INTERNATIONAL CONFERENCE
ON NUCLEAR POWER AND ITS FUEL CYCLE
SALZBURG, AUSTRIA • 2–13 MAY 1977

INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA-CN-36/66

FAST REACTOR FUEL REPROCESSING IN THE UK

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1. INTRODUCTION

Development work on the Liquid Metal Cooled Fast Breeder Reactor (LMFBR) has been carried out in the United Kingdom since the early 1950s. The experimental Dounreay Fast Reactor (DFR) of 60 MW(th) power has been operated as part of the development programme since 1960 and is now about to be shut down. The Dounreay Experimental Reactor Establishment (DERE) included plants specifically designed for the reprocessing, conversion and refabrication of highly enriched uranium fuel to provide a total fuel cycle capability to support the operation of DFR. Indeed DERE could be claimed to have been the first nuclear complex in which all fuel cycle services were situated on the same site as a single reactor.

The history of irradiated fast reactor fuel reprocessing in the UK therefore extends back to 1961 when the first discharge of irradiated fuel from DFR was reprocessed. Since then tens of tonnes of irradiated enriched uranium fuel of average burn-up of 2% of heavy atoms have been reprocessed latterly at a standard cooling time of 90 days from reactor discharge to feed to the reprocessing plant.

In 1966 a Prototype Fast Reactor (PFR) of 600 MW(th) output was sanctioned as the next stage in the development of the LMFBR system in the UK and after some commissioning difficulties in the steam generation equipment the reactor is now operating satisfactorily at power. The policy of reprocessing and refabricating the fissile material content of irradiated fast reactor fuel to demonstrate the feasibility and reliability of the total system was continued and in 1972 it was decided that the irradiated PFR fuel should be reprocessed at

DERE by modifying and extending the existing DFR fuel reprocessing plant for the purpose. Flowsheet and equipment development to support the design of the plant modifications and additions for this fully active demonstration plant have been carried out over the past four years and the modification programme is now nearing completion. Reprocessing of irradiated fuel will commence in the second half of 1977.

The development programme and design studies for PFR fuel reprocessing have identified areas in which direct scale up of the PFR designs would be inadequate or inappropriate for a large scale Commercial Fast Reactor reprocessing plant design. Consequently a parallel development programme has been initiated to provide data for the designs and active demonstration of the size and type of equipment which will be required to recycle the quantities of irradiated fuel produced from an expanding programme of CFR installation in the UK. The size and timing of the fuel cycle plants required to support a CFR programme is entirely dependent upon the initial reactor installation and the programme build up in relation to plutonium availability. For planning purposes the early demonstration of fully active full scale equipment in a plant module sized to support CDFR (perhaps on the same site) has been assumed.

2. REQUIRED REPROCESSING DEVELOPMENT WORK

The principal differences between irradiated fast reactor oxide fuel and thermal reactor oxide fuel are almost entirely attributable to the higher rating, burn-up and plutonium content of the fast reactor fuel.

The higher fuel rating required a fuel sub-assembly design which comprises small diameter pins closely packed within a wrapper or envelope and separated by grids or wire wraps. This form of sub-assembly is extremely difficult to disassemble prior to reprocessing so that the dissolvent can gain access to the contained fuel. (See Fig 1.)

The high burn-up of fissile material leads to a high fission product content in the irradiated fuel resulting in a high decay heat emission and in addition the formation of noble metal fission product alloys which are insoluble in the nitric acid medium normally used for the conventional PUREX type solvent extraction process by which uranium and plutonium are separated from fission products, purified and recovered for recycle. In addition the considerably higher burn-up achieved in LMFBR fuels compared to thermal reactor fuels reduces the evaporation factor attainable on the high level waste raffinate almost in the inverse ratio to the burn-up of the fuels, assuming that the liquor is to remain stable and free from solid precipitates. Thus, although thermal and LMFBR fuels are dissolved to give approximately molar solutions of uranium plus plutonium the limiting evaporation factor for a stable product is governed by the burn-up of the fuel in question.

The high plutonium content of the fuel affects the design of the fuel cycle plant and equipment because of the essential need to avoid accidental criticality excursions in the plants. It is necessary to resort to geometrically limited equipment or the introduction of fixed or soluble neutron absorbers as 'poisons' to control this situation and these constraints can influence the choice of process or equipment. Also the high concentration of plutonium is such that under certain PUREX flowsheet conditions it is possible that a plutonium rich third phase can form in the solvent extraction equipment. This situation must be avoided by careful flowsheet selection.

In addition the continuous recycle of high burn-up irradiated plutonium through the fast reactor core causes significant changes in the isotopic composition of the higher isotopes, eg Table 1 shows the calculated isotopic composition of rejected PFR plutonium.

