MINIMIZING THE INTEGRATED COLLECTIVE RADIATION DOSE AND THE TRANSMUTATION OF LONG-LIVED NUCLEAR WASTE

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ABSTRACT

For more than a year, ECN is engaged in a research programme on transmutation of actinides as well as of fission products by means of respectively fission and neutron capture. It is intended to contribute to international efforts, which evaluate recycling options of the nuclear waste problem. Besides studies on strategy and scenarios, efforts are made to demonstrate possibilities for transmutation of long-lived fission products as Tc-99 and 1-129. Global scenario studies on light water power reactors show that in principle such reactors could consume their own technetium production by means of neutron capture only in case that some surplus of technetium is being situated in the core. If the Tc would be homogeneously mixed through the LWR fuel, the inventory of Tc (a few hundred kg) would be too high to be practical in present scenarios for reprocessing this fuel. For inhomogeneous combinations of waste and fuel, the inventory would have to be even higher, and in general it can be stated that problems with reactor based incineration are complicated whereas only marginal advantages are to be expected. Therefore incineration schedules have been studied, which use an inventory as low as possible, and which would involve a high flux thermal neutron field for example at accelerator based reactor systems. Such hybrid systems are being considered as they may reduce the risk possibly without too high energy cost. Finally an experimental programme is presented, in which the Petten High Flux Reactor may be used to support comparative studies on targets for incineration.

1. Motivation for a study of transmutation of nuclear waste

There are various arguments in favour of nuclear transmutation of waste, each of which deserve examination. Here the discussion will be aimed towards the reduction of human exposure to leakage of waste repositories by removing long-lived components from the waste. This study is motivated by the wish to minimize the time integrated collective radiation dose due to nuclear waste. There is a growing tendency to extend the time interval of integration to infinity and consequently the interest in incineration of very long-lived waste (half lives from 10.000 years up to millions of

years) is increasing. This is especially so because it is rather difficult to guarantee integrity of repositories for such a long time, and therefore scenarios had to be considered, which involve leaching out by water, possibly after human intrusion of disposal sites. In these scenarios the risk is enhanced, of those long-lived components in waste, which have a large product of radio-toxicity and geochemical mobility. Especially the long-lived fission products Tc-99, I-129, and Cs-135 and the actinides Np and Am, may dominate the long-term risk of a waste repository [1-5] . Long-term risks are illustrated in table 1, which has been extracted from a UK study [4] on deep reposition in granite. This table presents, for a normal evolution scenario (without human intrusion) the collective dose in man Sv, due to 18.000 t of LWR spent fuel. This amount of fuel would arise from a hypothetical 20 GW(e) park of light water reactors, which operates for 30 years. Relevant nuclides have not only been presented for waste which has been vitrified, but also for directly stored fuel. In table 1 the dose is integrated over the first million years and over 108 years.

TABLE 1: INTEGRATED COLLECTIVE DOSE DUE TO 18 kt OF SPENT LWR FUEL, IN A REPOSITORY (according to ref. [4])

Period :	One million years		Hundred million years	
Storage:	Vitrified	Direct storage	Vitrified	Direct storage
Nuclides:				
Tc-99	100 %	98 %	17 %	46 %
I -129		2 %		1 %
Cs-135			10 %	24 %
U -235				6%
U -238				14 %
Np-237			71 %	5 %
Pu-239				4 %
Am-243			3 %	
man Sv	40	5 300 000	340 000	11 000 000

It has to be noted that the results of this UK study [4] rely heavily on assumptions on the effectiveness of vitrification and on geological characteristics of the repository. They may not be quite representative for other repositories, as calculations are based upon site dependent gee-chemical characteristics. If the waste is not in contact with oxygen, the mobility of Tc will be strongly reduced. In clay repositories [3] or in rock salt [5] - repositories the 1-129 risk could dominate, especially if the spent fuel is stored directly. Although there are differences among the deep geological repositories, it nevertheless can be

concluded that fission products will dominate the collective risk in case of direct storage of spent fuel for the first million years. As for example follows from [4], the removal of Tc and I from the waste, would reduce the collective one million year dose to a value less than only a few hundred man Sv in stead of 5.3 million man Sv.

