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A SIMPLE METHOD FOR RAPIDLY PROCESSING HEU FROM WEAPONS RETURNS*

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Abstract

A method based on the use of a high temperature fluidized bed for rapidly oxidizing, homogenizing and down-blending Highly Enriched Uranium (HEU) from dismantled nuclear weapons is presented. This technology directly addresses many of the most important issues that inhibit progress in international commerce in HEU; viz., transaction verification, materials accountability, transportation and environmental safety. The equipment used to carry out the oxidation and blending is simple, inexpensive and highly portable. Mobile facilities to be used for point-of-sale blending and analysis of the product material are presented along with a phased implementation plan that addresses the conversion of HEU derived from domestic weapons and realted waste streams as well as material from possible foreign sources such as South Africa or the former Soviet Union.

Introduction

The dilution of Highly Enriched Uranium (HEU) from dismantled nuclear warheads to fuel-grade uranium (3-5 wt. % ^{235}U) and its ultimate sale is important to both the United States and the Commonwealth of Independent States (CIS), both as a potential source of income and because of the strategic importance of HEU. In order for this process to become economically and politically feasible, the conversion from HEU to LEU must be accomplished to the satisfaction of military officials from each nation, members of the international safeguards community, as well as individuals representing the business interests of the buyer and the seller. If performed in an efficient and timely manner, this activity offers the exciting possibility of simultaneously increasing international security through the reduction of nuclear weapons stockpiles while engaging in profitable commerce in nuclear fuel.

The technical issues involved in such transactions are significantly different than those found in the present day fuel cycle. Specifically, the source of enriched product starts out in a metallic form, whose exact chemical and isotopic compositions are imprecisely known, rather than as UF_6 , which is easily blended and has long been the standard article of commerce for enrichment transactions. The flow sheet used to integrate this significant source of metallic enriched U into the civilian fuel cycle must address a variety of

differences inherent in the form, enrichment and location of this enriched feed stream. Specific areas of concern include:

- Chemical purity and processing considerations
- Isotopic composition (down-blending and minor isotope contribution)
- Transportation safety and costs
- Environmental impact
- Occupational and public safety

Many of these product integration issues are analogous to those which will be required for incorporation of (metallic) material produced by the Uranium Atomic Vapor Laser Isotope Separation (U-AVLIS) enrichment process into the fuel cycle. As a result, many of the unit processes that have been developed to address these issues within the U-AVLIS Program can be readily applied to the down-blending and sale of metallic HEU.

In addition to the normal scope of technical and business issues cited above, there are a number of international security issues which must also be addressed as a fundamental part of the product integration strategy. Specifically, because of the strategic importance of HEU, a high degree of assurance must be provided to ensure both that enriched material is not diverted and that transactions involving HEU or down-blended HEU represent material from weapons returns, rather than new production. The integration strategy must

recognize and take into account a number of important technical realities:

- Diversions of even small fractions of the HEU throughput streams constitute a significant national/international security threat.
- One cannot anticipate that materials from weapons with different specifications and/or production histories will be identical. The enriched material represented by the metallic HEU inventory can be heterogeneous, both with respect to isotopic and elemental composition (e.g., fraction impurities). Additional chemical processing will be required to ensure accurate material sampling and accountability.
- Because many of the sites proposed for weapons dismantlement and/or HEU processing are located at existing LEU production facilities, it is vital to assure that the product is derived from HEU and does not represent new Separative Work Unit (SWU) production.

Methodology

In the following sections, we outline a flow sheet for accepting, oxidizing, homogenizing, sampling and blending metallic HEU for ultimate introduction into the civilian nuclear fuel cycle. This process addresses many of the key business and national security issues involved in introducing a metallic source of enriched uranium into the commercial marketplace. The first four steps (materials acceptance, homogenization, sampling and analysis) would be common to any marketing scheme; these steps are shown in Figure 1.

The methodology presented is both cost effective and environmentally benign. The exact method chosen for the final, down-blending step depends on whether the buyer and seller wish to explore the economic advantages of an oxide based economy or remain captive to the UF₆ economy. A blending method used to support an oxide based economy is presented here for illustrative purposes.

