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SAFETY CONSIDERATIONS IN THE FAST REACTOR FUEL CYCLE

by

A R Baker, U K Atomic Energy Authority
W R Burton, U K Atomic Energy Authority
H A Taylor, British Nuclear Fuels Ltd

1. SYNOPSIS

The fuel cycle safety problems for fast reactors, as compared with thermal reactors, are enhanced by the higher fissile content and heat rating of the fuel. Additionally recycling leads to the build-up of substantial isotopes which contribute to the alpha and neutron hazards. The plutonium arisings in a nuclear power reactor programme extending into the next century are discussed.

A requirement is to be able to return the product plutonium to a reactor about nine months after the end of irradiation and it is anticipated that progress will be made gradually towards this fuel cycle, having regard to the necessity for maintaining safe and reliable operations.

Consideration of the steps in the fuel cycle has indicated that it will be best to store the irradiated fuel on the reactor sites while I 131 decays and decay heat falls before transporting and a suitable transport flask is being developed.

Reprocessing development work is aimed at the key area of fuel breakdown, the inter-relation of the fuel characteristics on the dissolution of the plutonium and a solvent extract cycle leading to a product suitable for a co-located fabrication plant.

Because of the high activity of recycled fuel it is considered that fabrication must move to a fully remote operation as is already the case for reprocessing, and a gel precipitation

process producing a vibro-compacted fuel is under development for this purpose.

The waste streams from the processing plants must be minimised, processed for recovery of plutonium where applicable and then conditioned so that the final products released from the processing cycle are acceptable for ultimate disposal.

The safety aspects reviewed include protection of operators, containment of radioactive materials, and regulation of discharges to the environment.

2. INTRODUCTION

The increasing need for nuclear power will be met, in most developed countries, first by thermal reactors and later by fast reactors. When the initial stock-pile of plutonium, produced in the first thermal reactors, is used up, the rate of installation of fast reactors will be determined by the plutonium production rate and there will be an incentive to reprocess short-cooled fuel. The additional safety considerations for a power programme including fast reactors arise from the need to reprocess short-cooled fuel and from the large-scale production of plutonium and some higher actinides.

The paper begins with a discussion of the characteristics of spent fuel to provide the basis for the remainder of the paper. There follows a brief indication of how the risk analysis that has been suggested for application to reactor safety might be adapted to cover the safety of transport, reprocessing and fabrication. Since ultimately the safety of chemical plants depends on their careful and reliable operation, the paper concludes with a discussion of the principles to be adopted for the design and operation of reprocessing and fabrication plants.

3. CHARACTERISTICS OF FAST REACTOR SPENT FUEL

Calculations have been performed for an illustrative UK nuclear power programme which provides for a rapid build-up of fast reactors near the turn of the century and gradual replacement of thermal reactors. It is assumed for the calculations that there is a common reprocessing plant in which all plutonium is isotopically mixed and americium and curium isotopes are rejected from the cycle. The results are displayed in Fig 1 where various quantities are plotted against the cumulative electrical energy generated in the programme. This choice of abscissa enables general conclusions to be drawn that do not depend on the exact form of UK programme assumed. For the particular programme assumed for the calculations, a cumulative electrical energy generation of 270 GW years corresponds to a generating capacity of 50 GWe and 1000 GW years corresponds to 135 GWe.

The gradual build-up of plutonium in the system (in stocks, reactors and spent fuel) is shown in Fig 1 for three variants of the illustrative programme as follows:

a. the base programme of fast and thermal reactors, with the installed capacity sub-divided as in the upper curve;

b. as a., but with additional thermal reactors substituted for the fast reactors and no recycle of plutonium in thermal reactors;

c. as a., but with additional thermal reactors substituted for fast reactors and plutonium recycled in thermal reactors.

The three curves show a substantical build-up of plutonium in the system whether fast reactors are introduced or not. The total system inventory is only reduced appreciably by the recycling of plutonium in thermal reactors involved in variant c.

