

5

**A Strategy for
Weapons-Grade Plutonium Disposition**

by

Kory William Budlong Sylvester

**B.S., Nuclear Engineering
Iowa State University**

**Submitted to the Department of Nuclear Engineering
and the Technology and Policy Program
in Partial Fulfillment of the Requirements for the Degrees of**

Master of Science in Nuclear Engineering

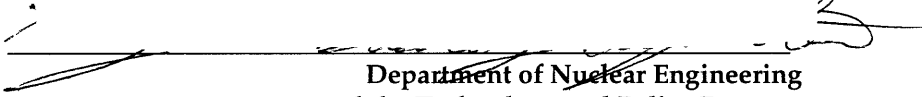
and

Master of Science in Technology and Policy

**at the
Massachusetts Institute of Technology
September 1994**

© Kory W. Budlong Sylvester, 1994. All rights reserved.
The author hereby grants to MIT permission to reproduce and to distribute
publicly paper and electronic copies of this thesis in whole or in part.

Signature of Author


Department of Nuclear Engineering
and the Technology and Policy Program

Certified by

Assistant Professor Scott A. Simonson
Department of Nuclear Engineering
Thesis Advisor

Certified by

Professor Marvin Miller
Department of Nuclear Engineering
Thesis Reader

Accepted by

Professor Allan F. Henry
Chairman, Department Committee on Graduate Students
Department of Nuclear Engineering

Accepted by

Professor Richard de Neufville
Chairman, Technology and Policy Program
Department of Civil Engineering

MASSACHUSETTS INSTITUTE
OF TECHNOLOGY

NOV 16 1994

Science

A Strategy for
Weapons-Grade Plutonium Disposition

by

Kory William Budlong Sylvester

Submitted to the Department of Nuclear Engineering
and the Technology and Policy Program
in Partial Fulfillment of the Requirements for the Degrees of
Master of Science in Nuclear Engineering and
Master of Science in Technology and Policy

ABSTRACT

A political as well as technical analysis was performed to determine the feasibility of glassification (vitrification) for weapons-grade plutonium (WGPu) disposition. The political analysis provided the criteria necessary to compare alternative storage forms. The technical areas of weapon useability and environmental safety were then computationally and experimentally explored and a vitrification implementation strategy postulated.

The Monte Carlo Neutron Photon (MCNP) computer code was used to model the effect of blending WGPu with reactor-grade Pu (RGPu). A mixture of 30% RGPu and 70% WGPu more than doubled the surface flux from a bare sphere of the mixture which assumedly correlates to a significantly increased predetonation probability. Rare earth diluents were also examined (using MCNP) for their ability to increase the compressed critical mass of the WGPu mixture. The rare earths (notably Eu) were effective in this regard.

As Pu-239 has a 24,100 yr. half-life, reactivity control in the long-term is an environmental safety issue. Rare earths were investigated as criticality controllers due to their neutron absorption capabilities and insolubility in aqueous environments. Thorium (a Pu surrogate) and the rare earths Eu, Gd, and Sm were added to two standard frits (ARM-1 and SRL-165) and formed into glass. Aqueous leach tests were performed (using MCC-1P guidelines) to measure rare earth leaching and determine the added elements' effects on glass durability. Europium was much more leach resistant than boron in the glasses tested. The elements had no negative effect on the environmental durability of the glasses tested at 90°C and minimal effect at room temperature. No fission product releases were detected in the ARM-1 compositions (which contained numerous simulated fission products).

Thesis Supervisor: Dr. Scott A. Simonson
Title: Assistant Professor of Nuclear Engineering

Acknowledgments

This thesis (although a labor of love) could not have been possible without the support and sacrifice of numerous individuals. Marriage to the most thoughtful, supportive, and genuinely fun woman I have ever met has proved to be invaluable during the course of my graduate work. Late nights at the lab are not ideal for newlyweds. Susan's understanding and encouragement made it all bearable. All this while I was responsible for uprooting her from a cushy job, stealing her from her family, and moving her to the hostile east coast. It should be made clear that such sacrifices are appreciated. Thanks for being yourself.

I owe much thanks to my parents, Jim and Barb. Throughout my life they have quietly made sure that I sought every option, pursued every interest, and strived to reach every goal. They have allowed me to soar as well as crash and burn, on my own. I have learned more from that freedom than from my 19 years (and counting) of classroom education. Their continuous support and stability has allowed me to take numerous chances. I could never repay the generosity they have lavished on me.

As an educator/wood carver, Dr. Scott A. Simonson is unparalleled. I don't think I could have asked for a better mentor (besides, no one else would have me). Intelligence and humility are sometimes difficult qualities to find together and Scott possesses both. His breadth and depth of knowledge make for invigorating discussions. Much of what is presented in this thesis stemmed from such interaction. Scott is a class act with a great family and I thank him for taking the time to shoot the breeze with me. I have benefited immeasurably from the experience.

I want to thank Dr. Marvin Miller whose frank discussions helped sort out the muddled issues surrounding WGPu disposition. I am also indebted to the many friends I have made at MIT. My roommates (Raj and Greg), the TPPers and the Nuclear Waste Group comprised a large fraction of that crew. Getting a few laughs out of this place is no small feat and they made it easy.

Finally I must thank the people that picked up the tab. That would be all of you. I was supported by the Nuclear Engineering/Health Physics Fellowship Program sponsored by the US Department of Energy and administered by the Oak Ridge Institute for Science and Education. I can assure you, your tax money was well spent.

Table of Contents

Table of Contents	5
List of Tables	7
List of Figures	8
Chapter 1. Introduction	10
Chapter 2. Background	13
2.1 Plutonium	13
2.2 Weapon Design	14
2.3 Current Dismantling Efforts	17
2.4 Barriers To Proliferation	18
Chapter 3. Political Analysis	21
3.1 International Concerns	21
3.1.1 Swift Implementation	21
3.1.2 Treatment Cost	22
3.1.3 Plutonium Fuel Value	22
3.1.4 Safety	22
3.1.5 Verification and Safeguarding	23
3.1.6 Flexibility and Insurance	24
3.2 Potential Treatments	24
3.2.1 The Burning Option	24
3.2.2 The Waste Option	27
3.2.3 The Storage Option	29
3.3 Discussion	31
Chapter 4. Technical Investigation	34
4.1 Scope	34
4.2 Degradation Effectiveness	35
4.2.1 Computational Procedures	36
4.2.2 Cases Examined	37
4.2.2.1 Isotopic Dilution	38
4.2.2.2 Chemical Dilution	38
4.2.3 Critical Mass Results	39
4.2.4 Surface Flux and Neutron Production Results	44

4.3 Environmental Safety	46
4.3.1 Experimental Procedures	47
4.3.2 Cases Examined	49
4.3.3 Durability Results	50
4.3.4 Thorium , Cesium and Europium Retention Results	54
4.3.5 Thorium Solubility in Glass	57
Chapter 5. Conclusions and Future Work	62
5.1 Technical Conclusions	62
5.1.1 Explosive Degradation	62
5.1.2 Environmental Safety	62
5.2 Discussion	63
5.3 An Implementation Strategy	64
5.4 Strategy Evaluation	68
5.5 Future Work	71
5.5.1 Computational Testing	71
5.5.2 Experimental Testing	72
5.5.3 Optimization	72
5.5.4 Alternative Storage Forms	73
5.5.5 Policy Analysis	73
Chapter 6. Bibliography	74
Appendix I. Sample Input Files for MCNP	77
Appendix II. Neutron Energy Spectrum.....	80
Appendix III. Surface Flux and Neutron Production Rate	83
Appendix IV. MCNP Data Summary	85
Appendix V. Glass Compositions.....	87
Appendix VI. Summarized Leach Test Results	89

List of Tables

Table 1 - Reference Plutonium Isotopic Compositions.....	13
Table 2 - Estimated Relative Ranking of Options.....	31
Table 3 - ICP Limits of Detection	50
Table A-1 - MCNP Results Summary.....	85
Table A-2 - Fabricated Non-Th Glass Compositions	87
Table A-3 - Fabricated Th Glass Compositions	88
Table A-4 - Pure SRL-165 Composition	88
Table A-5 - Pure ARM-1 Composition.....	88
Table A-6 - ARM-1 (90°C)	89
Table A-7 - 3% Rare Earths-ARM-1 (90°C).....	90
Table A-8 - 6% Rare Earths-ARM-1 (90°C).....	90
Table A-9 - 6% Rare Earths-2% ThO ₂ -ARM-1 (90°C)	90
Table A-10 - SRL-165 (90°C).....	91
Table A-11 - 6% Rare Earths-SRL-165 (90°C)	91
Table A-12 - 6% Rare Earths-2% ThO ₂ -SRL-165 (90°C).....	91
Table A-13 - ARM-1 (Room T.).....	92
Table A-14 - 3% Rare Earths-ARM-1 (Room T.)	92
Table A-15 - 6% Rare Earths-ARM-1 (Room T.)	92
Table A-16 - 6% Rare Earths-2% ThO ₂ -ARM-1 (Room T.).....	93
Table A-17 - SRL-165 (Room T.)	93
Table A-18 - 6% Rare Earths-SRL-165 (Room T.).....	93
Table A-19 - 6% Rare Earths-2% ThO ₂ -SRL-165 (Room T.)	94

List of Figures

Figure 1 - Compressed Critical Mass of Potential WGPu Storage Forms	40
Figure 2 - Rare Earth Effect on Compressed Critical Mass for WGPu.....	41
Figure 3 - Critical Mass of WGPu Mixture After Several Retrieval Attempts	42
Figure 4 - Compressed Critical Mass for a WGPu Mixture Containing 50% of a Single Rare Earth	43
Figure 5 - The Amount of Explosive Material Found in WGPu Glass Diluted With Various Amounts of Eu.....	44
Figure 6 - Isotopic Effect on Surface Neutron Flux for Pu Metal	45
Figure 7 - Boron Release from Various ARM Glass Compositions at 90°C	51
Figure 8 - Boron Release from Various SRL Glass Compositions at 90°C	52
Figure 9 - Rare Earth Effect on Silicon Release in SRL Glass at Room Temperature.....	53
Figure 10 - Silicon Release from SRL Glass Containing 2% Thorium Oxide and 6% Rare Earths at Room Temperature	54
Figure 11 - Reactivity Control Release in SRL Glass Containing 6% Rare Earths at 90°C	55
Figure 12 - Reactivity Control Release in SRL Glass Containing 6% Rare Earths at Room Temperature	56
Figure 13 - Europium Release From ARM Glass Containing 6% Rare Earths at Room Temperature	57

Figure 14 - Surface Photo of Devitrified Regions in ARM-1 Glass with 10% ThO ₂	58
Figure 15 - Grouping of Crystals in ARM-1 Glass with 10% ThO ₂	59
Figure 16 - Crystalline Structures in ARM-1 Glass with 10% ThO ₂	59
Figure 17 - Elemental Composition of the Bulk Region of ARM-1 Glass with 10% ThO ₂	60
Figure 18 - Elemental Composition from a Devitrified Region of ARM-1 Glass with 10% ThO ₂	61
Figure A-1 - MCNP Criticality Input File	78
Figure A-2 - MCNP Flux Input File	79
Figure A-3 - Neutron Energy Distribution in Surface Flux of Pure WGPu	80
Figure A-4 - Neutron Energy Distribution in Surface Flux of WGPu Mixed With Equal Amounts of Rare Earths	81
Figure A-5 - Neutron Flux From a Critical Mass of Various WGPu Mixtures	83
Figure A-6 - Source Terms for the Storage Forms	84

Chapter 1

Introduction

The signing of START II (Strategic Arms Reduction Treaty) in January of 1993 marked the end of the cold war and the beginning of a global effort to eliminate massive quantities of nuclear weapons. It is the latest in a series of international agreements on tactical and strategic nuclear arms reductions. Start II alone will reduce the number of strategic nuclear warheads to 3000 for the Former Soviet Union (FSU) and 3500 for the United States (US).¹ This should be contrasted with the 1990 totals of 12,000 and 11,000 strategic warheads for the US and FSU respectively.

While the treaty itself requires only the destruction of delivery systems, both countries are negotiating a "Safety, Security, and Dismantlement" agreement regarding the warheads themselves.² Dismantling would separate approximately 100 to 150 tonnes of weapons-grade plutonium (WGPu). At maximum dismantling capacity this translates into a buildup of more than 10 tonnes of separated WGPu per year in each country.

Simply stockpiling this material raises international security concerns as in unaltered form it may either be reinserted into a warhead or stolen by terrorists. While some of the plutonium will probably be held in reserve (how much is unknown) it is widely believed that the WGPu should be processed in a manner that renders it less attractive as an explosive, thus providing enhanced diversion protection over standard safeguarding efforts.³

The National Academy of Sciences (NAS) recently published a study in which it recommended adopting a spent fuel standard for the long-term disposition of the WGPu.⁴ This means that the final form of the plutonium

¹Isaacs, J., *A Bouquet for Bush*, The Bulletin of the Atomic Scientists, March 1993 pp.10, 46-7

²Lockwood, D., *GAO Study Outlines Obstacles To Soviet Warhead Dismantlement*, Arms Control Today, April 1993 p.25

³National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.112

⁴ibid. p.34

derived from weapons should be “roughly as inaccessible for weapons use as the much larger and growing stock of plutonium in civilian spent fuel.” They go on to identify reactor burning and vitrification (glassification) with high-level waste as the two most promising alternatives that satisfy this criteria.⁵

Most of the disposition research to date has focused on solutions that would use the Pu as fuel in a reactor.⁶ Less attention has been given to examining alternative storage options such as vitrification. In this thesis, an investigation is performed to determine the ability of a particular glass storage form to meet the diverse political and technical goals of the FSU and the US.

The prospect of vitrifying the Pu with or without high-level waste (HLW) is investigated. While a glass composition without HLW would apparently not satisfy the 'spent fuel standard' it may be more acceptable to the Russians and thereby accelerate the disposition process. It is possible that Pu stored in this 'clean' glass could be made extremely difficult to retrieve if chemically similar elements were included.

While the retrievability of glass in this form is not experimentally investigated in this thesis, the potential security gains from such processing are investigated computationally. The long-term environmental safety of the Pu storage form (with and without HLW) is also tested experimentally. The results are used to formulate a flexible strategy for WGPu processing that would provide increased security in the near term and environmental safety in the long-term.

The thesis begins with a background of the WGPu problem. Chapter 2 gives a technical description of plutonium and its use in weapon design. The current status of the disarmament process is described and the various barriers to proliferation are explained. The concerns of the international community as they relate to WGPu disposition are presented in Chapter 3 as well as a description of the available disposition technologies. Chapter 4 contains a description of the

⁵National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.224

⁶U.S. Department of Energy Plutonium Disposition Study, Technical Review Committee Report, Volumes 1 & 2, July 2 1993

computational and experimental work performed and their results. The conclusions gained from the technical investigation are presented in Chapter 5. These findings are used in combination with the identified political objectives to construct a potential implementation strategy for vitrification. This strategy is then evaluated and future areas of research are identified in the same chapter.

While this thesis should not be viewed as the definitive statement on WGPu disposition, it should expand our technical understanding of the vitrification option and its potential.

Chapter 2

Background

2.1 Plutonium

Plutonium is of interest as a weapon and a power source because it contains fissile isotopes. Fissile isotopes readily undergo fission; producing fission products and large amounts of energy. Plutonium itself however is not a naturally occurring element. It is produced in a uranium fueled nuclear reactor when U-238 captures a neutron and double beta decays to become Pu-239. A fraction of these Pu-239 isotopes will also capture neutrons to become the higher Pu isotopes 240, 241 and 242. It is the buildup of these higher isotopes that determines whether the Pu is labeled as reactor-grade (RGPu) or WGPu.

Plutonium composed of more than 94% Pu-239 is defined as weapons-grade. In contrast RGPu has less than 80% Pu-239.⁷ A typical example of the isotopic makeup of WGPu and RGPu is given in table one.

	WGPu	RGPu
% Pu - 239	95.0	62
% Pu - 240	3.0	21
% Pu - 241	1.5	13
% Pu - 242	0.05	4

Table 1 - Reference Plutonium Isotopic Compositions⁸

Civilian reactors were originally designed to operate with long burning cycles that optimized fuel efficiency and production of Pu (RGPu) which can be chemically separated (reprocessed), fabricated as fuel and reinserted into the reactor. Since chemical separation is no longer a goal of US utilities, the fuel is currently optimized for much higher burnup and economic efficiency. Military production reactors operate with short burning cycles to produce WGPu and

⁷DeVolpi, A., *Fissile Materials and Nuclear Weapons Proliferation*, Ann. Rev. Nucl. Part. Sci., V36, 1986 pp. 83-114

⁸Relative isotopics taken from Nuclear Chemical Engineering, McGraw Hill, Inc. 1981 p.88

tritium (for use in fusion weapons). The spent fuel from these reactors is reprocessed and the separated WGPu used in weapon manufacturing.

Weapons-grade Pu and RGPu also differ in the quantity separated. Although exact numbers do not exist, it is estimated that there are 200 tonnes of WGPu currently in the combined arsenals of the US and FSU and a total of 260 tonnes worldwide.⁹ As mentioned in the introduction nearly 150 tonnes of WGPu will be liberated if current dismantling schedules are fulfilled.

In comparison, approximately 120 tonnes of civilian RGPu had been separated from spent reactor fuel by the end of 1990. Of this amount, 50 tonnes had been recycled as nuclear fuel (mainly in fast reactors). The remaining 70 tonnes was stored principally at reprocessing facilities in France, the United Kingdom, and Russia in the form of sintered plutonium oxide. Russia has 25 tonnes of civilian Pu in storage.¹⁰

It is estimated that an additional 190 tonnes of civilian Pu will be separated by the year 2000 and of that approximately 120-130 tonnes of Pu will be added into storage.¹¹ This combined with the 1990 surplus equals 200 tonnes of stored Pu by the year 2000.

There exists even more RGPu in unprocessed spent fuel. It is estimated that 533 tonnes of RGPu were located in spent fuel worldwide in 1990.¹² The US portion of that figure was 175 tonnes and Russia left approximately 60 tonnes of RGPu in its spent fuel.

2.2 Weapon Design

In order to differentiate the problem of surplus RGPu from WGPu there is much debate in the literature regarding the weapons useability of RGPu. The

⁹Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992p.3

¹⁰National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.168

¹¹Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992 p.6

¹²ibid. p.33

issue can be clarified through an understanding of weapon design and the relevant physical characteristics of RGPu.

Nuclear weapons function in essence by taking fissile material in a sub-critical state and assembling it quickly into a super-critical state. A sub-critical weapon produces no energy. A weapon in a super-critical state however, will release energy at a rate that increases exponentially with time. Assembly is achieved by increasing the density of the fuel, usually by implosion with conventional high explosives. After a short period in a super-critical state though, expansion forces will cause disassembly and a return to sub-criticality, but not before the massive amounts of energy associated with the nuclear chain reaction is released.

Before a weapon can go super-critical however, it must contain at least a critical mass. A critical mass is the smallest amount of fuel required to sustain a fission chain reaction. At a minimum, a given weapon design must contain at least one critical mass in order to get any energy release. Therefore, the larger the critical mass the greater the amount of fuel required.

It is the concentration of fissile atoms in the fuel that largely determines its critical mass. In Pu the fissile isotopes are Pu-239 and Pu-241. Because the sum of these two isotopes is smaller in RGPu than in WGPu, its critical mass is larger. This means that for a given weapon design more RGPu is required to make a weapon than WGPu.

In addition the reduced concentration of Pu-239 in RGPu correlates to a higher concentration of the remaining Pu isotopes. These other isotopes cause numerous problems for weapons designers. The even numbered Pu isotopes (240 & 242) have a relatively high spontaneous fission rate which can cause predetonation. Predetonation occurs when the fission chain reaction is initiated prematurely, causing higher energy releases at the beginning of the compression stage. These early energy releases exert an expansion force that prevents the bomb from reaching design compression and its highest level of super-criticality. As a large fraction of the energy released during a nuclear explosion stems from these final stages of compression, predetonation greatly lowers the expected yield of the weapon.

Spontaneous fission (as well as other neutron producing reactions) does not guarantee that predetonation will occur, but it does reduce the expected yield of the weapon. Reactor-grade Pu also increases the variance in the estimated yields, making small yields more probable.¹³ This is why RGPu is often declared “non-weapons useable”.