Table 1, however, would hardly give arguments for transmutation of waste if one merely wished to reduce the one million year dose. Reduction of long-term risk by vitrification is very important. Further reduction is possible by transmutation of the fission products and only if one wishes to reduce the collective dose as integrated over the extremely long period of hundred million years, one should transmute Np and Am. Then the total dose will be reduced to less than thousand man Sv.

One should realise that table 1 also shows that the maximally expected hundred million year dose for directly stored fuel, is only five times higher than the dose, which would result from storing fresh fuel, which never has been in a reactor (this is the u-235 and u-238 contribution!) . This observation indicates that dose benefits from incineration of waste are relatively small with respect to the dose, which would result anyway for example from the mine tailings. One should therefore realise that transmutation of waste should only be considered if cost and risk is moderate, at least if one's only incentive would be reduction of the dose risk on the extremely long term.

2. Special motivation for study of fission Product transmutation

According to refs. [1] and [10], one may reduce the very-long-term ingestion hazard of actinides by neutron-induced fission of these actinides in an extremely high thermal flux. Then, however, fission product waste will be formed during this incineration process. If the neutrons, which are needed for transmutation, have been generated (partly) by the fission process, one even forms extra new fission product waste and new actinide waste. In view of environmental economy, it is clear that products of fission, spallation, or capture, which would be formed in the transmutation device, should be less bothersome than the original waste, which was produced by means of neutron capture or by fission in a reactor. Useful energy,. eventually produced in the transmutation device, should also be considered in this balance.

As was shown in the previous chapter, fission products dominate the very long-term risks of waste repositories. Although actinide incineration is not helpful in reducing the very long-term risk, it is favoured by quite respectable arguments. Such arguments may be first of all based upon the wish to reduce the source term of

radioactive waste and, correspondingly, to reduce the number of repositories. Secondly one may wish to destroy and peacefully use weapons material or limit the need for safeguarding in case of retrievable storage of actinide waste.

Scenario studies e.g. performed at ECN Petten [11] show how one may reduce the actinide activity by repeatedly recycling in power reactors. Hypothetically after a few cycles an extreme situation might develop in which the actinide radio-toxicity stabilizes towards an equilibrium value. Whereas light water reactors would reach a relatively high equilibrium alpha-activity (PWR-EQ in fig.1) after many recycling steps, fast reactors would reach their equilibrium (FBR-EQ) already rather soon. These studies indicate [11] that there are certainly possibilities to reduce the overall source term of radioactive waste by means of reactor based incineration systems. Besides risk reductions this scenario also will impose new risks for the present and future generation. First of all incineration of trans-uranium elements will increase the very long-term risk from fission products in the same way as the use of U-235 as a fuel would have done, but a net reduction will be reached in collective radiation dose because less mining would be needed per unit of produced energy. This reduction in collective radiation dose will however be offset, at least partially, by the build up of U-234 during the irradiation of the actinides, which can not be easily separated from the otherwise harmless depleted uranium and therefore will act for a long term as a source of Ra-226 in the tailings. As it is not yet clear how much the integrated radiation dose for the population decreases by actinide incineration in normal power reactors, further study is necessary. It should be investigated wether transmutation of fission products could be helpful in reducing the long-term radiation dose. It seems obvious that this would be so if one could transmute Tc-99 and I-129 by neutron capture to harmless Ru-100 and Xe-130.

3. Cost estimates for simple scenarios for Tc and I transmutation

Before one would decide on transmuting the long-lived fission products Tc-99 and 1-129, it should first be shown that the integrated dose indeed decreases by the transmutation and that neither energy cost nor short-range risk will exceed practical limits . Because costs and risks related to reprocessing will decrease with the radioactive inventory, scenarios with a relatively low inventory are to be preferred. Such scenarios use a high flux of neutrons from high flux reactors, - boosted spallation sources, accelerator-driven reactors, blankets of fast reactors or any other high flux devices, based on fission, fusion, spallation or a combination of these processes.

