount of LWR-SNF.
6 Reduction of TRU Inventory Contained in LWR-Spent Fuel

6.1 Introduction

In Chapter 5, we have dealt with a modification of WACOM to take into account the variation in the neutron spectrum of a transmuter core. It has been found that the implemented linear interpolating scheme enables the prediction of the time- and cycle-dependent evolution of fuel inventory in the transmuter core adequately.

In this chapter, the reduction of TRU inventory in LWR-SNF is investigated with the reference LBE-cooled transmuter which has been analyzed by modified WACOM in Chapter 5. The reference ATW plant illustrated in Figure 1.2 is applied, which includes eight LBE-cooled subcritical transmuters to process a specific amount of LWR-SNF for its life-time operation.

The mass and the composition of the TRU from the waste streams of the entire operation of the reference ATW plants are estimated and then compared with those of the TRU originally included in the LWR-SNF in the aspects of mass and toxicity reductions. Transmutations of long-lived fission products such as Tc and I are not treated here even though the reference ATW plant aims at the reduction of those as well.

6.2 Characteristics of the LBE-Cooled Transmuter

In Chapter 1, the key features of the reference ATW plant and its deployment scenario were overviewed, with its parameters listed in Appendix A. In this section, we replace the ATW plant parameters for the transmuter systems with the parameters of the LBE-cooled transmuter calculated by WACOM on the cycle-by-cycle basis.

It is found that the LBE-cooled transmuter in an equilibrium cycle consumes 246 kg/yr TRU taken from SNF to produce the power of 840 MWt. This consumption rate is compared to 237 kg/yr calculated by REBUS-3 code [Toppel, 1983] for the same LBE-transmuter in [AAA-RPO-SYS-01-0008, 2001]. The difference is due to a little discrepancy in the applied $k_{eff}$ constraint: $\Delta k_{eff} = 0.060$ in WACOM and $\Delta k_{eff} = 0.057$ in REBUS-3.

The consumption rate assumed for the reference ATW plant in [Hill, et al., 1999] is even 220 kg/yr. This is because the capacity factor of the reference ATW plant is assumed in [Hill, et al., 1999] to be 70% while that of the LBE-cooled transmuter is evaluated as 79% based on the applied $\Delta k_{eff}$. Other contributions may include difference in values of parameters such as average recoverable fission energy, target spallation energy, and average neutrons produced per fission.

In ATW fuel cycle analysis, a greater TRU consumption rate results in a shorter period of ATW plant operation than a 60-year life time for processing a specified amount of LWR-SNF. With the cycle-by-cycle calculation, WACOM shows that it takes about 112 cycles (56 years) for the LBE-transmuter to consume 13.25 MT of TRU under the assumed operating mode. Of the 13.25 MT of TRU taken from LWR-SNF, 13.214 MT is converted into FPs and 0.036 MT is lost from the separation process of discharged ATW fuel.

---

9 In the reference ATW plant, a transmuter is supposed to consume 13.25 MT of TRU for a 60-year life-time.

10 99.9% recovery yield is applied.
The performance characteristics of the LBE-cooled transmuter estimated by WACOM are summarized in Table 6.1.

<table>
<thead>
<tr>
<th>Multiplication factor</th>
<th>BOEC(^{11})</th>
<th>0.980</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>EOEC(^{12})</td>
<td>0.920</td>
</tr>
<tr>
<td>Burnup reactivity loss (%(\Delta k_{\text{eff}}))</td>
<td>6.1</td>
<td></td>
</tr>
<tr>
<td>Equilibrium % TRU fissioned</td>
<td>27.2</td>
<td></td>
</tr>
<tr>
<td>Equilibrium TRU waste from ATW fuel separation process (kg/year)</td>
<td>0.659</td>
<td></td>
</tr>
<tr>
<td>Equilibrium TRU consumption rate (kg/year)</td>
<td>246</td>
<td></td>
</tr>
<tr>
<td>Equilibrium loading (kg/year)</td>
<td>LWR-TRU</td>
<td>246</td>
</tr>
<tr>
<td></td>
<td>Recycled-TRU</td>
<td>658</td>
</tr>
<tr>
<td></td>
<td>Total TRU</td>
<td>904</td>
</tr>
<tr>
<td>TRU inventory (kg)</td>
<td>BOEC</td>
<td>2100</td>
</tr>
<tr>
<td></td>
<td>EOEC</td>
<td>1976</td>
</tr>
<tr>
<td>Fission products inventory (kg)</td>
<td>BOEC</td>
<td>617</td>
</tr>
<tr>
<td></td>
<td>EOEC</td>
<td>740</td>
</tr>
<tr>
<td>Zirconium inventory (kg)</td>
<td>BOEC</td>
<td>2398</td>
</tr>
<tr>
<td></td>
<td>EOEC</td>
<td>2398</td>
</tr>
</tbody>
</table>

### 6.3 Reduction of TRU Inventory Contained in LWR-SNF

After a complete operation of the reference ATW plant, the total mass of TRU, which would be included in the waste streams is quantified to be 5.291 MT. It comes from three sources: (1) 0.935 MT TRU included in the waste arising from reprocessing of 86,317 MT SNF, (2) 2.380 MT TRU included in the waste from ATW fuel processing in a 56-year operation period, and (3) 1.976 MT TRU included in the very last LBE-cooled transmuter core. For (1) and (2), composition change of TRU included in the waste stream by radioactive decay during the ATW operating period was not taken into account. For (3), it is assumed that any residual quantities of TRU remaining in the plant shut down would be fed into any ATW plants that remain in operating mode. It is noticeable that the separation waste from ATW fuel processing is longer than that from LWR-SNF reprocessing. The difference comes because the former gets to pass through the ATW fuel processing facility many times due to TRU-recycling in the ATW fuel cycle.

The isotopic compositions of the three different TRU waste sources are shown in Table 6.2. The isotopic composition of (1) is supposed to be the same as that of LWR-TRU because the same recovery fraction was applied for all LWR-TRU isotopes in LWR-spent fuel separation process. The isotopic compositions of (2) and (3) are similar to each other but quite different from that of (1). By recycling of TRU during the ATW plant operation period, mass fractions of \(^{237}\)Np, \(^{239}\)Pu, and \(^{241}\)Am of LWR-TRU decrease instead those of other TRU isotopes increase. The fractional mass of \(^{239}\)Pu, which is the most abundant isotope in LWR-TRU, decreases from \(~53\%\) to \(~24\%\) in ATW-TRU.

---

\(^{11}\) Beginning of equilibrium cycle
\(^{12}\) End of equilibrium cycle
waste while that of $^{240}$Pu increases from ~22% to ~35% and becomes most abundant in ATW-TRU waste.

Table 6.2  Isotopic Compositions of 0.935 MT, 2.380 MT, and 1.976 MT TRU

<table>
<thead>
<tr>
<th>Isotope</th>
<th>0.935 MT TRU</th>
<th>2.380 MT TRU</th>
<th>1.976 MT TRU</th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>0.000</td>
<td>0.224</td>
<td>0.260</td>
</tr>
<tr>
<td>U235</td>
<td>0.004</td>
<td>0.057</td>
<td>0.069</td>
</tr>
<tr>
<td>U236</td>
<td>0.002</td>
<td>0.072</td>
<td>0.106</td>
</tr>
<tr>
<td>U238</td>
<td>0.478</td>
<td>0.987</td>
<td>1.155</td>
</tr>
<tr>
<td>Np237</td>
<td>5.023</td>
<td>2.731</td>
<td>2.487</td>
</tr>
<tr>
<td>Pu238</td>
<td>1.272</td>
<td>5.546</td>
<td>5.420</td>
</tr>
<tr>
<td>Pu239</td>
<td>53.196</td>
<td>24.454</td>
<td>21.986</td>
</tr>
<tr>
<td>Pu240</td>
<td>21.534</td>
<td>35.168</td>
<td>35.360</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.782</td>
<td>6.261</td>
<td>6.332</td>
</tr>
<tr>
<td>Pu242</td>
<td>4.686</td>
<td>10.546</td>
<td>11.601</td>
</tr>
<tr>
<td>Am241</td>
<td>8.967</td>
<td>5.630</td>
<td>5.268</td>
</tr>
<tr>
<td>Am242m</td>
<td>0.014</td>
<td>0.429</td>
<td>0.413</td>
</tr>
<tr>
<td>Am243</td>
<td>0.926</td>
<td>3.244</td>
<td>3.718</td>
</tr>
<tr>
<td>Cm242</td>
<td>0.000</td>
<td>0.660</td>
<td>0.613</td>
</tr>
<tr>
<td>Cm243</td>
<td>0.002</td>
<td>0.064</td>
<td>0.061</td>
</tr>
<tr>
<td>Cm244</td>
<td>0.104</td>
<td>2.819</td>
<td>3.526</td>
</tr>
<tr>
<td>Cm245</td>
<td>0.009</td>
<td>0.803</td>
<td>1.064</td>
</tr>
<tr>
<td>Cm246</td>
<td>0.001</td>
<td>0.320</td>
<td>0.552</td>
</tr>
</tbody>
</table>

Thus, a total of 900 MT TRU separated from 86,317 MT of LWR-SNF has been reduced to 5.291 MT (i.e., by a factor of 170) by the assumed ATW plant deployment scenario. Table 6.3 shows the mass reduction factor of each TRU isotope, which is defined as the ratio of the mass of an isotope in 900 MT TRU to that in 5.291 MT TRU. The isotopic composition of 5.291 MT TRU is given by the three TRU waste sources described in the previous paragraph. The table shows that $^{237}$Np, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, and $^{241}$Am can be reduced more than a factor of one hundred by the reference ATW plant. Among those, mass reduction factors of $^{237}$Np, $^{239}$Pu, and $^{241}$Am are greater than the overall mass reduction factor, 170, which means that reduction of these isotopes could be done preferentially.

For $^{237}$Np and its precursor, $^{241}$Am, which are considered the main source of radiological hazards in a long-term period in a geological repository, their mass reduction factors are 281 and 251, respectively. This implies that radiological impacts of those isotopes from a geological repository could significantly be reduced by the reference ATW plant operation.

Table 6.4 shows the toxicity of each TRU isotope in 900 MT TRU and in 5.291 MT TRU, and the toxicity reduction factor of each isotope, which is defined as the ratio of the toxicity of an isotope in 900 MT TRU to that in 5.291 MT TRU. The toxicity reduction factor of each TRU isotope has the same value with the mass reduction factor of each.

---

\[^{13}\] 33,000 MWd/MT burnup PWR spent fuel with 25-year cooling time (99.995% of the uranium is removed in the UREX process)
TRU isotope as shown in Table 6.3 because the toxicity of an isotope is directly proportional to its mass. The overall toxicity reduction factor, however, is different from the overall mass reduction factor. This is because the toxicity fraction of an isotope is not identical to its mass fraction. For instance, the mass fraction of Pu-isotopes is ~82.0% in ATW-TRU waste while the toxicity fraction of those is ~32.4%. Isotopes with relatively short half-lives and/or small ALI values such as $^{238}$Pu, $^{241}$Am, $^{242}$Cm, and $^{244}$Cm have higher toxicity values per unit mass compared to others.

Figures 6.1 and 6.2 show the mass fraction and the toxicity fraction of each TRU isotope in 900 MT TRU and in 5.291 MT TRU, respectively. In both 900 MT TRU and 5.291 MT TRU, the mass fraction of Pu-isotopes is dominant but the most abundant isotope in 900 MT TRU is $^{239}$Pu, whereas in 5.291 MT TRU is $^{240}$Pu. In toxicity fraction, that of Pu-isotopes is dominant in 900 MT TRU while that of Cm-isotopes in 5.291 MT TRU. The toxicity fraction of Cm-isotopes in 5.291 MT TRU is ~59.8% even though the mass fraction of those isotopes is as small as ~4.2%.

**Table 6.3** Mass Reduction Factor of Each TRU-Isotope by the Reference ATW Plant

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>6.70E+17</td>
<td>2.67E+25</td>
<td>2.51E-08</td>
</tr>
<tr>
<td>U235</td>
<td>9.25E+25</td>
<td>6.98E+24</td>
<td>1.33E+01</td>
</tr>
<tr>
<td>U236</td>
<td>4.60E+25</td>
<td>9.78E+24</td>
<td>4.71E+00</td>
</tr>
<tr>
<td>U238</td>
<td>1.09E+28</td>
<td>1.28E+26</td>
<td>8.49E+01</td>
</tr>
<tr>
<td>Np237</td>
<td>1.15E+29</td>
<td>4.10E+26</td>
<td>2.81E+02</td>
</tr>
<tr>
<td>Pu238</td>
<td>2.90E+28</td>
<td>6.33E+26</td>
<td>4.59E+01</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.21E+30</td>
<td>3.82E+27</td>
<td>3.17E+02</td>
</tr>
<tr>
<td>Pu240</td>
<td>4.87E+29</td>
<td>4.34E+27</td>
<td>1.12E+02</td>
</tr>
<tr>
<td>Pu241</td>
<td>8.53E+28</td>
<td>7.71E+26</td>
<td>1.11E+02</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.05E+29</td>
<td>1.30E+27</td>
<td>8.11E+01</td>
</tr>
<tr>
<td>Am241</td>
<td>2.02E+29</td>
<td>8.05E+26</td>
<td>2.51E+02</td>
</tr>
<tr>
<td>Am242m</td>
<td>3.14E+26</td>
<td>4.58E+25</td>
<td>6.86E+00</td>
</tr>
<tr>
<td>Am243</td>
<td>2.07E+28</td>
<td>3.92E+26</td>
<td>5.28E+01</td>
</tr>
<tr>
<td>Cm242</td>
<td>1.97E+18</td>
<td>6.90E+25</td>
<td>2.86E-08</td>
</tr>
<tr>
<td>Cm243</td>
<td>4.47E+25</td>
<td>6.77E+24</td>
<td>6.60E+00</td>
</tr>
<tr>
<td>Cm244</td>
<td>2.32E+27</td>
<td>3.37E+26</td>
<td>6.87E+00</td>
</tr>
<tr>
<td>Cm245</td>
<td>2.00E+26</td>
<td>9.79E+25</td>
<td>2.04E+00</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.21E+25</td>
<td>4.62E+25</td>
<td>4.78E-01</td>
</tr>
</tbody>
</table>

Figure 6.3 shows the variation of the toxicity reduction factor with time after waste generation. It illustrates how the ratio of the toxicity of a TRU isotope in the 900 MT TRU feed to that in 5.291 MT TRU waste evolves with time. In the early-time domain, the toxicity reduction factor of the total TRU is as small as 40 at 0.1 year because the fractional toxicity of Cm-isotopes (mainly $^{242}$Cm and $^{244}$Cm) are significant in ATW-TRU waste as shown in Figure 6.4. However, most of the $^{242}$Cm and $^{244}$Cm isotopes decay out within 100 years and thus the factor increases up to ~100. This implies that if the cooling time of ATW-TRU were taken into account in a more realistic manner in the
model, the total toxicity reduction factor in early-time domain would be much larger than 40. The toxicity reduction factor of the total TRU increases up to ~170 in 10,000 years.

Table 6.4  Toxicity Reduction Factor of Each TRU-Isotope by the Reference ATW Plant

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>U234</td>
<td>5.42E+03</td>
<td>2.16E+11</td>
<td>2.51E-08</td>
</tr>
<tr>
<td>U235</td>
<td>2.60E+08</td>
<td>1.96E+07</td>
<td>1.33E+01</td>
</tr>
<tr>
<td>U236</td>
<td>3.89E+09</td>
<td>8.26E+08</td>
<td>4.71E+00</td>
</tr>
<tr>
<td>U238</td>
<td>4.83E+09</td>
<td>5.69E+07</td>
<td>8.49E+01</td>
</tr>
<tr>
<td>Np237</td>
<td>1.59E+15</td>
<td>5.67E+12</td>
<td>2.81E+02</td>
</tr>
<tr>
<td>Pu238</td>
<td>9.83E+18</td>
<td>2.14E+17</td>
<td>4.59E+01</td>
</tr>
<tr>
<td>Pu239</td>
<td>1.49E+18</td>
<td>4.70E+15</td>
<td>3.17E+02</td>
</tr>
<tr>
<td>Pu240</td>
<td>2.21E+18</td>
<td>1.96E+16</td>
<td>1.12E+02</td>
</tr>
<tr>
<td>Pu241</td>
<td>3.84E+18</td>
<td>3.47E+16</td>
<td>1.11E+02</td>
</tr>
<tr>
<td>Pu242</td>
<td>8.35E+15</td>
<td>1.03E+14</td>
<td>8.11E+01</td>
</tr>
<tr>
<td>Am241</td>
<td>1.39E+19</td>
<td>5.52E+16</td>
<td>2.51E+02</td>
</tr>
<tr>
<td>Am242m</td>
<td>6.62E+16</td>
<td>9.65E+15</td>
<td>6.86E+00</td>
</tr>
<tr>
<td>Am243</td>
<td>8.34E+16</td>
<td>1.58E+15</td>
<td>5.28E+01</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.75E+09</td>
<td>1.31E+17</td>
<td>2.86E-08</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.95E+16</td>
<td>4.47E+15</td>
<td>6.61E+00</td>
</tr>
<tr>
<td>Cm244</td>
<td>2.53E+18</td>
<td>3.69E+17</td>
<td>6.87E+00</td>
</tr>
<tr>
<td>Cm245</td>
<td>6.97E+14</td>
<td>3.42E+14</td>
<td>2.04E+00</td>
</tr>
<tr>
<td>Cm246</td>
<td>1.38E+14</td>
<td>2.88E+14</td>
<td>4.78E+01</td>
</tr>
<tr>
<td>Total</td>
<td>3.40E+19</td>
<td>8.45E+17</td>
<td>4.02E+01</td>
</tr>
</tbody>
</table>

In Figure 6.3, toxicity reduction factors of some isotopes are constant while others vary with time. Isotopes that have no precursors such as most Cm-isotopes (except $^{242}$Cm), $^{242m}$Am, $^{243}$Am show constant toxicity reduction factors and some isotopes that have precursors but their effects are negligible ($^{238}$U, $^{239}$Pu, $^{240}$Pu, $^{242}$Pu) also have nearly constant toxicity reduction factors. On the other hand, isotopes that have precursors and their effects are significant show noticeable variations with time.

Most U-isotopes (except $^{238}$U) and $^{242}$Cm show that their toxicity reduction factors increases with time. Mass fractions of U-isotopes and $^{242}$Cm are very small in LWR-TRU taken from the separation process of LWR-SNF because most of U-isotopes are separated by the UREX process and $^{242}$Cm decays out through the cooling period of LWR-SNF. Those isotopes, however, build up in the ATW transmuter and their mass fractions are higher in ATW-TRU waste than in LWR-TRU feed. Therefore, in early time, toxicity reduction factors of those isotopes are small but with time those increase due to decays of their precursors and approach to the toxicity reduction factors of their precursors except $^{234}$U. The sharp increase in $^{234}$U- and $^{242}$Cm- toxicity reduction factors in early time is due to the fact that the fractions of $^{234}$U and $^{242}$Cm in LWR-TRU are assumed to be zero in this study.
Figure 6.1 Mass fraction of TRU isotope in 900 MT TRU (before ATW treatment) and 5.291 MT TRU (after ATW treatment)

Figure 6.2 Toxicity fraction of TRU isotope in 900 MT TRU and 5.291 MT TRU
Figure 6.3 Variation of toxicity reduction factor with time after waste generation (not taken into account the actinides decay during time it takes to accumulate)

The toxicity reduction factors of $^{237}$Np, $^{238}$Pu, $^{241}$Pu, and $^{241}$Am decrease with time. The toxicity reduction factor of $^{237}$Np decreases from 280 to 200 in a time period between ~10 and ~10,000 years. In this time period, most of $^{241}$Pu decays into $^{241}$Am, and then into $^{237}$Np in both 900 MT TRU and 5.291 MT TRU but the fractional buildup of $^{237}$Np in the former is a bit less than that in the latter. After 10,000 years, the toxicity reduction factor of $^{237}$Np remains nearly constant because there is no more effect from its precursor. The toxicity reduction factor of $^{241}$Pu decreases sharply between ~100 and 1,000 years and after ~1,000 years it approaches to the toxicity reduction factor of its precursor ($^{245}$Cm). The half-life of $^{241}$Pu is ~13.2 years, so its toxicity reduction factor after ~100
years becomes same with that of its precursor. $^{241}$Am with a half-life of ~432 years also shows a similar pattern after 1,000 years and it approaches to the toxicity reduction factor of its precursor, $^{241}$Pu (or $^{245}$Cm). After 10,000 years, three isotopes ($^{241}$Pu, $^{241}$Am, and $^{245}$Cm) are in a secular equilibrium and thus have the same toxicity reduction factor. The toxicity reduction factor of $^{238}$Pu with a half-life of ~86 years begins to decrease after ~100 years and approaches to that of $^{242}$Cm, which has the same toxicity reduction factor with its precursor ($^{242m}$Am) after 10 years. So, after ~10,000 years, $^{238}$Pu, $^{242}$Cm, and $^{242m}$Am are also in a secular equilibrium.

From Figure 6.3, it is observed that the toxicity reduction factors of such isotopes that have shorter half-lives than their precursors approach to the toxicity reduction factors of their precursors as those decay out in early time ($^{238}$Pu, $^{241}$Pu, $^{241}$Am, $^{242}$Cm).

![Figure 6.4 Toxicity variation of TRU isotope with time after waste generation](image-url)
For isotopes that have longer half-lives than their precursors, two different patterns are observed: If the toxicity reduction factor of an isotope is greater than that of its precursor in early time, it keeps nearly constant or shows small variation with time (\(^{238}\text{U},^{231}\text{Np},^{239}\text{Pu},^{240}\text{Pu},\) and \(^{242}\text{Pu}\)). If the toxicity reduction factor of an isotope is less than that of its precursor in early time, it increases and approaches to the toxicity reduction factor of its precursor with time (\(^{234}\text{U},^{235}\text{U},\) and \(^{236}\text{U}\)). Once its toxicity reduction factor reaches to that of the precursor, it remains constant even after the precursor decays out as shown in the case of \(^{234}\text{U}\).

For isotopes with no precursors in this model, their toxicity reduction factors are constant with time (\(^{242m}\text{Am},^{243}\text{Am},^{243}\text{Cm},^{244}\text{Cm},^{245}\text{Cm},\) and \(^{246}\text{Cm}\)). Peaks or valleys shown in curves of \(^{241}\text{Pu},^{241}\text{Am},\) and \(^{242}\text{Cm}\) are graphical errors.

Figure 6.4 shows the toxicity variation of TRU isotope with time after waste generation. In the figure, the toxicity of TRU isotope in LWR-TRU is represented by a dotted line and that of ATW-TRU by a solid line.

For LWR-TRU, up to ~1,000 years the toxicity of \(^{241}\text{Am},\) between ~1,000 and ~10,000 years that of \(^{240}\text{Pu},\) and after ~10,000 years that of \(^{239}\text{Pu}\) is dominant. For ATW-TRU, until ~10 years the toxicity of \(^{244}\text{Cm},\) between ~10 and ~100 years that of \(^{238}\text{Pu}\) is dominant. However, within ~1,000 years most of Cm-isotopes and \(^{238}\text{Pu}\) decay out and between ~100 years and ~1,000 years \(^{241}\text{Am}\) becomes dominant in toxicity of ATW-TRU. Between ~1,000 and ~10,000 years \(^{240}\text{Pu}\) is most significant and after ~10,000 years major toxicity of ATW-TRU attributes to \(^{239}\text{Pu}\).

From this figure, it is expected that if we could reduce the mass of \(^{239}\text{Pu},^{240}\text{Pu},\) and \(^{241}\text{Am}\) further by the ATW transmuter, a larger toxicity reduction factor of the total TRU could be achieved after ~100 years. After ~100 years, the toxicity variations of LWR-TRU and ATW-TRU show a very similar pattern.

6.4 Discussions

In this chapter, the reduction of TRU inventory has been estimated based on the reference ATW plant deployment scenario. The reduction of TRU mass obtained in this analysis is smaller than the prediction (by factor of ~1,000) shown in the Roadmap [DOE/RW-0519, 1999]. Of the three TRU waste sources, TRU waste from ATW fuel processing was not estimated explicitly in the Roadmap as shown in Figure 1.2. According to the analysis of the ATW fuel cycle performed in the previous chapter, however, the amount of TRU waste which would be included in ATW separation waste is not negligible. It could be a major TRU waste source as estimated in Section 6.4.

The amount of TRU waste from ATW fuel processing could change if a different operating mode is applied for the performance analysis of the ATW fuel cycle. For example, the recovery fraction of TRU assumed in ATW fuel processing will be one of key parameters to the determination of the amount of TRU waste. However, for any case, it seems that a certain amount of TRU waste from ATW fuel separation process would be unavoidable. If TRU loss from ATW fuel fabrication process, which is assumed to be zero, is taken into account, more TRU waste would be generated.

The amount of TRU waste from the last transmuter could also be changed. Even though we assume that its operation is controlled by the reactivity constraint, it could be possible to allow more burnup for the last transmuter in its final operating cycle for the purpose of destructing TRU remaining in the core. It is also possible to modify the core
design, make it thermal or close to thermal, and thereby significantly reduce the TRU inventory needed for getting $k_{\text{eff}}$ of ~0.98. By doing so, we could reduce to some extent, but not completely, the amount of TRU waste from the decommissioning of the last transmuter.

For the estimation of the reduction of the toxicity of LWR-TRU, as mentioned in Section 6.4, by considering cooling time of ATW spent fuel before and during separation process, cooling time of TRU waste cumulated in waste stream, and the time required for refueling in each cycle it could be possible to get a greater reduction factor at the end of the reference ATW plant operation. However, in spite of the small toxicity reduction factor in early time domain due to aforementioned assumptions, we could get a profile describing a time-dependent variation of the toxicity reduction factor and in long-term period it provides us how much the radiological hazard of TRU isotope could be reduced by the reference ATW plant implementation.

6.5 Conclusions

In this study, we have investigated the performance of a reference ATW plant employing LBE-cooled subcritical transmuters as its transmuters.

The mass of TRU waste has been estimated to be 5.291 MT from the reference ATW system. But it should be noticed that the result shown here is a “snapshot” of one possible case out of infinite number of scenarios. If we use different operating conditions and make assumptions more realistically, the amount of TRU waste and its composition could be different from the results obtained here.

With respect to TRU reduction, the results show that the mass of LWR-TRU can be reduced by more than a factor of 170 and the toxicity of LWR-TRU can be decreased by more than a factor of 40 at the end of the reference ATW plant operation. After ~100 years since TRU waste generation, the overall toxicity reduction factor increases up to ~100 because most of Cm-isotopes (except $^{245}\text{Cm}$ and $^{246}\text{Cm}$) included in TRU waste decay out within that time period. After 10,000 years, it increases up to ~170 as $^{238}\text{Pu}$, $^{240}\text{Pu}$, and $^{241}\text{Am}$ decrease greatly while $^{239}\text{Pu}$ with a high toxicity reduction factor (~340) at that time begins to play a dominant role in the toxicity of TRU waste. So, in order to achieve a higher overall toxicity reduction factor in long term (after 10,000 years), $^{239}\text{Pu}$ should be transmuted more effectively.

