PYROPROCESSING AS A WASTE MANAGEMENT STRATEGY*

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ABSTRACT

Pyroprocessing, as incorporated in the Integral Fast Reactor (IFR) program, is being developed as a resource-efficient means of providing fuel for a reactor which has outstanding safety characteristics and performance potential. This process has been shown to be an effective way to partition uranium, transuranic elements, and fissions products. Being a gross separation process, it is not compatible with production of pure plutonium, but permits the recycle of essentially all the transuranic material to the Integral Fast Reactor for consumption.

For the IFR, stripping processes are available to reduce the transuranic content of the waste to very low levels, offering flexibility in approaches to meeting repository requirements. Effluents from the IFR should be minimal.

The pyroprocess looks so promising, the question naturally arises as to whether this process can be used to recover the IFR startup fuel from spent LWR fuel, rather than to rely on PUREX extraction. Conceptual flowsheets have been developed but not yet demonstrated. Should these processes be shown to be practicable, waste processing techniques similar to those of the IFR should be feasible, providing almost total recovery of the actinide content of the wastes. A natural segregation between the major heat generating elements and the noble metals (including Tc) permits considerable flexibility in high level waste strategy development. Thus, as a reactor development strategy, pyroprocessing offers a thorough and well considered fuel cycle starting from spent LWR fuel, producing power and ending with well characterized, minimum actinide wastes.

Whether this process can also significantly simplify the LWR waste disposal problem depends on the outcome of process development - estimated to take five years, and of the subsequent applications development programs. If successful, this program could influence the ultimate waste loading of the first repository and modify the requirements for a second repository.

BACKGROUND

The perceived public attitude today is that no new nuclear plants should be constructed until a suitable waste management strategy is available. For the once-through fuel cycle, this means the availability of a repository suitable for spent fuel, and the development of a suitable spent fuel disposal container. A properly selected geologic disposal site will certainly provide ample protection for the public, but providing credible evidence to a skeptical public is proving to be challenging.

The bulk of the hazardous radioactive material in these wastes, if properly separated, is high quality nuclear fuel. Extraction (partitioning) and consumption (transmutation or "actinide burning") of radioactive hazardous materials would clearly permit greater confidence and flexibility in geologic disposal of other long lived wastes and would provide fuel for nuclear power plants without the need for mining of uranium.

It is an established fact that a fast neutron spectrum is an effective means of consuming transuranic actinides. In a fast reactor, all of the actinides, including the minor actinides (Np, Am, Cm) are fuels.

AQUEOUS PROCESSING

One technology for extracting plutonium from spent LWR fuel is available and is in commercial use in France and the UK. This (PUREX) reprocessing technology is specific to uranium and plutonium recovery. Reprocessing permits putting the wastes into suitable waste forms, but because of the minor actinides and the typical 1-2% carryover of Pu into the waste stream, the fundamental hazard of the waste, while quantitatively reduced, is qualitatively unchanged. Various technologies for stripping of PUREX wastes have been developed (TRUEX for example), but have not been deployed on an industrial scale.

Many studies and assessments of actinide partitioning and transmutation were carried out in the late 1970's and early 1980's. An extensive bibliography of studies is contained in Ref. 1. In these studies, the partitioning process (based on PUREX reprocessing) was considered to be complicated and expensive, and the geological repositories were presumed to prevent the release of actinides for hundreds of thousands of years. As a result, it was concluded that waste management considerations alone would not

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provide a sufficient economic and safety incentive for partitioning and transmuting wastes (2).

PYROPROCESSING

A variety of recent developments combine to warrant a renewed interest in actinide recycle. The most fundamental factor is that pyroprocessing, as part of the IFR Program, appears to be a simple and effective means for recycling fast reactor fuels, permitting the total consumption of actinides. A unique aspect of this process is that it necessarily recovers an ensemble of all transuranic elements: plutonium, neptunium, americium, and curium. It is also not a "clean" process, in that there is necessarily a substantial uranium content and a partial carry-over of rare earth fission products in the product. The product is both highly radioactive and is unsuitable as a weapons material. Thus, the proliferation and diversion concerns associated with a commerce in separated plutonium are lessened. Further, techniques are available to strip the waste streams of their transuranics, greatly simplifying high level waste considerations. Being a much simpler process than PUREX, it is expected that the process will be substantially less costly than PUREX.

This process has four fundamental waste streams: cladding hulls (and scrap), a noble metal and rare earth waste stream, a chloride salt waste stream, and gaseous effluents. The cladding hulls, noble metal and rare earth waste streams will be combined in a metal (e.g. copper) matrix or alloy. The production of an appropriate waste billet appears straightforward. The gaseous effluents can be extracted from the well controlled cover gas by conventional cryogenic techniques. The chloride salt, after stripping of all actinides (which are returned to the process) will be further treated and converted to a suitable waste form; various waste forms are being evaluated. This stream will contain all of the Cs, Sr, and iodine fission products. A companion paper (3) describes the IFR fuel cycle and its waste streams, and the status of work on waste form development.

