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

INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA-CN-36/366

NUCLEAR TRANSMUTATION OF ACTINIDES OTHER THAN FUEL AS A RADIOACTIVE WASTE MANAGEMENT SCHEME

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1. THE PRINCIPLE OF NUCLEAR TRANSMUTATION

Nuclear transmutation might be applied for destroying radioactive nuclei if the natural decay process of selected nuclides can be accelerated noticeably and the generated radionuclides will have a shorter half-life and a smaller radiotoxicity than the original nuclides.

The quantity of transmuted nuclei ΔN of any isotope N may be described by the equation

$$\Delta N = (\lambda + \sigma\phi)N\Delta T$$

where the first term on the right hand side represents the natural decay whilst the second one corresponds to the reactions caused by the nuclear process under consideration. In the case of neutron reactions, σ means the sum of the effective capture and fission cross section and ϕ the effective neutron flux. Since σ is a natural constant for a given neutron spectrum, the only variables which influence the quantity of transmuted nuclei are the level of the neutron flux and the irradiation time ΔT. A feasibility study on nuclear transmutation involves consequently

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- the search for transmutation devices with neutron fluxes as high as possible,
- the search for devices with long operation periods without interruption, in order to diminish losses during reprocessing,
- the search for devices in which σ reaches optimum values, where optimum refers to the largeness as well as to the ratio of capture to fission and the number of neutrons released per fission.

The possibility of "burning" some selected fission products by recycling them through reactors was studied first by Steinberg and co-workers in 1964 [1]. Later, it was thanks to Claiborne that the concept of an ultimate disposal for higher actinides by transmutation was thoroughly discussed [2].

As nuclear transmutation devices, the fission reactor, intense accelerators (spallation reactor), and the controlled thermonuclear reactor (fusion reactor) have been proposed in literature. A survey of the problems involved in these techniques is described in [3].

In this paper, the discussion on transmutation is limited to fission reactors, firstly because at this stage they are the only devices available not requiring special additional development work and, secondly, because it seems to be attractive to realize the principle of auto-sufficiency. Because of the long decay chains with partly highly toxic daughter nuclei in the chain, transmutation of higher actinides will mainly aim at transmuting them to fission products.

2. REASONS FOR TRANSMUTATION OF ACTINIDES OTHER THAN FUEL

Up to now the management of high-level radioactive waste (HLW) resulting from the operation of a reprocessing plant for irradiated fuels has not yet been definitively resolved. It is true that the disposal of HLW in deep geological formations after solidification is considered to be the most likely concept to be applied. On the other hand, there are arguments which could favour an alternative waste disposal policy consisting of

- fractionating the HLW into individual categories with specific properties;
- nuclear transmutation of the long-lived species, and species with either long-lived daughter nuclei in their natural decay chains or strong radiotoxicity, to less hazardous ones, and
- geological disposal of the remaining wastes.

The arguments in favour of this disposal concept are:

- The bulk of fission products in the HLW decays to innocuous hazard levels within a time period of about one thousand of years depending slightly on the type of reactor operated and the considered pathway of radioactive material to man, whereas many actinides other than fuel represent hazards for geological time periods. This fact is illustrated by fig. 1 where ingestion has been assumed as a pathway to man. Nuclear transmutation of this category of waste would ensure that we will bequeath the radioactive waste to be generated by our nuclear energy production to

a minimum number of future generations of mankind and burden them potentially with the smallest possible risk. It corresponds best to the principle that the influence of technological processes on the natural equilibrium of the ecological system has to be kept "as low as practicable".

- It is quite probable that risk analyses demonstrate that the safety of repositories in selected underground geologic formations is high enough for some thousands of years. However, it is debatable whether these analyses of the tectonics of geologic formations can provide results that will remain valid over geological time-periods. This is,firstly, because an understanding of the evolution of the Earth in terms of glacial ages is necessarily rather difficult, and the effect of occurring natural phenomena is predictable only with smaller reliability. Secondly, the prognoses of the requirements of future generations of mankind after 10,000 years, and later,to the ecological system, and from this to the acceptable risk for those men, are completely speculative.

