



0149-1970(95)00035-6

## COMPARATIVE ANALYSIS OF ENVIRONMENTAL IMPACT OF FAST REACTOR FUEL CYCLES

B. Saha, A. R. Sundararajan and L. V. Krishnan

Safety Research and Health Physics Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603 102, India

### ABSTRACT

There is an increasing interest in the development of processes for separation of minor actinides from the fission products and their elimination through transmutation. Fast reactor systems have certain advantages over thermal reactors in achieving this and the overall success is influenced by the choice of the fuel cycle. A comparative analysis of the different options is attempted in this paper taking into account long term waste management, extraction of maximum fuel value and intercompatibility among the different fuel cycles. It is considered more appropriate that, from the long term environmental point of view, the choice of the fuel, core design and fuel cycle technology is based on considerations of ultimate disposition of radioactive wastes. The integrated metal fuel cycle employing the pyroprocess flow sheet and the associated waste processing schemes emerges as a more attractive option in this context.

### KEYWORDS

Fast reactors; fuel cycles; waste forms; actinide recycling; ceramic fuel; aqueous reprocessing; metallic fuel; pyrochemical process; thorium utilisation.

### INTRODUCTION

Commercial generation of electricity in nuclear power stations is only forty years old. In this period, several different reactor designs have come to be adopted. But, their selection has been based primarily on short term considerations. Easy availability of fuel has discouraged efforts to reprocess and reuse the fuel. In fact, the countries which have no plans for closing the fuel cycle presently account for half of the output of all nuclear power stations. This has resulted in accumulation of huge stocks of spent fuel. The few currently operating reprocessing plants are all based on the PUREX process in which long lived minor actinides (MA) along with some plutonium are carried into the high level liquid wastes (HLLW). The environmental impact arising out of ultimate disposal of spent fuel and HLLW is thus regarded as a matter of concern.

While thermal reactors which use natural or slightly enriched uranium are inevitable in the initial stage of nuclear power programme, it is also necessary to adopt practical ways for using the plutonium and other transuranic (TRU) elements produced in these reactors so as to render the wastes from nuclear power generation free of their presence.

Nuclear energy offers a compact and relatively clean energy source in comparison to coal. Realisation of the prospects for a revival of world interest in nuclear power generation will be facilitated by satisfactory resolution of the waste disposal problem. As an essential first step in this direction, the following guidelines seem appropriate:

- a) no permanent storage of spent fuel which contains transuranic elements (TRU),
- b) immobilisation of HLLW only after removal of TRU component and
- c) designs of future reactors to be governed by considerations of separation of TRU component from spent fuel and its recycling with the fuel.

Fast reactors are best suited for extraction of the energy potential in natural uranium to the fullest possible extent. Sodium cooled versions have emerged as the best choice. Occupational exposures and environmental discharges during their operation are significantly less compared to any other reactor type. Until recently, optimisation of the core design of fast reactors was done with the objective of demonstrating safety and achieving high burn up. The choice of oxide fuel and the specific core configuration was made accordingly and the designs have acquitted themselves creditably in performance. On grounds of familiarity, the PUREX process has been adopted for reprocessing irradiated fuel from fast reactors also. No problems have been encountered, although the experience is limited mainly to the first cores. The problem due to the presence of TRU component in HLLW persists.

For a brief while, there was focus on maximising the breeding ratio through use of other ceramic fuels. Attention has now however turned in all seriousness to the question of the ultimate disposal of long lived TRU wastes with the help of fast reactors. Emphasis is being laid on development of processes for separation of minor actinides from spent fuel and on core designs for their elimination through transmutation. It should be possible to gradually turn to reactors that greatly minimise production of higher actinides.

In the context of meeting the projected energy needs of the large Indian population, nuclear energy has been seen as the only available alternative (Paranjpe, 1992). The pressurised heavy water reactors (PHWR) enable natural uranium to be used and furnish the plutonium required to start the sodium cooled fast reactor programme. The well proven mixed oxide fuel and aqueous reprocessing scheme is to be used in the initial demonstration stage of the programme. It would be more appropriate that in later stages, from the long term environmental point of view, the choice of the fuel, core design and fuel cycle technology is based on considerations of the total picture including the ultimate disposition of radioactive wastes.

