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A new approach for the treatment and conditioning of excess nuclear materials and implications for repository disposal

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Summary. — One of the most critical, and historically overlooked, considerations when employing nuclear materials is the treatment and conditioning strategy for their disposal. Selecting the correct strategy depends on a broad range of factors from the characteristics and size of the inventory, to the available resources and infrastructure. However, equally important as the product, is its compatibility with both interim and final disposal requirements. While with smaller or less exotic inventories there are numerous options available, those options become more limited and challenging as inventories become larger, more hazardous, more unusual, or more proliferation-attractive. This paper will review several of the "established" methods for the treatment and conditioning of nuclear material inventories, noting their relative advantages and disadvantages, and highlight a new approach which may provide a safer, simpler, versatile and more cost-effective process for treating materials to prepare for disposal including the proliferation-attractive inventories. Furthermore, the paper will highlight the importance of defining interim storage and final disposal requirements and how this new treatment approach provides the flexibility to tailor the product to the requirements for interim storage and geologic disposal, which are still evolving in most countries. The paper will conclude by reviewing the implications of this approach for countries and entities considering options for the management of known and potential inventories of excess nuclear materials.

1. – Overview and context

Historically, the users of nuclear materials have focused on the production and utilization of those materials, with little consideration given to the management of scrap, waste, and excess materials, which eventually become part of their legacy material inventories. In recent decades, as research institutions and countries assess their nuclear legacies, future workscope, and the deactivation and decommissioning of facilities, more attention is being paid to the management of the legacy materials, including the treatment, conditioning, storage, and final disposition of these materials.

In many cases, these materials could be used by a malevolent actor to create an improvised nuclear device. As part of its efforts to minimize the threats posed by vulnerable nuclear materials, the U.S. Department of Energy/National Nuclear Security Administration's Office of Material Management and Minimization (M3) works with partner governments and material owners to eliminate the need for weapons-usable nuclear materials (WUNM), through the conversion of research reactors from highly enriched uranium (HEU) to low-enriched uranium (LEU), and the identification and execution of a variety of different consolidation and disposition approaches to ensure that these materials can never be used to create an improvised nuclear device. The approach to manage any given material inventory can vary widely, and depends largely on material-specific factors, the available infrastructure, the desired disposition pathway, and the economics. In many cases, the selected approach will achieve several different objectives, including consolidation of varied inventories, the minimization of their volume, improved safety characteristics, and a reduction in the attractiveness of the materials for weapons use.

1˙1. *Defining terms*. – Throughout this paper, several terms with specific meanings will be used. "Treatment" refers to the minimization of the volume of material to be disposed of. This is typically driven by logistical, practical, or economic drivers. "Conditioning" refers to the transformation of material to be disposed of into a more stable, less hazardous form. This is typically driven by safety considerations, which in some circumstances overlaps with nonproliferation considerations, including for example the downblending of HEU to LEU.

2. – Treatment and conditioning

There are several variables to be considered in identifying the ideal approach for the treatment and/or conditioning of a given inventory. They include the material (and its characteristics), the processing approach, the packaging strategy, and equally important, the interim and/or final disposition facility requirements. Typically, given that the material inventories and characteristics are fixed and known, the material owner or state only have the opportunity and ability to assess the treatment, storage, and disposal options.

The processing approach, packaging design, and the storage/repository disposal strategy and associated acceptance criteria thus become the primary areas that offer the material owner or state the flexibility to optimize the management of the excess materials. While the inventory, infrastructure, policy, economics, regulatory framework, and geology can restrict the options available to a material owner or state, to the extent possible, they should seek or preserve as much flexibility as possible, both to hedge against future uncertainty (*e.g.*, a need to dispose of unanticipated inventories) and to preserve the ability to utilize new technological or scientific advances (*e.g.*, a new treatment method).

2˙1. *Established approaches*. – There are a number of methods to treat and condition nuclear materials which have been generated over the years. While there are more than the four methods presented below, the four below have either been developed to a high level of technical maturity or used extensively around the world. Due to the inherent characteristics of a processing method, any given method will have certain features that are more or less flexible (see the table in fig. 1).

	Treat	Condition	Complex	Space Efficiency	Isotopic Dilution	Homogeneity	Relative Cost	Applicable Materials	Other Considerations
Direct Disposal	No	Yes	No	Low	No	Low	Low	Broad	High technical maturity for power reactor SNF
Compaction	Yes	No	No	High	No	Low	Low	Limited	More applicable to low-level contaminated items
Cementation	No/Yes	Yes	No/Yes	High	No (unless pre-treating)	Low/High	Low	Broad	Long-term durability a challenge
Dissolution & Vitrification	Yes	Yes	Yes	High	Yes	High	High	Broad	Actinide separation possible
Melt Processing	Yes	Yes	No	High	Yes	High	Low	Broad	Highly flexible

Fig. 1. – Table: different processing methods and their features.