TABLE 1

PLUTONIUM ISOTOPIC COMPOSITION AFTER IRRADIATION (INNER CORE)

Cycle	Pu238 %	Pu239 %	Pu240 %	Pu241 %	Pu242 %
1	0.14	70.09	24.96	3.86	0.96
2	0.18	66.62	27.86	4.07	1.27
3	0.23	63.81	29.98	4.35	1.57
4	0.27	61.73	31.53	4.62	1.86
5	0.31	60.03	32.67	4.86	2.14
6	0.34	58.71	33.52	5.05	2.40
10	0.40	55.59	35.16	5.60	3.25

This in turn has two major effects:

i the concentration of total plutonium in the fast reactor fuel has to be progressively increased to maintain the reactivity value of the sub-assembly, and

ii the curium 242 and 244 content of the irradiated fuel is increased thus increasing the heat and neutron emissions from the irradiated sub-assembly (see Fig 2).

Because of the characteristics of irradiated LMFBR fuel described above the development work for PFR fuel reprocessing at DERE has concentrated upon devising procedures and equipment to de-activate irradiated sub-assemblies from sodium, transport sub-assemblies from the irradiated fuel store to the reprocessing plant, disassemble the sub-assembly into component pins, chop the pins, leach out the fuel, remove wastes from the fuel breakdown cave and dissolver, clarify the active feed to the solvent extraction plant and treat or dispose of the insoluble residues. Solvent extraction flowsheet development work has been carried out to confirm decontamination factors and recovery efficiencies and to tailor the flowsheet to meet the constraints of the installed solvent extraction contactors.

In addition, the importance of installed instrumentation for monitoring and controlling the plant operating conditions and working environment was recognised and an instrument development programme specifically designed for the PFR fuel reprocessing plant project was initiated in conjunction with staff from the Atomic Energy Research Establishment (AERE) Harwell.

3. PFR FUEL REPROCESSING DEVELOPMENT

The target burn-up for PFR fuel is a mean of 7½% heavy atoms. As a preliminary to the design of the fuel transfer flask and the sub-assembly breakdown cave and equipment for PFR fuel reprocessing detailed calculations were carried out to assess the heat and neutron emissions and total radio-activity from an irradiated sub-assembly. The variations associated with the initial and makeup plutonium isotopiccom-positions, the plutonium enrichment and the number of irradiation cycles undergone and the total cycle time were calculated and a 'worst case' was defined to enable the plant designers to design the shielding against a permissible surface dose rate of 0.5 m rem/hour. The variations are illustrated in the table and the selected worst case was that of 6th cycle inner zone PFR sub-assemblies irradiated to 10% burn-up of contained heavy atoms and cooled for 180 days after discharge from the reactor and before reprocessing. Irradiated sub-assembly handling methods were designed to ensure that the centre pin clad temperature did not exceed 650°C to avoid internal pressurisation and rupture of intact pins. These design parameters enable PFR irradiated sub-assemblies having a heat emission of 3 kW per sub-assembly to be reprocessed and limit the cooling time to not less than 180 days, although it will be possible by special arrangements to investigate lower cooling times and higher heat outputs. Fig 3 shows a com-parison of the calculated heat outputs from PFR and CFR sub-assemblies against decay time.

3.1 Sub-Assembly Breakdown

Fig 1 shows a typical UK design of an LMFBR sub-assembly. The chosen method of disassembly of PFR fuel is for the end pieces to be cut off using a laser in the irradiated fuel caves attached to the reactor. This removes the mixer breeder section as well as the top end piece and it is not intended to reprocess this section at DERE because the plutonium content is low (0.3%). The topped and tailed sub-assembly is then steam cleaned to remove residual sodium canned and transferred to the reprocessing plant. The transfer flask has a water filled annular neutron shield and the heat rejection capability has been designed to accommodate up to 3.5 kW without forced cooling provision.

In the reprocessing plant the sub-assembly is removed from the can and the wrapper is removed by laser cutting The pins are then pulled out from the grids using an hydraulically operated draw table chopped into 2-4 cm lengths into a per-forated basket containing a thin brass containment liner.

The use of a laser for fuel breakdown purposes is novel and the plant unit has been set up in a laboratory situation in order to confirm the focussing arrangement, the attainable rate of cutting and the effects of the laser beam accidentally striking fuel in terms of volatilisation of plutonium and fission products of fusing fuel into an insoluble form. These tests have demonstrated that the 400 w continuous output laser to be installed in the plant can be focussed precisely so that the sub-assembly wrapper (3.25 mm thick) can be cut through and a longitudinal cutting rate of ca 0.5 metre per minute achieved without affecting the pins immediately adjecent to it. The effects of accidentally striking fuel are limited and can be accepted in terms of activity volatilisation or fusion of fuel. The laser is also used to cut sub-assembly hardware into suitable sections for removal from the breakdown cave in sealed 200 litre stainless steel containers which will be placed in retrievable or non-retrievable storage depending upon the measured alpha content of the waste.

