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Proliferation Resistant Reprocessing Methods

Guna S. Selvaduray
San Jose State University, gunas@email.sjsu.edu

L. Heising

sile uranium (mainly U-233) produced by neutron capture in thorium. This is recycled and successive generations of fuel require smaller amounts of external fissile material until an equilibrium situation is reached. Table 2 shows U-235 requirements for successive generations of a particular cycle.

The fuel burn-up for the successive generations is maintained constant at 29.3MWd/kg highly enriched uranium. After four generations, the U-235 requirements have reached an equilibrium level of a little over 4g/kg highly enriched with the remaining fissile loading coming from the spent fuel of the previous generation. This equilibrium level of external fissile requirements is a function of the desired fuel burn-up as shown in table 3 for U-235-topped thorium cycles[4].

The requirements for three thorium cycles are compared with those for the natural uranium, once-through cycle. The equilibrium U-235 concentration in fresh fuel increases with the desired burn-up. This has also been expressed as the Candu reactor lifetime requirements (assuming a 30-year life at an average load factor of 80 per cent) for natural uranium.

The figures for the thorium cycles were derived on the basis that the natural uranium is fed to an enrichment plant operating at 0.2 per cent tails and the uranium enriched to 93 per cent prior to mixing with the thorium. A tight control of reprocessing and fabrication losses, together with a one-year recycle delay time, is also assumed. Also shown are the thorium requirements assuming a 3 per cent loss during reprocessing, followed by a 10-year hold up period before refabrication.

A significant reduction in uranium requirements can be achieved by using the thorium cycle. Replacement reactors have even lower uranium requirements since a major part of the initial fissile inventory is already available in the spent fuel of the decommissioned reactor.

At present it is difficult to define a unique reference thorium cycle because the range of possible systems is broad and the characteristics would be tailored to the particular circumstances of implementation. High uranium utilization dictates a low burn-up cycle but economic considerations indicate a high burn-up cycle to reduce the impact of the reprocessing and refabrication charges. System studies[5] on the impact of introducing thorium cycles in Canada during the next century indicate that an intermediate burn-up cycle (20-30MWd/kg highly enriched) may be the most appropriate.

Table 4. Fissile inventory required for 1GWe reactor

	Amount (Mg)	Source
Highly enriched uranium (93%)	4.5	871Mg natural uranium ~1800Mg of spent Candu fuel, or ~800Mg of spent LWR fuel
Fissile plutonium	4.9	

Economic evaluation of thorium cycles is uncertain since costs can only be derived for conceptual designs of reprocessing and refabrication facilities which have not yet been demonstrated. Current studies indicate that the fuelling costs of the thorium cycle are greater than those for once-through natural uranium fuelling and will be equal only if the natural uranium price rises to at least $200/kg U.

The self-sufficient equilibrium thorium (SSET) cycle illustrated in table 3 is important from a strategic viewpoint. After an initial fissile inventory has been provided, this cycle offers the possibility of continued energy production fuelled only by thorium. The initial fissile inventory could be highly enriched uranium, plutonium or uranium-233. The quantities required for highly enriched uranium or plutonium initiation are given in table 4, together with the quantities of source material.

It is uncertain at present what burn-up can be obtained consistent with self-sufficiency. The cycle must breed slightly more fissible material than it consumes to compensate for reprocessing and refabrication losses. A more neutron-economic Candu design is possible to meet this requirement[6] but nuclear data are too uncertain to allow an accurate prediction of attainable burn-up. More experimental work is required in this area.

So, to summarize, use of thorium cycles in Candu reactors considerably increases the energy obtainable from a given uranium resource base. It seems that the proven Candu design can be adapted to the thorium cycle with little or no modification. The main developments required before a thorium cycle can be implemented are in the areas of fuel reprocessing and fabrication. The tactical advantages of using thorium fuel in once-through fuel cycles during the introductory phase are being investigated. For the recycle mode, fuel cycle costs are higher than those for uranium once-through systems but likely trends in uranium price could make thorium cycles economically attractive in the next century. The economic viability of the self-sufficient equilibrium thorium cycle is more uncertain but offers the potential of insurance against uranium shortages and an upper limit to nuclear energy costs from Candu.