- Also under the assumption that geological disposal has been proved to be sufficiently safe for the entire disposal time, there remains still an incentive for actinide transmutation. In fact, the safety of the disposal system will be determined by a probabilistic model: The risk of an individual failure of the system is the product of the consequences of the failure and the probability of its occurrence, where the probability of the failure is given per time unit, for instance per year. Consequently, the time-integrated risk that a disposal system will fail grows with the length of the necessary storage time of the waste. In addition to these arguments in favour of transmutation, there are further ones for partitioning the HLW:

- The safety requirements of the repository (burial ground) could be less incisive if alpha-free wastes have to be disposed of, i. e. wastes which contain alpha emitters in concentrations smaller than 10 nCi/g.

- Higher actinides may be considered as valuable materials for remote terrestrial and space application as power supplies, in medicine as cardiac pacemakers and as radiation sources for the treatment of tumors, and finally in industry for neutron radiography.

An estimate of the magnitude of the actinide problem is given in [4]. Assuming a forecast of the nuclear power generation in the member states of the European Communities, it has been demonstrated that the accumulation of Np, Am and Cm in the waste during the period from the year 1976 through 2000 will amount to about 54 t, 18 t and 4 t, respectively. Obviously, the specific generation rate per ton of irradiated fuel varies with the burn-up rate and the operated reactor type. Typical concentrations for irradiated LWR fuels are 480 g Np, 140 g Am, and 40 g Cm per ton.

3. SURVEY ON THE RESEARCH AND DEVELOPMENT WORK ON TRANSMUTATION

The US ERDA is sponsoring a three-year research programme in order to obtain a cost-risk benefit analysis on the transmutation concept of

actinides other than fuel in fission reactors and a conceptual flow sheet for actinide partitioning from HLW. This programme is being performed at the Oak Ridge National Laboratory in collaboration with other US laboratories. It was started on October 1976.

The actinide removal will be based on a combination of a modified Purex process and secondary processing of the waste stream [5]. The modifications of the Purex process aims at the reduction of the amounts of low- and intermediate level wastes by recycling chemical reagents. Processes which make Pu inextractable at low concentrations are being investigated: reactions with TBP, fission products and other impurities. Am, Cm and transcurium elements will be recovered isolating trivalent actinides and lanthanides and fractionating the product either by cation exchange chromatography or the Talspeak solvent extraction process. The effect of the resin material on the waste stream and the recovery of the salting agent in the TBP extraction process are open problems.

In a parametric study on suitable transmutation devices, items such as the actinide accumulation at continued recycling, the transmutation rate, the reactivity of actinide elements, the spontaneous fission neutron source in actinide fuel elements and the corresponding shielding requirements have been investigated [6].

A contract between the EEC, the UKAEA and the Reactor Centrum Nederland has been concluded, to carry out a study on the following subject: "Assessment of the state of the art on the removal of the actinides from radioactive wastes followed by their destruction by nuclear processes". The areas to be studied are, amongst others, the production of actinides in reactors, their occurrence in chemical plant wastes, their long-term hazards, their incineration and connected hazards, the chemical separation problems, as well as economic assessments.

At Hanford (ARHCO), flow sheets for the removal of actinides from acidic waste streams are under development, considering as extractants the neutral bidentate organophosphorous reagents DBDECMP (dibutyl-N, N-diethyl carbamyl methylene phosphonate) and its dihexyl analogue DHDECMP [7]. The first one can easily be purified but is relatively soluble in dilute acid solutions, whilst the second one is rather insoluble but cannot readily be purified.

Calculations of capture and fission cross sections for higher actinides have been performed by means of the Hauser-Feshbach nucleus model at Hanford (HEDL), in order to generate the improved version ENDF B-5.

At INEL, Idaho Falls, a flow sheet with DBDECMP diluted in xylene is being tested for removal of Np, Pu, Am from first-cycle high level raffinates to produce an actinide-free waste (alpha-activity smaller than 10 nCi/g)[8].

Work on actinides partitioning and actinides cross section has been performed at the Savannah River Laboratory in the framework of campaigns for the generation of ^{252}Cf. Higher actinides are extracted in a second solvent extraction cycle at low-acid state and high salt concentrations from the aqueous waste stream. The separation of lanthanides and actinides is made by rapid ion exchange [9].