The toxicity reduction factor is greater than the overall mass reduction factor, 170, for $^{239}\text{Pu}$, $^{237}\text{Np}$, and $^{241}\text{Am}$, which means that reduction of these isotopes could be done preferentially. This implies that the ATW deployment can decrease long-lived radionuclides, such as $^{237}\text{Np}$, which determine radiological impact of the repository in a preferential manner, compared to other TRU isotopes. After a successful operation of the reference ATW plant, both the inventory and the toxicity of $^{237}\text{Np}$ in LWR-TRU could be reduced by a factor of 281. In long-term analysis, the toxicity reduction factor of $^{237}\text{Np}$ decreases due to the decay of its precursor until the inventory of its precursor becomes negligibly small. In 1,000 years its value decreases up to 206 and after 10,000 years it almost remains constant as ~200.
7 Model for Conditioning of High-Level Waste

7.1 Introduction

From ATW fuel cycle, high-level radioactive wastes are to be generated mainly at a process that separates fission products from actinides. Such high-level wastes (HLW) from a separation process contain most of fission products and trace amounts of actinides usually in a liquid form. Before they are transferred to a geologic repository for final disposal, they need to be solidified. For solidification, liquid HLW is mixed with matrix materials such as borosilicate glass frit while satisfying many constraints. Figure 7.1 shows an ATW fuel cycle and HLW mass flow briefly.

The objective of this chapter is to develop a model to determine an optimized composition of solidified HLW for a given liquid HLW from a separation process and a given set of constraints for solidification. The model will also be able to determine a number of waste packages of given dimensions that would be generated by application of a separation process for a unit mass of irradiated fuel. These two outputs from the model, i.e., the composition of the solidified HLW and the number of packages per unit mass of processed irradiated fuel, are the key input data for repository impact analyses. The optimization is to be made by applying a linear-programming (LP) approach.

In Section 7.2, the process of HLW solidification and required considerations and regulations for US-defense HLW are reviewed. In Section 7.3, those regulations imposed on the solidification of defense HLW are converted into mathematical formulations in terms of two independent variables: the mass of HLW and that of glass frit. In Section 7.4, with derived formulations, a LP model is developed for HLW solidification and in Section 7.5, the solution methods of the LP problem are reviewed. In Section 7.6, the developed LP model is applied to a US-defense HLW and benchmarked. Discussion and conclusions for this chapter are followed in Section 7.7.

Figure 7.1 An ATW fuel cycle and HLW mass flow
7.2 Solidification of High-Level Waste (HLW)

7.2.1 Solidification Process of HLW

HLW mainly arises from separation processes of various nuclear fuel cycles and this contains most of the fission products, activation products and some of actinides. It is a reservoir of radioactivity and its hazard potential would last over a very long-term period. Therefore, a reliable technology for long-term isolation of HLW from the environment is a key to environmental protection against the consequences of nuclear power, and is also a key to the public acceptance of nuclear power [Benedict, et al., 1981].

The solidification of HLW is achieved by blending HLW with solidification materials such as glasses or ceramics, melting the mixture, and then pouring the melt into a solid container.

![Diagram of the HLW process flow](image)

**Figure 7.2 A simplified HLW process flow of the DWPF at the SRTC**

As an example, Figure 7.2 shows a simplified HLW process flow of the Defense Waste Processing Facility (DWPF) at the Savannah River Technology Center (SRTC) operated by the US-DOE [Bibler, et al., 2001]. The high level liquid waste stored in underground steel tanks at the SRTC is either a sludge of hydrous oxides or a nitrate salt slurry. The HLW sludge slurry is pretreated with nitric and formic acid to remove Hg and to adjust the rheology of the slurry in the Sludge Receipt and Adjustment Tank (SRAT) and then the sludge slurry is pumped to the Slurry Mix Evaporator (SME) where glass formers are added and the slurry thoroughly is mixed and concentrated to approximately 45 weight percent total solids. After sampling and confirming that the composition of the mixture meets process control requirements, the slurry is pumped to the Melter Feed Tank (MFT) which contains sufficient feed for eight canisters of glass, and then the mixture is pumped to the Melter operating at 1150°C. Finally, the melt is poured into a solid canister and then a filled canister is stored and cooled at an interim storage before its final disposal.
7.2.2 Requirements of Solidified Waste Products

7.2.2.1 General Considerations in HLW Solidification

A key feature of a solidified waste product is stability against destructive influences to which a highly radioactive solid may be exposed. The stability of the waste form provides an additional safety barrier in the geologic repository concept and thus the solidified waste product is supposed to be made as stable as practically achievable [Benedict, et al., 1981].

For HLW fraction in a solidification product, a higher fraction is desired on economic grounds and repository space availability. Costs of handling, packaging, and transportation are considerable and depend on the volume and the number of waste packages to be handled. In addition, less volume of waste products is favorable for the availability of repository space. On the other hand, the HLW concentration is limited by chemical reasons or by heat production reason. Chemical limitations are typical for glasses where either phase separations may occur or the product may not be a glass at all.

Fission products and actinides in any solidified HLW emit radiations by radioactive decay. Part of the irradiation energy is dissipated in elastic collisions with atoms from the solid material, thus displacing them and causing radiation damage. Among the possible consequences of radiation damage on solidified waste, energy storage is considered a potential risk. The temperature of the waste solid would suddenly rise if stored energy were released. Other possible effects of irradiation on the solidified waste are deterioration of mechanical properties and changes of volume due to radiation damage or as a consequence of helium formation from alpha decay.

Heat generation in the solidified waste causes a temperature elevation for more than 100 years. A cylindrical waste block with homogeneous heat source has a radial temperature gradient. Given the heat generation, the temperature difference between the surface and the centerline is a function of the heat conductivity of the material. A long-term effect promoted by high temperature is devitrification of glass and the devitrified product is less leach-resistant than the original glass. The temperature gradient will give rise to thermal stress in any monolithic material, which in turn may cause cracking. The stress at any point of a glass sample depends on the difference in temperature between this point and the average. So, the temperature difference needs to be controlled within a certain limit based on the thermal expansion coefficient of a solidified waste product.

The only chemical attack on the solidification products deserving serious consideration is leaching by water or brine, if such exist in the repository. Therefore, any solidified waste product should have a suitable leach resistance to water.

7.2.2.2 Specific Requirements for US-Defense HLW Solidification

Specific requirements may also be imposed on a certain type of HLW by a repository management system which is supposed to accept solidified waste products into its repository. Those could be concerned about a specific type of durability test, the concentration of a specific fissile material in a solidification product, criticality of a solidified product, radiation dose caused by radioactivity of a solidified waste, weight of a solidified waste product, geometry and material of a waste container and so on along with general considerations aforementioned. As an example of specific requirements, we

The US DOE is currently operating three facilities for the vitrification of HLW generated from a defense program: the West Valley Demonstration Project (WVDP), the Savannah River Site (Defense Waste Processing Facility, DWPF), and the Hanford Site. Those facilities have been producing vitrified HLW products before several years or preparing to vitrify HLW soon. Vitrified HLW produced at those facilities should satisfy specific requirements to be accepted into the Civilian Radioactive Waste Management System (CRWMS).

The US DOE developed the Waste Acceptance Product Specifications (EM-WAPS) which provides the technical specifications for the waste form producers to meet in order to ensure acceptance of their vitrified HLW. Followings are key specifications to be considered in our model developing [DOE/EM-0093, 1996].

**Waste Form Specifications**
- The waste form is borosilicate glass.
- The consistency of the waste form shall be demonstrated using the Product Consistency Test (PCT). For acceptance, the mean concentrations of lithium, sodium and boron in the leachate, after normalizing for the concentration in the glass, shall each be less than those of the benchmark glass described in the Environmental Assessment for selection of the DWPF waste form. Table 7.1 shows the DWPF-PCT Environmental Assessment limits.

**Table 7.1 Defense Waste Processing Facility PCT Environmental Assessment Limits [WSRC-IM-91-116-5, 1991]**

<table>
<thead>
<tr>
<th>Element</th>
<th>EA Limit [g/liter]</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>16.70</td>
</tr>
<tr>
<td>Na</td>
<td>13.35</td>
</tr>
<tr>
<td>Li</td>
<td>9.57</td>
</tr>
</tbody>
</table>

- Regarding the temperature control for phase stability, it shall be certified that after initial cool-down, the waste form temperature has not exceeded 400°C.

**Canister Specifications**
- The waste form canister, the canister label, and any secondary canister applied by the Producer shall be fabricated from austenitic stainless.
- The overall length of the unfilled canister, after accounting for the closure method, shall be 3.000 m (+0.005 m, -0.020 m), including the neck and lifting flange. The outer diameter of the unfilled canister shall be 61.0 cm (+1.5 cm, -1.0 cm).

**Canistered Waste Form Specifications**
- The Producer shall fill the canister to a height equivalent to at least 80% of the volume of the empty canister.
- The heat generation rate of each canistered waste form shall not exceed 1500 watts per canister at the year of shipment.
- The configuration, dimensions, and weight of the canistered waste form shall not exceed the maximum size and weight which can be received, handled, and emplaced in the repository. The weight of the canistered waste form shall not exceed 2,500 kg.
- The concentration of plutonium in each HLW standard canister shall be less than 2,500 g/m³.

7.3 Mathematical Formulation of Waste Acceptance Product Specifications

Specifications for defense HLW products summarized in Section 7.2 are formulated mathematically to be incorporated in linear programming model as follows.

7.3.1 Formulation of Product Consistency Tests (PCT) Limits

To incorporate the PCT limit of a waste glass product, we need an analytic model to predict glass dissolution rate under the same conditions that the PCT is subject to. To do so, we need to identify how the PCT is performed and to investigate how it can be described in a mathematical form.

7.3.1.1 Product Consistency Tests (PCT)

The PCT has been standardized by the American Society for Testing and Materials (ASTM) as standard test method C1285-02. Two types of the PCT, methods A and B, are available. Both evaluate the chemical durability of homogeneous glasses, phase separated glasses, devitrified glasses, glass ceramics, and/or multiphase glass ceramic waste forms by measuring the concentrations of the chemical species released to a test solution. For a consistency test for production of HLW glass, method A is used.

In test method A, the glass waste form is crushed and sieved to U.S. Standard ASTM-100 to +200 mesh (0.149-0.074 mm), the particles are cleaned of adhering fines, and an amount of sized and cleaned glass waste form that is greater than or equal to 1 g is placed in a Type 304L stainless steel vessel. An amount of ASTM Type I water equal to 10 ± 0.5 cm³/g of sample mass is added and the vessel is sealed. The vessel is placed in a constant temperature device at 90 ± 2°C. After 7 days (±3.4 h) the vessel is removed from the oven and cooled to ambient temperature. The remaining leachate is filtered and sent for analysis [ASTM C1285-02, 2002].

7.3.1.2 Analytic Model of PCT Method A

With the PCT method A, we can evaluate the concentration of a specific element dissolved in a solution during the test period under the test conditions. In order to predict it without performing the PCT method A, however, we need to introduce an analytic model which describes the behavior of glass dissolution under the conditions of the PCT method A. For this, we have investigated an analytic model, “Defense High-Level Waste Glass Degradation Model,” [CRWMS M&O 2000k, 2000] developed for radionuclide release from HLW glass and supposed to be integrated into Total System Performance Assessment (TSPA) calculations conducted to support site recommendation and license application for the Yucca Mountain site.

7.3.1.2.1 Abstracted Model for Defense HLW Glass Degradation

The dissolution rate of the glass imposes an upper bound on the radionuclide release rate because the release of radionuclides from the glass will depend on the prior dissolution of the glass. The approach taken to provide a bound for the radionuclide release is to develop models that can be used to calculate the dissolution rate of waste glass when contacted by water. The release rate of a particular radionuclide can then be calculated by multiplying the glass dissolution rate by the mass fraction of that
radionuclide in the glass. This approach for calculating the radionuclide release rate is based on the conservative assumption that the release of radionuclides is congruent with the degradation of borosilicate glass [CRWMS M&O 2000k, 2000; CRWMS M&O, 2000].

The glass dissolution model is implemented in the form of an analytical expression containing four parameters and two variables. The model parameters account for the pH, temperature, surface area, and the combined effects of glass composition and solution composition on the rate of glass corrosion. Conservative estimates of the values for the model parameters are provided based on experimental data. Implementation of the model for the TSPA analysis requires input of temperature and pH data.

Available experimental data concerning the degradation behavior of borosilicate-based glasses were analyzed to develop models for the dissolution and consequent radioisotope release from HLW glass under aqueous immersion conditions in the repository environment. The basic form of rate equation that describes dissolution of waste glass immersed in water is given in Section 6 of CRWMS M&O 2000k:

\[
Rate = S \cdot k_o \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a / RT) \cdot [1 - (Q / K)]
\]

where

- Rate = the dissolution rate of the glass, in units of mass/time,
- S = the surface area of glass immersed in water, in units of area,
- \( k_o \) = the intrinsic dissolution rate, which depends only on glass composition, in units of mass/(area-time),
- \( \eta \) = the pH dependence coefficient, which is dimensionless,
- \( E_a \) = the effective activation energy, in units of kJ/mol,
- \( R \) = the gas constant, which is 8.314×10^{-3} kJ/(mol-K),
- \( T \) = the temperature, in kelvins,
- \( Q \) = the concentration of dissolved silica in the solution, in units of mass/volume,
- \( K \) = a quasi-thermodynamic fitting parameter equal to the apparent silica saturation concentration for the glass, in units of mass/volume.

The rate expression contains two main parts: the forward rate, \( k_o \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a / RT) \), which represents the dissolution rate in the absence of feedback effects of dissolved silica, and the reaction affinity term, \((1 - Q / K)\), which quantifies the feedback effects. Because the value of \( Q \) can range between zero and \( K \), the value of the affinity term is mathematically constrained to values between one and zero. A glass will dissolve at the highest rate possible (at a given temperature and pH value) when the value of the affinity term is one (i.e., when \( Q = 0 \)). The dissolution rate will decrease as the value of the affinity term decreases (i.e., as the value of \( Q \) approaches \( K \)) until a minimum rate is reached.

Application of this rate expression requires knowledge of several parameter values that depend on the glass composition \((k_o, \eta, E_a, S, K)\) and of several variables related to the exposure conditions \((pH, T, Q)\). Evaluation of \( Q \) would be complex and subject to considerable uncertainty. A simpler bounding approach was, therefore, adopted. In this approach the affinity term \((1 - Q / K)\) was combined with the intrinsic dissolution rate constant, \( k_o \), to generate the following abstraction for aqueous degradation of HLW for the TSPA-SR analysis:

\[
Rate / S = k_{ef} \cdot 10^{\eta \cdot pH} \cdot \exp(-E_a / RT)
\]

where

\[
k_{ef} = k_o (1 - Q / K)
\]
Although \( k_{ef} \) is a complex function of glass composition and solution variables, the approach adopted in the abstraction is to provide a constant value for \( k_{ef} \) that conservatively bounds the anticipated range of the function \( k_{ef}(1-Q/K) \). This approach reduces the abstracted model to an equation involving four parameters (\( \eta, E_a, S, \) and \( k_{ef} \)) and two variables (pH and \( T \)). Available experimental data were used to obtain conservative estimates of each of these parameters given in Section 6 of CRWMS M&O 2000k.

The first step to estimate the model parameters is to analyze the dependence of the forward rate on the glass composition, pH, and temperature. These dependencies were measured under flow-through test conditions in which the value of the affinity term is maintained near one. On the basis of experimental results, the pH and temperature dependence of the forward rates were found to be independent of the glass composition, within the composition range of the glass tested, so the same values for the \( \eta, \) and \( E_a \) parameters are used for all waste glasses. The values of these parameters do, however, depend on pH. Analysis of the available data in Section 6.2 of CRWMS M&O 2000k resulted in the empirical estimates for values of the \( \eta \) and \( E_a \) parameters for acidic and basic conditions.

In the second step, a bounding value for the \( k_{ef} \) parameter was determined based on experimental observations of the behavior of the affinity term for test conditions that spanned a broad range of reaction progress. Experimental data show that the behavior of the corrosion rate with reaction progress can be characterized as proceeding through three stages.

- **Stage I** occurs when the value of affinity term is one and glass dissolves at the forward rate for the specific temperature and pH conditions involved.
- **Stage II** occurs as the value of the affinity term decreases significantly due to the accumulation of glass components particularly dissolved silica, in solution. The value of affinity term (and thus the dissolution rate) cannot become zero because glass is thermodynamically unstable and cannot equilibrate with solution.
- **Stage III** occurs when the dissolution rate increases due to the formation of alteration phases, effectively removing dissolved silica from the solution. The formation of alteration phases is believed to cause a decrease in the value of \( Q \) due to the consumption of dissolved silica as these silica-bearing phases form in Section 6.2.2 of CRWMS M&O 2000k.

This stage III behavior has not been observed for all glasses or all experimental conditions and the mechanisms involved remain unclear. It is not possible, therefore, to predict when Stage III occurs or to unambiguously predict the value of \( k_{eff} \) for use in performance assessment calculations. For these reasons a conservative bounding approach was adopted in evaluating \( k_{ef} \). After evaluating several options, an approach based on using the ASTM Product Consistency Test, method A (PCT-A) test results was selected in Section 6.2.3 of CRWMS M&O 2000k. This approach involved using the relatively short-term dissolution rate test results to obtain bounding values for \( k_{ef} \) that would conservatively encompass all three potential stages of dissolution behavior.

The abstracted bounding rate expressions are obtained when the estimated value for the model parameters are substituted into Eq. (7.2). The result is as follows:

For the low pH leg (pH<\( p_{H_m} \)),

--
\[
Rate / S = 10^{(0 \pm 1)} \cdot 10^{(-0.6 \pm 0.1) \cdot pH} \cdot \exp[-(58 \pm 15) / RT]
\]

(7.4)

For the high pH leg (pH≥pH_m),
\[
Rate / S = 10^{(6.9 \pm 0.5)} \cdot 10^{(0.4 \pm 0.1) \cdot pH} \cdot \exp[-(80 \pm 10) / RT]
\]

(7.5)

where, pH_m = the pH at which the minimum dissolution rate occurs = 2.1+1149/T

7.3.1.2.2 Application of Abstracted Model for PCT-A

With the abstracted model, we can estimate the bounding glass dissolution rate of PCT-A for a certain pH and temperature because conservatively estimated parameter values are based on the PCT-A experimental results. The temperature of PCT-A is kept constant at 90°C over the test duration (7 days) but the pH value of PCT-A solution varies with waste glass compositions. If we assume an extreme pH value, say pH=1 or pH=12, the estimated glass dissolution rate will be bounding the entire low pH leg region (pH<pH_m) or the entire high pH leg region (pH≥pH_m) because the glass dissolution rate increases as the pH value gets farther from pH_m.

For pH = 1 and T = 90°C, bounding dissolution rate of glass, Rate/S,
\[
Rate / S = 10^{(10)} \cdot 10^{(-0.5) \cdot (1)} \cdot \exp[-(73) / (8.314 \times 10^{-3}) (363)] = 0.10 [g / m^2 \cdot day]
\]

(7.6)

For pH = 12 and T = 90°C, bounding dissolution rate of glass, Rate/S,
\[
Rate / S = 10^{(7.4)} \cdot 10^{(0.5) \cdot (12)} \cdot \exp[-(90) / (8.314 \times 10^{-3}) (363)] = 2.81 [g / m^2 \cdot day].
\]

(7.7)

Thus, 2.81 [g/m^2-day] is the bounding dissolution rate of glass for pH=12 and T=90°C, and can be the bounding dissolution rate of PCT-A for the potential waste glasses over the entire pH range. Considering that the total surfaces area of 1g of glass particles sieved for PCT-A is ~1.99\times10^{-2} m^2, the equivalent ASTM type I water volume to 1 g of glass particles is 10^{-2} l (10 cm^3), and the test duration is 7 days [ASTM C 1285-02, 2002], the bounding concentration of the dissolved glass in the solution resulting from PCT-A would be 39.14 [g/l]. If we let the mass fraction of a specific element i in waste glass x_i the bounding concentration of the element in the solution would be the product of x_i and the bounding concentration of the dissolved glass (39.14 [g/l]). Therefore, EA limit for PCT-A specified in EM-WAPS can be formulated as follows:

For boron dissolution limit, from Table 7.1,
\[
39.14 x_B \leq 16.70 \ [g/liter],
\]

(7.8)

where x_B = the mass fraction of boron in a solidified waste glass.

Because the mass fraction of nuclide i in a solidified waste glass, x_i, is expressed in terms of the masses of a HLW and a matrix material (i.e., glass) and their compositions such as
\[
x_i = \frac{M_w x_{w,i} + M_G x_{G,i}}{M_w + M_G},
\]

(7.9)

Equation (7.8) can be rewritten as
\[
39.14 \left( \frac{M_w x_{w,B} + M_G x_{G,B}}{M_w + M_G} \right) \leq 16.70 \ [g/liter],
\]

(7.10)
where $M_W$ = the mass of a dried HLW sludge, [kg]  
$M_G$ = the mass of a matrix material (i.e., glass), [kg]  
$x_{W,i}$ = the mass fraction of nuclide $i$ in a dried HLW sludge,  
$x_{G,i}$ = the mass fraction of nuclide $i$ in a matrix material,  
$x_{W,B}$ = the mass fraction of boron in a dried HLW sludge,  
$x_{G,B}$ = the mass fraction of boron in a matrix material.

For sodium dissolution limit, from Table 7.1,

\[ 39.14 x_{Na} \leq 13.35 \text{ [g/liter]}, \quad (7.11) \]

where $x_{Na}$ = the mass fraction of sodium in a solidified waste glass.  
By replacing $x_{Na}$ such in (7.9), (7.11) is

\[ 39.14 \left( \frac{M_W x_{W,Na} + M_G x_{G,Na}}{M_W + M_G} \right) \leq 13.35, \quad (7.12) \]

where $x_{W,Na}$ = the mass fraction of sodium in a dried HLW sludge,  
$x_{G,Na}$ = the mass fraction of sodium in a matrix material.

For lithium dissolution limit,

\[ 39.14 x_{Li} \leq 9.57 \text{ [g/liter]}, \quad (7.13) \]

where $x_{Li}$ = the mass fraction of lithium in a solidified waste glass.  
By replacing $x_{Li}$ such in (7.9), (7.13) is

\[ 39.14 \left( \frac{M_W x_{W, Li} + M_G x_{G, Li}}{M_W + M_G} \right) \leq 9.57, \quad (7.14) \]

where $x_{W, Li}$ = the mass fraction of lithium in a dried HLW sludge,  
$x_{G, Li}$ = the mass fraction of lithium in a matrix material.

### 7.3.2 Formulation of Temperature Limit

#### 7.3.2.1 Temperature Distribution in a Cylinder Block with Heat Sources

To perform heat transfer analysis for a medium, three types of heat transfer modes are considered: conduction, convection and radiation heat transfer modes [Holman, 1990]. Temperature distribution in a medium resulting from conditions imposed on its boundaries can be determined by the heat transfer analysis. If a cylinder with uniformly distributed heat sources has a cladding with no heat sources such like a type of canister containing HLW and boundary conditions are $T = T_i$ at $r = r_i$ (interface) and $T = T_o$ at $r = r_o$ (outside surface), a general form of the heat-conduction equation in the cylinder is expressed as

\[ \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{1}{r^2} \frac{\partial^2 T}{\partial \phi^2} + \frac{\partial^2 T}{\partial z^2} + \frac{q}{k_r} = \frac{1}{\alpha} \frac{\partial T}{\partial t}, \quad (7.15) \]

where $T$ = the temperature at a location $(r, \phi, z)$ in a cylinder at a time $t$, [°C],  
$r$ = the distance from the origin of cylindrical coordinate in radial direction, [m],  
$\phi$ = the azimuthal angle,  
z = the distance from the origin of cylindrical coordinate in vertical direction, [m],
\( \dot{q} \) = the energy generated per unit volume, [W/m\(^3\)],

\( k_g \) = the thermal conductivity of a cylinder, which is assumed as a constant, [W/m\(^2\)K],

\( \alpha \) = the thermal diffusivity of a cylinder material, [m\(^2\)/sec],

\( t \) = the time, [sec].

If the cylinder is sufficiently long that the temperature may be considered a function of radius only, (7.15) is simplified as,

\[
\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\dot{q}}{k_g} = 0, \quad 0 < r \leq r_i. \tag{7.16}
\]

Applying additional boundary condition at the center line of a cylinder,

\[
\frac{dT}{dr}{|_{r=0}} = 0, \tag{7.17}
\]

we can obtain the solution of (7.16)

\[
T(r) = -\frac{\dot{q}}{4k_g} r^2 + \frac{\dot{q}}{4k_g} r_i^2 + T_i. \tag{7.18}
\]

The relation between the temperature, \( T_m \), at the center line of a cylinder \((r = 0)\) and \( T_i \) is

\[
T_m - T_i = \frac{\dot{q}}{4k_g} r_i^2. \tag{7.19}
\]

### 7.3.2.2 Temperature Distribution in Canister Wall without Heat Sources

The steady-state heat-conduction equation in cladding region with no heat sources is expressed [Incropera, et al., 1981] as

\[
\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} = 0. \tag{7.20}
\]

The solution of Eq. (7.20) can be either

\[
T(r) = \frac{(T_i - T_o)}{\ln(r_i / r_o)} \ln(r / r_i) + T_i \tag{7.21}
\]

or

\[
T(r) = \frac{(T_i - T_o)}{\ln(r_i / r_o)} \ln(r / r_o) + T_o. \tag{7.22}
\]

Now we have the temperature profiles in both waste glass with uniformly distributed heat sources and in cladding with no heat sources. The relation between the temperature, \( T_i \), at the waste glass-cladding interface and the temperature, \( T_o \), at the outside surface of cladding can be obtained by heat transfer balance at the interface. The heat passing out of a waste glass with heat sources is balanced by the heat passing through the cladding with no heat sources at the interface. The heat passing out of a cylinder with heat sources, \( q(r) \), is
\[ q(r) = \pi r^2 \dot{q} \]  

(7.23)

and at the interface \((r = r_i)\)

\[ q(r = r_i) = \pi r_i^2 \dot{q}. \]  

(7.24)

The heat passing through the surface area \(A\) in a medium with thermal conductivity \(k\)

\[ q(r) = -k A \frac{dT}{dr}, \]  

(7.25)

for the cladding of a cylindrical waste glass whose thermal conductivity is represented by \(k_c\),

\[ q(r) = -k_c 2\pi r L \frac{dT}{dr}. \]  

(7.26)

From (7.21)

\[ \frac{dT}{dr} \bigg|_{r=r_i} = \frac{T_i - T_o}{\ln(r_i / r_o)} \frac{1}{r_i}, \]  

(7.27)

So, (7.26) is

\[ q(r = r_i) = -\frac{2\pi L k_c (T_i - T_o)}{\ln[r_i / r_o]} . \]  

(7.28)

Letting \(r_o = r_i + c\), where \(c\) is the thickness of the cladding, if \(c \ll r_i\), Eq. (7.28) is approximated as

\[ q(r = r_i) \approx \frac{2\pi r_i L k_c (T_i - T_o)}{c}. \]  

(7.29)

Heat transfer balance at the interface requires that the heat passing out of a cylinder equals the heat passing through the cladding region:

\[ \pi r_i^2 \dot{q} = \frac{2\pi r_i L k_c (T_i - T_o)}{c}. \]  

(7.30)

Therefore, we obtain the relation between \(T_i\) and \(T_o\)

\[ T_i - T_o = \frac{cr_i \dot{q}}{2k_c}. \]  

(7.31)

To obtain the relation between the temperature at the outside surface of cladding \(T_o\) and the ambient temperature \(T_\infty\) we use heat transfer balance at the outside surface of cladding. At the outside surface region, the heat passing out of the cladding surface equals the heat passing through moving fluid surrounding a cylinder. Heat transfer through a moving fluid is done by convection and radiation heat transfer mechanism but here we only consider convection heat transfer which is the dominant heat transfer mode over radiation heat transfer mode. This gives us an overestimate temperature profile in a cylinder which is considered as a conservative estimate. To express the overall effect of convection, we use Newton’s law of cooling [Incropera, et al., 1981]:

69
\[ q = \bar{h} A (T_o - T_\infty), \quad (7.32) \]

where \( q \) = the heat transfer rate through a surface area \( A \), [W]

\[ \bar{h} = \text{the average convection heat-transfer coefficient, defined as } \bar{h} = \frac{1}{L} \int_0^L h(z) dz, \]

[W/m\(^2\)-K].

