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Overview of the Present UK Position on
Management of Minor Actinides

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This paper contains two interconnected messages; one is concerned with the recycle of minor actinides, the other is concerned with the development of process and equipment to support future fuel reprocessing business.

Minor actinide recycle (by which is meant Np and Am) has been well explored by groups in Europe and the US and more recently in Japan. Our present view is that although the technology could doubtless be developed to achieve minor actinide recycle, there is as yet no safety or economic justification for doing this.

If we look at existing UK plants at Sellafield we note that high level waste from first cycle extraction has been concentrated and stored since the early 1950's: some 1300 m³ has now accumulated. From July this year that concentrate is being processed through our new Vitrification Plant. In two years time, when THORP comes on stream its high level waste will be combined with existing stocks and processed in due course through the vitrification plant.. Thus there is no easy way of accessing minor actinides in this material. It is estimated that the amounts of neptunium and americium immobilised in this way from all past and future Magnox operations plus THORP baseload contracts are of the order 3.2 tonnes (Np) and 3.9 tonnes (Am). This will be associated with about 3.4 tonnes uranium and 0.3 tonne plutonium.

UK policy for the future management of vitrified waste of UK origin is to store it under safe and passive conditions for 50 or so years, pending a decision on whether to dispose of the waste finally or store it for further heat dissipation. The product store for this purpose has been brought into service in parallel with the vitrification plant.

The main message of this paper concerns development in support of our future business. The assumption is that fuel reprocessing will remain part of that business. We are looking at the plant to succeed THORP; in other words the time frame takes us to 2020 and beyond. Whether we build such a plant depends on its economics at that time in relation to the option of direct disposal of fuel. We assume that customers for fuel reprocessing will have a use for the products to be separated: such use will generate credits in relation to the alternative of starting with new uranium fuel and to those credits we may add the avoidable cost of direct fuel disposal. This logic leads to the concept of allowable reprocessing cost. It is very clear that present day reprocessing and waste management costs will need to be substantially reduced in order to be competitive in the future.

In order to achieve this result we are now embarking on a long range development programme. This draws on the results of previous work undertaken jointly with UKAEA, whose contribution is acknowledged. Our current programme has 3 principal elements: one is to simplify the head end by replacing mechanical operations with chemical dissolution of fuel and cladding; the second object is to reduce the complexity of the present sequence of solvent extraction cycles and thirdly, we want to reduce the size of process equipment. This last development is popularly known as process intensification. None of these developments is novel. All have been looked at over previous decades for a variety of different reasons. We wish to revisit them in the context of evolutionary development of established process and plant design.

The first picture (Overhead 1) shows development aims in relation to the head end. Present plant embodied in THORP includes chopping and leaching of fuel assemblies followed by batch dissolution in large dissolvers. The head end contains large items of mechanical plant which need to be taken off-line routinely for decontamination and refurbishment. The result of this is that the total size of the head end cell is some 4 or 5 times larger than that part which is actually devoted to handling and shearing the fuel. The head end in THORP is a major item of cost amounting to about one third of the total. In operation large volumes of intermediate level waste are generated and we have chosen (in response to lay out considerations on the Sellafield Site) to create a separate large intermediate level waste encapsulation plant.

We see potential benefits in substituting total chemical dissolution of fuel for mechanical plant. We are exploring two possible routes: one involving electrochemical dissolution in a nitric acid medium, the other employing more aggressive chemical reagents. We believe that either way we shall succeed in reducing the total size and investment in the head end and we shall eliminate large cost items associated with maintenance of mechanical plant.

The consequences of successful development include the possibility of reducing the size of the ILW encapsulation plant. We cannot eliminate it altogether because there will be other ILW streams arising elsewhere in the plant. We see attractive possibilities for alternative HLW immobilisation routes, because high level waste will now contain a large component of dissolved zircalloy, and we note that this is a major and essential constituent, for example, of Synroc.

If we now look at the second (Overhead 2) of our development aims, relating to the main chemical separation plant we see that at present these contain two or three complete extraction cycles. They are designed to produce separated U and Pu products meeting demanding specifications. The multiplicity of extraction cycles results in a multiplicity of effluent streams each of which requires treatment before very low active effluent can be disposed. Such treatment results in the generation of secondary waste which in its turn has to be handled, encapsulated, stored

and eventually disposed. The final point is that under present circumstances all minor actinides are rejected to high level waste and as we have seen, once they have gone down that route they become essentially inaccessible.