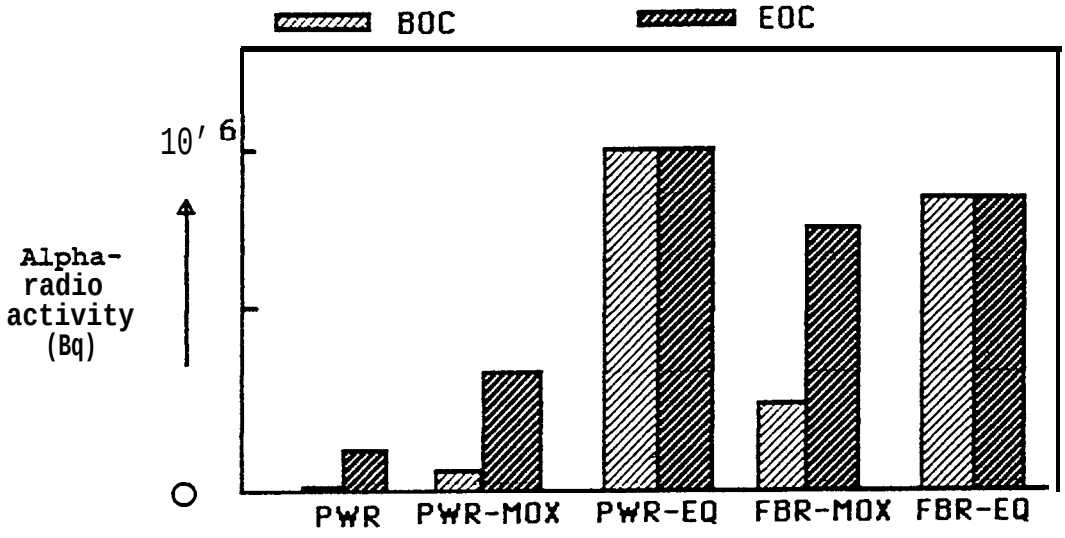

Figure 1: Alpha radio activity in PWR and FBR reactors [11]. Here BOC stands for "beginning of cycle" and EOC for "end of cycle", EQ stands for the equilibrium situation after recycling many times.

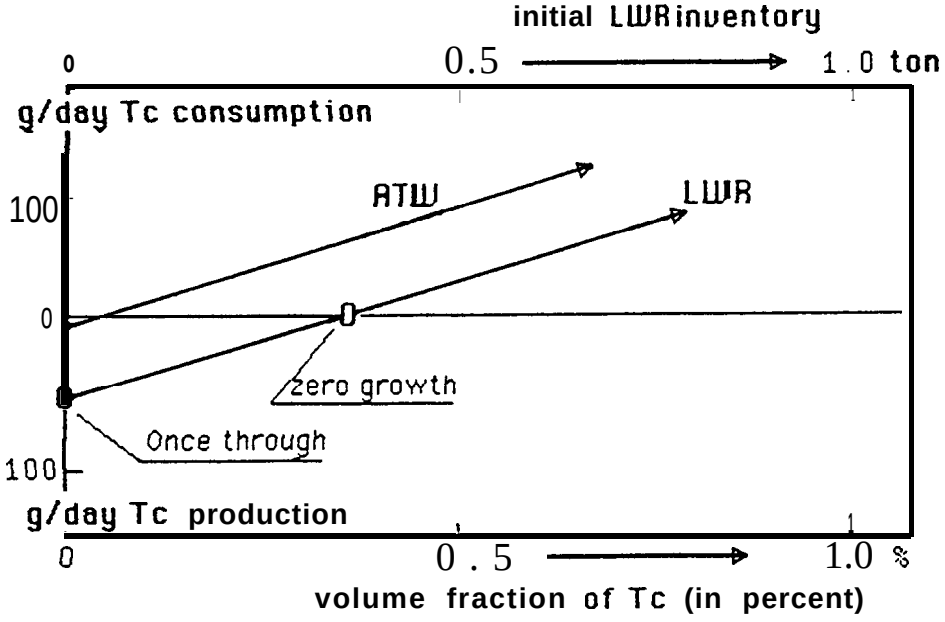

Figure 2: Transmutation of Tc in LWR and in ATW in function of its concentration. For the LWR the weight inventory is about 1000 kg for each percent of the volume fraction. For the corresponding ATW the active volume is about ten times smaller, which leads to an inventory of about 100 kg for one percent of the volume fraction.

