Conf-910413--

Consolidated Fuel Reprocessing Program

CONF-910413--2

DE91 007847

A NEW LOOK AT ACTINIDE RECYCLE*

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Paper submitted for presentation and for
publication in the "Conference Proceedings"
at the Third International Conference on
Nuclear Fuel Reprocessing and Waste Management
RECOD '91

April 14-18, 1991
Sendai, Japan

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*Research sponsored by the Office of Facilities, Fuel Cycle, and Test Programs, Department of Energy under Contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc.
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ABSTRACT

Over the past few years, worldwide interest has been revived in examining and assessing the value of recovering the minor actinides and certain fission products, in addition to plutonium, from spent light-water reactor (LWR) fuel and transmuting them in fast reactors, or by other means. The principal objective is perceived to be recovery of the actinides to reduce hazards of the wastes to be emplaced in geologic repositories. Other potential objectives are recovery of long-lived fission products that may be important hazards in the repository at some time and recovery of valuable noble metals for their commercial value. Obviously, where countries have planned a direct disposal of spent fuel, another consideration is long-term resource conservation (fissile plutonium).

This paper will address the justification for this reexamination and describe some of the technical progress that has been made since the major studies of a decade ago. During this time, the U.S. Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission have begun establishing detailed criteria and regulations for geologic repositories. An examination of the hazards of waste disposal relative to the EPA release standards reveals that removal of 99.9% of the actinides (Pu, Am, and Np) reduces these hazards quite close to the EPA standards after 300 years' decay of the strontium and cesium. It may be also useful to remove and separately manage and dispose of certain of the long-lived fission products, such as ^{99}Tc and ^{129}I. Much additional work is required to fully assess the appropriate target recoveries as the hazards and risks are more closely examined and as the standards are reworked and refined. The two decades before the projected start of the U.S. repository may present a window of opportunity to introduce several better management practices that act to simplify the repository safety issues.

From a technical standpoint, significant progress has been made on recovery of the actinides from aqueous wastes through use of the TRUEX process. Some of the development of that process applicable to waste management problems at Hanford are described in another paper at this conference. Additional work is required to demonstrate the application of the process to spent LWR fuels, but it appears straightforward. In addition, work at the Argonne National Laboratory on the liquid-metal reactor metal fuel cycle, also described in another paper here, shows the relative simplicity of recycle of the actinides in that fast reactor cycle. Much work remains to fully demonstrate that actinides from all secondary waste streams can be removed to the target levels from both the aqueous reprocessing of LWR fuel and the pyro processes for the metal-fueled fast reactor.

Finally, as this reassessment of many issues in the fuel cycle is made, the role of fast reactors, as well as possibly other transmutation options, should be examined to ensure the evolution of an optimum nuclear energy system that is responsive to public concerns.

INTRODUCTION AND BACKGROUND

Over the past few years, worldwide interest, epitomized by Japan's OMEGA Project, has been revived in reexamining "actinide burning." These efforts will assess various potential improvements in waste management. Included in the assessments is the potential value of recovering the minor actinides and certain fission products in addition to plutonium from spent light-water reactor (LWR) fuel. Implicit is the transmutation of the actinides in fast reactors or by other means. The principal objective is perceived to be recovery of the actinides to reduce hazards of the wastes to be disposed of in geologic repositories. Other potential objectives are recovery of long-lived fission products that are important hazards in certain repository sites and recovery of valuable noble metals for their commercial value. We will stress in this paper the view that an assessment of "actinide burning" is in reality a reexamination of a broad range of issues associated with improving the nuclear fuel cycle and the disposal of nuclear wastes, all of which may be crucial in the future of nuclear power. Coincidentally and importantly, is the role recovery of plutonium and other actinides that can play in resource utilization in the long-range use of nuclear power.

The authors of this paper have been involved in the early definitive studies done in the United States a decade ago[1] and have participated in recent initial studies.[2,3,4,5] While the Department of Energy (DOE) is undertaking an assessment of "actinide burning" as related to and in support of the ongoing liquid-metal reactor (LMR) program,[6] at this time, the specific program goals and dimensions of that effort are still being formulated. This paper, representing only the collective perspectives of the authors, will stress the importance of a comprehensive assessment to examine the potential of improving the disposal of nuclear wastes through selective partitioning of certain radionuclides from the wastes before disposal in one or more repositories.

CENTRAL THEME--IS THERE A BETTER FUEL CYCLE?