For the future we are aiming to simplify chemical separation down to one extraction cycle, part of this simplification involves a re-examination of product specifications appropriate for recycle to either thermal fuel or fast reactor fuel. Our joint programme with UKAEA has shown that a modified and optimised single cycle can theoretically achieve similar FP decontamination factors to those that will be achieved by THORP employing two cycles for each product stream. Very clearly a single extraction cycle will minimise the number and complexity of effluent and waste streams. A consequence of reduction to a single extraction cycle is that it provides the opportunity to examine the potential for internal recycle of minor actinides, a point to which I will return later.

The third development aim (Overhead 3) concerns process intensification, in other words the reduction of process plant to minimise size. Our present plant and layout for THORP include tall process columns with liquid stream residence times measured in minutes. These columns and their shielded cells have high aspect ratios which pose difficulties in applying seismic design criteria. Obviously, such columns and cells entail high mechanical and civil engineering costs. In addition the head end plant referred to previously contains large items of equipment for treatment of dissolver off gases. These scrubbing columns are sized to accept high ventilation flows emerging from the shear cell and they are a major element determining the elevation of the THORP plant. For the future, therefore, we see attractive possibilities in utilising miniaturised equipment. Such equipment will have typical residence times measured in seconds and its favorable aspect ratio will clearly ease the problem of seismic design. Shielded cells will be much smaller and civil engineering costs will be reduced.

One item under this heading is the centrifugal contactor supplied to UKAEA by Oak Ridge under the terms of the US/UK LMFBR Agreement. We are working with the UK Atomic Energy Authority in incorporating banks of these in the highly active cycle of the fast reactor fuel reprocessing plant at Dounreay. We expect this installation to be complete about 18 months from now and thereafter we anticipate some 5 years of operation under highly active plant conditions, this will be an invaluable source not just of process data but of engineering experience with these novel units. In parallel with this we are continuing to develop with the Authority an alternative concept involving fluidic devices in a horizontal array. With both of these options, the consequences are that we shall need to re-examine reaction kinetics under low residence time conditions, and we shall need to look at overall process control: we are accustomed to plants which are fairly forgiving and this may not be true with the fast acting systems that we are now developing.

I turn now to the matter of product specification. We have to keep in mind the possibility of product recycle either to thermal

reactor fuel or fast reactor fuel. In the first case uranium product will need to be free from neptunium, technetium and plutonium in order for economic recycle to conversion, enrichment and fabrication plants. It has to be noted that uranium product re-enrichment increases the level of U236 which, on irradiation, can result in a fivefold increase in neptunium content. This places additional demands on uranium purification to maintain low specified levels of Np in U product. Blending of fresh and recycled UO₂ fuel limits the rise in average Np content to about 50% above the level in fresh UO₂ fuel. There are corresponding increases in Pu238 content, which have implications for plutonium recycle. There are no significant changes in americium levels associated with uranium recycle.

There isn't a similar constraint on plutonium purity, small amounts of neptunium and uranium might be acceptable from the point of view of MOX fuel fabrication, but there is no LWR core performance benefit in recycling significant quantities of neptunium down this route. However, irradiation of thermal MOX can result in a fivefold increase in americium content compared with fresh UO₂ fuel at the same irradiation. Blending of UO₂ and MOX fuels will limit the rise in average americium content to about double the level in with fresh UO₂ fuel.

Specifications for recycle in the fast reactor fuel cycle are quite different. Uranium and plutonium will be recombined anyway in core fuel and it is not necessary to achieve high decontamination of either stream from the other component, nor is it necessary to achieve high separation of either stream from neptunium.

Indeed, studies now being undertaken within the EFR project are showing minor positive benefits in neptunium recycle to fast reactor fuel. The same is true of americium recycle if that could be achieved. The incorporation of modest quantities of neptunium and americium leads to an overall reduction in the required plutonium enrichment, it leads to an increase in plutonium breeding gain and a reduction in the rate of reactivity burn up, this leads in turn to a reduction in control rod investment. There are limits on the extent to which neptunium and americium can be recycled set by the increase in sodium void coefficient and reduction in Doppler coefficient. Another limit is posed by the resulting in-growth of plutonium 238 which presents handling problems at the next stage of fuel recycle.

