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Optimising Our Resources

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The long term future for the nuclear industry depends on better utilisation of the energy potential in uranium than the roughly 1% conversion achieved during a single pass through current reactors. Only fast neutron systems can achieve the order-of-magnitude improvement that is needed, but they are now a long way off. Consequently the technology needs to be sustained within the framework of shorter term programmes. Development effort must therefore be devoted not only to the means of recycling recovered fissile and fertile material in the short term through thermal reactors, but to other issues such as economics, environmental conservation and proliferation resistance on which the industry is rightly or wrongly seen to be vulnerable.

One of the few ethical insights peculiar to the nuclear age is that it has the power to ruin or gravely impair the material prospects of future generations, and must not do so. This insight has manifested itself chiefly in the principle of environmental responsibility: we must clear up the detritus of abandoned operations, manage our own wastes responsibly, and provide for eventual decommissioning in any new enterprise. Moreover, we should not leave to our successors any remediation that we could reasonably undertake ourselves.

So far, so good. Less firmly established in practice is the complementary obligation to conserve for those successors the means of maintaining the style of life to which we in the developed nations have become accustomed, and to which less privileged societies aspire. The key to this standard is energy, and the rate at which fuel resources are currently being exploited for the sake of short term profit is likely to be bitterly regretted some decades hence. Oil and gas resources are being depleted as rapidly as customers can be found for them, while operable coal mines have been abandoned, not because of exhaustion, nor because (true as it may be) coal is environmentally unfriendly, but through competition from cheap gas.

If challenged, this profligacy is often excused on the somewhat specious grounds of demand, with the added expectation that before supplies run out, “something will turn up”. New reserves of oil and gas could be discovered, solar energy might be harnessed on a large scale, or unlikely as it seems, thermonuclear fusion might become a practicable and almost inexhaustible source. We may ask whether the nuclear industry is conducting itself any better than other energy producers. On the one hand half of the world’s utilities discard fuel with its potential scarcely tapped. On the other hand, many have taken a creditably long term view in examining what provision can and should be made for the future.

Fully utilised, uranium and the plutonium bred from it would be equivalent in energy to about a million times their weight of oil (Table 1). In the once-through thermal reactor regime, they provide about two orders of magnitude less. Proposals for increasing burnup in a single pass (such as through higher enrichment with measures to control initial reactivity, improve retention of fission gases, and strengthen the cladding) address the economics of the cycle, not the overall utilisation. This can be improved to any great extent only by recycling the fissile and fertile materials remaining in discharged fuel.

Realising anything like the full potential requires fast neutron reactors, which, for various familiar

Table 1. Energy equivalents of uranium, with and without recycling.

	tonnes uranium	tonnes oil
Without recycle	1	10 000
With MOX recycle (single cycle)	1	13 000
With fast reactor	1	1 000 000

reasons, are unlikely to be widely deployed until well into the next century. As an example, 1 GWe / year supplied by multiple recycle in a fast reactor requires a feed of only about 1.2 t of depleted UO_2. This compares with about 200 t of fresh uranium for a once-through PWR. The current UK stockpile of depleted UO_2 would provide the energy equivalent of 35 x 10^9 t of oil. The challenge facing the nuclear industry is to maintain the long term option, to develop appropriate technology and in particular to gain acceptance for plutonium as a valuable energy resource rather than an inconvenient waste. In other words to be seen as a solution rather than a problem.

To achieve this long-delayed success, the industry must first survive against influential forces bent on destroying it. That requires meeting those criticisms which possess in some measure a rational basis, refuting the others, and of course distinguishing more accurately than sometimes in the past between the two categories. The chief grounds on which nuclear power is criticised are that it is currently expensive, that it generates large amounts of material potentially open to misuse in the form of weapons, and that its waste products are a perceived threat to the life and health of present or future human populations. It is no use complaining that on this last point the industry already sets a higher standard than its competitors: thanks largely to sensational journalism, the notion of unique dangers associated with artificial radiation sources has become firmly entrenched in the public mind, and education can make only limited headway against it.