3.2 Dissolution

The plant dissolver is a geometrically limited thermal recirculating tubular dissolver and its performance has been checked out by building a full scale glass model and carrying out inactive dissolution trials using heated baskets. Active dissolution has been carried out in a heavily shielded laboratory facility using 100 gm quantities of fuel contained in experimental fuel pins irradiated in DFR to 7½% burn-up of heavy atoms. These pins were not truly typical of PFR irradiated fuel because the fuel manufacturing route, the plutonium content of the fuel and the irradiation history of the pins were different but the fuel was fully active and dissolved readily in nitric acid without leaving any measurable fuel in the cladding.

3.3 Feed Clarification

These active dissolutions also provided gram quantities of insoluble fission products which have been examined and characterised at DERE and Harwell. Extrapolation to PFR conditions has resulted in the following conclusions:

a. the fission product insolubles in PFR fuel irradiated to 7½% mean burn-up will amount to 1% of the fuel charged to the dissolver (see Fig 4).

b. the insoluble material consists of alloys of Ru, Rh, Tc, Mo, Pd.

c. the heat emission of the insolubles will be about 1 watt/gm at 180 days cooling with a half life of about 1 year (see Fig 5).

d. the particle size is in the range 1-15 mm but the particles tend to agglomerate.

e. there is some evidence of the formation of an alloy (U, Pu) $(Pd, Rh)_3$ containing plutonium; in other respects there is in theory no absolute reason that the insolubles should contain plutonium.

f. the measured plutonium content of the samples examined so far does not exceed 0.1% w/w of the original plutonium content of the fuel.

g. there is some evidence that the mass of fission product insolubles is linearly related to fuel burn-up.

3.3 Feed Clarification

The activity and thermal emission of the fission product insolubles even if there is no significant plutonium content are such that it is essential that they be removed from the dissolver solution before it is fed to the solvent extraction process to avoid possible difficulties of enhanced preferential corrosion of the equipment or solvent degradation.

With the assistance of the UK National Tribology Centre a solid bowl high speed centrifuge has been developed with a bowl diameter of 5 cms capable of being accommodated within a geometrically limited in line plant enclosure (See Fig 6). Remote methods of changing the centrifuge bowl have been developed since the bowl will have solids and heat capacity to take only the insolubles likely to arise from one dissolver batch. Methods of handling the loaded bowl, determining the plutonium content and transferring it to a retrievable storage situation have also been developed.

3.4 High Level Liquid Waste Treatment

The first cycle reaffinate solution from the mini mixer settler experiments was subjected to evaporation trials which showed that the solution could be evaporated by a factor of only about 3:1 if fission product precipitation were to be avoided. It is intended to store the high level liquid waste from PFR reprocessing operations in simple cooled and jacketted stainless steel underground which will not be provided with agitation facilities and therefore evaporation will be limited to a factor of say 2.5:1.

3.5 Plant Experiments

The close proximity of the reprocessing plant to PFR will enable selected sub-assemblies specific fuel composition, irradiation history and cooling time to be used for carefully designed plant experiments to investigate dissolution behaviour and flowsheet variations.

It is anticipated that the active operation of the PFR fuel reprocessing plant in the UK will provide a wealth of information on the following areas of LMFBR fuel reprocessing technology.

a. sodium de-activation techniques for sub-assemblies at 3-6kW thermal output.

b. cladding and materials behaviour in head-end operations.

c. actual temperatures and cooling requirements for irradiated sub-assemblies with thermal outputs in the range 3-13 kW.

d. the quantity, thermal output, composition and particle size of insoluble fission product alloys and the nature and quantity of any associated insoluble plutonium from fast reactor fuel of standard composition, fuel manufacturing methods and irradiation history.

e. the effect of fuel fabrication routes and irradiation levels on insolubles.

f. the use of a laser as an operational fuel breakdown method and the quantity and nature of any volatilisation products.

g. the application of high speed contrifugation as a method of liquor feed clarification.

h. solvent degradation effects under long contact times.

i. plant confirmation of various possible solvent extraction flowsheet options.

3.6 Waste Management

The importance of controlling the radioactive wastes which would be produced from PFR fuel reprocessing was recognised during the plant design period and a new and comprehensive waste management scheme aimed at minimising radioactive waste generation and segregating the waste into different categories for treatment, storage or disposal was incorporated into the PFR fuel reprocessing project.

