NONPROLIFERATION ANALYSIS OF THE REDUCTION OF EXCESS PLUTONIUM AND HIGH-ENRICHED URANIUM*

Paul J. Persiani
Argonne National Laboratory
Argonne, IL 60439 USA

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Abstract

The purpose of this preliminary investigation is to explore alternatives and strategies aimed at the gradual reduction of the excess inventories of separated plutonium and high-enriched uranium (HEU) in the civilian nuclear power industry. The study attempts to establish a technical and economic basis to assist in the formation of alternative approaches consistent with nonproliferation and safeguards concerns. The analysis addresses several options in reducing the excess separated plutonium and HEU, and the consequences on nonproliferation and safeguards policy assessments resulting from the interacting synergistic effects between fuel cycle processes and isotopic signatures of nuclear materials.

Introduction

The strategic arms reduction treaty (START) and related bilateral agreements, and the increasing maturity of civil-use of plutonium recycle within the international commerce of the nuclear power industry, appear to have introduced additional challenges to the nuclear safeguards and nonproliferation research and development programs. The paper explores possible nonproliferation and safeguards alternatives and strategies and measurement techniques to address the gradual reduction of existing excess inventories of separated plutonium and high-enrichment uranium, in some stages of the international civilian nuclear power industry.

The analysis of the civilian nuclear fuel cycle is an important phase in implementing safeguards and nonproliferation alternatives and strategies in bringing the excess plutonium and HEU supply and demand into balance. The fuel cycle analysis identifies the synergistic interplay between fuel cycle processes and safeguards and nonproliferation concerns relating to the traceability of nuclear materials when assessed externally to the civilian power production cycle. A primary objective of the paper is to review, in part, an information/data base system of the fuel cycles necessary in the analysis and assessment of R & D programs. The acquisition of information/data and knowledge of the international commercial fuel cycle developments should include; plutonium and uranium inventory and flow balances, spent fuel policies, reprocessing operations and programs, mixed oxide activities, enrichment processes, fuel cycle service facility developments, and impacts on the economies of nuclear power generation, all of which would indicate safeguards development trends.

Transparency and Fuel Cycle Data

The international power industry has had an extensive history and experience in the strategy of a closed fuel cycle, spent fuel reprocessing, uranium and plutonium recycle, and waste management activities. The experience has been plutonium recycling by the use of U-Pu mixed-oxide (MOX) fueled assemblies in pressurized water reactors (PWRs).

The transparency aspects of nonproliferation commitments would be to utilize mutually agreed to information and data exchanges for validation of

declared information and for maintaining knowledge of nuclear material flows and inventory in the commercial power production cycles. The implementation of monitoring methods for fuel cycle data validation would depend on the cooperatively offered declarations and the access to data relating to the flows and inventories at key locations within the fuel cycle: UO_2 fuel and conversion processes, enrichment operations, fuel assembly fabrication plants, reactor power loading configurations, reprocessing plant, and spent fuel or separated plutonium storage facility operations including interim storage and geological repositories.

Reference annual mass flows and inventories for a representative 1400 Mwe pressurized water reactor (PWR) fuel cycle are presented in Figure 1 for three cases: the 100 percent UO_2 fuel loading once-through cycle and the 33 percent MOX loading configuration for a first and second plutonium recycle.