The convection heat-transfer coefficient has a dependence on the viscosity of the fluid in addition to its dependence on the thermal properties of the fluid (thermal conductivity, specific heat, density). This is because viscosity influences the velocity profile and, correspondingly, the energy-transfer rate in the region near wall. Here, instead of estimating the average heat-transfer coefficient by integrating along the vertical plate based on the conditions that produced waste packages are stored in an approximated equation of the average heat-transfer coefficient for free convection from a vertical plane surface to air at atmospheric pressure and moderate temperatures given in [Holman, 1990] are used. For laminar air flow, the approximated equation for the average heat-transfer coefficient is

\[ \bar{h} = 1.42 \left( \frac{T_o - T_\infty}{L} \right)^{1/4}, \quad (7.33) \]

for turbulent air flow, the approximated equation for the average heat-transfer coefficient is

\[ \bar{h} = 1.3 (T_o - T_\infty)^{1/3}. \quad (7.34) \]

Preliminary estimation for air flow surrounding a vertical plane 3 m in height shows that the type of air flow lies in near the boundary of laminar and turbulent region. It is known that the temperature profile inside a cylinder can be estimated more conservatively by using the approximation for laminar flow. So, at the surface of the cladding of a cylinder,

\[ q = 2.84 \pi r_o L^{3/4} (T_o - T_\infty)^{5/4}. \quad (7.35) \]

By the heat transfer balance at the surface area of the cladding,

\[ \frac{2 \pi r_i L k_c (T_i - T_o)}{c} = 2.84 \pi r_o L^{3/4} (T_o - T_\infty)^{5/4}, \quad (7.36) \]

and thus the relation between \( T_o \) and \( T_\infty \) is

\[ T_o - T_\infty = \left[ k_c r_i L^{1/4} \right] \left( \frac{1.42 r_o c}{r_i} \right)^{4/5} \left( \frac{r_i^2 L^{1/4}}{2.84 r_o} \right)^{4/5}. \quad (7.37) \]

The relation between \( T_m \) and \( T_\infty \) are obtained by addition of (7.19), (7.31), and (7.37):

\[ T_m - T_\infty = q \left( \frac{r_i^2}{4 k_g} + \frac{c r_i}{2 k_c} \right) + q^{4/5} \left( \frac{r_i^2 L^{1/4}}{2.84 r_o} \right)^{4/5}. \quad (7.38) \]

Now, we need to check the deviation of the average heat-transfer coefficient estimated from a vertical flat plate from that from a slender cylinder. Cebeci [1974]
provides experimental results on laminar free convection heat transfer from the outer surface of a vertical slender circular cylinder and the deviation of the average Nusselt number of a slender cylinder from that of a flat plate for various values of Prandtl number. For a cylinder with 3 m in height and 0.3 m in radius and air with Prandtl number of 0.72, the ratio of the average heat-transfer coefficient of a slender cylinder to that of a flat plate is estimated ~1.018. (the temperature of a slender cylinder was assumed to be 105°C).

For the maximum temperature limit specified in EM-WAPS, Eq. (7.38) is expressed as

\[
\dot{q} \left( \frac{r_i^2}{4k_g} + \frac{cr_i}{2k_c} \right) + \dot{q}^{4/5} \left( \frac{r_i^2 L^{1/4}}{2.84r_o} \right)^{4/5} + T_\infty \leq 400^\circ C.
\] (7.39)

### 7.3.3 Formulation of Heat Generation Limit

HLW contains fission products, actinides, and activation products generated from reactor operations, so it generates decay heat. Decay heat from those nuclides can be calculated by summing the products of the activity of each nuclide and the mean energy release per disintegration of that nuclide. The total decay heat \( q \) from the inventories of fission products, actinides, activation products is then given by [Oyamatsu, et al., 2001]:

\[
q = \sum_i CE_i \lambda_i N_i \ [W],
\] (7.40)

where \( C \) = conversion factor, 1.602×10^{-13} [W/MeV],
\( E_i \) = the mean energy release from the disintegration of nuclide \( i \), [MeV]
\( \lambda_i \) = the decay constant of nuclide \( i \), [1/sec]
\( N_i \) = the number of nuclide \( i \).

If the composition of a HLW is used, Eq. (7.40) can be expressed as

\[
q = \sum_i CE_i \lambda_i \frac{x_{W,i}}{A_i} N_A M_W
\] (7.41)

where \( x_{W,i} \) = the weight fraction of radionuclide \( i \) in HLW sludge,
\( A_i \) = the mass of 1 mol of radionuclide \( i \), [kg],
\( N_A \) = Avogadro’s number = 6.023×10^{23} [#/mol],
\( M_W \) = the mass of HLW, [kg].

For the heat generation limit in EM-WAPS, Eq. (7.41) is

\[
\zeta M_W \leq 1500 \ [W], \text{ where } \zeta = CN_A \sum_i E_i \lambda_i x_{W,i} / A_i.
\] (7.42)

### 7.3.4 Formulation of Solidified Waste Glass Volume Limit

The volume of a solidified waste poured into a canister is limited by the volume of an empty canister and by the minimum filling guideline, thus this can be formulated as

\[
V_{\text{min}} \leq V_{WG} \leq V_{\max}.
\] (7.43)

where \( V_{\text{min}} \) = the minimum volume limit of a solidified waste glass, [m^3],
\( V_{WG} \) = the volume of a solidified waste glass, [m^3].
\[ V_{\text{max}} = \text{the maximum volume limit of a solidified waste glass,} \ [m^3]. \]

The volume of a solidified waste glass consists of the volume of a HLW sludge and that of a glass frit and the minimum (or maximum) volume of a solidified waste glass can be expressed as the fractional volume of a canister.

\[ aV_C \leq \delta(V_W + V_G) \leq V_C , \] (7.45)

where \( a = \text{the minimum volume fraction, where} \ 0 \leq a \leq 1 , \)

\( V_C = \text{the volume of an empty canister,} \ [m^3], \)

\( V_W = \text{the volume of a dried HLW sludge,} \ [m^3], \)

\( V_G = \text{the volume of a glass frit,} \ [m^3], \)

\( \delta = \text{the volume reduction factor, which accounts for nonlinearity in mixing two different materials.} \)

The volumes of a HLW sludge and a glass frit can be rewritten in terms of their masses and densities as follow:

\[ aV_C \leq \delta \left( \frac{M_W}{\rho_W} + \frac{M_G}{\rho_G} \right) \leq V_C , \] (7.46)

where \( M_W = \text{the mass of a dried HLW sludge,} \ [kg], \)

\( M_G = \text{the mass of a glass frit,} \ [kg], \)

\( \rho_W = \text{the density of a dried HLW sludge,} \ [kg/m^3], \)

\( \rho_G = \text{the density of a glass,} \ [kg/m^3]. \)

For the solidified waste glass in EM-WAPS, Eq. (7.46) is

\[ 0.8V_C \leq \delta \left( \frac{M_W}{\rho_W} + \frac{M_G}{\rho_G} \right) \leq V_C . \] (7.47)

7.3.5 Formulation of Filled Canister Weight Limit

The weight limit of a solidified waste package can be formulated as

\[ M_W + M_G + M_C \leq M_{\text{max}} \ [kg], \] (7.48)

where \( M_C = \text{the mass of an empty waste package, which is assumed to be known,} \ [kg], \)

\( M_{\text{max}} = \text{the mass limit of a filled waste package,} \ [kg]. \)

For the filled waste package in EM-WAPS, Eq. (7.48) is

\[ M_W + M_G + M_C \leq 2500 \ [kg]. \] (7.49)

7.3.6 Formulation of Plutonium Concentration Limit

The concentration limit of a specific nuclide in a canister can be expressed as

\[ \frac{m_i}{V_C} \leq c_i \ [kg/m^3], \] (7.50)

where \( m_i = \text{the mass of a nuclide} \ i \ \text{in a solidified waste package,} \ [kg] \)

\( c_i = \text{the concentration limit of a nuclide} \ i \ \text{in a solidified waste package,} \ [kg/m^3]. \)

The mass of a nuclide \( i \) in a solidified waste package is the sum of the mass of a nuclide \( i \) in a HLW sludge and that in a glass frit, Eq. (7.50) can be written as
\[
\frac{M_W x_{W,i} + M_G x_{G,i}}{V_C} \leq c_i \text{ [kg/m}^3\text{]},
\]
where \( x_{W,i} \) = the mass fraction of a nuclide \( i \) in a HLW sludge,
\( x_{G,i} \) = the mass fraction of a nuclide \( i \) in a glass frit.

For the plutonium concentration limit in EM-WAPS, Eq. (7.51) is
\[
\frac{M_W x_{W,\text{Pu}}}{V_C} \leq 2.5 \text{ [kg/m}^3\text{]},
\]
where \( x_{W,\text{Pu}} \) = the mass fraction of plutonium in a HLW sludge.

7.4 Linear Programming Model for HLW Conditioning

In this section a mathematical model is developed, with which an optimized composition of a solidified waste product and the number of solidified waste packages are obtained based on the masses of a HLW and a glass frit and their compositions.

The mass, \( M_S \), of the solidified waste in a waste package is the sum of \( M_W \) and \( M_G \).

The composition vector, \( \tilde{N}_S \), of the solidified waste product can be calculated as
\[
\tilde{N}_S = \frac{M_W}{M_W + M_G} \tilde{N}_W + \frac{M_G}{M_W + M_G} \tilde{N}_G,
\]
where \( \tilde{N}_W \) is the composition vector of a HLW and \( \tilde{N}_G \) is the composition vector of a matrix material.

The number of solidified waste packages from the amount of HLW generated from a unit mass of irradiated fuel can be calculated by dividing the mass of the HLW generated by processing a unit mass of irradiated fuel by the mass \( M_W \) of HLW in a waste package.

Thus, if the composition vectors of a HLW and a matrix material are known, both the composition of a solidified waste product and the number of solidified waste products can be estimated by determination of two variables, \( M_W \) and \( M_G \).

To determine the two variables, \( M_W \) and \( M_G \), an optimization technique is employed. HLW solidification process can be illustrated by maximizing the waste loading into a waste package because by doing so, the number of waste products produced from a certain amount of HLW is minimized and in turn it can affect the availability of a repository space favorably. Also, as formulated in the previous sections, most constraints for the HLW solidification process and the solidified waste glass are expressed in the form of inequalities in a linear manner. \( M_W \) and \( M_G \) can be determined by solving a linear programming problem which describes an optimum waste loading in a waste package.

The linear programming problem consists of an objective function and several constraints. The objective function, \( f \), which is the mass \( M_W \) of HLW species in a waste package, for the HLW conditioning is formulated as
\[
\text{Maximize} \ f = M_W.
\]

This objective function is subject to constraints that are imposed on the HLW conditioning process. A system of constraint inequalities is expressed by
\[
A y \leq b \text{ and } y \geq 0
\]
where \( \mathbf{y} \) is the column vector of independent variables. In the present model, the vector consists of two components, \( M_W \) and \( M_G \). The vector \( \mathbf{b} \) is the column vector of constants, which are made up of the numbers of right-hand side of the inequalities, and \( \mathbf{A} \) is the matrix of coefficients of constraint inequalities.

To build matrix \( \mathbf{A} \) and vector \( \mathbf{b} \), we need to rewrite the formulations of the EM-WAPS developed in the previous section in a linear form.

- For PCT limit, the formulation of boron dissolution limit in Eq. (7.10) can be rewritten as
  \[
  M_W (x_{W,B} - 0.43) + M_G (x_{G,B} - 0.43) \leq 0,
  \]
  the formulation of sodium dissolution limit in Eq. (7.12) is rewritten as
  \[
  M_W (x_{W,Na} - 0.34) + M_G (x_{G,Na} - 0.34) \leq 0,
  \]
  and the formulation of lithium dissolution limit in Eq. (7.14) is rewritten as
  \[
  M_W (x_{W,Li} - 0.25) + M_G (x_{G,Li} - 0.25) \leq 0.
  \]

- For heat generation limit,
  \[
  \zeta M_W \leq 1500 \ [\text{W}].
  \]

- For temperature limit, heat generation rate per unit volume can be expressed as
  \[
  \dot{q} = \frac{q}{V_C} = \zeta M_W \frac{1}{V_C},
  \]
  and thus Eq. (7.39) is expressed as
  \[
  \alpha M_W + \beta (M_W) \frac{4/5}{+ T_x} \leq 400^\circ \text{C},
  \]
  where
  \[
  \alpha \equiv \left( \frac{\zeta}{V_C} \right) \left( \frac{r_i^2 + cr_i}{4k_g} + \frac{cr_i}{2k_c} \right), \quad \beta \equiv \left( \frac{\zeta}{V_C} \right) \left( \frac{r_i^2 L^{1/4}}{2.84 r_o} \right)^{4/5}.
  \]
  Eq. (7.60) is of a non-linear form but to construct LP problem it is required for Eq. (7.60) to be in a linear form. By assuming \( M_W \sim (M_W)^{4/5} \), Eq. (7.60) can be approximated in a linear form as
  \[
  (\alpha + \beta)M_W \leq 400 - T_x \ ^\circ \text{C}.
  \]
  It is observed that the approximated form, (7.61), gives a conservative temperature profile than (7.60).

- For solidified waste volume limit, Eq. (7.47) can be separated into two inequalities:
  \[
  0.8V_C \leq \frac{\delta}{\rho_W} M_W + \frac{\delta}{\rho_G} M_G,
  \]
  \[
  \frac{\delta}{\rho_W} M_W + \frac{\delta}{\rho_G} M_G \leq V_C.
  \]

- For filled canister weight limit,
  \[
  M_W + M_G \leq 2500 - M_C.
  \]

- For plutonium concentration limit,
Now, the matrix $A$ and the vector $b$ are written as follows.

$$A = \begin{bmatrix}
    x_{W,B} - 0.43 & x_{G,B} - 0.43 \\
    x_{W,Na} - 0.34 & x_{G,Na} - 0.34 \\
    x_{W,LI} - 0.25 & x_{G,LI} - 0.25 \\
    \alpha + \beta & 0 \\
    \zeta & 0 \\
    -\delta / \rho_W & -\delta / \rho_W \\
    \delta / \rho_W & \delta / \rho_W \\
    1 & 1 \\
    x_{W,PU} & 0
\end{bmatrix}, \quad
b = \begin{bmatrix}
    0 \\
    0 \\
    400 - T_c \\
    1500 \\
    -0.8V_c \\
    V_c \\
    2500 - M_C \\
    2.5V_C
\end{bmatrix}$$

(7.66)

### 7.5 Solution Methods for Linear Programming Problem

#### 7.5.1 Graphical Solution Method

As a solution method of linear programming (LP) problem, a graphical procedure can be used for solving problems which have two or three decision variables. For a two dimensional problem, the solution space is defined as a plane with $y_1 (=M_G)$ measured along the abscissa and $y_2 (=M_W)$ along the ordinate. Because they are linear, the constraints can be plotted on this plane as straight lines. If the linear programming problem was formulated properly (that is, it has a solution), these constraint lines will delineate a region, called the feasible solution space, encompassing all possible combinations of $y_1$ and $y_2$ that obey the constraints and hence represent feasible solutions. The objective function for a particular value of $f$ can then be plotted as another straight line and superimposed on this space. The value of $f$ can then be adjusted until it is at the maximum value while still touching the feasible space. This value of $f$ represents the optimum solution. The corresponding values of $y_1$ and $y_2$, where $f$ touches the feasible solution space, represent the optimal values of decision variables [Hiller, et al., 1990].

Because they are limited two or three dimensions, graphical solutions have limited practical utility. However, they are very useful for demonstrating some basic concepts that underlie the general algebraic techniques used to solve higher-dimensional problems with the computer.

#### 7.5.2 The Simplex Method

For solving LP problems, although graphical solutions are useful for a two or three dimension problem, most applications involve numerous variables and many constraints. Therefore, other methods must be used to analyze these models. A frequently used method is the simplex method. The method itself is an algorithmic procedure in which the optimal solution to a linear programming problem is found after the steps of the algorithm have been a finite number of times [Wu, et al., 1981].

As the first step to apply the simplex method for solving a linear programming problem, the original form of the linear programming problem which is in inequality
form is converted into an equality form by introducing slack (or surplus) variables. Therefore, if we have the following original form of an LP problem:

\[ \text{Maximize } f = cy, \text{ subject to } Ay \leq b \text{ and } y \geq 0. \]  

(7.67)

the inequality system can be converted as follows:

\[ [A \ I] \begin{bmatrix} y \\ y_s \end{bmatrix} = b \text{ and } y, y_s \geq 0, \]  

(7.68)

where \( c \) is the row vector

\[ c = [c_1, c_2, \ldots, c_n], \]  

(7.69)

\( y, y_s, b, \) and \( \theta \) are the column vectors such that

\[ y = \begin{bmatrix} y_1 \\ y_2 \\ \cdot \cdot \cdot \\ y_n \end{bmatrix}, \quad y_s = \begin{bmatrix} y_{n+1} \\ y_{n+2} \\ \cdot \cdot \cdot \\ y_{n+m} \end{bmatrix}, \quad b = \begin{bmatrix} b_1 \\ b_2 \\ \cdot \cdot \cdot \\ b_m \end{bmatrix}, \quad 0 = \begin{bmatrix} \cdot \\ \cdot \\ \cdot \cdot \cdot \\ \cdot \end{bmatrix}. \]  

(7.70)

\( A \) is the matrix of coefficients of constraint equations,

\[ A = \begin{bmatrix} a_{11} & a_{12} & \cdot \cdot \cdot & a_{1n} \\ a_{21} & a_{22} & \cdot \cdot \cdot & a_{2n} \\ \cdot \cdot \cdot & \cdot \cdot \cdot & \cdot \cdot \cdot \\ a_{m1} & a_{m2} & \cdot \cdot \cdot & a_{mn} \end{bmatrix}, \]  

(7.71)

and \( I \) is the \( m \times m \) identity matrix. In the augmented form of the linear programming problem, introduced slack (or surplus) variables is expressed as the column vector, \( y_s \).

The general approach of the simplex method is to obtain a sequence of improving basic feasible solutions until an optimal solution is reached. One of the key features of the simplex method involves the way in which it solves for each new basic feasible solution after identifying its basic (non-zero values) and nonbasic variables (zero values). Given these variables, the resulting basic solution is the solution of the \( m \) equations

\[ [A \ I] \begin{bmatrix} y \\ y_s \end{bmatrix} = b, \]  

(7.72)

in which the \( n \) non-basic variables from among \((n + m)\) elements of \( y_s \) are set equal to zero. Eliminating these \( n \) variables by equating them to zero leaves a set of \( m \) equations in \( m \) unknowns (the basic variables). This set of equations can be denoted by
\[ B y_B = b, \tag{7.73} \]

where the vector of basic variables,

\[
y_B = \begin{bmatrix} y_{B1} \\ y_{B2} \\ \vdots \\ \vdots \\ y_{Bn} \end{bmatrix}, \tag{7.74} \]

is obtained by eliminating the non-basic variables from \( y \), and the basic matrix,

\[
B = \begin{bmatrix} B_{11} & B_{12} & \cdots & B_{1m} \\ B_{21} & B_{22} & \cdots & B_{2m} \\ \vdots & \vdots & \ddots & \vdots \\ B_{m1} & B_{m2} & \cdots & B_{mn} \end{bmatrix}, \tag{7.75} \]

is obtained by eliminating the columns corresponding to coefficients of non-basic variables from the matrix \([A I]\). For the zero-th iteration step, the original variables are likely selected as the initial nonbasic variables (set equal to zero) and the slack (or surplus) variables are selected as the initial basic variables. So, the initial \( y_B \) and \( B \) are

\[
y_B = \begin{bmatrix} y_{n+1} \\ y_{n+2} \\ \vdots \\ \vdots \\ y_{n+m} \end{bmatrix}, \quad B = \begin{bmatrix} 1 & 0 & \cdots & 0 \\ 0 & 1 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{bmatrix}. \tag{7.76} \]

The simplex method introduces only basic variables such that \( B \) is nonsingular, so that \( B^{-1} \) always will exist. Therefore, to solve \( B y_B = b \), both sides would be multiplied by \( B^{-1} \).

\[
B^{-1} B y_B = B^{-1} b, \tag{7.77} \]

\[
y_B = B^{-1} b. \tag{7.78} \]

Let \( c_B \) be the row vector whose elements are the objective function coefficients (including zeroes for slack variables) for the corresponding elements of \( y_B \). The value of the objective function for this basic solution is then
\[ f = c_B y_B = c_B B^{-1} b . \]  

(7.79)

The initial \( y_B = b \) because \( B = B^{-1} \) for identity matrix and the initial \( c_B \) is a null vector because the coefficients of the slack variables in the original objective function are zeros. Thus, the initial value of the objective function would be zero.

The subsequent iteration for another basic feasible solution is followed by determining the entering basic variable by selecting the nonbasic variable that would increase \( f \) at the fastest rate. Based on the entering basic variable, the leaving basic variable is determined by selecting the basic variable that reaches zero first as the entering basic variable increased. According to the entering basic variable and the leaving basic variable, the vector \( y_B \), the matrix \( B \), and \( c_B \) are updated and then new \( y_B \) and the value of \( f \) are identified. Iterations are repeated until the objective function obtains an optimal value.

7.6 Implementation of LP Model to a US-Defense HLW

7.6.1 Compositions of HLW and Borosilicate Glass

A specific type of HLW sludge and borosilicate glass frit were chosen to apply the developed linear programming model. Table 7.2, Table 7.3, and Table 7.4 show the details of selected HLW sludge and glass frit which were used for frit development and model assessment activities at the Defense Waste Processing Facility (DWPF) of the Savannah River Technology Center (SRTC) [Peeler, et al., 2001].

Table 7.2 Composition of Nominal MB3 sludge Used for Initial Model Assessments and Frit Development Activities [Peeler, et al., 2001].

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Wt %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃</td>
<td>16.846</td>
</tr>
<tr>
<td>CaO</td>
<td>3.743</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.000</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>43.087</td>
</tr>
<tr>
<td>MgO</td>
<td>0.241</td>
</tr>
<tr>
<td>MnO</td>
<td>2.824</td>
</tr>
<tr>
<td>Na₂O</td>
<td>18.740</td>
</tr>
<tr>
<td>NiO</td>
<td>1.926</td>
</tr>
<tr>
<td>SiO₂</td>
<td>2.018</td>
</tr>
<tr>
<td>U₃O₈</td>
<td>10.575</td>
</tr>
<tr>
<td>Total</td>
<td>100.000</td>
</tr>
</tbody>
</table>

Table 7.3 Oxide Composition of Glass Frit 320 [Peeler, et al., 2001]

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>B₂O₃</td>
<td>8.00</td>
</tr>
<tr>
<td>Li₂O</td>
<td>8.00</td>
</tr>
<tr>
<td>Na₂O</td>
<td>12.00</td>
</tr>
<tr>
<td>SiO₂</td>
<td>72.00</td>
</tr>
<tr>
<td>Total</td>
<td>100.00</td>
</tr>
</tbody>
</table>
Table 7.4  Concentration, Decay Constant, and Average Decay Energy of Several Radionuclides in the HLW Sludge in DWPF MB3 [Bibler, et al., 2002]

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Wt%</th>
<th>( \lambda ) [1/sec]</th>
<th>Average decay(^{14}) energy [MeV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-59</td>
<td>3.88E-04</td>
<td>2.93E-13</td>
<td>6.700E-03</td>
</tr>
<tr>
<td>Ni-63</td>
<td>3.97E-05</td>
<td>2.00E-10</td>
<td>1.700E-02</td>
</tr>
<tr>
<td>Se-79</td>
<td>8.95E-05</td>
<td>3.38E-13</td>
<td>4.200E-02</td>
</tr>
<tr>
<td>Sr-90</td>
<td>3.32E-03</td>
<td>7.55E-10</td>
<td>1.958E-01</td>
</tr>
<tr>
<td>Zr-93</td>
<td>5.41E-03</td>
<td>1.44E-14</td>
<td>1.960E-02</td>
</tr>
<tr>
<td>Tc-99</td>
<td>7.35E-04</td>
<td>1.04E-13</td>
<td>8.460E-02</td>
</tr>
<tr>
<td>Pd-107</td>
<td>8.10E-05</td>
<td>3.38E-15</td>
<td>1.000E-02</td>
</tr>
<tr>
<td>Sn-121m</td>
<td>6.77E-06</td>
<td>4.00E-10</td>
<td>3.380E-01</td>
</tr>
<tr>
<td>Sn-126</td>
<td>1.58E-04</td>
<td>2.20E-13</td>
<td>1.135E+00</td>
</tr>
<tr>
<td>I-129</td>
<td>4.29E-06</td>
<td>1.40E-15</td>
<td>7.800E-02</td>
</tr>
<tr>
<td>Cs-135</td>
<td>9.34E-05</td>
<td>9.55E-15</td>
<td>5.630E-02</td>
</tr>
<tr>
<td>Cs-137</td>
<td>3.17E-04</td>
<td>7.30E-10</td>
<td>1.866E-01</td>
</tr>
<tr>
<td>Pm-147</td>
<td>1.40E-05</td>
<td>8.39E-09</td>
<td>6.051E-02</td>
</tr>
<tr>
<td>Sm-151</td>
<td>6.75E-04</td>
<td>2.44E-10</td>
<td>1.978E-02</td>
</tr>
<tr>
<td>Eu-154</td>
<td>2.96E-06</td>
<td>2.56E-09</td>
<td>1.509E+00</td>
</tr>
<tr>
<td>Th-232</td>
<td>3.44E-02</td>
<td>1.56E-18</td>
<td>4.084E+00</td>
</tr>
<tr>
<td>U-233</td>
<td>1.11E-04</td>
<td>1.38E-13</td>
<td>4.904E+00</td>
</tr>
<tr>
<td>U-234</td>
<td>5.72E-04</td>
<td>8.97E-14</td>
<td>4.859E+00</td>
</tr>
<tr>
<td>U-235</td>
<td>3.04E-02</td>
<td>3.12E-17</td>
<td>4.418E+00</td>
</tr>
<tr>
<td>U-236</td>
<td>1.46E-03</td>
<td>9.38E-16</td>
<td>4.570E+00</td>
</tr>
<tr>
<td>Np-237</td>
<td>1.88E-03</td>
<td>1.03E-14</td>
<td>5.156E+00</td>
</tr>
<tr>
<td>U-238</td>
<td>7.53E+00</td>
<td>4.92E-18</td>
<td>4.279E+00</td>
</tr>
<tr>
<td>Pu-238</td>
<td>2.25E-04</td>
<td>2.50E-10</td>
<td>5.591E+00</td>
</tr>
<tr>
<td>Pu-239</td>
<td>1.24E-02</td>
<td>9.11E-13</td>
<td>5.199E+00</td>
</tr>
<tr>
<td>Pu-240</td>
<td>1.05E-03</td>
<td>3.35E-12</td>
<td>5.253E+00</td>
</tr>
<tr>
<td>Pu-241</td>
<td>2.71E-05</td>
<td>1.53E-09</td>
<td>5.230E-03</td>
</tr>
<tr>
<td>Am-241</td>
<td>9.53E-04</td>
<td>5.08E-11</td>
<td>5.604E+00</td>
</tr>
<tr>
<td>Am-242m</td>
<td>1.13E-06</td>
<td>1.45E-10</td>
<td>6.664E-02</td>
</tr>
<tr>
<td>Pu-242</td>
<td>1.16E-04</td>
<td>5.89E-14</td>
<td>4.982E+00</td>
</tr>
<tr>
<td>Am-243</td>
<td>2.68E-04</td>
<td>2.98E-12</td>
<td>5.423E+00</td>
</tr>
<tr>
<td>Cm-243</td>
<td>1.16E-06</td>
<td>7.71E-10</td>
<td>6.189E+00</td>
</tr>
<tr>
<td>Cm-244</td>
<td>5.48E-05</td>
<td>1.21E-09</td>
<td>5.901E+00</td>
</tr>
</tbody>
</table>

7.6.2  Construction of LP Problem and Graphical Solution

Based on the compositions of the HLW sludge and the glass frit shown above tables and the specifications of the canister described in EM-WAPS, undetermined coefficients of constraints inequalities in \( A \) and \( b \) are determined.

