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ELECTROMETALLURGICAL TECHNIQUES FOR DOE SPENT FUEL TREATMENT

STATUS REPORT ON ARGONNE NATIONAL LABORATORY'S R&D ACTIVITY THROUGH SPRING 1997

National Research Council

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Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment

Board on Chemical Sciences and Technology
Commission on Physical Sciences, Mathematics, and Applications
National Research Council

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Preface

The National Research Council (NRC) was asked by the Department of Energy (DOE) to provide an ongoing evaluation of Argonne National Laboratory's (ANL's) research and development (R&D) activity on electrometallurgical techniques for treatment of DOE spent fuel, including the specific application of the technology to Experimental Breeder Reactor II (EBR-II) spent fuel. The committee charged with this task has continued to review the program and has prepared this report based on the progress of the program through spring of 1997.

The committee held two meetings in 1997 (the appendix gives the agendas) to review the R&D program at Argonne National Laboratory-East (ANL-E) and evaluate the progress of the EBR-II demonstration at Argonne National Laboratory-West (ANL-W). The first meeting was held on April 7-8, 1997, at the National Research Council in Washington, D.C. Briefings by ANL, DOE, and other interested groups were presented. Subsequently, the committee formulated technical questions on the program in areas that required further clarification by ANL and DOE.

The committee's second meeting, on May 15-16, 1997, was held in Argonne, Illinois, in conjunction with a site visit. The first day consisted of presentations by ANL-E and ANL-W researchers and a laboratory tour to examine the operations. On the second day, the committee discussed the technical details and deliberated the issues that have formed the basis of the recommendations in this report.

The committee meetings and site visit to ANL-E were particularly productive. The committee found the level of detailed technical discussions to be very useful and the information provided by ANL researchers to be important for its evaluation of the program.

Contents

Executive Summary

Background

In September 1994, in response to a request from the Department of Energy (DOE), the National Research Council (NRC) established the Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment. The committee was charged with evaluating the technical advantages and disadvantages of a proposed Argonne National Laboratory (ANL) R&D program on the use of electrometallurgical technology to treat DOE spent fuel, including the specific application of the technology to Experimental Breeder Reactor II (EBR-II) spent fuel. A preliminary report of the committee was issued in February 1995 (NRC, 1995a), followed by a more extensive report in July 1995 that recommended that the technology of electrometallurgical processing appears sufficiently promising for treating a variety of DOE spent fuels that continued R&D would be warranted (NRC, 1995b, p. S-1).

DOE subsequently requested that the Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment continue its activity by carrying out two tasks. The first task involved monitoring the scientific and technical progress of ANL's R&D program on electrometallurgical techniques for the treatment of DOE spent nuclear fuel, including both the redirected research program at ANL-East (ANL-E) and the fuel treatment program at ANL-West (ANL-W) associated with the ongoing shutdown of the EBR-II. The second task was to evaluate the scientific and technological issues associated with extending this R&D program to handle plutonium, should DOE decide that an electrometallurgical treatment option for the disposition of excess weapons plutonium is worth pursuing. The second task was addressed by the committee in the 1996 report, *An Evaluation of the Electrometallurgical Approach for Treatment of Excess Weapons Plutonium* (NRC, 1996a). The success of the planned demonstration of the process as applied to the treatment of EBR-II spent fuel was deemed essential in determining the future of the program. Two additional reports (NRC, 1996b and 1997a) have been submitted to DOE in response to the first task of continued monitoring and evaluation of the electrometallurgical program.

Ongoing Evaluation Activity

As ANL's R&D program has developed over the past several years, changes in its details and scope have been necessitated by, for example, DOE's 1996 Environmental Assessment (EA) (DOE, 1996). In response to these changes, DOE requested that the committee, as part of its ongoing evaluation, revisit the question in its original charge: "Do pyrometallurgical techniques represent a potentially viable technology for DOE spent fuel treatment that warrants further research and development?" In the following sections, the committee provides its evaluation of the program and the associated activities at ANL-E and ANL-W through the spring of 1997.

Status of the EBR-II Spent Fuel Treatment Demonstration for 1999

The ANL program must be considered in terms of an R&D project that is focused on the treatment of EBR-II spent fuel by the electrometallurgical technology. Within that context, the program is making acceptable progress in providing a sufficient database for DOE to make a technical decision on the application of this technology to treatment of other DOE spent fuels. Thus, the committee reaffirms its overall recommendation of the July 1995 report (NRC, 1995b, p. S-11):

> ANL should proceed with its development plan in support of the EBR-II demonstration. . . . If the EBR-II demonstration is not accomplished successfully, the ANL program on electrometallurgical processing should be terminated. On the other hand, if the EBR-II demonstration is successful, the DOE should revisit the ANL program at that time in the context of a larger, "global" waste management plan to make a determination for possible continuance.

This reaffirmation of the 1995 recommendation is based on the quality and commitment of the involved ANL-E and ANL-W personnel, and on the progress in both the ANL-E R&D and the ANL-W demonstration. The present status of the demonstration project indicates that a strong and committed R&D staff continues to be an important factor. A focused R&D program must be maintained for the successful demonstration of the electrometallurgical technology. The committee encourages ANL to proceed aggressively to resolve the R&D issues and move rapidly into a demonstration phase that identifies process definitions and conditions.

Demonstration Project Implementation Plan

In the March 1997 report, the committee noted the benefits to be gained by formalizing the organizational arrangements of the demonstration project, both at ANL-E and ANL-W, and between the two ANL sites (NRC, 1997a). The committee is pleased to note that increased organizational structure is being brought to the project by the Work Breakdown Structure process (ANL, 1997). A less detailed project implementation plan can further clarify how programs are being conducted in an integrated and coordinated manner. *The committee looks forward to receiving the demonstration project implementation plan after it is approved by DOE.* It is recommended that the plan be written in a form that can be easily understood and that allows for an analysis of the efficacy of the new organizational structure.