The arisings of the important long-lived alpha-emitting higher actinides from the base programme, variant a., are compared with those from variant c., an alternative all-thermal-reactor programme, in the lower curves of Fig 1. The consumption of plutonium by recycling in thermal reactors produces for disposal appreciably more americium (Am 241 and 243) and curium (mainly Cm 244) than in the thermal-fast system. The uranium-fuelled thermal-reactor system without reprocessing (variant b) produces much more of heavy elements for disposal in the form of unusable plutonium.

Though the thermal-fast system as a whole creates less americium and curium, the spent fuel from a fast reactor contains a higher ratio of these elements to fission products than fuel from U-fuelled thermal reactors. The activities from americium and curium must therefore be taken into account in fast reactor fuel operations; for example, there is additional decay heat from the alpha activity and the spontaneous neutron emission leads to a requirement for additional shielding. The small content of Cm 242 can dominate the hazard in the unlikely event of vaporisation of spent fuel in an incident. As the irradiation period in a fast reactor is a factor of 2 or 3 smaller than in a thermal reactor, a smaller fraction of fission products decays during the irradiation and so spent fuel from a fast reactor has higher decay heat and activity per unit of reactor power in the first few years of cooling.

In summary, three special features of fast reactor fuel processing are:

a. the alpha and neutron activities due to americium and curium in spent fuel;

b. the high heat ratings in the early years of fuel cooling;

c. the high throughputs of plutonium.

The required break-down of the elapsed time for the fuel cycle, corresponding to a total out-of-reactor time of nine months, as shown to be desirable by economic studies, is given in Table 1. The restrictions at the various stages due to the

high activity or heat output are indicated in the final column.

4. RISK ANALYSIS

A probability approach to risk analysis for reactors has been suggested by F R Farmer (1, 2) which makes use of the risk-consequence curve reproduced as Fig 2. Although the application of this approach in reactor licensing has not yet been agreed with responsible authorities in the UK, it is instructive to examine the implications of its extension to cover the fast reactor fuel cycle. It would require the expression of incident severity in terms of equivalent I 131 release.

Fairbairn et al (2) have suggested a method for calculating factors to use for calculating equivalent amounts of I 131 based on the ICRP recommendations for maximum permissible concentrations in air. According to this method (but with re-evaluated numerical values), 1 Ci of I 131 is equivalent to 0.07 g of Pu 239 or 0.024 g of plutonium from an Advanced Gas-cooled reactor (AGR). The contributions to the equivalent I 131 activity of an irradiated sub-assembly initially containing approximately 11 kg of AGR plutonium, for a decay period of 10^7s, are as follows:

Pu	0.5 MCi I 131
Am + Cm	2.9
fission products	0.8
	4.2

The large contribution from Cm arises from the assumption of four years' storage of the plutonium before irradiation.

As an illustration, examination of Fig 2 shows that the required probability for an incident allowing the release of 1% of the contents of a sub-assembly after a decay time of 10^7s is $p = 2 \times 10^{-5}$ per reactor-year. This probability would have to be made more remote when applied to a transport flask to allow for passage by railway through heavily populated areas. Similarly, the probability of an incident allowing the release of 1% of the equivalent of a whole core (300 SA) in, say, a storage pond is $p = 10^{-7}$ per reactor-year. Although this value is similar to that required for a reactor whole-core incident, there are fewer sequences available for a major accident in the course of the fuel cycle since most fault sequences analysed lead to damage only within the plant.

It seems feasible to produce containments during transport and processing consistent with the probability estimates in the previous paragraph; strong basic structures will be provided in any case for gamma shielding.

5. TRANSPORT

The main requirements in the design of the transport flask

are containment, shielding, impact resistance and fire resistance. In order to provide for an out-of-reactor time of nine months, transport must be possible after about 160 days (Table I). The UK proposals for a five sub-assembly flask to be carried by railway were published in 1972 (3). To provide a greater margin on the total heat dissipation of 40 kW, the design has been reworked recently to accommodate only three sub-assemblies.