The mass flows and inventories include the uranium feed requirements and the SWU requirements for the enrichment process in the once-through and first and second plutonium recycle. The fuel management program adopted is the current commercial practice of reloading 1/3 of the core assemblies (about 60 to 65 assemblies). The once-through fuel cycle produces some 295 kg of fissile plutonium with the fissile plutonium being about 60 to 70 percent of the total plutonium element. Since the isotopic composition of the plutonium would synergistically impact on fuel cycle fabrication and storage operations and related safeguards and nonproliferation measurement techniques, the isotopics at each phase of the cycle are included in the flows and inventories of the power cycle. The relevant isotopic data are presented in tabular forms (see Tables I-II) corresponding to the letters A, B, C,....etc., as denoted in the flow charts. The spent fuel assemblies are usually stored at reactor sites or transferred to facilities for reprocessing and MOX fabrication. The majority of the once-through spent fuel isotopics (Ref. 1-3) are representative nominal concentrations for discharge burnup levels of about 33 GWd/T. These are the stores from which MOX fuels are being fabricated for plutonium recycle. To review the range of implementing safeguards nondestructive assay (NDA) and destructive analysis (DA) measurement techniques, the measured isotopic compositions for the US Zion-1 PWR (Table III) are included as reference data for burnup levels ranging from 32 to 64 GWd/T. The higher burnup levels are currently the goals to be achieved by electric power utilities to improve the fuel cycle economics. However, higher levels of the Pu-238 concentrations on the order of 3 to 4 percent of the plutonium element, can affect the front-end of the MOX fuel fabrication process and consequently safeguards measurement techniques. Referring to Table IV, the plutonium specific neutron yields are increased by factors of 2 or more just from the plutonium isotopic composition changes as a function of burnup. These increases in neutron yields will be maintained for the first and second plutonium recycles. Neutron NDA methods for material control and accountancy will be further burdened by the calibration requirements as a consequence of the changes and the uncertainties in the changes of the Pu-238 and Pu-242 isotopic concentration buildup. The Transuranium Institute (TUI) are investigating methods to determine the Pu-242 isotopic content in high burnup reactor fuels (Ref. 4).

UO_2 ASSEMBLY LOADING		
	INITIAL FEED (A)	DISCHARGE 33 GWd/T LWR STORES (B)
234 U	0.03	0.018
235 U	4.0	1.035
236 U	-	0.44
238 U	95.95	98.49
238 Pu	-	1.9
239 Pu	-	58.9
240 Pu	-	22.8
241 Pu	-	10.9
242 Pu	-	5.7