Adequacy of the Criteria for Evaluating the Demonstration

The committee believes that DOE must determine and approve of the criteria by which the ANL's R&D program will be evaluated at the end of the demonstration in 1999. *Before the demonstration is* *completed, DOE should establish criteria for success in the demonstration phase to allow evaluation of the electrometullurgical technology for further use in treating DOE spent fuel.*

The current plan for application of electrometallurgical technology to the treatment of EBR-II fuel is limited by the terms of the current EA, which specifies the amount of fuel that can be processed (DOE, 1996). Processing of the remaining EBR-II spent fuel would require preparation of an EA plan within the next 2 years and its approval prior to further, and possibly expanded, utilization of the electrometallurgical technology. *A new EA will be required before additional EBR-II spent fuel can be treated. DOE should begin plans for such an EA now so that its preparation does not become the source of a major operational delay, if the current demonstration project is successful.* Alternatively, if the demonstration does not satisfactorily meet the criteria for success, other technologies will be required for treatment of the remaining EBR-II spent fuel.

Electrometallurgical Techniques for Treatment of Other DOE Spent Fuels

The committee continues to believe that successful demonstration of the electrometallurgical process for treating EBR-II fuel is essential to support development of applications of this technique to treatment of other DOE spent fuels. ANL's research efforts have involved the investigation of the electrometallurgical technology for treatment of non-EBR-II fuels such as the N-reactor fuel. However, the DOE Office of Environmental Management (EM) may proceed with plans for the N-reactor fuel that do not include the use of electrometallurgical technology. Since the current approach of DOE-EM is to develop project plans for implementation within the next 10 years, *the offices of Nuclear Energy (NE) (which funds the present program) and Environmental Management (EM) should maintain close contact to ensure proper coordination of their activities.*

Waste Forms

ANL has achieved satisfactory progress in the preparation, characterization, and testing of development-scale ceramic and metal waste forms. *DOE should establish acceptance criteria for waste forms scheduled for storage in a geologic repository.* The electrometullurgical technology program currently is forced to assume that its own definition will be acceptable to the U.S. Nuclear Regulatory Commission. DOE should provide its best guidance to ensure that the Argonne approach is useful.

External Technical Experts

At the current stage of the ANL R&D program, *the committee suggests that ANL utilize external technical experts in specific scientific areas of the program. These technical experts should be recognized for their in-depth knowledge in particular technical areas.* The committee suggests establishing more formal and intensive interactions with experts in particular fields for the benefit of the program.

Background

In September 1994, in response to a request from the Department of Energy (DOE), the National Research Council (NRC) established the Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment. The committee was charged with evaluating the technical advantages and disadvantages of a proposed Argonne National Laboratory (ANL) R&D program on the use of electrometallurgical technology to treat DOE spent fuel, including the specific application of the technology to Experimental Breeder Reactor II (EBR-II) spent fuel. A preliminary report of the committee was issued in February 1995 (NRC, 1995a), followed by a more extensive report in July 1995.

The July 1995 report, *An Assessment of Continued R&D into an Electrometallurgical Approach for Treating DOE Spent Nuclear Fuel* (NRC, 1995b), resulted from the committee's full investigation of the potential viability of the technology for treatment of DOE spent fuel. The committee concluded that "the technology of electrometallurgical processing appears sufficiently promising for treating a variety of DOE spent fuels that continued R&D would be warranted" (p. A-2). Further, the committee provided the following overall recommendation regarding the Experimental Breeder Reactor II (EBR-II) demonstration, which is now expected to conclude in June 1999 (NRC, 1995b, p. S-11):

> ANL should proceed with its development plan in support of the EBR-II demonstration. . . . If the EBR-II demonstration is not accomplished successfully, the ANL program on electrometallurgical processing should be terminated. On the other hand, if the EBR-II demonstration is successful, the DOE should revisit the ANL program at that time in the context of a larger, "global" waste management plan to make a determination for possible continuance.

DOE further requested that the Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment continue its activity by carrying out two tasks. The first task was to provide an ongoing evaluation of ANL's R&D activity on electrometallurgical techniques for the treatment of DOE spent fuel, including their specific application in both the EBR-II spent fuel redirected research program at ANL-E and the fuel treatment program at ANL-W associated with the ongoing shutdown of the EBR-II. The second task was to evaluate the scientific and technological issues associated with extending this R&D program to handle plutonium, should the DOE decide that an electrometallurgical treatment option for the disposition of excess weapons plutonium would be worth pursuing. The second task was addressed by the committee in the 1996 report, *An Evaluation of the Electrometallurgical Approach for Treatment of Excess Weapons Plutonium* (NRC, 1996a).

In response to the first task, continuing evaluation, the committee provided technical status reports on progress in July 1996 (NRC, 1996b) and March 1997 (NRC, 1997a). Both status reports identified specific areas in which ANL should focus its effort in order to maximize R&D progress and achieve successful demonstration of the electrometallurgical process for treatment of EBR-II spent fuel. The committee stated (NRC, 1997a, p. 1):

> The EBR-II fuel conditioning work at ANL-W is still in the process evaluation and equipment development phases, as can be expected in the initial stages of such a demonstration and with the new limitations set by the EA [Environmental Assessment].

In 1996 DOE issued an Environmental Assessment (EA) of the electrometallurgical treatment research and demonstration project in the Fuel Conditioning Facility at Argonne National Laboratory-West (DOE, 1996). The EA necessitated a variety of changes in the details and scope of the EBR-II demonstration program. In response to these changes, DOE requested that the committee, as part of its ongoing evaluation, revisit the question in its original charge: "Do pyrometallurgical techniques represent a potentially viable technology for DOE spent fuel treatment that warrants further research and development?" This report provides the committee's evaluation of the ANL's R&D program on the use of electrometallurgical technology for treatment of spent fuel as of the spring of 1997.