## NATURE OF THE PROBLEM

If spent fuel is to be stored away without reprocessing, the radioactivity associated with it remains about 10,000 fold higher than the release limits prescribed by the US Environmental Protection Agency (EPA), even after 100,000 years (Burch *et al.*, 1991) indicating that the integrity of the isolation barrier must be maintained for more than a million years.

Reprocessing by the PUREX process produces HLLW with about 0.5% of unrecovered plutonium from spent fuel along with all the minor actinides and the fission products (Fig.1). When the HLLW is immobilised in borosilicate glass as currently planned or practised by many countries, these components are carried into the glass blocks. A high degree of isolation of the glass becomes necessary for about 100,000 years if the HLLW is from uranium fuel used in LWRs and for longer period from MOX fuel irradiated in fast or thermal reactors.

Fast reactor wastes from the PUREX process contain about nine times more of alpha emitters after 1000 years compared to thermal reactors. Noble metal (NM) content is about 50% higher and the use of stainless steel (SS) cladding results in leaching of Ni and Cr during dissolution stage. The higher NM inventory accentuates the problem of their precipitation in the glass matrix enhancing the possibility of stress corrosion cracking (Ringwood *et al.*, 1988).

The presence of Mo and Cr leads to formation of a leachable phase if the vitrification is carried out under oxidising conditions. On the other hand, in an attempt to minimise formation of the undesirable phase under chemically reductive environment, there could be accumulation of Ni and relatively noble metals in the bottom of the melter (Ringwood *et al.*, 1988). Leach rates from glass matrices being

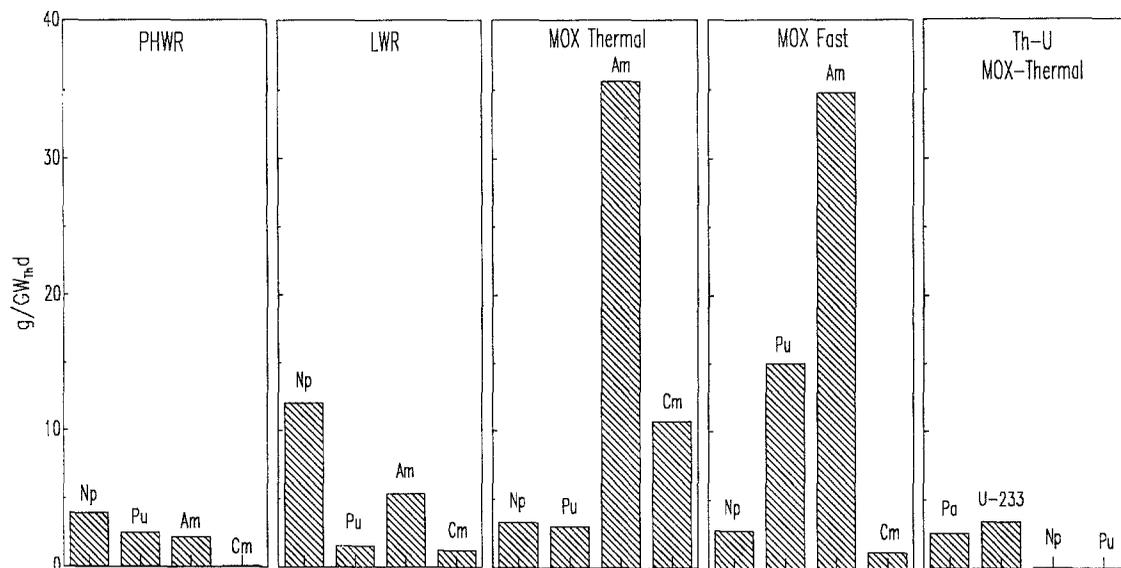


Fig. 1. Typical discharges of actinides to HLLW from different reactor types per unit energy generation. [Pu: 0.5% (PUREX); U-233: 0.5% (THOREX); MA: 100%]

strongly temperature dependent, there are limitations to the depths at which the repository can be located.