In contrast to the burn-up of actinides little energy is released during the transmutation of fission-products, and it is clear that the energy cost of fission product transmutation should be small with respect to the energy generated during creation of this waste. Further one should also consider the extra costs related to irradiation, recycling and even mining due to the extra fuel, which is needed. Obviously besides a scenario-study, one also needs some practical demonstrations in pilot plants in order to test the presumed (low) level of these costs.

Simple incineration scenarios have been studied at ECN [9, 11] on light water reactors, high flux reactors and spallation sources. First of all a standard LWR with 1000 MW electrical power is considered, in which the technetium is homogeneously mixed through the fuel in a concentration between 0 and 1% . Consider one fuel batch (1/3 of the core) staying in the reactor for three years. In this period an amount of 30 kg technetium is produced in an LWR by fission in the batch. In that case a “zero growth” scenario, i.e. burnup of technetium equivalent to its production by the fission, would be realized when the initial Tc-enrichment would be about 0.3%, and the technetium inventory in the LWR core would be 300 kg (see figure 2) . For the LWR the energy-economy is obviously good, as introduction of a fraction of technetium in the fuel of about one percent will only lead to a negative reactivity effect of the order of a few percent. Because the standard 1000 MWe LWR produces somewhat less than 60 g/day of technetium, it would be easy to reach the zero-growth option at an expense of only a few percent in the fuel price.

Although the above given LWR transmutation scenario seems quite feasible, the high Tc concentration in the fuel could be less attractive from the point of view of the cost and the risk of reprocessing and recycling. Nearly 25 % of the technetium in spent reactor fuel is present in the fines, in the form of hardly soluble intermetallic compounds of the elements Mo-Tc-Ru-Pd, whereas most of the rest will end up in the high level waste after reprocessing. Selective extraction of this technetium from the high level waste streams seems possible, but this requires intensive study as it will present a major cost factor. This is especially so because in the LWR scenarios, the technetium would have to be reprocessed many times, and the LWR option for Tc transmutation seems not realistic because of the high inventories and the extra reprocessing costs. For iodine, the latter argument is much less valid, as the availability problem does not exist, because iodine is now already isolated on a large scale in standard reprocessing facilities for LWR fuel. In some special heterogeneous irradiation scenarios, partitioning costs might be reduced. It seems however to be likely that in any heterogeneous scenario much higher inventories are needed because effective

cross sections are lowered by self shielding. As the inventory related costs (size of apparatus, source term risks etc.) will dominate the partitioning costs, the balance may shift in favour of high flux devices with a small inventory.

As a second scenario a hypothetical High Flux Reactor has been considered, with about a factor ten higher thermal flux, but about five times lower power than the standard LWR. Apart from discussions on the feasibility of such a reactor, one might discuss the features of a hypothetical scenario in which such a reactor is used for the burnup of waste. There are advantages in this scenario: first of all the much lower power of this HFR has as consequence that the technetium production by the fission process itself could be less than the LWR value indicated as "once through" in fig.2. Further the use of special fuel or the use of special Tc targets could reduce reprocessing costs. In this hypothetical HFR only a limited amount of excess neutrons per fission could be available for transmutation, and from the yield of long-lived fission products per fission in the LWR (about *0.1)* and from the excess neutrons per fission in the HFR (less than 0.5 [8]), it follows that this HFR, if it were to transmute all the long-lived fission products produced by a single standard LWR, should have at least 20 % of the thermal power of this standard LWR. This limit would be proportionately lower if one would limit one self to incineration of 1-129 for which the yield is less than 1 %, but in that case the inventory of 1-129 should be rather high because of the smaller value of the cross section.

