OPPORTUNITIES FOR FUEL CYCLE DEVELOPMENT

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ABSTRACT

Today the Nuclear Industry is faced with several key challenges, not least that in all it does it has to achieve the highest levels of safety, pursue waste management options acceptable to the industry and general public, operate within varying political climates, and tackle these within a cost regime competitive with fossil fuel generated electricity.

These are undoubtedly testing challenges, but on looking to the future there are certain trends which may prove beneficial to the industry and assist in mitigating them - namely that global energy demands are forecast to steadily increase over the next twenty years and beyond, and that in meeting this demand, the desire is to do so with minimal environmental impact.

This paper will describe the part nuclear generation can play in servicing future energy demands, how the nuclear resource can best be utilised, how the 'holistic fuel cycle' philosophy can provide a framework for tackling the challenges faced by the industry, and the extent by which international co-operation can support certain advances in the fuel cycle.

THE FUTURE ROLE OF NUCLEAR

A joint World Energy Council (WEC) and International Institute for Applied Systems Analysis (IIASA) study¹, supported by other authoritative organisations, predicts that primary energy, electricity and fossil fuel consumption will continue to increase over the next 50 years, and that this energy demand will double over the next 20-30 years alone. If we look at the supply trend, and make certain assumptions, an interesting picture emerges for nuclear: If the assumption is made that due to environmental pressures, fossil fuel generated electricity plateaus after the year 2000, that renewable energy sources (predominantly hydropower) can double their share of the electricity market by 2050, and account is taken of the capacity of nuclear reactors currently in the planning phase, then a mis-match between demand and supply appears of the order of ~8000 TWh by 2050. Indeed, it is worth bearing in mind that this forecast is at the lower end of the range of the options covered in the WEC/IIASA study and makes full allowance for energy efficiency and conservation issues. Thus in terms of bridging this gap, it is clear that energy conservation and renewables are insufficient on their own to assure sustainable economic development - leaving a clear opportunity for nuclear.

Underlying these projections and running in parallel with the above argument, is the environmental impact of fossil fuels. At the 'Earth Summit' in Rio in 1992, the United Nations Framework Convention on Climate Change was agreed and parties to this Framework recognised the long term need to stabilise greenhouse gas concentrations globally (particularly CO₂), whilst at the same time not impairing economic and social development. At Kyoto in 1997, the compromise reached was that developed countries should aim for a 5% reduction of 1990 emission levels by 2012; in the UK the target is 8%, which is well within the 20% target set by the Government itself.

The overall aim of the World Energy Council is for a balanced energy policy which utilises a mix of energy resources, and also implies in its approach that an increase of the percentage of sources of energy that limit emissions is desirable. The nuclear industry needs to become more pro-active in supporting this long term

aim. Nuclear power is a well-developed source of energy, with nearly 450 nuclear reactors operating world-wide in some 32 countries, providing approximately 17% of global electricity supply². These operating nuclear power plants already avoid the emission of 2.3 billion tons of CO₂ per year - this figure is ~10% of current total CO₂ emissions. As an illustration, the generation of 10,000 TWh by nuclear power in 2050 (which is very much within the WEC/IIASA forecast range) would avoid about 7.6 billion tons of CO₂ emissions.

There is clearly an economic opportunity for the nuclear industry - to be able to seize this, the industry needs to maintain its safety record and internationally bring up to the highest standard parts of the industry which are lagging behind. It needs to ensure that it can maintain a cost competitive position, and this means a better utilisation of our basic resource and a more efficient operation of fuel cycle services.

RESOURCE UTILISATION

When compared to estimates of fossil energy resources, the uranium resource is fairly small if used in a once - through fuel cycle (Figure 1).



Figure 1. Estimated World Primary Energy Resources³

However, with multiple recycle, the energy yield from the uranium could exceed that from the total combined fossil source by a factor of three. It is clear therefore that any long-term future for the nuclear industry depends upon a better utilisation of the energy potential in uranium than the roughly 1-2% conversion achieved during a single pass through current reactors.

In the once-through thermal reactor regime, one tonne of uranium equates to $\sim 10,000$ tonnes of oil, whilst single recycle as thermal MOX fuel is an interim step which provides a modest but useful gain of about 30% on this. But, fully utilised in fast reactors, uranium and the plutonium bred from it would be equivalent in energy to about a million times their weight of oil. Indeed, the current UK stockpile of depleted UO₂ would provide the energy equivalent of 35 billion tonne of oil if used in this way.

However, for various familiar reasons, fast reactors are unlikely to be deployed until well into the next century. Hence, we must ensure that in what we do now and in the near future, we keep this option open and thereby secure the long term viability of the industry. A key challenge facing the industry is that appropriate low cost recycle technology has to be developed and that plutonium has to be accepted as a valuable energy resource. Implicit in this is the need to counter the criticisms directed at the nuclear industry with respect to safety, environmental performance and nuclear material proliferation whilst retaining the necessary capabilities and skill base long term. In addressing these issues and in future

developments of the fuel cycle it is essential that an holistic approach is taken (Figure 2) - that is consideration of the fuel cycle as an integrated whole and the inter-relationship of each stage understood and considered in any developments that are made.