In this context, the word weapon is used to describe an explosive that meets military reliability standards for sophisticated weapon states. A standard fission explosive (designed to use WGPu) fueled with RGPu is not considered a weapon due to its uncertain yield even though it probably would result in a several kiloton explosion. However it seems a more sophisticated weapon could be designed utilizing RGPu that “would be assured of having higher yields”.¹⁴

The higher concentrations of Pu-241 in RGPu has both benefits and drawbacks in weapon design. As Pu-241 is a fissile isotope, increased quantities of it help offset the reduction of fissile Pu-239 in RGPu. But plutonium-241 has a short half-life (13.2 yrs) and beta decays into Am-241. This isotope of americium decays by alpha emission with an associated high energy gamma. The increase in alpha emission is a source for self heating of the Pu, promoting metallurgical phase changes that could damage the weapon. The high energy gamma is also a health hazard, increasing shielding requirements and thereby increasing the cost of handling.¹⁵

In summary, while WGPu is clearly preferred over RGPu for weapons use due to its low probability of predetonation, smaller critical mass, lower temperature and handling ease, a fission explosive can still be designed with RGPu. The NAS study recognized the security danger posed by both reactor grades. It declared vitrification (with HLW) a “leading contender for long-term plutonium disposition”, meeting its spent fuel standard despite any isotopic

¹³Mark, J., *Reactor-Grade Plutonium's Explosive Properties*, Nuclear Control Institute, August 1990 p.4

¹⁴National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.33

¹⁵DeVolpi, A., *Denaturing Fissile Materials*, Progress in Nuclear Energy 10:161, 1982 p.187

alteration.¹⁶ This statement de facto gives little security credit to processes that would alter the isotopics of the WGPu.

Nuclear weapons are also fueled with highly enriched uranium (HEU). As spontaneous fission in HEU is rare, it can be used in simpler "gun type" weapons.¹⁷ These weapons function by colliding (or shooting) one mass of fissile material into another, forming a super-critical mass.¹⁸ The idea is the same as an implosion weapon only assembly is slower.

The demilitarizing of HEU is a much easier task than what is required for WGPu. The HEU can be isotopically diluted or 'denatured' by blending it with natural or depleted uranium. This low enriched uranium (LEU) product is suitable for use in civilian nuclear reactors with no potential for weapon use. The US has agreed to pay approximately \$10 billion for the LEU derived from the Former Soviet Union's dismantled warheads.¹⁹

2.3 Current Dismantling Efforts

Before any sort of Pu treatment can be performed, the weapons themselves must be returned from the field and dismantled. While this is not a political hurdle for the US, dismantling the former Soviet Union's weapons will require cooperation between four countries with wavering trust of each other. The result has been a series of agreements contingent on certain actions.

Both Russia and the US have made the implementation of START I conditional on Belarus, Ukraine, and Kazakhstan acceding to the non-proliferation treaty (NPT) as non-nuclear-weapons states.²⁰ Implementation also requires Russia to make bilateral agreements with these three new countries regarding weapon withdrawal and destruction. As of May 1994, Belarus and Kazakhstan had ratified START I and voted to accede to the NPT.

¹⁶National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.195

¹⁷ibid. p.33

¹⁸DeVolpi, A., *Fissile Materials and Nuclear Weapons Proliferation*, Ann. Rev. Nucl. Part. Sci., V36, 1986 p.85

¹⁹von Hippel, et. al., *Eliminating Nuclear Warheads*, Scientific American, August 1993 p.47

²⁰Lockwood, D., *GAO Study Outlines Obstacles To Soviet Warhead Dismantlement*, Arms Control Today, April 1993 p.25

Ukraine has not officially ratified START I but has begun shipping warheads to Russia as part of a three-way agreement between Ukraine, US and Russia.²¹ The agreement calls for Ukraine to transport all of its strategic nuclear warheads (~1800) to Russia over a three year period. In exchange Russia is to send 100 tons of LEU to Ukraine for power generation. This is to be financed by the US in the amount of \$60 million. Russia will also forgive a large portion of Ukraine's natural gas debt as payment for tactical warheads previously returned to Moscow.

While its actions are promising, Ukraine's parliament (Rada) has voted down resolutions that would have Ukraine join the NPT.²² Russia's new neighbor is concerned about issues of security, international power, and financial compensation. Nuclear weapons provide the most impressive deterrent mankind has ever conceived. For a country that has a history of imperial enslavement, they represent unquestioned sovereignty and protection from territorial challenges in the future. If "their" weapons are to be removed however, they want to be paid for them as evidenced by the tri-lateral agreement.

While determination of a fair value for the uranium from nuclear weapons is relatively easy to compute, the value of Pu is more difficult to ascertain. This is especially true if the final use of the Pu is unknown. If it is waste, then there is little to persuade US or Russia to pay Ukraine anything for it (other than to speed the disarmament process). If it is to be used as fuel, Ukraine will have to be assured that profits (if any) will be shared. In any case, a selection of a disposition technology would simplify negotiations and promote continued disarmament.

2.4 Barriers To Proliferation

Assuming the problems currently existing with warhead dismantlement are resolved, we can focus on the question of what to do with this highly

²¹Keeny, S. M., *Will Political Realism Prevail in Kiev?*, Arms Control Today, January/February 1994 p.2

²²Deni, J., *Chronology of U.S.-Soviet-CIS Nuclear Relations*, Arms Control Today, June 1994 p.33

sensitive material. The highest priority is assumed security. There exists several physical and structural barriers that can be used to prevent proliferation.

The implied objective of START II is to ensure that the WGPu removed from the warheads is never refabricated into an explosive of any type. The most obvious solution would be to eliminate the Pu altogether. This option requires for example, bombarding the Pu with neutrons until it is destroyed (creating other elements) or jettisoning the material into space. The outer space solution seems rather impractical but engineered sources of neutrons are available for Pu destruction.

According to the Academy, elimination is achieved if the option ensures “that retrieving enough plutonium for a nuclear explosive from whatever remains would be extremely difficult.”²³ Therefore if the Pu is diluted to a high enough level it could effectively be considered eliminated.

If the WGPu is not destroyed, it can be altered in ways that make it less desirable to explosive engineers. The isotopics of the Pu itself can be changed to reduce the fissile content and increase the spontaneous fission rate, giving it the same problematic characteristics of RGPu. This can be achieved by exposing the Pu to a neutron source of the type mentioned earlier or by simply blending the WGPu with RGPu. But again, this does not eliminate Pu’s explosive potential.

Mixing the WGPu with chemically similar elements would make it technically difficult to separate, providing another obstacle to proliferation. A diluted mixture of this type could be designed that would prevent fabrication as a fission explosive unless the Pu was purified. If partial separation was achieved, the critical mass of the mixture would be relatively high. If radioactive elements were chosen as diluents, a lethal radiation barrier could also be created.

However, the NAS study notes that the radiation barrier associated with spent fuel diminishes as fission products decay.²⁴ They state that after fifteen years the dose rate at a distance of one meter from the center of the fuel bundle

²³National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.145

²⁴ibid. p.151

would be ten times less than that after one year. This dose rate (2000 rads per hour) is still extremely dangerous but will decrease by “50 percent every 30 years”.²⁵ The time at which remote processing would become unnecessary would depend upon the level of risk the radiation worker would be willing to accept and what protections were afforded them.²⁶ This information is included not to discredit the value of a radiation barrier but rather to note the fact that the security it provides will fade with time.

The chemical mixing described previously could be achieved through a variety of processes. For example, the Pu could be directly mixed with measured concentration of diluents, burned in a reactor, or added to the HLW scheduled for vitrification. The final form of each would include elements chemically similar to Pu but present in different concentrations.

Finally, barriers can be utilized that do not alter the Pu itself but prevent would-be proliferators access to Pu repositories. Armed guards, fences, concrete barriers, alarm systems, etc... may all be used to reduce the risk of proliferation. Simply storing the Pu in close proximity to highly radioactive spent nuclear fuel would provide a radiation barrier that would also deter unauthorized access. Clearly any Pu processing will necessarily be done with substantial structural barriers in place.

The ultimate decisions on which barriers to be used will be made by policy makers. The following chapter describes the political complexities surrounding WGPu disposition. The major concerns of the international community and the technical options available to satisfy those concerns are presented. This information combined with an understanding of the relevant technical issues will allow the formulation of a rational disposition policy.

²⁵National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.151

²⁶ibid. p.151

Chapter 3

Political Analysis

3.1 International Concerns

The previously mentioned barriers provide a technical and structural arsenal for defense against proliferators. If security was the only criteria, assumedly all of the available barriers would be used to minimize the risk of proliferation. Other factors however will influence the choice of barriers to be used. The issues themselves and the power of the organizations that champion them will determine the acceptable solutions.

3.1.1 Swift Implementation

Clearly of interest to all is timeliness. The risk of theft is perceived to increase with time, therefore the quicker a treatment plan is selected and implemented the better. This risk is magnified in the FSU. Russia is in great political upheaval at the present time, creating instability which could reduce the institutional control of the separated nuclear material. This, combined with the economic stresses placed on FSU workers, makes the sale of sensitive material a real concern.²⁷ Another reason for expedition is the mere fact that the longer we wait, the greater opportunity for each country to renege on its commitments. Missing this window of opportunity for real arms reductions would be unfortunate to say the least.

Attention will have to be given to the timely availability of facilities, materials, and expertise as well as all necessary licenses and permits. For example, a treatment process requiring the construction of a nuclear reactor in the US may be vulnerable to severe licensing delays. If reciprocity were required, such delays could induce Russia to postpone processing even if they had a facility ready to operate.

²⁷von Hippel, et. al., *Eliminating Nuclear Warheads*, Scientific American, August 1993 p.44

3.1.2 Treatment Cost

The financial compensation issues that have surfaced during the present dismantling negotiations will not evaporate for disposition talks. It seems clear that Russia will continue to seek economic aid (in billions of dollars) from the West to carry out any Pu processing. The exact amount will depend on the technology selected. Therefore a strategy will have to be chosen with attention given to the cost/security gained tradeoffs.

3.1.3 Plutonium Fuel Value

This concern should be accounted for in the preceding cost section but the immense economic, human, and environmental sacrifice associated with the production of WGPu makes options that would treat the Pu as waste less attractive.

The US and the FSU however have conflicting views on the issue of Pu fuel. The US abandoned Pu fuel for power production by banning civilian reprocessing under the Carter administration, viewing the associated proliferation risks as being too high. Although the ban has since been lifted, the Clinton administration has officially stated its opposition to civilian Pu use.²⁸

The FSU views the future of Pu fuel in a different light. The head of the Russian Federation's Ministry of Atomic Energy (MINATOM), Viktor Mikhailov, has informed the US that MINATOM wants to use the WGPu recovered from its nuclear warheads to fuel its commercial reactors as part of its fast reactor program.²⁹ Construction of two BN (fast) reactors has been stalled due to financial constraints but plans exist to build more.

3.1.4 Safety

Radioactive material processing is inherently dangerous. Plutonium is a carcinogen that is well known for its toxicity.³⁰ As an alpha emitter Pu is dangerous when it is ingested. Alpha particles deposit a great deal of their

²⁸White House Press Release, September 27, 1993

²⁹Lockwood, D., *GAO Study Outlines Obstacles To Soviet Warhead Dismantlement*, Arms Control Today, April 1993 p.25

³⁰Hu, H., et al, *Plutonium: Deadly Gold of the Nuclear Age*, International Physicians Press, Cambridge, Massachusetts 1992 pp.8-18

energy as they travel through tissue, causing biological damage. One study (based on animal testing) estimates that about a millionth of an ounce of Pu-239 deposited in the lungs would “cause lung cancer with a probability approaching 100 percent.”³¹

In addition Pu metal is pyrophoric.³² This means that it can ignite spontaneously in the presence of air. Burning would create small particles of Pu, thus providing an easy mechanism for ingestion.

It is clear that adequate protection must be given to the general public in either country during all Pu disposition activities. In addition to the risks the Pu itself presents, disposition activities themselves could create risks from other sources. The selection of a processing strategy that minimizes the risk to humans and the environment is therefore required. The risk to future generations as well as present ones must be considered as Pu-239’s half-life is more than twenty-four thousand years.

3.1.5 Verification and Safeguarding

A strategy for Pu processing must also include provisions for verification and safeguarding. Both sides must be adequately assured that equal quantities of WGPu are being processed in a safe, secure fashion. Since only a small amount of Pu is needed to make an explosive, strict material accounting is needed to assure that none of the WGPu is siphoned off for reuse or simply misplaced.

Verification and safeguarding is the primary concern of the International Atomic Energy Agency (IAEA). The IAEA was formed in the 1950s to promote and safeguard the peaceful use of the atom. From its conception the IAEA was envisioned to play a role as a repository for surplus fissile materials. A failed attempt was made in 1982 to reach consensus on an international Pu storage regime for civilian RGPu. Nonetheless, the agency has a continued interest in fulfilling this role. It views the present disarmament efforts as an “occasion for gaining agreement on a system to effectively control and verify weapons-usable

³¹ibid. p.14

³²ibid. p.5

nuclear material” which could be applied to excess civilian Pu.³³ The US government appears to support the idea of a processing strategy that would address both the RGPu and WGPu problem.³⁴

3.1.6 Flexibility and Insurance

The only certainty concerning Pu disposition is that much is unknown about the appeal of the various options. In addition political and societal priorities are certain to waver over the dismantling and disposition time period. Selection of a disposition technology and how that technology will be used must be done in a way that provides a sufficient level of flexibility.

3.2 Potential Treatments

Having described the barriers that can be used to deter proliferation and the diverse interests influencing disposition strategy formulation, this section describes the primary treatment options and their effectiveness. While much uncertainty exists concerning specific advantages and disadvantages of each option, the perceived positives and negatives are stated.

3.2.1 The Burning Option

Elimination, isotopic degradation, and chemical dilution could be achieved by fabricating the WGPu into fuel elements for use in existing reactors or future government or private reactor designs. Reactors would fission a certain fraction of the WGPu and isotopically degrade the remaining Pu. To what degree depends upon how long the Pu is left initially in the reactor and whether or not the fuel is reprocessed for reinsertion.

After the last burning cycle, the Pu could be left with the fission products in the spent fuel assemblies. This would chemically dilute the Pu and provide an intense radiation barrier at least for several decades. Thus, reactor burning options would provide effective proliferation protection by utilizing all possible physical barriers.

³³Scheinman, L., *Managing the Coming Glut Of Nuclear Weapon Materials*, Arms Control Today, March 1992 p.9

³⁴White House Press Release, September 27, 1993

Existing US light water reactors (LWRs) or partly completed reactors are the preferred choice of the NAS should the reactor option be chosen. However, implementation could be plagued with delays. Plutonium fuels are generally fabricated with natural or depleted uranium in what is called mixed-oxide fuel (MOX). Current LWR designs in the US do not utilize MOX fuel and therefore no MOX fabrication facilities exist here. If a MOX facility was built, the LWRs could accept 1/3 core loadings of MOX without significant technical problems.³⁵ One study estimates that after all permits are issued, 18 LWRs could process 50 tonnes of surplus WGPu in approximately 10 years.³⁶

Indeed the more reactors used the faster the Pu could be processed, but finding utilities in the US willing to do this and receiving the appropriate licensing may cause additional delays. A realistic time frame for the implementation of this reactor option would be between 10 and 20 years. The high cost of MOX fabrication places the preliminary cost estimates at approximately a billion dollars for this option.³⁷

If the appropriate licenses could not be attained the NAS recommends the construction of new government reactor(s) built on federal property.³⁸ The NAS further recommends using a well-proven design to avoid any technical or licensing uncertainties. The amount of time and money required to process the WGPu in this scenario would depend on the length of construction, number of reactors constructed and Pu loading in the fuel. However, it is assumed that they would "require more time and money" for implementation.³⁹

The environmental effects of the burning option could be considered marginal but not benign. Waste would be created at the fuel fabrication facility and the spent fuel would present the same disposal problems as other spent fuel

³⁵U.S. Congress, Office of Tech. Assessment, Dismantling the Bomb and Managing the Nuclear Materials, U.S. Government Printing Office, September 1993 p.89

³⁶ibid. p. 90

³⁷Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992 p. 29

³⁸National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.143

³⁹ibid. p.156

in the US. Given the larger volume of HLW waste already generated in this country, this small addition of spent fuel seems less of a factor.

If civilian reactors were used, verification and safeguarding efforts may be complicated. Each reactor would have to be intensely monitored. Federally owned reactors at a single location would be easier to monitor. Use of Pu in the civilian sector for WGPu disposition may also pave the way for a Pu fuel cycle in the US. This is viewed as a diversion threat and is opposed by the current Clinton administration.⁴⁰ This fact alone may prove fatal for the civilian burning option in the US.

Plutonium burning in Russia is welcome but may face problems of a different kind. The implementation time, cost and safety of reactor burning in Russia is difficult to assess. The main concern is the high security risk associated with transporting and processing WGPu at numerous locations.⁴¹ This transportation risk could potentially be minimized as the MOX fabrication facility currently under construction is located at a "major nuclear weapons facility."⁴² All the WGPu could therefore be made into MOX at a single secure facility.

Additionally the Russian reactor of choice for safe WGPu burning (as identified by the NAS) is the VVER 1000. Multiple reactors of this type are often located at a single site. As they apparently are easily modified to accept full core MOX loadings, a single facility (with several reactors) may be able to burn the WGPu in a timely fashion. Two such reactors (with full core loadings) would be able to burn 50 tonnes of WGPu in 30 years.⁴³ This again would limit the diversion risks from transportation.

Each country could build new advanced reactors to treat the Pu but this option is not supported by the NAS. These reactors would invoke substantial

⁴⁰White House Press Release, September 27, 1993 p.4

⁴¹National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.167

⁴²ibid. p.170

⁴³ibid. p.168

development and demonstration costs (one estimate is around \$10 billion).⁴⁴ Potential designs include an advanced liquid metal reactor (ALMR), high-temperature gas-cooled reactor (HTGR), and an accelerator based converter (ABC). These designs would take 10 to 25 years to develop and deploy.⁴⁵ In the US such a project would probably meet opposition from anti-nuclear and environmental interest groups almost ensuring further delays.

These political obstacles probably do not exist in Russia (yet). Building more nuclear reactors is also in line with their nuclear energy policy. The problem is that they do not have any money to do so by themselves. Aid from the west would most likely be solicited. The US however, may be hard pressed to justify such expenditures. Russia's disappointing environmental and safety record to date warrants caution in promoting an expansion of its nuclear program at this time.⁴⁶

3.2.2 The Waste Option

Weapons-grade Pu could be treated as waste to be disposed of in a geologic repository. Depending on the elements mixed with the Pu, virtually all of the same barriers to proliferation could be applied as reactor burning (save elimination). The degree of isotopic degradation is limited only by the availability of separated RGPu, which in Europe appears to not be a problem. Weapons-grade plutonium could be disposed of "as is" in special containers or encapsulated with a variety of chemically similar constituents to hamper separation.

The direct disposal option provides only a structural barrier to proliferation and presents serious safety concerns. The major dangers associated with Pu disposal are the potential for a criticality accident and/or contamination of the environment through groundwater interaction. Direct plutonium disposal would provide little protection from such accidents.

⁴⁴Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992 p. 29

⁴⁵U.S. Congress, Office of Tech. Assessment, Dismantling the Bomb and Managing the Nuclear Materials, U.S. Government Printing Office, September 1993 pp.91-96

⁴⁶Leskov, S., *Lies and Incompetence*, Bulletin of the Atomic Scientists, Vol. 49 num. 5, June 1993 pp.13,55

A storage matrix that retarded Pu movement would be preferred. A number of encapsulation technologies have been examined for HLW to limit its mobility in the environment. Glassification (vitrification) is the most developed. A vitrification facility exists in the US at the Savannah River Site (SRS) in South Carolina which uses borosilicate glass as its waste form.