Direct disposal is a method that involves encapsulation of the nuclear material in a robust package. The material is typically conditioned prior to encapsulation in an effort to minimize corrosion, criticality, and other undesirable phenomena once emplaced in a repository. Conditioning may change the surface characteristics of the material but not the intrinsic characteristics. As direct disposal does not effectively involve processing or otherwise changing the form of the material, this method does not allow for the modification of a given material, whether to isotopically dilute or homogenize it. With that said, this approach can be very cost effective as it involves loading the material as it is into a package and then emplacing it in a repository.

Compaction is typically used to consolidate materials that are contaminated with nuclear materials, rather than the materials themselves. While it has broad applicability given the significantly larger volumes of these types of low-level wastes, its application for the disposal of bulk amounts of nuclear materials are very limited and rare.

Cementation is a widely used approach with a relatively high degree of flexibility, particularly when coupled with a pre-treatment step. Absent this step, cementation can be used to encapsulate nuclear materials in solid form or to stabilize and immobilize liquids. Without pre-treatment, it has a lower cost and higher throughput rate compared to other treatment processes and is widely applicable to a range of materials. Pre-treatment can make the process significantly more complex, while also allowing it to be more effective. For example, a pre-treatment step could involve the crushing/grinding of solids to a much smaller size, allowing them to be mixed with other additives to achieve, for example, isotopic downblending. This type of pre-treatment also allows for a more thorough blending of the nuclear material and the cement matrix, producing a waste form from which it is much more challenging to extract the nuclear material. As with any cement product, cemented waste forms can suffer long-term durability challenges, but there are a number of methods that can be used to mitigate this issue.

Dissolution and vitrification is the most complex of the methods presented here. It also tends to be more expensive when compared to other processes. However, it can be an extremely flexible process, as different chemical flow sheets can be adapted to process different materials. This approach is typically better suited for inventories where there is a significant amount of similar material as the initial capital investment and the flow sheet development can require significant resources. Besides cost and complexity, the other concern with this approach is that it can also be readily adapted to remove fission products and separate fissile materials, presenting a potential nonproliferation concern.

Any one of the methods may be suitable and ideal in many cases, despite areas of inflexibility, depending on the existing infrastructure and capabilities at a given facility. In fact, M3 has worked with partners globally to implement the treatment or conditioning of nuclear materials using many of these methods; however, there may be circumstances, where more flexibility is required due to material characteristics, economics, or other considerations.

2˙2. *New approach.* – One of the challenges M3 has encountered is inventories of exotic nuclear materials from legacy fuel cycle research, including HEU-thorium mixtures. These typically exist in relatively small amounts (tens of kilograms of heavy metal), but there are several examples of larger inventories (hundreds of kilograms of heavy metal). While relatively small in the context of global inventories of nuclear material, they can present an outsized challenge to manage due to the unique characteristics presented by their form and the inability to achieve effective economies of scale for their treatment and/or conditioning using the established approaches. Moreover, in the context of nonproliferation considerations and M3's mission, despite the challenges associated with disposition, these materials may also be weapons-usable, thus providing an important incentive to identify a method capable of reducing the proliferation threat they pose.

Using HEU-thorium mixtures as an example, M3 sought to identify a method by which we could enable our partners around the world to prepare their inventories of these materials for disposition in a way that is 1) able to achieve isotopic downblending of the HEU to LEU, 2) able to produce a product that can be tailored to the requirements of a variety of different disposal facilities, and 3) cost effective. With these parameters in mind, M3 selected melt processing as the ideal method, as it can achieve the objectives discussed above, is technologically mature (facilitating acceptance by different national regulators), capable of producing a relatively homogenous waste product, and applicable to a broad range of unirradiated and irradiated nuclear material inventories, including: metallic, alloy, oxide, or silicide fuels as well as materials clad in aluminum, magnesium, stainless steel, or zirconium alloys. A picture of the melt processing system at Savannahis given in fig. 2 and the cross section and microstructure of a melt processed ingot is shown in fig. 3. Given this versatility, the method is ideal for M3's mission, as it can be applied to a broad range of different materials, in different countries, with different regulatory and disposal requirements

Fig. 2. – Melt processing system at Savannah River National Laboratory.