Although the various relevant topics interact, and must be optimised holistically, aspects of them may first be considered separately. Concern about costs, of course, runs through the whole discussion. This paper concentrates first on technological aspects, but later refers specifically to the key issue of environmental protection.

Trends in Development

Because of technical inertia imposed by concern for safety, developments for some considerable time are likely to be evolutionary and incremental, refining methods already understood in principle. Thereafter, they may gradually be supplanted by more radical techniques beyond current capabilities, or not yet imagined.

With LWRs, which will almost certainly remain predominant for the coming twenty years or so, technological development and political acceptance of plutonium as an energy source depend crucially on recycling in MOX fuel. Although the energy released thereby, including the recycle of uranium, is only a modest 30% beyond that already obtained, it is a useful increment. It also leaves the remaining plutonium still available for future fast reactors, and above all establishes the credentials of plutonium as a valuable material in its own right.

The next few decades are likely to see evolutionary changes connected with LWRs, and probably some synergy with other reactor types. Development or increasing use may be expected for:

- thermal MOX fuel in current reactor types;
- recycled uranium in LWRs, CANDUs, etc. to take advantage of the residual enrichment;
- advanced PWRs or BWRs adapted to full MOX fuel cores;
- CANDUs and other pressurised heavy water reactors, which have especially good neutron economy and are therefore capable of using fuel with low reactivity, eg. reusing fuel after discharge from LWRs (the ‘DUPIC’ concept);
- perhaps accelerator-driven sub-critical assemblies, if their merits are seen to outweigh the complexity of the external drive.

If desired, thorium based fuels could possibly be introduced as an interim measure into reactors of conventional type, for reasons considered later. Thereafter, attention may well shift gradually to:

- fast neutron reactors, which currently offer the best established way to realise the full energy potential of uranium and can sustain a balance between production and consumption of plutonium, perhaps most cost-effectively in conjunction with a larger number of LWRs;
- high temperature reactors, capable of better thermodynamic efficiencies (particularly important if thermal pollution should become a serious consideration);
- possibly molten salt systems, as a means of simplifying the whole cycle.

Developments in Recycling

Whatever the benefits of recycling, utilities will not be convinced if it requires a significant increase in overall generating cost. Reprocessing represents a relatively small proportion of the total, but it competes directly with other ways of managing discharged fuel, such as direct disposal or retrievable storage. Wasteful as these may be in the long term, and at best delaying a policy decision for a generation or two, their immediate advantages may appear compelling unless reprocessing and refabrication can be made economically competitive. A great deal of development effort is therefore directed to this end.

Currently the Purex process — solvent extraction with tributyl phosphate (TBP) in a hydrocarbon diluent — is dominant for the main line of processing irradiated fuel with no serious commercial rival. After decades of use the scope for further refinement is limited, concentrated largely on simplification and on reducing the cost of ancillary processes.

Specifications for reprocessed products have traditionally been based on that for fresh uranium and on requirements for manipulating plutonium without inconvenient shielding, and the necessary degree of decontamination can be achieved only by successive process cycles. It has no relevance to reactor operation, which almost immediately generates higher proportions of fission products; in any case, with increasing irradiation levels, the inherent beta–gamma radioactivity of the products or their daughter nuclides becomes significant. Attention is therefore being concentrated on reduced processing schemes, with a single solvent extraction cycle from which the products would be refabricated remotely. Such schemes would also have the advantages of eliminating wastes and improving proliferation resistance.

To refabricate fuel from recycled material, it is possible to adapt present methods of fuel manufacture, with stages of precipitation, filtering, calcination, milling, pelleting, sintering, grinding to size, fitting to cladding and final assembly — indeed, it has been done. However, the resulting mechanical complexity, however successful, is inherently expensive. For the sake of improved economy and reliability, methods of refabrication need to be intrinsically better suited to remote operation, such as sol–gel processes with vibro packing, for which the causes of disappointing early performance are claimed to have been overcome by developments in Russia.

A sharp separation of plutonium from uranium, besides being undesirable in terms of proliferation resistance, is also futile if the two elements are to be partially recombined in MOX fuel. Processes may therefore be adapted to produce some uranium as pure oxide and a mixture of plutonium with the rest. A major task is to build on information already available in developing a reliable finishing method for a homogeneously mixed product suited to refabrication.