LWR PYROPROCESSES

A logical extension to this pyroprocessing work is to establish whether this type of processing can be applied to the extraction of actinides from spent oxide fuels from LWRs or from oxide fuelled LMRs as a means of providing the initial fuelling for IFRs. The processes being considered are compatible with direct production of IFR fuel materials; few new waste streams are introduced. Based on initial laboratory scale work, two apparently suitable pyrochemical processes have been identified, but engineering feasibility has not yet been established. If successful, this technology would permit a full complementarity between LWRs and IFRs, with the LWRs providing the initial fuelling for IFRs which would subsequently operate on a closed fuel cycle, fuelled only with uranium which would also be recovered from spent LWR fuel. If sufficient recovered LWR actinide fuel is not available, the cycle can be started with other available enriched material and fed with natural or depleted uranium.

WASTE IMPLICATIONS

It is evident that extraction of the transuranics would dramatically and fundamentally change the demands that nuclear fuel cycle wastes place on a repository. Without recycle, the once-through nuclear fuel cycle is burdened with a waste that is orders of magnitude more toxic than the original ore. With an actinide decontamination factor of 10^3 to 10^4, as is available from the IFR cycle, waste disposal criteria can focus on appropriate means of assuring control of the Cs and Sr for their much more limited lifetime and of the minor long lived materials (Tc, C-14, I-129). Since there will always be a trace residue of actinides and the current pyroprocesses do not address the long lived fission products Tc-99, I-129, and C-14, it is clear that deep geological disposal of residual high level wastes will be required. The current repository program is also necessary for defense wastes, LWR wastes not ultimately reserved for actinide recovery, and other high level wastes.

A frequently quoted repository risk assessment (ORNL-5566) (2) which showed that Tc-99 and I-129 dominate the long-term risk, rather than actinides, is based on a particular assumed leach incident at the repository. The model used for risk analysis predicts that Tc-99 and I-129 migrate readily whereas actinides are sorbed in the geosphere. The dose rate calculated for Tc is only slightly above the currently projected EPA limit. Thus, only minor retention factors are required. On the other hand, the Pu-239 risk is projected to be low because it has both low solubility and because its sorption characteristics limit its transport. Release fractions over 100,000 years are projected to be 2×10^{-11} per year.

The burden for the repository developers is to provide assurance that these extremely low release fractions remain valid over hundreds of thousands of years, for the actinides placed in the repository. For Tc, only modest improvements are required. If actinides are recycled and burned, the technical requirements on the repository are reduced correspondingly. In addition, in the IFR fuel cycle, Tc will be in an alloy form, providing retention under any assumed pathway analysis.

A flexibility is also available in terms of the repository heat load requirements. Traditional repository designs are limited by the maximum heat burden that the site can handle without damage to the rock (chemistry, structure, and mechanical strength). The strength of the rock and the overlying strata also limits the number and spacing of tunnels and holes that can be mined for emplacing the waste canisters. The most efficient use of the repository would occur when the limit on the total heat generated corresponds to the

structural strength limit of the repository. Since the short term heat load is dominated by the Cs and Sr, actinide separation does little for the repository heat load for immediate disposal. However, if engineered storage for 50 to 100 years is assumed, elimination of the transuranics will significantly reduce the repository heat load.

For the IFR waste streams, the metal waste form (noble metals and cladding hulls) would initially have high heat but short half life. With as little as a 10 year holding period, the heat load will not control its disposition. For the salt stream, the heat loading will be controlled by Cs and Sr; thus, the appropriate strategy can be specifically aligned to their 30 year half life.

STATUS

The IFR fuel cycle has progressed sufficiently to consider that the technical feasibility of the concept has been demonstrated; beginning late in 1991, an industrial scale demonstration will begin, to determine the commercial potential of this concept. The EBR-II reactor is currently fully fuelled with an IFR type fuel, and design studies in the U.S. ALMR program have demonstrated the adaptability of the concept (4, 5). The development of pyrochemical processes for recovery of actinides from LWR fuel is in the development stage and the developers project that technical feasibility should be demonstrated by 1995, with commercial feasibility demonstration requiring perhaps another 10 years. Given that no country is currently planning irrevocable action relative to disposal of spent fuel during this time period and given the adequacy of stocks of spent nuclear fuel, this technology development can proceed in parallel with available reprocessing and with characterization of high level waste disposal sites.

CONCLUSIONS

The ultimate waste management significance of the development of pyroprocessing technologies cannot be assessed at this time. The IFR, as an environmentally more benign fuel cycle, will not dominate, or even particularly influence repository requirements which are set by defense wastes and (currently) by unreprocessed reactor fuel. The recovery of actinides from LWR fuel for the initial fuelling of IFRs would influence repository development only if all LWR fuel was put through this process. However, if the process of extraction of actinides proves to be technically feasible (projected date 1995) and economically attractive (perhaps by about 2005), then it would be appropriate to realign disposal priorities, favoring defense wastes and other unrecoverable wastes, saving the "good" fuel for recycle. While the actinide recycling does not change the need and timing of the first repository, it could alter the total hazard burden of this repository and would be in ample time to have a dramatic influence on the second repository.

The need to make effective use of our natural resources is fundamental; it is only a question of timing as to when it will be necessary to deploy reactors which more efficiently utilize the world's uranium resources. The fundamental technologies, including reprocessing and reactor designs are available but further work is necessary to simplify designs and to reduce costs. Thus, programs to assure that the technology and industrial capability to deploy fast breeders are available should continue. The potential for the IFR fuel cycle to simplify waste disposal argues for aggressive programs to develop and demonstrate pyroprocessing technologies.

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