Table I. UO_2 Assembly Nominal Isotopic Concentrations

ANNUAL MASS FLOW AND INVENTORY FOR REFERENCE (1400 MWe) PWR FUEL CYCLE

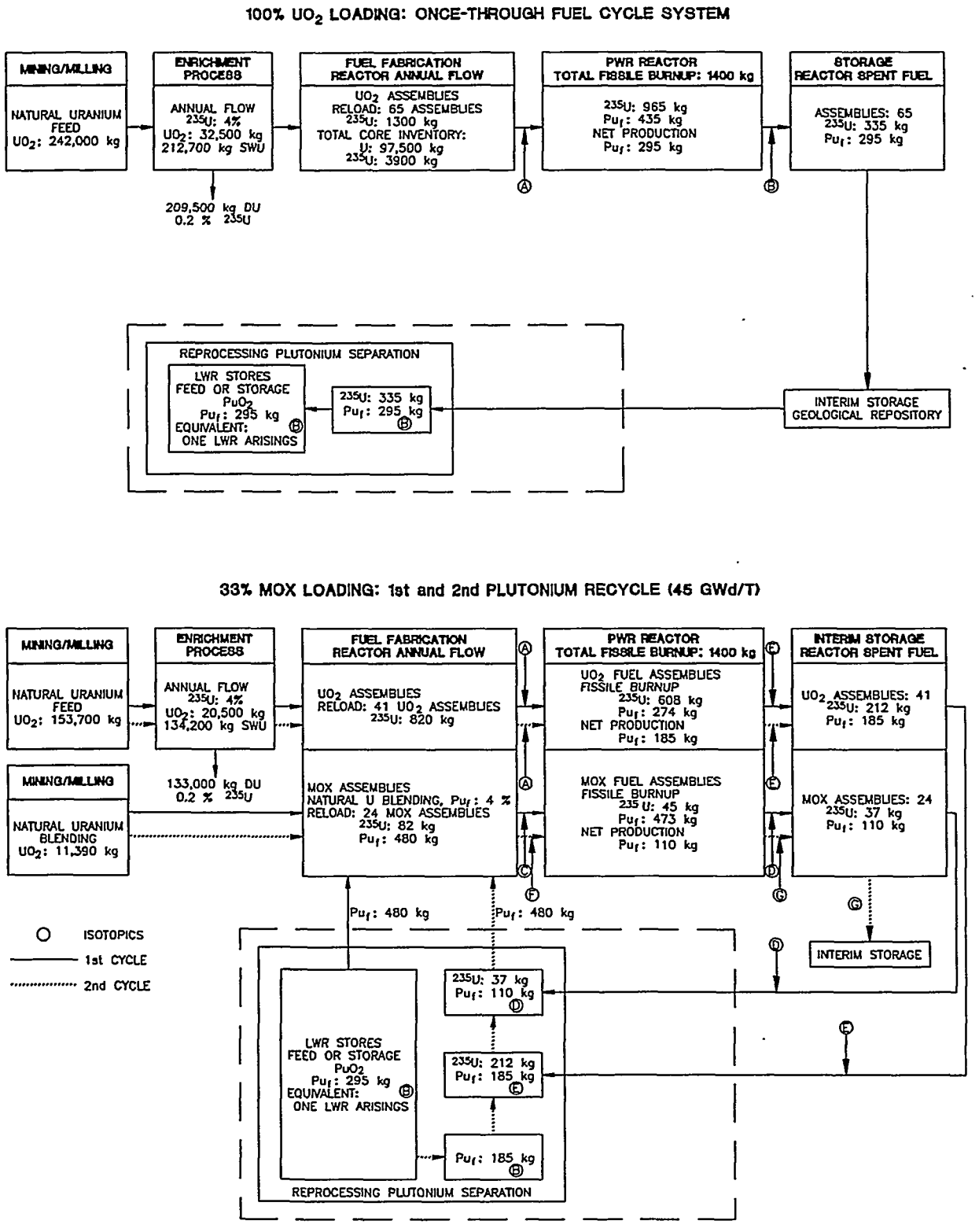

Figure 1. *Annual Mass Flow and Inventory for Reference (1400 Mwe) PWR Fuel Cycle*

1ST Pu RECYCLE					
UO_2 ASSEMBLY LOADING			MOX ASSEMBLY LOADING		
INITIAL FEED (A)		DISCHARGE 45 GWd/T (E)	INITIAL FEED (C)		DISCHARGE 45 GWd/T (D)
234 U	0.03	0.018	234 U	0.0055	0.011
235 U	4.0	0.67	235 U	0.72	0.32
236 U	-	0.57	236 U	-	0.08
238 U	95.97	98.74	238 U	95.275	99.55
238 Pu	-	3.1	238 Pu	1.9	2.8
239 Pu	-	55.8	239 Pu	58.9	39.4
240 Pu	-	23	240 Pu	22.8	32.3
241 Pu	-	11.7	241 Pu	10.9	13.8
242 Pu	-	6.4	242 Pu	5.7	11.6

Table II. MOX and UO_2 Assembly Nominal Isotopic Concentrations (w/o)

ZION-1 (PWR)[1]					
BURNUP MWD/T					
ISOTOPICS W/O	32000	36000	43000	51000	64000
238 Pu	1.635	2.042	2.694	3.620	4.823
239 Pu	57.12	54.12	49.91	46.52	42.71
240 Pu	25.88	26.64	27.88	28.11	26.79
241 Pu	9.67	10.16	10.77	11.42	12.72
242 Pu	5.69	7.03	8.78	10.33	12.96

1. ZION - 1 Measurements are for selected segments of fuel pins in reactor assemblies after 5 operating power cycles.

Table III. Table of Measured Isotopic Values

ZION-1 (PWR)						
Neutron Yield (Neuts/kg-sec)						
BURNUP MWD/T						
Isotopics	22000	32000	36000	43000	51000	64000
238 Pu	22.1	42.5	53.1	70.1	94.1	125.4
239 Pu	-	-	-	-	-	-
240 Pu	198.1	229.1	235.8	246.7	246.8	237.1
241 Pu	-	-	-	-	-	-
242 Pu	51.8	99.2	122.6	153.1	180.1	225.9
NEUTRON YIELD NEUTS/kg-sec	2.7×10^5	3.7×10^5	4.1×10^5	4.7×10^5	5.2×10^5	5.9×10^5