3. – Considerations for interim storage and final disposal

A discussed above, there are a number of options available for the treatment and conditioning of nuclear materials, each with different degrees of flexibility and challenges. When seeking to identify the optimum method for the treatment and/or conditioning of an inventory of nuclear materials, there are a number of different variables at play. Key considerations include the characteristics of the material in its initial state and the desired end product characteristics, which can be tailored to meet the disposal requirements.

3˙1. *Current vs. desired form.* – First and foremost, the activity of the material is the most important characteristic to consider. Materials that have been heavily irradiated, or that contain alpha emitters, will need to be handled, processed, and disposed of differently than materials that are unirradiated. With this defined, the next step is to review the waste acceptance criteria (WAC) for the interim and (if available) final disposal location(s). This will define the other key characteristics of the waste form. Building flexibility into the WAC will increase the available treatment/conditioning options. WAC requirements that are based on product performance standards, rather than those that prescribe particular treatment/conditioning methods or material forms provide material owners flexibility to tailor the treatment or conditioning method to produce a waste form that meets the WAC.

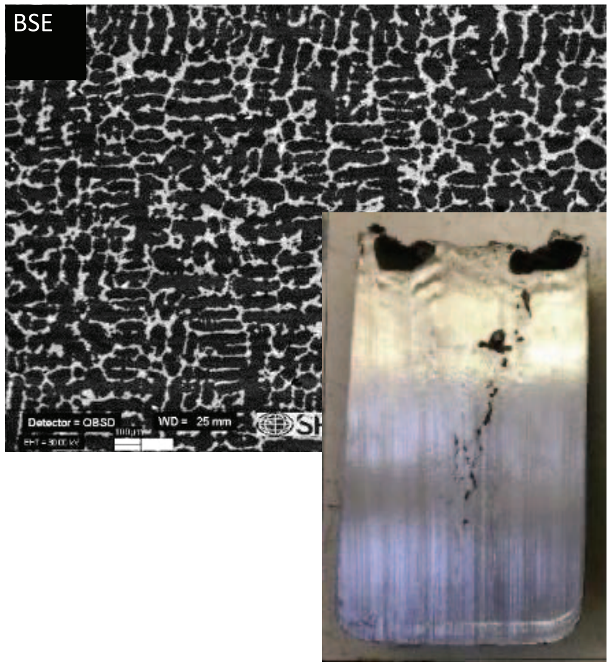

Fig. 3. – Cross section and microstructure of a melt processed ingot (provided by A. Duncan, SRNL).

Another important factor to consider is the total system performance assessment of the final repository. This assessment examines all anticipated materials for the repository and the repository characteristics in order to analyze how the system as a whole will perform on geologic timescales. This assessment guides the development of optimal requirements and characteristics for the various waste forms and waste packages that will be placed in the repository.

With activity known and the WAC understood, the preferred treatment and/or conditioning method can be identified and used as the basis to define the waste form and disposal/waste package configuration. This package must meet both the interim and final disposal facility requirements to avoid post-storage processing. Modeling and analysis can demonstrate a waste package's compliance with these requirements and physical testing can validate the waste form's performance. In some cases, international standards (*e.g.*, ISO, ASTM, etc.) exist and can be referenced in evaluating the waste form instead of developing new testing and analytical methods. For example, ASTM C-1431-99 standard guide for corrosion testing details the waste form test protocol for aluminum-based fuel destined for repository disposal.

3·2. *Existing resources*. – The early stages of this process can be quite daunting; the siting of a repository, the development of a WAC, and conducting all of the required analysis is a significant undertaking. Fortunately, there are a number of resources available to material owners or states to assist in every step of the process. The International Atomic Energy Agency has published a wide variety of technical documents, safety standards, and other references to assist states with the definition of national requirements, from best practices relating to the management of spent fuel, to highly technical guides to assist with the definition of interim and final disposal facility requirements (and the technical basis to underpin them). For example, these documents can help a material owner or state define not only the repository requirements but also the general requirements for the waste form and waste package (dimensions, mass, etc.), corrosion resistance, criticality, thermal performance, shielding requirements, accident conditions, etc.).

4. – Implications and conclusions

The disposition of nuclear material was not given much priority consideration in the early decades of fuel cycle research which focused on fuel development and performance. Fortunately, since then, significant work has been done to help material owners or states plan for the disposition of their materials, in some cases before they receive them. There are also now many technically mature and accepted options available to treat and/or condition nuclear materials for disposal. There are a variety of considerations that inform selection of the optimal and most cost-effective method for a given material inventory and material owner. Lessons learned from successful repository disposal programs show that stakeholder feedback on both technical and policy considerations and requirements play an important role in the success of a repository disposal program.