Savannah River has looked at the possibility of including WGPu in their operations. They examined options ranging from vitrifying the Pu by itself to incorporating it with their HLW. Processing could (optimistically) begin in eight years at the earliest.⁴⁷ Including 1% WGPu concentration in their HLW glass would allow 50 tonnes of Pu to be processed in 11 years of operation. Their rough cost estimations for processing 50 tonnes of WGPu range from \$0.7 billion (small logs, without HLW), to \$1.6 billion (large logs, with or without HLW).⁴⁸

The NAS study states that 'clean' vitrification (options that do not include HLW) should not be pursued "except possibly as a first step toward adding radiological or physical barriers as well."⁴⁹ However they do identify several potentially significant benefits to be gained from such processing. "If the initial step of vitrifying the plutonium separately before later revitrifying it with HLW were an alternative to longer storage of plutonium in pit form, and could be accomplished quickly and for modest additional cost, this might be a useful approach."⁵⁰

Savannah River characterizes this approach as being "a relatively quick and effective way of producing a proliferation-resistant and stable form that could be stored and monitored in any approved special nuclear materials storage location."⁵¹ In their concept, the WGPu would be vitrified in a small melter,

⁴⁷McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p.20

⁴⁸ibid. p.24

⁴⁹National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.228

⁵⁰ibid. p.189

⁵¹McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p.2

forming small logs or sheets of glass suitable for crushing into a frit. The frit would be relatively easy to revitrify with HLW if a decision was made to do so.

Scenarios that would allow for large 'clean' logs were also presented by Savannah River. One option would be to wait until all SRS HLW had been vitrified so that the same melter could be used for WGPu. Alternatively a separate facility at SRS could be modified to include a new melter of any size to process the Pu.

There are concerns, however, over the use of borosilicate glass for Pu disposal. In the presence of water, it appears that boron leaches at a higher rate than Pu.⁵² Boron is used for criticality control (it absorbs neutrons) in the waste matrix. In the long-term, sufficient amounts of boron could exit the waste form leaving the Pu in an unsafe state.

The former Soviets have been experimenting with phosphate glass at their reprocessing facility in Chelyabinsk. The Pu capacity is low in this facility, less than half that of Savannah River.⁵³ In addition, the phosphate glass appears to be inferior to the borosilicate glass used in the US in terms of durability and safety. It appears that a new glassification facility would have to be constructed in Russia for WGPu disposition, again with financial aide.

As fewer facilities are projected to be necessary, this option would appear to have less verification and safeguarding problems. But treating the WGPu as waste is against the wishes of Russia, arguing that this option throws away the fuel value of Pu. Economic reality, however, is that value is determined by the marketplace. Uranium fuel is currently plentiful and inexpensive and MOX fuel cannot compete, therefore Pu has no current economic value.

3.2.3 The Storage Option

The default option of simply storing the WGPu is often mentioned. Storage in unaltered form would present only structural barriers to proliferation. It is worth mentioning that the processes outlined above would provide various

⁵²Taubes, G., *No Easy Way to Shackle The Nuclear Demon*, Science, Vol. 263, February 4, 1994 p.631

⁵³Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992 p.98

storage options too. The Pu could be stored indefinitely as fuel for existing or future reactors or in a vitrified form. Even if the WGPu is burned in a reactor, the spent fuel cask (containing a fraction of the original Pu) is simply another storage form. Inserting the cask in a repository provides another structural barrier, but it does not make the Pu go away. Nevertheless, most references to the storage option assume that the WGPu will be stored in metal or oxidized form and that is what I will discuss here.

Separated WGPu from retired weapons is typically stored in 'pit' form. The pit is the core of the bomb, a spherical shell containing the WGPu.⁵⁴ The pit is sealed in a protective metal jacket to prevent oxidation (spontaneous ignition).⁵⁵ The container is then placed into a reinforced bunker called an "igloo". Pits have proven to be stable in this form for many years and safe storage for several decades is viewed as technically feasible.⁵⁶

Storage has been occurring for a number of years at the Pantex facility in the US. As any disposition strategy will require intermediate storage, efforts are currently underway to expand the capacity of this facility.⁵⁷ Russia has four facilities for dismantling its weapons but it faces similar storage problems. It appears that in both cases capacity will be expanded through redesign of existing facilities, utilization of other governmental facilities or through the construction of new facilities.

Both countries continue to perform these dismantling and storage operations under the highest level of security and secrecy. The need for treaty verification though may change all that. Of most importance in the verification process is assurance that weapons scheduled for dismantling, are dismantled. Those efforts are for naught, however, if the separated fissile materials are not closely guarded. Russian officials have expressed willingness to allow monitoring of weapon dismantlement but only on a reciprocal basis. This reciprocal monitoring was opposed under the Bush administration, labeling such

⁵⁴Cameron, K., *Taking Apart the Bomb*, Popular Science, April 1993 p.67

⁵⁵National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.118

⁵⁶ibid. p.119

⁵⁷ibid. p.113

an arrangement as “intrusive, expensive, and unnecessary”.⁵⁸ The Clinton administration though has stated its intention to allow IAEA inspection of fissile material no longer needed as a deterrent. This is a major change in policy and is likely to spur dismantling efforts.

If physical barriers are not imposed on the WGPu, continued verification and safeguarding will be required. This will be costly and obviously delay any meaningful disposition efforts. Currently RGPu stored in Europe is safeguarded by a European organization, Euratom. The cost of such storage is estimated between \$1000 - \$4000 per kilogram per year.⁵⁹ The cost of storing 50 tonnes of WGPu at these rates would be between \$50 to \$200 million per year. The actual costs will be higher as WGPu will require stiffer security measures, but these values show that even simple storage is not cheap.

3.3 Discussion

The only conclusion this author can reach is that there is no obvious disposition choice. Each option has its strengths and weaknesses. Table 2 is an attempt to summarize in an admittedly vague and contestable manner the ranking of each option in several categories of concern. Highest, medium and lowest values represent the relative ability of each option to satisfy the identified concern. The uncertainty surrounding each option prevents a more definitive treatment.

	Time	Cost	Fuel Value	Safety	Verification
Burn	10-20 yrs	~\$1-10 b	highest	lowest	lowest
Waste	10-20 yrs	\$0.7-1.6 b	lowest	highest	medium
Storage	0	\$200 m/yr	medium	medium	highest

Table 2 - Estimated Relative Ranking of Options

It should be emphasized that the values in the table are purely speculative. The ranking of the options may be meaningless if the span of the assigned values is within an acceptable range. For example, while reactor burning may be given

⁵⁸Lockwood, D., *GAO Study Outlines Obstacles To Soviet Warhead Dismantlement*, Arms Control Today, April 1993 p.25

⁵⁹Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992 p.9

the lowest mark in the safety category, the associated risk of that option may still be quite minimal.

The next chapter attempts to remove some of the technical uncertainty associated with the vitrification option. This option was chosen for analysis as relatively little research has been performed in this area and its potential benefits warrant additional consideration. While its cost, safety and verification aspects may make vitrification an attractive disposition option, its flexibility and potential for fast implementation are its key advantages.

Glass compositions that would contain HLW as well as 'clean' glasses were investigated. As mentioned previously it would be relatively quick and easy to vitrify the Pu without HLW. Those of us in the Plutonium Research Group at MIT believe that while a WGPu glass containing HLW would be among the most proliferation proof options, un-spiked glass storage may promote expedient disposition as well as present substantial barriers to non-governmental diversion.

Russian opposition to vitrification may also be removed as this form would allow future retrieval (by an industrialized organization) and use as fuel. If durably designed for long-term storage, this clean glass would also be suitable for additional mixing with HLW or direct disposal into a repository should the economic value of Pu fuel remain low. This flexibility could speed disposition as parties would not be worried about being locked into one option.

Compared to reactor burning vitrification also has several advantages. The NAS study states the main benefits gained from reactor burning stem from the chemical mixing of the plutonium with difficult to separate, radioactive fission products.⁶⁰ Virtually all the same barriers provided by spent fuel could be designed into a glass form (including elimination if dilution were high enough). Moreover, a concentration of fission products higher than in spent fuel may be incorporated into the glass.

⁶⁰National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.154

While a clean glass would not provide a radiation barrier, it has been noted that this barrier will weaken with time. It may also be possible to include sufficient amounts of chemically similar elements with dilute concentrations of Pu to successfully deter recovery by a sub-national group. Clean vitrification would not link disposition to spent fuel disposal in this country either, which has implementation problems of its own.

If vitrification instead of reactor burning were used in Russia, new facilities would probably need to be built, but disposition would not be tied to the expansion of their nuclear industry. The Clinton administration would presumably have fewer reservations about funding such an operation as it (in general) does not support the use of Pu in reactors. In any respect the potential of this option warrants further investigation.

Chapter 4

Technical Investigation

4.1 Scope

For vitrification of any design to be considered a viable disposition option it must provide hurdles for would be weapon manufacturers (particularly sub-national groups). This means that however the Pu is altered, the final product must be extremely problematic if not unusable as an explosive. Additionally, the Pu storage form should be environmentally acceptable over an indefinite storage period. The storage form should maintain its integrity in a repository type environment, successfully avoiding a criticality accident and/or an unplanned release of any harmful elements.

Regarding weapon usefulness, the vitrification option has the potential for isotopic as well as chemical dilution of the WGPu thereby degrading its explosive potential. The degree of degradation depends on the diluents used and their ease of separation. Elements chemically similar to WGPu (and thereby difficult to separate) as well as RGPu were chosen for investigation as potential diluents. A computer code was then used to examine the weapon characteristics of these mixtures and identify those diluents most damaging to weapon performance.

It should be noted that no processing option (other than elimination) is entirely irreversible. Theoretically, chemicals can always be separated and even RGPu could be isotopically enriched through laser isotope separation.⁶¹ However, certain elements are decidedly more difficult to separate from Pu than others, invoking economic and time penalties to a would be proliferator. The issue of retrievability was not examined experimentally in this thesis but should be a portion of future work.

⁶¹Mark, J., *Reactor-Grade Plutonium's Explosive Properties*, Nuclear Control Institute, August 1990
p.6

Another perceived benefit of vitrification relates to environmental and human safety. The ability of glass to retain its constituent elements in the long-term decreases the risk of human exposure to its constituents. Potential WGPu glass compositions were fabricated and tested to determine their stability and leach resistance.

The environmental durability of the WGPu glass is significant as it may contain HLW. Adding the Pu and any other diluents must not promote fission product release. Elemental leaching may also cause the loss of reactivity control in the long-term.⁶² As mentioned earlier if significant amounts of neutron absorbers leach from the waste form, a criticality accident may occur. An ideal waste form would contain absorbers that leached at the same rate as the Pu, thus controlling reactivity until the Pu decays away.

Related to the question of environmental safety is the limit of Pu loading into a glass frit. The more Pu that can be dissolved into the glass the faster and cheaper it can be disposed. There is a limit however. At some point the Pu may have detrimental effects on the properties and phase stability of the glass. Determination of that limit is briefly examined experimentally.

The remaining sections of this chapter explain in detail the experiments performed in an effort to generate new information on the technical capability of vitrification to meet weapon degradation and environmental safety objectives.

4.2 Degradation Effectiveness

In order to perform an objective evaluation of the various diluents, specific parameters important in weapon design had to be selected as a metric for comparison. The next section explains the parameters chosen as well as the computational methods and procedures used to calculate values for those parameters. The specific cases investigated and the obtained results are then presented.

⁶²Taubes, Gary, *Plutonium Disposal: No Easy Way to Shackle the Nuclear Demon*, Science, Vol. 263, Feb. 4 1994 p.631

4.2.1 Computational Procedures

The computer code MCNP (Monte Carlo Neutron Photon transport) was used to calculate critical mass and neutron flux data for various WGPu storage matrices. Critical mass is a relevant parameter as this value affects the size of the weapon and therefore the amount of material necessarily diverted for weapon manufacture.

The surface flux from one compressed critical mass of material was also calculated for different WGPu mixtures. This value provides relative information about the flux of neutrons in the material itself, thus gauging each mixture's susceptibility to predetonation. The neutron production rate per kg was also used as a measure of predetonation potential.

Monte Carlo works essentially in the following way. Monte Carlo traces individual particle movements through a specified material composition and geometry. During each particle 'walk', statistical distributions are used to determine what physical interactions occur and when. Thousands of particles are tracked and the results are averaged to determine system characteristics. This should be contrasted with typical deterministic methods which solve equations for average particle behavior. MCNP calculates numerous system parameters. Calculating k_{eff} allows the determination of critical mass and the surface flux (as mentioned previously) is used as a measure of predetonation potential.

For all MCNP runs used in my analyses a bare sphere geometry was assumed. The mixture was also assumed to be homogeneous and surrounded by a vacuum. Obviously weapons do not exist in a vacuum (they are actually surrounded by neutron reflectors and explosives for implosion). However this simplification was assumed adequate for comparative purposes.

For the criticality calculations the density for each mixture was calculated and then doubled in an effort to simulate the compression of an implosion weapon. The Pu was assumed to be in alpha phase possessing an uncompressed

density of 19.5 g/cc.⁶³ Most critical mass references in the literature are for an uncompressed bare sphere. Compression was used in the analyses because it could easily be entered into MCNP and represented a slightly more realistic configuration. An initial critical radius of the material was estimated and then adjusted until a k_{eff} of one was reached.

Neutron flux data was calculated by tracking the number of neutrons that exited a solid sphere of each of the mixtures. The radius of the sphere was such that one compressed critical mass of the material was formed. However, as the material was at its uncompressed density it remained sub-critical. This scenario attempts to determine the neutron population in the precompression stages of detonation, assuming a weapon designer would fuel his/her device with only one compressed critical mass. Sample MCNP input and output files are included in Appendix I.

A source term was then calculated with the aid of the program SFALPYLD (Spontaneous Fission/ALpha neutron YieLD) developed by L. C. Leal at Argonne National Laboratory. This program provides the number of neutrons per second generated from spontaneous fission and alpha absorptions that produce neutrons for a specified mixture of radioactive material. MCNP generates data on a per source particle basis so this information is necessary for the surface flux calculation.

4.2.2 Cases Examined

As mentioned before, the degree to which vitrification successfully degrades the weapon useability of WGPu depends upon what is included in the melt. Isotopic dilution has the benefits of increasing the critical mass of Pu as well as increasing the probability of predetonation (as explained in Chapter 2). Chemical dilution reduces the fissile atom density thereby increasing the critical mass. Additionally, if the nuclear properties of the chemical diluents are such that they steal neutrons from the fission chain reaction, critical mass is increased further. This section explains the specific compositions examined and why they were selected.

⁶³DeVolpi, A., *Fissile Materials and Nuclear Weapons Proliferation*, Ann. Rev. Nucl. Part. Sci., V36, 1986 p.93

4.2.2.1 Isotopic Dilution

The approach used to determine the marginal effects of isotopic blending was to assume reasonable WGPu and RGPu isotopic concentrations (see Table 1) and mix them together at different weight percents. Blending with RGPu is the only mechanism for isotopically diluting the WGPu other than reactor burning. Stockpiles of RGPu do exist in Europe and elsewhere which could be used for this purpose.

4.2.2.2 Chemical Dilution

The effect of chemical dilution was also examined by assuming the Pu was maintained as weapons-grade and inserted in different storage forms. The matrices examined for these calculations included those currently in use today as well as prospective storage forms.

We began by examining metallic Pu and Pu oxide as these are the most common configurations for separated Pu. Plutonium pits are metallic and RGPu is stored as sintered PuO₂. Data from these forms provided a base case to compare the other alternatives.

Fuel forms were studied to determine the security gains by simply treating the WGPu as fuel in reactors and storing it. Mixed-oxide fuel (MOX) is the most common fuel used to burn Pu globally. Calculations were performed on a typical, low enriched (4% fissile) MOX fuel composition. The results of these tests allow us to compare advanced storage forms with that of a standard Pu fuel on their ability to degrade weapon performance.

Another potential storage form is as fuel for the IFR (Integral Fast Reactor). This fuel is composed of 20% WGPu, 70% U, and 10% Zr by weight.⁶⁴ The IFR has been promoted as an advanced reactor by Argonne National Laboratory. They view WGPu disposition as a way for launching their technology. We are interested in the ability of the fuel form (unburned) to hamper weapon use.

⁶⁴Conversation with Dr. Jim Laidler, Argonne National Laboratory, Argonne, Illinois Aug. 13, 1993

The waste form investigated was a vitrified mixture of rare earths. Rare earths are found in the lanthanide series and are chemically similar to Pu, thereby hampering recovery. The rare earths gadolinium, samarium and europium were initially selected for testing. Thorium was included in the analysis as it also is difficult to separate from plutonium. These elements were mixed with WGPu so that each element occupied 20% by weight of the mixture.

4.2.3 Critical Mass Results

The critical mass of the mixtures depends on the concentration of the fissile elements (Pu-239 & 241), the density of the mixture and the diluents' cross sections for processes competing with fission. As noted previously, the 2x compressed critical mass of each of the storage forms was calculated as this value is more representative of the material required in an implosion type nuclear explosive. Isotopic blending was examined first, followed by chemical dilution.

Isotopic dilution, through blending, has a minimal effect on critical mass. The compressed critical mass was calculated to be 2.7 kg for WGPu and 3.4 kg for RGPu. The small critical mass of RGPu demonstrates its explosive potential. If it were not for the spontaneous fission from the even isotopes, RGPu would be almost as desirable as WGPu.

Chemical dilution is achieved by placing the WGPu in one of the selected storage forms. The effect of such dilution on the compressed critical mass of the WGPu mixture (as calculated by MCNP) is shown in Figure 1.

Decreasing the density from 19.5 g/cc for metallic Pu to 11.46 for crystalline PuO₂ increases the critical mass of WGPu from 2.7 to 7.1 kg. This increase is relatively significant but easily reversed. Thus the critical mass calculations for glass were made assuming metallic weight fractions for the rare earths even though in glass they exist as oxides. Similarly, MOX fuel was assumed to be in reduced form.

A comparison of the rare earth and IFR storage forms in Figure 1 shows the effect of neutron absorbing diluents. For the same WGPu loading (20%), the critical mass of the rare earth mixture is 30 times greater. The MOX fuel critical

mass dominates the comparison due to its relatively low Pu concentration (approximately 4%). However, the process for separating U and Pu by solvent extraction is widely known.⁶⁵

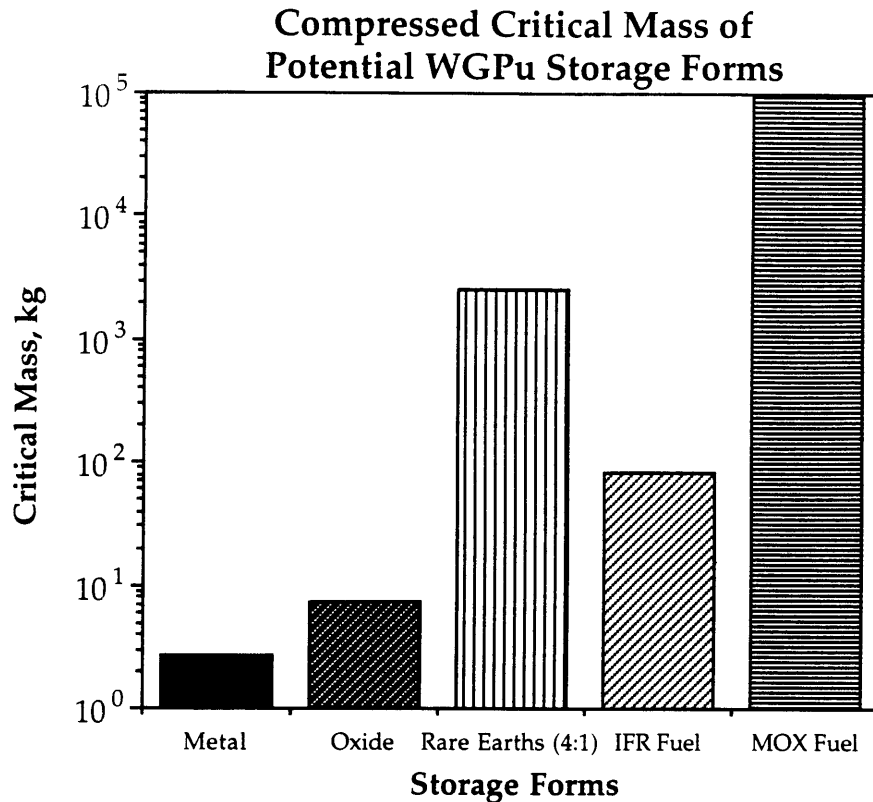
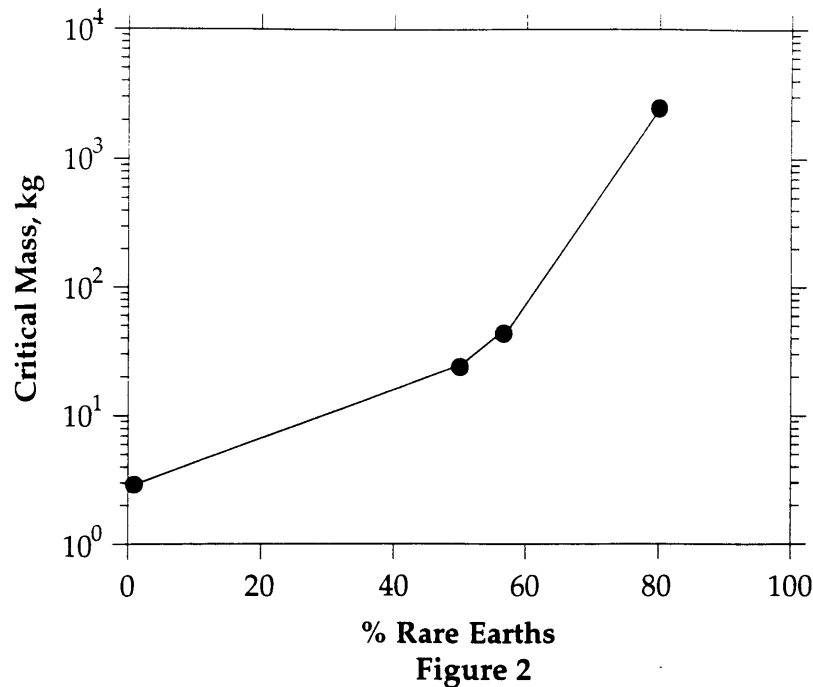


Figure 1

As the percentage of rare earths increases, the compressed critical mass of the mixed glass increases in the manner described in Figure 2. Critical mass increases greatly when the rare earth loading is in the 50-80% range. However, these beneficial effects from rare earths dilution are useful only as long as they remain in the Pu mixture.