Over the past 20 years, the U.S. policy on nuclear waste disposal has switched from: (1) the historic "closed" fuel cycle in which plutonium would be recovered and used first in thermal reactors, then fast reactors; (2) to a once-through cycle in which spent fuel would be disposed of directly in deep geologic repositories; and (3) to a period now, in which the principal focus is still on direct disposal, but at a time frame at least 20 years in the future.

The recently announced extension in the repository schedule appears to afford an excellent opportunity for reexamination of several aspects of waste disposal without seriously impeding progress on the ongoing repository program to determine if there might be a better nuclear fuel cycle. Those who argue for this reexamination fully recognize the potential for complicating the present repository program. Few question that wastes should be and will be disposed of eventually in geologic repositories; potentially, removal of actinides and certain other long-lived radionuclides could both simplify long-term waste disposal and provide a highly positive image to the public regarding this important issue. At this stage, the answers are unknown. A substantive and comprehensive program must be carried out if responsible answers are to be found.

OBJECTIVES OF NEW ASSESSMENTS

The initial new looks at these issues have tended to examine the conclusions of the earlier studies[1] and to question why results would be different today. Several points are relevant in this regard. First, the basis for the earlier study was a comparison of two closed LWR fuel cycles, one with removal of actinides and one without, not comparison of a throw-away cycle with a closed cycle. Secondly, all risks associated with the front end of the fuel cycle, mining and milling, were omitted.

And finally, the repository risk assessment was based on a hypothetical site with the assumption that licensing, building, and operating a geologic repository was a straightforward activity. Since that time, significant steps have been taken in quantifying licensing processes and regulations, and the program to site and build a repository has encountered many problems.

Thus, a decade after the major early study, many perspectives are changed. Today, some even question whether nuclear power has a place in the future. Clearly, options for dealing with nuclear wastes must be better understood and considered acceptable before public opinion can be swayed into strong endorsement for nuclear power in the future. New conclusions may be reached from an assessment based on today's picture.

While many views concerning this new look at "actinide burning" are quite similar, all are not in common. We would argue the principal objectives for the U.S. undertaking a new program are as follows:

1. To assess and compare options for the nuclear fuel cycle and nuclear waste disposal, including the present direct disposal option, seeking means to better and acceptable options which could foster a climate for future nuclear power.

2. To carry out broad development programs for relevant recovery and transmutation options in order to resolve the technical issues and understand the costs for such processes.

3. To organize and implement comprehensive systems analyses programs in order to compare the relevant fuel cycle options along with the associated reactor power programs.

4. To directly interface and integrate these efforts with the ongoing repository program with the objective of reaching orderly decisions over the coming decade for the long-term disposal of nuclear wastes in the United States.

Such a program would be appropriately structured in 3- to 4-year phases where tentative decisions might be reached at the end of each phase. Final decisions on what options to choose should not be expected before the end of this decade. Almost certainly such a process and the decisions reached interact, depend on, and affect choices that the country makes for the future of nuclear power. Properly structured, such an effort could mesh well with the ongoing quest for a nuclear future and with the ongoing repository program. Ideally, an orderly long-term option and method for disposal could be chosen as a clearer vision of the role of nuclear power in the future is seen.

IMPORTANT PROBLEMS AND ISSUES TO ADDRESS

This section will attempt only a brief perspective on some of the important issues.

1. Would Removal of Actinides and Certain Fission Products Have Significant and Positive Effects on Geologic Disposal?: While the attributes of a specific repository site must be assessed in detail for definitive answers, some general perspectives can be drawn. A. G. Croff has examined[5] the situation where certain fractions of the actinides are removed, comparing the resulting inventory of radionuclides in the repository with release limits in the emerging Environmental Protection Agency (EPA) standards and criteria. Figure 1 shows that after

decay of the short-lived fission products, removal of 99.9% of the actinides lowers the inventory to within about an order of magnitude of the EPA release limits. As Croff explains, the limits are based on probabilistic assessments, and exact factors cannot be specified. In Figure 2, from Croff's paper also, the contribution of various radionuclides to the overall risk indices is shown. Here, one can see that several long-lived fission products and reactor activation products control. It may be important to remove portions of these special radionuclides also. Removal of more than 99.9% of the actinides appears to be of little value unless certain of those fission products are also removed.