As a third scenario, incineration was considered to take place in a high flux of thermal neutrons from a boosted spallation source. In ref [1] it is claimed that transmutation is quite feasible in an intense thermal neutron field *of* a spallation neutron source, in which high energy protons (1 GeV) , hit a target system. This so called Accelerator Transmutation of Waste system (ATW) would not only incinerate the fission products from 10 LWR's, but also their minor actinide waste. This can be understood by realising that in this system fission of actinides produces excess neutrons at an energy dissipation value of only about 100 MeV per neutron. This is much better than in a fission reactor, wherein only say 0.25 excess neutrons could become available per fission at an energy dissipation of 100 MeV. For. hybrid reactor-accelerator systems many neutrons are freed by fission, and the number of excess neutrons for a given energy dissipation may be expected to lie in between the two extreme values. In the above treatment of the ATW the released energy at the target is not including the energy consumption of the accelerator itself, and therefore a quite different reasoning is applied in order to derive the cost of the transmutation process.

For accelerator-based transmutation, . the energy costs will be directly related to the electricity bill. Therefore the overall efficiency of the system (the number of MWe needed to produce one MWe of beam power) is highly relevant. If advocates of the ATW scheme are correct in stating that the net energy balance for the ATW can be made positive by simultaneously breeding Pu-239 fuel [1], the case of the ATW is strong. Independent upper estimates of the costs can be based on earlier studies [7] of accelerator breeding. From these studies it has been concluded that the neutron costs and handling costs "are not supposed to exceed practical limits". As it seems likely that the cost per atom is relatively low for 1-129 as well as for Tc-99, the relative transmutation cost of long-lived fission products would be less than their yield in the fission process, and thus less than 10% of the cost of the original accelerator bred Pu-239 fuel.

One should realize that in accelerator-booster systems about 100 MeV per excess neutron [1] is dissipated. Although this value is at least twice to four times as low as for nuclear reactors a quite large amount of heat will be liberated. This power, which originates from fission power to a substantial fraction, should be usefully applied, for example to generate the electricity needed to run the accelerator. In this case the cost of transmuting Tc-99 will only be a fraction of the cost of the fuel from which the Tc originated, just as for earlier treated cases of transmutation in an LWR. When this ideal is reached remaining factors in the selection procedure for specific scenarios will be related to cost of partitioning rather than of transmutation.

A more modest scenario based on the same principle would be the smaller version of the ATW, which has been described as the "Hanford system" in ref [1] . For this system the product of the high neutron flux with the number of available Tc nuclei and the effective capture cross section would be about the same as for the standard LWR. This Hanford system would transmute just as well as the standard LWR, but with much lower energy dissipation. In such a system one may also use a lower radioactive inventory because for the same effective cross section, the Tc inventory needed will be inversely proportional with the flux.

4. Experimental programme in the HFR at Petten

In most scenarios one could limit the integrated collective dose by performing reprocessing and irradiation of Tc and I. Therefore some demonstrations are needed, and it would be desirable to have access to facilities for irradiation and for post-irradiation experiments. The HFR in Petten has been in use as a material testing reactor for a long period, and on-site experience is available in irradiations as well as in the post irradiation

experiments. As has been more fully described in ref. [9], some demonstrations should support scenario studies of transmutation. For the HFR the total flux, integrated over the full spectrum turns out to be about 10^{15} n/cm^2 s, and substantial increase of this value is not feasible within the frame of the present programme. In small samples a thermal flux of $4*10^{14}$ n/cm^2 s can be achieved and for larger samples the flux may be about a factor four lower (for comparable samples this is still about ten times higher than in an LWR) .