Figure 2: Holistic Fuel Cycle

With this approach in mind, we can consider the principal stages of the Fuel Cycle, and possible future developments therein:

FUEL

Accepting that LWRs will almost certainly be predominant for the next 20 years or so, developments are likely to be evolutionary rather than revolutionary.

The economies of thermal reactor fuel will depend upon both the 'front end' and the 'back end'. In the 'front end', the economics improve with burn-up to about 55-60GWd/t. Beyond this level, the separative work function rises steeply and the corresponding irradiation times imply a greater discounting penalty, both of which restrict the likely economic burn-up. In the 'back-end', both reprocessing and direct disposal costs are likely to increase with burn-up. When all these effects are taken into account, the economic optimum burn-up of thermal reactor fuels are not much different from today's technological capabilities - up to about 60GWd/t.

As to the future technical limitations on fuel burn-up, it is considered that for a reprocessing cycle, the likely range will be about 55-60GWd/t. Extending burn-ups beyond this range may not be optimal because of increased difficulty and expense of reprocessing more active fuels, decreasing fissile quality of plutonium (particularly if considered for re-use in thermal reactors) and low U-235 assay. Therefore, minor developments in fuel design and cladding are all that will be required.

For the once-through cycle, greatly increased 'back-end' costs may drive burn-ups towards 80 or even 100GWd/t. However, current cladding has little prospect of achieving such levels of burn-up and a radical alternative - such as ceramic cladding will be required. Furthermore, the much higher power peaking factors associated with high burn-ups will take the fuel too close to its fundamental limits, such as melting point. A new fuel design will therefore be required.

Looking further into the future at high temperature reactors possessing helium cooled graphite moderator, higher melting point fuel is possible, for example fuel based on coated particles - uranium dioxide or MOX

inside a silica or silicon carbide shell, possibly pressed into blocks incorporating graphite or maybe as a pebble bed. Fission gas retention should be good and there is no requirement for cladding, hence there should be few barriers to achieving very high burn-ups and setting the economic standards against which the LWRs will have to compete.

There are several other fuel options that may to be relevant in the future:

- Use of recycled uranium. Current enrichment processes enrich U-236 (a neutron absorber) and U-232 (with strong gamma emissions from its daughter products making fuel fabrication difficult) as well as U-235. Selective enrichment of U-235, for example by laser isotope enrichment, would allow use of recycled uranium of lower assay.
- New reprocessing technologies may choose not to separate U and Pu to present levels of purity with consequential effects on fuel manufacture. Examples may be pyroprocessing followed by vibro-manufacture or concentrating Pu to about 20% in uranium and finishing of fuel by a sol-gel route.
- Thorium-based fuel cycles may form a minor part of the overall nuclear capacity. Manufacturing the fuel from virgin stock appears to be a relatively straightforward adaptation of current processes (although not in current plant), but a sustained thorium-U-233 cycle requires reprocessing, thorium-based fuels are intractable and moreover, recycling is hampered by severe radiological problems. In practical terms, industrial application may be possible some decades ahead but the development requirements in terms of cost and resources are very substantial, thus it remains more a topic for low-key research at present.

It can be concluded therefore that advances in fuel technologies, certainly over the next 50 years, will be driven by the prevailing reactor mix, by economics and by the need to integrate fuel manufacturing with adjacent segments of the fuel cycle, and particularly the output from reprocessing plants.

REPROCESSING

Whatever the benefits of reprocessing/recycling, they will not convince the utilities if they require 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 are liable to appear compelling unless reprocessing and refabrication can be made more economically competitive. A great deal of development effort is therefore directed to this end.

The World's current reprocessing technology is the 50-year old PUREX process - solvent extraction with tributyl phosphate in a hydrocarbon diluent. After decades of use, the scope for further refinement of this aqueous based process is limited, and is concentrated largely on simplification and on reducing the cost of ancillary processes.

However, there is considerable R&D ongoing world-wide looking at options for more radical fuel cycles. Under consideration are:

- Relaxed reprocessing plant production specifications: Attention is concentrated on reduced schemes, with a simple solvent-extraction cycle from which the products would be refabricated remotely.
- Relaxed requirements for sharp separation of plutonium from uranium: Processes could be adapted to produce some uranium as pure oxide and a mixture of plutonium with the rest.

- Alternatively total co-processing of a mixed U and Pu stream is feasible.
- Developments in remote operation to allow more extensive recycling of material and integrated fuel refabrication (e.g. sol-gel coupled with vibro-packing mentioned earlier)

In terms of the actual reprocessing, newer more radical technologies are based on non-aqueous flowsheets, namely molten salts, fluoride volatility and DUPIC technologies.