With weight fractions of B, Na, and Li in Table 7.2 and 7.3, Eq. (7.56), (7.57), and (7.58) become

\[
-0.430M_W - 0.405M_G \leq 0 ,
\]  

\[ -0.201M_W - 0.251M_G \leq 0 , \quad (7.57a) \]

\[ -0.250M_W - 0.213M_G \leq 0 , \quad (7.58a) \]

The coefficient \( \zeta \) in Eq. (7.59) is determined based on Table 7.4, thus Eq. (7.59) is

\[ 0.010M_W \leq 1500 . \quad (7.59a) \]

For the canister and the glass frit, the thermal conductivity of the canister made of AISI 304L stainless steel [DOE-EM-0093, 1999] is assumed to be 14.9 \([\text{W/m}^{\circ}\text{K}]\) [Incropera, et al., 1990] and that of the borosilicate waste glass is assumed to be 0.9 \([\text{W/m}^{\circ}\text{K}]\)\(^{15}\). The canister specified in EM-WAPS has a cylindrical geometry with a height of 3 m, an outer diameter of 0.61 m, and a wall thickness of 0.0095 m. Then, \( \alpha \) and \( \beta \) are determined as

\[
\alpha = \left( \frac{\zeta}{V_C} \right) \left( \frac{r_i^2}{4k_g} + \frac{cr_i}{2k_c} \right) = \left( \frac{0.010}{0.818} \right) \left( \frac{(0.2955)^2}{4(0.9)} + \frac{(0.0095)(0.2955)}{2(14.9)} \right) = 2.977 \times 10^{-4},
\]

\[
\beta = \left( \frac{\zeta}{V_C} \right) \left( \frac{r_i^2 L^{1/4}}{2.84 r_o} \right)^{4/5} = \left( \frac{0.010(0.2955)^2(3)^{1/4}}{0.818} \right)^{4/5} = 5.861 \times 10^{-3}.
\]

Assuming the ambient temperature \( T_{\infty} = 25^\circ\text{C} \), Eq. (7.61) is

\[ (6.159 \times 10^{-3})M_W \leq 375. \quad (7.61a) \]

With aforementioned geometry, the volume of an empty canister \( V_C \) is calculated to be \( \sim 0.818 \text{ m}^3 \). Assuming \( \rho_W = 2,800 \text{ kg/m}^3 \), \( \rho_G = 2,200 \text{ kg/m}^3 \), and \( \delta = 0.8 \), Eq. (7.62) and (7.63) are

\[ -M_W - 1.273 M_G \leq -2290, \quad (7.62a) \]

\[ M_W + 1.273 M_G \leq 2863. \quad (7.63a) \]

For the density of AISI 304L stainless steel, 7,900 \( \text{kg/m}^3 \) (at 300\(^{\circ}\text{K} \)) [Incropera, et al., 1990], the mass \( M_C \) of the empty canister is calculated to be \( \sim 467 \text{ kg} \). Then, Eq (7.64) is

\[ M_W + M_G \leq 2033. \quad (7.64a) \]

With the combined weight fraction of plutonium isotopes in Table 7.4, Eq. (7.65) is

\[ (1.38 \times 10^{-4}) M_W \leq 2.045. \quad (7.65a) \]

Now, we have the matrix \( A \) and the vector \( b \) as follows.

\(^{15}\) The thermal conductivity of a material varies with temperature, and it is known that the thermal conductivity of borosilicate waste glass ranges from \( \sim 0.9 \) to \( \sim 1.3 \) \([\text{W/m}^{\circ}\text{K}]\) in the temperature range 100 to 500\(^{\circ}\text{C} \) [Benedict, et al., 1981]. However, the effect of its variation on the constraint on temperature limit is not significant due to small contribution of \( \alpha \) in the coefficient of Eq. (7.61): For example, for \( k_g = 0.9 \), \( \alpha + \beta \approx 0.263 \zeta \) and for \( k_g = 1.3 \), \( \alpha + \beta \approx 0.254 \zeta \). By taking \( k_g = 0.9 \), the mass of HLW is evaluated conservatively.
Figure 7.3 shows a graphical solution to the linear programming problem formulated above. All constraints can be plotted on the solution space as lines by replacing the inequality by an equal sign and the inequality is represented by a small solid arrow on the plot. So, we have a feasible solution space satisfying all constraints, where optimum values of the objective function exist. From this figure, we observe that only two constraints related to a filled canister weight and a waste glass product volume delineate the feasible solution space while other constraints such as heat generation, temperature, Pu-concentration, and PCT-limit do not touch the feasible solution space.

\[
A = \begin{bmatrix}
-0.430 & -0.405 \\
-0.201 & -0.251 \\
-0.250 & -0.213 \\
6.159 \times 10^{-3} & 0 \\
0.010 & 0 \\
-1 & -1.273 \\
1 & 1.273 \\
1 & 1 \\
1.380 \times 10^{-4} & 0 \\
\end{bmatrix}, \quad b = \begin{bmatrix}
0 \\
0 \\
375 \\
1500 \\
-2290 \\
2863 \\
2034 \\
2.045 \\
\end{bmatrix}
\]

Figure 7.3 The graphical solution of the linear programming problem for a US-defense HLW
For PCT constraints, Figure 7.3 shows that any combination of $M_W$ and $M_G$ satisfies the PCT-limit constraint as long as waste glass products can be made even though no glass material is blended with HLW sludge. This is because the analytical model of PCT limit constraint estimates the dissolution rate of a nuclide from a glass matrix based on the concentration of the nuclide in the glass matrix and in any combination of the HLW sludge and the glass frit, concentrations of B, Na, and Li do not exceed the limit of the EA glass shown in Table 7.1.

Constraints of heat generation, temperature, and Pu-concentration are plotted far away from the feasible solution space, which mean those constraints are not essential for the conditioning of the selected HLW sludge here. Practically, heat generation from the defense HLW of the SRTC is not a significant concern in HLW conditioning because it has been cooled for decades in the SRTC-HLW storage tanks. The optimum value of the objective function occurs at $M_W = 1,096$ [kg] and $M_G = 938$ [kg], where the value of $M_W$ itself is the optimum, which is equivalent to 53.9 wt% waste loading.

However, it should be pointed out that results are subject to the constraints and assumptions used. For constraint lines of a waste glass product volume, which form a band with a width, we used an arbitrary value of $\delta = 0.8$, which takes into account nonlinearity in a volume summation of different materials. This value is expected to vary with the blending ratio of HLW sludge to matrix material while it is used as a constant here. An estimation has been done to check how the variation of $\delta$ affects the optimum value of the objective function and it was observed that larger value of $\delta$ makes lines of constraints on a waste glass product volume move downward keeping the same slope resulting in a greater optimum value of the objective function while in adverse case, resulting in a smaller optimum value. For $\delta = 1.0$, the optimum value of the objective function occurs where $M_W = 2,033$ [kg] and $M_G = 0$ [kg], which means 100 wt% waste loading. However, this can not take place in reality because there is no glass matrix to confine HLW sludge. For $\delta < 0.71$, there is no feasible solution space because the constraint band lies outside of the region defined by the filled canister weight constraint. Therefore, in this model, in order to apply the constraint on waste glass volume rigorously, it is crucial to know the range of $\delta$ and its dependency on the blending ratio of HLW sludge to matrix material.

The DWPF had estimated the fractional waste loading range for the same HLW sludge and glass frit specified in Table 7.2, Table 7.3, and Table 7.4 based on its own constraints. The fractions of the maximum allowable and the minimum allowable waste loading estimated were 30.0 wt% and 22.0 wt%, respectively. To figure out what makes the difference between the optimum waste loading estimated by the LP model and the maximum allowable waste loading estimated by the DWPF, it is necessary to understand constraints employed by the DWPF.

### 7.6.3 Constraints of the Defense Waste Processing Facility (DWPF)

The Defense Waste Processing Facility (DWPF) of the SRTC has produced HLW glass products since 1996. The facility uses glass as a matrix material and borosilicate wasteform is generated. A simplified HLW process flow of the DWPF is illustrated in Figure 7.2. While producing borosilicate wasteform, several product and process constraints should be satisfied. These include constraints on the process melt to assure
that the material is processable and on the quality of the resulting waste glass product [Brown, 2002].

The decision whether a particular waste-glass blend will produce a melt that will satisfy the aforementioned constraints is made based on sampling and measurement subject to uncertainties. For the acceptability decision on a melt, the DWPF developed a statistical system called the Product Composition Control System (PCCS). The waste-glass mix and melt properties that must be controlled are related to the composition of vitrified waste product. Therefore, the PCCS function is to blend waste with glass and then monitor the composition of the feed in the Slurry Mix Evaporator (SME) in Figure 7.2. The SME is both the first control point in the DWPF process wherein all necessary constituents are present and the last control point at which any change to them can be effected. The PCCS thus deals with monitoring the blended SME batch and then the monitoring of the SME is accomplished by sampling its contents.

For each SME batch, a set of samples is taken to begin an acceptability decision. Each of these samples is vitrified and the chemical compositions of the resulting waste glasses are measured. The average of the measured chemical compositions for a minimum number of samples is determined, and this average composition serves as the basis for the acceptability decision for the SME batch. The constraints applicable to the DWPF acceptability decisions are provided in Table 7.5.

Table 7.5  Constraints Applicable to DWPF Acceptability Decisions [Brown, 2002]

<table>
<thead>
<tr>
<th>Name</th>
<th>Constraint</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low conservation</td>
<td>95 wt% ≤ Σ(Major oxides)</td>
<td>No uncertainty applied</td>
</tr>
<tr>
<td>High conservation</td>
<td>Σ(Major oxides) ≤ 105 wt%</td>
<td>No uncertainty applied</td>
</tr>
<tr>
<td>Durability</td>
<td>PCT (B, Na, Li) ≤ EA</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Alumina</td>
<td>3.0 wt% + MU16 ≤ Al2O3 content</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Homogeneity</td>
<td>4.0 wt% + MU ≤ Al2O3 content</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Low frit loading</td>
<td>70 wt% + MU ≤ Σ(Frit oxides)</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>High frit loading</td>
<td>Σ(Frit oxides) ≤ 85 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Liquidus temperature</td>
<td>TL ≤ 1050°C</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Low viscosity</td>
<td>20 poise ≤ Viscosity (η)</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>High viscosity</td>
<td>Viscosity (η) ≤ 110 poise</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>TiO2</td>
<td>≤ 1.0 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>NaCl</td>
<td>≤ 1.0 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>NaF</td>
<td>≤ 1.0 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Cr2O3</td>
<td>≤ 0.3 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>SO4</td>
<td>≤ 0.4 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
<tr>
<td>Cu</td>
<td>≤ 0.5 wt% - MU</td>
<td>Uncertainty (95% confidence level)</td>
</tr>
</tbody>
</table>

Table 7.5 shows that the DWPF uses different constraints from those specified in EM-WAPS except the durability constraint which is directly related to the product consistency test (PCT). The DWPF takes into account composition-related constraints such as conservation, alumina, homogeneity, frit loading, and several chemical

---

16 MU: measurement uncertainty
compounds (TiO₂, NaCl, NaF, Cr₂O₃, SO₄, and Cu) as well as property-related constraints such as durability, liquidus temperature, and viscosity.

To estimate a maximum allowable waste loading for the compositions of the HLW and the glass frit listed in Table 7.2 and 7.3, a similar linear programming method could be applied based on the constraints listed in Table 7.5. Mathematical formulations are possible for constraints except for conservation and property-related constraints (liquidus temperature and viscosity).

For the durability constraint, mathematical formulations developed in Section 7.3 can be applied. So, Eq. (7.56a), (7.57a), and (7.58a) are valid.

The alumina content constraint is formulated as

\[ \frac{m_{\text{Al}_2\text{O}_3}}{M_W + M_G} \geq 0.03, \]  \hspace{1cm} (7.80)

and the homogeneity constraint as

\[ \frac{m_{\text{Al}_2\text{O}_3}}{M_W + M_G} \geq 0.04, \]  \hspace{1cm} (7.81)

where \( m_{\text{Al}_2\text{O}_3} \) = the mass of Al₂O₃ in a solidified waste glass. \( m_{\text{Al}_2\text{O}_3} \) can be represented in terms of \( M_W \) and \( M_G \),

\[ m_{\text{Al}_2\text{O}_3} = x_{\text{W,Al}_2\text{O}_3} M_W + x_{\text{G,Al}_2\text{O}_3} M_G, \]  \hspace{1cm} (7.82)

where \( x_{\text{W,Al}_2\text{O}_3} \) = the weight fraction of Al₂O₃ in HLW,

\( x_{\text{G,Al}_2\text{O}_3} \) = the weight fraction of Al₂O₃ in glass frit.

Inserting (7.82) into (7.80) and (7.81) results in:

\( (x_{\text{W,Al}_2\text{O}_3} - 0.03)M_W + (x_{\text{G,Al}_2\text{O}_3} - 0.03)M_G \geq 0, \)  \hspace{1cm} (7.83)

\( (x_{\text{W,Al}_2\text{O}_3} - 0.04)M_W + (x_{\text{G,Al}_2\text{O}_3} - 0.04)M_G \geq 0. \) \hspace{1cm} (7.84)

As alternative forms to (7.80) and (7.81), the following can also be possible:

\[ \frac{m_{\text{Al}}}{M_W + M_G} \geq 0.016, \]  \hspace{1cm} (7.80a)

\[ \frac{m_{\text{Al}}}{M_W + M_G} \geq 0.021, \]  \hspace{1cm} (7.81a)

where \( m_{\text{Al}} \) = the mass of Al in a solidified waste glass. \( m_{\text{Al}} \) can be represented in terms of \( M_W \) and \( M_G \),

\[ m_{\text{Al}} = x_{\text{W,Al}} M_W + x_{\text{G,Al}} M_G, \]  \hspace{1cm} (7.82a)

where \( x_{\text{W,Al}} \) = the weight fraction of Al in HLW,

\( x_{\text{G,Al}} \) = the weight fraction of Al in glass frit.

Inserting (7.82a) into (7.80a) and (7.81a) results in:

\( (x_{\text{W,Al}} - 0.016)M_W + (x_{\text{G,Al}} - 0.016)M_G \geq 0, \)  \hspace{1cm} (7.83a)
\begin{align}
(x_{W,Al} - 0.021)M_W + (x_{G,Al} - 0.021)M_G \geq 0. \tag{7.84a}
\end{align}

With the weight fraction of Al$_2$O$_3$ in Table 7.2 and 7.3, (7.83) and (7.84) are:
\begin{align}
-M_W + 0.217M_G \leq 0, \tag{7.83b}
-M_W + 0.311M_G \leq 0. \tag{7.84b}
\end{align}

The low frit loading and the high frit loading constraints are formulated as
\begin{align}
\frac{M_G}{M_W + M_G} \geq 0.7, \tag{7.85}
\frac{M_G}{M_W + M_G} \leq 0.85. \tag{7.86}
\end{align}

Rearranging (7.83) and (7.84) results in:
\begin{align}
M_W - 0.429M_G \leq 0, \tag{7.85a}
M_W - 0.177M_G \leq 0. \tag{7.86a}
\end{align}

The TiO$_2$ constraint is formulated as
\begin{align}
\frac{m_{TiO_2}}{M_W + M_G} \leq 0.01, \tag{7.87}
\end{align}
where $m_{TiO_2}$ = the mass of TiO$_2$ in a solidified waste glass.

The NaCl constraint is formulated as
\begin{align}
\frac{m_{NaCl}}{M_W + M_G} \leq 0.01, \tag{7.88}
\end{align}
where $m_{NaCl}$ = the mass of NaCl in a solidified waste glass.

The NaF constraint is formulated as
\begin{align}
\frac{m_{NaF}}{M_W + M_G} \leq 0.01, \tag{7.89}
\end{align}
where $m_{NaF}$ = the mass of NaF in a solidified waste glass.

The Cr$_2$O$_3$ constraint is formulated as
\begin{align}
\frac{m_{Cr_2O_3}}{M_W + M_G} \leq 0.003, \tag{7.90}
\end{align}
where $m_{Cr_2O_3}$ = the mass of Cr$_2$O$_3$ in a solidified waste glass.

The SO$_4$ constraint is formulated as
\begin{align}
\frac{m_{SO_4}}{M_W + M_G} \leq 0.004, \tag{7.91}
\end{align}
where $m_{SO_4}$ = the mass of SO$_4$ in a solidified waste glass.
The Cu constraint is formulated as
\[
\frac{m_{\text{Cu}}}{M_W + M_G} \leq 0.005, \tag{7.92}
\]
where \( m_{\text{Cu}} \) = the mass of Cu in a solidified waste glass.

Now, we have the matrix \( \mathbf{A} \) and the vector \( \mathbf{b} \) as follows:
\[
\mathbf{A} = \begin{bmatrix}
-0.430 & -0.405 \\
-0.201 & -0.251 \\
-0.250 & -0.213 \\
-1 & 0.217 \\
-1 & 0.311 \\
1 & -0.429 \\
1 & -0.177
\end{bmatrix}, \quad \mathbf{b} = \begin{bmatrix}
0 \\
0 \\
0 \\
0 \\
0 \\
0 \\
0
\end{bmatrix}.
\]

The graphical solution for the above LP problem is shown in Figure 7.4. Due to unavailability of measurement uncertainty (MU), it is assumed to be zero. Constraints on \( \text{TiO}_2, \text{NaCl, NaF, Cr}_2\text{O}_3, \text{SO}_4, \) and Cu are not plotted because they are not included either in the HLW or the glass (Table 7.2 and 7.3). Essential constraints forming the feasible solution space are constraints on the low frit loading and the homogeneity. The constraint line on the low frit loading indicates upper limit of waste loading fraction, here 30.0 wt\%, and the constraint line on the homogeneity sets lower limit of waste loading, here 23.6 wt\%. So, the waste loading range for the used HLW and glass is estimated as between 23.6 wt\% and 30.0 wt\% from the graphical solution. This well agrees with that estimated by the DWPF in [Peeler, et al., 2001], which is between 22.0 wt\% and 30.0 wt\%. Small discrepancy in lower limits seems to arise from measurement uncertainty (MU), which is neglected in the graphical solution. Two property-related constraints which are not taken into account here seems likely nonessential for the estimation of waste loading range.

![Figure 7.4 The graphical solution for the DWPF-HLW conditioning](image-url)
However, it is noticeable that the feasible solution space determined by two constraints is open-ended. This implies that the maximum (or minimum) allowable waste loading, in terms of mass, could be infinite even though its fraction remains constant. Practically, infinite waste loading is impossible at any case due to the limited volume of a canister.

This type of feasible solution space usually means that the LP problem is under-constrained and optimum values are not identified. To make the LP problem complete, additional constraints need to be considered. Of course, two property-related constraints that are not taken into account for the construction of the LP problem could play such roles that make the feasible solution space close-ended. However, considering that constraints of the DWPF have been employed for the purpose of acceptability decision in the process of waste glass production, constraints listed in Table 7.5 might be insufficient for the determination of optimum waste loading. If it is so, some other constraints such like specified in EM-WAPS could be employed for the determination of optimum waste loading. This could be a reasonable choice for optimizing of the solidification of US-defense HLW because all HLW producers including the DWPF should meet the specifications of EM-WAPS for their HLW products to be accepted into the Civilian Radioactive Waste Management System (CRWMS) [DOE/EM-0093, 1996].

![Figure 7.5 Revised graphical solution for the DWPF-HLW conditioning incorporating constraints of both EM-WAPS and the DWPF](image)

Figure 7.5 shows the revised graphical solution for the DWPF HLW conditioning which includes constraints of both EM-WAPS and the DWPF. Now, the feasible solution

| $M_W$ [kg] | $M_W \leq 150,000$ [kg] (Heat generation) |
| $M_W \leq 60,887$ [kg] (Temperature) |
| $M_W \leq 14,819$ [kg] (Pu-concentration) |
| $M_W + M_G \leq 2,033$ (Filled canister weight) |
| $M_W + 1.27M_G \leq 2,863$ (Filled waste glass volume) |
| $M_W + 1.27M_G \geq 2,290$ |
| $M_W + 1.25M_G \geq 0$ (Na-concentration) |
| $M_W + 0.94M_G \geq 0$ (B-concentration) |
| $M_W + 0.85M_G \geq 0$ (Li-concentration) |
| $M_G$ [kg] | $M_G = 610$ [kg] |
| $M_G = 1,423$ [kg] |
| $M_W - 0.43M_G \leq 0$ (Low frit loading) |
| $M_W - 0.31M_G \geq 0$ (Homogeneity) |
| $M_W - 0.22M_G \geq 0$ (Al-content) |
| $M_W - 0.18M_G \geq 0$ (High frit loading) |
space is of a closed shape and essential constraints are identified as those on the filled waste glass weight, the filled waste volume, the low frit loading, and the homogeneity. The maximum waste loading, 610 kg (30.0 wt%), is determined by the intersection of the filled waste weight and the low frit loading constraints while the minimum waste loading, 448 kg (23.6 wt%), is done by the intersection of the filled waste glass volume (lower limit) and the homogeneity constraints.

As shown in Figure 7.5, by incorporating of both EM-WAPS and the DWPF constraints, the optimum waste loading and the fractional waste loading range in solidification of a DWPF-HLW could be estimated more accurately. This implies that the combined constraints contribute to the construction of a more rigorous LP model for optimization of US-defense HLW conditioning.

7.7 Discussions and Conclusions

In this chapter, HLW conditioning model for determination of an optimized composition of solidified HLW has been developed based on the regulations described in EM-WAPS and the constraints of the DWPF. As a mathematical method, a linear programming model was implemented and formulations were derived for key constraints.

Most of the regulations and guidelines identified could be formulated in a linear inequality in terms of the mass of HLW and that of glass frit, but some of those such as constraints on radiation dose limit and nuclear criticality limit specified in EM-WAPS could not be formulated in an appropriate form. So, if an optimized waste glass is produced based on the calculation by the LP model developed, its radiation dose and the criticality need to be checked and confirmed prior to final disposal.

For the constraint on the filled waste volume limit, a constant volume reduction factor was assumed because the appropriate relation between the selected HLW and glass frit is unavailable. Therefore, for the case that an optimum waste loading is determined by the constraint on the filled waste volume limit, the uncertainty of the optimum waste loading could be significant. To avoid this, the volume reduction factor needs to be evaluated with respective to the mixing ratio of a selected HLW and a glass frit. In this study, however, doing so is beyond the scope. It will remain as a future work.

With the implementation of the LP model to the solidification of a US-defense HLW, it was found that by incorporating of both EM-WAPS and the DWPF constraints, the optimum waste loading of US-defense HLW can be evaluated more reliably.

With the graphical solution of the LP problem, essential and non-essential (or redundant) constraints for determining a feasible solution region were clearly figured out. It also shows how redundant constraints would affect on the shape of the feasible solution space with the compositional variation of HLW or glass frit.
8 Solidification of HLW Generated from ATW Fuel Cycle

8.1 Introduction

In the previous chapter, a linear programming (LP) model for determining the optimum waste loading and the composition of solidified HLW has been developed and applied for the determination of optimum waste loading of a US-defense HLW.

In this chapter, the developed LP model is applied to the solidification of HLW to be produced from the ATW fuel cycle. For this, first, the mass and the composition of HLW being generated from the ATW fuel cycle are quantified and then the LP model is applied to determine optimum waste loadings for the HLW quantified.

In Chapter 6, we quantified the mass and the composition of TRU in the waste streams of a reference ATW system employing LBE-cooled transmuters. Other than TRU, liquid HLW generated from the separation of the ATW fuel cycle also includes several constituents such as FPs, process chemicals, corrosion products, and oxygen. Those constituents are quantified based on a specific condition and a reference reprocessing design before applying the LP model.

8.2 HLW Generated from ATW Fuel Cycle

According to the reference ATW plant deployment scenario described in [Hill, et al., 1999], three major HLW streams are identified: (1) HLW from the separation process of LWR-spent fuel, (2) HLW from separation process of ATW-spent fuel, and (3) HLW from the last transmuter to be decommissioned at the end of ATW plant operation. In this study, long-lived fission products (\(^{99}\)Tc and \(^{129}\)I) are assumed not to be separated. Figure 8.1 shows high-level waste streams of the ATW fuel cycle.

For (1), a total of 86,317 MT of LWR-spent fuel is processed. The burnup of LWR-spent fuel is assumed to be 33,000 MWd/MT and a 25-year cooling period is assumed after discharge before reprocessing. Uranium and TRU are separated from fission products with recovery yields of 99.995% and 99.9%, respectively. Thus, all fission products and residual TRU are contained in the waste stream. The amounts of separated uranium and TRU from 86,317 MT of LWR-spent fuel are estimated as 82,314 MT and 900 MT, respectively. Consequently, the amount of fission products and residual TRU contained in the waste stream is 3,103 MT (3,102.1 MT for FPs and 0.9 MT for TRU).

For (2), a TRU-Zr dispersion fuel is assumed as an ATW fuel type and TRU separated from LWR-spent fuel is blended with zirconium. The TRU content of recharged fuel at an equilibrium cycle of the LBE-cooled transmuter is calculated to be \(~53\) wt% by WACOM. If 53 wt% TRU enrichment is assumed for ATW fuel fabrication, this results in \(~1,700\) MT of TRU-Zr fuel for 900 MT TRU for a once-through fuel cycle. Discharged TRU-Zr fuel, however, is reprocessed and the separated TRU and Zr are refabricated with makeup material. Thus, the cumulative mass of discharged TRU-Zr fuel to be processed in the separation facility is more than \(~1,700\) MT, it is estimated to be 6,579 MT by WACOM: 6,507 MT is discharged from 67 transmuters (97.12 MT from a transmuter), 72 MT from the last transmuter.

For discharged ATW-spent fuel, TRU and Zr are assumed to be separated with recovery yields of 99.9% and 100%, respectively. All fission products and residual TRU will be contained in the waste stream. The applied discharge burnup for TRU-Zr fuel is...
27.2 wt% and a 25-year cooling period is assumed. The amount of fission products and residual TRU in the waste stream is estimated as 895 MT (885.4 MT from 67 transmuters, 9.6 MT from the last transmuter) and 2.4 MT, respectively.