A consistent cross section set for higher actinides appearing in the ^{252}Cf generation chain has been elaborated for thermal reactors [10]. According to this set, the accuracy of all predicted actinide concentrations was within 10%.

The Battelle Pacific Northwest Laboratories have set up a research programme which aims to evaluate the incentives for partitioning, to compare waste management schemes with and without partitioning, to develop flow sheets for removing long-lived nuclides from fission products and solidified waste, to study release pathways of greatest risk and retention properties of soils, etc. [11].

At General Atomic, the high-temperature gas-cooled reactor with its high burn-up potential has been investigated as plutonium burner and higher actinide transmutation device. This is considered to be a likely solution to the actinide disposal problem, if continual recycle of actinides is applied [12].

At General Electric, a study on higher actinide transmutation from three light-water reactors and one fast breeder by means of a fast breeder has been studied [13].

Work on chemical separation of higher actinides has also been performed at the GfK Karlsruhe [14] and is under way at CTH Goeteborg, Sweden.

4. RESEARCH ACTIVITY IN THE FIELD OF ACTINIDE TRANSMUTATION AT THE JOINT RESEARCH CENTRE (JRC) OF EURATOM

In 1973, the Council of Ministers of the European Community approved a first four-year research programme on the Management and Disposal of Radioactive Waste. At the end of 1976 a decision has been taken to continue the research programme during the period 1977 through 1980. The programme is subdivided into a number of activities which cover all areas of interest for the elaboration of the transmutation concept. By means of a fault-tree analysis methodology, the real long-term risk represented by geological disposal of HLW in different geologic formations will be evaluated. Research on stability of conditioned HLW and interaction of actinides with the environment will contribute to this evaluation. Assessment studies on physical and technological problems contributed to the quantification of the magnitude of the actinide problem in high-level radioactive wastes (HLW) from different reactor types, to the identification of gaps in nuclear data needed for calculating the actinide evolution, and to the determination of relations between recovery degrees of individual actinides from the HLW and the potential radiotoxic risk. The neutron-physical feasibility of actinide transmutation in thermal, as well as fast reactors has been proved. Further work will illustrate the implications of the presence of higher actinides in the fuel cycle and propose a design for fuel elements containing higher actinides.

About 80 possible flow-sheets for the separation of higher actinides have been identified, and by means of utility analysis techniques it is planned to determine their effectiveness in terms of various parameters, such as waste arising, extraction efficiency, R&D costs, R&D time, operating costs etc. The choice of useful separation processes will thus be restricted to a few flow-sheets, which will be submitted to fully active, laboratory scale verification, possibly during 1979-1980.

As a result of the physical, chemical and technical studies to be carried out at the JRC, the technical feasibility and the net benefits of transmutation as an alternative in the HLW management scheme in terms of risks and costs will be established. If the net benefits justify the application of this concept in practice, development work of the same order as for the commercial reprocessing technique will still be required. Estimates give figures of at least 15 years to integrate the partitioning process into a reprocessing plant.

5. PHYSICAL STUDIES AT THE JRC

The reliability of the calculational results for the build-up and destruction of higher actinides depends essentially on the nuclear cross sections. In order to see how sensitive final results may be to inaccuracies in cross sections, extensive sensitivity studies have been performed. A way for calculating sensitivities has been developed, consisting of analytical formulae for all isotopes of interest, which contain explicitly the main parameters and permit easy assessment of the effect of the secondary conditions [15]. Results for the sensitivity of the long-term hazard of actinide waste to nuclear cross sections show, in the case of a typical light water reactor, the great importance of the capture cross sections of ^{241}Am, ^{237}Np, ^{243}Am, ^{244}Cm, ^{242}Cm, and of the fission cross section of ^{242m}Am. The sequence of importance for a fast reactor is similar: capture cross sections of ^{241}Am, ^{243}Am, ^{237}Np, ^{242m}Am, ^{242}Cm, and of fission cross sections of ^{241}Am, ^{243}Am, ^{242}Cm, ^{237}Np. Conversely, the fission cross sections ^{241}Am, ^{243}Am, ^{237}Np, ^{244}Cm and ^{245}Cm are of the greatest importance for calculating the transmutation rate in a fast reactor. In order to be able to decide which nuclear data has to be measured or theoretically determined, the status of the nuclear data must be reviewed. A good survey on the state of the art was given by the IAEA Advisory Group Meeting on Transactinium Nuclear Data at Karlsruhe in 1975. A systematic collection of these and all other available data permits the probable errors in the nuclear constants to be determined and, combined with the sensitivity results, conclusions concerning the accuracy of the calculational results for build-up and destruction of higher actinides may be drawn.