Pyroelectrochemical reprocessing using molten salts involves the dissolution of irradiated fuel in molten salts (mainly molten alkali chlorides at 450-700°C), followed by decontamination of the fuel by electrolysis or some other similar method. Such processing means that only minimal treatment of the products (such as removal of salts and crushing, melting or liquid metal vacuum extraction) is required prior to remanufacture of the fuel.

This approach has been applied to oxide fuel (RIAR, Russia), metal fuel (Argonne National Laboratory, US) and nitride fuel (JAERI, Japan) with suitable modifications. The process benefits are seen as:

- high chemical stability
- high dissolution capacity
- no neutron moderator
- improved proliferation resistance
- generally performed in one compact device, leading to lower production costs
- minimised HLW volume through concentration of fission products from the molten salts
- final products are ready for fuel manufacture
- suitable for wide range of fuel types without the need to alter equipment

Though the technology was developed for Fast Breeder Reactor fuel (e.g. for oxide fuel at RIAR or metal fuel at Argonne) there is sufficient experimental data to demonstrate the applicability for LWR fuel. Indeed, these processes have been tested on a semi-industrial level and are ready for commercialisation.

A second non-aqueous technology is fluoride volatility. This well-known process has some safety drawbacks as a large portion of the radioactive materials is transferred into the gaseous phase. However, techniques are available which can allow only the uranium to be extracted (as UF6) from the irradiated fuel, keeping all other products solid. If necessary, a pyrochemical treatment can extract the plutonium dioxide from the fluorination residues (the CENTAUR process). This process is particularly suitable for reprocessing long-cooled fuel in order to extract good quality uranium suitable for further enrichment.

In future, there may be limited application of methods for direct repeated irradiation of fuel. The DUPIC (Direct Use of PWR fuel in CANDU) programme undertaken by Canada and South Korea is an example. Though this method has yet to be demonstrated on a large scale, feasibility studies are encouraging. Similar principles can be used for internal recycle of FBR fuel especially for fuel containing minor actinides for transmutation.

All these technologies have been experimentally proven to differing degrees. They can be modified depending on the reactor types used in future and will permit the reduction in recycle duration by integrating reprocessing and fuel manufacture.

WASTE AND ENVIRONMENTAL IMPACT

One of the most complex and challenging areas of the fuel cycle is that of waste management - this includes reducing the generation and diversity of wastes, its retrieval, treatment, storage and ultimate disposal. As has been indicated earlier, advances in this area are linked to developments and advances in other parts of the fuel cycle.

Today's challenges in the waste management area include:

- treating the large volume of historic waste
- devising acceptable schemes for disposing of solid wastes
- reducing the current discharges whilst tackling the historic wastes,

and the fact that these solutions have to be tackled with due regard to making them sensitive and robust to potential wastes generated from advanced fuel cycles.

In particular, concern over waste management issues has focused attention on 'clean technologies', that is, designing processes from the start for minimal, tractable waste streams rather than finding ways retrospectively to deal with whatever wastes are produced - indeed the holistic approach in action.

In terms of waste management, the most widely accepted methods for immobilisation of High Level and Intermediate Level Wastes resulting from reprocessing are vitrification in glass matrices for HLW and encapsulation, typically cement, for ILW. As a result, development work is predominantly geared at refining these technologies, although other avenues are being explored internationally, for example the use of ceramics for immobilising HLW.

In terms of waste minimisation, volume reduction is a key area of activity, with specific technology developments in the areas of sorting and categorisation, declassification, 'super' compaction ability, and higher incorporation levels in final immobilised forms. Similarly, in dealing with liquid effluents, research is centred on technologies that have the ability to treat large volumes of primary and secondary wastes whilst removing low levels of radionuclides.

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 has increased to reconsider this option and substantial programmes are underway, particularly in Japan and France, developing this concept.

CONCLUSION

In conclusion, the nuclear industry has a clear opportunity to revitalise itself over the next 50 years, based on the forecast gap between supply and demand and from the benefits it can derive as a result of the general international desire to meet that need with managed atmospheric emissions. Nuclear has a demonstrable record of efficient, safe electricity generation with little or no CO₂ emissions - the industry needs to be more proactive in promoting these positive aspects.

The industry also needs to improve its economies whilst maintaining the highest levels of safety - this requires considering the fuel cycle as a whole and ensuring developments in one area are beneficial and not detrimental in others. This integrated approach best places the industry to optimise the fuel cycle and to enable the best use of the uranium resource. Realising the full potential of this resource must involve recycle and ultimately use within fast reactors - all that is done in the interim needs to support this ultimate goal.

This paper has indicated options and strategies for developing the fuel cycle as a whole highlighting in certain areas where increases are expected to be incremental and where in others more rapid programmes are being pursued. In exploring many of these potential advancements, collaboration and co-operation is of primary importance; realisation of many of the challenges seen by the industry will necessitate large R&D expenditure, and it should be recognise this is most effectively met through willing collaboration on an international level.

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KEY WORDS

Fuel cycle, Development, Resource, Holistic, Recycle.