Materials Acceptance

The first step in weapons destruction would consist of mechanically reducing the size of the original components by the seller. This step serves the primary

function of providing material in a convenient size for subsequent handling. It also serves to inhibit the ability of other parties to obtain geometric information related to the design of specific weapons systems. While information of this type could, in principle, be recovered, we propose allowing visual, but not hands on verification of the transfer of metallic HEU components.

Following size reduction, material originating from one or more weapons would be assembled in a stream of critically safe batches and inspected both visually and by non-destructive assay techniques, such as gamma-ray spectroscopy. While non-destructive assay techniques are not sufficiently precise (or accurate) to insure accurate materials accountability, they can be effective in preventing the accidental or intentional substitution of undesirable radionuclides into the materials stream.

If desired, it is possible for all parties to independently weigh each batch of material on their own equipment. The material would then be loaded, in the presence of the interested parties, into a batch-type fluidized bed oxidizer shown in Figure 2. For accounting purposes, the weight of the empty and full reaction vessel would be recorded and witnessed prior to connection to the remainder of the fluidizer assembly.

Homogenization

Oxidation of metal is a necessary first step in any of the uranium blending or chemical purification schemes known to us at this time. Oxidizing offers several technical benefits, the most important being the ability to convert metallic HEU into a chemically stable powder that can easily be homogenized. Homogenization leads directly to robust assay verification procedures. In order to assure that the source of LEU is in fact HEU and not new production, it is imperative that assays be obtained as close to the "front end" of the process as possible. The most effective time to obtain this representative assay is after homogenization of the highly enriched oxide when small (~0.1g) samples can be made available to each of the interested parties.

The compact, fluidized bed oxidizer (shown in Figure 2) is the central component of the proposed metal integration system. It provides (at 600°C or higher) a rapid technology for simultaneously oxidizing and homogenizing metal in a simple unit with no moving parts. High pressure air is introduced at the bottom of the reaction chamber and directed up through the uranium

pieces. Spent gas is exhausted through a HEPA filter. At temperatures above 400°C, U spontaneously ignites. Oxide particles, which tend to naturally spall off of the solid material, are sheared off and entrained in the moving gas stream. Part way up the column, the chamber diameter is increased, resulting in a net decrease in gas velocity and disentrainment of the powdered oxide. The particles then fall back to the surface of the bed where they are re-entrained with freshly oxidized material. This process results in homogenization of the batch and is accompanied by repeated contact between particles that result in attriting (mechanical size reduction) of the oxide particles. This further blends and homogenizes the oxide within the system.

The small size of a facility required to process uranium metal to U_3O_8 is emphasized in the artist's conception of a complete plant inside a sea-air-and cargo container (Figure 3). The only operating requirement for this facility is a source of electrical power to operate air compressors, heaters, and HEPA filtered blower units. A small plant comprised of 12 fluidized bed assemblies processing 8 kg per day each, 365 days per year, is capable of processing approximately 35 MT of HEU per year. The removal of three samples from each batch means that the analytical sample load would be 36 samples per day, a workload within the capability of a single instrument (see below).

Sampling

Experience in the U-AVLIS Program, where the product assay varies (for a number of technical reasons) over a narrow range, has indicated that statistical, or spot sampling is simply inadequate to assure accurate commerce in uranium metal. The ability to extract a representative sample of each batch of HEU to precisely establish its isotopic and elemental (e.g., uranium) composition from the starting material is essential for assuring that subsequent diversions do not occur.

Following oxidation/homogenization, product will be made available to each party for sampling. Three 0.1g samples taken from random locations should be sufficient to assure complete blending. Each party has the option to acquire and analyze its own sample set. Mass spectral analysis of the blended material will allow precise determination of the ratio of fissile to non-fissile isotopes. The fraction of total uranium in each sample could be precisely established using isotope dilution techniques. This information, coupled with the

starting weight of metal, will allow each party to precisely establish the fissile isotope content. The number of ^{235}U atoms and therefore, the value of each batch, can be determined with great precision by averaging all known measurements at the time of transfer. The minor isotopic composition (e.g., ^{235}U , ^{236}U) of the material could be further used as a method of establishing a unique fingerprint to perhaps tag each batch of material.