In collaboration with the GfK Karlsruhe, the differential fission cross section of ^{241}Am is being measured at the Van de Graaff accelerator of Karlsruhe for the energy range from 50 keV to 1.3 MeV. Preliminary results have shown that ENDF/B-4 data are at least for a factor 3 too small for energies of 180 keV to 450 keV and substantially too large below 100 keV.

A contract for the development of a computer programme which will permit the calculation of cross-sections as function of energy by nuclear models has been concluded. Moreover, integral cross-section measurements by irradiation of actinide samples have been performed and are planned by the Transuranium Institute Karlsruhe in its experiments TACO and FACT, respectively. With TACO, pure higher actinide samples were irradiated in the French reactor Rapsodie and the isotopic composition will be analyzed, whereas FACT aims to determine the build-up of higher actinides in specially designed fuel elements.

The concept of the JRC for transmuting higher actinides is based on the assumption that if possible no special burner reactors should be developed but that actinides should be recycled through normal power reactors. It is scheduled to investigate in detail any suitable thermal reactor and a fast breeder. In the case of thermal reactors, a procedure for the appropriate treatment of nuclear cross sections for self-shielding effects has still to be developed. The requirements to the reactor to be chosen as transmutation device are a transmutation rate per irradiation cycle as high as possible and a minimum production of transcurium isotopes which would increase the radiation problems in the fuel cycle. Moreover, it would be desirable that actinide recycling contributes only to the radiation already existing in the fresh fuel. This requirement is partly fulfilled in the Pu fuel cycle, where ^{238}Pu and ^{241}Am are constituents of the normal fuel and in the uranium-thorium fuel cycle, where the isotope ^{232}U occurs. Regarding the fuel cycle the following questions have to be answered:

- Have higher actinides to be recycled necessarily in special fuel elements or may they be homogeneously mixed with normal fuel? In this context the mass and the composition of higher actinides, their heat release, their spontaneous fission neutron source, their α-radiation and the effect of these parameters on the fuel cycle costs has to be evaluated. Preliminary results indicate that the mass of higher actinides during continuous recycling remains smaller than 1% of the mass of fuel.

- If a heterogeneous recycle scheme for actinide fuel elements were applied, should they then be separately reprocessed or be blended with normal fuel before reprocessing? Important quantities to be assessed in this context are the attainable extraction efficiencies for individual higher actinides from the HLW and the acceptable losses which must be determined by a long-term risk analysis.

- If actinide fuel elements are reprocessed in a proper plant, should its Pu content then be extracted? Preliminary calculations for a fast breeder indicate that at equilibrium the Pu content in actinide fuel elements amounts to about 20%. It strongly increases the thermal power rating and the reactivity of actinide fuel elements.

6. CHEMICAL STUDIES AT THE JRC

Industrial experience on the recovery of actinides from irradiated fuels is limited to uranium and plutonium by the Purex process. Semi-industrial processes were, however, developed for the separation of higher actinides. The techniques utilized and the results obtained form a useful basis for developing flow-sheets for actinide removal from HLW, but they cannot be applied for the purpose of an alternative waste management concept without major improvements. In fact, all of them aim at recovering valuable higher actinides in a relatively pure state, while the completeness of the recovery is not important. Extraction degrees of 80 or 90% are frequently considered as an optimum. In addition, the generation of secondary alpha-contaminated wastes is also unimportant.

On the other hand, for waste decontamination purposes the requirements to the flow-sheets are completely different. The primary objectives are recovery degrees as high as possible and minimum secondary waste arisings. Therefore, the chemical schemes have to be reconsidered in order to meet these new objectives. Theoretical studies as well as experiments with simulated waste streams described in literature have been evaluated [16].