These considerations prompt re-examination of methods for minor actinide recycle. That is to say: recycle to plutonium product when reprocessing thermal reactor fuel in order to transfer minor actinides to the fast reactor fuel cycle, or recycle within the fast reactor fuel cycle. In the case of neptunium this may prove to be relatively easy particularly if the search for a single cycle flowsheet is successful. Neptunium extraction behaviour in TBP depends on the mix of valency states and the rate at which they interconvert. We visualise conditioning of neptunium to cause the six valent form to extract. This will be followed by reduction to the five valent form which would then emerge with the reduced and separated plutonium product.

It seems unlikely that this internal recycle possibility can be applied to americium. It is normally inextractable in PUREX first cycle flowsheets, although it has been shown to be extractable under high salt/low acid conditions. Total dissolution of fuel and cladding could provide the necessary high salt conditions, but it seems likely that a separate contactor will be required to extract americium from high level waste after low acid conditions have been established. Once extracted, the americium stream will need to be scrubbed free of associated fission products and for this purpose a high salt low acid scrub stream will be required. In due course the americium can be returned and combined with the plutonium product from the main process. We are likely, therefore, to be constrained to parallel processing of americium rather than internal recycle.

It is clear that a number of problems remain to be investigated, the question of feed conditioning to oxidise neptunium to its extractable form needs examination to ensure that plutonium extraction is not itself prejudiced. We note that conditions suitable for americium extraction will also favour the extraction of lanthanide fission products and decontamination from these must rely on differential partition and may require many extraction and scrub stages. The penalty may be tolerable if intensified extraction equipment is available from our other line of development.

Even if we can devise process chemistry suitable for recycle of actinides it remains to demonstrate that they can be successfully incorporated in ceramic oxide fuel and we should note that historically fuel demonstration takes many years.

In conclusion therefore: BNFL expects to remain in the nuclear fuel cycle business for a long time: we expect this business will include fuel reprocessing and waste management: we recognise the need to control and reduce the costs of these operations: we have in place a long range development programme which addresses three targets for cost reduction. If we are successful in all three then minor actinide recycle by simplified routes becomes feasible and could in due course be undertaken if there is good reason to do so. We must meanwhile avoid commitments to engage in actinide recycle for dubious reasons of waste management at costs which would jeopardise the economics of the fuel cycle.

DEVELOPMENT AIMS

1. HEAD END

PRESENT:

CHOP/LEACH WITH **BATCH** DISSOLVERS
LARGE ITEMS OF MECHANICAL PLANT
NEED FOR OFF-LINE MAINTENANCE
MAJOR ELEMENT OF CAPITAL COST
LARGE ASSOCIATED ILW ENCAPSULATION
PLANT

FUTURE:

POSSIBLE TOTAL CHEMICAL DISSOLUTION
ROUTE

REDUCE HEAD END PLANT SIZE &
INVESTMENT

ELIMINATE MECHANICAL PLANT &
MAINTENANCE NEEDS

CONSEQUENCES:

REDUCE SIZE OF ILW ENCAPSULATION
PLANT

SEEK ALTERNATIVE HLW IMMOBILISATION
ROUTE

DEVELOPMENT AIMS

2. CHEMICAL SEPARATION PLANT

PRESENT:

TWO OR THREE EXTRACTION CYCLES

**DEMANDING SPECIFICATIONS FOR
U & Pu PRODUCTS**

MULTIPLICITY OF EFFLUENT STREAMS

SECONDARY WASTES

**TOTAL REJECTION OF MINOR ACTINIDES
TO HLW**

FUTURE:

ONE EXTRACTION CYCLE

**PRODUCT SPECIFICATION REFLECTS
PRODUCT END USE**

MINIMISE EFFLUENT AND WASTE STREAMS

CONSEQUENCES:

**OPPORTUNITY FOR INTERNAL RECYCLE OF
MINOR ACTINIDES**

DEVELOPMENT AIMS

3. PROCESS INTENSIFICATION

PRESENT:

TALL PROCESS COLUMNS

TYPICAL RESIDENCE TIMES IN MINUTES

HIGH ASPECT RATIO POSES DIFFICULTIES
IN APPLYING SEISMIC DESIGN CRITERIA

LARGE SHIELDED CELLS

HIGH CIVILS COSTS

FUTURE:

SMALL EQUIPMENT IN HORIZONTAL ARRAY

TYPICAL RESIDENCE TIMES IN SECONDS

FAVORABLE ASPECT RATIO FOR SEISMIC
DESIGN

SMALL SHIELDED CELLS

MODERATE CIVIL COSTS

CONSEQUENCES:

NEED RE-EXAMINATION OF REACTION
KINETICS

REVIEW OVERALL PROCESS CONTROL