Analysis

From a commerce and accountability point of view, the importance of obtaining accurate isotope assays and purity data cannot be underestimated. The "real-time" analysis capability made possible by placing an analysis facility at the blending site is a vital element of good business practice. This facility need not be complicated. Commercially available instrumentation could easily be modified to allow field deployment. A complete laboratory that also fits into a standard sea-air-land cargo container shown in Figure 4. The heart of this laboratory is a high throughput Inductively-Coupled-Plasma-Mass-Spectrometer (ICP-MS) configured for isotopic analysis. The use of isotope dilution techniques in such a system will assure that uranium assay measurements are made to high accuracy. Alternately, using standard calibration techniques, assays of better than five parts in 1000 can be obtained routinely. The ability to analyze for all isotopes of nearly all of the metallic elements in a single instrument offers a powerful tool for identifying other impurities.

Downblending

The physical characteristics of the oxide that lead to easy homogenization also make it possible to blend with lower assay oxide. Downblending with oxide offers the immediate advantage of converting the material to a form that is less attractive to diversion for (para) military uses. If blended to ≤ 20 wt. % ^{235}U , it offers a further benefit of being subject of less restrictive NRC shipping regulations. (Presently, there are no provisions for the shipment of HEU other than by the national defense establishment.) However, in order to minimize blending losses, it is more economical to blend with Low Enriched Uranium (LEU). The oxide product of the U-AVLIS process would be an ideal source for such material as it is not enriched in ^{234}U and, if produced from virgin material, would be free of ^{236}U .

The low assay material could be either of domestic or

foreign origin. The U.S. mining industry has protested vehemently over the importation of uranium from the CIS. One possible compromise would be to allow the use of low assay U from the CIS for blending down to ~20 wt.% ²³⁵U with the final blending occurring in the U.S. with U-AVLIS produced material.

Transportation

The production and shipment of a blended oxide product form has a number of secondary advantages. As a chemically stable solid, it is less likely to disperse in the event of a transportation accident than is UF₆. Additionally, since U₃O₈ contains a higher mass of U per kg (847 g U), than does UF₆ (667 g U), the oxide should be less costly to ship. The absence of F₂ and HF in the proposed process has significant environmental and occupational safety advantages.

An additional feature of the fluidized bed oxidizer is the external plenum for air cooling the highly exothermic oxidation reaction. This plenum also acts as a secondary containment wall for the process material. If desirable, such a fluidized bed could serve as its own shipping container. A distinct advantage to this concept is that it eliminates the need to provide an infrastructure for unloading powdered material at the point-of processing site. Instead, containers equipped with tamper indicating seals could be returned to a central facility in the U.S. for unloading and chemical conversion into fuel. Considerable cost savings can be realized by shipping lower volumes of material at assays of 20 wt.% ²³⁵U instead of fully diluted 4 wt.% material. If deemed necessary, a second inspection team can verify container assays and weights at this facility to insure no diversions occurred enroute.

Environmental Concerns

Unlike other processes, such as those involving UF₆, the present proposal does not require HF or F₂ storage, handling, processing or disposal. The only reagent required for the entire process is air. There is no waste stream. The small size and simplicity of the hardware will ultimately result in relatively low decontamination and decommissioning costs at the end of the program.

A final, but not insignificant, benefit of oxide production early in the process is related to the reduced consumption of nitric acid required to put the material in a form amenable to purification and ultimate conversion to

ceramic grade UO₂. As indicated by equations (1) and (2) the use of oxide starting material instead of metal reduces consumption of HNO₃ by approximately 50% while production of NO (and subsequently NO_x) is reduced by nearly 90%.

Metallic Dissolution:

Oxide Dissolution:

Both of these effects have economic and environmental advantages.