A critical feature in the development of separation flow-sheets is the specification of the "target decontamination yield" of the separation process. Extraction efficiencies for higher actinides which would lead to long-term risk levels of HLW corresponding to naturally occurring high-grade uranium ore represent a desirable goal. However, the specification of realistic values for the recovery degrees requires a comprehensive analysis of the overall waste management policy, particularly of the conditioning techniques and the disposal options for all types of radioactive waste generated at the individual steps of the fuel cycle. Such an analysis is at present under way in the JRC. For orientative purposes an analysis of the effect of increasing separation yields on the complexity of the chemical flow-sheet has been carried out. It led to the choice of decontamination factors of 10^3 for Pu, Am and Cm, and of 10 for Np as targets for the experimental studies [17].

In the field of chemical separation of nuclear materials, solvent extraction is known as the most flexible basic technology applied in large-scale plants. Hence, this method can be rightly considered as a promising candidate for waste partitioning.

Usually, trivalent actinides are being recovered from low-acidity feed solutions containing variable amounts of salting and/or complexing agents. Tributylphosphate (TBP) and di-(2-ethylhexyl)-phosphoric acid (HDEHP) are the extractants which have found large application. In contrast to this, organic tertiary amines are considered as unattractive extractants for removing Am and Cm from HLW raffinates, owing to the highly corrosive feed (concentrated LiCl solutions) required by such a type of process. The extractant dibutylbutyl phosphonate (DBBP) is used for Am recovery from liquor wastes previously adjusted to low-acidity. Due to their high extractive properties under very acid conditions, bidentate compounds could be usefully employed for extracting trivalent as well as tetravalent and hexavalent actinides from concentrated nitric acid solutions without any addition of salting agents.

As a first approach, the modification of solvent extraction processes currently employed for recovering actinides and their adaption to waste partitioning needs was considered. According to this approach, some extractants which seem to be potential candidates for waste partitioning have been identified [18]. They may be used for low-acidity feed solutions as well as for acid feeds. Process steps of some tentative HLW partitioning flow-sheets based on HDEHP and TBP extractants have been experimented to verify their feasibility and the possibility of integrating them in the current HLW management scheme. The flow-sheet for HDEHP is reproduced in fig. 2. The properties and the behaviour of HDEHP and TBP under operation conditions at the different process steps of the flow-sheets

have been tested. As a possible alternative to HDEHP, a hydroxamic acid has also been experimentally investigated [19]. The experiments were carried out using a synthetic, suitably traced HLW solution.

The use of real HLW solutions is scheduled to confirm the results obtained. The problems connected with the presence of non-extractable actinide forms (α-bearing solids, polymers and complexes) have been pointed out and further experimental investigations are being prepared.

Recovery of actinides from waste streams by precipitation is of interest, since a large amount of basic data is available in literature and it might resolve the problem of unextractable forms of Pu in the waste stream. Experiments with the OXAL process have been performed with simulated waste solutions as well as real waste solutions from fuels irradiated to 26,000 MWd/t and 3,500 MWd/t, respectively [16]. Depending on the pH-value of the solution, the losses of residual Pu to the waste stream were in the range between 1 and 2%, whereas the Am/Cm losses amounted to 0.1% up to 1%. It can be noted that there is a good agreement between recovery degrees obtained by simulated and real waste solutions. The precipitate containing fission products such as Zr, Mo, Ru, which is formed during the denitration and concentration steps, carries away a part of the actinides. Whilst Am and Cm can easily be recovered by washing with nitric acid, for Pu more drastic methods such as dissolution of the precipitate in HCl or HF and successive separation from the fission products must be applied. At laboratory-scale, the fresh oxalate precipitate can be easily processed by HNO_3, 10M, for preparing the feed for the successive separation of actinides from rare earths. The quantity of additional low- and intermediate-level waste generated by the OXAL process has proved to be very small.

7. CONCLUSIONS

Despite world-wide activities in the field of higher actinide transmutation, it is still too early for a realistic judgement of the advantages and consequences of this high-level radioactive waste disposal concept in terms of risk. Undoubtedly, the potential long-term risk will be reduced. But this benefit will be partly compensated by increasing the short-term risk inherent in handling the actinides in the fuel cycle. When all technical implications of the actinide recycle will have been explored in greater detail, assessments on the costs involved in this concept and a risk-benefit analysis will be possible. Probably, these results will be available after a period of three or four years.

