# External Assessment of the U.S. Sodium-Bonded Spent Fuel Treatment Program

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Abstract. Some advocates of electrometallurgical reprocessing (or "pyroprocessing") of spent nuclear fuel argue that the technology has many advantages relative to aqueous reprocessing methods, including cost savings, safety benefits, and increased proliferation resistance. However, to date there has been very little actual operating experience with production-scale electrometallurgical processes, making it difficult to validate these claims. Since 1996, U.S. researchers have been implementing a program to pyroprocess 26 metric tons of sodiumbonded, metallic spent fuel (both driver and blanket) from the shutdown EBR-II and FFTF fast reactors at the Fuel Conditioning Facility at present-day Idaho National Laboratory. In 2000, the U.S. Department of Energy (DOE) asserted that the campaign would be completed within a decade. However, as of October 2016, only about 15% of the inventory had been processed, and it appears likely that several more decades will be needed to finish the job. DOE has released very little information to the public on the program and the reasons why it is experiencing such severe delays. Documents recently obtained by the Union of Concerned Scientists (UCS) under the U.S. Freedom of Information Act shed light on the operational issues of pyroprocessing technology that have contributed to the problems experienced during the campaign. It is apparent from this information that the technology is neither as efficient nor as clean as some claim. The Republic of Korea and other nations that have expressed a deep interest in the development of pyroprocessing technology would be well-advised to take note of the formidable challenges associated with its practical implementation.

Key Words: sodium-bonded, EBR-II, pyroprocessing.

#### 1. Introduction

The metallic-fueled, liquid-metal cooled fast reactor and its associated fuel cycle have been subjects of intense interest for United States researchers for decades. The Experimental Breeder Reactor-II (EBR-II), a fast spectrum research and test reactor with a maximum power rating of 62.5 MWth at Argonne National Laboratory-West (now part of the Idaho National Laboratory), was in operation from 1961 to 1994. Many different metallic fuel types were tested over the course of EBR-II operation.

Since the mid-1960s, and coincident with their development of metal fuels for fast reactors, Argonne National Laboratory (ANL) researchers also conducted studies on electrometallurgical approaches for reprocessing the spent fuel (commonly known as pyroprocessing). Pyroprocessing entailed the dissolution and separation of metallic spent fuel in a molten salt bath and the extraction of uranium and transuranic elements on cathodes.

In 1983, ANL proposed a concept called the Integral Fast Reactor (IFR), which consisted of a sodium-cooled, metal-fueled fast breeder reactor with a co-located pyroprocessing and fuel refabrication facility. As they tried to secure U.S. government funding for the project, Argonne scientists became vocal advocates for the technology, which they argued would be cheaper, more efficient and more proliferation-resistant than aqueous reprocessing. In preparation for pyroprocessing operations as part of the IFR program, a large hot-cell complex called the Fuel Conditioning Facility (FCF), which was adjacent to the EBR-II, underwent extensive refurbishment in the late 1980s-early 1990s.[1] These upgrades included installation of fuel processing equipment and enhancements to the critically important in-cell cranes used for remote handling. The FCF contained two engineering-scale electrorefiners, the Mark-IV and Mark-V, which were meant to be used concurrently.

Ultimately, the IFR project was cancelled and the EBR-II was shutdown in 1994. However, the reactor left a legacy in its wake: a substantial inventory of spent fuel containing sodium, a reactive metal that some argued could not be disposed of in a geologic repository for spent fuel. In 2000, DOE decided to move forward with pyroprocessing of the EBR-II spent fuel as a method of waste treatment that would remove and neutralize the sodium and would separate uranium from fission products. However, the campaign has proceeded far more slowly than originally projected.

Pyroprocessing continues to be championed as a breakthrough technology by ANL, the Republic of Korea, and other passionate advocates around the world. However, when one examines the actual experience with pyroprocessing as it has been used to treat the Department of Energy's sodium-bonded metal spent fuel inventory, the picture is a lot less rosy. Despite decades of development by ANL and other DOE national laboratories, the results of the pyroprocessing campaign have fallen far short of expectations. The technology has turned out to be more complicated and challenging in practice than the idealized image put forth by its promoters, and it is becoming apparent that the technology is not ready for production-scale deployment. DOE has made very little information about the project available to the public over the last sixteen years to explain why the program has been delayed. Recently, the Union of Concerned Scientists (UCS) has obtained information through a Freedom of Information Act (FOIA) request that sheds some light on the problems that have contributed to the failure of this program to meet its goals.