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Proliferation-resistant reprocessing methods

By Dr Guna S. Selvaduray* and Dr Carolyn D. Heising-Goodman†

Although no reprocessing plant can be made completely diversion proof choice of reprocessing system can help make a plant proliferation resistant. This article examines techniques for deciding which methods most hinder the would-be divertor.

Although there clearly cannot be a technical fix to answer the nuclear proliferation problem, significant differences between reprocessing technologies exist with respect to their relative diversion resistance. Many methods for the reprocessing of light water reactor and/or

*San Jose State University.
†Massachusetts Institute of Technology, Massachusetts.

breeder spent fuel have been shown to be technically feasible. These techniques include CIVEX, AIROX, pyrometallurgical processes such as the tin nitride process, and high-volatility methods.[1] Recently, these methods have gained increased attention in the United States and elsewhere because of concern over the potential for diversion of reactor-grade plutonium from the relatively purified product stream of the PUREX process.[2]

Methods of processing

The PUREX process was developed from military operations in the United States during the 1940s at Hanford. Because PUREX was designed for weapons applications some people have suggested that other methods should be developed for civilian applications.[3] These methods involve either modifications of the PUREX process, such as co-processing,[4] Pu-238 spiking methods[5] and the CIVEX process for breeder fuel,[6] or are based on entirely separate techniques such as salt transport and pyrometallurgy.[7]

In comparing these technologies, it must be remembered that PUREX remains the most technically advanced. So, for other methods to compete successfully, they must not only be shown to appreciably increase diversion resistance but to do so without adverse economic effects.

To assess the relative diversion resistance of the various reprocessing methods, it is useful to establish a set of generic characteristics that define a process that is acceptably diversion resistant. This paper describes two methods for doing this. In a hypothetical diversion attempt, four major steps are involved (as shown in the illustration above). This paper is limited to a discussion of the difficulties of the first three steps in this process.

Diversion resistance characteristics

The purpose of making diversion resistance a design criterion is to minimize the likelihood of a successful diversion of nuclear material. Resistance can be increased either by making it almost impossible to obtain such material, and/or ensuring that the product of the process is not easily convertible to a weapons usable status. The first characteristic can be achieved by strengthening international safeguards, e.g., physical surveillance, installation of detection instruments, etc. The second characteristic is a function of the inherent properties of the reprocessing technology itself.

While safeguards are important, this paper explores the possibility for increasing diversion resistance as a function of this second characteristic. With reference to the illustration it is steps 1 and 3 that most concern us here; step 2 is subject to physical containment while step 4 is outside the scope of this analysis.

Favourable properties

We can define three principal attributes of inherent process diversion resistance: dilution of the weapons usable material in the product-waste streams; physical inability to extract weapons usable material from the streams (or a limited extraction capability); and conversion process and handling difficulties.

A product stream of weapons grade quality is most desirable for would-be divertors. Product streams offering less than weapons grade offer some diversion resistance. Co-processing of plutonium with uranium does not affect the recyclability of the recovered material to power reactors but does increase the amount of additional processing necessary by a potential divertor. Low decontamination factors (of the order of 10^3) not only pose health hazards to divertors but means they need a knowledge of remote handling equipment. A low concentration of weapons usable material means large volumes must be diverted to obtain significant quantities. This would essentially eliminate a "one-shot deal" and statistically increase the probability of detection.

If the piping network is complex a potential divertor needs a more comprehensive knowledge of the process before he can successfully obtain the material. However, it also makes detection by international authorities more difficult; it is easier to monitor and detect accounting errors when there are comparatively few product/effluent streams. So from an "easier-to-detect" viewpoint processes with less complex processing sequences are considered more diversion resistant.