Table IV. Table of Neutron Yields of Pu Isotopic Concentrations

The partial MOX loading of 33 percent of the core for the 1st plutonium recycle are for the fuel burnup levels of 45 GWd/MTHM. The reload management schedule is the removal and replacement of 1/3 of the core (65 assemblies) with MOX fuel assemblies. The plutonium recycle results in a reduction of about 80,000 kg of natural uranium feed requirements and about 79,000 kg-SWU requirements in the enrichment process, a 30 percent reduction in both processes.

Incentives for Draw-down of Excess Separated Plutonium

Cost estimates are introduced to indicate the basis for the expected trend of deploying separated plutonium (from spent fuel arisings and from weapons dismantlement agreements) into the international civilian power cycle. The scoping of the front-end fuel cycle costs would have to include the comparison between these cost reductions with the increased costs of the MOX fabrication process. Preliminary estimates indicate that a break-even or slight increase in front-end fuel cycle costs between the two options could be expected depending on the near-term costing developments in the processes involved. Referring to Table V and Fig. 2, the front-end fuel cost differentials were found to range between -0.5 to +0.2 mills/kWh for the partial MOX loading case. The unit process cost estimates used were based on the current market pricing reported in Nuclear Fuel, Vol. 20, No. 12, June 5, 1995 and estimates in "The Economics of the Nuclear Fuel Cycle", Nuclear Energy Agency, NEA, 1992; UF6 value of $35/kg U; $70 to $85/kg-SWU in the enrichment process; UO_2 fuel fabrication cost of $200 to $275/kg U; and $800 to $1100/kg for the MOX fuel fabrication process. In the case of the draw-down of weapons-plutonium stores (Fig. 3.), the front-end cost differentials were found to range between -0.2 to +0.65 mills/kWh for the partial MOX loading. The cost differentials include an estimated cost of $300 to $400/kg of PUO_2 for the weapons-plutonium metal to oxide conversion process.

The method for the transfer of ownership, control, costs, and safeguarding of the weapons-plutonium stores in the MOX fabrication process and reactor core loading operation would have to be developed for the specific implementation of this option in the disposition of plutonium.

Draw-Down of Excess Plutonium Stores Front-End fuel Cycle Costs[1] Mills/kWh			
	Once-Through Cycle	Plutonium Recycle	
Enrichment $/kg-SWU	Fabrication Price		Cost Differential With Once-Through (mills/kWh)
	UO_2 Fuel ($275/kg)	MOX Fuel ($1100/kg)	
70	2.94	3.08	+0.14
85	3.23	3.26	+0.03
100	3.52	3.44	-0.08
120	3.90	3.68	-0.22
Enrichment $/kg-SWU	Fabrication Price		Cost Differential With Once-Through (mills/kWh)
	UO_2 Fuel ($200/kg)	MOX Fuel ($800/kg)	
70	2.72	2.61	-0.11
85	3.01	2.80	-0.21
100	3.30	2.97	-0.33
120	3.68	3.22	-0.46
Enrichment $/kg-SWU	Fabrication Price		Cost Differential With Once-through (mills/kWh)
	UO_2 Fuel ($200/kg)	MOX Fuel ($1100/kg)	
70	2.72	2.94	+0.22
85	3.01	3.12	+0.11
100	3.30	3.30	+0.00
120	3.68	3.55	-0.13

[1] Current Uranium Pricing: UF6 value $35/kg; UO_2 Fabrication $200 to $275/kg; MOX Fabricating $800 to $1100/kg; $70 to $120/kg-SWU. Based on electricity generation: 11×10^9 kWh