## 2. Pyroprocessing Demonstration Project

After the IFR program was cancelled in 1994 and the EBR-II was shut down, about 26 metric tons (MT) of spent driver and blanket fuel remained. At the time, the standard EBR-II driver fuel element was an alloy of metallic highly enriched uranium and zirconium, clad in stainless steel, which was the design chosen for development for the IFR program. (Some driver elements contained "fissium," an alloy of zirconium and other metals.) EBR-II blanket fuel elements consisted of depleted uranium metal, also clad in stainless steel. Both the driver and blanket fuel elements contain metallic sodium that was used to improve heat transfer across the fuel-cladding gap.

DOE, under pressure by ANL and its supporters in Congress, began to evaluate whether it could use the equipment installed in the FCF to pyroprocess the sodium-bonded spent fuel from the EBR-II, as well as from the legacy spent fuel from other experimental fast reactors (including the Fast Flux Test Facility and Fermi Unit 1). The primary argument for processing the spent fuel was that the bonding layer of metallic sodium between the fuel and the cladding, a reactive material, rendered the spent fuel unsuitable for direct disposal in the Yucca Mountain geologic repository. However, a significant underlying motivation was to find another mission that could keep the pyroprocessing program at ANL alive.

At the time, DOE asked the National Academy of Sciences to review the technical viability of using pyroprocessing to treat DOE-owned spent fuel, including the sodium-bonded spent fuel inventory. Eventually, the study focused on evaluation of a three-year project that ANL initiated in 1996 to demonstrate the technology for both driver and blanket fuel from EBR-II. The original demonstration project goal, processing 200 kg of spent fuel per day continuously for 30 days, turned out to be wildly unrealistic and was abandoned. In its place, in 1998 the NAS approved a criterion that "125 EBR-II assemblies can be treated in a fuel-conditioning

facility (FCF) within 3 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 per month sustained for 1 month."[2] This goal represented an average rate more than 30 times lower than the original goal. It was understood that the 125 assemblies comprised 100 driver assemblies, with a total mass of about 400 kgHM, and 25 driver assemblies, with a total mass of about 1200 kgHM.

The demonstration project, as defined by the NAS, took place from June 1996 to June 1999.<sup>1</sup> The demonstration failed to meet the 125-assembly processing goal criterion approved by the NAS in 1998. Only 105 EBR-II assemblies—100 driver and 5 blanket—were processed in this time.<sup>3</sup> Yet when the NAS issued its final report in 2000 on the demonstration project, it stated that "ANL has met all of the criteria developed for judging the success" of the project (NAS 2000). How was this possible?

As pointed out in a letter sent by the Nuclear Control Institute to the NAS in 2000, this was possible only because the NAS changed the relevant demonstration project success criterion mid-stream.[4] Sometime in 1999, as it became clear that the project was far from achieving its goal of processing 25 blanket assemblies within 3 years, DOE suggested changing the success criterion from showing that "125 EBR-II assemblies can be treated ... within 3 years" to "100 driver and *up to* 25 blanket ... assemblies can be treated ... within 3 years ...," [emphasis added]. The NAS committee agreed to the change, in spite of the fact that inserting the phrase "up to" would render the goal essentially meaningless. According to the new criterion, processing any number of blanket assemblies fewer than 25, including zero, would then be judged a success. The acceptance of this change by the NAS committee was highly improper and ranks among the worst violations of scientific integrity that this author has encountered during his career. In fact, if the NAS instead had taken note of ANL's inability to meet the original processing goal of the demonstration program, it might have foreseen the major problems that lay ahead for the production-scale program, and advised caution.