⁶⁵Benedict, M., et al, Nuclear Chemical Engineering, McGraw-Hill, Inc., New York, 1981 pp.157-215

Rare Earth Effects on Compressed Critical Mass for WGPu



It is important to know the gains made by weaponeers from incremental separation attempts to separate the WGPu from the glass. Thus, the compressed critical mass of the mixture remaining after incremental decontamination attempts was calculated and is displayed graphically in Figure 3. The purification levels were chosen to span a range of decontamination levels and do not represent results from laboratory testing or actual processes. The actual extraction would roughly follow the process for extraction of Th from ores but this has not been verified or tested.

The first stage represents converting the glass to a reduced metal mixture. The next step assumed 90% of the rare earths were removed but none of the Th. The third stage removed all but trace elements of the rare earths and the final step removed 99% of the Th. The uncompressed critical masses are included to give an idea of the care required in processing to protect against a criticality accident. This graph shows the importance of separability to security maintenance.

Critical Mass of WGPu Mixture After Several Retrieval Attempts

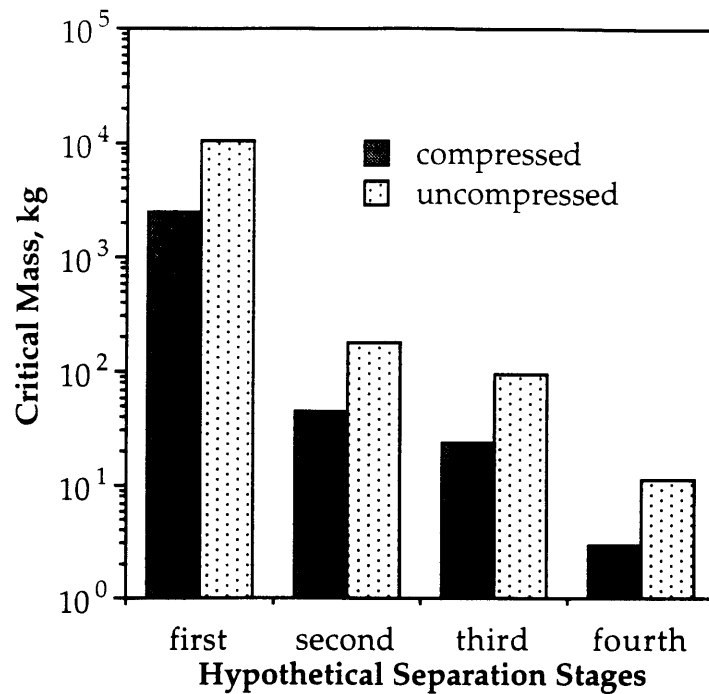


Figure 3

In order to investigate each individual rare earth's contribution to the increased compressed critical mass Figure 4 was generated. The fission neutron energy spectrum as well as the relevant capture cross sections for these rare earths is included in Appendix II. Europium's relatively large neutron capture cross section (~0.1 barns at 1 MeV) combined with its low density (5.26 g/cc) make it a good diluent.⁶⁶ At a loading of 50% the compressed critical mass of the Eu/WGPu mixture is over twice that of Gd or Sm. In the interest of mapping out the maximum potential benefit from rare earth dilution, Eu was selected for use in the remaining chemical dilution analyses.⁶⁷

⁶⁶Cross section taken from ENDF/B Cross Sections, Brookhaven National Laboratory, Upton, New York, November 1972.

⁶⁷Europium is present in the earth's crust at a concentration of 2.1 ppm. This is approximately the same as tin. Gold is present at 0.004 ppm. [Source - The Elements, John Emsley, Oxford Univ. Press, NY 1989]

**Compressed Critical Mass
for a WGPu Mixture Containing
50% of a Single Rare Earth**

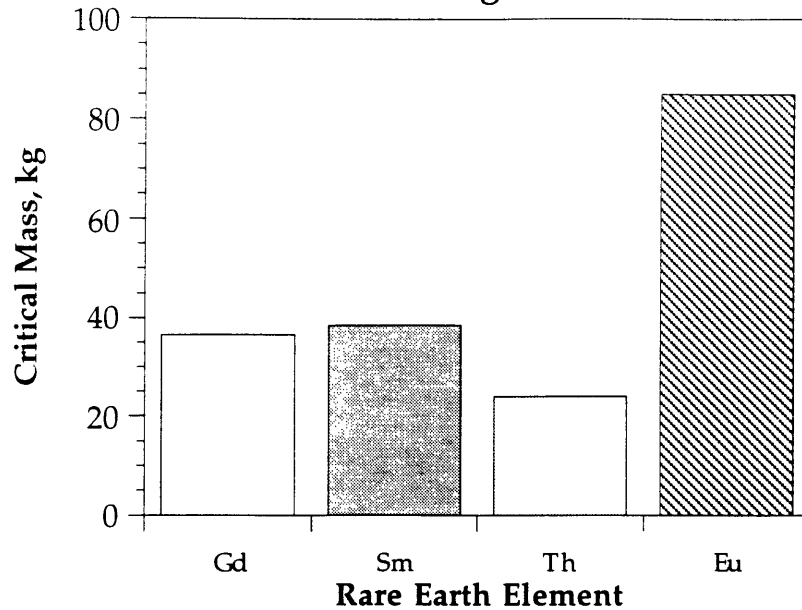


Figure 4

The critical mass data describes the minimum amount of material necessarily diverted for weapon use. If the facilities similar to those at Savannah River are used the Pu will be sequestered in 1800 kg glass logs. The higher weight fraction of Eu, the greater amount of the Eu/Pu mixture required to form one critical mass. If the Eu loading is high enough more than one glass log will be needed. Figure 5 displays this relationship for Pu with an isotopic mixture of 70% WGPu-30% RGPu. The heavy line shows the amount of Eu/Pu mixture necessary for detonation in a design similar to Trinity's (three critical masses).⁶⁸ As the fraction of Eu increases the required material also increases as one would expect.

However, the amount of Eu/Pu material present in each log is variable. For example if the WGPu is blended with 20% Eu and mixed into a glass frit so that the Pu comprises 2% of the entire glass, thieves would have to steal more than one log to obtain one Eu/WGPu critical mass. Alternatively, if weaponeers obtained a glass log loaded with 2% WGPu and copious amounts of Eu they

⁶⁸Conversation with Dr. Marvin Miller, Professor of Nuclear Engineering, Mass. Institute of Technology.

would have to refine the Eu/WGPu portion below 23% Eu to obtain the required amount of material. The refining break points for 1% and 0.5% Pu loadings in the log are also shown in the figure.

The Amount of Explosive Material Found in WGPu Glass Diluted With Various Amounts of Eu

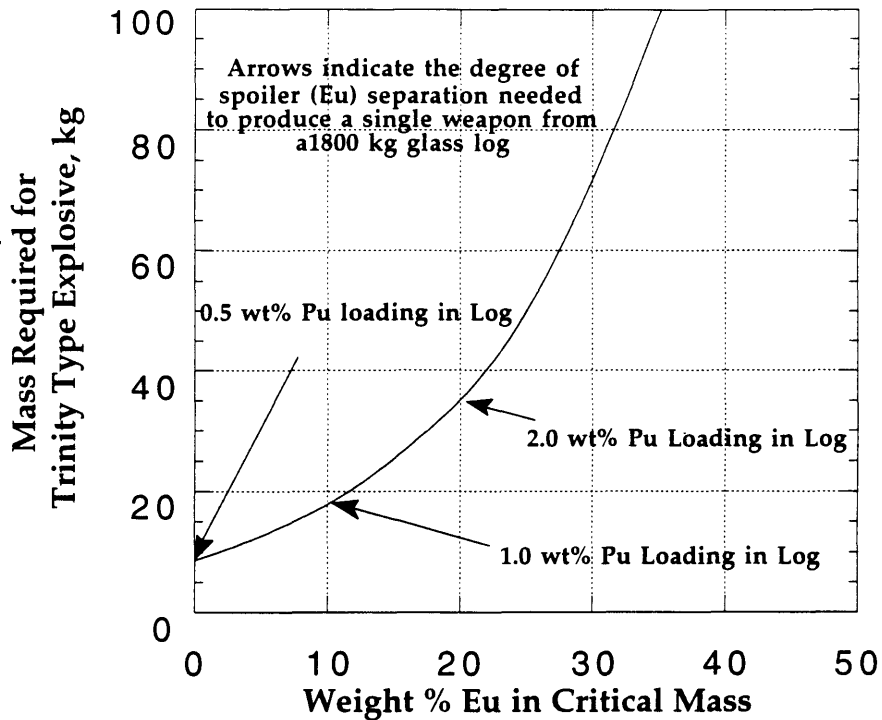


Figure 5

As the Pu loading in the glass is increased the material requirements for successful weapon fabrication are reduced. Yet there are economic and other incentives for doing so. The amount of time and effort spent on processing the material would be reduced. As the Pu would be in a significantly improved form, verification and safeguarding efforts could be relaxed sooner, averting additional storage costs. Therefore, there exists a trade-off between added security and cost/speed of implementation.

4.2.4 Surface Flux and Neutron Production Results

The results on neutron flux from blending WGPu with RGPu is shown in Figure 6. In this case a constant mass of Pu was assumed instead of calculating

the flux based on a single compressed critical mass. This is because blending changes that value. An uncompressed Pu mass of 9.033 kg was therefore chosen (from a rough calculation aimed at selecting a slightly sub-critical mass).

Blending relatively small amounts of RGPu into WGPu (maintaining a constant mass of 9.033 kg) increases the relative percentage of isotopes Pu-240 and 242. These even isotopes increase the spontaneous fission rate of the Pu. A blend of 30% RGPu more than doubles the neutron flux at the surface of the sphere.

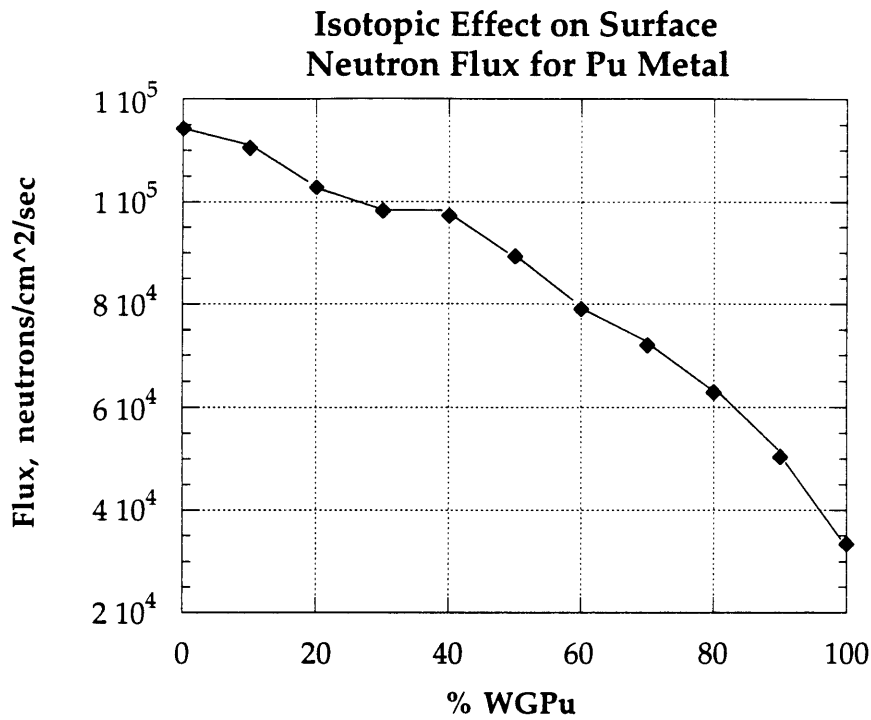


Figure 6

While the isotopic blending has a noticeable effect on surface flux and therefore predetonation, chemical diluents have less an effect. Comparing the flux from one compressed critical mass, the mixture of 80% rare earths increased the surface flux from 2934 n/cm² (for pure WGPu) to 3483 n/cm².

In this case the increased amount of WGPu is almost completely offset by the added absorption and decreased density of the mixture. On a per kilogram basis the rare earth dilution obviously reduces the neutron production rate by

1/5 (Pu occupies 20% of the mixture). Similar analyses were performed on the other storage forms and the results are included in Appendix III. A summary of all MCNP results is in Appendix IV. In general dilution does not increase the predetonation potential unless neutron producing diluents are used.

4.3 Environmental Safety

While a rare earth glass may provide security benefits for WGPu storage it must also prove to be environmentally safe. If radioactive fission products are incorporated into the glass along with the Pu, the glass must still retain excellent environmental durability characteristics. In addition, borosilicate glasses may not provide sufficient reactivity control in the long-term for high WGPu loadings.

This is because boron preferentially exits (leaches) from borosilicate glass at a rate much higher than that of Pu.⁶⁹ As the relative fraction of boron is reduced the potential for criticality increases. Rare earth elements absorb neutrons but leach at a rate similar to Pu, thus providing a potentially safer storage form. The leaching characteristic of these elements were explored experimentally and are described in this section.

The leaching of elements from glass has been well studied in recent history. Elemental release rates depend on numerous factors such as :

- 1) how a given element is incorporated into the glass structure;
- 2) how specific sites in the glass react with water;
- 3) how the composition and reactivity of the leachate influence glass reactivity;
- 4) how the structure and reactivity of the glass changes in surface alteration layers;
- 5) how glass dissolution modifies the chemistry of the leachate; and
- 6) the flow rate of water into and out of the vicinity of the waste.⁷⁰

⁶⁹National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994 p.191-92

⁷⁰Bunker, Bruce C., "Waste Glass Leaching: Chemistry and Kinetics," Scientific Basis for Nuclear Waste Management X, Vol. 84, 1987 pp.493-507

Instead of exploring the specific mechanisms for boron or rare earth release, identical tests on glasses containing both elements were performed to uncover any differences. The next section explains the testing procedures used on the selected glasses compositions.

4.3.1 Experimental Procedures

Rather than designing a new glass frit from scratch, two borosilicate glasses were altered to include these chemicals. Glasses comprised of rare earths and an analog to Pu (i.e. thorium) were fabricated. The presence of both boron and europium in the same glass sample provide the comparative data necessary to determine the relative advantages or disadvantages of using Eu for long-term criticality control.

Advanced Reference Material (ARM-1) and Savannah River Laboratory-165 frit (SRL-165) were selected for our experiments. The exact composition of each glass is shown in Appendix V. ARM-1 is a borosilicate glass that contains simulated fission products. This is an important glass for laboratory tests as it has been prepared to simulate a typical HLW composition. Using the ARM-1 glass in our experiments represents the option of mixing the WGPu directly with highly radioactive waste, introducing a deadly radiation barrier. SRL-165 is a 'clean' borosilicate glass used at the Savannah River Laboratories Defense Waste Processing Facility (DWPF). While the SRL-165 glass would not present a radiation barrier, the incorporation of WGPu would assumedly be a simpler and less hazardous undertaking.

Both the ARM-1 and the SRL-165 were used in the same manner. Measured amounts of rare earth elements and thorium (in nitrate form) were mixed in with the standard glass compositions and inserted into a furnace. The powdery mixtures were heated at 1150° C until they melted, poured into a graphite mold and left covered to cool. They were then annealed at 500° C and cut into samples for use in the Materials Characterization Center testing procedure (MCC-1P).⁷¹

⁷¹MCC-1P Static Leach Test Method, Materials Characterization Center, Revision 9-30-83

MCC-1P is a standard procedure for gaining leach rate data on various vitrified materials. The sample preparation and testing procedures required by MCC-1P were followed. Cleaned samples of glass were placed on mesh baskets in Teflon containers and submerged in deionized water. All compositions were tested for 7, 14, and 28 days at room temperature and 90°C.

At the termination of the testing period the glass samples were removed from their containers, rinsed and dried. The leachate was then removed and filtered with a syringe for analyses using inductively coupled argon plasma spectroscopy (ICP).

The ICP spectrometer is used to measure the concentration of certain elements in an aqueous mixture. A peristaltic pump generates a flow of leachant which is injected with argon gas to create a liquid-vapor mist. This mist is passed through an argon flame which is magnetically contained for optimum detection. As energy is transferred to the mist, the elements present are excited. The de-excitation of these elements create x-rays of characteristic wavelength. The spectrometer records the number of x-rays detected and reads the concentration from a concentration vs. count curve.

The curve used for comparison is generated from standards of known concentrations. The concentration of an element is inputted into the controlling software and its associated spectrum recorded. A calibration curve is then generated using a linear polynomial equation. The sample spectrum is compared with values on the calibration curve to determine the appropriate concentration.

Knowing the concentration of the element in the leachant, the volume and density of the leachant, and the surface area of the glass, it becomes possible to determine the amount of element released per unit surface area of the glass (g/m^2). This is the standard method for expressing the results of a MCC-1 leach test thus allowing our data to be compared to other tests.

The upper boundary for Pu loading in the ARM glasses was also tested. Plutonium has been shown to be soluble at 7% and it has been suggested that

10% could be an upper limit.⁷² An ARM glass mixture was made with 10% thorium oxide to identify any phase transformations that may occur at that loading. A liquid-liquid phase separation (one glass containing mixtures of differing composition) is undesirable as it could cause a change in the physical properties of the glass and/or decrease the elemental leach and bulk glass dissolution rates.

Visual examination of all glass formulations were performed to check for gross crystallinity. An energy dispersive X-ray micro-analyzer (EDAX) was used to determine the chemical composition at various locations in the glass to identify inhomogeneities. A scanning electron microscope (SEM) was used to examine the structure of the glass at roughly 10^6 magnification to locate any micro-crystallization. Crystallization is an undesirable phase transformation as these sites are more susceptible to leaching.

4.3.2 Cases Examined

The elements selected for analyses with the ICP were silicon, sodium, boron, thorium, europium and cesium. Silicon is a network former meaning that as molten silica cools it enters a solid, vitreous state as opposed to a crystalline state.⁷³ As a major structural component of glass, the leaching of silicon gives an indication of the overall decomposition of the glass.

Sodium is a network modifier which means it cannot form a glass by itself, but when mixed with other network formers it is capable of entering the glassy state. As such it is the principle flux in glass manufacturing. Sodium is very soluble and its leaching characteristics will again give us some idea of the overall dissolution of the glass.

Boron oxide is a glass former but it has a high solubility and is not expected to precipitate. We are interested in its leaching characteristics in order to compare them to europium's. This will indicate Eu's potential for controlling reactivity (relative to boron) in the long-term.

⁷²McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p. 8

⁷³Scholes, S. R., *Modern Glass Practice*, CBI Publishing Company, Inc., Boston 1975 pp. 33-34

The thorium release data will provide an indication of plutonium's retention in the glass. Cesium data was obtained to determine the affect of ARM modification on the leaching of a representative fission product. This data should indicate the capability of two currently used glass frits to retain elements of potential use in WGPu disposition while continuing to sequester other wastes.