Table V. Draw-Down of Excess Plutonium Stores

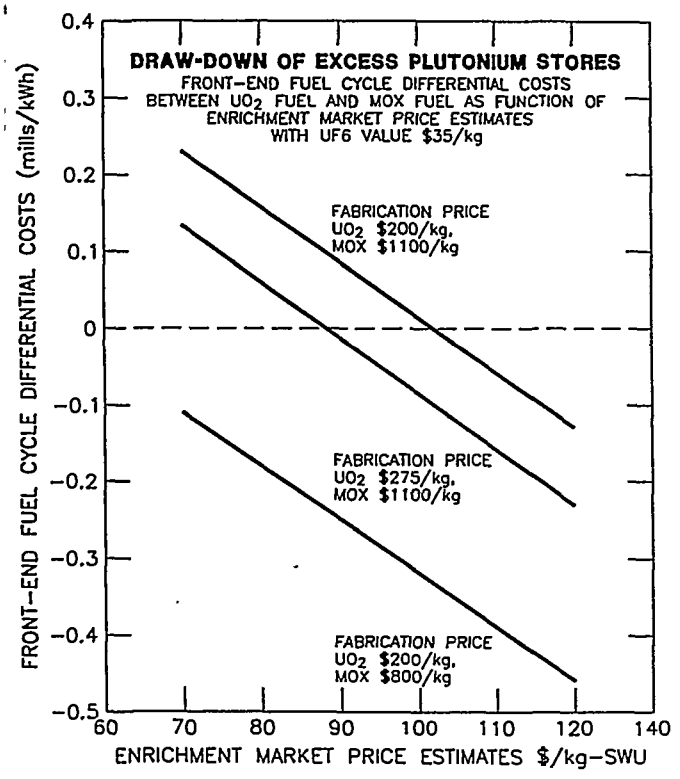

Fig. 2. Draw-Down of Excess Plutonium Stores

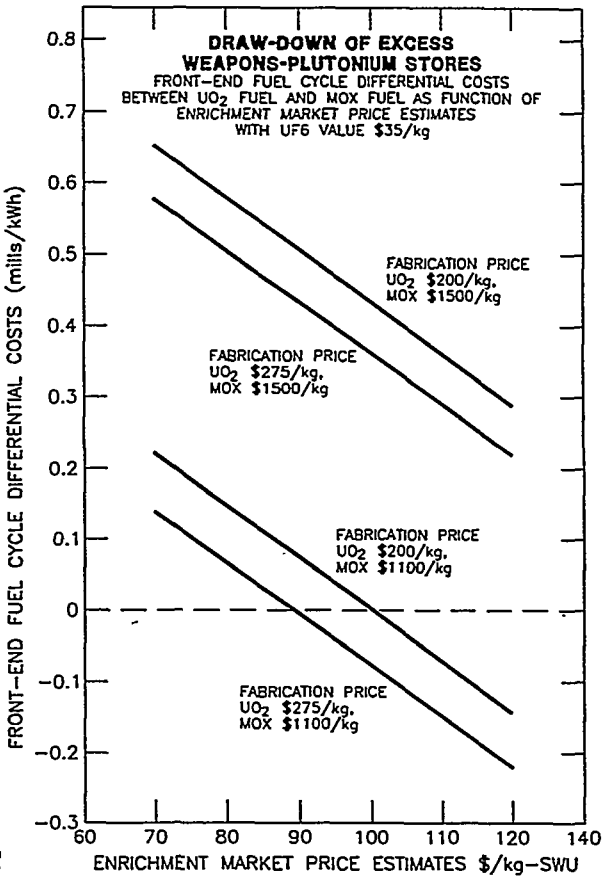

Fig. 3. Draw-Down of Excess Weapons-Plutonium Stores

The revenue generating potential in separated civil-plutonium stores could be a compelling economic incentive to draw-down the excess plutonium. Separated plutonium stores of about 10 MT (7 MT of fissile plutonium) could represent a more than $4 billion revenue generating investment, assuming an electricity retail rate of 70 mills/kWh for the 60 X 10^9 Kwh energy content in the stored separated plutonium. In an analogous manner, assuming weapons-plutonium stores of about 50MT (47 MT of fissile plutonium), the potential revenue generating investment of some $27 billion could be realized.