Clearly this question is extremely complex. The original studies showed that water-controlled migration of certain isotopes, principally ^{99}Tc, controlled eventual doses. Studies from the Yucca Mountain site[7] show similar results. All now recognize that the role of some half-dozen radionuclides, in addition to the actinides, must be examined to provide a comprehensive perspective on repository risks. Some view that removal and temporary surface storage of the heat-producing ^{90}Sr and ^{137}Cs might be desirable. Reprocessing affords an opportunity for separation and either special disposal methods or transmutation of these higher-risk nuclides.

2. Are Technical Options Viable for Recovery and Disposition of the Actinides and Other Radionuclides that Require Special Treatment?: This can be answered generally "yes" for the bulk of the nuclides of interest, where methods are fully demonstrated or in an advanced state of development, but with the added caveat that some further development and demonstration at significant scale is required. Section V below addresses the issues specifically in reprocessing/partitioning. No attempt will be made here to elaborate on transmutation questions. Fast reactors are clearly recognized now as the most likely options for transmutation of actinides presuming that in the long-term recycle of the fissile actinides will be a source of needed fuel for future nuclear power plants. Some view accelerator-driven subcritical target assemblies as likely options, possibly only a limited role for transmuting the minor actinides or certain special isotopes like ^{99}Tc.[3]

3. Can Costs be Quantified Now, and Are They Within Acceptable Ranges?: This is another extremely complex issue which should be addressed as an important component of the overall systems assessments recommended. Both reprocessing/partitioning and transmutation components must be addressed. Most experts believe that fast reactors will be somewhat more expensive than present thermal reactors, and critics predict that high reprocessing costs will make plutonium fuel for the fast reactors prohibitive. For example, T. H. Pigford[7] presented a recent view on future reprocessing costs, which we believe is too pessimistic. Mainly in reaction to these views about high reprocessing costs, the DOE, supported in part by Japan through Central Research Institute of Electric Power Industry (CRIEPI), is undertaking a program to examine reprocessing of thermal fuels using pyrometallurgical methods. It will require large resources and considerable time before comparisons between aqueous and pyro methods are definitive. It would seem appropriate to seek some progress in assessing this cost comparison over the next few years.

In our view, we believe the following perspective on costs:

a. When deployed in large numbers, capital costs for fast reactors will be somewhat more expensive than present thermal reactors.