The SpectroFlame ICP used in this experiment has the following limits of detection for these elements:

Element	Limit of Detection, ppm
Boron	0.0015
Silicon	0.0063
Thorium	0.0109
Europium	0.0006
Sodium	0.016
Cesium	19

Table 3 - ICP Limits of Detection

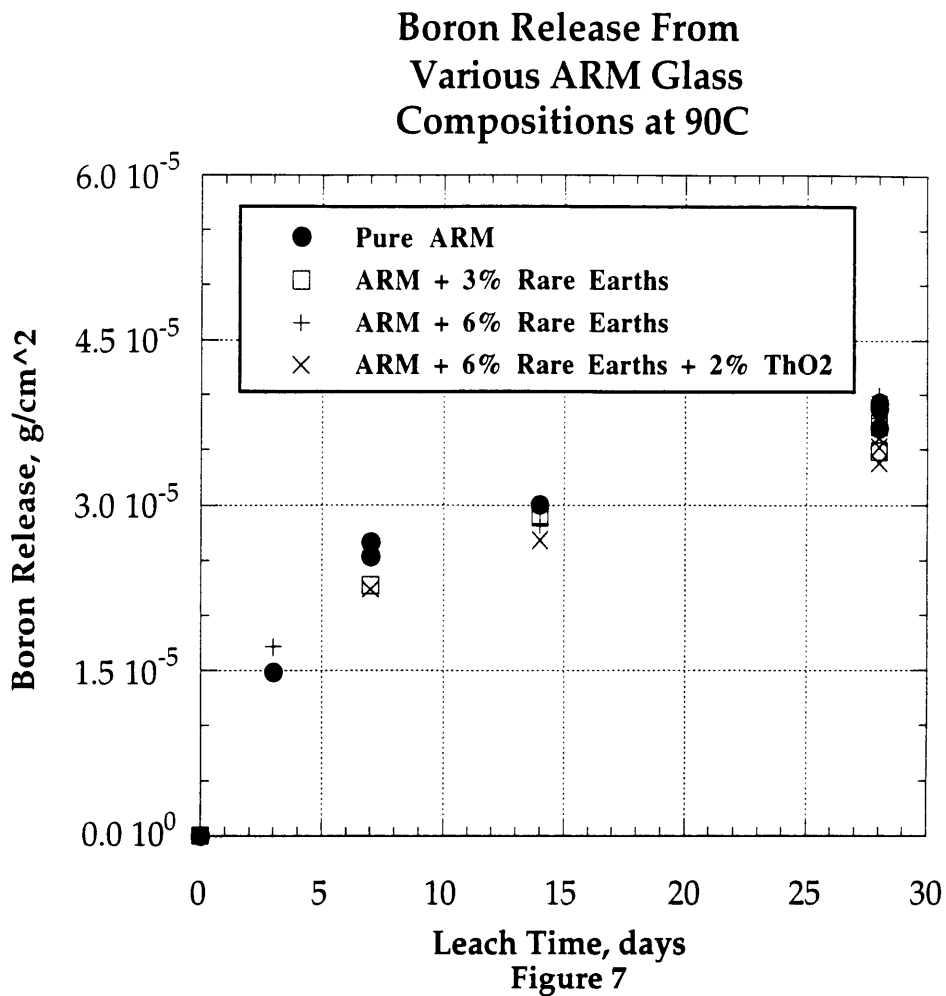
4.3.3 Durability Results

Several rare earth and thorium loadings were fabricated and tested using both the ARM-1 and SRL-165 glass frit. ARM glass was made with a 1% and 2% loading of each (3% and 6% total) of the rare earths. Following these successful fabrications, an additional ARM glass with 6% rare earth loading was made containing 2% thorium. The exact compositions of all the glasses tested are included in Appendix V.

Figure 7 displays boron leach data at 90°C for clean ARM and each of the subsequent rare earth - ARM compositions (3%, 6%, and 6% with Th). The trends observed are representative of the elemental leach data for silicon and sodium. Appendix VI includes the summarized results from all the leach tests performed.

The figure shows that the inclusion of thorium and rare earths in the ARM glass had no detectable negative effect, and possibly a slight benefit on the structural durability of the glass but there is no statistical basis for this claim.

The release curves for Si, Na and B were all very similar over the 28 day period tested for all of the ARM glasses. This was true for both the 90°C and room temperature tests. The ARM glass has shown itself to be a quality glass capable of containing a variety of diluents.



The SRL glass also performed well. Mimicking the ARM glass compositions, SRL glasses were made containing 2% of each of the rare earths Sm, Gd, and Eu. An SRL glass including 6% total rare earths and 2% thorium was also manufactured. Figure 8 shows the boron release over 28 days at 90°C for the SRL glass compositions. Again, the curves for Si and Na release are similar to those for boron.

Boron Release From Various SRL Glass Compositions at 90C

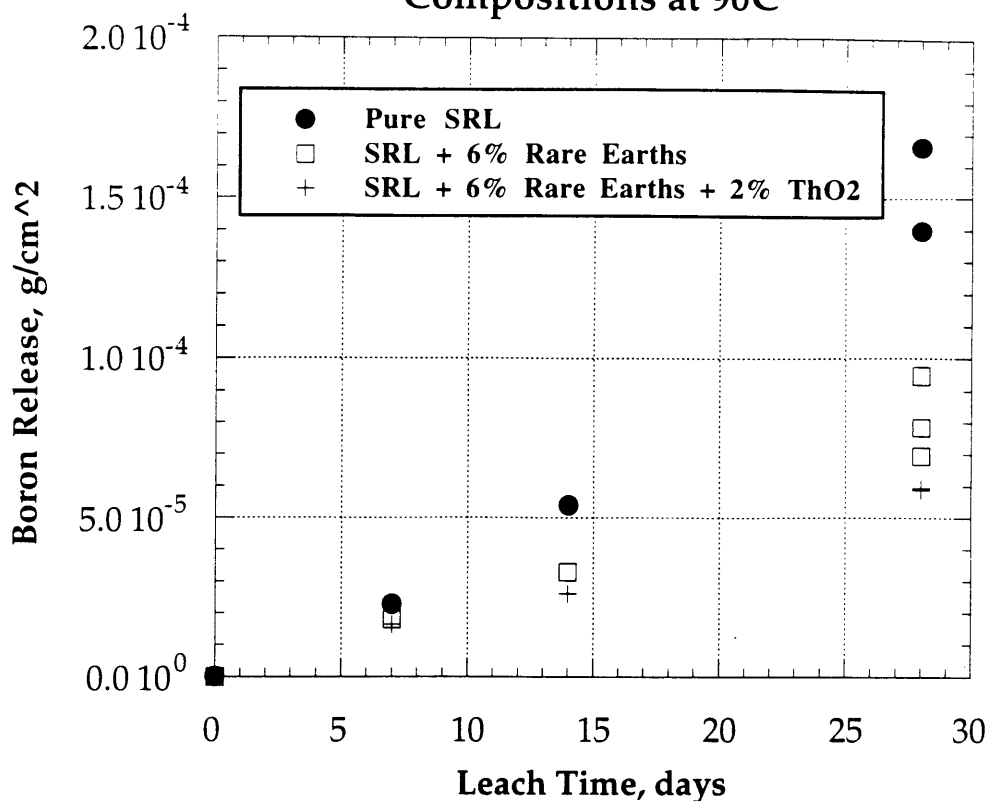


Figure 8

The SRL frit's durability at 90°C also appeared to be unaffected by the added elements. At 90°C the SRL glass containing the rare earths had B, Si and Na releases slightly less than that of pure SRL. With the addition of thorium, these releases became approximately 1/2 those of pure SRL. Thorium apparently aides the formation of the glass network. Thorium has +4 valence state which may allow it to form a tetrahedral geometry with four oxygen atoms similar to silicon in the glass, thus joining with the silicon network and contributing to the network structure.

At room temperature the SRL glasses did experience some loss in durability due to rare earth inclusion. Of the three elements B, Si, and Na, the silicon release rate was increased the most. Figure 9 shows the Si leaching curve for pure SRL and SRL with 6% rare earths. Boron release increased slightly and sodium release remained constant. The magnitude however of these releases

was small. Even the Si concentration in the 6% rare earth SRL glass was below 2 ppm. This should be contrasted with 80 ppm Si in the leachate of the same glass at 90°C.

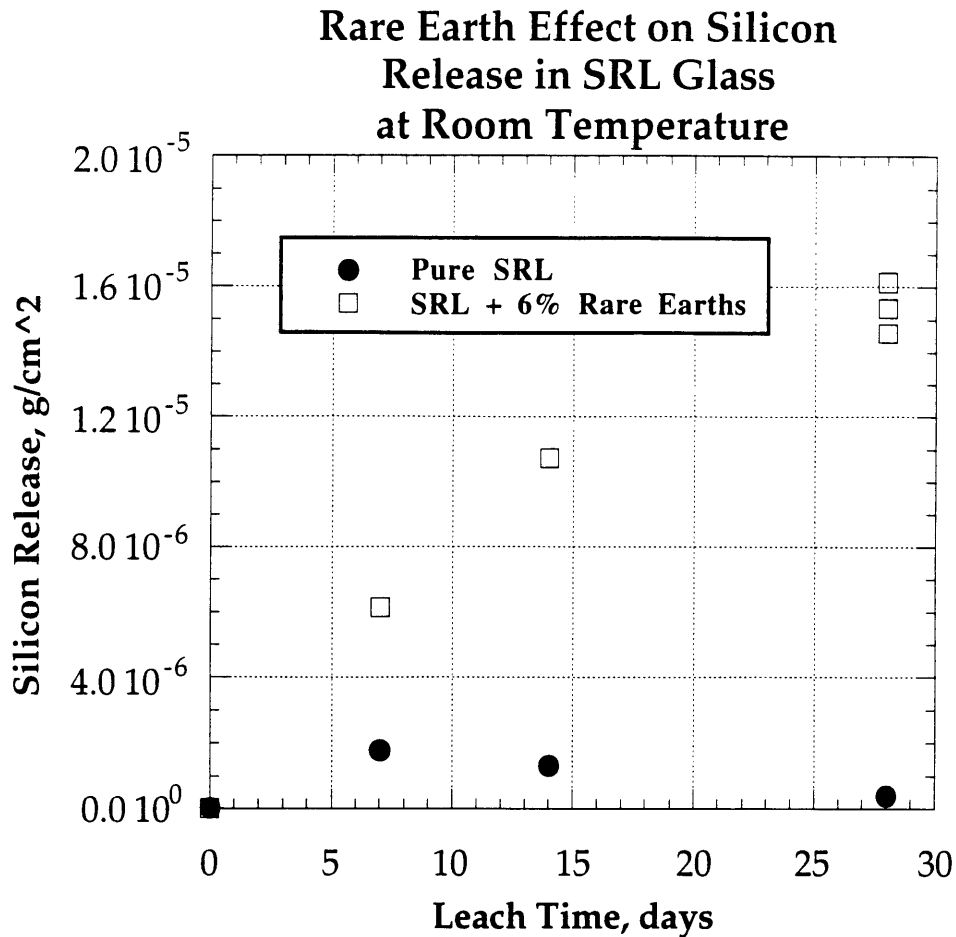
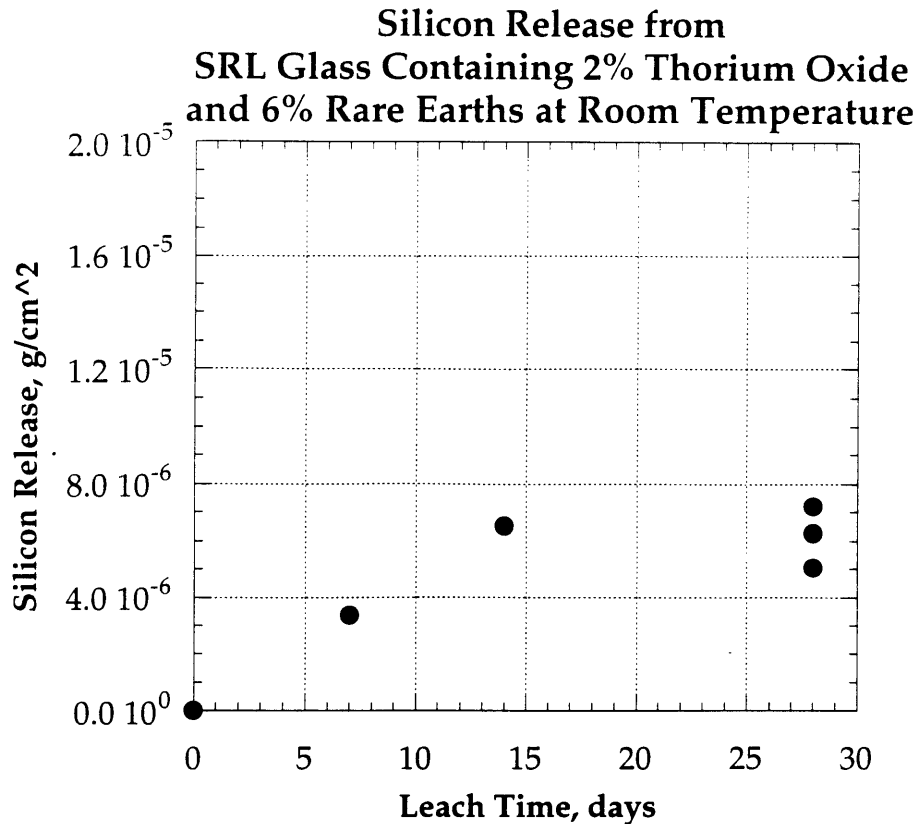


Figure 9

When thorium was added to the rare earth - SRL glass, the retention of boron, silicon and sodium was improved. Figure 10 shows the Si release from this thorium glass. It is only slightly higher than that of the pure SRL displayed in Figure 9. Boron and sodium release is also slightly reduced. This is another example of thorium's ability to increase the durability of a glass.

In the category of durability, the ARM glass outperformed the SRL glass at both 90°C and room temperature. The ARM glass retained a greater fraction of B, Si, and Na initially present compared to the SRL glass. The fission product

simulants have been recognized for some time for their ability to increase the durability of the glass.⁷⁴



4.3.4 Thorium , Cesium and Europium Retention Results

As an analog of plutonium, thorium retention in the glass is of great importance. Fortunately both the SRL glass and the ARM glass were able to hold thorium releases below detectable levels (0.0109 ppm). This was true in both the 90°C and room temperature tests. This reflects the general insolubility of all actinides in aqueous solutions.⁷⁵

The release of Cs was undetectable in all cases as well. This is an important finding as the inclusion of WGPu and any diluents should not affect the ability of the glass to serve its original purpose -- isolate waste.

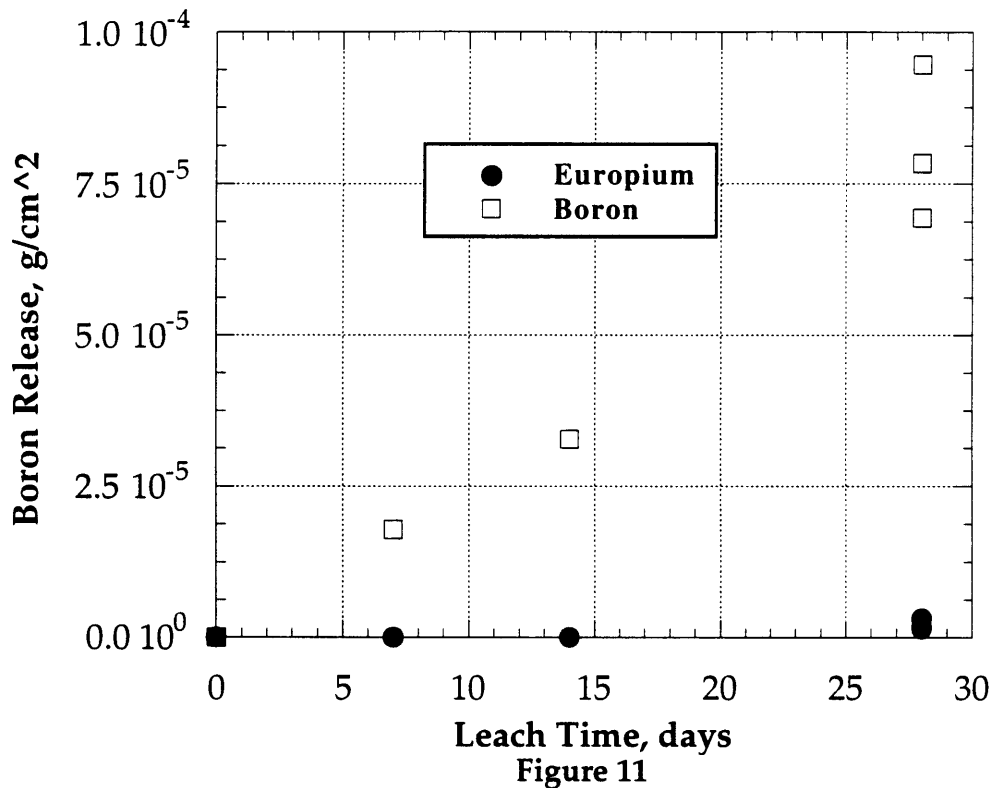
⁷⁴Lutze, W., and R. C. Ewing, ed., Radioactive Waste Forms for the Future, Elsevier Science Publishing Company, Inc., New York, 1988 pp.559-60

⁷⁵ibid. pp.497-499

Unfortunately this effect must be confirmed by further experiments as the limit of detection of Cs by the ICP is quite high (19 ppm). Another fission product should be selected with a lower limit of detection or a more accurate measuring device utilized.

In most of the leachate samples europium was undetectable. When it was found it was present in very small quantities (low ppbs). The SRL glass with 6% rare earths at 90°C had the highest Eu concentration in its leachate and its leach data is displayed in Figure 11. Europium was found at a concentration around 300 ppb in this glass. For comparison purposes, the leaching profile for boron in the same glass is shown in the same figure. The Eu release is exceedingly small compared to boron. When thorium was added to the glass the Eu release in the SRL became undetectable again.

Reactivity Control Release in SRL Glass Containing 6% Rare Earths at 90C



At room temperature the Eu release from 6% rare earth-SRL glass was reduced to 35 ppb. The associated leach data for Eu and B is displayed in Figure 12. Again, the boron release in the same glass is significantly higher. Adding thorium to this composition only slightly reduces (by 5 ppb) the Eu concentration found in the leachate, but again it should be noted that these are very small absolute quantities.

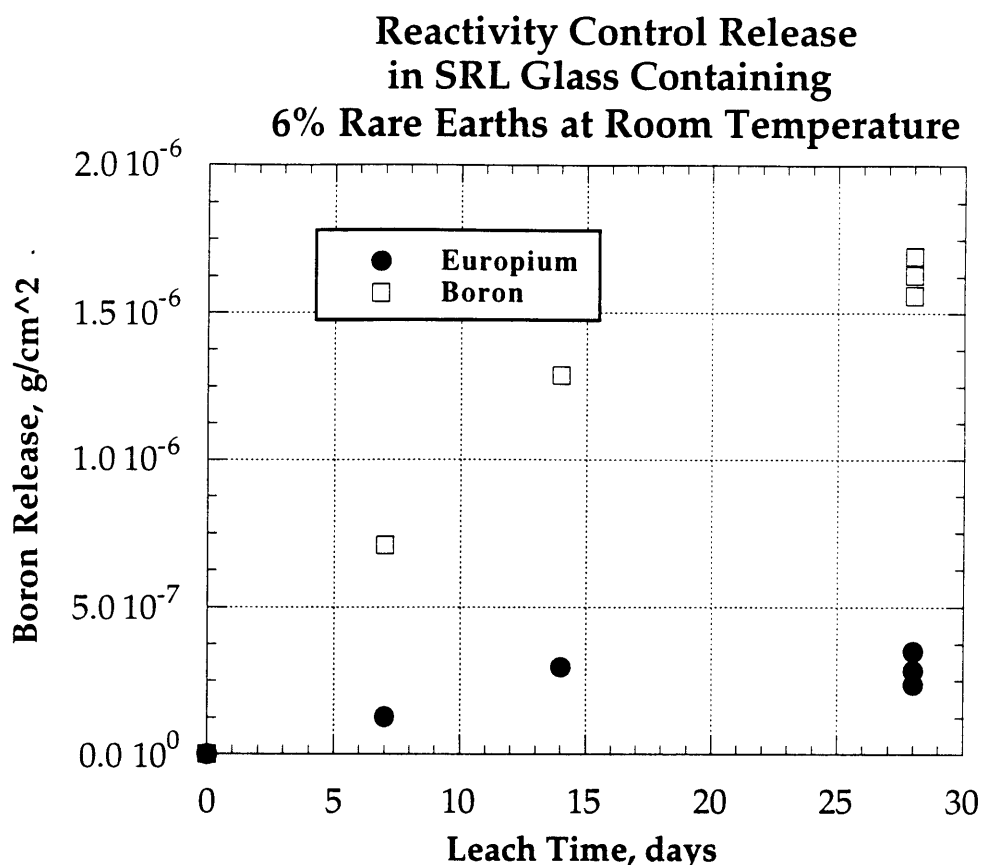


Figure 12

Interestingly, this case where the room temperature Eu release was less than that at 90°C is the only result that behaves as expected with an increase in temperature. In all the other compositions (SRL and ARM) the glasses were more effective at containing the europium at 90°C. It should be stressed that these room temperature releases were still very small (<35 ppb) but this phenomena warrants further investigation.