Material that is difficult to handle is less likely to be successfully diverted. High radiation levels (low decontamination factors) affect the handling difficulty. High temperature liquid metals and salts are harder to handle than room temperature aqueous solutions making diversion less likely. Gaseous streams are harder to tap and disperse easily in the event of a loss of containment; this is particularly true for high pressure processes.

Just as important as the factors discussed above is the ease by which the process can be diluted and/or fission product spiked. If diluents and/or spiking agents can be introduced into the process stream diversion can be made extremely difficult and any diverted material can be rendered essentially useless for actual explosive purposes.

Decision analysis techniques

In general, techniques available for selecting between processes can be categorized into two groups; the sieve approach and the ranking approach. The Sieve approach consists of sequential elimination of candidates through successive application of criteria. These criteria are usually defined in rank order as primary, secondary and tertiary criteria. This approach has been widely used by geologists; for example, in the selection of a high-level waste repository.

In the sieve approach, a process continues to be a candidate only if it continues to meet the higher order criterion. It emerges as a potential candidate if and only if it meets *all* the criteria. If in the course of the analysis any one criteria is not met the process is eliminated and the next process analyzed. This method of analysis defines those processes that meet all required criteria but does not give an indication of the relative merits of the processes analyzed.

The ranking approach, developed in the formal discipline of Bayesian decision analysis,[8] allows for a quantitative relative comparison of alternatives. Reprocessing methods can be evaluated for their performance under a set of criteria (or attributes) and provision can be made for difference in importance of each attribute quantified in the analysis.

The principal difference between the two approaches is that while the sieve

Steps in the process of an attempt to divert nuclear materials from a nuclear power fuel facility for explosive purposes.

Step 1	Step 2	Step 3	Step 4
Obtain Material from Product Stream	Transport Material to Laboratory Location	Process and Further Refine material to Weapons Usable Quality	Fabricate Nuclear Explosive

Application of the ranking decision analysis approach to the reprocessing technology selection process.

Reprocessing technology	Development time (year)		Warning period % of task to be completed		Radiation level (R/hr at 1m from source)	Criticality problem level	Development cost (10^6 1975)	Overall ranking	
	Overt*	Covert	Overt	Covert				Overt	Covert
AIROX	1.5	5	90	1-3	6.6	Low	5.5-8	-0.31	-0.23
Halide volatility	0.8	5	80	1-2	10^{3}-10^{4}	Low	4.5-7	-0.24	-0.18
PUREX	0.5	5	<1	1	10^{4}-10^{6}	Medium	2	-0.09	-0.18
Salt transport	0.02†	5	<1	1	10^{3}-10^{4}	Medium	1	-0.12	-0.24
Tin nitride	1.5	5	90	1-3	0.66	Low	5.5-8	-0.31	-0.23

*Overt and covert refer to whether or not a non-weapons state attempts to divert material from the commercial plant either covertly or overtly
†In the salt transport case the plutonium is in the metallic form in one of the process streams in the plant. If diversion occurs at that point no outside processing is necessary to acquire usable material for an explosive. In all other materials, many successive stages are necessary.

approach evaluates a process against a set of criteria applied sequentially, the ranking approach evaluates a process against multiple criteria simultaneously. The ranking approach has the advantage of being able to quantitatively compare alternatives relative to each other while the sieve approach must rely on a more absolute determination of an acceptable alternative.

To demonstrate these techniques, five reprocessing methods have been selected for analysis. These include AIROX, halide volatility, PUREX, salt transport and the tin nitride processes. This is not a complete list of available alternatives, but these five have been chosen to represent the wide range of process types available.