No major technological problems are envisaged for technetium transmutation [9] but anyway the technical feasibility should be demonstrated by experiments on targets of a relevant scale. Most likely, the samples to be irradiated in a pilot plant will be technetium metal or oxide. However, some aspects have to be kept in mind. For homogeneous options, one- should also give attention to the behaviour of a fuel pin with substantial fractions of technetium, whereas the behaviour of canning and target materials at high exposure (i.e. 10^{21} n/cm^2) can be a concern for the heterogeneous option.

Incineration- induced risks are present during and shortly after irradiation of 1-129. During the irradiation time one has to deal with the iodine, compounds of which are corrosive, volatile and unstable at high temperature. Further, the transmutation product gaseous xenon could cause swelling and problems with target rod pressurization. For these reasons the transmutation of iodine has been considered to be only marginally feasible for quite some time [2]. Some new ideas, involving vented pins and use of either ceramic iodides [6], or eutectic mixtures of iodides with metals such as lead or sodium as a target [9] may show a way out.

Few iodine compounds are chemically stable especially under the irradiation conditions, therefore compatibility with cladding materials is mandatory. Iodides, with suitable properties have been selected but in all cases the effect of gamma-radiation on the stability of the samples needs further study. As most promising liquid the metal-iodide eutectic Pb-I_x system was selected. It seems desirable to first investigate the phase diagrams and the miscibility. Also the systems Sri-I or Zn-I may be suited in case additions of a second iodide (NaI) will lower the melting points at the iodine rich side of the metal-iodide phase diagram. Major advantages over the use of Sn as a metallic component are, in case of Pb, the lower volatility and in case of Zn the higher thermal stability. Especially lead di-iodide is attractive as this is the compound used in reprocessing plants for storing the radioactive fission product I-129 . In case that a molten state of the matrix would not be essential, magnesium di-iodide or cerium tri-iodide seem to be good choices in view of

stability, low vapour pressure and high iodine weight content. Experience on vented ceramic and liquid targets is available from earlier projects, which aimed at research on gaseous emission during neutron. irradiation.

In 1991 a programme received funding for the following items :

- detailed characterization of the irradiation facilities,
- selection of sample materials to be irradiated,
- selection of geometry of the samples and canning,
- neutron-physics calculations for optimization of the various options for a facility with the highest possible thermal flux for irradiation of small samples over long periods,
- Considering test-irradiations with stable 1-127 compounds in order to study their stability under gamma radiation,
- Supporting nuclear data compilation and evaluation.

Regarding the nuclear data programme it can be mentioned that this work mainly relates to nuclear data on actinides and long-lived fission products. Existing codes and libraries (based upon JEF-2) are being improved. In order to support work presented in this chapter, the following nuclear data are required: capture and possibly also (n,2n) cross sections of Tc-98, Tc-99, Ru-100, 1-128, I-129, Xe-130, and also of elements in carrier materials and cladding. These data are being extracted from the available libraries. For reactor physics and burnup calculations, the codes ORIGEN and FISPACT will be adapted together with their working libraries of cross sections, risk data and decay constants. Furthermore, a contribution to accelerator based transmutation schemes will be made by evaluating intermediary energy nuclear physics data, using experience in pre-equilibrium and multi-step direct reaction models.

5.Conclusions

Neutrons can be used for incinerating actinides by fission, and transmuting long-lived fission products by capture. If one would transmute actinides by neutron-induced fission, in a reactor, one produces additional energy as well as those fission products, which dominate the very long-term risk of waste in a repository. If one uses high power accelerator-based neutron boosters, one would produce much less new waste than in the standard LWR. If the dissipated energy is usefully applied to run the accelerator and if reprocessing costs are small enough, transmutation of long-lived fission products would cost only a fraction of the price of nuclear fuel from future breeders. In order t-o reduce reprocessing costs, one aims for low inventory systems. Energy costs to run the accelerator are reduced for the highly boosted hybrid accelerator-reactor system (ATW), with a high neutron density. For the moment the most promising system, which is based

on accelerator technology, seems to be this ATW system described in ref [1] . In developing such a system, some questions regarding targets and irradiation conditions may arise, which may be answered by demonstrations in the HFR Petten.

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