The following principles were adopted for the revised radioactive waste management strategy.

i Solid Radioactive Wastes

a. Waste of alpha content greater than current UK ground burial authorisations should be segregated from other wastes and stored in a readily retrievable form to enable subsequent decontamination and/or plutonium recovery processes to be applied to it.

b. Waste of low alpha content should be disposed of into silos or ground burial areas situated within the DERE site.

c. A development programme should be initiated to investigate waste treatment processes and equipment and to develop instrumentation sensitive enough to measure low levels of alpha contamination.

ii Liquid Radioactive Wastes

a. High level liquid radioactive wastes should be stored in cooled and jacketted underground stainless steel tanks at such a concentration that the precipitation of insoluble fission products will be avoided.

b. Low level liquid radioactive wastes should be subjected to an effluent treatment process which would reduce the radioactivity content substantially before discharge to sea.

c. Development work should be carried out to confirm the applicability of the British Nuclear Fuels Limited (BNFL) HARVEST vitrification process to fast fast reactor fuel reprocessing wastes and to evolve methods of treating and packaging for disposal the solids arising from the effluent treatment process.

iii Gaseous Radioactive Wastes

Revised ventilation systems should be installed minimising ventilation flows and using fluidic control devices to reduce the volume of gas to be treated. All exhaust gas flows from the facilities should be monitored and treated before discharge to atmosphere.

Facilities and procedures based on these principles have been introduced at DERE in support of the PFR fuel reprocessing project and data resulting from these radioactive waste management operations will also be available in support of the design of future CFR fuel reprocessing plant including all solid, liquid and gaseous radioactive waste treatment, packaging, storage and disposal requirements.

3.7 Instrument Development

The instrument development programme has resulted in the production of prototype instruments which are capable of remotely measuring the concentration of uranium and plutonium in active liquors without breaching plant containment and this type of instrument is being installed in the reprocessing plant for evaluation and refinement under active operating conditions. The same principle can be used for determining liquor levels and the interface between organic and aqueous phases and this application will also be investigated under plant conditions. Instruments capable of measuring very low levels of plutonium contamination on solid wastes in the presence of significant quantities of fission product activity have also been developed successfully and will be installed as part of the waste management scheme described above. Personal instruments

designed to monitor neutron dose rates and air contamination levels are also at an advanced stage of development.

3.8 Further Development

Nevertheless the plant design and process and equipment development work carried out for the PFR fuel reprocessing project have also identified areas in which further work will be required to enable a large scale LMFBR fuel reprocessing plant to be designed to operate with high availability and reliability in support of a substantial LMFBR programme.

4. CFR FUEL REPROCESSING DEVELOPMENT

On present evidence it is judged that a conventional solvent extraction process will be capable of reprocessing highly rated high burn-up LMFBR fuels although it may be necessary to adjust the flowsheet or equipment design to minimise solvent aqueous contact times in the plant. The other major problems which must be tackled to provide adequate design data for a large LMFBR fuel reprocessing plant are mainly related to the unit operations needed to cater for fuel of higher rating and burn-up than the current designs and the reduced cooling time before reprocessing which may be considered necessary for improved economics or uranium conservation. Consequently the UK has embarked upon a parallel development programme to supplement the information which will come from the PFR fuel reprocessing by carrying out design studies on alternative methods of fuel disassembly, dissolution and solid waste treatment which will culminate in full scale testing of the preferred methods and equipment resulting from these design studies. Further solvent extraction flowsheet and equipment studies will be made if PFR fuel reprocessing experience show these to be necessary.

The development programme includes:

i heat transfer aspects of sub-assembly handling and storage and transport in a variety of media.

ii consideration of alternative sub-assembly breakdown techniques including mechanical and chemical alternatives to laser cutting.

iii alternative fuel exposure methods and equipment related to dissolver feed mechanisms and dissolver design.

iv assessment of alternative feed clarification methods.

v flowsheet and equipment development studies for a solvent extraction system aimed at a total liquid recycle and minimum environmental discharge specification.

vi consideration of additional liquid and gaseous waste treatment and recycle methods.

vii hazard assessment of the preferred process and equipment.

The timing, size and siting of the first CFR fuel reprocessing plant module will be influenced by social, economic and technical factors related to the fast reactor installation programme. In these circumstances although it has been calculated that a single line of solvent extraction equipment could process up to 300 t (U+Pu) per annum (equivalent to the core fuel throughput from 15-20 GW(e) of installed fast reactors) present thoughts are that a full scale demonstration of the system should include a CFR and a single module of full scale equipment for fuel reprocessing and fabrication plants. It is considered that a 50 t reprocessing plant module would enable the full scale head and equipment to be proved and its reliability assessed before a large plant was needed. Such a plant module would also provide information on the related aspects of demonstration of satisfactory safety and environmental discharge from the fast reactor fuel cycle.