Ongoing Evaluation Activity

The ANL program must be considered in terms of an R&D project that is focused on the treatment of EBR-II spent fuel by the electrometallurgical technology.[1] Within that context, the program is making acceptable progress in providing a sufficient database for DOE to make a technical decision on the application of this technology to treatment of other DOE spent fuels. Thus, the committee reaffirms its overall recommendation of the July 1995 report (NRC, 1995b, p. S-11):

> ANL should proceed with its development plan in support of the EBR-II demonstration. . . . If the EBR-II demonstration is not accomplished successfully, the ANL program on electrometallurgical processing should be terminated. On the other hand, if the EBR-II demonstration is successful, the DOE should revisit the ANL program at that time in the context of a larger, "global" waste management plan to make a determination for possible continuance.

[1] See, for example, Argonne National Laboratory, Nuclear Technology, EBR-II Spent Fuel Treatment Program Monthly Reports, November 1996 through March 1997.

This reaffirmation of the 1995 recommendation is based on the quality and commitment of the involved ANL-E and ANL-W personnel, on the progress in both the ANL-E R&D and the ANL-W demonstration, and on the assumption that the demonstration will meet the criteria established by ANL. The present status of the demonstration project indicates that a strong and committed R&D staff continues to be an important factor. A focused R&D program must be maintained for the successful demonstration of the electrometallurgical technology. The committee encourages ANL to proceed aggressively to resolve the R&D issues and move rapidly into a demonstration phase that identifies process definitions and conditions. The current status of the program is discussed below.

Argonne National Laboratory has provided the committee with the following information: the Driver Assembly Process Flow Diagram and mass balance; the Blanket Assembly Process Flow Diagram and mass balance; the two Ceramic Waste Process Flow diagrams and mass balances for the "throw away" and "batch" exchange operations; and the Spent Fuel Treatment Project Schedules. The committee, in addition, requested flow sheets with more detail. The project schedules were very helpful for the committee's understanding of expectations related to the decision points in June 1999.

Status of the EBR-II Spent Fuel Treatment Demonstration for 1999

Status of Electrorefiners

The committee recognizes that the work being carried out on the Mark IV electrorefiner (Mark IV) in the Fuel Conditioning Facility (FCF) as well as on the Mark V High Throughput Electrorefiner (HTER), both at ANL-W, and the HTERs under development at ANL-E, is still in the R&D phase.

The committee's present understanding is that the Mark IV electrorefiner at ANL-W has an inner diameter of about 40 inches. There are four 10-inch-diameter ports on its top, two for loading and unloading the cathode mandrel on which uranium is deposited and collected and the other two for loading and unloading the anode baskets in which chopped fuel is placed and from which accumulated cladding hulls must be removed. Mark IV is operating with 400 to 450 kg of KCl/LiCl salt and has a liquid cadmium pool. It has a batch size of 16 kg (8 kg per anode) of uranium and is being used to treat the EBR-II driver fuel. Separation of uranium and zirconium is accomplished by controlling the current-time (coulomb) history during deposition and monitoring the uranium potential on the cathode. Separation is optimized by control of electrode configuration and operating conditions.

The Mark V HTER at ANL-W is undergoing testing prior to installation in the FCF. It has the same diameter salt container as the Mark IV, along with four 10-inch-diameter ports in its top, one for each of four cathode/anode pairs. It will be used to treat the EBR-II blanket assemblies during the last phase of the demonstration project (see below). Each cathode/anode pair uses a combination of two rotating anodes and a stationary cathode arranged in a concentric, cylindrical configuration. In contrast to the Mark IV, uranium is removed from the cathode mandrels continuously by being scraped into a removable wire-screened buckets positioned under the cathodes. Mark V has a batch size of 150 kg of uranium when all four electrode pairs are operating. The concentric anode/cathode design (curved anode baskets located in the annuli between cathode cylinders) gives increased throughput by allowing increased current densities. The increased current densities result from increased electrode surface area and decreased distance between anode and cathode. Of utmost importance is the scraping of the uranium from the cathode as it is deposited. Considerable work is being carried out at ANL-E to evaluate scraper configurations and conditions for the Mark V. However, ANL-E has an 8-inch HTER, and a 25-inch HTER, the inner portion of which is to be used to mimic the operation of the individual 10-inch ports in the Mark V at ANL-W.

The 25-inch HTER under development at ANL-E has anode/cathode modules of approximately the same configuration as those in Mark V. This electrorefiner also has a batch size of 150 kg of uranium. ANL is varying both the length (10 and 26 inches) and the number (20 and 8) of anode baskets as part of its parametric studies related to the use of the HTER for other DOE spent fuels. While various operating conditions are being researched, one typical set tried is an anode rotation speed of 50 rpm and a current density of 0.07 amps per cm^2. These parameters are being studied to find the best operating conditions.

The Mark IV and Mark V electrorefiners have similarities but also differ in significant ways. Mark IV collects uranium on the cathodic mandrel. During the deposition, a scraper shapes the cathode deposit. A cadmium pool at the bottom of the electrorefiner catches and dissolves any uranium that either falls from or is scraped off the cathode. That uranium is subsequently redeposited on the cathodic mandrel. Entrapped salt and cadmium are removed from the uranium by distillation, and the uranium is cast into an ingot. In contrast, Mark V does not employ a cadmium pool and does not collect the majority of the uranium on the cathode. Instead, the uranium is continuously scraped off the cathode and collected in a basket below the cathode. The collected uranium is then melted, excess salt is distilled off, and an ingot is cast. The design, testing, and production of satisfactory scrapers appear to be vital to the success of the Mark V and other HTERs.

Uranium trichloride will be added to the Mark V electrorefiner to provide a mechanism for transporting the uranium from the anode compartment (dissolver) to the cathodic mandrel. This function is served by the addition of cadmium chloride to the process salt in the Mark IV electrorefiner. ANL-W appears to be considering producing UCl_3 for this purpose.

Dissolution efficiencies are in the range of 88 to 99.9 percent, in the best cases exceeding the design basis of 99.5 percent.[2] However, reproducibility of the dissolution step is uncertain enough that additional R&D may be warranted.