The AIROX[9] process is a low decontamination reprocessing technique that takes the spent fuel through successive oxidation and reduction stages using oxygen and hydrogen. The oxidation step is also used for decladding as UO_2 expands 30 per cent by volume when it changes to U_3O_8. The halide volatility technique converts the spent fuel to the hexafluorides and then separates them by the difference in their boiling points. The great majority of fission products form fluorides which volatilize at high temperatures. Of the fluorides which volatilize more readily than uranium hexafluoride, only tellurium hexafluoride is a gas at ordinary temperatures so it can be separated readily. Uranium hexafluoride boils at 56.2°C and plutonium hexafluoride at 62.3°C so it is easy to separate them.

The PUREX process is the only commercially available reprocessing technique. It is an aqueous solvent extraction technique and is described in detail in several sources[10] and so will not be described here.

The salt transport process was designed for plutonium recovery from LMFBR fuels. Decladding is accomplished by immersing fuel subassemblies in liquid zinc at about 850°C. The uranium oxide and plutonium oxide in the fuel are converted to metal before going through a series of liquid metal-molten salt solvent extractions yielding metallic uranium and plutonium. The tin nitride process involves dissolving the spent fuel in liquid tin followed by selective nitriding of uranium. The fission products either form nitrides or intermetallics both of which float; the uranium nitride precipitates out along with the plutonium and other actinide nitrides.

A sample quantitative analysis

The sieve and ranking methods will now be applied to the five reprocessing technologies described earlier to show how the method works and compare the results. The first, or primary, criterion applied in this example is that plutonium must not be separated out during reprocessing. This criterion is met by only two of the five processes; AIROX and tin nitride. The halide volatility, PUREX and salt transport processes aim at producing a relatively pure stream of plutonium and therefore violate the primary criterion. AIROX and tin nitride, however, not only do not separate out plutonium but *cannot* do so. In fact, the applicability of the oxidation-reduction reactions proposed in the AIROX process for uranium fuels have not been experimentally verified for plutonium.

Both AIROX and tin nitride are low decontamination processes and as such handling is difficult. The process temperatures are also high. The materials in the tin nitride process are probably harder to handle since they are dissolved in molten tin at around 1500°C. The AIROX process, because it uses hydrogen in one of the unit operations, could force the divertor to face explosion risks. Though neither of these processes are extremely complex, they lend themselves to dilution as a means of reducing the concentration of fissile material in the product stream. In the AIROX process, dilution could be affected after decladding is complete and the uranium oxide pellets are pulverized.

The tin nitride process offers easier dilution as it is merely necessary to reduce the input of spent fuel into the molten tin bath at the time of initial dissolution. Spiking agents can also be added at dilution making the fissile material less desirable to the potential divertor. However, increasing the number of spiking agents can adversely effect the reactivity coefficient in a reactor causing significant economic enrichment penalties to compensate for a reduction in reactivity. These penalties are sufficient enough to warrant a careful cost-benefit comparison of process economics with diversion resistance.

From this analysis the AIROX and tin nitride processes are found to be most diversion resistant; both technologies offer handling difficulties due to low decontamination and high process temperatures. The AIROX process poses an explosion danger to the divertor whereas the tin nitride process is more amenable to dilution.

The ranking approach employed to distinguish between the sample reprocessing technologies is that developed by Papazoglu *et al*[11] at Massachusetts Institute of Technology. This method is based on the principles of multi-attribute decision theory wherein a set of indices or attributes which characterize the proliferation resistance of the technologies is defined and evaluated.

For this particular application, a set of five attributes are considered important to a potential divertor's decision to use a given technology to derive weapons material. The first attribute is the development time, or the time it takes from start to finish to develop a nuclear explosive using diverted nuclear material. The warning period, defined as the percentage of the development task left to complete at the time of detection by outside agents, is the second attribute. The third is the inherent difficulty of utilizing the technology as a source of nuclear fissile material, defined further by a breakdown into three sub-attributes

– the radioactivity level of the process, the status of scientific and technical information known about the process by the potential proliferator, and the level of criticality problem associated with the process. The fourth attribute is the weapons material quality, defined as the type of nuclear material diverted (i.e., either weapons or reactor grade plutonium, or enriched uranium (U-233 or U-235)), and the fifth is the development cost of the explosive construction attempt.