Figure 8.1  HLW streams of the ATW fuel cycle

For (3), the HLW (spent fuel) of the last transmuter at the end of ATW plant operation is assumed to be directly disposed of in a geological repository. The amount of FPs and TRU contained in the last transmuter is supposed to be less than 2.6 MT.

Here, TRU loss from the ATW fuel fabrication process is assumed to be zero.

Major constituents of liquid HLW generated from separation processes of LWR- and ATW-spent fuel are fission products, residual TRU, corrosion products, and process chemicals added in reprocessing. Gaseous fission products such as Kr, I, and Xe are not included in liquid HLW and thus assumed not to be solidified\(^{17}\). Most elements in liquid HLW exist in an oxide form. The mass and composition of FPs and TRU in HLW can be estimated based on an applied burnup and a cooling time. But those of corrosion products and process chemicals being contained in HLW depend on the design of a specific separation facility and/or the processes of a HLW processing facility. In this study for

---

\(^{17}\) Krypton and xenon isotopes are currently released from reprocessing plants to the atmosphere after holding for some periods for decays. All iodine isotopes except \(^{129}\)I decay into stable nuclides prior to reprocessing (for permanent fixation of \(^{129}\)I, adsorption on silver-loaded adsorbents, such as zeolites, silica, or alumina, could be used).
corrosion products and process chemicals in HLW, reference design data of a PUREX plant are employed due to unavailability of design data of the ATW separation process: as process chemicals 22.3 kg-Na and 0.393 kg-P, and as corrosion products 4.2 kg-Fe, 1.1 kg-Ni, and 1.09 kg-Cr are contained in the liquid HLW generated from the separation process of 1 MT of LWR-spent fuel [PNC SN8410 90-061, 1990].

The composition and the mass of FPs and residual TRU generated from the separation process of the LWR-spent fuel are calculated by ORIGEN2.1 code. It is estimated that ~69 kg of HLW is generated from the separation process of 1 MT of LWR-spent fuel, consisting of 43.0 wt% FPs (excluding gaseous FPs), 0.016 wt% TRU, 33.8 wt% corrosion products and process chemicals, and 23.2 wt% oxygen. The heat generation rate per kg-HLW is 5.494 [W/kg-HLW].

The composition and the mass of FPs and residual TRU generated from the separation process of the ATW-spent fuel are calculated by WACOM and MOCUP codes. The cumulative masses of FPs and TRU and the composition of TRU in the waste stream are quantified by WACOM based on cycle-by-cycle analysis, and the composition of FPs is calculated by MOCUP\(^1\) based on the composition of an equilibrium cycle due to the limitation of WACOM. From the separation process of 1 MT of ATW-spent fuel, ~178 kg of HLW is generated, containing 65.7 wt% FPs (excluding gaseous FPs), 0.21 wt% TRU, 13.1 wt% corrosion products and process chemicals, and 21.0 wt% oxygen. The heat generation rate per kg-HLW is 2.917 [W/kg-HLW].

HLW is melted with a glass frit and poured into a stainless steel canister. As a glass matrix, a borosilicate glass frit developed by Japan Nuclear Cycle Development Institute (JNC) [Yamashita, et al., 1995] is assumed. This borosilicate glass provides an empirical formula for the density of the vitrified waste and thus the constraint on the filled waste volume is reformulated based on it.

### 8.3 Determination of Optimum Waste Loadings

#### 8.3.1 Empirical Formula for Density of Solidified HLW

According to EM-WAPS, the solidified waste volume should be between 80% and 100% of the volume of an empty canister and it was formulated in (7.47) of Section 7.4 by introducing \(\delta\) to account for nonlinearity in volume summation. If a relation for the density of the solidified waste, \(\rho_{WG}\), is available, (7.47) can be reformulated as

\[
0.8V_C \leq \frac{M_w + M_G}{\rho_{WG}} \leq V_C \quad [m^3].
\] (8.1)

An empirical formula is available for the density of the solidified waste, which is vitrified with the glass frit, PF798 [Yamashita, et al., 1995; Sasage, et al., 1995]. It is formulated as

\[
\rho_{WG} = 1.230 \left( \frac{M_w}{M_w + M_G} \right) + 2.419 \quad [kg/m^3].
\] (8.2)

---

\(^{18}\) See Table E.1 in Appendix E
\(^{19}\) See Appendix D for input data cards of MOCUP
\(^{20}\) See Table E.2 in Appendix E
Substituting (8.2) into (8.1) and for \( V_C = 0.82 \text{ m}^3 \), following are obtained:

\[
(M_W + M_G)^2 - (2992.2M_W + 1983.6M_G) \leq 0
\]  
(8.3)

\[
(M_W + M_G)^2 - (2393.7M_W + 1586.9M_G) \geq 0
\]  
(8.4)

By applying the empirical formula for \( \rho_{WG} \), two quadratic inequalities for the constraint on the solidified waste volume limit are obtained. LP model, however, requires constraints of a linear form and thus approximates of (8.3) and (8.4) which are in a linear form need to be employed.

![Figure 8.2 Quadratic equalities and linear approximates for the filled waste volume constraint](image)

In Figure 8.2, the quadratic forms and its linear approximates are plotted for entire waste loading region, 0% ~ 100%, even though the empirical formula was developed based on waste glass samples with a limited fractional range of waste loading, i.e., between \(~20\%\) and \(~40\%\). Dots represent the quadratic equation and the solid line is its linear approximate. It is observed that the linear approximate taken does not deviate significantly from the quadratic form, further by doing so, the optimum waste loading (here, maximum waste loading) could be estimated more conservatively. In this chapter,
thus, the formulation of the filled waste volume limit introduced in section 7.4 is replaced by followings:

\[
M_w + 1.508M_G \leq 29922 \quad (8.5)
\]

\[
M_w + 1.508M_G \geq 23937 \quad (8.6)
\]

8.3.2 LP Problem for HLW Generated from Separation Process of LWR-SNF

The compositions of HLW generated from separation process of LWR-SNF and the glass frit developed (PF798) by JNC are listed in Table 8.1 and Table 8.2. Based on Table 8.1 and Table 8.2 and the revised formulation of the filled waste volume, LP problem is constructed and solved by the graphical solution method.

Figure 8.3 shows the graphical solution of the LP problem constructed for HLW generated from the separation process of LWR-spent fuel. It is found that the constraint on the heat generation rate determines the optimum values of the objective function. Up to 273 kg of HLW can be loaded into a canister. The mass of glass frit ranges between 1,406.3 kg and 1,516.7 kg, which corresponds to 16.3 wt% and 15.3 wt% waste loading. The lower limit of waste loading is 255 kg (15.0 wt%), and this is determined by the intersection of two constraints on the high frit loading limit and the lower volume limit of the filled waste glass.

Table 8.1 Composition of HLW Generated from Separation Process of LWR-SNF with a Burnup of 33,000 MWd/MT and a 25-year cooling period 21

<table>
<thead>
<tr>
<th>Element</th>
<th>wt%</th>
<th>Element</th>
<th>wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se</td>
<td>8.752E-04</td>
<td>Ce</td>
<td>3.651E-02</td>
</tr>
<tr>
<td>Rb</td>
<td>5.679E-03</td>
<td>Pr</td>
<td>1.719E-02</td>
</tr>
<tr>
<td>Sr</td>
<td>1.024E-02</td>
<td>Nd</td>
<td>6.205E-02</td>
</tr>
<tr>
<td>Y</td>
<td>7.213E-03</td>
<td>Pm</td>
<td>3.373E-06</td>
</tr>
<tr>
<td>Zr</td>
<td>5.845E-02</td>
<td>Sm</td>
<td>1.326E-02</td>
</tr>
<tr>
<td>Nb</td>
<td>1.430E-07</td>
<td>Eu</td>
<td>1.738E-03</td>
</tr>
<tr>
<td>Mo</td>
<td>5.127E-02</td>
<td>Gd</td>
<td>1.861E-03</td>
</tr>
<tr>
<td>Tc</td>
<td>1.187E-02</td>
<td>Na</td>
<td>3.231E-01</td>
</tr>
<tr>
<td>Ru</td>
<td>3.270E-02</td>
<td>P</td>
<td>5.694E-03</td>
</tr>
<tr>
<td>Rh</td>
<td>7.181E-03</td>
<td>Fe</td>
<td>6.085E-03</td>
</tr>
<tr>
<td>Pd</td>
<td>1.936E-02</td>
<td>Ni</td>
<td>1.594E-03</td>
</tr>
<tr>
<td>Ag</td>
<td>1.076E-03</td>
<td>Cr</td>
<td>1.579E-03</td>
</tr>
<tr>
<td>Cd</td>
<td>1.499E-03</td>
<td>U</td>
<td>1.304E-04</td>
</tr>
<tr>
<td>Sn</td>
<td>1.319E-03</td>
<td>Np</td>
<td>1.280E-06</td>
</tr>
<tr>
<td>Te</td>
<td>7.290E-03</td>
<td>Pu</td>
<td>2.278E-05</td>
</tr>
<tr>
<td>Cs</td>
<td>3.162E-02</td>
<td>Am</td>
<td>2.458E-06</td>
</tr>
<tr>
<td>Ba</td>
<td>3.060E-02</td>
<td>Cm</td>
<td>1.672E-08</td>
</tr>
<tr>
<td>La</td>
<td>1.871E-02</td>
<td>O</td>
<td>2.323E-01</td>
</tr>
</tbody>
</table>

21 Process chemicals (Na, P) and corrosion products (Fe, Ni, Cr) are taken into account based on [PNC SN8410 90-061, 1990]. For isotopic composition of radionuclides, see Table E.1 in Appendix E.
Table 8.2  Composition of the Glass Frit, PF798, Developed by JNC [Yamashita, et al., 1995]

<table>
<thead>
<tr>
<th>Oxide</th>
<th>wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>62.30</td>
</tr>
<tr>
<td>B₂O₃</td>
<td>19.00</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>6.70</td>
</tr>
<tr>
<td>Li₂O</td>
<td>4.00</td>
</tr>
<tr>
<td>CaO</td>
<td>4.00</td>
</tr>
<tr>
<td>ZnO</td>
<td>4.00</td>
</tr>
</tbody>
</table>

Figure 8.3  The graphical solution of the linear programming problem for HLW generated from separation process of LWR-SNF

Figure 8.3 shows the graphical solution of the LP problem constructed for HLW generated from the separation process of LWR-spent fuel. It is found that the constraint on the heat generation rate determines the optimum values of the objective function. Up to 273 kg of HLW can be loaded into a canister. The mass of glass frit ranges between 1,406.3 kg and 1,516.7 kg, which corresponds to 16.3 wt% and 15.3 wt% waste loading. The lower limit of waste loading is 255 kg (15.0 wt%), and this is determined by the intersection of two constraints on the high frit loading limit and the lower volume limit of the filled waste glass.
8.3.3 LP Problem for HLW Generated from Separation Process of ATW-SNF

Table 8.3 Composition of HLW Generated from Separation Process of ATW-SNF with a Burnup of 27.2 wt% and a 25-year cooling period

<table>
<thead>
<tr>
<th>Element</th>
<th>wt%</th>
<th>Element</th>
<th>wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se</td>
<td>1.682E-04</td>
<td>Ce</td>
<td>1.065E-02</td>
</tr>
<tr>
<td>Br</td>
<td>2.890E-04</td>
<td>Pr</td>
<td>2.785E-02</td>
</tr>
<tr>
<td>Rb</td>
<td>5.409E-03</td>
<td>Nd</td>
<td>8.420E-02</td>
</tr>
<tr>
<td>Sr</td>
<td>1.557E-03</td>
<td>Pm</td>
<td>9.050E-06</td>
</tr>
<tr>
<td>Y</td>
<td>5.298E-03</td>
<td>Sm</td>
<td>2.840E-02</td>
</tr>
<tr>
<td>Zr</td>
<td>6.664E-02</td>
<td>Eu</td>
<td>3.060E-03</td>
</tr>
<tr>
<td>Mo</td>
<td>8.345E-02</td>
<td>Gd</td>
<td>3.792E-03</td>
</tr>
<tr>
<td>Tc</td>
<td>2.170E-02</td>
<td>Tb</td>
<td>5.130E-05</td>
</tr>
<tr>
<td>Ru</td>
<td>8.154E-02</td>
<td>Dy</td>
<td>3.035E-05</td>
</tr>
<tr>
<td>Rh</td>
<td>2.589E-02</td>
<td>Na</td>
<td>1.247E-01</td>
</tr>
<tr>
<td>Pd</td>
<td>4.276E-02</td>
<td>P</td>
<td>2.197E-03</td>
</tr>
<tr>
<td>Ag</td>
<td>7.345E-03</td>
<td>Fe</td>
<td>2.348E-03</td>
</tr>
<tr>
<td>Cd</td>
<td>2.749E-03</td>
<td>Ni</td>
<td>6.149E-04</td>
</tr>
<tr>
<td>In</td>
<td>2.107E-04</td>
<td>Cr</td>
<td>6.093E-04</td>
</tr>
<tr>
<td>Sn</td>
<td>7.507E-04</td>
<td>U</td>
<td>5.396E-05</td>
</tr>
<tr>
<td>Sb</td>
<td>1.429E-04</td>
<td>Np</td>
<td>6.526E-05</td>
</tr>
<tr>
<td>Te</td>
<td>3.022E-03</td>
<td>Pu</td>
<td>1.667E-03</td>
</tr>
<tr>
<td>Cs</td>
<td>9.261E-02</td>
<td>Am</td>
<td>2.856E-04</td>
</tr>
<tr>
<td>Ba</td>
<td>2.510E-02</td>
<td>Cm</td>
<td>4.632E-05</td>
</tr>
<tr>
<td>La</td>
<td>3.279E-02</td>
<td>O</td>
<td>2.100E-01</td>
</tr>
</tbody>
</table>

Figure 8.4 The graphical solution of the linear programming problem for HLW generated from separation process of ATW-SNF
The compositions of HLW generated from separation process of ATW-SNF is listed in Table 8.3 and the graphical solution of the LP problem for HLW generated from the separation process of ATW-spent fuel is illustrated in Figure 8.4. As in Figure 8.3, the optimum waste loading is determined by the constraint on the heat generation rate, but it allows much higher waste loading, 514 kg because the heat generation rate per unit mass of HLW is smaller than that of LWR-HLW. The corresponding glass frit ranges between 1,247 kg and 1,519 kg resulting in 29.2 wt% and 25.3 wt% waste loading, respectively.

8.4 Effects of Composition Variation on the Optimum Waste Loading

In this section, the effects of the variation in the composition of HLW and glass frit on the optimum waste loading are investigated. Under the applied conditions the optimum waste loading estimated for HLW generated from LWR-spent fuel was ~273 kg and that for HLW generated from ATW-spent fuel was ~514 kg. These values vary with applied conditions to HLW to be vitrified. So, in order to know the sensitivity of the optimum waste loading to the composition of HLW or glass frit, parametric studies were undertaken.

8.4.1 Feasible Solution Space Bounded by Composition-Independent Constraints

Feasible solution space is a region that satisfies all constraints of a LP problem, and thus giving optimum values of an objective function. As shown in Figure 8.3 and Figure 8.4, a feasible solution of a LP problem can be bounded by several key constraints, while other constraints loosely encompass the feasible solution space (called as “redundant constraints”). Some constraints move with the variation of HLW or glass frit composition, while others remain constant. Among constraints shown in Figure 8.3 or 8.4, those on PCT-limit (Na-, Li-, and B-concentrations), Pu-concentration limit, heat generation rate limit, temperature limit, and homogeneity limit are composition-dependent; constraints on filled canister weight limit, filled waste volume limit, and glass frit loading limit are considered as composition-independent.

The feasible solution space bounded by the composition-independent constraints is shown in Figure 8.5. Those encompassing the feasible solution space are the filled canister weight limit, the lower limit of the filled waste volume, and the high- and the low-frit loading limits. For the given objective function (maximizing \( f = M_w \)) subject to the composition-independent constraints only, the optimum waste loading is determined at the upper corner point of the feasible solution space, which is ~610 kg. This value can be considered as the “maximum achievable” waste loading for the solidification of US-defense HLW because the feasible solution space can not be further expanded by adding any composition-dependent constraints. Figure 8.5 also shows that a minimum waste loading (~255 kg) is set by the composition-independent constraints. This implies that if a composition-dependent constraint does not satisfy the minimum waste loading, waste vitrification is not allowed.

---

22 For isotopic composition of radionuclides, see Table E.2 in Appendix E.
Figure 8.5 The feasible solution space bounded by the composition-independent constraints only for solidification of US-defense HLW

8.4.2 Composition Dependency of PCT-Limit Constraints

Constraints on PCT-limit are derived and formulated in Section 7.4. All those inequalities are a function of both $M_W$ and $M_G$ and pass through the origin. Slopes and signs of inequalities vary with fractional masses of B, Na, and Li in HLW or glass frit.

The formulations for B-, Na-, and Li-concentration limit are shown in (7.56), (7.57), and (7.58), respectively.

The formulations imply that if both coefficients of $M_W$ and $M_G$ have negative signs (e.g., for boron, the fractional mass in both HLW and glass frit are less than 0.43), the inequality lines are plotted like “A” in Figure 8.6. Then, those inequalities always satisfy the feasible solution space bounded by composition-independent constraints. For this case, the constraints on PCT-limit are considered as a redundant. On the other hand, if both coefficients have positive signs (e.g., for boron, the fractional mass in both HLW and glass frit are greater than 0.43), the inequality lines are plotted like “B” in Figure 8.6. Thus there is no feasible solution space satisfying composition-independent constraints and constraints on PCT-limit, i.e., indicating not acceptable for solidification.

When coefficients of two variables have different sign, there are two cases:

- If the coefficient of $M_W$ is positive and that of $M_G$ is negative, inequality line is plotted like “C” in Figure 8.6. If the ratio of the coefficient of $M_W$ to that of $M_G$ is equal to or greater than the slope of the low frit loading, 0.43, this always satisfies the feasible solution space bounded by composition-independent constraints, but in the case that the ratio is less than the slope of the high frit loading, 0.18, the LP problem has no feasible solution space (not acceptable for solidification).
If the coefficient of $M_W$ is negative and that of $M_G$ is positive, inequality lines are plotted like “D” in Figure 8.6. If the ratio of the coefficient of $M_W$ to that of $M_G$ is equal to or less than the slope of the high frit loading, 0.18, those inequalities always satisfy the feasible solution space bounded by composition-independent constraints, but in the case that the ratio is greater than the slope of the low frit loading, the LP problem has no feasible solution space.

By checking the fractional masses of B, Na, and Li in both HLW and glass frit, it is possible to estimate if those constraints are redundant for the feasible solution space bounded by composition-independent constraints (thus, allowing maximum achievable waste loading) or making the solidification of HLW impossible or working as an essential constraint for determining an optimum value.

![Figure 8.6](image)

**Figure 8.6** The variation of the inequality of boron concentration limit with the variation of fractional boron in HLW and glass frit

### 8.4.3 Composition Dependency of Homogeneity Constraint

Homogeneity constraint is formulated in terms of alumina content in Section 7.6 and the formulation is similar to those for PCT-limit constraints except the sign of inequality. It always passes through the origin. If both coefficients of $M_W$ and $M_G$ have positive signs (i.e., fractional masses of Al in HLW and glass frit are greater than 0.02) the inequality line is plotted like “A” in Figure 8.6 and it is redundant for the feasible solution space bounded by the composition-independent constraints. When both coefficients have negative signs (i.e., fractional masses of Al in HLW and glass frit are less than 0.02) the inequality line is plotted like “B” in Figure 8.6, and there is no feasible solution for the
LP problem. For both coefficients with a different sign, the case that the coefficient of $M_W$ is negative and that of $M_G$ is positive corresponds to the inequality line “C” in Figure 8.6. The other way around corresponds to the inequality line “D”.

### 8.4.4 Composition Dependencies of Constraints on Pu-concentration, Heat Generation Rate, and Temperature Limits

In the LP model developed, inequalities for plutonium concentration limit, heat generation limit, and temperature limit are only a function of $M_W$ as shown in Section 7.4 because glass frit contains neither Pu element nor radioactive isotopes emitting heat. So, those inequality lines move up or down according to the fractional mass of Pu or radioactive isotopes in HLW.

For Pu-concentration limit, as long as the fractional mass of Pu in HLW is equal to or less than $3.355 \times 10^{-3}$, this constraint always satisfies the feasible solution space bounded by composition-independent constraints and locates above a line “A” in Figure 8.7. If, however, the fractional mass of Pu in HLW is greater than $8.039 \times 10^{-3}$, this constraint does not satisfy any portion of the feasible solution space bounded by composition-independent constraints and locates under a line “B” in Figure 8.7. Here, $8.039 \times 10^{-3}$ is considered as a “maximum allowable” mass fraction of Pu in HLW for solidification and $3.355 \times 10^{-3}$ or a smaller fraction could be considered as a “favorable” mass fraction of Pu in HLW in order to get the “maximum achievable” waste loading.

For the limit of heat generation rate, as long as the heat generation rate per unit mass of HLW is equal to or less than $2.455$ [W/kg], this constraint always satisfies the feasible solution space bounded by composition-independent constraints and locates above the line “A” in Figure 8.7. If, however, the heat generation rate per unit mass of HLW is greater than $5.882$ [W/kg], inequality line locates under the line “B” in Figure 8.7 and thus HLW solidification is not allowed. Here, $5.882$ [W/kg] is considered as a “maximum allowable” heat generation rate of unit mass of HLW and $2.455$ [W/kg] or a smaller value could be considered as a “favorable” heat generation rate of unit mass of HLW.

![Figure 8.7](image.png) The variation of the inequality lines for plutonium concentration limit, heat generation limit, and temperature limit
A formulation developed in Section 7.4 for the temperature evolution in the center line of a filled canister has a coefficient, which is a function of the heat generation rate of unit mass of HLW. For the regulation and guidelines set for US-defense HLW, it is observed that if the heat generation rate limit satisfies the feasible solution space bounded by the composition-independent constraints, the temperature limit also does. So, this constraint can be considered as a “redundancy” for the constraint of the heat generation limit.

8.4.5 Necessary and Favorable Conditions of Composition-Dependent Constraints

So far, it has been investigated how the inequality lines of the composition-dependent constraints change with the composition variations of HLW and glass frit. From observed results it is found that in order that the LP problem has a solution, each composition-dependent constraint should satisfy a certain condition. Otherwise, the LP problem has no solution, indicating that solidification of HLW is not allowed. Those conditions are considered as “necessary” conditions for solidification of HLW. The investigations, also, show a certain condition that makes a composition-dependent constraint redundant for the feasible solution space bounded by the composition-independent constraints, and thus allowing the objective function of the LP problem to have a “maximum achievable” waste loading. These conditions are considered as “favorable” conditions. The “necessary” conditions of the composition-dependent constraints for HLW solidification are summarized in Table 8.4, and the “favorable” conditions of the composition-dependent constraints for redundancy in Table 8.5.

Table 8.4 Necessary Conditions of Composition-Dependent Constraints for HLW Solidification

<table>
<thead>
<tr>
<th>Constraint</th>
<th>Necessary conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>B-concentration</td>
<td>[x_{W,B} \text{ and } x_{G,B} &lt; 0.43 \text{ or } x_{W,B} &gt; 0.43 \text{ and } x_{G,B} &lt; 0.43, \right(\frac{x_{W,B} - 0.43}{x_{G,B} - 0.43}\right) \geq 0.18 \text{ or} \right(\frac{x_{W,B} - 0.43}{x_{G,B} - 0.43}\right) \leq 0.43 \text{ or} \right(\frac{x_{W,B} - 0.43}{x_{G,B} - 0.43}\right) &gt; 0.43]</td>
</tr>
<tr>
<td>Na-concentration</td>
<td>[x_{W,Na} \text{ and } x_{G,Na} &lt; 0.34 \text{ or } x_{W,Na} &gt; 0.34 \text{ and } x_{G,Na} &lt; 0.34, \right(\frac{x_{W,Na} - 0.34}{x_{G,Na} - 0.34}\right) \geq 0.18 \text{ or} \right(\frac{x_{W,Na} - 0.34}{x_{G,Na} - 0.34}\right) \leq 0.43 \text{ or} \right(\frac{x_{W,Na} - 0.34}{x_{G,Na} - 0.34}\right) &gt; 0.43]</td>
</tr>
<tr>
<td>Li-concentration</td>
<td>[x_{W,Li} \text{ and } x_{G,Li} &lt; 0.24 \text{ or } x_{W,Li} &gt; 0.24 \text{ and } x_{G,Li} &lt; 0.24, \right(\frac{x_{W,Li} - 0.24}{x_{G,Li} - 0.24}\right) \geq 0.18 \text{ or} \right(\frac{x_{W,Li} - 0.24}{x_{G,Li} - 0.24}\right) \leq 0.43 \text{ or} \right(\frac{x_{W,Li} - 0.24}{x_{G,Li} - 0.24}\right) &gt; 0.43]</td>
</tr>
<tr>
<td>Homogeneity</td>
<td>[x_{W,Al} \text{ and } x_{G,Al} &gt; 0.02 \text{ or } x_{W,Al} &gt; 0.02 \text{ and } x_{G,Al} &lt; 0.02, \right(\frac{x_{W,Al} - 0.02}{x_{G,Al} - 0.02}\right) \leq 0.43 \text{ or} \right(\frac{x_{W,Al} - 0.02}{x_{G,Al} - 0.02}\right) &gt; 0.43]</td>
</tr>
<tr>
<td>Pu-concentration</td>
<td>[x_{W,Pu} \leq 8.038 \times 10^{-3}]</td>
</tr>
<tr>
<td>Heat generation rate</td>
<td>[\zeta \leq 5.882 \text{ [W/kg]}]</td>
</tr>
</tbody>
</table>

The graphical solutions of the LP problem shown in Figure 8.3 and 8.4, which are obtained for HLW generated from the ATW fuel cycle, show that all composition-dependent constraints except for the heat generation rate limit (maybe temperature limit also) are favorable (redundant). If the heat generation rate limit were also redundant, the “maximum achievable” waste loading (~610 kg) would be obtained. In order to make the constraint redundant, it is necessary to reduce the heat emission rate of unit mass of HLW down to 2.455 [W/kg]. Reducing the heat emission rate of unit mass of HLW can be
achieved either by diluting the concentration of radioactive isotopes adding non-radioactive materials in HLW or by allowing a longer cooling period for discharged spent fuels. Adding non-radioactive materials in HLW, however, causes an increase in the mass of HLW to be vitrified.