Consideration is being given to higher standards for surface dose rates than required in the current IAEA regulations (4) to meet the requirements of the reprocessing plant since flask handling could be the full-time employment of some workers.

6. FUEL PROCESSING: INTRODUCTION

The background against which the UK requirements for future fast reactor fuel processing are considered includes the continuing build-up of thermal fuel reprocessing technology and the small-scale plutonium fabrication operations for PFR fuel. A recent paper by Franklin (5) reviews the broad field. The introduction of the fast reactor fuel cycle involves two new main issues, as compared with a thermal reactor cycle:

a. The breakdown, dissolution and separation of a more active irradiated fuel with a higher heat output; and

b. The refabrication of the plutonium into fuel (together with treatment of plutonium-contaminated materials) on a larger scale than at present.

When considering the development programme, the complete fuel cycle must be taken into account. It is convenient to consider that there will be one plant, under one management, which receives irradiated fuel assemblies and produces new fuel assemblies. While this gives security advantages which are outside the scope of this paper, there are also significant advantages in respect of inter-stage product specifications, plutonium handling procedures and integrated process control.

Additionally, since the nature of the fuel to be reprocessed and fabricated can affect the viability of processing operations, it is necessary to consider the best balance that can be found - that is, established with continuing practical experience - between the sometimes competing requirements of fuel performance and fuel plant performance.

7. FUEL REPROCESSING

The safety aspects of reprocessing are also discussed in a paper by Donoghue et al (6). Process development considerations and the operations to be carried out for the reprocessing at Dounreay of PFR fuel are covered in a paper by Allardice et al (7). The forward development programme includes:

a. consideration of alternative sub-assembly breakdown techniques, including mechanical and chemical

alternatives to the laser cutting used in the plant for PFR fuel.

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

In addition to the planned process development work, consideration will be given to the inter-relation with fuel design and the fabrication plant operations. The four examples given below are typical of the future points to be considered.

A key parameter in the operation of a reprocessing plant will be the heat output per sub-assembly at break-down. Table I, based on preliminary economic assessments (necessarily preliminary because of the limited amount of practical work), indicates that break-down will take place after about 186 days cooling at 6.3 kW per sub-assembly. The first fast reactor reprocessing plant will initially operate during a period of plutonium surplus, such that the case for rapid turn-round of plutonium does not apply, and there is little doubt that reprocessing operations will commence with relatively long-cooled fuel so that experience can be gained under the easier operating conditions of a lower heat output. As such experience is acquired, the cooling time will be progressively lowered, but a careful balance will be maintained between the advantages and disadvantages of rapid turn-round.

A compromise also has to be found in the requirements in the fuel design set by reactor operation on the one hand and by fuel break-down, dissolution, and the monitoring, treatment, packaging and disposal of waste products on the other. The possible methods for fuel break-down include: shearing a complete sub-assembly together with the wrapper; removing the wrapper from a gridded sub-assembly and then shearing; or shearing small bundles of pins taken from a sub-assembly containing wire-wrapped pins.

The effect of the fuel design on fuel dissolution is also receiving careful consideration. It appears likely that insolubles in unirradiated fuel will not improve during irradiation and thus a good fuel solubility in as-fabricated fuel is required in order to assist reprocessing operations. In general, this is obtained from a high standard of micro-homogeneity, which in turn is obtainable by a wet-blending process giving a solid solution, such as by co-precipitation or gel precipitation, or by an adequate milling process for mixed PuO_2 and UO_2.

The feed material for a wet fabrication process is a mixture of plutonium and uranium nitrates. For the integrated processing plant discussed above, this is also the end-product of the reprocessing operations, and steps may be taken, as practicable, to process plutonium in the liquid phase as rapidly as possible into finished fuel. Thus, the reprocessing operations may be optimised to produce the required nitrate mixes, either by complete separation and re-addition of uranium or by some partial separation.

