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Application of the Electrometallurgical Treatment Technique to Long-Term Disposition of DOE Spent Fuel*

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INTRODUCTION

The DOE inventory of spent nuclear fuel consists of approximately 2700 tonnes heavy metal (MTHM), containing over 100 different fuel types. The current plan for the disposition of this fuel is to condition it for dry storage until it can be placed in a geological repository. However, the variation in the physical condition and chemical composition of DOE spent fuel complicates the task of qualifying the fuel for repository disposal. Each type or category of fuel must be characterized and certified to meet repository disposal criteria, an expensive and time-consuming process. Some of the fuel types contain chemically reactive components (such as metallic sodium), which must be stabilized prior to long-term storage or disposal. Finally, some of the fuel is damaged or declad, and some has already been altered by its present storage environment, making it difficult to qualify that general type of fuel for disposal. The electrometallurgical (EM) treatment technique developed at Argonne National Laboratory (ANL) has the potential to convert many of these spent fuel types into a uniform set of three product streams (uranium metal, metal waste form, ceramic waste form). This treatment would simplify the process of preparing and qualifying these fuels for repository disposal. This paper reviews work done on evaluating the applicability of the EM technique to the treatment of the types of

DOE spent fuels currently being stored at the Idaho National Engineering and Environmental Laboratory (INEEL).

PROCESS OVERVIEW

The EM treatment process separates the major constituents of spent nuclear fuel: uranium, cladding, transuranics, and fission products. This separation is performed by selectively electrotransporting the constituents of the fuel in a molten salt electrolyte. The uranium is collected as a pure metal that can be held in interim storage until its ultimate disposition is decided. The cladding and noble metal fission products become part of a metal waste form. The transuranics, alkalis and alkaline earths are dissolved in the electrorefiner salt and incorporated into a glass-zeolite ceramic waste form. Both of the metal and ceramic waste forms are being designed for ultimate repository disposal.

TREATMENT OF DOE SPENT FUEL TYPES

When determining whether a given fuel type can be treated with the EM process, it is the chemical composition of the fuel (i.e., composition of the fuel matrix and cladding) that is of primary concern. The chemical composition of the fuel determines whether a pretreatment step (e.g., oxide reduction) is required prior to electrorefining and also determines the operating conditions (e.g., voltage) of the electrorefining step. In terms of chemical composition, spent DOE fuel can be classified as being metallic, oxide, or "other" (hydride, carbide, or nitride). The salt from the Molten Salt Reactor Experiment (MSRE) falls into a class of its own. Figure 1 provides a general breakdown of DOE spent fuel by chemical characteristics, along with a representative example of each category.[a] The information used to prepare the classifications comes from the Integrated Spent Nuclear Fuel Database System (ISNFDS). [1]

The EM process can be applied to all the metallic fuel types, with the exception of the aluminum matrix fuels, without substantial changes to the existing process. Electrometallurgical treatment of U-Al alloy fuels is possible with some modifications to the current electrorefining technique; theoretical studies and experiments with simulated U-Al alloy fuel have been used to develop a number of viable process options. A large-scale demonstration of the ability of the EM process to separate the components of the EBR-II fuel is in progress at ANL-West. The ability of the electrorefiner to separate the components of Zircaloy-clad uranium metal fuel has been demonstrated in engineering-scale tests using unirradiated N-Reactor fuel.[2] It is anticipated that U-Mo alloy fuel (FERMI-I) can be treated in a manner similar to the N-Reactor fuel. Aluminum-clad uranium metal fuels (Single Pass Reactor-type) have been successfully treated by an initial mechanical process that opens the cladding and exposes the fuel, followed by electrorefining.

Before electrorefining, oxide fuels require a preliminary treatment step to reduce the oxide components of the fuels to a metallic form. A process using metallic lithium dissolved in molten lithium chloride has been developed to accomplish this reduction. The ability of the lithium reduction process to reduce the principal components of oxide fuel has been demonstrated at the engineering scale[3]; current work involves gaining an improved understanding of the parameters involved in scaling up the process. At present, the lithium reduction process can be used to

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[a] Thoria-based fuels and other fuels containing BeO, Gd_2O_3, etc. have been excluded from consideration because the current oxide reduction process cannot be used to reduce these oxides to the metallic form--a necessary step prior to electrorefining.

reduce the clad oxide fuels consisting primarily of UO_2. The existing process can also be used to reduce UO_2-ZrO_2 ceramic matrix fuels, and work is in progress to apply the lithium reduction process to simulated TMI-2 fuel debris.[4]

For the other categories of fuel, work has been successfully completed on demonstration of the EM process to treating MSRE salt.[5] The U-C, U-Zr-H, and U-N fuel types will require an initial head-end step--analogous to the lithium reduction step for the oxide fuels--prior to electrorefining. Theoretical evaluation of treatment options is underway for these three fuel types.