Table 8.5  Favorable Conditions of Composition-Dependent Constraints for Redundancy

<table>
<thead>
<tr>
<th>Constraint</th>
<th>Redundancy Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>B-concentration</td>
<td>$x_{W,B}$ and $x_{G,B} &lt; 0.43$ or $x_{W,B} &gt; 0.43$ and $x_{G,B} &lt; 0.43$, $(x_{W,B} - 0.43)/(x_{G,B} - 0.43) \geq 0.43$ or $x_{W,B} &lt; 0.43$ and $x_{G,B} &gt; 0.43$, $(x_{W,B} - 0.43)/(x_{G,B} - 0.43) \leq 0.18$</td>
</tr>
<tr>
<td>Na-concentration</td>
<td>$x_{W,Na}$ and $x_{G,Na} &lt; 0.34$ or $x_{W,Na} &gt; 0.34$ and $x_{G,Na} &lt; 0.34$, $(x_{W,Na} - 0.34)/(x_{G,Na} - 0.34) \geq 0.43$ or $x_{W,Na} &lt; 0.34$ and $x_{G,Na} &gt; 0.34$, $(x_{W,Na} - 0.34)/(x_{G,Na} - 0.34) \leq 0.18$</td>
</tr>
<tr>
<td>Li-concentration</td>
<td>$x_{W,Li}$ and $x_{G,Li} &lt; 0.24$ or $x_{W,Li} &gt; 0.24$ and $x_{G,Li} &lt; 0.24$, $(x_{W,Li} - 0.24)/(x_{G,Li} - 0.24) \geq 0.43$ or $x_{W,Li} &lt; 0.24$ and $x_{G,Li} &gt; 0.24$, $(x_{W,Li} - 0.24)/(x_{G,Li} - 0.24) \leq 0.18$</td>
</tr>
<tr>
<td>Homogeneity</td>
<td>$x_{W,Al}$ and $x_{G,Al} &gt; 0.02$ or $x_{W,Al} &lt; 0.02$ and $x_{G,Al} &lt; 0.02$, $(x_{W,Al} - 0.02)/(x_{G,Al} - 0.02) \leq 0.18$ or $x_{W,Al} &gt; 0.02$ and $x_{G,Al} &gt; 0.02$, $(x_{W,Al} - 0.02)/(x_{G,Al} - 0.02) \geq 0.43$</td>
</tr>
<tr>
<td>Pu-concentration</td>
<td>$x_{W,Pu} \leq 3.355 \times 10^{-3}$</td>
</tr>
<tr>
<td>Heat generation rate</td>
<td>$\zeta \leq 2.455$ [W/kg]</td>
</tr>
</tbody>
</table>

8.4.6  Effects of Cooling Time on Heat Generation Rate

![Figure 8.8](image.png)  Dependency of heat generation rate on cooling time of discharged spent fuel (HLW generated from separation process of LWR-spent fuel with a burnup of 33,000 MWd/MT; process chemicals and corrosion products based on [PNC SN8410 90-061, 1990])
In Figure 8.3 and Figure 8.4, the optimum waste loadings are limited by the heat generation rate constraint. By reducing the heat generation rate per unit mass of HLW, the inequality line for this constraint moves upward and thus a greater waste loading can be achieved in both Figures. Figure 8.8 shows a dependency of heat emission rate on cooling time and shows that it decreases almost exponentially with cooling time. A dotted line marked as 5.882 [W/kg-HLW] represents a “maximum allowable” heat generation rate in order that HLW solidification is permitted. Corresponding cooling time is ~22 years. Another dotted line marked as 2.455 [W/kg-HLW] is considered as a “favorable” heat generation rate, which enables us to get a “maximum achievable” waste loading, 610kg. Corresponding cooling time is ~60 years and after then the heat generation rate is no longer essential for the HLW solidification.

8.4.7 Effects of Process Chemicals and Corrosion Products on Heat Generation Rate

Heat generation rate can also be reduced by adding non-radioactive materials in HLW and thus diluting radioactive materials contained in unit mass of HLW. In Section 8.3, as non-radioactive materials added in liquid HLW, process chemicals and corrosion products were considered.

Figure 8.9 shows the dependency of heat generation rate on the content of process chemicals and corrosion products in HLW to be solidified. The content of process chemicals and corrosion products in HLW varies from zero wt% to 33.8 wt%, which is corresponding to the design data specified in [PNC SN8410 90-061, 1990], in HLW.

![Figure 8.9 Dependency of heat generation rate on content of process chemicals and corrosion products (HLW generated from separation process of LWR-SNF with a burnup of 33,000 MWd/MT and a 25-year cooling time)](image-url)
It is observed that the heat generation rate decreases almost linearly as the content of process chemicals and corrosion products increases. To reduce the heat generation rate down to a “maximum allowable” heat generation level, ~31 wt% of process chemicals and corrosion products needs to be added, and to reduce down to a “favorable” heat generation level ~56 wt% of those needs to be added, even though this level is out of the design data. As shown in Figure 8.9, to reduce the heat generation rate by adding non-radioactive materials into liquid HLW results in a significant increase in HLW mass.

8.5 Discussions and Conclusions

An implementation of the LP model to the solidification of HLW from the ATW fuel cycle gave two different optimum waste loadings for two high-level waste streams of the ATW fuel cycle with a specified condition.

For HLW from the LWR-spent fuel, essential constraints bounding the feasible solution space are the heat generation rate limit, the lower limit of the filled waste volume, and the high frit loading limit. For HLW from ATW-spent fuel, the filled canister weight limit is important in addition to those for HLW from the LWR-spent fuel.

The feasible solution space, however, is subject to the compositions of HLW and glass frit. The composition of HLW varies with several conditions such as a burnup of discharged spent fuel, cooling time, the amount of added process chemicals. If the compositions of HLW and glass frit do not satisfy any of the “necessary” conditions listed in Table 8.4, the LP problem has no solution. For Pu-concentration limit, HLW from ATW-spent fuel contains about 0.2% of Pu as shown in Table 8.3. If a lower burnup for ATW fuel or a smaller recovery yield for Pu is applied, a greater fraction of Pu in HLW from ATW-spent fuel would result. Thus, when the mass fraction of Pu in HLW gets greater than the “necessary condition” for Pu-concentration in Table 8.4 (~0.8%), the LP problem has no solution.

The optimum waste loadings in both Figure 8.3 and 8.4 are determined by the heat generation rate limit. The difference in the optimum waste loadings arises from the difference in heat generation rate per unit mass of HLW. But, for ATW-spent fuel, since the composition of the discharged ATW-spent fuel is represented by that of the homogenized core at the end of each cycle in the current study, it deviates from the composition of the ATW-spent fuel with a 1/6-discharge fraction.

As illustrated in Figure 8.8 and 8.9, it is found that in order to achieve a higher waste loading, reducing the rate of heat generation per unit mass of HLW is crucial. To reduce heat generation rate per unit mass of HLW, we could allow a longer cooling period for discharged fuel before reprocessing, or we could dilute the concentration of radioactive waste by adding more non-radioactive material such as process chemicals into HLW. The latter, however, causes greater mass of HLW to be solidified. For the former, if we allow a long cooling period for both LWR- and ATW-spent fuel such that the constraint on the heat generation rate is no longer essential, the optimum waste loading will be determined by the intersection of two constraint lines corresponding to filled canister weight limit and low frit loading limit. This enables us to obtain the maximum achievable waste loading (610 kg).

The investigations on characteristics of constraints have shown that all constraints could be categorized into composition-independent and composition-dependent constraints. The feasible solution space bounded by composition-independent constraints
is unaffected by composition variation in HLW or glass frit. In order that the LP problem be solvable, composition-dependent constraints should satisfy at least a part of the feasible solution space bounded by the composition-independent constraints. If all composition-dependent constraints are redundant for the feasible solution space bounded by the composition-independent constraints, the LP problem always has a “maximum achievable” waste loading. Therefore, if conditioning HLW for solidification in a facility is focused on a direction such that “favorable” conditions can be met, it would be desirable in terms of a greater waste loading.

The maximum achievable waste loading determined by two composition-independent constraints will not change as long as regulations and guidelines specified in [DOE/EM-0093, 1996; Brown, 2002] are not revised.

As the next step of this study, the results and observations obtained in this chapter will be used to investigate the impacts of the ATW fuel cycle on the Yucca Mountain Repository (YMR) capacity.
9 Expansion of the Yucca Mountain Repository Capacity by ATW Fuel Cycle

9.1 Introduction

To investigate the impacts of the ATW fuel cycle on a geologic repository, two models have been developed in the previous chapters: (1) the model for the cycle-by-cycle simulation of fuel inventory evolution in the ATW fuel cycle and (2) the model for the determination of an optimized composition of solidified high-level waste for a given set of constraints.

With the model for the ATW fuel cycle, the amount of FPs and TRU contained in the waste streams of the ATW fuel cycle and the composition of TRU waste were quantified. With the model for HLW solidification, an optimized composition of vitrified HLW generated from the ATW fuel cycle were obtained.

In this Chapter, based on results and observations made in Chapter 8, the impacts of the ATW fuel cycle on the Yucca Mountain Repository (YMR) capacity is quantitatively evaluated.

9.2 Statutory YMR Capacity

According to [DOE/RW-0539-1, 2002], the YMR has a 70,000 MT capacity, 63,000 MT designated for commercial-spent nuclear fuel (CSNF) and the remaining 7,000 MT reserved for defense HLW and DOE-owned spent nuclear fuel (SNF). To be stored in the YMR, all waste is sealed in highly durable containers, referred to as waste packages. The YMR is currently projected to store 11,760 waste packages: (1) 7,886 waste packages for CSNF, (2) 3,521 waste packages for co-disposal, which contains 5 canisters of vitrified HLW and 1 canister of DOE-owned SNF, and (3) 353 waste packages for naval SNF. For CSNF, it is estimated that a waste package contains 7.989 MT of CSNF on average.

9.3 Waste Packages Generated from HLW Solidification

Impacts on the YMR capacity by ATW fuel cycle can be evaluated by comparing the number of waste packages for the direct disposal of CSNF with that for processing CSNF by the ATW system. The number of waste packages generated from solidification of a certain amount of HLW is calculated as

\[ \text{Number of waste packages} = \frac{M_{HLW}}{n_{can} m_{loaded}} \]  

(9.1)

where \( M_{HLW} \) = a total mass of HLW to be solidified, [MT],

\( n_{can} \) = the number of canisters contained in a waste package,

\( m_{loaded} \) = the mass of HLW loaded into a canister, [MT].

In the YMR, for HLW a waste package contains 5 HLW canisters, and thus the number of waste packages can be determined based on both the total mass of HLW to be solidified and the mass of HLW loaded into a canister. Figure 9.1 illustrates an overview of waste-package generation paths from the ATW fuel cycle. In addition to FPs and TRU waste generated from separation processes, process chemicals, corrosion products, and oxygen are vitrified together in a vitrification facility. As mentioned in Chapter 8,
gaseous fission products are treated separately. Waste packages containing 5 canisters in each are finally disposed of in the YMR.

**Figure 9.1** Overview of waste-package generation paths from the ATW fuel cycle

**Table 9.1** Constituents and Mass of HLW for Table 8.2 and Table 8.3, and Waste Packages Generated: Per 1 MT of SNF, 22.3 kg-Na, 0.393 kg-P, 4.2 kg-Fe, 1.1 kg-Ni, and 1.09 kg-Cr Contained in HLW, 25-Year Cooling after Discharge before Reprocessing Applied

<table>
<thead>
<tr>
<th>Constituent</th>
<th>HLW from processing 86,317 MT of LWR-spent fuel (Table 8.2)</th>
<th>HLW from processing 6,579 MT of ATW-spent fuel (Table 8.3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission products (FP)</td>
<td>3,102.1 MT</td>
<td>895 MT</td>
</tr>
<tr>
<td>Gaseous FP (Kr, I, Xe)</td>
<td>542.6 MT</td>
<td>125.7 MT</td>
</tr>
<tr>
<td>TRU</td>
<td>0.935 MT</td>
<td>2.439 MT</td>
</tr>
<tr>
<td>Process chemicals</td>
<td>1,958.8 MT</td>
<td>149.3 MT</td>
</tr>
<tr>
<td>Corrosion products</td>
<td>55.2 MT</td>
<td>4.2 MT</td>
</tr>
<tr>
<td>Oxygen</td>
<td>1,383.8 MT</td>
<td>246.0 MT</td>
</tr>
<tr>
<td>Total mass of HLW</td>
<td>5,957.9 MT</td>
<td>1,171.3 MT</td>
</tr>
<tr>
<td>Optimum waste loading</td>
<td>273 kg</td>
<td>514 kg</td>
</tr>
<tr>
<td>Number of canisters</td>
<td>21,904</td>
<td>2279</td>
</tr>
<tr>
<td>Number of waste packages</td>
<td>4,381</td>
<td>456</td>
</tr>
<tr>
<td>Total waste packages</td>
<td>4,837</td>
<td></td>
</tr>
</tbody>
</table>
Applying (9.1) to the results obtained in Chapter 8, a total of 4,837 waste packages are generated. Constituents and mass of HLW corresponding to Table 8.2 and Table 8.3 are listed in Table 9.1 as well as waste packages. About 90.6% of waste packages is generated from processing of LWR-spent fuel while 9.4% of those generated from processing of ATW-spent fuel. The ratio of waste packages from LWR-spent fuel to those from ATW-spent fuel is two times greater than that of the respective total HLW mass. This is due to the difference in their optimum waste loadings shown in Figure 8.3 and Figure 8.4.

This observation, however, is subject to the assumptions made in Chapter 8.

9.3.1 Determination of Mass of HLW for Solidification

Determination of the mass of process chemicals and corrosion products included in the waste stream is as important as those of fission products and TRU when the composition of the solidified waste is determined. The mass loading of HLW constituents and heat generation rate per unit mass of solidified HLW could significantly be affected by those materials.

In Chapter 8, design data of a PUREX plant of JNC for determination of process chemicals and corrosion products contained in the waste streams of the ATW fuel cycle were used. This data set represents only one of many possibilities. The amount of those materials in the waste streams depends on the design of separation facility and/or waste vitrification facility. However, the detailed design of those facilities for the ATW system is not yet available [DOE/RW-0519, 1999].

In this study, to identify major components of process chemicals and corrosion products that could be included in the waste stream, several design data of reprocessing plants were examined. Two major components which could significantly increase the mass of HLW were figured out. Sodium as mentioned in Chapter 8 is a major process chemical in the reprocessing plant of JNC [PNC SN8410 90-061, 1990]. It arises from successive washing processes of solvent containing contaminants in a PUREX process because a well-designed PUREX plant aims for as complete recycle of solvent as possible to minimize costs of solvent makeup and disposal [Benedict, et al., 1981]. The other is gadolinium which is used as homogeneous poison for criticality control in the reprocessing plant of the Allied General Nuclear Services in the US [Benedict, et al., 1981].

Sodium concentration of liquid HLW from the PUREX process of JNC is 44.6 g/liter, and 500 liter of liquid HLW is generated from processing 1 MT of LWR-spent fuel [PNC SN8410 90-061, 1990]. Liquid HLW generated from the US-reprocessing plant contains 24 g/liter of gadolinium, and 1,375 liter of liquid HLW is generated from processing of 1 MT of nuclear spent fuel [Benedict, et al., 1981]. Other process chemicals and corrosion products are of a relatively small concentration compared to those of sodium and gadolinium in liquid HLW and thus neglected for the considerations to be followed.

For determination of the HLW mass before final solidification, among many possible cases, two founding cases are considered. In case (1), hypothetically no process chemicals are assumed to be contained in liquid HLW. In case (2), as process chemicals Na and Gd are contained based on design data specified in [PNC SN8410 90-061, 1990; Benedict, et al., 1981]. Case (1) results in a minimum amount of HLW that would be generated from the ATW fuel cycle. This HLW has a relatively high heat emission rate.
per unit mass in early cooling time and thus makes waste loading into a canister lower. Case (2) results in a greatly increased amount of HLW compared to Case (1), but a relatively low heat emission rate per unit mass in early cooling time and thus a greater mass loading of HLW in a canister.

For Case (1), 3,208 MT of HLW oxides from separation of LWR-spent fuel and 962 MT of HLW oxides from separation of ATW-spent fuel are vitrified after removing gaseous FPs and adding oxygen. The material balance of Case (1) is shown in Table 9.2.

For Case (2), 9,086 MT of HLW oxides from separation of LWR-spent fuel and 1,410 MT of HLW oxides from separation of ATW-spent fuel are solidified after removing gaseous FPs and adding oxygen. The material balance of Case (2) is shown in Table 9.3. The mass of process chemicals added in LWR reprocessing is relatively large, compared with that added in ATW partitioning because the throughput of LWR reprocessing is large due to large amount of uranium.

Due to the compositional variation of HLW with cooling time and subsequent change in oxide forms of HLW constituents, total mass of HLW could vary within a range of several metric tons with a cooling time of the spent nuclear fuel.

**Table 9.2  Constituents and Mass of HLW for Case (1): No Process Chemicals Contained in HLW, 25-Year Cooling after Discharge before Reprocessing Applied**

<table>
<thead>
<tr>
<th>Constituent</th>
<th>HLW from processing 86,317 MT of LWR-spent fuel</th>
<th>HLW from processing 6,579 MT of ATW-spent fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission products (FP)</td>
<td>3,102.1 MT</td>
<td>895 MT</td>
</tr>
<tr>
<td>Gaseous FP (Kr, I, Xe)</td>
<td>542.6 MT</td>
<td>125.7 MT</td>
</tr>
<tr>
<td>TRU</td>
<td>0.935 MT</td>
<td>2.439 MT</td>
</tr>
<tr>
<td>Sodium</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Gadolinium</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Oxygen</td>
<td>647.7 MT</td>
<td>189.9 MT</td>
</tr>
<tr>
<td>Subtotal</td>
<td>3,207.8 MT</td>
<td>961.7 MT</td>
</tr>
<tr>
<td>Total mass of HLW</td>
<td></td>
<td>4,169.5 MT</td>
</tr>
</tbody>
</table>

**Table 9.3  Constituents and Mass of HLW for Case (2): 22.3 kg Na/MT-SNF and 33 kg Gd/MT-SNF Contained in HLW, 25-Year Cooling after Discharge before Reprocessing Applied**

<table>
<thead>
<tr>
<th>Constituent</th>
<th>HLW from processing 86,317 MT of LWR-spent fuel</th>
<th>HLW from processing 6,579 MT of ATW-spent fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission products (FP)</td>
<td>3,102.1 MT</td>
<td>895 MT</td>
</tr>
<tr>
<td>Gaseous FP (Kr, I, Xe)</td>
<td>542.6 MT</td>
<td>125.7 MT</td>
</tr>
<tr>
<td>TRU</td>
<td>0.935 MT</td>
<td>2.439 MT</td>
</tr>
<tr>
<td>Sodium</td>
<td>1,924.9 MT</td>
<td>146.7 MT</td>
</tr>
<tr>
<td>Gadolinium</td>
<td>2,848.5 MT</td>
<td>217.1 MT</td>
</tr>
<tr>
<td>Oxygen</td>
<td>1,752.2 MT</td>
<td>274.1 MT</td>
</tr>
<tr>
<td>Subtotal</td>
<td>9,085.6 MT</td>
<td>1,409.7 MT</td>
</tr>
<tr>
<td>Total mass of HLW</td>
<td></td>
<td>10,495.3 MT</td>
</tr>
</tbody>
</table>
### 9.3.2 Optimum Waste Loadings and Waste Packages

In Chapter 8, it is identified that most of the composition-dependent constraints except on the heat generation limit are redundant for the feasible solution space bounded by composition-independent constraints in solidification of HLW generated from ATW fuel cycle. Thus, optimum waste loadings in a canister are determined by the constraint on heat generation rate limit.

Figure 9.2 and 9.3 show the variation of heat generation rate with cooling time for Case (1) and Case (2), respectively.

Figure 9.2 implies that HLW generated from reprocessing LWR-spent fuel should be cooled at least ~47 years for Case (1) and ~11 years for Case (2) in order that the heat generation rate reduces down to a “maximum allowable” level, 5.882 [W/kg-HLW]. To obtain a “maximum achievable” waste loading (610 kg, 30 wt%), the HLW is required to be cooled ~82 years for Case (1) and ~40 years for Case (2), at which the heat generation rate becomes 2.455 [W/kg-HLW].

From Figure 9.3, it is estimated that HLW from reprocessing ATW-spent fuel should be cooled at least ~12 years for Case (1) and ~8 years for Case (2) in order that the heat generation rate reduces down to 5.882 [W/kg-HLW]. To obtain the “maximum achievable” waste loading (610 kg), the HLW is required to be cooled ~39 years for Case (1) and ~25 years for Case (2), respectively.

![Figure 9.2](image)

**Figure 9.2** Heat generation rate of HLW generated from reprocessing LWR-spent fuel with cooling time of discharged LWR-spent fuel

For Case (1), assuming a sufficiently long cooling time such that the heat generation rate per unit mass is no longer essential in determination of optimum waste loading (thus allowing a “maximum achievable” waste loading, 610 kg), 1,365 waste packages are...
generated for a total 4,169.5 MT of HLW oxides. This case could be considered as an “ideal” case in terms of minimizing waste packages. For a minimum cooling time such that the optimum waste loading is identical with a minimum waste loading (255 kg), 3,270 waste packages are generated.

For Case (2), if we solidify these HLW after a minimum cooling time, it gives 8,232 waste packages for 10,495.3 MT of HLW oxides. This case could be an example of unfavorable cases in terms of minimizing waste packages. For a sufficiently long cooling time, 3,435 waste packages are generated.

These observations indicate that reducing the amount of process chemicals added and allowing a sufficient cooling time before solidification are effective in reducing the number of waste packages.

![Graph](image)

**Figure 9.3** Heat generation rate of HLW generated from reprocessing ATW-spent fuel with cooling time of discharged ATW-spent fuel

### 9.3.3 Reduction of the Number of Waste Packages

Based on the statutory YMR capacity, it is estimated that a waste package designed for CSNF from light-water reactors contains 7.989 MT of CSNF on average, and this estimation results in 10,805 waste packages for 86,317 MT of LWR-CSNF.

Reduction of the number of waste packages containing 86,317 MT of CSNF by the ATW fuel cycle can be evaluated by comparing it with the number of waste packages generated from the ATW fuel cycle.

In Case (1) assuming a sufficiently long cooling time, 10,805 waste packages containing 86,317 MT of LWR-CSNF would be reduced to 1,365 waste packages containing vitrified HLW or by a factor of 7.916. In Case (2) assuming a minimum
cooling time, the total number of waste packages is reduced to 8,232 waste packages or by a factor of 1.313.

Reduction of waste packages can also be evaluated on the electricity generation basis. According to [Form RW-859, 2002; DOE/EIA-0384, 2004], US cumulative spent fuel discharge is 47,023.4 MT and corresponding cumulative electricity generation is 1,597.0 GWe-year as of 2002. Assuming that the spent fuel generation per GWe-year is 27.03 MT/GWe-year, which is estimated for the year 2002, a total of 3,050.7 GWe-year electricity would be generated from 86,317 MT of CSNF. This indicates that 3.542 waste packages need to be disposed of at the YMR per 1 GWe-year electricity generation.

In the case that 86,317 MT of CSNF is processed by the ATW system, the ATW system is designed to produce 752.3 GWe-year from 900 MT of TRU separated from 86,317 MT of CSNF. Consequently, a total of 3,803 GWe-year electricity would be generated from 86,317 MT of CSNF by employing the ATW system. 1 GWe-year electricity generates 0.359 waste packages in Case (1) (for 1,365 waste packages resulting from 3,803 GWe-year electricity generation) and 2.165 waste packages in Case (2) (for 8,232 waste packages resulting from 3,803 GWe-year electricity generation).

Thus, the number of waste packages per unit electricity generation is reduced by a factor of 9.866 for Case (1) and by a factor of 1.636 for Case (2).

### 9.4 Expansion of the YMR Capacity by ATW Fuel Cycle

In the present plans for YMR, of a total of 11,760 waste packages, 7,886 waste packages (67%) are for CSNF while 3,874 waste packages (33%) are in the defense HLW category. If the reduction factors evaluated in the previous section are applied to CSNF waste packages, 7,886 CSNF waste packages are reduced to: (1) 996 packages for a reduction factor of 7.916 and (2) 6,006 packages for a reduction factor of 1.313. So, a total of 11,760 waste packages are reduced to 4,870 waste packages (by a factor of 2.415) for (1), and 9,880 waste packages (by a factor of 1.190) for (2).

For a geological repository for only CSNF waste, the expansion factor of the repository capacity would be the same as the reduction factor of CSNF waste packages.

On electricity generation basis, the reduction factors, 9.866 for Case (1) and 1.636 for Case (2), are still valid for the YMR as long as the defense HLW keeps its statutory limit. This implies that the current YMR supports 2,188 GWe-year electricity generation from 63,000 MT of CSNF for a direct disposal case, but by employing the ATW system it could support nearly ten times greater electricity generation for Case (1) but only 1.6 times greater electricity generation for Case (2).

### 9.5 Discussions

In this chapter, two reference cases were investigated to evaluate expansion of the YMR capacity made possible by the ATW fuel cycle.

The extent of the YMR capacity expansion evaluated here is based on many assumptions. When a sufficient cooling time is assumed for discharged spent fuels, however, the number of waste packages resulted from a HLW solidification process is solely determined based on the mass of HLW. Thus, for a more reliable estimation of waste packages, the mass of HLW to be solidified should be quantified accurately.

The mass of HLW generated from the ATW fuel cycle is a function of several variables such as burnup, recovery yield, the amount of process chemicals added from
separation processes, and the amount of process chemicals removed from conditioning processes of HLW for final solidification.

The amount of FPs contained in LWR-spent fuel increases approximately linearly with a burnup. In the current study, a burnup of 33,000 MWd/MT was assumed for 86,317 MT of LWR-spent fuel. If a greater burnup were applied (implying a larger electricity generation from 86,317 MT of LWR-SNF and/or a delayed accumulation of LWR-SNF), more FPs would be contained in HLW, resulting in an increased number of waste packages. On the basis of electricity generation, however, the reduction factor of waste packages would not be affected because electricity generation also increases by an increased burnup.

The amount of actinides (U+TRU) included in HLW depends on a recovery yield of an actinide isotope in separation processes. Here, for LWR-spent fuel, a recovery yield of 99.995% for uranium and 99.9% for TRU were applied, and for ATW-spent fuel, a recovery yield of 99.9% is applied to all actinides. If a separation process for LWR-spent fuel or ATW-spent fuel has a smaller recovery yield for actinides, more mass of actinides would be contained in HLW.

Process chemicals to be included HLW depends on a design of the reprocessing plant. A PUREX plant design in Japan shows that a significant amount of Na is included in HLW from reprocessing LWR-spent fuel, while that in US shows that a significant amount of Gd is contained in HLW. In a German reprocessing plant a significant amount of salt is added to the HLW. So, the amount of process chemicals to be added in HLW needs to be quantified reliably based on a specific design of reprocessing plants used for processing the LWR and the ATW fuels.

The amount of process chemicals included in HLW could also be changed in the process of conditioning HLW before final solidification. A certain amount of chemicals could be added in liquid HLW for adjustment of pH, or some fraction of process chemicals added from separation process could be removed before final solidification. So, for determination of the mass of process chemicals, the design of a vitrification facility employed by the ATW system should also be taken into account.

Among the assumptions made in this model, the zirconium used as ATW fuel matrix was assumed to be recycled with a recovery yield of 100% for ATW fuel fabrication. If we allow some fractional loss of zirconium to the waste stream, more HLW would be generated from reprocessing ATW-spent fuel.

Therefore, a total mass of HLW determined for Case (1) can vary with a burnup and recovery yields applied to actinides and zirconium, and that for Case (2) can vary with all variables aforementioned.

The number of waste packages evaluated for Case (1) represents a scenario in which a sophisticated effort is made to minimize process chemicals contained in liquid HLW. This could be achieved by designing a reprocessing plant, from which HLW with zero-process chemicals is produced, or building a vitrification facility, in which process chemicals are completely removed from HLW before final solidification. On the other hand, the number of waste packages evaluated for Case (2) represents a scenario that could be the result of a lack of special efforts to reduce the amount of process chemicals in HLW.
Summary and Conclusions

In this study, the TRU inventory contained in LWR-SNF and the waste package volume associated with LWR-SNF by ATW fuel cycle have been quantified. A multi-cycle burnup code for ATW system has been developed. For determination of the number and composition of waste packages related to LWR-SNF, a linear programming model has been developed.