8. FUEL FABRICATION

The conventional process for the manufacture of fast reactor fuel is derived from the considerable experience of uranium pellet-fuel fabrication, modified with the object of containing plutonium-bearing powders within the primary containment of the processing equipment, the secondary containment of the glove-box, and the tertiary containment of the cell or building. Experience is, however, that plutonium powders are not adequately contained by the equipment and transfer items, and the glove-box becomes the primary containment. Consideration may therefore be given as to whether the processing of a dusty powder is the best approach to mixed-oxide fuel manufacture, taking note of:

a. the requirement to minimise operator dosage;

b. the dispersibility of PuO_2 powder;

c. the contamination of materials within the glove-box and their subsequent disposal as waste;

d. the future recycling of more active plutonium; and

e. the future applicability of a powder process to the manufacture of carbide fuel, via, say, a pyrophoric carbide powder.

An alternative approach is to select a plutonium conversion process for the specific purpose of eliminating powder handling, as far as practicable, and then to examine the nature and performance of the fuel that results from this process.

A typical example, now being examined in the UK, is the gel precipitation process, where mixed nitrates, conditioned with a gelling agent, are precipitated directly to microspheres. A possible disadvantage is the lower smear density normally obtainable with particulate fuel as compared with pelleted fuel. However, the reduced number of process stages, the wet nature of the operations, and the non-dusty properties of the product could reduce the contamination within glove-boxes and alternative containment designs, and hence operator dosage and arisings of plutonium-contaminated materials. The gel granules have a relatively high resistance to dispersal when compared with powders and have the additional security advantage of a diluted plutonium content as compared with PuO_2.

In the longer term, gel precipitation offers the possibility of fully remote operations to alleviate dosage from more active plutonium, and a reduction in inter-stage storage of plutonium owing to the reduced number of operations. The gel precipitation process may be applied to the manufacture of carbide fuel. An oxide pilot plant is under construction at Windscale to make fuel for irradiation proving in PFR.

Irradiation proving of UO_2-fines fuel, where the PuO_2 is restricted to the larger granules in vibro-compacted fuel, is

in hand. The objective is to reduce plutonium handling operations to the minimum.

9. PLUTONIUM STORAGE

Currently, plutonium arising from reprocessing is stored as PuO_2 powder until required for fuel fabrication. Three alternative fabrication routes may be considered for PuO_2, which has been stored for a number of years:

a. Process directly to pellets without removal of the in-grown americium, with the possibility of increased dosage levels;

b. Dissolve the stored PuO_2, extract the americium and re-convert to PuO_2 for fuel fabrication. This involves double handling of plutonium with penalties of cost, dosage and plutonium-contaminated materials;

c. Dissolve the stored PuO_2 for wet processing to fuel with or without americium removal, again involving increased processing.

An alternative that may be considered is to fabricate the plutonium, as it arises from reprocessing, directly into a suitable fuel type for storage, with the object of minimising subsequent plutonium operations. This concept might call for the establishment of a reactor core design that could use stock compositions of mixed oxide granules or pellets, without the need for further processing. For adjustment purposes when the stock is in the form of granules, additional small and large granules as required could be made from newly separated plutonium at the time of fuel canning. Increased radiation levels in the pin-filling plant and higher curium levels in the reprocessing plant would have to be taken into account. During the period of run-down of the plutonium stock-pile, when this concept applies, irradiated fuel could be cooled for longer periods than normal to reduce the curium level.

10. PLUTONIUM-CONTAMINATED MATERIALS

The fabrication of mixed oxide fuel on the commercial scale will give rise to plutonium-contaminated materials (PCM) and these require careful consideration. The philosophy adopted in the UK for PCM is that not only must arisings be minimised but the contained plutonium must be recovered to the maximum extent practicable, leaving the minimum quantity of plutonium unavoidably remaining as waste for eventual disposal. In support of this philosophy, development is being directed towards the reduction of PCM arisings, improved plutonium measurement techniques, plutonium recovery methods and conditioning treatments for storage and disposal.