CONCLUSIONS

The EM treatment provides a flexible way to prepare the DOE spent fuel types that require stabilization for long-term repository disposal. The process produces a standard set of stable waste forms from a wide variety of input fuel types and conditions. It is currently being demonstrated at a large scale on actual EBR-II spent fuel and has also been demonstrated on other simulated fuel types, including TMI-2 fuel debris and the MSRE salt on a smaller-scale.

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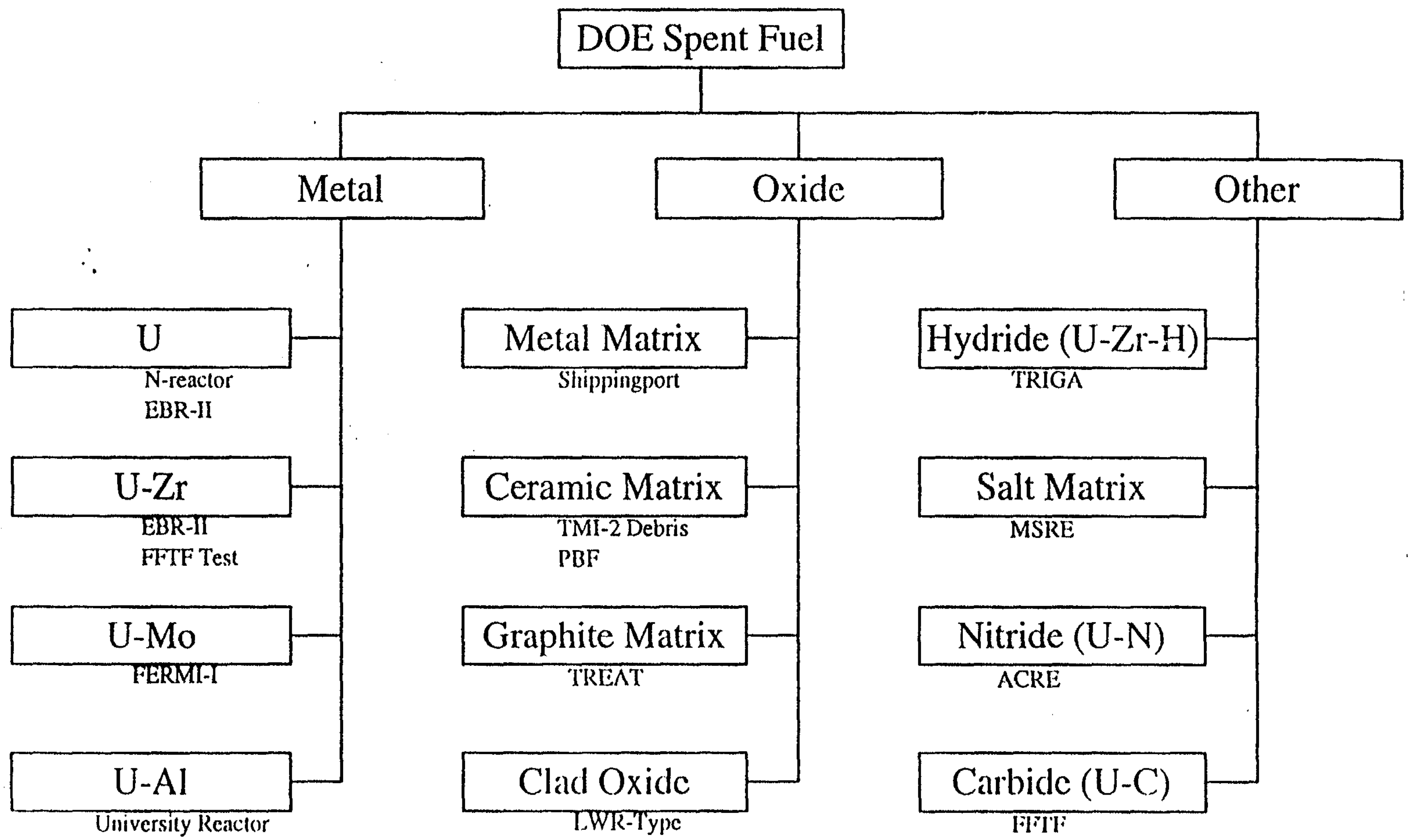

Fig. 1. Classification of DOE Spent Fuel Types by Chemical Composition