Material balances are good for both Mark IV and Mark V: about 98 percent for driver fuel assemblies and about 95 percent for blanket assemblies. However, many uncertainties remain. It is not

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[2] Dissolution efficiency: ratio of undissolved uranium to total amount of uranium.

clear what the purity of the uranium deposit on the cathode mandrel will be, nor what will happen to plutonium and the other elements present.

The committee notes that the difference between demonstrating the viability of the process and optimizing the process seems to be blurred. The committee believes that ANL should be clear about directing its efforts to the former. The committee makes this observation in light of what it perceives to be significant effort devoted to various electrorefiner designs, and it questions whether they are required for experimental verification or for optimization.

Process Development

Process development work being carried out in the Mark IV has involved both irradiated and unirradiated materials and has used both depleted uranium and depleted uranium with zirconium metal present.[3] The presence of zirconium metal appears to improve both uranium collection efficiency and the nature of the uranium deposit. The electrorefining process is being carried out at constant current to a fixed voltage cutoff. ANL fully understands the importance of establishing and stabilizing the process operating conditions that will permit anodizing the uranium while leaving the zirconium mostly in the anodization baskets, and it has made good progress in this area.

ANL is studying a number of important process operating parameters, such as rotation rates, scraper configuration, and current densities, on all of the electrorefiners. One parameter specific to the Mark IV is the "path" followed by the uranium during its deposition. The first path is what ANL calls "direct." In this path, uranium in the chopped driver fuel is anodized from anode baskets into the melt, followed by its reduction and deposition as metallic uranium at the cathode. The second path ANL calls "deposition." In this path, the uranium in the chopped driver fuel is anodized from the anode basket into the molten process salt, reduced to metal into the cadmium pool (which is operated as a cathode), anodized back into the process salt from the cadmium pool (which is now operated as an anode), and finally reduced and deposited as metallic uranium at the cathode. $CdCl_2$ is added to the electrorefiner to oxidize the uranium in the cadmium to U(III) so as to effect its transfer to the process salt. Care must be taken because an excess of the cadmium salt apparently can corrode iron components of the electrorefiner.

Electrorefiner operating results of particular interest are the morphologies of the uranium deposits and the uranium collection efficiencies. The morphologies of the deposits as shown in several photographs of the cathodes from the Mark IV electrorefiner appear more dendritic than the deposits seen by the committee at ANL-E. However, since the uranium deposits are treated further in the cathode processor and the casting furnace, this does not appear to be a serious problem.

One possible benefit of the Mark V electrorefiner would be the elimination of the casting furnace process step because the cathode processor, operating on uranium scraped from the cathode, can produce

[3] Argonne National Laboratory, Nuclear Technology, EBR-II Spent Fuel Treatment Program Monthly Report, March 1997.

an acceptable uranium ingot. (Since there is no cadmium involved in the operation of the Mark V electrorefiner, the operation of the cathode processor is simplified.)

Collection efficiencies for uranium deposits in the Mark IV range from 42 to 78 percent. They are stated to be less than 100 percent because of loss of uranium that falls from the cathode during deposition. (This uranium is not actually lost but falls into the cadmium cathode, from which it may be recovered by subsequent anodization into the molten process salt and then deposited onto the cathode mandrel.)

Crucibles

Material for the cathode processor crucible is still being evaluated to minimize the uranium loss (more than the design specification of 1 percent) to the crucible itself. When process salt and cadmium salt are present in addition to the uranium, graphite crucibles are attacked, with changes in the quality of the uranium ingot, formation of loose dross, appearance of a heavy surface scale, and poor release of the ingot from the crucible. Beryllia crucibles were stated in a presentation to the committee to "look favorable," but it is not clear from data so far in evidence that beryllia is the answer. Large beryllia crucibles were found to be subject to thermal stress failure, and small crucibles exhibited poor ingot release. Since the Mark V will not have one of the troublesome elements--cadmium--it is not clear whether the crucible failure problems will persist with that unit.[4]

Project Schedule

As indicated in the Spent Fuel Treatment Project Schedules supplied by ANL, blanket assembly treatment will not start until March 1998 at the earliest, although installation of the Mark V unit at ANL-W was scheduled to begin in the summer of 1997. Heretofore it has not been clear to the committee whether both of these electrorefiners are to operate at the same time during the EBR-II fuel treatment demonstration. Further, it was not clear what effect, if any, the EA-mandated reduced scale of the demonstration would have on the operation of the equipment. Based on presentations to the committee, it now appears that there will be no adverse effect, either in electrorefiner operation or in demonstrating the success of the process. In any event, both electrorefiners are needed to complete the demonstration on the projected schedule.

Demonstration Project Implementation Plan

In the March 1997 report (NRC, 1997a), the committee noted the benefits to be gained by formalizing the organizational arrangements of the demonstration project, both at ANL-E and ANL-W and

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[4] Presented by R. W. Benedict, Argonne National Laboratory, on May 15, 1997, Argonne, Ill.

between the two ANL sites. The committee is pleased to note that increased organizational structure is being brought to the project, and it believes a more focused, well-coordinated demonstration will result.

Clarification of responsibilities at ANL-E and ANL-W is being achieved by increased use of the Work Breakdown Structure (WBS) process (ANL, 1997). It could be helpful to augment the WBS process with a less detailed project implementation plan that addresses in particular the sequence in which tasks are performed at the two sites and their interactions and interdependencies. In this way, significant task overlaps and inefficiencies may be more readily noted and avoided.

The committee looks forward to receiving the demonstration project implementation plan after it is approved by DOE. The plan should be easy to understand, allowing for analysis of the efficacy of the new organizational structure and the general manner in which ANL-E and ANL-W programs are being conducted in support of integration and coordination.