Reduction of PCM arisings is being pursued by the introduction of improved design and operational techniques, including the development of the gel precipitation process as above.

Techniques for the recovery of plutonium are under investigation, and a pilot plant incinerator is in active operation at Windscale and a complementary ash-leaching pilot plant is under construction. Alternatives such as acid digestion and pyrolysis are also being examined.

Long-term retrievable storage for PCM will be required to stock the material until the plutonium recovery processes are available on a production scale. The planned stores will provide safe, secure, ventilated storage with facilities for fire and criticality detection.

In keeping with the integrated fuel cycle approach discussed above, the processing of PCM for recovery of plutonium is visualised as a part of the reprocessing and fabrication operation and in this sense is analogous, for both siting and management purposes, to the recycle and recovery of scrap fuel.

There are two main options for the disposal of very low level PCM which has a plutonium content too low for recovery operations or which may have resulted from such recovery operations. These are disposal in deep geological formations and disposal in the deep ocean. No suitable geological disposal site has been identified within the UK, although investigational work has begun. For the sea dumping of future quantities of PCM, the UK is discussing with other NEA member states the ways in which that organisation's experience can best be utilised. It is hoped that these discussions will lead to the establishment of agreements which will ensure that appropriate categories of PCM can be disposed of to the deep ocean with negligible additions to the naturally occurring alpha-activity in man's environment or in marine resources.

11. CONCLUSIONS

The large-scale use of plutonium is a necessary consequence of a large nuclear programme, though the total inventory is somewhat greater if the plutonium produced in thermal reactors is used in fast reactors rather than recycled in thermal reactors. The production of the higher actinides, americium and curium, for disposal is smaller when fast reactors are included in the programme.

The probability approach to safety assessment should be useful in future consideration of fuel cycle plants. The probability of serious incidents involving widespread activity should be extremely low because of the substantial shielding provided for most operations.

The UK's considerable expertise in processing thermal reactor fuels is being extended to cover the special features introduced by fast reactors: the high decay heat and activity of irradiated fuel and the processing of substantial quantities of plutonium. Priority is being given to establishing the preferred balance of fuel performance and fuel plant performance, to minimising the number of plutonium processing operations and to developing safe and reliable fuel cycle plant. Areas

identified for attention include fuel break-down, fuel dissolution, fuel fabrication, plutonium storage and plutonium-contaminated materials. The anticipated long-term outcome is a fully remote reprocessing and fabrication facility with integrated processing of plutonium-contaminated materials.

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TABLE I

Estimates of the time required for each stage of the fuel cycle

Description	For average fuel, elapsed time (days)	Heat output of SA (a) (kW)	Restriction
Despatch			
Shut-down	0		
Reactor ready for refuelling	4	43	
Unload SA, store at reactor site	6	37	
Transport			
Insert SA into welded canisters	145		
Load canisters into flask	159	6.9	For heat dissipation by natural circulation from the external surface the heat output for a flask containing 3-5 SA must be limited to about 40 kW.
Despatch flask	161		
Receipt and storage			
Receive flask at processing plant, unload and store canisters under water	166		At present, steam-cleaning can only be done satisfactorily for a least output per SA of a few kW.
Remove canisters and steam-clean SA, followed by storage of bare SA under water	181	6.3	
Disassembly and shearing	186	6.2	The risk of melting following a break-down of cutting equipment will limit the allowable heat output of fuel.
Possible fuel pre-treatment			
Removal of tritium	191		
Fuel dissolution	196		Decay heat will complicate the process for leaching of fuel.