#### 3. Environmental Impact Statement and Sodium-Bonded Spent Fuel Pyroprocessing Campaign

Based on the flawed NAS assessment that the demonstration program was a success and that there were no significant technical barriers to the use of pyroprocessing to treat all EBR-II spent fuel, DOE moved forward with a plan to use the pyroprocessing equipment at INL to treat the remaining EBR-II spent fuel inventory, including 3.1 metric tons heavy metal (MTHM) of driver and 22.4 MTHM of blanket fuel, as well as some addition miscellaneous sodium-bonded spent fuel, including 250 kgHM from the Fast Flux Test Facility (FFTF). To address non-proliferation concerns, DOE decided to modify the pyroprocessing flowsheet to eliminate the group extraction of transuranic elements onto a liquid cadmium cathode. The transuranics would remain largely dissolved in the molten chloride salt. The highly enriched uranium to produce a low-enriched uranium product. The salt would eventually be stabilized for geologic disposal in a ceramic waste form. A metal waste form containing zirconium and noble metal fission products would also be produced.

<sup>&</sup>lt;sup>1</sup> There appears to have been a difference in interpretation regarding the length of the demonstration program. The NAS interpreted it to be from June 1996-June 1999, consistent with a three-year period. However, ANL has interpreted it as lasting from June 1996-August 1999, over which time 13 blanket assemblies were pyroprocessed.

In the July 2000 *Final Environmental Impact Statement for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel* (FEIS), DOE projected that the driver fuel would be reprocessed at a rate of 600 kgHM per year and the blanket fuel would be processed at a rate of 4,400 kgHM per year beginning as soon as 2000.[5] At this rate, totaling 5,000 kgHM per year, the total spent fuel inventory would be reprocessed in 6 years, and be completed by 2006-2007.

The time to complete this project was critically important because of the 1995 Idaho Settlement Agreement, in which DOE committed to removing all spent fuel from the State of Idaho by 2035 and to treating all high-level waste at the INL, in preparation for final disposal elsewhere, by a target date of 2035. This could be interpreted as allowing pyroprocessing of sodium-bonded spent fuel, provided that the resulting high-level waste would be converted to a final disposal form suitable for transport out of the state by 2035. DOE's analysis indicated that its preferred alternative could convert all the sodium-bonded spent fuel in Idaho to a form suitable for geologic disposal well before the 2035 date. DOE estimated that the other alternatives, such as mechanical removal of the sodium from the blanket fuel, would have taken longer than pyroprocessing, although they still could have been completed before 2035.

However, DOE assumed an average monthly processing rate in the FEIS that was more than three times greater than the rate that had been achieved for the driver fuel, and more than twice that achieved for the blanket fuel, yet it provided no justification for these assumptions. And in fact, documents released under FOIA indicate that DOE knew from the beginning that these rates represented a long-term goal and not a realistic near-term throughput for the process. The non-public ANL Spent Fuel Treatment Implementation Plan, dated October 2000, reveals that the throughput rate for the process in its existing configuration was 2,200 kg of heavy metal per year, if the facility were operated on an unrealistic 24 hours per day, 7 day per week schedule.[6] When operated on a normal 8 hour per day, 5 day per week schedule, the throughput would only be 600 kg heavy metal per year. In order to achieve the 5,000 kgHM per year rate assumed in the FEIS, INL not only would have to operate the facility 24 hours a day, 7 days a week, but it also would have to implement major process improvements and equipment upgrades. The implementation plan assumed that all activities needed to increase the throughput to production-scale rates would be entirely successful. In other words, the FEIS reprocessing schedule was based on attaining a processing rate from the beginning that DOE knew could not be achieved for years, if ever, and would also require running the facility nonstop.

The ANL treatment implementation plan provided two schedules that were both significantly different from the FEIS timeline. The first assumed that the throughput would ramp up from 600 kgHM per year to 5,000 kgHM per year by FY 2005, resulting in completion of the 24.75 MTHM inventory by FY 2010 for a total unescalated cost of \$435 million (or an average of \$17,750 per kg HM). The second assumed that the peak throughput achieved was 3,500 kgHM per year in FY 2007, resulting in completion of the campaign by FY 2013 for an unescalated cost of \$537 million (or an average of \$21,900 per kgHM).