Adequacy of the Criteria for Evaluating the Demonstration

The May 15, 1996, Environmental Assessment (EA) (DOE, 1996) stipulated the amount of fuel that can be processed and defined a treatment technology that does not separate plutonium. The impact of the EA was to limit the processing operations to 100 driver assemblies and 25 blanket assemblies and to render the criteria established in 1995 inadequate (NRC, 1995b). In March 1997 the committee recommended, "A well-defined set of performance criteria needs to be developed [in light of the modified scope as defined by the EA].... The achievement of those objectives would better position ANL to request approval to proceed to additional applications of its electrometallurgical technology program" (NRC, 1997a, pp. 1-2).

In light of the EA, ANL has redefined its criteria for evaluating the success of the EBR-II demonstration to be concluded in 1999. The proposed criteria presented by ANL[5] are as follows:

1. Demonstration that the 100 driver and 25 blanket EBR-II assemblies can be treated in FCF within three years, with a throughput rate of 16 kg/month for driver assemblies sustained for a minimum of 3 months and a blanket throughput rate of 150 kg for one month.
2. Quantification (for both composition and mass) of recycle, waste, and product streams that demonstrate projected material balance with no significant deviations.
3. Demonstration of overall dependable and predictable process, considering uptime, repair and maintenance, and operating of linked process steps.

[5] Presented by R. W. Benedict, Argonne National Laboratory, on May 15, 1997, Argonne, Ill.

4. Demonstration that safety risks, environmental impacts, and nuclear materials accountancy are quantified and acceptable within regulatory limits.

The committee generally concurs that meeting these criteria would provide DOE with sufficient information for decision making on the success of the 1999 EBR-II demonstration. However, some clarification of the criteria appears to be necessary. While the above criteria define general objectives, further clarification and well-articulated definitions may provide a stronger basis for ANL to demonstrate the effectiveness of electrometallurgical technology in treating spent fuel. For example, it is not clear what "significant deviations" means in the second criterion. Further, in the third criterion, how will "dependable" and "predictable" be defined? In the fourth criterion, who will determine acceptability in the areas mentioned? *Before the demonstration is completed, DOE should establish criteria for success in the demonstration phase to allow evaluation of the electrometullurgical technology for further use.* In 1999, ANL's achievements should clearly prove its competence to complete the processing of the remaining EBR-II spent nuclear fuel and provide DOE with confidence that the electrometallurgical technology will be a viable option for processing other DOE spent fuel.

The committee acknowledges that there must be a measure of subjectivity in deciding whether the criteria for success have been met, especially in an R&D program, and that criteria should not be too prescriptive. Nonetheless, there are several possible ways to define better the process for establishing success. To better define the goals in specific technical areas, ANL may consider, for example, involving external technical experts.

The current plan for application of the electrometallurgical technology to EBR II fuel is limited by restrictions on the amount of fuel that can be processed as set by the current EA. If the demonstration is successful, it would seem reasonable that the remaining EBR-II spent fuel would be treated by this technology. Processing of the remaining EBR-II spent fuel would require preparation of an EA plan within the next 2 years and its approval prior to further, and possibly expanded, utilization of the electrometallurgical technology. *A new EA will be required to go beyond the currently approved amount of fuel. DOE should begin plans for such an EA now so that its preparation does not become a major delay, if the current project is successful.* Alternatively, if the demonstration does not meet the criteria satisfactorily, other technologies will be required for treatment of the remaining EBR-II spent fuel.

Electrometallurgical Techniques for Treatment of Other DOE Spent Fuels

Electrometallurgical technology has been proposed as a process having the potential to treat successfully other DOE spent fuels. Among the earlier incentives to proceed with R&D on the electrometallurgical technology was its potential for handling a variety of different spent fuels, such as N-reactor fuel from Hanford, Molten Salt Reactor Experiment (MSRE) residues, and Savannah River Site fuels. In its 1996 report, the committee recommended that "upon satisfactory completion of the demonstration with EBR-II fuel, the electrometallurgical technique should be evaluated in the broader context of alternative technologies for processing spent nuclear fuel" (NRC, 1996a, p. 2).

The major DOE spent fuel type is the N-reactor fuel at the Hanford site, which makes up nearly 80 percent of the DOE spent fuel inventory. The deteriorated condition of part of this fuel necessitates reduction of uranium oxide to the metal before its treatment using the basic electrometallurgical process. The ANL ancillary oxide reduction process using lithium metal must be demonstrated before the electrometallurgical process can be considered for use with this N-reactor fuel. When the requisite ancillary processes have been demonstrated, DOE should consider the electrometallurgical process in context with other competing processes for dealing with the DOE spent fuel problem. These competing processes include aqueous processes as well as long-term storage, which is planned and has been partially implemented for use with spent fuel at some DOE sites.

The committee continues to believe that successful demonstration of the electrometallurgical process for treating EBR-II fuel is essential to support development of applications of this technique to other DOE spent fuels. If this technology is to be considered by DOE for treatment of other reactor spent fuel, DOE should evaluate the upcoming results of the spent fuel treatment demonstration and initiate planning for the post-1999 period. DOE should begin to consider the criteria for success in the demonstration and the spent fuel to which the technology would be most applicable and to determine the R&D data and "hot" cell demonstration requirements for reaching a decision based on technological and economic considerations. This process should be initiated soon if DOE is to utilize the results of the 1999 demonstration effectively and if this technology is to be employed in the DOE-EM program. The committee wishes to restate its position that ANL, upon meeting the appropriate criteria for success in processing EBR-II driver fuel and blanket assemblies, should successfully demonstrate the processes ancillary to the central electrometallurgical process itself, for example, lithium reduction of oxide fuels, before the process can be considered seriously for use with other DOE spent fuel.

It appears highly unlikely that electrometallurgical techniques will be applied to the treatment of MSRE salts from Oak Ridge National Laboratory (NRC, 1997b). R&D dealing with extraction of zirconium and uranium may, however, have applications to spent fuels other than MSRE residues such as N-reactor fuel.