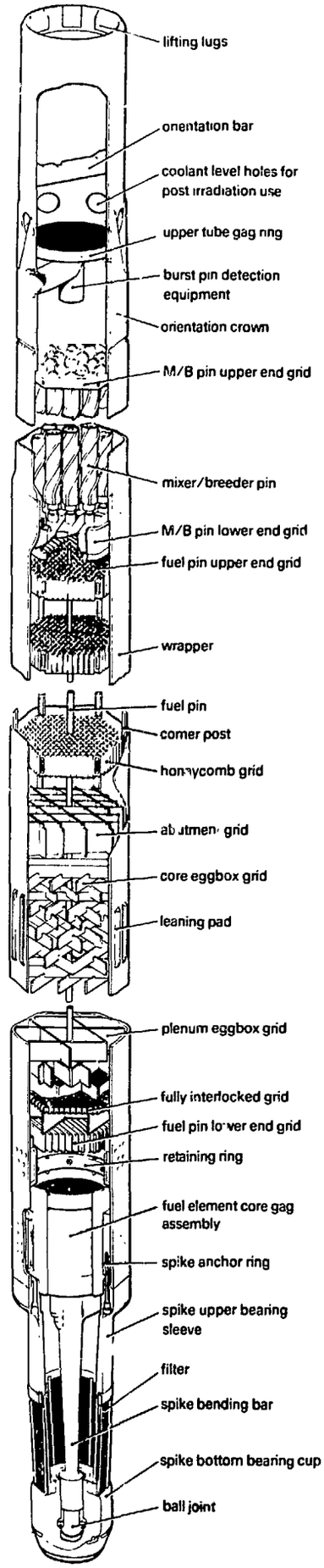

FIGURE 1
PFR SUB ASSEMBLY

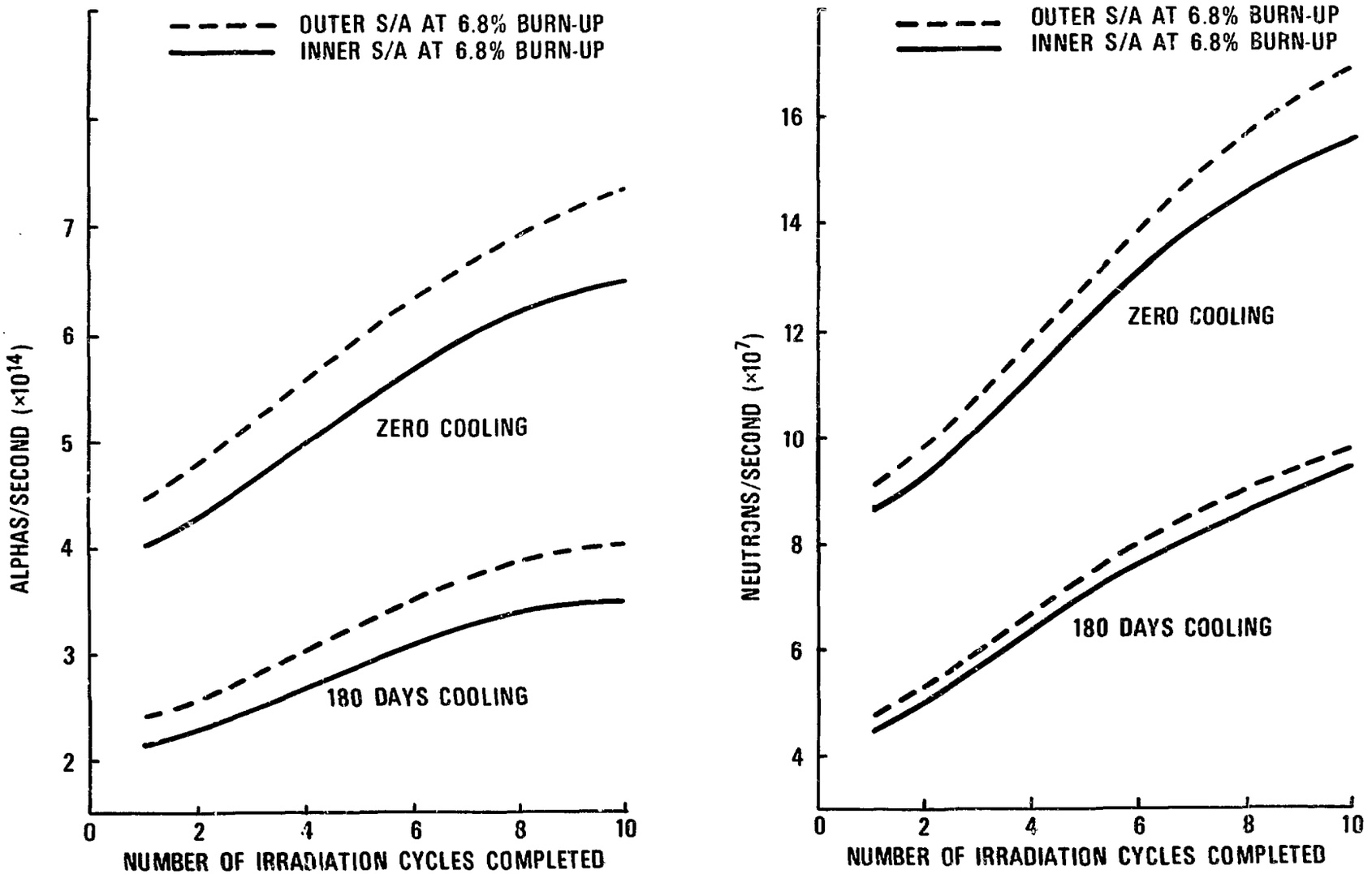

Fig.2. ALPHA AND NEUTRON EMISSION FROM