Description	For average fuel, elapsed time (days)	Heat output of SA (a) (kW)	Restriction
Solvent extraction	201		Decay heat complicates the design of the solvent extraction process, especially when dealing with a plant stoppage.
Control of plant effluents	-		The consequences of FP dispersion can be reduced by allowing an elapsed time sufficient for I131 decay.
Refabrication			
Manufacture fuel	206		
Store SA at processing plant	227		
Despatch new fuel	238		
Reloading into reactor			
Store SA at reactor site	241		
Load SA into reactor	271		
Prepare reactor for start-up	273		
Start up reactor	274		

a. The heat output given is for a maximum-rated SA, containing 60 kg of mixed oxide fuel, with 2 significant figures given to show the dependence on elapsed time.

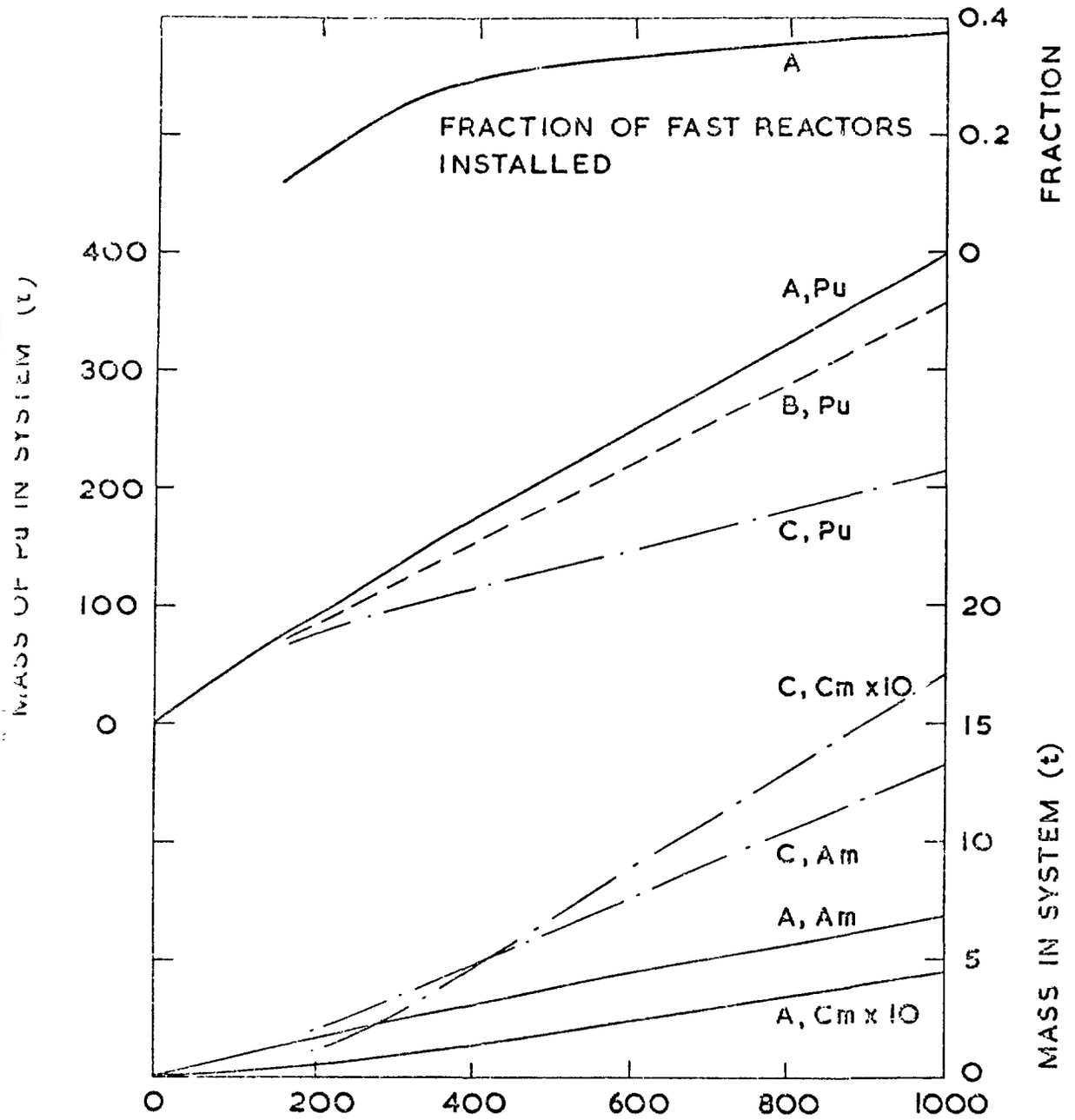

FIG. 1 INVENTORIES OF Pu, Am AND Cm IN A UK POWER REACTOR PROGRAMME

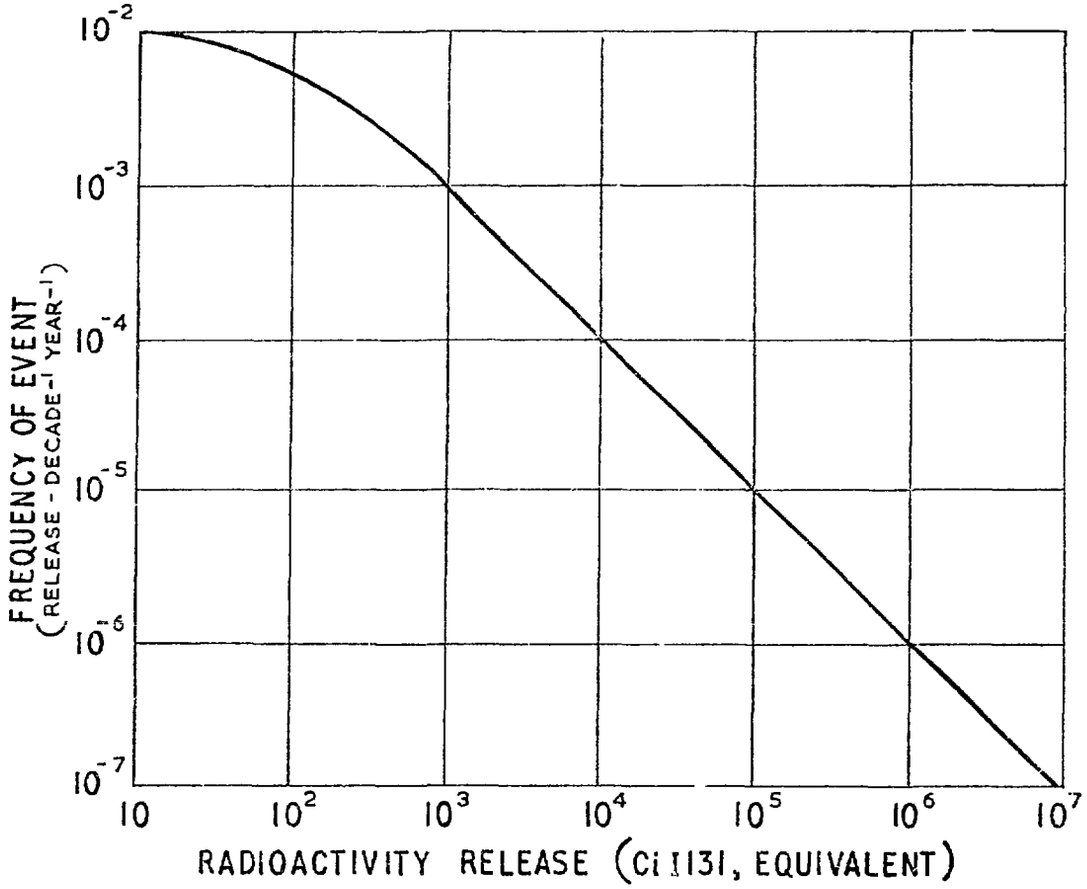

FIG.2. PROPOSED DEPENDENCE OF FREQUENCY OF OCCURRENCE ON MAGNITUDE OF RADIOACTIVITY RELEASE