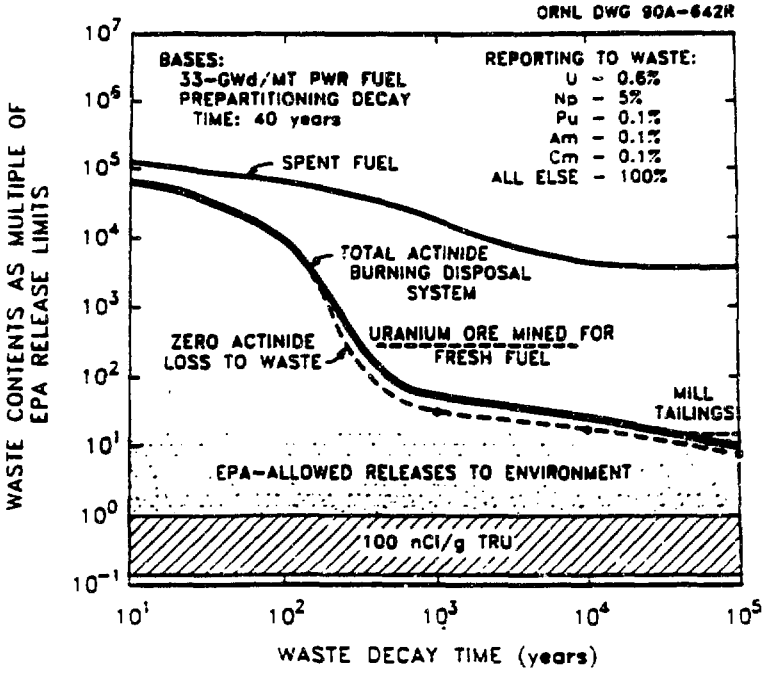

FIGURE 1. Radionuclide inventory of repository relative to EPA disposal standard

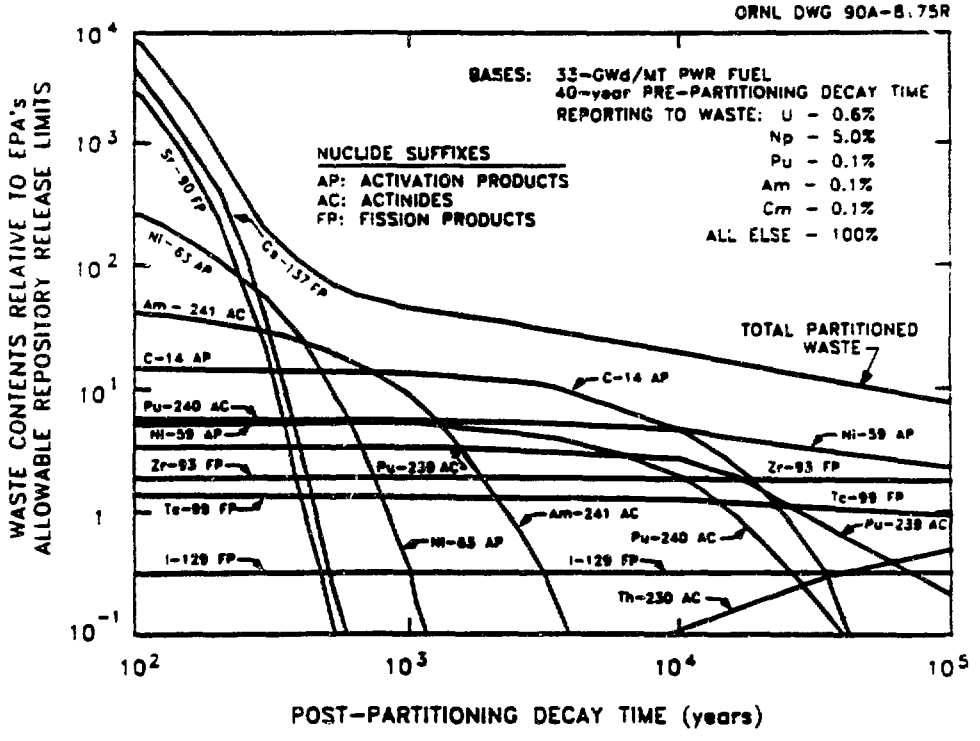

FIGURE 2. Principal radionuclide contributors to repository inventory relative to EPA disposal standard

b. Conventional aqueous reprocessing will become somewhat cheaper in the future, possibly in the range of 50 to 70% of the costs experienced by the new large plants in France and in the United Kingdom. R. H. Allardice recently reported[8] on British Nuclear Fuels Limited, plc (BNFL) plans for new research and development programs seeking to improve and make reprocessing cheaper. While some still believe that reprocessing costs are continuing to escalate rapidly, a recent BNFL study reported that costs for THORP had increased only 12%, in addition to escalation due to inflation in the period from 1973 when the original design and cost estimate were prepared to final costs as known today.

c. Incremental costs for removing the minor actinides should not add more than 10% to the base cost of reprocessing.

d. The authors of this paper believe that it is unlikely that alternate methods for reprocessing thermal oxide fuels, including the pyro methods being examined by the Argonne National Laboratory (ANL) in the DOE/CRIEPI-sponsored program, will be less costly than present known aqueous methods.

e. Overall, economics will not be the controlling factor as to whether actinide recovery is done. Ultimate decisions will be based more on such factors as the future role of nuclear power where long-term use will dictate recycle and the assessment and perceptions as to whether removal of actinides aids in waste management.

STATUS OF TECHNOLOGY

Aqueous-based processes which are relatively well developed for oxide fuels can be used for the principal separations of actinides and certain fisssion products from spent LWR fuel. This section will summarize briefly the status of such processes and point out where additional development is needed.

Conventional Reprocessing Steps

Shearing methods, followed by nitric acid dissolution, are used to prepare solvent extraction feed for the classic PUREX process. Most of the ^{129}I, which is a significant risk in some very long-term scenarios, is volatilized and recovered during dissolution. The ^{14}C is also volatilized and may be removed from the dissolver off-gas. PUREX is then used to recover U and Pu. Flowsheet conditions are adjusted to drive Np to the high-level waste (HLW) along with the other minor actindes, which will subsequently be removed using the TRUEX process. The ^{99}Tc is also driven to the HLW.

TRUEX Process

The recently developed TRUEX solvent extraction process[9] is used to extract the minor actinides along with the fission product lanthanides, the residual Pu and U from the acidic HLW using octyl (phenyl),N,N,diisobutylcarbomolymethylphosphine oxide (CMPO) diluted in tributyl phosphate and a suitable hydrocarbon. Commercially available, CMPO has been shown to be satisfactorily resistant to both hydrolysis and radiolysis. Experience has shown that the solvent will extract nearly quantitatively all actinides from any HNO_3 solution. The plus 3 actinides, along with the lanthanides, readily strip into dilute nitric acid. By use of suitable reagents, Am and Cm can be partitioned from Pu, Np, and U. Tc closely follows U and can be co-stripped into $(NH_4)_2CO_3$, which

simultaneously removed CMPO degradation products. A primary amine solvent extraction process[2] also could be used for separation of U and Tc from either the PUREX uranium product or the U-Tc stream from TRUEX.