A SUB-ASSEMBLY AT MEAN DISCHARGE BURN-UP

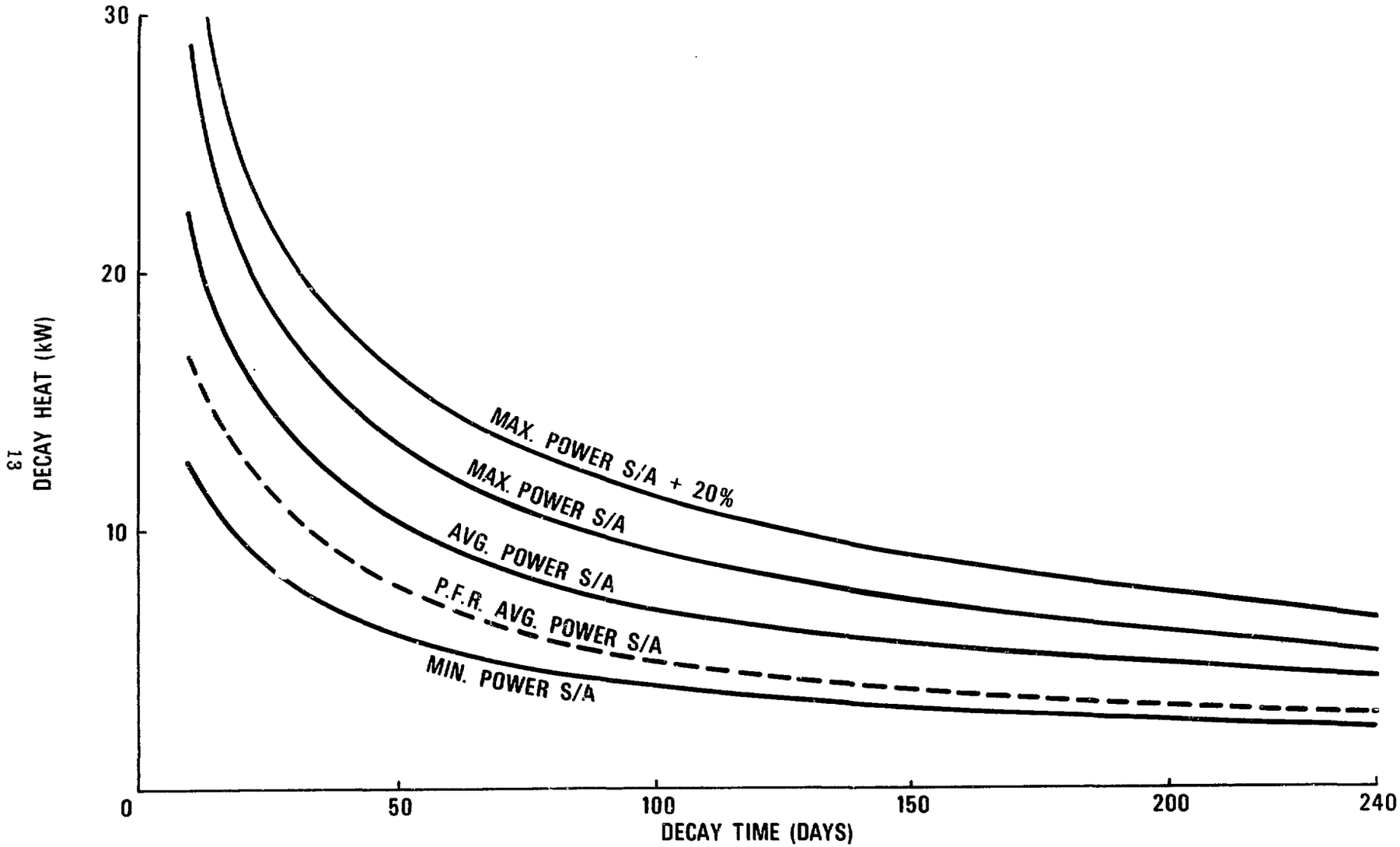

Fig.3. C.F.R. FUEL SUB-ASSEMBLY DECAY HEAT 10 TO 240 DAYS.