Features of mathematical model and computer code, WACOM, developed for the analysis of the ATW fuel cycle include the following:

- It is capable of a cycle-by-cycle calculation of fuel inventory evolution in a homogeneous ATW transmuter with fuel recycling.
- It takes into account constraints on the transmuter core design and the variation with recycling, the neutron spectrum and, hence, effective one-group cross sections in the transmuter core.
- The numerical solution, predictor-corrector method, chosen for the calculation of time-dependent fuel inventory evolution within a cycle was validated by comparing with ORIGEN2.1 code.
- A linear interpolation scheme implemented in WACOM to account for the variation of neutronics in transmuter core was found to be adequate.
- WACOM was benchmarked against REBUS-3 code for a reference LBE-cooled transmuter with multi-cycles. TRU consumption rate calculated by WACOM well matched that evaluated by REBUS-3 for the same burnup reactivity loss.
- The time it takes WACOM to simulate the complete ATW transmutation campaign is a few seconds while the time it would have taken “commercial” codes such as MOCUP to do so is at least several hours.

Based on a reference ATW plant deployment scenario that is using LBE-cooled subcritical cores, the reduction of TRU included in LWR-spent fuel was estimated and following observations were made:

- TRU waste generated from the reference ATW plant operations is estimated to be 5.3 MT, which indicates that the mass of TRU contained in 86,317 MT of LWR-SNF can be reduced to a factor of 170. This value, however, depends on the assumed ATW operating mode and a greater reduction may be possible by employing other operating modes and transmuter design options.
- The toxicity of LWR-TRU can be decreased more than a factor of 40 at the end of the reference ATW plant operation. After ~10,000 years since TRU waste generation, the overall toxicity reduction factor would increase up to ~170 due to the decay effect of TRU isotopes with half-lives shorter than 10,000 years.
- The toxicity reduction factors of several TRU isotopes such as $^{239}$Pu, $^{237}$Np, and $^{241}$Am are greater than the overall mass reduction factor. This implies that the ATW plant deployment can decrease not only the toxicity of TRU isotopes but also long-lived radionuclides, such as $^{237}$Np, which determine long-term radiological impact of the repository in a preferential manner, compared to other TRU isotopes.
With quantification of FPs and TRU contained in HLW streams generated from the ATW fuel cycle, the following were obtained:

- From reprocessing 86,317 MT of LWR-spent fuel, 3,102 MT of FPs and 0.9 MT of TRU are evaluated to be contained in the high-level waste stream.
- 895 MT of FPs and 2.4 MT of TRU are evaluated to be contained in the HLW generated from the separation process of ATW-spent fuel.
- The combined mass of FPs and TRU contained in the last transmuter, which is to be directly disposed of in a geological repository at the end of ATW operation, is likely to be less than 2.6 MT. The ratio of TRU and FPs depends on an applied burnup for the last transmuter.

A linear-programming model for determination of an optimized composition of solidified HLW was developed for a given set of regulations and guidelines, and it was applied to a US-defense HLW solidification. The optimum waste loading obtained by the LP model was compared with the maximum allowable waste loading estimated by the Defense Waste Processing Facility and it was found that those are in good agreement. The LP model was, then, applied to the solidification of HLW generated from the ATW fuel cycle and the following were found:

- Liquid HLW generated from reprocessing LWR- or ATW-spent fuel contains process chemicals and corrosion products as well as FPs and TRU, and those exist in a form of oxide. Determination of process chemicals and corrosion products contained in liquid HLW plays a key role for the quantification of overall HLW mass that needs to be solidified.
- As an example for quantification of HLW, process chemicals and corrosion products in HLW were determined based on the design data of a PUREX plant of Japan. The mass of HLW from 86,317 MT of LWR-spent fuel was evaluated to be 5,958 MT and that from 6,579 MT of ATW-spent fuel was evaluated to be 1,171 MT. The optimum waste loadings for the HLW from LWR-spent fuel and that from ATW-spent fuel are 273 kg and 514 kg per canister, respectively. The difference is due to the heat generation rate per unit mass of HLW.
- Parametric studies show that the heat generation rate per unit mass of HLW decreases exponentially with cooling time of discharged fuel and linearly with the fraction of non-radioactive materials such as process chemicals and corrosion products. This indicates that when the optimum waste loadings are dictated by the heat generation rate limit, a greater waste loading could be achieved by allowing a sufficiently long-cooling time or by diluting the concentration of radioactive materials in HLW with non-radioactive materials.
- Investigations of the properties of constraints implemented in the LP model show that the constraints could be categorized into composition-independent and composition-dependent constraints. By checking whether the composition of HLW and glass frit satisfy all “necessary” conditions or not, we could determine if the LP problem is solvable or not. In a case that the composition-dependent constraints satisfy all “favorable” conditions, the optimum value of the LP problem is identical to the maximum achievable waste loading (here, 610 kg).
To span the number of waste packages that could be produced from reprocessing 86,317 MT of LWR-spent fuel by the ATW fuel cycle and to evaluate resulting repository expansion effects, two reference cases were investigated. One case corresponds to a sophisticated effort to minimize process chemicals contained in liquid HLW. The other case would result from a lack of special efforts to reduce the amount of process chemicals in HLW. The following are concluded:

- For the former case, assuming a sufficiently long cooling time, 10,805 waste packages containing 86,317 MT of LWR-spent fuel could be reduced by the ATW to 1,365 waste packages containing vitrified HLW, i.e., by a factor of 7.916. For the latter case, assuming a minimum cooling time, a total 10,805 waste packages could be reduced to 8,232, i.e., by a factor of 1.313.

- On the basis of electricity generation, the number of waste packages per unit electricity generation is reduced by a factor of 9.866 for the former case and by a factor of 1.636 for the latter case.

- For the YMR capacity, even though CSNF waste packages could be reduced by a factor of 7.916, the overall expansion of the YMR capacity would be less than a factor of 3 due to existence of the DOE-owned SNF and HLW. On the basis of electricity generation, however, the YMR still could support about 10 times greater electricity generation than the current YMR for direct disposal as long as the statutory capacity limit for DOE-owned SNF and HLW is not changed.

- Reducing the amount of process chemicals getting into the waste and allowing for a sufficient cooling time before solidification are effective in increasing the repository expansion factor.
**11 Future Works**

Additional works are necessary to perform the works done in this study more reliably and efficiently and to carry out the analysis of environmental impacts of ATW fuel cycle.

First, a module for the calculation of fission products inventory in WACOM is required to obtain a reliable composition of fission products based on a cycle-by-cycle calculation. With this, the heat generation rate of HLW and a required cooling time for its solidification could be evaluated more accurately.

Second, to simplify calculation procedures to obtain the linear fits of effective one-group cross sections, a method to couple WACOM and MCNP needs to be developed.

Third, to investigate the environmental impacts of a fuel cycle on a geologic repository, a nuclide transport model in geological media needs to be coupled to WACOM.
REFERENCES


ENDF/B-IV Library, National Nuclear Data Center, Brookhaven National Laboratory, Upton, N.Y., October, 1974.

ENDF/B-V Library, National Nuclear Data Center, Brookhaven National Laboratory, Upton, N.Y., October, 1979.


Oyamatsu, K., H. Takeuchi, M. Sagisaka, and J. Kataura, *New Methods for Calculating Aggregate Fission Product Decay Heat with Full Use of Macroscopic-


Spencer, B. W., The Rush to Heavy Metal Reactor Coolants-Gimmick or Reasoned, Proc. 8th Intl. Conf. on Nuclear Engineering, ICONE8, Baltimore, MD, USA, April, 2000.


Appendix A: Reference ATW Plant Parameters in the Roadmap

Reference ATW plant parameters used in the roadmap for the separation facility, the accelerator, and the transmuter systems are summarized in Tables A.1 through Table A.3 [Hill, et al., 1999].

Table A.1 Reference ATW Plant Parameters for the Separation Facility

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>TRU loss fraction per pass</td>
<td>0.001</td>
</tr>
<tr>
<td>Total processing loss</td>
<td>0.0033</td>
</tr>
<tr>
<td>Tc &amp; I processing loss</td>
<td>0.05</td>
</tr>
<tr>
<td>TRU Throughput</td>
<td>1765.8 kg/yr</td>
</tr>
<tr>
<td>$^{99}$Tc Throughput</td>
<td>135.6 kg/yr</td>
</tr>
<tr>
<td>I Throughput</td>
<td>37.9 kg/yr</td>
</tr>
<tr>
<td>Spent Fuel Throughput</td>
<td>169.2 MT/yr</td>
</tr>
</tbody>
</table>

Table A.2 Reference ATW Plant Parameters for the Accelerator

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton energy</td>
<td>1 GeV</td>
</tr>
<tr>
<td>Proton current</td>
<td>90 mA</td>
</tr>
<tr>
<td>Number of beam lines</td>
<td>2</td>
</tr>
<tr>
<td>Beam power</td>
<td>90.0 MW</td>
</tr>
<tr>
<td>Power required-accelerator</td>
<td>304.0 MWe</td>
</tr>
<tr>
<td>Accelerator net efficiency</td>
<td>29.6 %</td>
</tr>
<tr>
<td>Power required-plant</td>
<td>378.6 MWe</td>
</tr>
</tbody>
</table>

Table A.3 Reference ATW Plant Parameters for the Transmuter Systems

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Multiplication factor ($k_{eff}$)</td>
<td>0.97</td>
</tr>
<tr>
<td>Average neutrons/fission</td>
<td>2.95</td>
</tr>
<tr>
<td>Energy per fission</td>
<td>208 MeV</td>
</tr>
<tr>
<td>Thermodynamic efficiency</td>
<td>37.0%</td>
</tr>
<tr>
<td>Plant capacity factor</td>
<td>70.0%</td>
</tr>
<tr>
<td>Fission/year/target</td>
<td>5.52E+26</td>
</tr>
<tr>
<td>kg TRU fissioned/year/target</td>
<td>220 kg TRU/yr</td>
</tr>
<tr>
<td>Tc conversion efficiency</td>
<td>0.7</td>
</tr>
<tr>
<td>I conversion efficiency</td>
<td>0.7</td>
</tr>
<tr>
<td>Fission heat</td>
<td>832.1 MWt</td>
</tr>
<tr>
<td>Target/blanket total heat</td>
<td>840.0 MWt</td>
</tr>
<tr>
<td>Electricity per target/blanket</td>
<td>310.8 Mwe</td>
</tr>
<tr>
<td>Net plant efficiency</td>
<td>31.4 %</td>
</tr>
</tbody>
</table>
Appendix B: Input Data Card for MCNP Calculation for Startup-Core

ATW-LBE Cooled Transmuter with 31.8% Actinides-68.2% Zirconium Fuel

C Cylindrical reactor, aligned with z-axis

C Cell Cards
1  110 2.9360E-02 2 -5 -10 imp:n=1 tmp=8.617E-8 $ source/reflector
11 101 3.57873e-02 2 -5 10 -13 imp:n=1 tmp=8.617E-8 $ homogenized fuel zone
20 110 2.9360E-02 1 -2 -13 imp:n=1 tmp=6.0319E-8 $ reflector under fuel
21 110 2.9360E-02 5 -6 -13 imp:n=1 tmp=6.0319E-8 $ reflector over fuel
22 110 2.9360E-02 1 -6 13 -14 imp:n=1 tmp=6.0319E-8 $ reflector outside fuel
4  0 -1:6:14 imp:n=0 $ outside void

C Surface Cards
1  pz 0.000 $ bottom edge of reactor
2  pz 196.660 $ bottom edge of fuel region
5  pz 303.340 $ top edge of fuel region
6  pz 500.000 $ top edge of core
10  cz 36.941 $ outer edge of source/reflector region
13  cz 123.105 $ outer edge of fuel region
14  cz 152.783 $ outer edge of reflector region

C Data Cards
ksrc  0 0 200

C Material data
C Fuel region
m101 26000.55c 7.70210E-03
  24000.50c 1.11940E-03
  28000.50c 4.31280E-05
  42000.60c 5.27910E-05
  25055.60c 5.53300E-05
  14000.60c 7.21530E-05
  6000.78c 8.44370E-05
  74000.55c 1.37780E-05
  23000.60c 2.98350E-05
  82000.50c 9.72520E-03
  83209.60c 1.20290E-02
  90230.92c 1.69963E-13
  92234.86c 1.00000E-15
  92235.16c 2.98790E-08
  92236.92c 1.48760E-08
  92238.16c 3.52550E-06
  92327.60c 3.72040E-05
  94238.92c 9.38170E-06
  94239.16c 3.90700E-04
  94240.92c 1.57500E-04
  94241.92c 2.75460E-05
  94242.92c 3.39890E-05
  95241.60c 6.53110E-05
  95242.51c 1.01550E-07
  95243.88c 6.68890E-06
  96242.60c 6.37380E-16
  96243.60c 1.44470E-08
  96244.60c 7.48160E-07
  96245.60c 6.44790E-08
  96246.60c 7.13520E-09
40000.80c 4.12730E-03
35081.55c 1.00000E-15
36083.59c 1.00000E-15
36084.50c 1.00000E-15
37085.55c 1.00000E-15
37087.55c 1.00000E-15
39089.60c 1.00000E-15
40091.86c 1.00000E-15
40092.86c 1.00000E-15
40093.86c 1.00000E-15
40094.86c 1.00000E-15
40095.86c 1.00000E-15
40096.86c 1.00000E-15
42095.50c 1.00000E-15
42097.41c 1.00000E-15
42098.41c 1.00000E-15
42100.41c 1.00000E-15
43099.60c 1.00000E-15
44101.50c 1.00000E-15
44102.37c 1.00000E-15
44103.50c 1.00000E-15
44104.37c 1.00000E-15
45103.86c 1.00000E-15
45105.86c 1.00000E-15
46104.37c 1.00000E-15
46105.50c 1.00000E-15
46108.50c 1.00000E-15
47109.84c 1.00000E-15
48110.86c 1.00000E-15
48113.86c 1.00000E-15
48114.86c 1.00000E-15
49115.37c 1.00000E-15
53127.86c 1.00000E-15
53129.86c 1.00000E-15
54131.86c 1.00000E-15
54132.86c 1.00000E-15
54134.86c 1.00000E-15
54135.54c 1.00000E-15
55133.86c 1.00000E-15
55134.86c 1.00000E-15
55135.86c 1.00000E-15
55137.86c 1.00000E-15
57139.37c 1.00000E-15
59141.50c 1.00000E-15
60143.50c 1.00000E-15
60144.37c 1.00000E-15
60145.50c 1.00000E-15
60146.37c 1.00000E-15
60147.50c 1.00000E-15
60148.50c 1.00000E-15
60150.37c 1.00000E-15
61147.50c 1.00000E-15
61148.50c 1.00000E-15
61149.50c 1.00000E-15
62147.50c 1.00000E-15
62149.94c 1.00000E-15
C Target region, Reflector region
m110 26000.55c 1.0000E-15
24000.50c 1.0000E-15
28000.50c 1.0000E-15
42000.60c 1.0000E-15
25055.60c 1.0000E-15
14000.60c 1.0000E-15
82000.50c 1.3125E-02
83209.60c 1.6235E-02

fq0 ef
C Tallies for normalization purposes
f114: N 11 11
fm114 (-6.097036E+05 101 (-6) (-7 -6) (-2))

C Tallies for power distribution purposes
f214: N 11
fm214 (-1 101 (-6) (-7 -6) (-2))
C begin_mocup_flux_tallies
C time dependent flux
fc14 Volume average flux tally
f14:n
11
C end_mocup_flux_tallies
mode n
kcode 800 1 60 260
prdmp 260 260 260
print
## Appendix C: Neutronics Used for Linear Fits and Formulations

### Table C. 1 Effective One-Group Cross Sections Used for Linear Fits

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Capture ($n,\gamma$) cross section [b]</th>
<th>Fission ($n,f$) cross section [b]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st BOC (31.8wt% TRU)</td>
<td>100th BOC (43.0wt% TRU)</td>
</tr>
<tr>
<td>U235</td>
<td>5.428E-1</td>
<td>4.520E-1</td>
</tr>
<tr>
<td>U236</td>
<td>4.723E-1</td>
<td>3.730E-1</td>
</tr>
<tr>
<td>U238</td>
<td>3.709E-1</td>
<td>2.870E-1</td>
</tr>
<tr>
<td>Np237</td>
<td>1.597E+0</td>
<td>1.320E+0</td>
</tr>
<tr>
<td>Pu238</td>
<td>7.624E-1</td>
<td>6.300E-1</td>
</tr>
<tr>
<td>Pu239</td>
<td>5.213E-1</td>
<td>3.990E-1</td>
</tr>
<tr>
<td>Pu240</td>
<td>5.314E-1</td>
<td>4.230E-1</td>
</tr>
<tr>
<td>Pu241</td>
<td>4.565E-1</td>
<td>3.740E-1</td>
</tr>
<tr>
<td>Pu242</td>
<td>4.741E-1</td>
<td>3.740E-1</td>
</tr>
<tr>
<td>Am241</td>
<td>1.731E+0</td>
<td>1.430E+0</td>
</tr>
<tr>
<td>Am242m</td>
<td>4.254E-1</td>
<td>3.140E-1</td>
</tr>
<tr>
<td>Am243</td>
<td>1.536E+0</td>
<td>1.230E+0</td>
</tr>
<tr>
<td>Cm242</td>
<td>3.558E-1</td>
<td>2.560E-1</td>
</tr>
<tr>
<td>Cm243</td>
<td>2.718E-1</td>
<td>2.020E-1</td>
</tr>
<tr>
<td>Cm244</td>
<td>8.806E-1</td>
<td>7.240E-1</td>
</tr>
<tr>
<td>Cm245</td>
<td>3.183E-1</td>
<td>2.710E-1</td>
</tr>
<tr>
<td>Cm246</td>
<td>2.509E-1</td>
<td>1.950E-1</td>
</tr>
<tr>
<td>Zr</td>
<td>2.887E-2</td>
<td>2.133E-2</td>
</tr>
<tr>
<td>FP</td>
<td>2.662E-1</td>
<td>2.364E-1</td>
</tr>
<tr>
<td>Isotope</td>
<td>Capture (n,γ) cross section [b]</td>
<td>Fission (n,f) cross section [b]</td>
</tr>
<tr>
<td>----------</td>
<td>---------------------------------</td>
<td>---------------------------------</td>
</tr>
<tr>
<td></td>
<td>(X = \text{weight fraction of TRU in fuel})</td>
<td></td>
</tr>
<tr>
<td>U234</td>
<td>-1.5054X +1.1713</td>
<td>0.1679X+0.2828</td>
</tr>
<tr>
<td>U235</td>
<td>-0.8107X+0.8006</td>
<td>-1.7679X+2.4902</td>
</tr>
<tr>
<td>U236</td>
<td>-0.8866X+0.7542</td>
<td>0.0435X+0.0700</td>
</tr>
<tr>
<td>U238</td>
<td>-0.7491X+0.6091</td>
<td>0.0234X+0.0200</td>
</tr>
<tr>
<td>Np237</td>
<td>-2.4732X+2.3835</td>
<td>0.1705X+0.2647</td>
</tr>
<tr>
<td>Pu238</td>
<td>-1.1821X+1.1383</td>
<td>-0.1339X+1.1876</td>
</tr>
<tr>
<td>Pu239</td>
<td>-1.0920X+0.8685</td>
<td>-0.9554X+2.1308</td>
</tr>
<tr>
<td>Pu240</td>
<td>-0.9679X+0.8392</td>
<td>0.1464X+0.3260</td>
</tr>
<tr>
<td>Pu241</td>
<td>-0.7366X+0.6907</td>
<td>-2.3750X+3.3213</td>
</tr>
<tr>
<td>Pu242</td>
<td>-0.8938X+0.7583</td>
<td>0.1429X+0.2116</td>
</tr>
<tr>
<td>Am241</td>
<td>-2.6875X+2.5856</td>
<td>0.1420X+0.2000</td>
</tr>
<tr>
<td>Am242m</td>
<td>-0.9946X+0.7417</td>
<td>-5.2321X+5.8298</td>
</tr>
<tr>
<td>Am243</td>
<td>-2.7321X+2.4048</td>
<td>0.1196X+0.1526</td>
</tr>
<tr>
<td>Cm242</td>
<td>-0.8911X+0.6392</td>
<td>0.0902X+0.0992</td>
</tr>
<tr>
<td>Cm243</td>
<td>-0.6232X+0.4700</td>
<td>-3.5179X+3.9527</td>
</tr>
<tr>
<td>Cm244</td>
<td>-1.3982X+1.3252</td>
<td>0.1759X+0.3664</td>
</tr>
<tr>
<td>Cm245</td>
<td>-0.4223X+0.4526</td>
<td>-3.0179X+3.4677</td>
</tr>
<tr>
<td>Cm246</td>
<td>-0.4991X+0.4096</td>
<td>0.1482X+0.1983</td>
</tr>
<tr>
<td>Zr</td>
<td>-0.2661X+0.3508</td>
<td>-</td>
</tr>
<tr>
<td>FP</td>
<td>-0.0673X+0.0503</td>
<td>-</td>
</tr>
</tbody>
</table>
Appendix D: MOCUP Input Data Cards for Evaluation of End of Equilibrium Cycle Composition in ATW Transmuter

D.1 Input Data Card for MCNP

ATW-LBE Cooled Transmuter in Equilibrium (41.0% Actinides, 12.1% FP, 46.9% Zirconium)

C Cylindrical reactor, aligned with z-axis
C Cell Cards
1 110 2.9360E-02 2 -5 -10 imp:n=1 tmp=8.617E-8 $ source/reflector
11 101 3.61624e-02 2 -5 10 -13 imp:n=1 tmp=8.617E-8 $ homogenized fuel zone
20 110 2.93600E-02 1 -2 -13 imp:n=1 tmp=6.0319E-8 $ reflector under fuel
21 110 2.9360E-02 5 -6 -13 imp:n=1 tmp=6.0319E-8 $ reflector over fuel
22 110 2.9360E-02 1 -6 13 -14 imp:n=1 tmp=6.0319E-8 $ reflector outside fuel
4 0 -1:6:14 imp:n=0 $ outside void

C Surface Cards
1 pz 0.000 $ bottom edge of reactor
2 pz 196.660 $ bottom edge of fuel region
5 pz 303.340 $ top edge of fuel region
6 pz 500.000 $ top edge of core
10 cz 36.941 $ outer edge of source/reflector region
13 cz 123.105 $ outer edge of fuel region
14 cz 152.783 $ outer edge of reflector region

C Data Cards
ksrc 0 0 200

C Material data
C Fuel region
m101 26000.55c 7.70210E-03
24000.50c 1.11940E-03
28000.50c 4.31280E-05
42000.60c 5.27910E-05
25055.60c 5.53300E-05
14000.60c 7.21530E-05
6000.78c 8.44370E-05
74000.55c 1.37780E-05
23000.60c 2.98350E-05
82000.50c 9.72520E-03
83209.60c 1.20290E-02
90230.92c 1.69963E-13
92234.86c 2.85911E-06
92235.16c 7.59241E-07
92236.92c 1.15104E-06
92238.16c 1.27900E-05
93237.60c 3.04324E-05
94238.92c 5.96207E-05
94239.16c 2.72366E-04
94240.92c 3.93490E-04
94241.92c 7.01406E-05
94242.92c 1.26226E-04
95241.60c 6.24681E-05
95242.51c 4.39706E-06
95243.88c 3.99332E-05
96242.60c 6.50580E-06
96243.60c 6.46145E-07
C Target region, Reflector region
m10 26000.55c 1.0000E-15
   24000.50c 1.0000E-15
   28000.50c 1.0000E-15
   42000.60c 1.0000E-15
   25055.60c 1.0000E-15
   14000.60c 1.0000E-15
   82000.50c 1.3125E-02
   83209.60c 1.6235E-02
C Individual materials
m1  94238.92c 1.0000E-00
m2  94239.16c 1.0000E-00
m3  94240.92c 1.0000E-00
m4  94241.92c 1.0000E-00
m5  94242.92c 1.0000E-00
m6  92234.86c 1.0000E-00
m7  92235.16c 1.0000E-00
m8  92236.92c 1.0000E-00
m9  92238.16c 1.0000E-00
m10 95241.60c 1.0000E-00
m11 95242.51c 1.0000E-00
m12 95243.88c 1.0000E-00
m13 96242.60c 1.0000E-00
m14 96243.60c 1.0000E-00
m15 96244.60c 1.0000E-00
m16 96245.60c 1.0000E-00
m17 96246.60c 1.0000E-00
m18 93237.60c 1.0000E-00
m19 90230.92c 1.0000E-00
m20 35081.55c 1.0000E-15
m21 36083.59c 1.0000E-15
m22 36084.50c 1.0000E-15
m23 37085.55c 1.0000E-15
m24 37087.55c 1.0000E-15
m25 39089.60c 1.0000E-15
m26 40091.86c 1.0000E-15
m27 40092.86c 1.0000E-15
m28 40093.86c 1.0000E-15
m29 40094.86c 1.0000E-15
m30 40095.86c 1.0000E-15
m31 40096.86c 1.0000E-15
m32 42095.50c 1.0000E-15
m33 42097.41c 1.0000E-15
m34 42098.41c 1.0000E-15
m35  42100.41c  1.0000E-15
m36  43099.60c  1.0000E-15
m37  44101.50c  1.0000E-15
m38  44102.37c  1.0000E-15
m39  44103.50c  1.0000E-15
m40  44104.37c  1.0000E-15
m41  45103.86c  1.0000E-15
m42  45105.86c  1.0000E-15
m43  46104.37c  1.0000E-15
m44  46105.50c  1.0000E-15
m45  46106.37c  1.0000E-15
m46  47109.84c  1.0000E-15
m47  48110.86c  1.0000E-15
m48  48112.86c  1.0000E-15
m49  48114.86c  1.0000E-15
m50  49115.37c  1.0000E-15
m51  50117.86c  1.0000E-15
m52  51119.86c  1.0000E-15
m53  52121.86c  1.0000E-15
m54  53123.86c  1.0000E-15
m55  54125.86c  1.0000E-15
m56  55127.86c  1.0000E-15
m57  56129.86c  1.0000E-15
m58  57131.86c  1.0000E-15
m59  58133.86c  1.0000E-15
m60  59135.86c  1.0000E-15
m61  60137.86c  1.0000E-15
m62  61139.86c  1.0000E-15
m63  62141.86c  1.0000E-15
m64  63143.86c  1.0000E-15
m65  64145.86c  1.0000E-15
m66  65147.86c  1.0000E-15
m67  66149.86c  1.0000E-15
m68  67151.86c  1.0000E-15
m69  68153.86c  1.0000E-15
m70  69155.86c  1.0000E-15
m71  70157.86c  1.0000E-15
m72  71159.86c  1.0000E-15
m73  72161.86c  1.0000E-15
m74  73163.86c  1.0000E-15
m75  74165.86c  1.0000E-15
m76  75167.86c  1.0000E-15
m77  76169.86c  1.0000E-15
m78  77171.86c  1.0000E-15
m79  78173.86c  1.0000E-15
m80  79175.86c  1.0000E-15
m81  80177.86c  1.0000E-15
m82  81179.86c  1.0000E-15
m83  82181.86c  1.0000E-15
m84  83183.86c  1.0000E-15
m85  84185.86c  1.0000E-15
m86  85187.86c  1.0000E-15
m87  86189.86c  1.0000E-15
m88  87191.86c  1.0000E-15
m89  88193.86c  1.0000E-15
m90  89195.86c  1.0000E-15
m91  74000.55c 1.0000E-15
m92  23000.60c 1.0000E-15
m93  40000.80c 1.0000E-15
m94  82000.50c 1.0000E-15
m95  83209.60c 1.0000E-15
fq0  e f
c  Tallies for normalization purposes
f114:N 11 11
fm114 (-6.097036E+05 (-6) (-7 -6) (-2))
C  Tallies for power distribution purposes
f214:N 11
fm214 (-1 101 (-6) (-7 -6) (-2))
c  begin_mocup_flux_tallies
c  time dependent flux
fc14  Volume average flux tally
f14:n
  11
c  end_mocup_flux_tallies
c  begin_mocup_reaction_rate_tallies
c  time dependent reaction rates
fc314 Reaction rates
f314:n
  11
fm314 (1 1 (16) (17) (19:20) (102))
  (1 2 (16) (17) (18) (102))
  (1 3 (16) (17) (19:20) (102))
  (1 4 (16) (17) (18) (102))
  (1 5 (16) (17) (18) (102))
  (1 6 (16) (17) (19:20) (102))
  (1 7 (16) (17) (18) (102))
  (1 8 (16) (17) (19:20) (102))
  (1 9 (16) (17) (19:20) (102))
  (1 10 (16) (17) (18) (102))$
  (1 11 (16) (17) (19:20) (102))$
  (1 12 (16) (17) (19:20) (102))$
  (1 13 (16) (17) (19:20) (102))$
  (1 14 (16) (17) (19:20) (102))
  (1 15 (16) (17) (19:20) (102))
  (1 16 (16) (17) (18) (102))
  (1 17 (16) (17) (18) (102))
  (1 18 (16) (17) (18) (102))
  (1 19 (16) (17) (18) (102))$
  (1 20 (102))$
  (1 21 (16) (17) (102) (107))
  (1 22 (16) (102) (107))$
  (1 23 (102))$
  (1 24 (102))
  (1 25 (102) (107))
  (1 26 (102) (107))$
  (1 27 (102) (107))$
  (1 28 (102))$
  (1 29 (102) (107))$
  (1 30 (102))
  (1 31 (16) (102) (107))$
  (1 32 (102))$
  (1 33 (16) (17) (102) (107))$