ANL's research efforts have involved the investigation of electrometallurgical technology for non-EBR-II fuels such as the N-reactor fuel. However, the DOE-EM may proceed with plans for the N-reactor fuel that do not include the use of electrometallurgical technology. Since the current approach of DOE-EM is to develop project plans for implementation within the next 10 years, *the offices of Nuclear Energy (NE) and Environmental Management (EM) should maintain close contact to ensure proper coordination of their activities.*

Waste Forms

Since the fall of 1997, ANL has achieved satisfactory progress in the preparation, characterization, and testing of development-scale ceramic and metal waste forms. The committee has concerns, however, regarding characterization of product homogeneity and scale-up of the manufacturing process for the ceramic waste form, as well as whether batch loading options may supplant ion-exchange column loading of radionuclides into zeolite or sodalite.

A key portion of the electrometallurgical technology is the creation of two novel waste forms. One is a metallic waste form composed predominantly of cladding metal. The second is a "ceramic" waste form composed of synthetic zeolite (or sodalite) loaded with fission products and trans-uranium elements and then encapsulated within a nonradioactive borosilicate glass-bonded matrix (also called glass-bonded zeolite, or GBZ) by hot isostatic pressing (HIP).

Waste Form Preparation: Demonstration of Column Ion-Exchange

Scale-up, remote design, and "cold" operation of the column ion-exchange system for the loading of radionuclides into zeolite (or sodalite) are planned in parallel with the EBR-II demonstration. The committee learned, however, that no "hot" zeolite or sodalite will be generated by column ion-exchange. Part of the rationale for this approach is that the preparation of fully loaded "hot" ceramic waste forms must wait until the disposable fission products and transuranic ions have increased to higher levels in the process salt. With the quantities of fuel planned for the demonstration as reduced by the Environmental Assessment (100 driver and 25 blanket assemblies), the radionuclides will not increase to the level that requires use of the column ion-exchange until after the completion of the planned EBR-II demonstration.

The committee remains concerned about whether at the conclusion of the EBR-II demonstration (June 1999) there will be sufficient information available on ion-exchange loading of zeolite or sodalite under radioactive operating conditions and at relevant radionuclide concentrations to provide a basis for a decision to proceed with using electrometallurgical technology to treat spent fuel. In particular, it is worrisome that "hot" column ion-exchange tests will not be part of the EBR-II demonstration and that no fully loaded samples of the expected final ceramic waste form will be produced or characterized. DOE should consider whether the absence of such information will create difficulties in reaching a decision to proceed with this technology after the EBR-II demonstration is completed.

Selection and Characterization of a Reference Crystalline Matrix

ANL plans to select a reference crystalline matrix for incorporation of radionuclides by October 1997. Alternative crystalline host phases being considered include the following:

1. Linde Type-A (LTA) zeolite alone, or LTA zeolite with chabazite (CHA) zeolite, or
2. Zeolite converted to sodalite (a felspathoid).

The conversion of the radionuclide-loaded zeolite to a denser form (e.g., sodalite) will require particular attention to the uniformity of the distribution of the salt between zeolite crystals. Because the denser phase contains less salt, it may be necessary to limit the highest salt loading in each individual zeolite crystal to no greater than the amount that can be contained in the individual sodalite crystals. Otherwise, portions of the salt (and radionuclides) can be expected to reside outside of the sodalite and, possibly, to be more easily leached from the final waste form. ANL should remain alert to issues regarding the scale-up of the manufacturing process for any of these waste forms, especially with regard to the conditions (time, temperature, mixing, etc.) required to achieve an adequate degree of uniformity of salt loading inside each of the crystalline host phases.

Preparation of Ceramic Waste Forms

ANL expects to produce radioactive samples of its ceramic waste form by the end of the EBR-II demonstration period in 1999. It plans to demonstrate all of the key steps for ceramic waste form preparation, including creating radionuclide-loaded zeolite (or sodalite) and HIP of these crystalline phases with glass to manufacture the final waste form. The samples planned are as follows:

1. A Pu-238 loaded zeolite, to be tested to assess potential alpha-recoil damage;

2. A zeolite loaded from the "spent" processing salt of the EBR-II demonstration at ANL-W (the so-called "Throw-Away Option"); and

3. A zeolite batch ion-exchange equilibrated with salt in the electrorefiner, also to be done in parallel with the ANL-W demonstration (the so-called "Batch Equilibration Option").

None of these samples closely matches the composition and radionuclide loadings to be expected in zeolites from actual column ion-exchange operations. Such samples are postulated, however, as providing preliminary and representative information regarding the performance of radionuclide-bearing zeolites that will be prepared in actual column ion-exchange operations after the EBR-II demonstration.

DOE must, therefore, consider the question of whether this approach will provide adequate information to proceed beyond the June 1999 demonstration. Implementation of the "Throw-Away

Option" or "Batch Equilibration Option" as an alternative to column ion-exchange for the preparation of the final ceramic waste form would concern the committee. In particular, these options significantly increase the volumes of radioactive waste forms produced and should be carefully evaluated.

Preparation of Cladding Metal Waste Forms

Small-scale samples of the metal waste form have been fabricated by melting the cladding residues at 1600°C in an inert atmosphere environment and subsequently casting them into ingots. Noble-metal fission products, cladding, and alloying zirconium constituents from the original EBR-II fuel are expected to reside in this metal waste form. Tests to date show satisfactory corrosion, metallurgical, and mechanical properties.[6] Production of "hot" metal waste forms from the demonstration products is scheduled to commence in April 1998 in order to evaluate scale-up issues.

Because of the extremely low corrosion rate observed for the metal waste form (similar to rates for stainless steel and Zircaloy), no formal waste form qualification testing procedures are being developed or applied. While this assumption of waste form acceptability does not appear to be unreasonable, the committee believes that by June 1999, ANL ought to apply its corrosion rate data to the same conceptual water contact and release models that are being developed for the ceramic waste form.