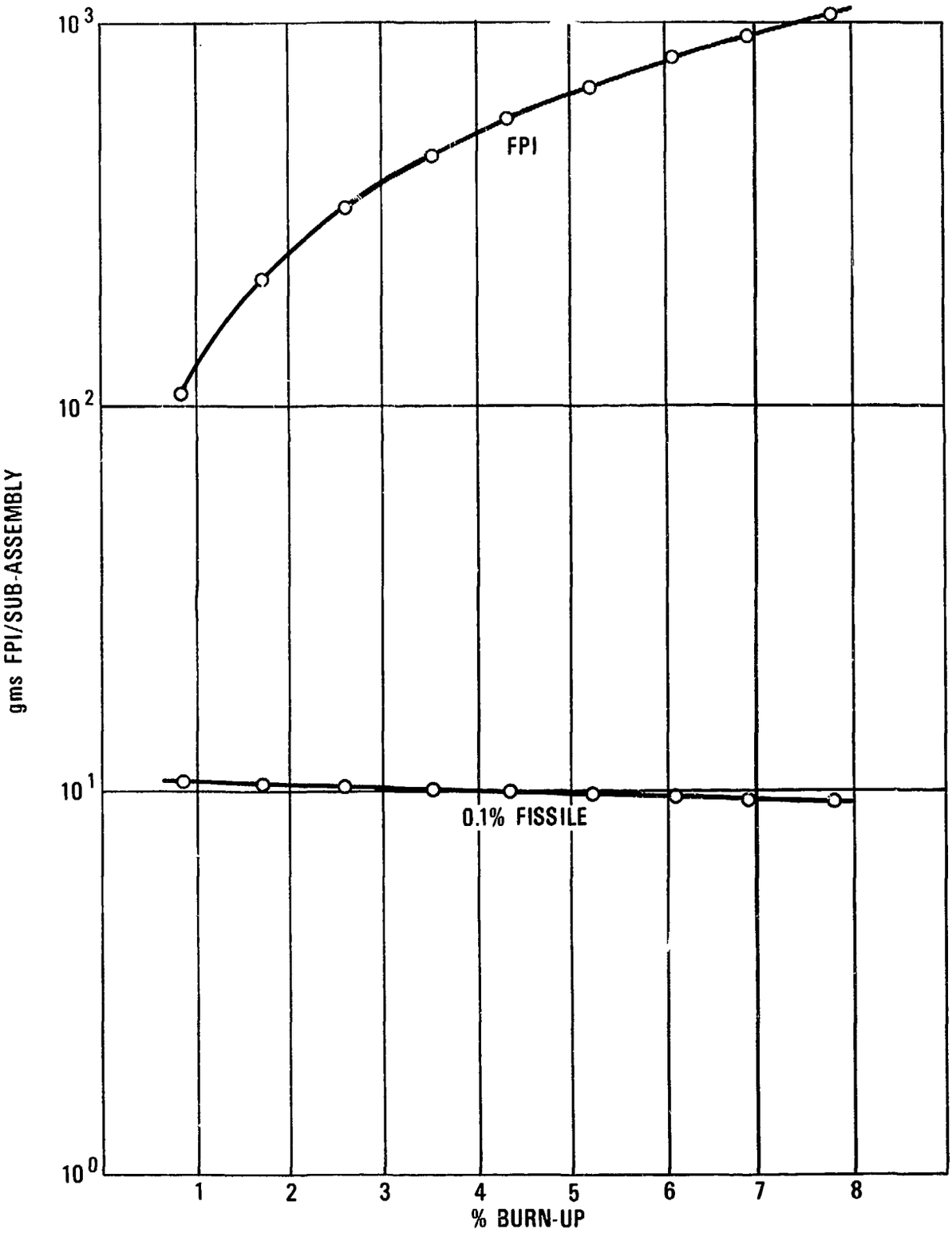

Fig.4. MAXIMUM POSSIBLE YIELD OF FPI FROM 46.3kg INNER SUB-ASSEMBLY. (WORST CASE AT 200W.g^{-1} 1% TO 8% BURN-UP)