Purex itself and its ancillary processes, although effective, generate an inconvenient number of bulky aqueous and organic wastes that incur substantial costs in treatment and disposal. Some of these might be avoided by a change to a solvent more resistant to radiation, or more tolerant of degradation products, and itself readily decomposed to innocuous products when no longer usable. Given the extent of experience with TBP, however, any replacement would have to show convincing advantages in order to gain commercial acceptance.

More radically, non-aqueous routes for reprocessing are being considered, notably fluoride volatility and molten salt methods. The volatility of uranium hexafluoride has long been utilised in methods of purification and isotopic enrichment. Adapting it to reprocessing is complicated by the similar volatility of certain fission product fluorides, and by the relative instability of plutonium hexafluoride — a property that offers a separative route but may cause premature deposition.

Molten salt processes have lately been associated particularly with the integrated fast reactor (IFR) in the USA, but since the withdrawal of funding for this project have been conceptually applied to LWR fuels. Electrochemical separations now seem to be favoured over selective chemical reduction. A significant difference from aqueous processes is a relatively straightforward separation of lanthanide fission products from secondary transuranics, which follow the plutonium. The importance of such a separation is considered later.

Improved Efficiency

The total energy extracted from uranium can certainly be improved by the use of full MOX cores. However, energy emerges from the reactor as heat, which is of interest chiefly as a means to the end of generating electricity. For thermodynamic reasons, the conversion efficiency is less than 50%, which perhaps might be improved by raising the outlet temperature of the coolant. Ways to achieve this include:

- raising the temperature limit on the fuel cladding;
- homogenising fuel ratings within the core so that the mean cladding temperature is closer to the limit;

Then, given these improvements:

- raising the limit on the interior temperature of

the fuel, by changes to chemical form or microstructure;
- using fuel pins with an annular cross-section to avoid an overheated centre;
- increasing the thermal conductivity of the fuel substance (eg. by the use of additives, or by replacing oxide with nitride or metal alloy) so that the outer temperature can be brought closer to the interior limit.

All of these possibilities are being actively pursued.

Environmental Impact

Environmental detriment from any industrial activity has three elements:

- routine discharges of waste;
- accidental releases of noxious material;
- slow dispersion of materials consigned to a repository.

In this respect, the nuclear industry is exceptional only in that its wastes are more easily detected and thus responsibility more clearly identifiable than elsewhere. Except in the case of direct disposal, the amounts involved are minuscule with little direct bearing on resource utilisation, but unsatisfactory management could arouse sufficient animosity to undermine the whole principle of recycling.

Concern about waste management has focused attention on “clean technology”, ie. designing processes from the start for minimal, tractable waste streams rather than finding ways retrospectively to deal with whatever wastes are produced. Holistic consideration of the fuel cycle is becoming increasingly important.

For the long term, particular concern has arisen over the possible diffusion into the environment of long lived radionuclides such as the minor actinides (neptunium, americium and curium) and some fission products. In the 1980s the idea of separating them, for specially secure disposal or transmutation into shorter lived species, was dismissed as an unwarrantable diversion of financial and intellectual resources. However, pressure to reconsider has more recently become intense, along with a suggestion that the energy obtainable through fission of the actinides would cover the cost of separation from highly active waste. The arguments are open to question, but need to be kept under review.

For neutronic and chemical reasons, recovered secondary transuranics can only be blended to a limited extent with uranium and plutonium. They may have to be formed into special targets for irradiation, perhaps dispersed in inert matrices such as magnesium oxide. Similar considerations would apply to long lived fission products if they too were to be transmuted. Since consumption in a single pass would be incomplete, methods of reprocessing and refabrication would have to be devised.