By and large the ARM glass fared better than the SRL at retaining the europium. At 90°C the europium was undetectable in the leachate. The room temperature ARM glass with the 6% rare earth loading had the highest Eu concentration (of the ARM glasses) in its leachate. Still, its concentration was less than 10 ppb after 28 days. Figure 13 shows the europium leach profile for this glass. Note the smaller scale of this graph relative to the other europium curves. The affect of thorium addition did little to alter the Eu leach rate.

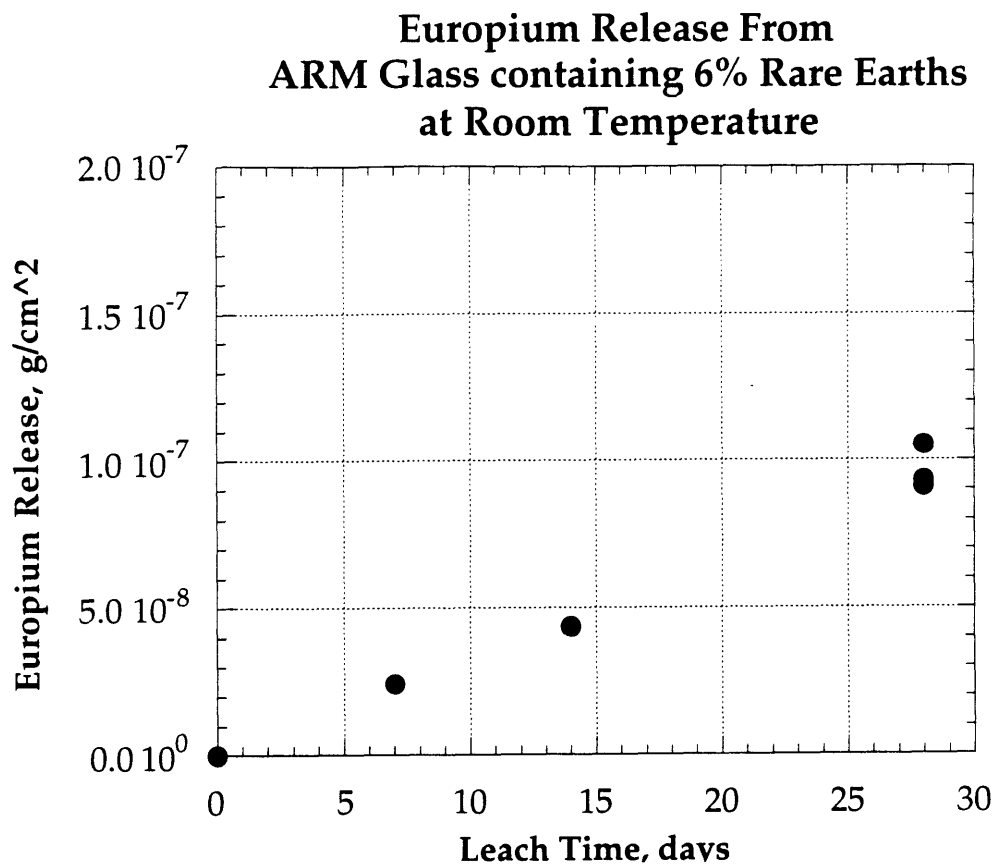


Figure 13

4.3.5 Thorium Solubility in Glass

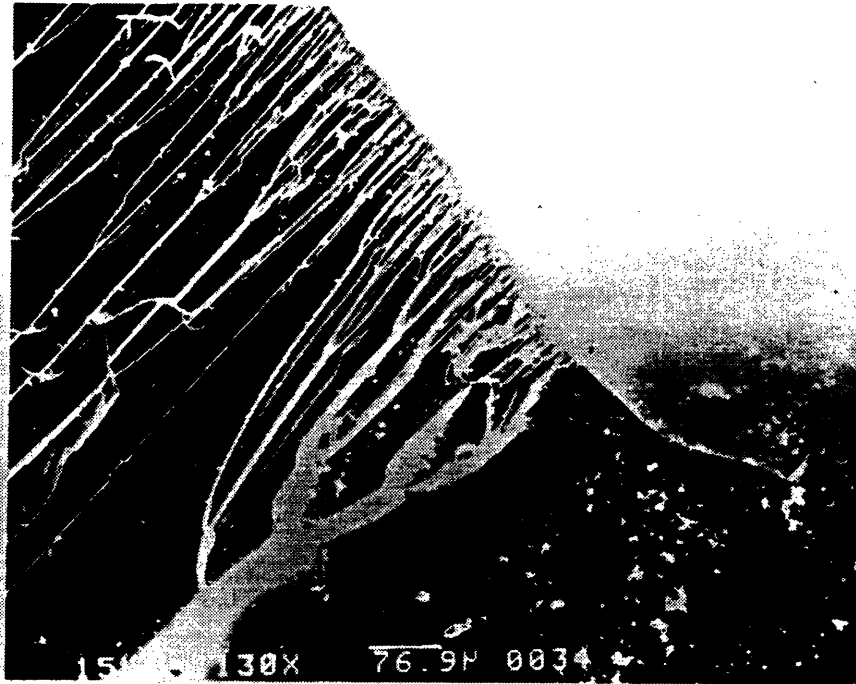
The higher the Pu loading into the glass the fewer number of glass logs required to treat the WGPu. This translates into a faster disposition schedule with potential cost savings. However, increasing the concentration of Pu in the glass increases the risk of a criticality accident or devitrification. It is the later concern that we addressed in our experiments.

The ARM glass frit was mixed with 10% ThO₂ as Th(NO₃)₃·(H₂O)₆ and melted at 1175°C. After the melt was poured and allowed to cool, it was examined visually. Figure 14 is a surface photograph of this composition. Note the strains of a white milky substance. They run the length of the glass bar in a wave-like fashion. This appears to be the onset of crystallization of the glass.



**Figure 14 - Surface Photo of Devitrified Regions
in ARM-1 Glass with 10% ThO₂**

To determine if this was indeed devitrified material, the glass was cracked in a glove box and the fractured surface examined under the SEM. Figures 15 and 16 show that phase separation did indeed occur. Figure 15 is a magnified picture of the fracture itself. The finger-like strains are merely cracks in the glass. However, in the lower right portion of Figure 15 there is a grouping of small dots which are likely to be crystals. The bulk glass upward and to the left of these clusters is free from such material which is typical as crystals tend to form in localized regions, leaving other regions unchanged. Figure 16 is a higher magnification photo of a cluster of these crystalline structures.

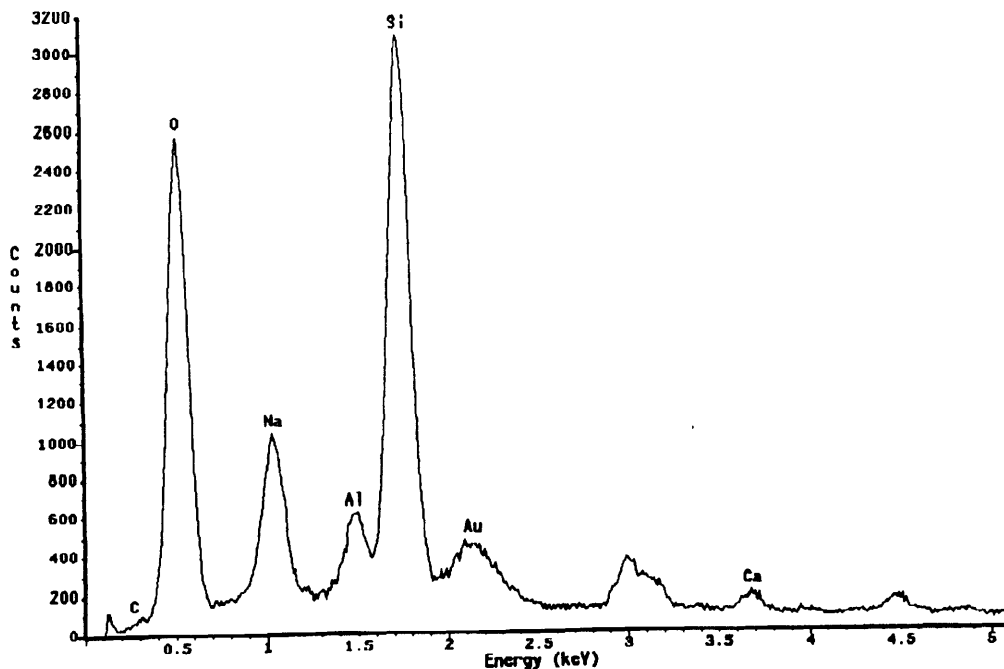


**Figure 15 - Grouping of Crystals
in ARM-1 glass with 10% ThO₂**



**Figure 16 - Crystalline Structures
in ARM-1 Glass with 10% ThO₂**

Figures 17 and 18 contain the EDAX readouts for a portion of the bulk glass as well as a devitrified region respectively. The elemental composition in the bulk glass (Fig. 17) shows the elements we would expect: Si; O; Ca; Al; Na; and Th (the 2.98 keV peak). A gold peak is observed as the sample was coated with it to improve its conductivity under the SEM.



**Figure 17 - Elemental Composition
of the Bulk Region of
ARM-1 Glass with 10% ThO₂**

In Figure 18 the EDAX showed higher relative Si and Ca peaks for the devitrified region than that for the bulk glass. Smaller peaks for oxygen and sodium were also observed. This is consistent with devitrification as Si tends to exclude other elements upon crystallization. It should be noted that while devitrification did occur, Th was not found in the devitrified crystals. This suggests that Th (and presumably Pu) is rather stable in this glass even at such a high loading.

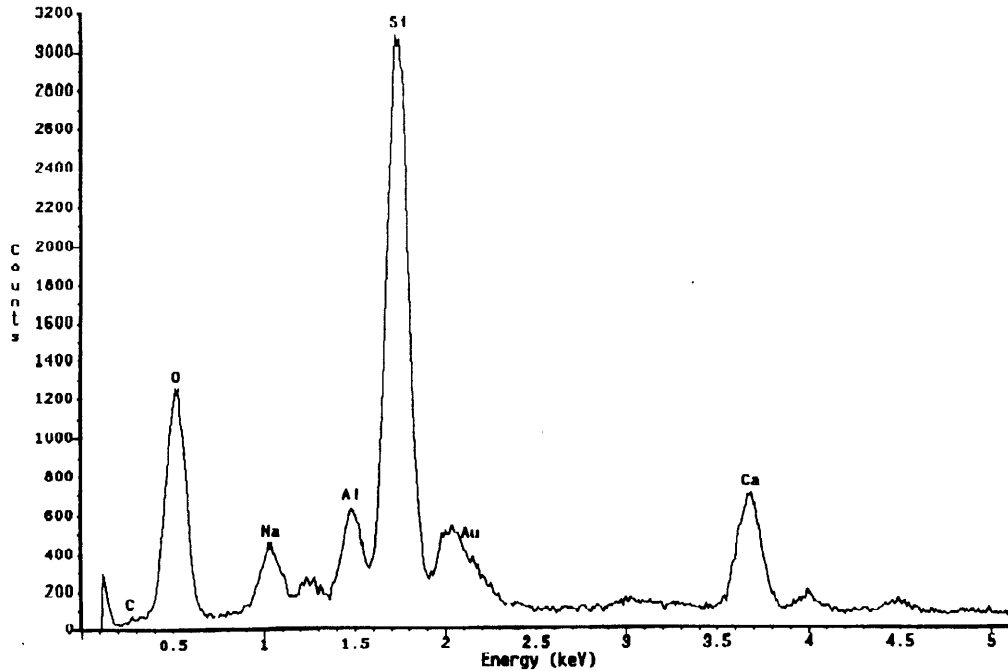


Figure 18 - Elemental Composition from a Devitrified Region of ARM-1 Glass with 10% ThO₂

The conclusions gained from these investigations remove some of the uncertainty associated with WGPu vitrification. In the next and final chapter, these technical conclusion are stated and used in the formulation of an implementation strategy. An effort is made to create a robust strategy that includes alternative options at key processing decision points. Areas of future work are then identified.

Chapter 5

Conclusions and Future Work

This chapter presents the conclusions that can be made through examination of the data obtained through computational and experimental research. These results are then discussed in the context of the policy analysis performed in Chapter 3. A potential strategy for utilizing vitrification technology in WGPu disposition is presented and evaluated in the subsequent section and finally, areas for future work are discussed.

5.1 Technical Conclusions

5.1.1 Explosive Degradation

Neutron production levels approaching that of RGPu can be attained with a minimal amount of blending. With the addition of 30% RGPu the neutron flux from a nine kg sphere of Pu more than doubles. Chemical diluents can dramatically increase the critical mass of the Pu mixture. This effect is meaningful if the Pu is mixed with elements difficult to separate. Radioactive elements may also be added to provide a radiation barrier similar to that in spent fuel. It was also shown that an appropriate amount of europium doping and Pu loading will require the theft of multiple logs of Pu glass to obtain sufficient explosive material. The shear size of these logs would require industrial equipment to move them, thus presenting another proliferation barrier.

5.1.2 Environmental Safety

If the ultimate decision is to treat the Pu as waste, it can be done so safely with current technologies. As shown experimentally, ARM and SRL-165 glass frits have proven themselves to be very accommodating. The addition of 2% Th and 6% rare earth elements into either glass caused no visual damage to the glass (i.e. gross devitrification). The environmental durability of the altered glass was not detrimentally effected. Silicon, boron and sodium release rate were often marginally decreased with the addition of these elements. However, additional testing should be performed to provide confirmation of fission product retention.

The results from the Eu leach tests show that insoluble elements have the potential for controlling WGPu reactivity in the long-term. Europium was consistently more leach resistant than boron.

The solubility limit of Th in ARM-1 glass is higher than 2% but less than 10%. This information suggests that the WGPu loading limit is below that level too. This is important information but it is unlikely that the Pu would be loaded at that high a level due to criticality concerns. Remember, low enriched MOX fuel has less than half the fissile isotope concentration of such a glass.

5.2 Discussion

The problem of WGPu disposition is not a simple one. Imbedded in it are concerns over proliferation, national sovereignty, environmental protection, and energy policy. It would be premature (and beyond the scope of this thesis) to present a detailed plan for WGPu disposition. However, an understanding of the major political factors and the technical potential of the available technologies allows us to construct a general strategy for the use of one technology, vitrification.

What is apparent from the literature is that economic factors will play a major role in WGPu disposition decisions. From the ratification of START II to the ultimate WGPu disposition strategy, the availability of sufficient financial incentives will allow arms reduction to proceed in whatever manner deemed appropriate by the financiers.

The US has great control over WGPu disposition (assuming it is willing to exercise it) because they hold the purse strings. When the US did not provide timely payment of the disarmament funds it agreed to pay in the tri-lateral agreement, Russia halted its shipments of LEU to Ukraine.⁷⁶ This jeopardized the continuation of Ukrainian disarmament. Therefore if the US wants the disarmament process continue running smoothly, it must adequately compensate the disarming parties. But it does not want to be gouged too deeply.

⁷⁶Lockwood, D., *Ukraine Begins Transfer Of Strategic Nuclear Warheads*, Arms Control Today, April 1994 p.20

As mentioned earlier, a rational mechanism for setting the price of the Pu would be welcomed as it would give starting point in compensation negotiations.

Aside from the economic disincentives for US action, there is much to be gained by setting a course for WGPu disposition. A timely selection of a disposition technology could help determine how much Ukraine should be compensated for its weapons, thereby helping the disarmament process move forward. The sooner a path is chosen the faster the actual processing can be performed and risk of proliferation reduced. But hasty action could lock the US into a less than optimal treatment technology.

Given the clear proliferation risks and the uncertainty surrounding the ultimate fuel use or non-use of the Pu, it appears that whatever disposition technology is selected preference would be given to a process that provided the greatest combination of security and flexibility. The final product must provide a high level of proliferation resistance while recognizing the competing goals and vast uncertainty involved.

It is clear that reactor burning (with reprocessing and reinsertion) can provide the same barriers to proliferation that civilian RGPu in spent fuel possesses. The technical analysis presented in this thesis shows the glass option has nearly the equivalent potential (the same if the Pu is highly diluted).

Inherent in the vitrification option is the selectiveness of the process. Any barrier (sans elimination) can be designed into the process. This flexibility is a great advantage over the burning option. The vitrification option also meaningfully degrades the WGPu in the near term while buying time to see how the demand for Pu develops.

5.3 An Implementation Strategy

The US and Russia are two vastly different countries. For vitrification to become applicable in either country a strategy for its use must be adapted that addresses these differences. The following are the major components of a basic disposition program that could be pursued. Portions of this program face real

obstacles to implementation. These obstacles are identified and contingent plans suggested.

1. Collocate

It is estimated that storage will be needed for at least ten years no matter what disposition technology is selected. The timing may be right to give international storage of separated RGPu another try. If enacted, this arrangement would provide a perfect opportunity for storing RGPu and WGPu together.

As shown in the criticality calculations, RGPu (while not as desirable as WGPu) has great explosive capabilities. Storing both Pu grades would recognize this fact and assure adequate proliferation protection.

The projected surpluses of RGPu in Europe will certainly strain safeguarding efforts of Euratom. They may be supportive of collocation as they would be able to share the watchdog burden with the IAEA. Russia however would probably object to foreign storage of its WGPu.

An alternative would be a facility in Russia for housing only domestic RGPu and WGPu under IAEA supervision. As no separated RGPu is available in the US its WGPu would have to be housed alone but IAEA supervision could be granted. The Clinton administration is apparently open to IAEA inspection of surplus WGPu and it would seem that Russia would not object as long as the US was reciprocating.

2. Isotopically Blend

If a Russian collocation agreement could be reached, the WGPu could be isotopically degraded. If equal amounts of each Pu grade were blended, the resulting concentration of Pu-239 would be less than 80%, which would effectively give the Pu greater predetonation problems and almost eliminate its potential reuse in Russian arsenals.

While blending would be ideal for preventing reassembly into warheads its use is unlikely due to the lack of reciprocity. As mentioned above the US does not have the required RGPu and is highly unlikely to obtain foreign RGPu for this purpose.

3. Vitrify

The Pu (blended or not) could be vitrified using existing technologies and slightly modified glass frits. It is the author's opinion that the initial glass should be free from any radioactive diluents but include reactivity control elements. This would allow future recovery and fuel use of the Pu if it indeed becomes valuable.

The vitrification product would provide considerable proliferation resistance assuming it is not easily reversed. The unseparated critical mass of the heavy metal constituents could be made excessively large (as shown in our calculations). The size of the contained logs should be designed large enough to prevent the material from 'walking off' but not so large as to make governmental recovery unduly difficult.

The inclusion of an appropriate reactivity control element(s) would allow for safe permanent disposal should the economic viability of Pu remain unrealized. At that point in time a decision could be made whether or not to include HLW in the glass. Europium has been demonstrated as a candidate element that could potentially control the WGPu reactivity without jeopardizing its environmental durability (even if HLW were included). However, less expensive alternatives to Eu could be pursued.⁷⁷

⁷⁷The 1991 price of Eu was about \$7500/kg. Source: CRC Handbook of Chemistry and Physics 71st Ed. 1990-91 p.4-122

5. Provide Fuel Compensation

If the above mentioned arrangements are acceptable to both countries, that would be the end of the story. However, concerns may still exist over the ability to economically recover and burn the Pu. Currently Pu is of little or no economic value but the Russians are betting that the future value of the Pu may be significant. This fear is more acute in Russia as the US has no intention of creating a MOX based nuclear industry. For this reason Russia is hesitant to place the potential fuel in any form that would require additional costs to fabricate into fuel.