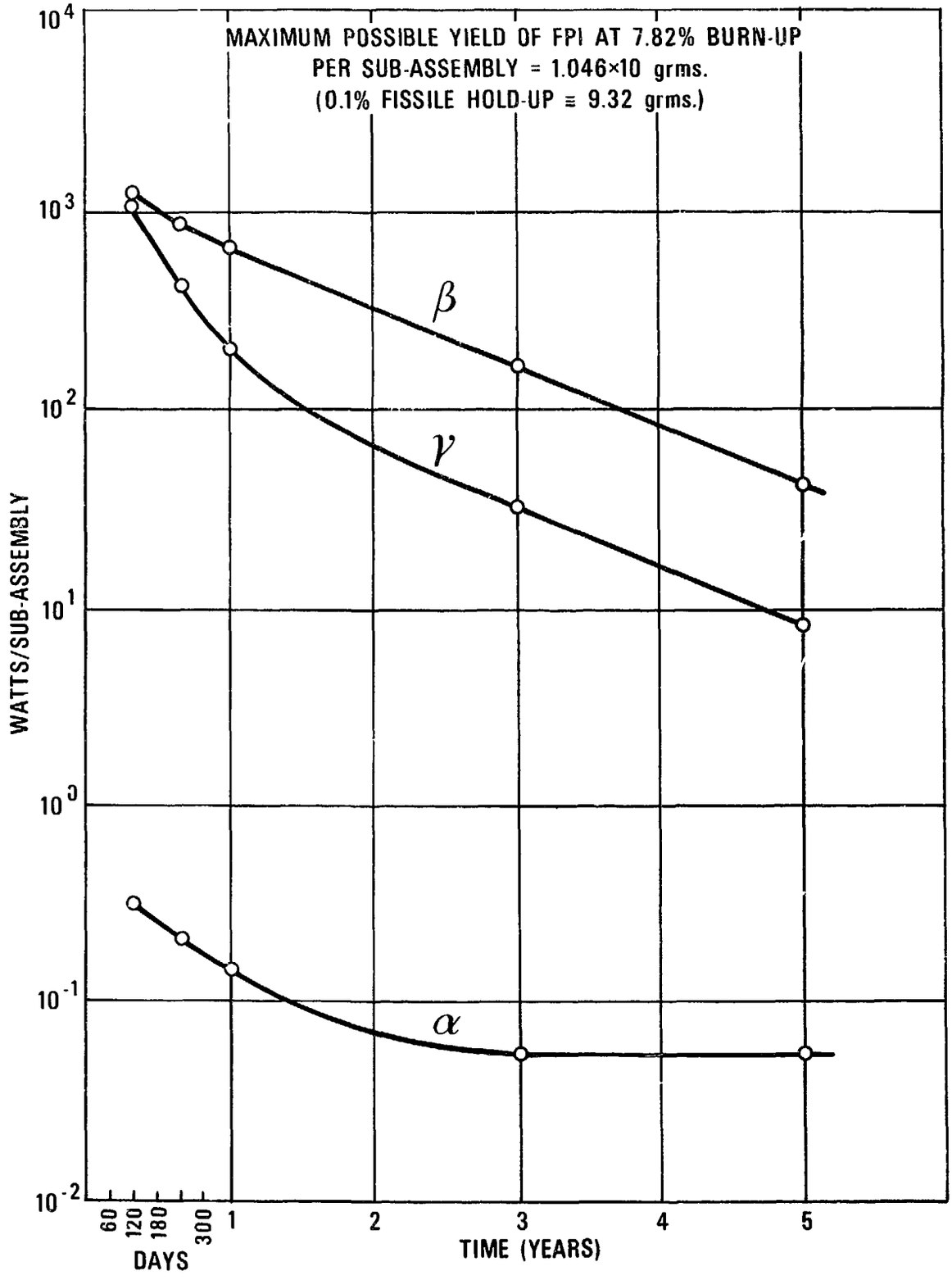

Fig.5. HEAT OUTPUT OF MAXIMUM POSSIBLE YIELD OF FPI FROM 46.3 kg INNER CORE SUB-ASSEMBLY. 120 DAYS TO 5 YEARS COOL. (WORST CASE AT 200 $W.g^{-1}$ TO 7.82% BURN-UP)