Progress on Waste Form Testing and Qualification

ANL is now implementing a Qualification Testing (QT) program for its proposed ceramic waste form, with less emphasis on qualification testing of its proposed metal waste form. The committee is awaiting ANL's detailed project plan to assess the adequacy and scheduling of QT activities. ANL has indicated that such QT activities are likely to include conceptual model development to identify important performance characteristics of the waste form and evaluation of characteristics under simulated repository disposal conditions.

Conceptual Model for Ceramic Waste Form Performance

ANL has recognized the importance of developing a conceptual model to assess how its proposed ceramic waste form will behave under long-term conditions in a deep geologic repository. ANL's preliminary conceptual model for reaction of the ceramic waste form with repository groundwater and subsequent transport of released radionuclides is divided into three steps:

- Step I: contact of the ceramic waste form with water and/or water vapor;
- Step II: primary release of radionuclides from the ceramic waste form; and

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[6] Argonne National Laboratory, Nuclear Technology, EBR-II Spent Fuel Treatment Program Monthly Report, March 1997.

• Step III: subsequent migration of radionuclides, either into secondary solid phases or as mobile aqueous species (or colloids) undergoing diffusive or advective-diffusive transport.

With respect to Step I, one issue to be resolved is whether the encompassing, nonradioactive glass binder will provide physical isolation of the radionuclide-loaded zeolite/sodalite, limiting the available area of the ceramic contacted by water. Demonstrating physical protection for long periods of time may be difficult and should be based on consideration of the full range of thermal, mechanical, hydrological, and chemical conditions of planned geologic repositories.

In contrast to its physical effects, the presence of the glass binder will undoubtedly have an impact on the chemical reactions occurring between water and the radionuclide-bearing zeolite/sodalite. For example, glass dissolution may be a source of solvated cations that could expedite the subsequent ion-exchange release of radionuclides from the ceramic waste form. Furthermore, water-glass reactions typically lead to increases in pH and alkaline conditions, which could increase the dissolution rate of zeolitic materials.

ANL does not yet have enough QT data on its proposed ceramic waste forms to unambiguously distinguish which release mechanism (Step II)--ion-exchange or dissolution--controls the primary release of radionuclides. Ion-exchange is now acknowledged as one likely mechanism, supported by comparison of the release behavior of nonradioactive ceramic waste forms by ANL using dilute brine contacting waters. ANL plans an extensive set of both long-term and short-term tests (see following section) to attempt to evaluate which primary release mechanism dominates at what time period.

Step III relates to the impact of transport conditions of a repository on the long-term rate of migration of radionuclides released from the waste packages. Information on the expected flow and transport conditions from DOE's high-level waste (HLW) repository program is needed so that reasonable test conditions can be simulated in planned long-term QT of the ceramic waste form.

Qualification Testing Methods

ANL has listed a broad range of planned testing techniques that will form the initial phase of QT for its ceramic waste form. ANL noted that many of the same staff and much of the same analytical equipment necessary to support QT must be shared with parallel activities such as waste form preparation and characterization.

In view of ANL's limited human and facility resources, it would be prudent for ANL to involve external technical experts to review the need for a full set of tests to support QT. There may be needless overlap and redundancy among these proposed tests. Furthermore, alternative unsaturated test methods[7] may provide QT capabilities that are complementary or superior to the vapor-hydration and drip-test methods developed by ANL.

Many of the proposed QT testing methods also specify instructions for post-test analyses of solids and solutions. As part of its QT investigations, ANL ought to examine the necessity and sufficiency of such proposed solids and solution analyses with respect to the unique characteristics of the ANL ceramic waste form.

Summary on Waste Form Qualification

ANL is apparently decoupling its waste form QT program from the criteria for successful demonstration of its electrometallurgical process by June 1999. The committee believes that this separation can be justified only if ANL maintains a sustained, high-level commitment to its proposed QT program up to and after the June 1999 demonstration. It is important that progress on QT with respect to both ceramic and metal waste forms be demonstrated by June 1999. Delays, redirection, or postponements of planned QT activities could have serious and potentially adverse implications for the feasibility of wide application of ANL's electrometallurgical processing technique. Any proposed programmatic changes in waste form qualification should be carefully evaluated by DOE.

DOE should establish acceptance criteria for waste forms scheduled for storage in a geologic repository. Although the final acceptance criteria will have to be approved by the U.S. Nuclear Regulatory Commission and be consistent with the yet-to-be-published repository regulations, ANL's electrometallurgical technology program currently is forced to assume that its own definition will be acceptable. DOE should provide its best guidance to ensure that the Argonne approach is useful.

External Technical Experts

At the current stage of the ANL R&D program, *the committee suggests that ANL utilize external technical experts in specific scientific areas of the program. These technical experts should be recognized for their in-depth knowledge in particular technical areas.* Whereas the National Research Council committee has provided an overall program evaluation and policy recommendations, the proposed experts would examine technical details of the R&D. They would address such issues as:

- process and equipment flow sheets,
- materials of construction,
- equipment operability,

[7] Examples include unsaturated flow simulation using ultracentrifugation (Conca et al., 1997) and the partially unsaturated flow (PUF) test (McGrail et al., 1997).

- nuclide loading of zeolites,
- waste forms' acceptability and costs, and
- testing and performance assessment of waste forms.

The essential elements of the process are that it be carried out by experts in the requisite fields, be independent, and be external to the process developers. Such experts could also assist ANL in defining the adequacy of the EBR-II demonstration criteria.

The committee also suggests active interaction with the international research groups engaged in similar work (e.g., on issues of zeolite technology, characterization of materials, and qualification testing). Currently, ANL researchers' interactions occur at professional meetings and scientific conferences. The committee suggests more formal and intensive interactions with experts in particular fields to maintain the highest level of science in the program.