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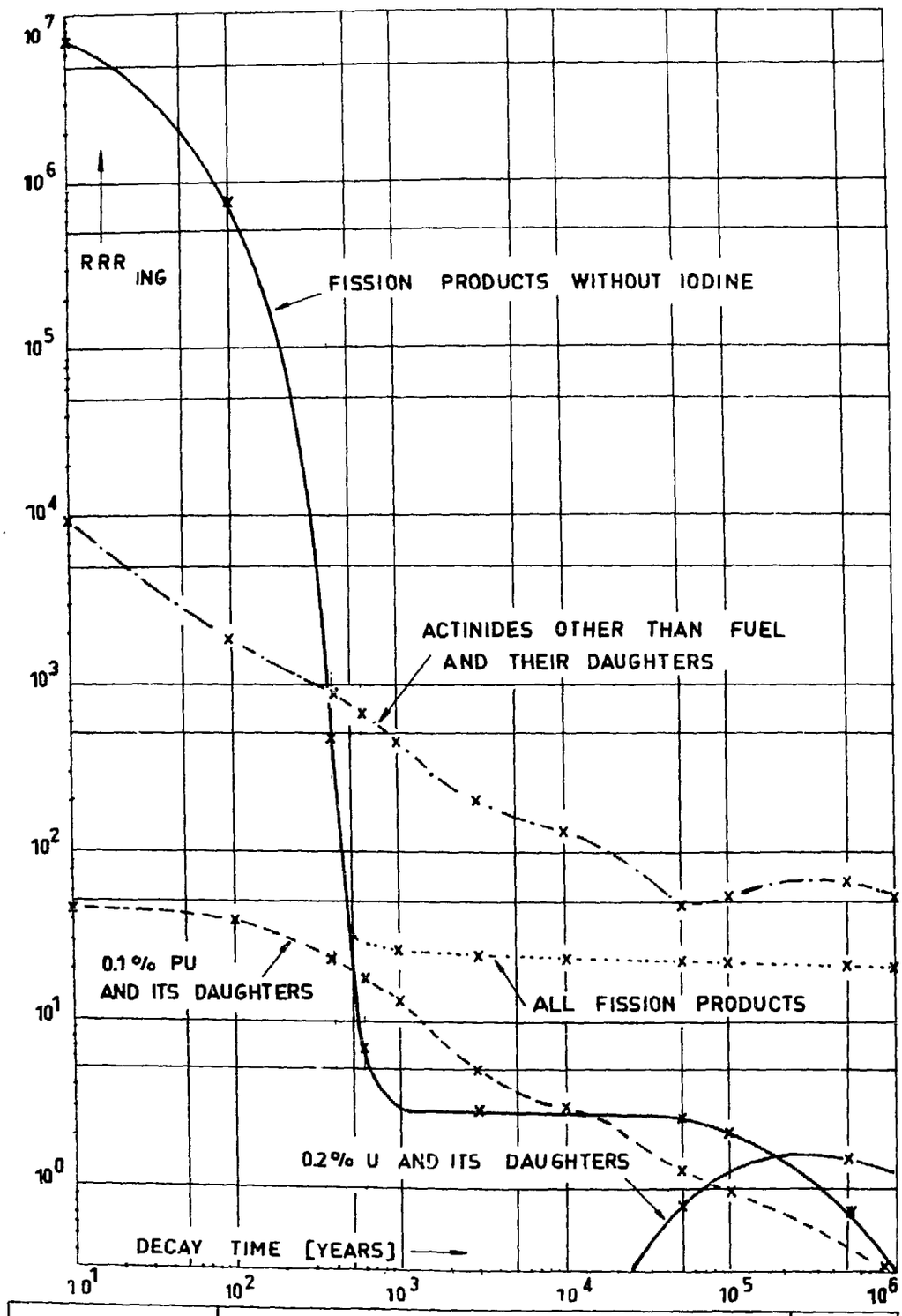
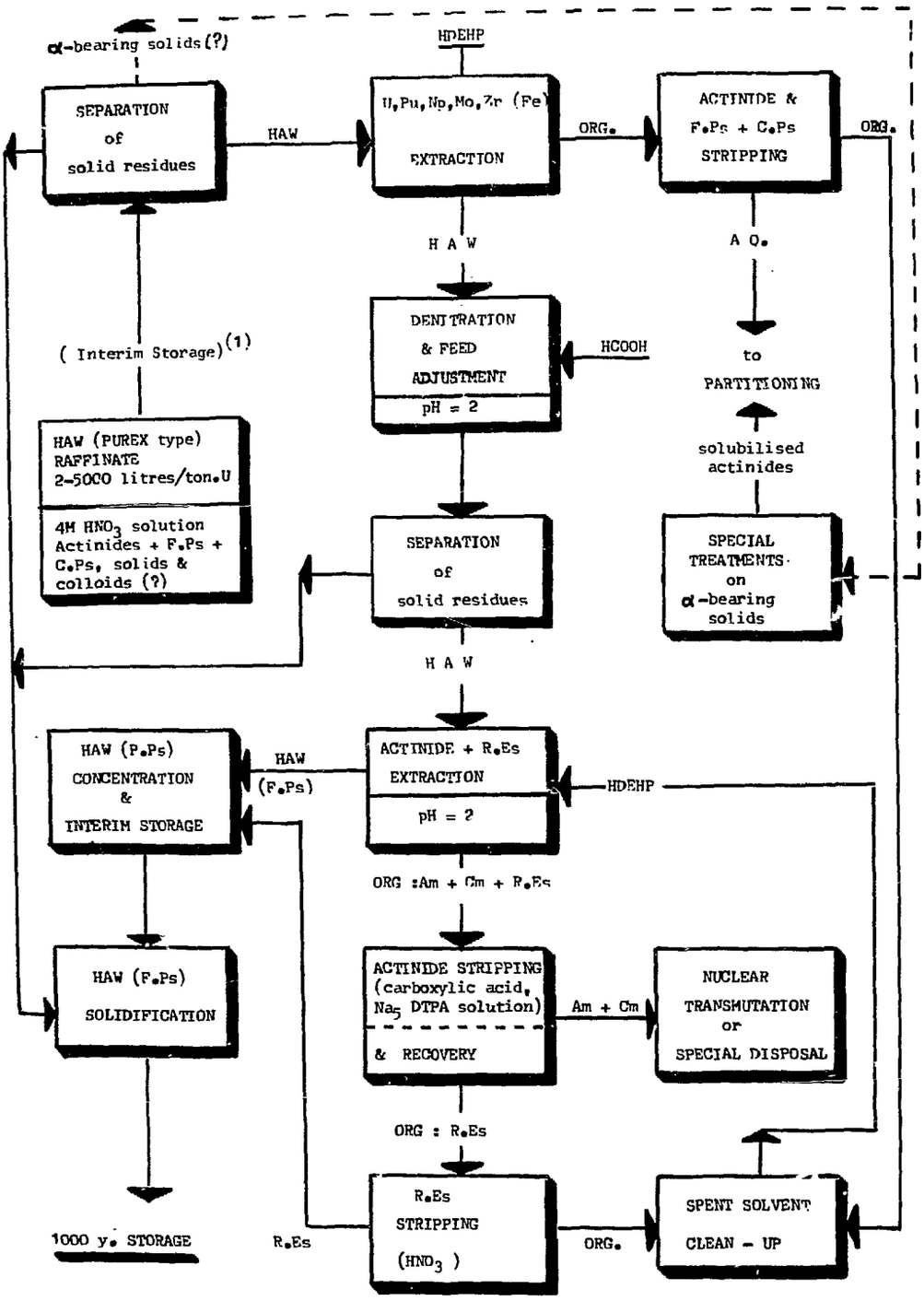


CCR-EURATOM ISPRA	RELATIVE RADIOTOXIC RISK (RRR) BY INGESTION FOR HLW FROM AN U-FED LWR (33000 MWD/T)	Fig. 1



(1) to cool the HAW and to allow, if needed, the growing of 238Pu daughter

CCR-EURATOM ISPRA	TENTATIVE FLOW-SHEET FOR HLW PARTITIONING BASED ON ACTINIDE EXTRACTION BY HDEHP	fig. 2