Ultimately, neither the FEIS schedule nor the ANL implementation plan schedules came anywhere near the mark. The highest total throughput that was ever achieved was in FY 1999, when 114 kg of driver fuel and 600 kg of blanket fuel was pyroprocessed. The annual quantity pyroprocessed sank to lows of 0 kgHM in FY 2012 and 15 kgHM in FY 2013. By the end of FY 2016 (the latest date for which we have data), less than 4.9 MTHM (including

219 kg of FFTF fuel) had been pyroprocessed (at an average rate of 240 kgHM per year), leaving nearly 21 MTHM, or 85% of the initial quantity, still to go. Even assuming the maximum throughputs for driver and blanket fuel that were historically achieved, and simultaneous pyroprocessing of both driver and blanket fuel, it would take about 14 years to finish the driver fuel and 31 years to finish the blanket fuel, extending the program's completion to 2047. This would be 12 years after the Idaho Settlement Agreement deadline. INL internal documents confirm that in 2014 it was aware that at the current pyroprocessing rate of only 500 kgHM per year, it would take 44 years (that is, to 2058) to treat the remaining fuel and therefore that "the production capability of the existing facility will not support the Idaho Settlement Agreement milestone to remove all SNF from the State of Idaho by 2035."[7] Ominously, the document includes a comment that "according to [name redacted] EBR-II elements are not subject to the SA [Settlement Agreement]," a position that is not consistent with public statements on this issue.

How did initial estimates of the duration of the pyroprocessing campaign turn out to be so far off? INL blames a "lack of sufficient funding to support production operations" as the reason for the substantial lengthening of the time frame for the project.[8] However, although there is no doubt that DOE failed to provide the funding levels that had been specified as necessary to support the Implementation Plan activities, this is far from the only reason. A review of the records implicates a variety of factors: an overly optimistic assessment of the capabilities of the installed equipment from the outset, vagaries in programmatic direction and funding, a failure to successfully carry out modifications that would have increased throughput, and frequent outages due to a wide variety of equipment failures and other reasons (such as discoveries of flaws in safety analyses). The manipulators used for remote-handling operations were particularly unreliable and required frequent repairs.

For one thing, the projected throughputs were unrealistic from the start. The 1996-1999 demonstration project, upon which the NAS based its judgment that pyroprocessing could be successfully implemented for all the sodium-bonded spent fuel, utilized a 7 day per week, 12 hour per day (84 hour per week) work schedule. The schedule was ramped up even more for a short time in order to demonstrate the goal throughputs of 19 kgHM per month for three months for the driver fuel and 164 kgHM in one month for the blanket fuel. It is now clear that these rates required a surge to 7 day per week, 24 hour per day operation, with a 100% capacity factor, based on the Implementation Plan's statement that the current configuration for such operation yielded a throughput of 2,200 kgHM per year (or 183 kgHM per month). (This is also confirmed by a monthly progress report provided by INL that stated that "the rate achieved during the one month throughput test during the demonstration [for the blanket fuel] was approximately 210 g/hr." The total quantity pyroprocessed during the month was over 150 kgHM, which could only have been possible if the process were run nonstop.)

However, immediately after the demonstration project ended in 2000, DOE cut staffing to allow only a normal 40-hour work week.[9] Thus the throughput objectives of the demonstration were only met by utilizing a schedule that would be completely unachievable for a production-scale program. Even if DOE had provided the funding necessary to staff the FCF round the clock, the schedule left no downtime for maintenance, which would be unworkable for any nuclear facility, not to mention one that had been plagued with significant equipment failures. And even if DOE had provided full funding for the facility upgrades needed to more than double its throughput to (5,000 kgHM per year within a few years), there was no assurance that those upgrades would work, given the numerous problems that were

encountered with the existing setup (and despite the Implementation Plan's assumption that they would be entirely successful).

DOE has not been helpful in trying to clarify the actual performance of the FCF relative to initial estimates. In October 2003, it sent a report to Congress that revealed to the public that all was not well within the pyroprocessing project at INL. It proposed a new baseline plan that "represented a realignment of the schedule detailed in the initial SFT program plan that was backed by the Record of Decision."[10] The new plan would focus on pyroprocessing the remaining driver fuel while considering non-pyroprocessing alternatives for the blanket fuel. However, the report provided erroneous information to Congress by stating that the driver fuel could be pyroprocessed at a rate of 200 kgHM per year on a 40-hour per week schedule, so that the remaining 2.7 MTHM could be treated in 14 years, or by FY 2017. In reality, only 26 kgHM of driver fuel had been pyroprocessed in FY 2003. Later documentation (the 2007 INL Preferred Disposition Plan) indicates that the typical rate of driver fuel pyroprocessing was around 108 kgHM for a 40-hour week.