While much of the TRUEX development has been done with fission-product-free feeds, some batch extraction experiments, done by J. L. Swanson at PNL,[2] have confirmed the applicability to spent fuel. Swanson carried out batch extractions on dissolved 7-year old commercial spent fuel, using first PUREX, then TRUEX. The final product contained only 2 nCi/gram, and a DF of 4.5×10^5 was achieved from TRUEX feed to raffinate. These early results are very encouraging, but much additional work is needed both at laboratory and larger scale to fully understand and demonstrate this process.

Other Processes

To produce suitably-purified actinide fraction for reactor fuel existing ion-exchange methods or, perhaps, some new yet-to-be-developed method would be used to separate the lanthanides from the actinides. The actinides could be prepared as separate transmutation targets or recombined with the plutonium product as dictated by other technical or proliferation considerations.

If required to reduce heat content in repository waste packages, ^{90}Sr and ^{137}Cs may be removed by processes which have been fully developed and used in the past. The ANL has made recent progress developing an improved ^{90}Sr extraction process, and they are also developing a method of partitioning ^{137}Cs from acidic HLW.

The flowsheet is designed to minimize the amount of secondary waste streams. Secondary leaching of the hulls is probably required. Depending on the recovery factors needed, and the ultimate acceptance criteria for waste packages, these secondary treatment processes may be very complex or quite simple. Until recovery criteria are determined and other attributes are assessed, it is not possible to specify the specific streams nor the exact treatment needed.

Needed Technology Development

While work to date has either fully demonstrated or indicates the technical feasibility of the processes described to meet perceived recovery criteria, additional development and demonstration are required. The following are the most important technical needs:

1. Development/demonstration of TRUEX using PUREX HLW.
2. Separation of Tc from U.
3. Separation of actinides from lanthanides.
4. Separation of Sr and Cs from PUREX HLW.
5. Secondary waste streams treatment.

RELATION TO LMR PROGRAM IN THE UNITED STATES

Since another paper at this conference will describe in more detail the relationship of the emerging assessment of "actinide burning" in the United States to the ongoing LMR program, this

will only be noted briefly here. The ANL and General Electric have begun ongoing assessments of "actinide burning" and the issue of fissile fuel for start-up of LMRs. For a long time, the U.S. LMR program placed limited priority on the fuel cycle and on interactions between the LWR fuel cycle and the potential deployment of LMRs. Thus, this new effort is a positive step in the United States to now examine appropriate methods of providing fissile material for LMRs and to couple the two fuel cycles. "Actinide burning" is opening into a broader framework in the United States as DOE recognizes and addresses the interfacing issues with the repository program as well as with related issues associated with managing the existing wastes at production sites.

IMPORTANCE OF UNDERTAKING THIS ASSESSMENT

As J. A. Rawlins recently pointed out[3], the future course of nuclear power will impact the assessment of the potential role for "actinide burning" in the United States. Should a decision be made to phase out nuclear power, the incentives for "actinide burning" may be marginal. However, there still might be a role in transmuting the existing actinides. For that limited role, accelerators might be the option of choice for transmutation. If nuclear power continues at approximately the present level, thermal reactors with direct disposal could still be utilized for a long time without recycle of actinide fissile material. On the other hand, if nuclear power plays an increasingly more important role, then recycle of Pu will ultimately be required. With that scenario, it is considerably simpler to also implement recovery and transmutation of the minor actinides and recovery and special handling of other important fission and activation products. Thus, these studies must address a spectrum of future nuclear scenarios, in addition to the specific pros and cons of improving waste management through "actinide burning" and related fuel cycle options that deal with other special radionuclides.

We strongly endorse a major undertaking in the United States, coupled to and working closely with similar programs elsewhere in the world to reexamine a broad range of options for dealing with the nuclear fuel cycle. Included with the options will be "actinide burning," but the studies should look well beyond just that limited option. The technical community now recognizes the depth of the issues with nuclear waste disposal. Examining and assessing all credible options is mandatory. Answers are unknown. Many things can change over the coming decade as this assessment is done. Nothing less than a broad, comprehensive program to examine these issues is acceptable.

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