Numerical values of resistance

To derive a quantitative indicator of the relative diversion resistance of each technology, a value function for each attribute has been defined so that a dimensionless numerical indicator for each technology can be calculated. The numerical indicators for each attribute are then multiplied by weighting factors and summed over the number of attributes to arrive at a single numerical indicator for each fuel cycle strategy. Basically, the purpose of the value function is to provide a numerical measure of the relative attractiveness of the various proliferation pathways available to the would-be proliferator.

The results of the analysis show, on a scale of −1 to 0, that the AIROX and tin nitride processes are most diversion resistant overall and that PUREX is the least resistant (see the table). These results occur because of the greater development time necessary for explosive construction, the lower status of information concerning the processes and the higher development costs. Also, the higher radiation levels make these processes more diversion resistant. The salt transport process shows good diversion resistance for the covert case, but not for the overt case. Its overall ranking of −0.24 is close enough to that of AIROX and tin nitride (both −0.23) to consider them essentially the same. Although the salt transport process produces metallic plutonium, the medium level criticality problem posed to the potential divertor contributes to its diversion resistance.

However, that the ultimate choice between reprocessing technologies must not be based solely on a single criterion such as diversion resistance but on other important selection criteria as well, such as economics, environmental and safety aspects, is evident. The nuclear industry today is at a cross road and the proliferation issue is only one among many other major concerns. Technologies that do not meet diversion resistance standards need to be clearly identified and alternate technologies developed that can simultaneously optimize all important selection criterion. Other aspects that can be expected to influence the decision include: the effect a choice of an alternate reprocessing technology will have on existing fuel cycle facilities and economics; impact on reactivity and reactor design (e.g., effect on enrichment levels and burnable poison management); waste management implications; and the stage of development of the alternatives.

Research and development decisions must not be based solely on political considerations; they must also be based on sound technical analysis. Utilization of quantitative decision analysis techniques can provide the required firm foundation for technical decisions. Application of these methods compels policy makers to define clearly the criteria upon which they evaluate alternatives and to record their personal preferences and biases towards the importance of each criteria. Equally as important is the mechanism that decision analysis provides for updating the evaluation in the light of new data and experimental evidence. All of these factors suggest that such methods should be more widely applied and accepted in the policy making arena.

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Plutonium processing for the fast reactor fuel cycle

By R. H. Allardice* and H. A. Taylor†

Current Dounreay and Windscale experience in reprocessing, fabrication and plutonium waste operations together with the continuing UK development programme to establish process and plant design for larger scale operations is described in this paper. The necessity for considering these fuel cycle operations as an entity and avoiding the independent optimization of individual areas, in order to obtain the preferred balance for safe and reliable Pu processing operations is emphasized.

Following the decision made in the late 1960s to construct the Prototype Fast Reactor – PFR (600MWth, 250e) on the same site at Dounreay as the Experimental Fast Reactor – DFR (60MWth, 15e), it was decided in 1970/71 to complete the fuel cycle by reprocessing the irradiated fuel in the near term rather than at a later date. (A typical fast breeder reactor fuel cycle is shown in Fig. 1.) This policy was adopted not only to reduce the PFR plutonium inventory and to take the reactor operation through the full fuel recycle stages, but more significantly to demonstrate and prove the viability of the whole fuel cycle and to explore all facets of the reprocessing and associated waste treatment.

It was decided to modify and extend the existing DFR metallic uranium fuel reprocessing plant which also offered the opportunity to demonstrate the possibility of extending the life of redundant highly active plant. A schematic illustration of the modified building is shown in Fig. 2. These modifications were completed in 1979. The plant will start reprocessing high burn-up fuel (at 8-10

*United Kingdom Atomic Energy Authority, Dounreay.
†British Nuclear Fuels Limited, Risley.