According to one estimate, each glass block of the size proposed to be produced from relatively low burn up fuel (Hanford wastes) becomes a strong and sustained source of neutrons emitting  $10^{13}$  n/s (Goldberg, 1990). The emission would be higher in fast reactor wastes.

Ceramic waste forms are being examined as alternatives for glass (e.g. Synroc), which have certain distinct advantages (Ringwood and Kelly, 1986). However, the suitability of glass, Synroc or of composite ceramics for wastes from fast reactors with fresh and recycled U-Pu cores remains a matter for further study in detail.

Assurances of safety and integrity of waste forms over geological periods of a million years or more appear difficult to comprehend or accept. On the other hand, time periods of a few thousand years are easy to visualise, as historical records are likely to be available. Many monuments and archaeological objects are known to have remained in an excellent state of preservation over such periods without the benefit of any special care. After separation of the TRU component, radioactivity levels in HLLW fall to that in uranium ore within a few centuries. Recycling of the TRUs with the fuel provides a means for their destruction through fission, at the same time adding calorific value to the fuel.

#### ACTINIDE TRANSMUTATION

Different possibilities exist for elimination of MA through fission such as in (a) subcritical assemblies driven by accelerators, (b) specially designed MA burner reactors (both fast and thermal) or (c) present designs of fast power reactors. Thermal reactors with U-Pu fuel have also been proposed for burning plutonium, but this involves continued production of the higher actinides.

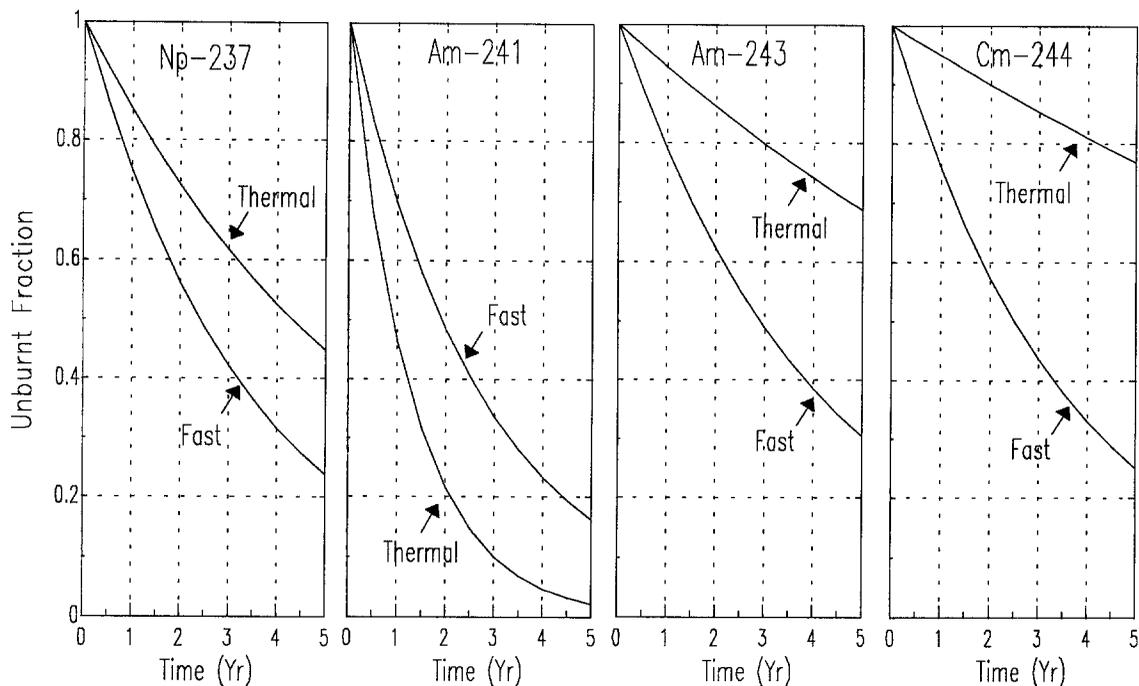


Fig. 2. Unburnt fraction of initial loading of MA as a function of residence time in typical fast and thermal reactors.