There may be a way around this problem. The US might be able to negotiate an agreement with its allies who reprocess. In the agreement the US would purchase an amount of RGPu equivalent to the fuel value (fissile content) of any WGPu that Russia would vitrify (under appropriate verification procedures).

In this scenario HLW would necessarily be added to the frit to ensure that Russia did not in the future attempt to recover the Pu for reuse in their arsenals. Reciprocity would probably require that the US modify its glass to include HLW also.

The purchased RGPu would come from the current or projected surpluses. While this material is obviously spoken for, it seems that sufficient incitement could be given to the owners to sell. As contracts exist for future reprocessing (and increased storage) it is unlikely that they would run out.

The Russian infrastructure for Pu burning is still small and delivery of the RGPu could be immediate (and stored with Russia's own RGPu) or set for a future date. If 75 tonnes of WGPu were vitrified approximately 100 tonnes of RGPu would be required to provide the equivalent fissile content. This represents half of the current stored inventory, so the required material does exist.

An alternative solution would not necessarily require the transport of any Pu. The US could purchase an option to buy RGPu from its European owners for a fixed price at a future date. It seems that the price would be fairly low given the Pu has no value currently. The delivery time could be determined through negotiations with Russia and could correspond to a period when they feel their energy program will be heavily dependent on Pu fuel. If Pu becomes a valuable commodity of the future, they can cash in their option. If not, they have lost nothing.

Either agreement would allow for WGPu processing and guarantee Russia the fuel value of its Pu. Russia would then have no reason to delay any disposition treatment. The US may favor this option as it provides a rational way of determining an appropriate price for the Pu.

This action should not be viewed as the creation of a Pu market. It would be a one-time transaction to aid dismantling and disposition efforts. The transport of RGPu is not unheard of and has been transported from Europe to Japan.

In summary, the US should open Pantex or another WGPu storage facility to 24 hour IAEA and Russian supervision. The Russians should take equivalent steps. Vitrification into large 'clean' logs could be performed with melters brought on-site or in other acceptable facilities. The WGPu should be vitrified with reactivity control elements that do not effect its environmental durability. If Russia objects to this treatment of its Pu, replacement RGPu should be purchased from European reprocessors to compensate for Russia's lost fuel. This presumably would allow processing to proceed. Radioactive waste would then be added to both the US and Russia's glasses. All of this activity would most likely be funded by the US or international contributors.

5.4 Strategy Evaluation

The proposed strategy offers a realistic way for vitrification technology to be used in WGPu disposition. It provides an effective proliferation deterrent

while addressing the concerns of the primary parties as well as the international community. If HLW is eventually included in the glass, the potential for reuse in either country's arsenal would be diminished as large, shielded and remotely operated facilities would be required.⁷⁸

If not, retrieval by either government would be rather straightforward. This fact could actually speed disposition as it may calm Russian fears of throwing away a valuable material. While this would also lessen the obstacles to retrieval for military use, such activity would be visible due to the oversight of the storage facility by IAEA inspectors.

Vitrification perhaps should not be viewed as a way of preventing national reuse. Even if HLW were included in the glass either country could make the costly effort to retrieve the Pu. A radiation barrier, while a nuisance, would not eliminate retrieval by a nation-state. Only through isotopic dilution would either government be deterred as the Pu would not meet their very high weapon standards.

If Russia or the US truly wanted the material there would be little to stop them. However, the separation would assumedly be extremely difficult for a non-government organization, especially if chemically similar elements such as Eu were used for reactivity control. This must be confirmed experimentally though.

The author believes this to be a realistic approach as he believes there is little chance of the WGPu ever leaving the boundaries or control of either country. If this is the case, no amount of diluents will guarantee non-reuse. Therefore, a policy of promoting actions that would virtually eliminate terrorist diversion while discouraging reuse seems rational.

A vitrification scenario as described provides alternatives at several key decision points. These options provide a robust menu of options for decision makers including a path of least resistance (vitrify with reactivity control in

⁷⁸McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p.7

readily processable logs) thereby allowing swift implementation. This is desirable as the plutonium pits in their present form provide no material barrier to proliferation. If even this option was rejected, it would seem vitrification has little chance of being used.

If collocation could be achieved (even domestically) it would provide a starting point for a program of increased security for RGPu. Reactor-grade Pu's low critical mass warrant our security attention. While current safeguarding efforts appear sufficient, the growth of separated RGPu surpluses cause international concerns to grow. Collocation would be a first step in recognizing their similarities.

Collocation would also keep open the isotopic blending option should Russia allow it. If, in the future, the world turns away from Pu fuel, concessions may be made to use the surplus RGPu as part of the global demilitarization effort. The presented strategy would allow future processing of the glass to include sufficient RGPu to prevent reuse in either countries arsenal.

The cost of such a program would be significant. Similar vitrification efforts are estimated by the Westinghouse Savannah River Company to cost \$0.5-1 billion in this country.⁷⁹ This estimate is the incremental cost of adding Pu vitrification at completed or nearly completed facilities. If construction of new facilities were required, this estimate would assumedly be higher.

It is clear that no processing will be cheap. Estimates for reactor burning are around \$10 billion. Some reactor costs may be recouped through revenues from reactor operations. (As a caveat, the costs of vitrification can also be offset by constructing reactors and selling the energy generated.) Compared to the price of a single stealth bomber (~\$1 billion) the opportunity to reduce the arsenals of the two major superpowers by more than one-half appears to be a good security investment.

⁷⁹McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p.24

In sum, there is a great deal of uncertainty surrounding WGPu disposition. Vitrification give us a relatively low cost way to increase the material security while buying time. Whether we process it for additional proliferation protection, dispose of it as is, or retrieve it for use, vitrification allows us to set a course of action today that will reduce the associated risks of diversion in the near term. To construe an argument typically used against vitrification, (WGPu was produced at too great a cost to throw away) WGPu was produced at too great a cost to waste on generating expensive kwhs.

5.5 Future Work

Clearly there is much more work to be done in examining the technical and political feasibility of vitrifying WGPu. The computational and experimental research described in this thesis examined a small range of options to prove the potential of the concept. Additional work must be done to confirm these findings as well as optimize the glass composition for a number of criteria. Additional comments on the political feasibility of the proposed strategy are also needed and could be gained through interviews with individuals close to the disarmament process.

5.5.1 Computational Testing

A continued effort could be made to provide more appropriate information on the weapon usefulness of different WGPu mixtures. Compressed critical mass and neutron production data are useful but do not wholly represent the explosive desirability of a material. An analysis that included the effect of different tampers, compression ratios, the dynamics of the explosion, etc..., would be helpful.

A wider variety of neutron absorbers and elements that spontaneously fission should be identified. A close examination of the fast fission spectrum for WGPu as well as the capture and neutron production cross sections for diluents in that energy region would aid the selection.

5.5.2 Experimental Testing

The leach tests performed should be duplicated in order to confirm the findings presented here. As a part of that effort the specific leaching mechanisms for Eu as well as the other elements should be identified.

A variety of potential elements for reactivity control should be selected and tested for impact on environmental durability. An entirely new glass composition specifically designed for WGPu disposition could also be designed.

The solubility of Pu in glass must also be investigated further. The one experiment performed here implies that the solubility limit lies below 10%. Continued testing would provide a more accurate limit. The specific mechanisms for devitrification under various environmental conditions should also be determined.

As noted several times, the reversibility of the vitrification effort will be of paramount importance. If sufficiently pure WGPu can be easily retrieved using laboratory size equipment, the vitrification option will be less appealing. A detailed study of potential separation processes is needed. This information should also impact the selection of potential diluents. If retrieval is easier than separating U from Pu and HLW is not included in the glass, storage as MOX fuel would be as desirable.

5.5.3 Optimization

Perhaps the most daunting task will be to design a glass frit that optimizes all of the above mentioned factors. Data must be generated on the production possibilities of the glass. This means that different combinations of elements must be tested to map out their ability to beneficially impact parameters important in WGPu disposition. These criteria should include: compressed critical mass; neutron production rate; reactivity control; elemental leach rate; durability; retrievability and costs. Additional criteria should also be sought.

An important result of this analysis will be the identification of trade-offs between added security and reversibility (if no HLW is included in the glass). This is a fine line as it may be desirable to place the WGPu in a form unretrievable for non-governmental bodies but less burdensome for either

government. The results could effect whether the vitrified WGPu was stored as a glass frit or in large logs.

5.5.4 Alternative Storage Forms

The scope of this thesis was limited to vitrification. There exists other physical storage forms which may have desirable characteristics. Fabricated monazite which contains numerous rare earths is one example. A scoping study of these forms would be useful. This may provide insight into safer ways of storing RGPu also.

5.5.5 Policy Analysis

A more definitive strategy could be formulated with a more complete understanding of the political workings of the entire disarmament negotiations. This would be quite difficult if not impossible to obtain first hand, however, interviews with decision makers in both governments would be invaluable.

While some understanding of the Clinton administration's views are obtainable in white house press releases and other literature, equivalent views from Russian decision makers are scarce. The identify of those decision makers and confirmation of their commitment to burning the WGPu would be of paramount importance.

The feasibility of collocation and international oversight must also be investigated further. It is believed that IAEA supervision has potential due to the US government's apparent support but the potential for collocation seems dim. The views of European reprocessors and utilities concerning collocation as well as the purchase of surplus RGPu is also needed.

Chapter 6

Bibliography

1. Barnaby, F., Plutonium and Security, St. Martin's Press, New York, 1992
2. Benedict, M., et al, Nuclear Chemical Engineering, McGraw-Hill, Inc., New York, 1981
3. Berkhout, F., et al., *Disposition of Separated Plutonium*, Science and Global Security, Vol. 3, 1992
4. Bunker, Bruce C., "Waste Glass Leaching: Chemistry and Kinetics," Scientific Basis for Nuclear Waste Management X, Vol. 84, 1987 pp.493-507
5. Cameron, K., *Taking Apart the Bomb*, Popular Science, April 1993 p.64-69,102-103
6. Deni, J., *Chronology of U.S.-Soviet-CIS Nuclear Relations*, Arms Control Today, June 1994 p.33
7. DeVolpi, A., *Fissile Materials and Nuclear Weapons Proliferation*, Ann. Rev. Nucl. Part. Sci., V36, 1986
8. DeVolpi, A., *Denaturing Fissile Materials*, Progress in Nuclear Energy 10:161, 1982
9. Feiveson, H., and von Hippel, F., *Dismantling the Doomsday Machine*, Technology Review, May/June 1992 pp.62-69
10. Garwin, R. L., *Safe Secure Disposal of 50,000 Excess Nuclear Weapons*, Paper presented at the NATO Special Workshop on Global Stability Through Disarmament, August 14, 1993
11. Hu, H., et al, Plutonium: Deadly Gold of the Nuclear Age, International Physicians Press, Cambridge, Massachusetts 1992
12. Isaacs, J., *A Bouquet for Bush*, The Bulletin of the Atomic Scientists, March 1993 p.10,46-7
13. Keeny, S. M., *Will Political Realism Prevail in Kiev?*, Arms Control Today, January/February 1994

14. Leskov, S., *Lies and Incompetence*, Bulletin of the Atomic Scientists, Vol. 49 num. 5, June 1993 pp.13,55
15. Lockwood, D., *GAO Study Outlines Obstacles To Soviet Warhead Dismantlement*, Arms Control Today, April 1993
16. Lockwood, D., *Ukraine Begins Transfer Of Strategic Nuclear Warheads*, Arms Control Today, April 1994 p.20
17. Lutze, W., and R. C. Ewing, ed., Radioactive Waste Forms for the Future, Elsevier Science Publishing Company, Inc., New York, 1988 pp.559-60
18. Mark, J., *Reactor-Grade Plutonium's Explosive Properties*, Nuclear Control Institute, August 1990 p.4
19. McKibben, J. M., et al, *Vitrification of Excess Plutonium*, Predecisional Draft, Plutonium Vitrification Task Group, Westinghouse Savannah River Company, WSRC-RP-93-755, May 1993 p.20
20. National Academy of Sciences, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington D.C., 1994
21. Rochlin, G. I., Plutonium, Power, and Politics, University of California Press, Ltd., London, England, 1979
22. Scheinman, L., *Managing the Coming Glut Of Nuclear Weapon Materials*, Arms Control Today, March 1992 p.9
23. Scholes, S. R., Modern Glass Practice, CBI Publishing Company, Inc., Boston 1975 pp. 33-34
24. Simonson, S., *Don't Be Fuelish with Warheads*, Insight, January 24, 1994 pp.25-27
25. Taubes, G., *No Easy Way to Shackle The Nuclear Demon*, Science, Vol. 263, February 4, 1994 p.631
26. U.S. Congress, Office of Tech. Assessment, Dismantling the Bomb and Managing the Nuclear Materials, U.S. Government Printing Office, September 1993
27. U.S. Department of Energy Plutonium Disposition Study, Technical Review Committee Report, Volumes 1 & 2, July 2 1993
28. von Hippel, et. al., *Eliminating Nuclear Warheads*, Scientific American, August 1993

29. White House Press Release, September 27, 1993
30. Woolf, A., *The START and START II Arms Control Treaties: Background and Issues*, Congressional Research Service Report for Congress, 93-617 F, June 30, 1993

Appendix I

Sample Input Files for MCNP

Figures A-1 and A-2 display sample MCNP input files for criticality and flux calculations respectively. The criticality sample file is for a mixture of equal weights of rare earths and WGPu. The flux calculation is for pure RGPu. The input files are shown in their entirety. The output files are far too large to be displayed easily so only the most important portions are shown.

Sample input file for a criticality calculation on MCNP

```
krarew
1 1 -17.08 -1
2 0 +1
1 so 32.50
```

```
mode n
imp:n 0
kcode 1000 1.0 10 40
m1 52149 -0.20
53000 -0.20
54000 -0.20
30232 -0.20
94239 -0.19
94240 -0.006
94241 -0.003
94242 -0.001
```

Print

This metallic mixture includes equal weight percents of Eu, Gd, Sr, Th and WGP1.

The compressed critical mass = 17.08 * 4/3*P1*32.50^3
 = 245589.8 g or 2456 kg

Sample output file for the above input file

```
estimator cycle 40 ave cf 30 cycles combination simple average combined average corr
k(collison) 0.978687 1.002672 0.0027 k(co./abs) 1.004205 0.0022 0.050
k(absorption) 0.991258 1.006925 0.0035 k(abs/tk ln) 1.005456 0.0024 0.076
k(trk length) 0.983477 1.004408 0.0030 k(tk ln/csl) 1.002243 0.0027 0.92E
rem life(col) 2.7728E+00 2.7677E+00 0.0069 k(co./abs/tk ln) 1.003765 0.0024
rem life(abs) 2.7765E+00 2.7765E+00 0.0065 life(col/abs) 2.7722E+00 0.0067 0.991
source points generated 982
```

Figure A-1 - MCNP Criticality Input File

Sample input file for a flux calculation in MCNP

```
refer
  1  1  19.5  1      imp:n=1
  2  0          +1    imp:n=0

  1  so  4.0

mode  n
sdef  pos 0 0 0   erg d1 wgt=1  tmc=0
sp1   -3  0.799  4.903
f2:n  1
nps   10000
m1    94239  0.62
      94240 -0.21
      94241 -0.13
      94242 -0.04

print
```

Sample output file from the above input file

```
ltally  2          nps =    10000
        tally type 2  particle flux averaged over a surface.      units  1/cm**2
        tally for  neutrons

        AREA
        surface:

                2.89529E+03

surface  1

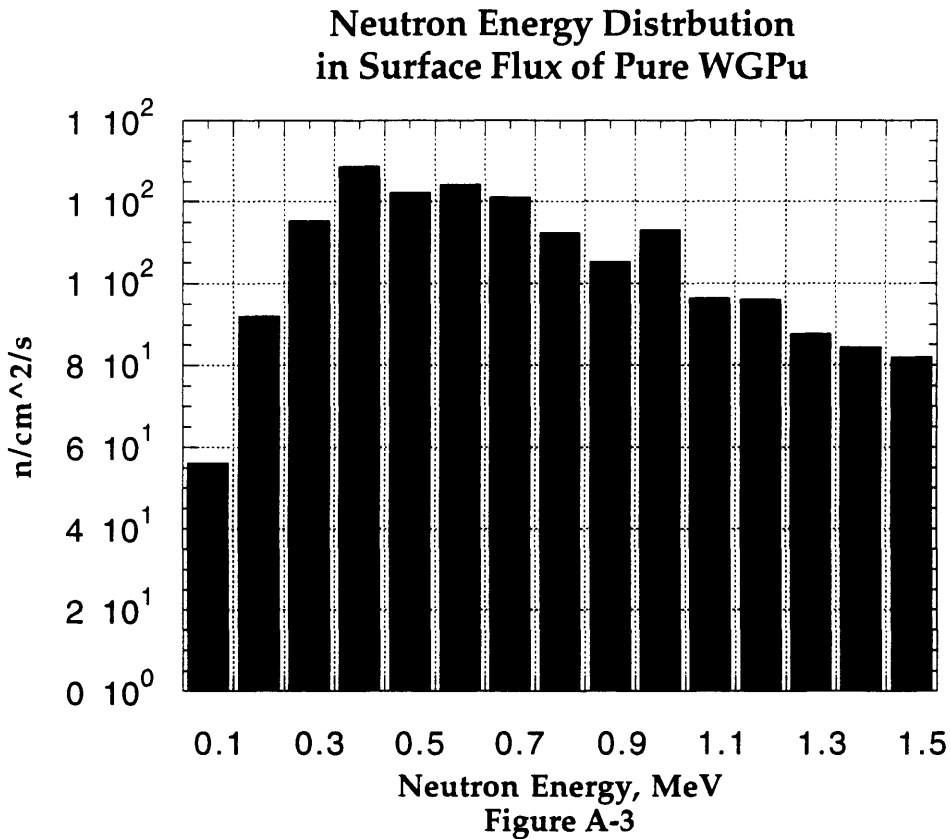
                4.75034E-02  0.0327
```

Figure A-2 - MCNP Flux Input File

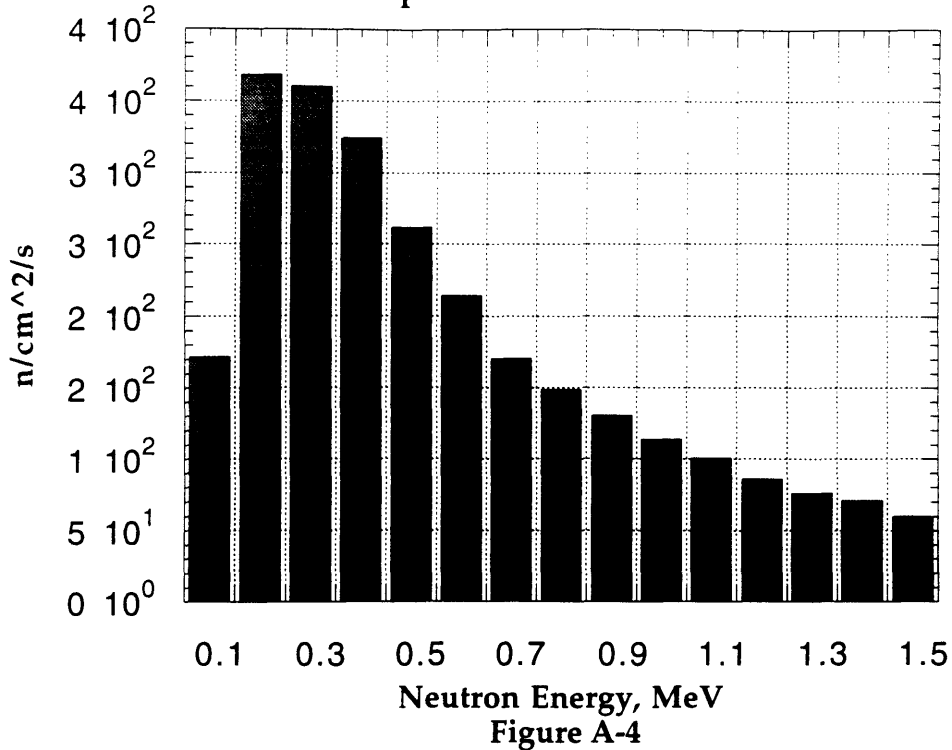
Appendix II

Neutron Energy Spectrum

Figures A-3 and A-4 show the neutron energy distributions for a compressed critical mass of pure WGPu and a mixture of equal fractions of WGPu, Th and the rare earths Eu, Gd, and Sm. Flux calculations were performed on a single compressed critical mass of each material (in uncompressed form) to generate this distribution. The number of neutrons exiting the surface of the sphere (per cm² per second) as well as their energy was recorded. The energy value on the x-axis represents the top limit for the energy bin. For example, the 0.6 MeV value in Figure A-3 represents the flux of neutrons leaving the sphere with an energy between 0.5 MeV and 0.6 MeV.