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Appendix A: Meeting Agendas

Committee Meeting
National Academy of Sciences
Washington, D.C. 20418
April 7-8, 1997

Agenda

Monday, April 7, 1997

8:00 a.m.	Breakfast
8:30	Introductory Business • Preliminary Discussion of Committee Activities

Open Session

10:00	Introductory Remarks and Background Information • Welcome and Introductions (Gregory R. Choppin) • William D. Magwood, Department of Energy • Sterling Franks/Dan Funk, Department of Energy
10:30	Status of ANL R&D Activity on the Electrometallurgical Technology for DOE Spent Fuel Treatment • James J. Laidler, Argonne National Laboratory • Yoon Chang, Argonne National Laboratory • Robert W. Benedict, Argonne National Laboratory
12:00 p.m	Other Perspectives on the Electrometallurgical Technology Program
12:15	Lunch

Executive Session

1:00	Committee Discussions
5:00	Adjourn
6:00	Committee Dinner

Tuesday, April 8, 1997

8:00 a.m.	Breakfast
8:30	Committee Discussions
12:00 p.m.	Lunch
1:00	Discussion of Key Issues
2:00	Future Meeting Plans
3:00	Adjourn

Committee Meeting
Argonne National Laboratory - East Site Visit
May 15-16, 1997

Agenda

Thursday, May 15, 1997 - Argonne National Laboratory - East Site Visit

7:15 a.m.	Meet in Hotel Lobby
7:30	Bus Leaves for ANL
	• Continental Breakfast at ANL
8:30 a.m. to	ANL Site Visit and Presentations
5:45 p.m.	EBR-II Spent Fuel Treatment Demonstration Project Plans
	• Robert W. Benedict, Argonne National Laboratory
	Ceramic Waste Development: Status and Plans
	• J. P. Ackerman
	ANL-West Waste Equipment Tests and Plans
	• K. M. Goff
	Waste Testing and Qualifications: Status and Plans
	• D. J. Wronkiewicz
	ANL-West Waste Testing: Status and Plans
	• S. G. Johnson
	EBR-II Demonstration Test Results
	• Robert W. Benedict
	High-Throughput Electrorefining
	• E. C. Gay
	Oxide, Aluminum, and MSRE Treatment Process Development Status
	• C. C. McPheeters
6:30	Return to Hotel
7:00	Working Dinner:
	• Committee Discussion on Site Visit
	• Identification of Topics for Committee Report

Friday, May 16, 1997 - O'Hare Hilton Conference Center (Room 2047)

7:30 a.m.	Breakfast
8:00	New Membership
	Discussion of Report
	Recommendations
	Schedule for Report
	Date for ANL-W Site Visit
3:00 p.m.	Adjourn

Appendix B: Abbreviations Used

The following abbreviations are used in the main text of this report.

ANL ....Argonne National Laboratory
ANL-E ....Argonne National Laboratory-East (Illinois)
ANL-W...............Argonne National Laboratory-West (Idaho)
D&D ....Decommissioning and Decontamination
DOE ...................U.S. Department of Energy
EA.......................Environmental Assessment
EBR-II ....Experimental Breeder Reactor II
EM......................Environmental Management, Office of (DOE)
FCF ....Fuel Conditioning Facility
HIP......................hot isostatic pressing
HLW ....high-level waste
HTER..................High Throughput Electrorefiner
MSRE ....Molten Salt Reactor Experiment
NE.......................Nuclear Energy, Office of (DOE)
NRC ....National Research Council
QT.......................Qualification Testing
R&D ....research and development
WBS ....Work Breakdown Structure

Appendix C: Project History

The following discussion of the ANL R&D program and its relationship to treatment of EBR-II spent fuel provides background information to clarify the committee's approach to these independent but related activities.

The ANL R&D program under review by the Committee on Electrometallurgical Techniques for DOE Spent Fuel Treatment is considering techniques for the treatment of various DOE spent fuels for ultimate disposal using an electrometallurgical process. The 5-year program funded by DOE is intended to yield sufficient information to permit DOE to select by 1999 a fuel treatment technology to treat its inventory of spent fuels. It is estimated that more than 80% of that inventory consists of metallic spent fuel from EBR-II and oxide spent fuel from the N-Reactor at Hanford.

The R&D program on electrometallurgical techniques for processing of spent fuel is being directed by ANL-E. The technology utilizes the processes developed for the ANL Integral Fast Reactor (IFR) concept that included fission and fast reactors together with fuel processing and fabrication operations all on one site. Government and industry rejected the IFR concept, and the terminated activity is no longer a viable option. The portion of the IFR strategy that employed electrometallurgical techniques for the processing of spent fuels is the foundation of the present ANL R&D program that is intended to show that this technology can be successfully applied to treat various DOE spent fuels. This committee is charged with evaluating this ANL R&D effort.

For more than 25 years, EBR-II was successfully and continuously operated as a research reactor primarily by ANL-W in Idaho. That facility was recently shut down, and DOE has authorized program funds for its decommissioning and decontamination (D&D). This independent program of EBR-II cleanup continues to be the principal activity of ANL-W, which this committee does not review.

In the evolution of the ANL-E R&D program, some aspects of its theoretical analyses were confirmed by laboratory and bench-scale experiments, while others remained unanswered. Because of the nexus between the two ANL programs, the ANL-E 5-year R&D plan included pilot plant testing of the electrometallurgical concept and demonstration of the process and equipment viability by treating a small portion of the EBR-II spent fuel. The original small portion was recently reduced substantially as defined in DOE's 1996 Environmental Assessment.

The ANL-E R&D program includes experimental verification of the electrometallurgical process. This committee has strongly endorsed this experimental verification of the electrometallurgical process and believes it is necessary if this technology is to be considered by DOE as being a viable option for treatment of its spent fuels. If such verification or demonstration cannot be accomplished successfully in the time frame required by DOE, this committee has recommended that the electrometallurgical process not be considered further.

This committee has sought to keep separate the requirements and activities of the ANL-E R&D from those of the ANL-W D&D program. There are obvious overlaps. Nevertheless, it must be kept in mind that this committee has concluded that the ANL-E R&D program requires experimental proof or demonstration, and it is logical to use EBR-II spent fuel in that pursuit while acknowledging that the D&D of EBR-II does not require a successful electrometallurgical technology since other cleanup options may be available.

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