131
c    end_mocup_reaction_rate_tallies

Reactions

fc414 Reaction rates
f414:n
11
fm414 (1 84 (16) (102))
(1 85 (16) (102))
(1 86 (16) (102))
(1 87 (16) (102))
(1 88 (16) (102))
(1 89 (16) (102))
(1 90 (16) (102))
(1 91 (16) (102))
(1 92 (16) (102))
(1 93 (16) (102))
(1 94 (16) (102))
(1 95 (16) (102))
c nu_bar of actinide isotopes
fc514 nu_bar
f514:n
  11
fm514 (1 1 (-7))
  (1 2 (-7))
  (1 3 (-7))
  (1 4 (-7))
  (1 5 (-7))
  (1 6 (-7))
  (1 7 (-7))
  (1 8 (-7))
  (1 9 (-7))
  (1 10 (-7))
  (1 11 (-7))
  (1 12 (-7))
  (1 13 (-7))
  (1 14 (-7))
  (1 15 (-7))
  (1 16 (-7))
  (1 17 (-7))
  (1 18 (-7))
  (1 19 (-7))
f614:n
  20
fm614 (1 94 (16) (17) (102))
  (1 95 (16) (17) (102))
mode n
kcode 800 1 60 260
prdmp 260 260 260
print
### D.2 Input Data Card for ORIGEN2.1

-1
-1
-1

<table>
<thead>
<tr>
<th>TIT</th>
<th>STAR LWR</th>
</tr>
</thead>
<tbody>
<tr>
<td>BAS</td>
<td>Task1a (2cm diameter)</td>
</tr>
<tr>
<td>LIP</td>
<td>0 0 0</td>
</tr>
<tr>
<td>LPU</td>
<td>902300 922340 922350 922360 922380 932370 942380 942390 942400 942410 942420 952410 952421 952430 962430 962440 962450 962460 -1</td>
</tr>
<tr>
<td>LPU</td>
<td>350810 360830 360840 370850 370870 390890 400910 400920 400930 400940 400950 400960 400970 420950 420960 420970</td>
</tr>
<tr>
<td>LPU</td>
<td>441040 451050 461050 461080 471090 481100 481130 491150 531270 531290 541310 541320 551330 541340 551350 551370 571390 591410 601430 601440 601450 601460 601470 611470</td>
</tr>
<tr>
<td>LPU</td>
<td>621470 601480 611480 611490 621490 601500 621500 621510 621520 631530 631540 631550 641550 641560 641570 -1</td>
</tr>
<tr>
<td>LIB</td>
<td>0 0 2 3 0 -322 -323 9 50 0 4 0</td>
</tr>
<tr>
<td>OPTL</td>
<td>8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8 8</td>
</tr>
<tr>
<td>OPTA</td>
<td>8 8 7 8 8 8 8 8 8 8 3 3 3 3 3 3 3 3 3 3 3</td>
</tr>
<tr>
<td>OPTF</td>
<td>8 8 7 8 8 8 8 8 8 8 8 8 8 3 3 3 3 3 3 3 3</td>
</tr>
<tr>
<td>CUT</td>
<td>3 1.0E-24 28 1.0E-75 -1</td>
</tr>
<tr>
<td>INP</td>
<td>1 2 -1 -1 1 1</td>
</tr>
<tr>
<td>IRP</td>
<td>30.000000 8.400000E+02 1 2 4 2</td>
</tr>
<tr>
<td>IRP</td>
<td>60.000000 8.400000E+02 2 3 4 0</td>
</tr>
<tr>
<td>IRP</td>
<td>90.000000 8.400000E+02 3 4 4 0</td>
</tr>
<tr>
<td>IRP</td>
<td>120.000000 8.400000E+02 4 5 4 0</td>
</tr>
<tr>
<td>IRP</td>
<td>150.000000 8.400000E+02 5 6 4 0</td>
</tr>
<tr>
<td>IRP</td>
<td>180.000000 8.400000E+02 6 7 4 0</td>
</tr>
<tr>
<td>IRP</td>
<td>200.000000 8.400000E+02 7 8 4 0</td>
</tr>
<tr>
<td>PCH</td>
<td>0 8 8</td>
</tr>
<tr>
<td>MOV</td>
<td>8 1 0 -1.0</td>
</tr>
<tr>
<td>TIT</td>
<td>Cooling of Discharged Fuel (25 years)</td>
</tr>
<tr>
<td>HED</td>
<td>1</td>
</tr>
<tr>
<td>DEC</td>
<td>5.0 1 2 5 4</td>
</tr>
<tr>
<td>DEC</td>
<td>10.0 2 3 5 0</td>
</tr>
<tr>
<td>DEC</td>
<td>15.0 3 4 5 0</td>
</tr>
<tr>
<td>DEC</td>
<td>20.0 4 5 5 0</td>
</tr>
<tr>
<td>DEC</td>
<td>25.0 5 6 5 0</td>
</tr>
<tr>
<td>OUT</td>
<td>6 1 0 0</td>
</tr>
<tr>
<td>STP</td>
<td>4</td>
</tr>
<tr>
<td>322</td>
<td>902300</td>
</tr>
<tr>
<td>2</td>
<td>902300</td>
</tr>
<tr>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>
### Appendix E: Heat Generation Rate of HLW from LWR- and ATW-SNF

#### Table E. 1  Heat Generation Rate per kg-HLW from LWR-SNF (25-year cooling)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life [s]</th>
<th>Average decay energy [MeV]</th>
<th>Wt% in HLW</th>
<th>Heat generation rate [W/kg-HLW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc 79</td>
<td>2.05E+12</td>
<td>4.20E-02</td>
<td>9.05E-05</td>
<td>1.57E+06</td>
</tr>
<tr>
<td>Rb 87</td>
<td>1.48E+18</td>
<td>1.41E-01</td>
<td>3.84E-03</td>
<td>2.81E+10</td>
</tr>
<tr>
<td>Sr 90</td>
<td>9.19E+08</td>
<td>1.95E-01</td>
<td>4.70E-03</td>
<td>7.46E+01</td>
</tr>
<tr>
<td>Y 90</td>
<td>2.30E+05</td>
<td>9.35E-01</td>
<td>1.18E-06</td>
<td>3.55E+00</td>
</tr>
<tr>
<td>Zr 93</td>
<td>4.82E+13</td>
<td>1.96E-02</td>
<td>1.12E-02</td>
<td>3.28E+06</td>
</tr>
<tr>
<td>Tc 99</td>
<td>6.77E+12</td>
<td>8.46E-02</td>
<td>1.18E-02</td>
<td>1.01E+04</td>
</tr>
<tr>
<td>Ru106</td>
<td>3.18E+07</td>
<td>1.00E-02</td>
<td>9.56E-11</td>
<td>1.90E-08</td>
</tr>
<tr>
<td>Pd107</td>
<td>2.05E+14</td>
<td>1.00E-02</td>
<td>3.05E-03</td>
<td>9.29E-08</td>
</tr>
<tr>
<td>Ag110M</td>
<td>2.15E+07</td>
<td>2.81E+00</td>
<td>1.37E+16</td>
<td>1.08E+11</td>
</tr>
<tr>
<td>Sn126</td>
<td>3.15E+12</td>
<td>1.13E+00</td>
<td>4.04E-04</td>
<td>7.73E-05</td>
</tr>
<tr>
<td>Sb125</td>
<td>8.74E+07</td>
<td>5.27E-01</td>
<td>4.34E-07</td>
<td>1.40E-03</td>
</tr>
<tr>
<td>Te123</td>
<td>3.15E+20</td>
<td>1.71E-02</td>
<td>1.05E-07</td>
<td>3.11E-18</td>
</tr>
<tr>
<td>Te125M</td>
<td>5.01E+06</td>
<td>1.41E-01</td>
<td>6.07E+09</td>
<td>9.19E-05</td>
</tr>
<tr>
<td>Cs134</td>
<td>6.50E+07</td>
<td>1.71E+00</td>
<td>4.18E-07</td>
<td>5.50E-03</td>
</tr>
<tr>
<td>Cs135</td>
<td>7.25E+13</td>
<td>5.63E-02</td>
<td>3.90E-03</td>
<td>1.49E-06</td>
</tr>
<tr>
<td>Cs137</td>
<td>9.46E+08</td>
<td>1.86E+01</td>
<td>1.03E-02</td>
<td>9.93E-01</td>
</tr>
<tr>
<td>Ce144</td>
<td>2.45E+07</td>
<td>1.11E+00</td>
<td>1.58E-12</td>
<td>3.34E-09</td>
</tr>
<tr>
<td>Pm147</td>
<td>8.27E+07</td>
<td>6.05E-02</td>
<td>3.36E+06</td>
<td>1.12E-03</td>
</tr>
<tr>
<td>Sm147</td>
<td>3.37E+18</td>
<td>2.31E+00</td>
<td>3.14E-03</td>
<td>9.78E-10</td>
</tr>
<tr>
<td>Sm148</td>
<td>2.53E+23</td>
<td>2.01E+00</td>
<td>2.32E-03</td>
<td>8.37E-15</td>
</tr>
<tr>
<td>Sm151</td>
<td>2.84E+09</td>
<td>1.97E+02</td>
<td>1.77E+04</td>
<td>5.46E-04</td>
</tr>
<tr>
<td>Eu152</td>
<td>4.29E+08</td>
<td>1.27E+00</td>
<td>1.10E-07</td>
<td>1.43E-04</td>
</tr>
<tr>
<td>Eu154</td>
<td>2.71E+08</td>
<td>1.50E+00</td>
<td>7.60E-05</td>
<td>1.84E-01</td>
</tr>
<tr>
<td>Eu155</td>
<td>1.56E+08</td>
<td>1.22E+01</td>
<td>6.22E+06</td>
<td>2.10E-03</td>
</tr>
<tr>
<td>U 234</td>
<td>7.71E+12</td>
<td>4.85E+00</td>
<td>3.13E+09</td>
<td>5.63E-10</td>
</tr>
<tr>
<td>U 235</td>
<td>2.22E+16</td>
<td>4.41E+00</td>
<td>1.26E+06</td>
<td>7.15E+11</td>
</tr>
<tr>
<td>U 236</td>
<td>7.39E+14</td>
<td>4.57E+00</td>
<td>5.79E+07</td>
<td>1.01E+09</td>
</tr>
<tr>
<td>U 238</td>
<td>1.41E+17</td>
<td>4.27E+00</td>
<td>1.28E+06</td>
<td>1.09E+09</td>
</tr>
<tr>
<td>Np237</td>
<td>6.75E+13</td>
<td>5.15E+00</td>
<td>1.28E-06</td>
<td>2.76E+08</td>
</tr>
<tr>
<td>Pu238</td>
<td>2.76E+09</td>
<td>5.59E+00</td>
<td>2.75E+07</td>
<td>1.56E-04</td>
</tr>
<tr>
<td>Pu239</td>
<td>7.59E+11</td>
<td>5.19E+00</td>
<td>1.40E-05</td>
<td>2.69E-05</td>
</tr>
<tr>
<td>Pu240</td>
<td>2.06E+11</td>
<td>5.25E+00</td>
<td>6.44E-06</td>
<td>4.55E-05</td>
</tr>
<tr>
<td>Pu241</td>
<td>4.54E+08</td>
<td>5.23E+03</td>
<td>9.83E-07</td>
<td>3.15E-06</td>
</tr>
<tr>
<td>Pu242</td>
<td>1.22E+13</td>
<td>6.66E+02</td>
<td>9.92E+07</td>
<td>3.82E-06</td>
</tr>
<tr>
<td>Am241</td>
<td>1.36E+10</td>
<td>5.60E+00</td>
<td>2.29E-06</td>
<td>2.61E-04</td>
</tr>
<tr>
<td>Am242M</td>
<td>4.79E+09</td>
<td>6.66E+02</td>
<td>1.00E-09</td>
<td>3.88E-09</td>
</tr>
<tr>
<td>Am243</td>
<td>2.32E+11</td>
<td>5.42E+00</td>
<td>1.64E-07</td>
<td>1.05E-06</td>
</tr>
<tr>
<td>Cm242</td>
<td>1.40E+07</td>
<td>6.21E+00</td>
<td>2.43E-12</td>
<td>2.97E-07</td>
</tr>
<tr>
<td>Cm243</td>
<td>8.99E+08</td>
<td>6.18E+00</td>
<td>2.97E-10</td>
<td>5.63E-07</td>
</tr>
<tr>
<td>Cm244</td>
<td>5.71E+08</td>
<td>5.90E+00</td>
<td>1.50E-08</td>
<td>4.23E-05</td>
</tr>
<tr>
<td>Cm245</td>
<td>2.68E+11</td>
<td>5.59E+00</td>
<td>1.27E-09</td>
<td>7.30E-09</td>
</tr>
<tr>
<td>Cm246</td>
<td>1.49E+11</td>
<td>5.52E+00</td>
<td>1.34E-10</td>
<td>1.35E-09</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td></td>
<td>5.494</td>
</tr>
<tr>
<td>Isotope</td>
<td>Half-life [s]</td>
<td>Average decay energy [MeV]</td>
<td>Wt% in HLW</td>
<td>Heat generation rate [W/kg-HLW]</td>
</tr>
<tr>
<td>----------</td>
<td>--------------</td>
<td>---------------------------</td>
<td>------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Sc 79</td>
<td>2.050E+12</td>
<td>4.200E-02</td>
<td>2.396E-05</td>
<td>4.154E-07</td>
</tr>
<tr>
<td>Rb 87</td>
<td>1.482E+18</td>
<td>1.410E-01</td>
<td>3.799E-03</td>
<td>2.778E-10</td>
</tr>
<tr>
<td>Sr 90</td>
<td>9.190E+08</td>
<td>1.958E-01</td>
<td>7.098E-04</td>
<td>1.125E-01</td>
</tr>
<tr>
<td>Y 90</td>
<td>2.304E+05</td>
<td>9.350E-01</td>
<td>1.780E-07</td>
<td>5.366E-01</td>
</tr>
<tr>
<td>Zr 93</td>
<td>4.828E+13</td>
<td>1.960E-02</td>
<td>1.408E-02</td>
<td>4.123E-06</td>
</tr>
<tr>
<td>Tc 99</td>
<td>6.772E+12</td>
<td>8.460E-02</td>
<td>2.190E-02</td>
<td>1.878E-04</td>
</tr>
<tr>
<td>Ru 106</td>
<td>3.181E+07</td>
<td>1.003E-02</td>
<td>1.033E-10</td>
<td>2.054E-08</td>
</tr>
<tr>
<td>Pd 107</td>
<td>2.050E+14</td>
<td>1.000E-02</td>
<td>2.755E-03</td>
<td>8.398E-08</td>
</tr>
<tr>
<td>Ag 110M</td>
<td>2.159E+07</td>
<td>2.817E+00</td>
<td>6.305E-16</td>
<td>5.001E-11</td>
</tr>
<tr>
<td>In 115</td>
<td>1.577E+22</td>
<td>2.420E-01</td>
<td>2.088E-04</td>
<td>1.863E-15</td>
</tr>
<tr>
<td>Sn 126</td>
<td>3.154E+12</td>
<td>1.135E+00</td>
<td>2.271E-04</td>
<td>4.343E-05</td>
</tr>
<tr>
<td>Sb 125</td>
<td>8.741E+07</td>
<td>5.274E-01</td>
<td>2.738E-07</td>
<td>8.837E-04</td>
</tr>
<tr>
<td>Te 123</td>
<td>3.156E+20</td>
<td>1.710E-02</td>
<td>2.255E-08</td>
<td>6.643E-19</td>
</tr>
<tr>
<td>Te 125M</td>
<td>5.011E+06</td>
<td>1.418E-01</td>
<td>3.828E-09</td>
<td>5.795E-05</td>
</tr>
<tr>
<td>Cs 134</td>
<td>6.507E+07</td>
<td>1.717E+00</td>
<td>5.115E-07</td>
<td>6.735E-03</td>
</tr>
<tr>
<td>Cs 135</td>
<td>7.258E+13</td>
<td>5.630E-02</td>
<td>4.039E-02</td>
<td>1.552E-05</td>
</tr>
<tr>
<td>Cs 137</td>
<td>9.467E+08</td>
<td>1.866E-01</td>
<td>1.952E-02</td>
<td>1.873E+00</td>
</tr>
<tr>
<td>Ce 144</td>
<td>2.456E+07</td>
<td>1.119E-01</td>
<td>6.339E-13</td>
<td>1.341E-09</td>
</tr>
<tr>
<td>Pm 147</td>
<td>8.279E+07</td>
<td>6.051E-02</td>
<td>9.154E-06</td>
<td>3.051E-03</td>
</tr>
<tr>
<td>Sm 147</td>
<td>3.377E+18</td>
<td>2.310E+00</td>
<td>8.668E-03</td>
<td>2.697E-09</td>
</tr>
<tr>
<td>Sm 148</td>
<td>2.525E+23</td>
<td>2.014E+00</td>
<td>2.190E-03</td>
<td>7.893E-15</td>
</tr>
<tr>
<td>Sm 151</td>
<td>2.840E+09</td>
<td>1.978E-02</td>
<td>3.030E-03</td>
<td>9.344E-03</td>
</tr>
<tr>
<td>Eu 152</td>
<td>4.292E+08</td>
<td>1.276E+00</td>
<td>6.697E-07</td>
<td>8.758E-04</td>
</tr>
<tr>
<td>Eu 154</td>
<td>2.714E+08</td>
<td>1.509E+00</td>
<td>8.362E-05</td>
<td>2.024E-01</td>
</tr>
<tr>
<td>Eu 155</td>
<td>1.565E+08</td>
<td>1.227E-01</td>
<td>3.030E-05</td>
<td>1.025E-02</td>
</tr>
<tr>
<td>U 234</td>
<td>7.716E+12</td>
<td>4.859E+00</td>
<td>2.820E-05</td>
<td>5.068E-06</td>
</tr>
<tr>
<td>U 235</td>
<td>2.221E+16</td>
<td>4.418E+00</td>
<td>1.555E-06</td>
<td>8.799E-11</td>
</tr>
<tr>
<td>U 236</td>
<td>7.389E+14</td>
<td>4.570E+00</td>
<td>3.496E-06</td>
<td>6.127E-09</td>
</tr>
<tr>
<td>U 238</td>
<td>1.410E+17</td>
<td>4.279E+00</td>
<td>2.066E-05</td>
<td>1.763E-10</td>
</tr>
<tr>
<td>Np 237</td>
<td>6.753E+13</td>
<td>5.156E+00</td>
<td>6.523E-05</td>
<td>1.410E-06</td>
</tr>
<tr>
<td>Pu 238</td>
<td>2.769E+09</td>
<td>5.591E+00</td>
<td>1.085E-04</td>
<td>6.146E-02</td>
</tr>
<tr>
<td>Pu 239</td>
<td>7.594E+11</td>
<td>5.199E+00</td>
<td>5.216E-04</td>
<td>9.974E-04</td>
</tr>
<tr>
<td>Pu 240</td>
<td>2.063E+11</td>
<td>5.253E+00</td>
<td>7.773E-04</td>
<td>5.499E-03</td>
</tr>
<tr>
<td>Pu 241</td>
<td>4.544E+08</td>
<td>5.230E-03</td>
<td>3.569E-05</td>
<td>1.143E-04</td>
</tr>
<tr>
<td>Pu 242</td>
<td>1.221E+13</td>
<td>6.664E-02</td>
<td>2.219E-04</td>
<td>8.547E-04</td>
</tr>
<tr>
<td>Am 241</td>
<td>1.364E+10</td>
<td>5.604E+00</td>
<td>2.096E-04</td>
<td>2.389E-02</td>
</tr>
<tr>
<td>Am 242M</td>
<td>4.797E+09</td>
<td>6.664E-02</td>
<td>8.029E-06</td>
<td>3.093E-05</td>
</tr>
<tr>
<td>Am 243</td>
<td>2.329E+11</td>
<td>5.423E+00</td>
<td>6.771E-05</td>
<td>4.345E-04</td>
</tr>
<tr>
<td>Cm 242</td>
<td>1.408E+07</td>
<td>6.216E+00</td>
<td>2.109E-08</td>
<td>2.574E-03</td>
</tr>
<tr>
<td>Cm 243</td>
<td>8.994E+08</td>
<td>6.189E+00</td>
<td>7.594E-07</td>
<td>1.439E-03</td>
</tr>
<tr>
<td>Cm 244</td>
<td>5.715E+08</td>
<td>5.901E+00</td>
<td>2.246E-05</td>
<td>6.341E-02</td>
</tr>
<tr>
<td>Cm 245</td>
<td>2.682E+11</td>
<td>5.598E+00</td>
<td>1.654E-05</td>
<td>9.442E-05</td>
</tr>
<tr>
<td>Cm 246</td>
<td>1.493E+11</td>
<td>5.523E+00</td>
<td>6.500E-06</td>
<td>6.548E-05</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td></td>
<td>2.917</td>
</tr>
</tbody>
</table>
Appendix F: Application of LP Model for Solidification of HLW from PWR

F.1 Introduction

Recently, a linear programming (LP) model for determining the composition of solidified high-level waste (HLW) by considering constraints for solidification was developed [Ahn, et al., 2004].

In this study, the developed model has been applied for the HLW vitrification process developed by Japan Nuclear Cycle Development Institute (JNC). Major constituents of high-level liquid wastes (HLLW) generating from PUREX reprocessing of LWR spent fuel are fission products, minor actinides, corrosion products, and process chemicals added in reprocessing. These are melted with glass frit for solidification.

F.2 Formulation of LP Problem

To apply the LP model for JNC-HLW solidification, we need to prepare (1) the set of constraints expressed in terms of the mass $M_W$ of HLW oxides and the mass $M_G$ of glass frit, (2) the composition vector $\vec{N}_W$ of HLW oxides, and (3) the composition vector $\vec{N}_G$ of the glass frit.

F.2.1 Constraints for JNC-HLW Conditioning

For the canister for solidified HLW developed by JNC [JNC TN1400 99-021, 1999], we consider the following seven constraints.

1. The volume of the solidified HLW should not exceed the volume of an empty canister, 0.15 m$^3$.
2. The weight of the solidified HLW should not exceed 400 kg.
3. The total heat generation rate from the solidified HLW is limited by 2,300 W.
4. The admissible maximum temperature for the solidified HLW is 450 °C.
5. To keep stability/durability of the waste glass, the mass $M_W$ should not exceed 30 wt% [Tsuboya, 1992],
6. The content of MoO$_3$ in waste matrix is limited by 2 wt% [Kawamura, 1995], and
7. The content of Na$_2$O in waste matrix is limited by 10 wt% [Kawamura, 1995].

These constraints need to be formulated in terms of $M_W$ and $M_G$.

Constraint (1) can be formulated based on the total mass of the solidified HLW and its density. For the density, an empirical formula reported in [Yamashita, et al., 1995; Sasage, et al., 1995] taking into account the dependency of waste loading fraction is employed. The formula derived using the empirical formula, however, is of a quadratic form, which has been approximated in a linear form.

Constraint (3) is formulated by summing the decay heat of each isotope, which is expressed in terms of the composition vector $\vec{N}_W$ for the waste oxides, the decay heat and the decay constant of each nuclide.

For Constraint (4), we solved a heat transfer equation for a cylindrical glass block surrounded by stainless cladding, with a uniform heat source. Because the solution is a function of volumetric heat source, the center-line temperature can be expressed in terms of $M_W$. 


Constraints (6) and (7) can be formulated by the composition vectors $\mathbf{N}_W$ and $\mathbf{N}_G$.

### F.2.2 Composition Vectors for HLW and Glass Frit

For HLW, we assume the PWR spent fuel with 4.0% enrichment and a burnup of 28,000 MWD/MTU. A 180-day cooling period after discharge and before reprocessing, and 5-year cooling before solidification have been assumed. The spent-fuel composition was obtained by ORIGEN2.1. Recovery fractions of U and Pu are assumed 0.604% and 0.297%, respectively [PNC SN8410 90-061, 1990]. Other actinide elements and all fission-product radionuclides except for noble gas elements are completely transferred to HLW. No noble gas element is included in HLW. Data shown in [PNC SN8410 90-061, 1990] have been assumed for concentrations of corrosion products and process chemical in the HLLW. The mass of HLW oxides from 1 metric ton (MT) of the irradiated fuel has been calculated to be 78.1 kg.

Glass frit PF798 [Yamashita, et al., 1995] developed by JNC was assumed.

![Figure F.1 Optimum waste loading estimated by the graphical solution method for JNC-HLW conditioning](image)

**Figure F.1** Optimum waste loading estimated by the graphical solution method for JNC-HLW conditioning

### F.3 Results

Figure B.1 shows graphically how the maximum waste loading has been determined. The maximum HLW loading $M_W$ is obtained as 96.3 kg. The mass $M_G$ of the glass frit can range between 230.9 kg and 299.1 kg. Thus, the HLW loading fraction ranges between 24.4 wt% and 29.4 wt%. The typical waste loading applied for the vitrification process developed in JNC is ~25 wt%, which is found to be in this range. This benchmarks the LP model, and indicates that the vitrified HLW from the JNC process has the optimized waste loading.
Essential constraints encompassing the feasible solution space are identified to be constraints (1), (4), and (7). However, other redundant constraints lie closely around the feasible solution space. This implies that a small change in the HLW composition or the composition of the glass frit could affect the optimum HLW loading. For instances, a higher molybdenum content in HLW makes the slope of the line for constraint (6) becomes smaller, and this constraint could become essential.