Neutron Energy Distribution in Surface Flux of WGPu Mixed With Equal Amounts of Rare Earths



As evidenced by the figures, the addition of the rare earths softens the neutron spectrum. The largest number of neutrons exiting the rare earth mixture have energies between 0.1 and 0.2 MeV. The larger mass of material in the rare earth mixture allows for more neutron collisions, thereby decreasing the average energy of the neutrons exiting the sphere. By examining the ENDF/B cross sections (available from Brookhaven National Laboratory) we see that the inelastic scattering cross section of the rare earths as well as Th is around 2 barns in the 0.6 MeV region. Scattering with these elements serves to decrease the neutron energy.

The ENDF/B data also shows the increased capture by Eu as the neutron energy is reduced. This serves to increase the critical mass of the WGPu mixture. The capture cross section for Eu is around 0.2 barns at 0.6 MeV and increases to around 0.4 barns at 0.4 MeV. Europium's neutronic properties in this energy range (combined with its low density) make it superior to the other diluents examined in increasing critical mass.

Gadolinium's capture cross section is around 0.1 barns in this region (0.4-0.6 MeV) and does not increase significantly until the neutron energy is below 0.3 MeV. Thorium's capture cross section is approximately the same as Eu at 0.6 MeV but remains constant until neutron energies fall below 0.2 MeV. The capture cross section distribution for samarium-149 is similar to Eu's in this energy range. (Sm-149 was used in the analyses as its cross sections were the only ones available.) However, samarium's higher density make it less effective at increasing the mixture's critical mass.

Appendix III

Surface Flux and Neutron Production Rate

This appendix contains graphical representations of the neutron flux and neutron source data mentioned in Section 4.3. Figure A-5 shows how the flux from one compressed critical mass of MOX (in uncompressed form) is nearly twice that of pure WGPu. This is because there is a much greater amount of WGPu present (more than 3500 kg) in one compressed critical mass of MOX fuel.

The mixture of 80% rare earths (as mentioned in Chapter 4) produces a surface flux slightly higher than that of pure WGPu. The increased amount of WGPu present is almost completely offset by the added absorption and decreased density of the mixture. A compressed critical mass of the IFR fuel contains less WGPu than the rare earth mixture and has a surface flux lower than that of pure WGPu.

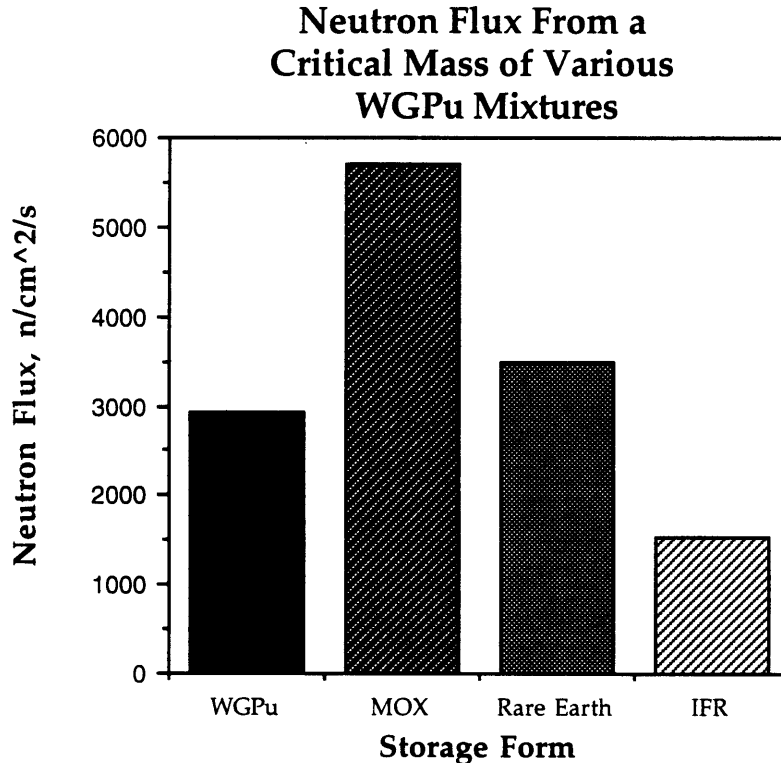
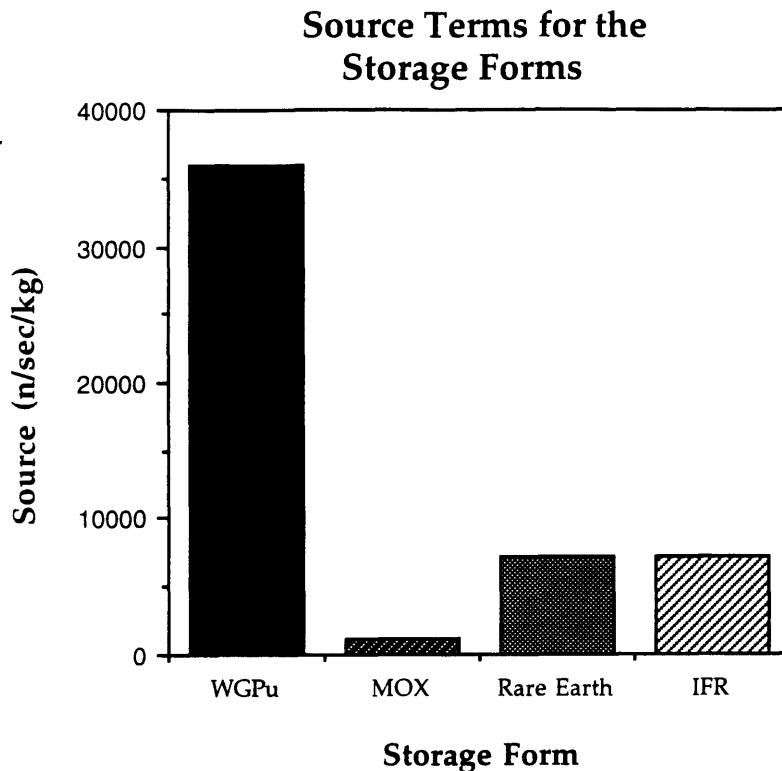


Figure A-5

Figure A-6 shows the neutrons generated per second per kilogram of material from the spontaneous fissioning of Pu and U in the mixtures. In some way this is a better measure of preinitiation potential. It is unlikely that a weaponeer would fashion a several ton bomb out of dilute plutonium. He or she would more likely refine the material, increasing the relative percent of Pu present. As this occurred the critical mass would decrease. The source term per kg would increase but would never exceed that of WGPu. Therefore it can be concluded that chemical dilution (with non-neutron producing elements) is ineffective at increasing the probability of preinitiation.



Storage Form
Figure A-6

Appendix IV

MCNP Data Summary

Table A-1 in this appendix provides a summary of the key compressed critical mass and neutron flux data presented in the body of this thesis as well as information on the surface gamma flux and deposited energy for the forms studied.

The gamma flux and energy deposited data was initially retrieved from MCNP for future comparison with other, radioactive diluents. At the initiation of our investigation, we investigated this data to determine if these characteristics would be a deterrent to potential proliferators. The magnitude of both parameters was relatively low providing virtually no barrier to proliferation. As diluents that would increase these values were not included in our investigation, these results were not presented with the other technical analyses. They are included here only for informative purposes.

		Compressed Critical Mass (kg)	Neutron Flux (n/cm ² /s)	Gamma Flux (gam/cm ² /s)	Deposited Energy (MeV/g/s)
Pu Metal	RG:	3.4	1.14 x 10 ⁵	3.85 x 10 ⁴	2.00 x 10 ⁵
	WG:	2.7	3.33 x 10 ⁴	1.17 x 10 ⁴	6.10 x 10 ⁴
PuO ₂	RG:		2.82 x 10 ⁴	9.02 x 10 ³	5.20 x 10 ⁴
	WG:	7.1	8.99 x 10 ³	3.05 x 10 ³	1.77 x 10 ⁴
MOX Fuel	RG:		1.25 x 10 ³	82	5.14 x 10 ²
	WG:	9.6 x 10 ⁴	3.48 x 10 ²	24	1.49 x 10 ²
Rare Earth Glass	RG:		2.42 x 10 ³	490	1.42 x 10 ³
	WG:	2.5 x 10 ³	6.01 x 10 ²	106	3.92 x 10 ²
IFR Fuel	RG:		6.16 x 10 ³	729	4.43 x 10 ³
	WG:	83.5	9.28 x 10 ²	111	7.12 x 10 ²

Table A-1 - MCNP Results Summary

For the flux and deposited energy calculations, a constant mass of 9.033 kg of Pu was used to obtain the tabulated values. This should be contrasted with

the values discussed in Section 4.3. Those flux values were for a single compressed critical mass, therefore the amount of Pu present was allowed to vary.

Appendix V

Glass Compositions

In this appendix the exact compositions of the glasses tested are listed. Two batches of each composition were needed to obtain the required number of samples for the numerous leach tests. Experimental error resulted in slightly different compositions and all variations are included here.

Tables A-2 and A-3 contain the weight percents of the oxides contained in all the fabricated glasses. Tables A-4 and A-5 show the compositions of the base frits SRL-165 and ARM-1 respectively. The SRL-165 composition was obtained from Radioactive Waste Forms for the Future, Elsevier Science Publishing Company, Inc., New York, 1988 (p.23). The ARM-1 composition was obtained from Pacific Northwest Laboratory.

	3% RE- ARM(#1)	3% RE- ARM(#2)	6% RE- ARM(#1)	6% RE- ARM(#2)	6% RE- SRL(#1)	6% RE- SRL(#2)
% Eu ₂ O ₃	0.998	0.998	2.01	2.05	1.99	1.99
% Gd ₂ O ₃	1.02	1.01	2.00	2.08	1.99	2.01
% ThO ₂	0.00	0.0	0.00	0	0.00	0
% Sm ₂ O ₃	0.998	0.990	2.00	2.07	1.99	2.01
% ARM	97.0	97.0	94.0	93.79	0.00	0
% SRL	0.00	0	0.00	0	94.0	93.99

Table A-2 - Fabricated Non-Th Glass Compositions

	6% RE 2% Th- ARM(#1)	6% RE 2% Th- ARM(#2)	6% RE 2% Th- SRL(#1)	6% RE 2% Th- SRL(#2)
% Eu ₂ O ₃	1.92	1.98	1.92	1.99
% Gd ₂ O ₃	2.01	1.98	2.00	1.99
% ThO ₂	1.99	1.98	2.00	2.01
% Sm ₂ O ₃	2.00	1.98	2.00	1.99
% ARM	92.1	92.1	0.00	0.00
% SRL	0.00	0.00	92.1	92.0

Table A-3 - Fabricated Th Glass Compositions

% SiO ₂	68.0
% B ₂ O ₃	10.0
% Li ₂ O	7.0
% Na ₂ O	13.0
% MgO	1.0
% ZrO ₂	1.0

Table A-4 - Pure SRL-165 Composition

% SiO ₂	46.50
% Al ₂ O ₃	5.59
% CaO	2.23
% TiO ₂	3.21
% Na ₂ O	9.67
% B ₂ O ₃	11.3
% Nd ₂ O ₃	5.96
% Li ₂ O	5.08
% ZnO	1.46
% P ₂ O ₅	0.65
% ZrO ₂	1.80
% BaO	0.66
% MoO ₃	1.67
% CeO ₂	1.51
% Cs ₂ O	1.16
% SrO	0.45

Table A-5 - Pure ARM-1 Composition

Appendix VI

Summarized Leach Test Results

The various leachates obtained during the glass testing periods were run through the ICP spectrometer. This machine gives the concentration (in ppm) of selected elements found in an aqueous solution. Knowing the volume of the leachate, the surface area of the glass and assuming a density of one for the solution it becomes possible to present the concentration data in units of grams/cm². A blank was included for all tests and its concentration was subtracted to determine the elemental release from the glass only. The following tables provide a summary of all the data (in grams/cm²) obtained during testing. Tables A-6 through A-12 contain the 90°C leach test results and tables A-13 through A-19 contain the room temperature results.

The ICP spectrometer fixes concentrations through a curve fitting process. This allows for negative concentrations to be obtained. As this is a non-physical result, zeros were inserted in the following tables whenever this occurred. However, outputted results that were actually below detection limits are included.

90°C Leach Test Results

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
3	1.483e-05	8.740e-05	0	0	2.866e-05	2.542e-07
7	2.536e-05	1.468e-04	5.185e-08	0	4.823e-05	0
7	2.661e-05	1.502e-04	5.263e-08	0	5.238e-05	3.237e-08
14	3.009e-05	1.777e-04	1.114e-07	2.987e-09	5.997e-05	9.880e-08
28	3.701e-05	2.016e-04	0	0	7.319e-05	0
28	3.926e-05	2.156e-04	9.102e-09	1.012e-09	7.608e-05	1.922e-07
28	3.876e-05	2.141e-04	0	7.649e-13	7.740e-05	0

Table A-6 - ARM-1 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	2.276e-05	1.281e-04	6.801e-08	0	4.276e-05	0
14	2.890e-05	1.648e-04	4.456e-08	0	5.677e-05	0
28	3.713e-05	2.006e-04	2.170e-08	0	7.189e-05	0
28	3.749e-05	2.054e-04	0	0	7.401e-05	0
28	3.491e-05	1.936e-04	4.548e-08	1.011e-09	6.839e-05	0

Table A-7 - 3% Rare Earths-ARM-1 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
3	1.715e-05	9.446e-05	0	0	3.326e-05	0
7	2.665e-05	1.436e-04	0	1.010e-09	5.272e-05	8.401e-08
7	2.583e-05	1.383e-04	0	0	4.970e-05	0
14	2.832e-05	1.534e-04	0	0	5.720e-05	0
28	3.733e-05	1.931e-04	4.241e-08	1.024e-09	7.268e-05	0
28	3.746e-05	1.956e-04	6.665e-08	2.426e-08	7.346e-05	2.437e-07
28	3.991e-05	2.068e-04	3.836e-08	1.029e-09	7.713e-05	0

Table A-8 - 6% Rare Earths-ARM-1 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	2.237e-05	1.163e-04	0	0	4.323e-05	0
14	4.436e-07	6.163e-07	0	3.336e-08	0	0
28	3.524e-05	1.779e-04	0	0	6.958e-05	0
28	3.608e-05	1.810e-04	0	0	7.138e-05	0
28	3.384e-05	1.702e-04	0	0	6.633e-05	0

Table A-9 - 6% Rare Earths-2% ThO₂-ARM-1 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	2.261e-05	2.494e-04	2.645e-08	0	7.030e-05	5.215e-08
14	5.397e-05	6.078e-04	1.590e-07	0	1.716e-04	1.882e-07
28	1.398e-04	1.531e-03	7.189e-07	2.341e-12	4.456e-04	0
28	1.664e-04	1.846e-03	5.007e-07	0	5.359e-04	0

Table A-10 - SRL-165 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	1.782e-05	1.897e-04	0	0	5.547e-05	0
14	3.274e-05	3.411e-04	5.670e-08	7.087e-09	1.013e-04	1.883e-07
28	9.454e-05	9.866e-04	5.072e-08	3.153e-06	2.952e-04	3.293e-06
28	7.830e-05	8.119e-04	5.010e-09	1.749e-06	2.448e-04	1.678e-06
28	6.935e-05	7.315e-04	4.549e-08	1.517e-06	2.172e-04	1.602e-06

Table A-11 - 6% Rare Earths-SRL-165 (90°C)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	1.618e-05	1.731e-04	0	0	5.044e-05	0
14	8.192e-07	6.515e-06	0	2.489e-07	1.807e-06	0
28	5.930e-05	6.180e-04	0	0	1.882e-04	0
28	5.894e-05	6.104e-04	0	0	1.881e-04	0
28	5.865e-05	5.963e-04	0	0	1.832e-04	0

Table A-12 - 6% Rare Earths-2% ThO₂-SRL-165 (90°C)

Room Temperature Leach Test Results

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	6.718e-07	7.468e-07	4.458e-08	0	1.193e-06	0
14	1.187e-06	1.564e-06	0	3.959e-09	8.681e-07	0
28	4.640e-08	5.616e-07	0	1.822e-08	8.588e-07	2.636e-07
28	7.228e-07	1.575e-06	0	3.044e-09	1.840e-06	2.674e-07
28	5.350e-07	8.383e-07	9.302e-08	1.260e-12	1.304e-06	0

Table A-13 - ARM-1 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	5.581e-07	1.234e-06	3.642e-08	8.091e-09	1.364e-06	0
14	8.263e-07	1.523e-06	1.315e-08	1.920e-08	6.103e-07	3.027e-07
28	0	2.560e-07	3.828e-08	1.008e-08	6.593e-07	8.857e-07
28	6.982e-08	7.382e-07	0	1.717e-08	8.152e-07	6.185e-07
28	4.202e-07	9.843e-07	0	1.819e-08	1.390e-06	3.992e-08

Table A-14 - 3% Rare Earths-ARM-1 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	5.053e-07	1.141e-06	2.052e-09	2.429e-08	1.077e-06	1.720e-08
14	7.741e-07	1.861e-06	7.274e-08	4.346e-08	8.121e-07	5.737e-07
28	7.058e-07	1.665e-06	0	1.050e-07	1.402e-06	2.535e-07
28	5.916e-07	1.590e-06	0	9.100e-08	1.514e-06	5.388e-09
28	6.167e-07	1.791e-06	0	9.297e-08	1.603e-06	2.844e-07

Table A-15 - 6% Rare Earths-ARM-1 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	1.979e-07	3.700e-07	0	2.738e-08	5.138e-07	2.578e-07
14	4.436e-07	6.163e-07	0	3.336e-08	0	0
28	4.967e-07	6.868e-07	1.003e-09	7.788e-08	8.815e-07	0
28	4.206e-07	9.837e-07	8.089e-08	1.173e-07	1.022e-06	1.972e-07
28	2.985e-07	7.632e-07	4.056e-09	7.390e-08	6.401e-07	9.017e-08

Table A-16 - 6% Rare Earths-2% ThO₂-ARM-1 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	8.385e-07	1.769e-06	5.874E-08	8.978E-12	3.256e-06	0
14	1.185e-06	1.322e-06	0	0	3.397e-06	0
28	1.074e-06	0	0	0	4.259e-06	0
28	1.573e-06	3.846e-07	0	0	5.157e-06	7.327E-08

Table A-17 - SRL-165 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	7.083e-07	6.146e-06	4.255E-08	1.257e-07	3.182e-06	0
14	1.288e-06	1.073e-05	4.049E-08	2.965e-07	3.702e-06	8.151E-07
28	1.630e-06	1.534e-05	3.709E-08	3.491e-07	5.874e-06	2.497E-07
28	1.696e-06	1.614e-05	2.338E-08	2.837e-07	6.146e-06	2.280E-07
28	1.561e-06	1.458e-05	2.025E-08	2.360e-07	5.547e-06	3.647E-07

Table A-18 - 6% Rare Earths-SRL-165 (Room T.)

Leach Time, Days	Boron, g/cm ²	Silicon, g/cm ²	Thorium, g/cm ²	Europium, g/cm ²	Sodium, g/cm ²	Cesium, g/cm ²
0	0	0	0	0	0	0
7	4.657e-07	3.365e-06	3.746e-08	1.255e-07	2.077e-06	0
14	8.192e-07	6.515e-06	0	2.489e-07	1.807e-06	0
28	1.038e-06	6.270e-06	3.224e-08	3.069e-07	2.579e-06	0
28	9.816e-07	7.193e-06	0	2.768e-07	2.834e-06	3.611e-07
28	7.410e-07	5.029e-06	0	2.333e-07	2.059e-06	3.560e-07

Table A-19 - 6% Rare Earths-2% ThO₂-SRL-165 (Room T.)