Transmutation in fast reactor is more effective in reduction of MA inventory (Fig. 2) since the harder neutron spectrum leads to shorter composite depletion half-lives (fission + capture + natural decay) (Koch, 1986). Further, as the fission to capture ratio increases with neutron energy, the share of transmutation by fission is highest in metal fuelled fast reactors compared to ceramic fuels. The fuel worth of MA is almost equivalent to plutonium in metal fuel cores but less in the MOX core. To maintain the same reactivity, plutonium content in the metal core can be reduced by about the same amount as the addition of MA. The compensation provided by MA in the oxide core is about 70 to 80 % less (Inoue *et al.*, 1991).

### REPROCESSING OF CERAMIC FUELS OF FAST REACTORS

There is considerable operating experience with the PUREX process in treating first cores of oxide fuel from fast reactors. Available experience is however limited with respect to high burn up, short-cooled spent fuel. Recycling leads to enhanced levels of MA and Pu-238. The content of the latter radionuclide in plutonium is limited to levels below 5 % in French practice to avoid excessive radiolysis of solvent. Recovery of plutonium could be affected in the dissolution stage by incomplete acid digestion and in the extraction stage by inadequate stripping from radiation degraded TBP phase. The aqueous process has been proposed for other ceramic forms of fast reactor fuel like carbide and nitride, with slight modifications in the dissolution stage.

Separation of the low concentrations of MA from PUREX generated HLLW requires additional processing steps and several schemes with new extractants are under investigation (e.g. CTA process of Sweden (Liljenzin *et al.*, 1984), TRUEX of US (Horwitz and Schulz, 1989). The multivalency of Np and Pu as compared to the trivalency of the higher actinides (Am, Cm) and the chemical similarity of trivalent actinides with trivalent rare earths add to the complexity of the process.

In aqueous processes, the heavy metals assume different chemical forms such as oxides, nitrates, complexes with the organic extractants and as oxalates (or any other precipitable form for powder or

sol preparation). Consequently, the number of parameters to be controlled is many and is likely to affect overall recovery. It also leads to generation of multiple types of liquid wastes.

## METAL FUEL REPROCESSING

Operating experience of EBR II in US with the metal alloy fuel (U-Pu-Zr) is indicative of good performance up to as high as 18 atom percent burn up (Till *et al.*, 1990). Some aspects of the inherent safety of a metal fuel core have also been demonstrated. The highest breeding gain offered by such a core, due to the hard spectrum, yields the maximum potential whether for breeding fuel or for burning MA. The nonaqueous reprocessing scheme based on electrorefining under development in US as an adjunct to the metal core achieves separation of U and TRU from the undesirable fission products effectively in a single stage (Battles *et al.*, 1991) in contrast to the multistep aqueous process. The heavy metals assume only two chemical states, during the entire fuel cycle campaign, namely the metallic form itself and as chlorides.

The use of the combination of molten salt (KCl+LiCl) and molten cadmium in a compact vessel eliminates generation of primary liquid wastes and the absence of hydrogenous materials eases the problem of accidental criticality. The reactive fission products such as the alkali metals, alkaline earths and halogens remain in the salt phase. The nobler metals (Tc, Ru, Pd, Rh, Zr etc.) stay in the metal phase. Through simple judicious alterations of the oxidation and reduction conditions in the biphasic system, 99.9% recovery of actinides is likely to be achievable and the lanthanides can be driven to the desired phase.

Uranium and the TRU materials recovered during electro refining are consolidated by melting and/or retorting. For recycling back into the reactor, a fuel alloy slug is produced by injection casting of composition adjusted molten fuel. This is a remote operation, the feasibility of which has been demonstrated (Stevenson, 1987). The fissile material recovered is present in the denatured form because of inherent dilution with uranium and the accompanying radioactivity, providing a good deterrent against any diversion for undesirable applications.

As this nonaqueous process makes use of simple inorganic process materials, which are stable even in intense radiation fields, high burn up, short cooled fuel can be processed without any loss in separation efficiency.

All the TRUs are present as trivalent chlorides in the molten state with their Gibbs energies of formation clustered together and their corecovery along with plutonium is rendered very simple. A small fraction of the rare earth elements goes with the TRU stream. Possible adverse effect on performance of the recycled fuel in the core with respect to Fuel Cladding Chemical Interaction (FCCI) needs to be studied and acceptable limits for rare earths firmly established.