The issue of secondary transuranics has become more serious since the inauguration of the Japanese OMEGA project, and the passing of a French law in 1991 requiring further attention to long lived radioactive wastes. Nuclear properties relevant to incineration by fission or transmutation have been extensively studied, but the difficult separation of these elements from the chemically similar and much more abundant lanthanides is a prerequisite not yet established in solvent extraction systems. It has been achieved experimentally only through an extension to the Purex process, generating additional wastes that would themselves present problems in disposal. Separation factors would have to be very high if the actinide contamination were not to be spread over a larger volume. The operational risks would also have to be set against any supposed long term benefit. Whether the net improvement would justify the cost, or indeed would exist at all, is very debatable.

Largely because of concern over these elements, and more particularly the proliferation risks rightly or wrongly associated with civil plutonium, interest has revived in thorium as a fuel. Theoretically representing an enormous energy reserve, and originally proposed as a replacement for uranium once it became scarce, it is now favoured chiefly because it forms practically no transuranic elements. Manufacturing the fuel from virgin stock appears to be a relatively straightforward adaptation of current processes (although could not be done not in current plant), but a sustained thorium / U-233 cycle would require reprocessing, and thorium based fuels are intractable. Moreover, recycling thorium and U-233 is hampered by severe radiological problems.

Thorium oxide cannot be dissolved in nitric acid without highly corrosive additives, while subsequent solvent extraction has been achieved only under experimental or makeshift arrangements that would not be commercially acceptable. Dry methods might be preferred: fluoride volatility is inapplicable, but according to theoretical calculations and some early trials, molten salt processes could be adapted. In practical terms, industrial application may be possible some decades ahead; meanwhile it remains more a topic for low-key research.

Concern about accidental releases from operating plants is usually concentrated on reactors. For example, in a fast reactor an overheating accident might conceivably cause fuel to slump into a more reactive configuration than normal, with a consequent breach of containment. There have been several proposals, notably Carlo Rubbia's combination of various relevant ideas, for sub-critical assemblies of fissile and fertile materials with a fission chain initiated and controlled by neutrons generated in the impact of a high energy proton beam on a heavy metal target. Such a system would reduce the risk of accidental release, although whether it would actually have all the advantages claimed for it remains to be demonstrated. In order to avoid generating transuranic elements, including plutonium, Rubbia's initial proposal assumed thorium as the fertile content, but in other respects uranium would be equally effective and easier to process.

Conclusion

The question of whether the nuclear industry is adequately facing its responsibilities for the future has no clear-cut answer. The practice of once-through passage of nuclear fuel, widely adopted for economic or political reasons, wastes nearly all the potential of the original uranium, which is some day almost certain to be needed. Recycling recovered uranium and plutonium in thermal systems, though better than discarding them, achieves only modest gains. Fast reactors are the only complete solution in sight, but are still a long way off. Meanwhile the industry must be sustained against attacks on its performance in environmental preservation or resisting weapon proliferation, while commercial practice makes it susceptible to economic competition from fuels such as natural gas with short term advantages if little long term prospect.

The UK has no reactors of advanced designs, and currently does not use MOX fuel in those it does have. However, it still retains the policy of storing separated uranium and plutonium for future use, and continues to recover fissile and fertile materials from discharged fuel. British Nuclear Fuels (BNFL) is actively supporting the use of both reprocessed uranium and MOX fuels, with a substantial investment in new plants to utilise the products from its THORP reprocessing plant at Sellafield.

At BNFL's Springfields site a new plant is being constructed to convert reprocessed uranium to UF_6, while LWR fuel can be fabricated from re-enriched UF_6 in the Oxide Fuel Complex. Reprocessed uranium with a U-235 content of nominally 0.9% is also ideally suited to recycling in reactors of the CANDU type, and this opportunity is being explored by BNFL together with partners in Korea and Canada.

The new 120 t/year Sellafield MOX Plant will substantially expand BNFL's capability in plutonium recycling and build on the experience gained in operating the current 8 t/y MOX Demonstration Facility. With a view firmly fixed on the longer term, BNFL also sustains the UK involvement in international development of fast reactors.

Meanwhile, improvements are evolutionary, with the hope of more fundamental changes in a few decades. It is vital for the industry to remain economically competitive with other energy sources that have been optimised for short term considerations, while meeting the severe constraints on its products and wastes. This is the object of most current development effort.