Implementation Strategy

We propose a two phase plan to establish the operational and economic baseline for this process prior to a third phase deployment activity. Phase I consists of the construction and activation of a single, pre-prototype, fluidized bed oxidizer that incorporates the essential feature of the system described above. This unit will be proof-tested by processing 8kg batches of depleted uranium tails "splats" from the recently downsized U-AVLIS Program. Isotopic and elemental blending will be verified by placing up to 1kg each of natural U and Fe respectively into the test batches. The duration of Phase I would be approximately eight months and would require the following resources:

Project Leader	0.3 FTE
Design Engineer	0.5 FTE
Technician	0.5 FTE
Analytical Chemist	0.3 FTE
	1.6 FTE

Pre-prototype Reactor	50K
Safety Systems	15K
Misc. Operating Expenses	25K
	80K

A top-level schedule for Phase I is shown in Figure 5.

Phase II would result in a fully engineered oxidizer incorporating procedures for loading and unloading HEU, and integration of material produced by this process into the uranium economy. To meet this goal, we propose to oxidize and blend a substantial fraction of material currently held at Y-12 by the Central Scrap

Management Office (CSMO). We further propose to blend this material to 4-5 wt.% ^{235}U for eventual incorporation into the fuel cycle. This is a potential method to realize profits from what is now regarded as waste. Attainment of this goal would require the following resources:

Project Leader	1 FTE
Design Engineer	1 FTE
Technician	1 FTE
Analytical Chemist	1 FTE
ES&H Support	<u>2 FTE</u>
	4.2 FTE
Pre-Prototype Reactor	150K
Safety Systems	30K
Blending Stock HEU	50K
Blending Stock LEU	<u>50K</u>
	280K

A top level schedule for Phase II is shown in Figure 5. If desired, Phase II activities can be accelerated and carried out in parallel with the latter stages of Phase I.

For Phase III we propose to apply the fluidized bed technology to the task of oxidizing, blending and sampling both the remaining CSMO "scrap" and the 40 MT of HEU material from returned U.S. warheads that the United States Enrichment Corporation (USEC) has requested to offset the costs of purchasing HEU from the CIS at above market value. The modular, portable nature of this technology also makes it an attractive candidate for transport to South Africa where it would be used to neutralize their HEU stockpile and put the proceeds onto the civilian nuclear fuel market.

Conclusions

The compact size and modular nature of the fluidized bed system, coupled with its independence from chemical reagent usage and nonexistent waste stream, make it ideal for packaging as a portable factory suitable for point-of-disassembly use. This makes issues related to disarmament, treaty verification, or commerce straightforward and efficient to administer. If it became desirable to speed-up the conversion process, replication of the basic unit is simple and inexpensive.

The proposed system is particularly attractive when compared to other competing options, particularly those

based on remitting scrap followed by conversion first to oxide and then to UF_6 . In addition to a host of environmental, transportation and safety issues, the potential diversion of HEU accompanied by substitution of newly produced LEU in the product streams make such an option difficult to monitor with a high degree of certainty.

About the Authors

William McLean II (Ph.D., Physical Chemistry, 1977) is Group Leader for Materials Technology for the U-AVLIS Program at Lawrence Livermore National Laboratory. He is responsible for identifying and developing technologies to rapidly and accurately determine the ^{235}U mass balance from multi-million dollar U-AVLIS technology demonstrations. Between 1983 and 1993, he and his staff have homogenized and sampled the product and tails stream from experimental programs generating several metric tons of uranium metal.

Philip E. Miller (Ph.D., Analytical Chemistry, 1985) is Group Leader for Inorganic Analysis for the Chemistry and Materials Science Division at Lawrence Livermore National Laboratory and Lead Chemist for the U-AVLIS Program. He directs a fully automated mass spectroscopy facility that performs up to 10,000 isotopic assays per year.

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Figure 1. Sample acquisition, homogenization and analysis flowchart.

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Figure 2. Fluidized bed oxidizer module.

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Figure 3. 35 Metric tons per year oxidation plant. Note: The entire plant is packaged in a SEA-AIR-LAND transportainer.

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Figure 4. Complete, self-contained, mobile sampling laboratory

Figure 5. Top-level schedule for process demonstration.

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