Gaseous discharges from the electrorefining process are much cleaner than the dissolver off gas from PUREX. Tritium and Kr-85 alone are released to the argon cover gas. The pyrophoricity of actinides and toxicity of cadmium call for effective control over system integrity. Considerable experience is available in the safe handling of molten sodium systems in fast reactors that indicates this can be easily achieved.

While the electrorefining process was developed primarily for the metal fuel, it can be adapted for treating the spent oxide fuel with the introduction of prior pyrochemical steps (McPheeters *et al.*, 1993) for enabling extraction of TRUs for their subsequent recycling in metal fuel cores.

Simpler means are also being examined for isolating the waste actinides from accumulated HLLW through pyrochemical routes e.g. denitration, chlorination and liquid metal extraction to obtain a TRU enriched alloy feed for the electrorefiner (Inoue *et al.*, 1991; Hijikata *et al.*, 1993)

## ULTIMATE WASTE FORMS

A comparison of the collective effective dose commitment from the various stages of the nuclear fuel cycle is presented in Fig.3. The largest single contribution is from uranium milling operations (UNSCEAR, 1993). It is estimated that a total of about one million tonnes of uranium have been

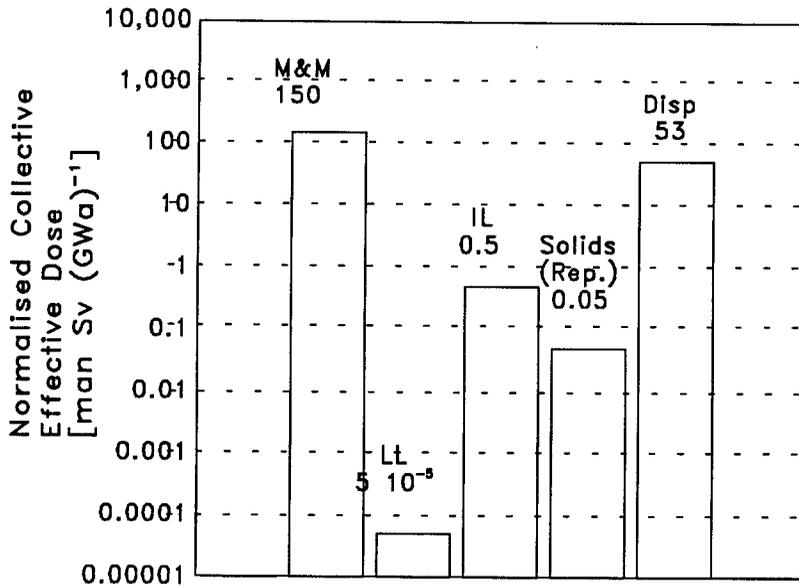


Fig. 3. Collective effective dose commitment to the general population from nuclear fuel cycle (over 10,000 years, normalised to power generation). [M&M: Mining and milling; LL: Low level; IL: Intermediate level; Disp.: Global dispersion]

produced so far, of which 400 kTe have been utilised for power generation and a stockpile of 600 kTe is available in the civilian and military sectors put together. Mining of this amount of uranium has already led to a collective dose commitment of 0.75 million manSv so far. This may appear to be a small part in comparison to the collective dose of 26 million manSv estimated to come from all the past atmospheric nuclear tests. Further additions to the dose commitment can be minimised by reducing the dependence on uranium mining. This is made possible by recycling the residual fissile actinides present in spent fuel along with depleted uranium initially and with thorium subsequently in fast reactors. What remains then is the contribution from the long lived fission products to the collective dose commitment, which have to be immobilised in the solid waste form with very low leach rates to the biosphere, to minimise their contribution to population dose commitment.