Draw-down Rates of Separated Plutonium

The characteristic plutonium flows and inventory requirements for the 1st and 2nd recycle mode indicate that the recycle operation could be used

to draw-down the excess separated plutonium stores at an annual rate of about 185 kg of fissile plutonium or 250 to 300 kg of plutonium. Assuming stores of about 9 to 10 MT of separated civil-plutonium, about 40 reactor-years (10 MT/0.250 MT/reactor year) would be required to reduce the excess separated plutonium. For the assumed stores of 50MT of weapons-plutonium, a 250 reactor-year cycle (50MT/0.2MT/reactor year) is estimated to reduce the weapons-plutonium stores. The balance between demand and supply could be effected over several years utilizing 10 reactors that are licensed for 1/3 core civil plutonium MOX loading operations. Another alternative is utilizing a plutonium recycle once-through fuel management schedule. The draw-down of excess separated plutonium would result in a 15 reactor-year cycle (10 MT/0.650 MT/reactor-year) for civil plutonium, and a 95 reactor-year cycle (50MT/0.52MT/reactor-year) for the weapons-plutonium stores.

For reactors licensed to operate with a 100% MOX core loading, the annual rate of fuel requirements would be at the level of 1300 kg of fissile plutonium or 1700 kg of civil plutonium. A once-through fuel cycle management operation would result in about a 6 reactor-year cycle (10 MT/1.7MT reactor-year), and about a 40 reactor-year cycle (50MT/1.35MT/reactor-year) for the weapons-plutonium stores.

The estimated draw-down rates of the excess separated plutonium stores will eventually depend on reactor operating capacity factors.

High-Enriched Uranium (HEU)

The assessment and analysis of accessible isotopic data for high-enriched uranium (HEU) have included isotopic specifications in the research reactor and production reactor programs. The availability of HEU from the dismantlement weapons program, and the re-use of the uranium from the processed spent HEU introduces the possibility of blending these fuels with natural or depleted uranium and then re-enriching to satisfy the fuel element specification requirements of research reactor or power production operations. The analysis focuses on identifying isotopic signatures of fuels having been produced by the blending and/or enrichment process. Preliminary analysis indicates that the correlations of the minor uranium isotopic concentrations may distinguish the isotopics between blended and enriched fuels.

Minor uranium isotopic compositions for all reactor fuels can be investigated to determine the differences between isotopic specifications of fuels enriched to LEU levels and those fuels blended down to LEU from HEU levels. Scoping studies of uranium isotopes from gaseous diffusion enrichment processes (Ref. 5) using a reference simple cascade model were compared with the compositions of the stock of uranium fuels used in the zero-power-critical (ZPR) facilities at ANL (see Figure 4). The U-234, U-235 and the U-236 weight percents for uranium oxide or metallic fuels, as a function of U-235 enrichment ranging from 1.8 to 93 % were examined to determine the dependence on uranium enrichment in a reference gas diffusion process. The isotopics of the feed uranium assumed an initial concentration of U-236 at 0.007 percent. The U-234 concentration exhibited an increasing enrichment functional relationship with respect to the U-235 enrichment, and the U-236 concentration increase exhibited a negative curvature. The isotopic ratios for low-enriched LEU and high-enriched HEU uranium fuels indicates that the isotopic signature measurements made at the product and tail streams of the enrichment process can become an important safeguards data validation procedure for enrichment plants.