For instance, in FY 2001, no driver fuel was pyroprocessed because flaws in the anode baskets in the Mark-IV electrorefiner were discovered, and the baskets had to be redesigned. Therefore, the goal that had been set for pyroprocessing 600 kgHM in FY 2001 had to be met solely by treating blanket fuel in the Mark V electrorefiner. Ultimately, only 575 kgHM was pyroprocessed that year, 25 kgHM short of the goal, despite efforts to increase the Mark-V throughput.

## 4. Waste Treatment

The FOIA documents obtained by UCS have revealed another obstacle to meeting the goals for disposal of the EBR-II spent fuel, including the terms of the Idaho Settlement Agreement: the fact that INL does not currently have available hot cell space for the equipment needed to produce the ceramic waste form containing the fission products and transuranic elements separated from the uranium.[11] The space that was originally designated for this equipment is currently occupied by equipment being used for oxide fuel pyroprocessing research and development as part of the U.S-Republic of Korea Joint Fuel Cycle Study. This space was made available by removing the equipment that had previously been used for ceramic waste form development. While DOE still maintains that it intends to install the ceramic waste form equipment when it will be needed, there is increased uncertainty regarding the qualification, production and disposition of the ceramic waste.

## 5. Conclusions

The bottom line is that unless an average increase in throughput of at least a factor of two is achieved soon, INL's pyroprocessing project is likely to fail to meet DOE's commitments under NEPA and to the state of Idaho under the Settlement Agreement.

It is also important to note that the pyroprocessing process flowsheet that INL has attempted to execute, as complex and difficult as it is, is far simpler than and not representative of the process that would be needed for pyroprocessing spent fuel for the purpose of producing new fuel for a metal-fueled fast reactor. The driver fuel that has been pyroprocessed was composed largely of highly-enriched uranium, a completely different composition than the driver fuel of an IFR, which would contain approximately 20% plutonium and other actinides. Also, the current INL process does not involve extracting plutonium and other actinides from

the molten salt and refabrication of fresh fuel. And it does not involve the additional reduction steps that would be necessary to convert oxide spent nuclear fuel from today's commercial power reactors to a metallic form suitable for pyroprocessing.

The lessons learned from this experience are highly relevant in assessing claims that pyroprocessing technology is ready for commercial development. First of all, this demonstrates that developing a new reprocessing technology is not quick, easy or cheap. It requires sustained multi-decade investments in a program that will necessarily involve much trial-and-error.

## 6. References

- [1] GOFF, K.M., et al., "Electrochemical Processing of Used Nuclear Fuel," Nucl. Eng. Tech. **43** (2011) 335.
- [2] NATIONAL RESEARCH COUNCIL, NATIONAL ACADEMY OF SCIENCES, Electrometallurgical Techniques for DOE Spent Fuel Treatment: Final Report, Washington, DC (2000), p. 72.
- [3] NATIONAL RESEARCH COUNCIL, ibid, p 86.
- [4] LYMAN, E. AND LEVENTHAL, P., Letter to Bruce Alberts, President, United States National Academy of Sciences, April 28, 2000.
- [5] UNITED STATES DEPARTMENT OF ENERGY, Final Environmental Impact Statement for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel, DOE/EIS-0306 (2000).
- [6] ARGONNE NATIONAL LABORATORY, Spent Fuel Treatment Implementation Plan, F0000-0061-ES-00 (2000).
- [7] IDAHO NATIONAL LABORATORY, INL Environmental Liabilities Register, February 18, 2016.
- [8] SIMPSON, M.F., Developments of Spent Nuclear Fuel Pyroprocessing Technology at Idaho National Laboratory, INL/EXT-12-25154 (2012).
- [9] IDAHO NATIONAL LABORATORY, Idaho National Laboratory Preferred Disposition Plan for Sodium-Bonded Spent Nuclear Fuel (2007).
- [10] UNITED STATES DEPARTMENT OF ENERGY, Report on the Preferred Treatment Plan for EBR-II Sodium-Bonded Spent Nuclear Fuel (2003).
- [11] IDAHO NATIONAL LABORATORY, EBR-II Blanket Disposition Alternatives, Technical Evaluation, TEM-10300-1, Revision 3, TEV-2200 (2012).