During acid dissolution of the ceramic fuel, nuclides such as H-3, C-14, I-129 and Kr-85 are released to the off gas. Schemes have been proposed for trapping them. This is important especially for C-14 as its contribution to the long term dose commitment is relatively large, if global dispersion occurs through atmospheric release. Production of C-14 in MOX fuelled fast reactors has been estimated to be about 6 GBq per tonne at 30 GWd/Te burnup. In comparison, the mixed carbide fuels produce about 55 times more of C-14 and the yield from mixed nitrides is 55 times higher still (Matzke, 1986). Reduction is possible for nitride fuel through use of the stable heavier form of nitrogen N-15 enriched to 99%, but the level would still be about 30 times more than in MOX fuel. Stable solid matrices like barium carbonate or bismutite have been identified for fixing C-14 and bismuth oxyiodide for I-129.

Borosilicate glass has been extensively investigated as the matrix for ultimate storage of high level waste from ceramic fuelled reactors. Release of H-3 and Ru-106 during calcination and vitrification has to be dealt with in the vitrification process. For effective isolation over very long time periods, the loading of Mo and other nobler metals may need to be limited to moderate levels. After removal of TRUs, the requirement for very long term integrity of the matrix is eased considerably.

Current proposals (Taylor *et al.*, 1993) for nonaqueous processing of metal fuel call for the radionuclides in the salt stream to be occluded in zeolite. The long lived nuclides Se-79, I-129, and Cs-135 are also among them, but at relatively low concentrations. The leach resistance of the zeolite appears to be good. It is also possible to have the waste finally turned to a form similar to sodalite, a

chloride containing insoluble aluminosilicate mineral with crystal structure and molecular cage like zeolite. The other radionuclides which go into the metal stream are placed in copper aluminium alloy which takes up noble metals, lighter rare earths and unrecovered TRUs (~0.1 %). The long-lived nuclides C-14, Ni-63, Zr-93, Tc-99 and Pd-107 are immobilised in this medium. No separate schemes are needed for trapping I-129 and C-14.

## THORIUM BASED FUELS

Nuclear and material aspects of the thorium fuel cycle and possible designs of systems have received considerable attention (Rodriguez and Sundaram, 1981; Srinivasan and Kimura, 1991). Thorium based fuels, with U-233 as fissile material, generate far less quantities of the higher actinides, which require four or more successive neutron captures for their formation from U-233. By far the highest yield is that of the lighter actinide Pa-231, which arises from (n,2n) reaction in thorium. It is as important as the higher actinides, from population exposure considerations. In quantitative terms, production of MA in LWRs with a mixture of U-233 and Th-232 is reported to be about a tenth of that if enriched uranium fuel is used, more than 95% of it being Pa-231 with Np-237 constituting the rest (Naka and Takeda, 1991). Levels of higher actinides are lower by several orders of magnitude.

Various strategies for thorium utilisation have been examined in the Indian context. Recycling of plutonium in current design of PHWRs for priming the thorium cycle involves minimum development effort and is likely to lead to earlier exploitation of energy from thorium, to the extent of about 17 GWe on a sustainable basis (Balakrishnan, 1991).

Use in LMFBRs of the plutonium available from PHWRs, for example, with mixed (U,Pu) carbides initially and later with mixed (Th,U) carbides is estimated to support a level of power generation an order of magnitude higher (Lee *et al.*, 1991). Of this about 75 % is expected to come from the latter. Substantial production of Pa-231 is to be expected from overall nuclear power generation at such levels.

Thorium has been subjected to irradiation in the Indian research and power reactors. The THOREX process has been used for separation of U-233 from the irradiated thorium. Protactinium is expected to move with the raffinate stream and end up in the high level aqueous waste. No values of protactinium levels in this stream have been reported in literature surveyed by us, possibly because of the low burn up achieved in these irradiations. Methods for partitioning this element from the aqueous waste have not yet been developed.

Irradiation of thorium metal based alloys are understood to have progressed to 100 GWd/Te in US (IAEA, 1987). The extension of the nonaqueous separation process to metal cores of U-233 and Th-232 needs to be investigated to eliminate Pa-231 from the final waste stream.

## CONCLUSIONS

The choice of pyrochemical processes for electrorefining and recycling of the actinides from spent fuel, the adoption of metal fuel in fast reactor system and an early switch to thorium as a fertile material seem to offer a better approach for achieving substantial reduction of the very long term impact on the environment from nuclear power generation.

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