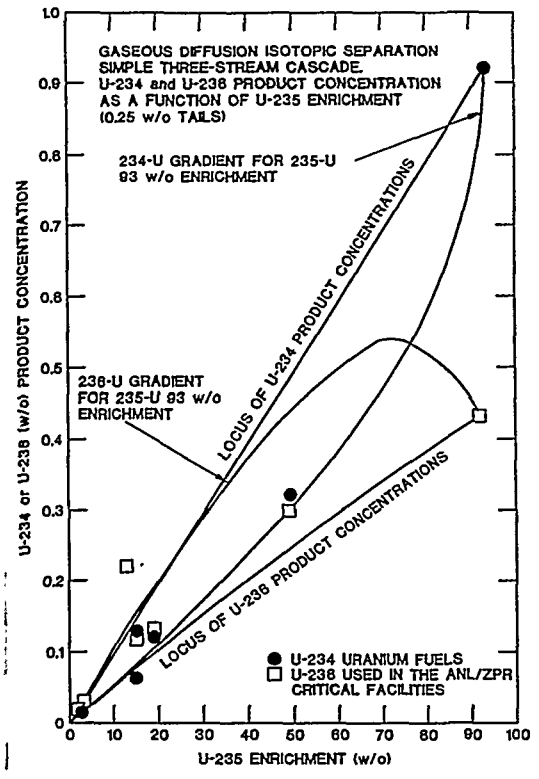

Figure 4. Gaseous Diffusion Isotopic Separation

Minor isotopic concentrations could be used to establish the initial sources (origin) of nuclear material, enrichment, and/or blending processes to explain the many currently different isotopic specifications and those specifications anticipated in the near-term reduced-enrichment fuels program. The results in Figure 4 indicate that if the side withdrawals are small with respect to the product rate, then the concentrations approach the cascade gradients. This mode of cascade operations could explain, in part, the many different isotopic specifications found in the inventories of enriched fuels used in reactor facilities.

HEU of Unknown Origin

The analysis of HEU reported in Nucleonics Week, February 16, 1995, indicated traces of the U-236 (see also Ref. 6). The isotopic concentrations reported in weight percent were 1.078, 87.768, 0.210, and 10.944 of U-234, U-235, U-236 and U-238, respectively. The isotopic signature of U-236 at the product level of about 0.210 percent indicates that in the enrichment process the initial feed concentration could have ranged in the 0.004 to 0.007 percent level, and the depleted uranium stream could have U-235 tails ranging between 0.2 to 0.3 percent. This concentration level is indicative of uranium reprocessing streams from very low-burnup fuels. The U-234 concentration is indicative of a product stream from a typical gas diffusion enrichment process with a natural uranium feed. However, the reported U-234 and the U-236 isotopics seems to indicate that the HEU sample is the product of a gas diffusion isotopic separation process of a uranium product stream from reprocessed low-burnup fuels. The feed to the re-enrichment process could also include some blending with natural uranium. A more detailed study would be required for a more definitive diagnostic analysis. The above scoping investigation does emphasize that the isotopic signatures and correlation techniques can be implemented to establish a basis for declared measurement data validation and safeguards relating to the synergism of potential anomalies and traceability of nuclear material.

Summary

As a consequence of the anticipated near-term fuel cycle activities in the reduction of excess separated plutonium and high-enriched uranium, the assessment of the impact on nonproliferation and safeguards measurement techniques and subsequent R & D programs will require an expanded information/database and knowledge in reactor design, fuel cycle analysis, reactor operations, dynamics of isotope separations, reprocessing, and in the synergism of these technologies with the flow and inventories of nuclear material. The total systems analysis required for identifying anomalies in the nuclear fuel cycle suggests that:

1. The safeguards at the front-end of the fuel cycles involving unirradiated LEU and HEU feed and MOX fuels should be emphasized, as well as at the back-end involving plutonium in the spent fuel.

2. Safeguards agencies should form an internal expert group to develop fuel cycle analyses and assess anomalies.

3. Safeguards agencies should not concentrate safeguards on certain technologies in preference to other existing mature nuclear technologies (calutrons) on the basis of effectiveness or efficiency. This nonproliferation regime would, in effect, force the choice of the less efficient technologies by countries pursuing a clandestine weapons program (The Iraqi Experience). (See Ref. 7.)

4. The isotopic signatures and diagnostic correlation techniques should be implemented to assist in identifying anomalies in the fuel cycle and traceability of nuclear materials.

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