MOTIVATION FOR TRANSMUTING LONG-LIVED RADIOACTIVE PRODUCTS

K. Abrahams

Netherlands Energy Research Foundation (ECN), Petten, The Netherlands
P.O. Box 1, NL-1755 ZG Petten, The Netherlands
Tel: ++31 2246 4340, Fax ++31 2246 3490
E-mail: abrahams@ecn.nl

Abstract
In the Netherlands the efforts on waste transmutation are coordinated in a research programme called RAS. One of the aims of this RAS program is to inform the public and advise the authorities on methods for transmutation/conditioning of nuclear waste, and on techniques which are being developed. Any new way to treat waste should of course not lead to significant risks for the present population. Small risks might be accepted, but these should sufficiently be compensated for. Benefits for the present generation are related to the better exploitation of the full energy content of the actinides, which will reduce fuel costs and waste streams from mining as well as from spent fuel. Future generations might profit from the fact that the waste has been cleaned from actinides and that proliferation risks are eliminated. Another benefit could be that transmutation also could lead to a reduction of dose-risks by leakage of mobile elements such as Rn-222 and the metalloid fission products like technetium and iodine. It is shown in this paper that the balance of benefits and risks is quite different for long-lived fission products than for actinides.

1. GENERAL CRITERIA TO JUDGE TRANSMUTATION SCENARIOS

Spent fuel from light water reactors contains about 100 kg of long-lived fission products, about 300 kg of toxic actinides and 900 kg of short-lived fission products for each GWe year generated. For the actinides the mass balance depends strongly on the fuel cycle history, for the fission products this is less so. About one third of the long-lived fraction of fission products consists of geochemically mobile fission products Tc-99 and I-129. Partitioning and transmutation (P & T) of fission products will have different benefits and risks than P&T of actinides, as may be judged by means of the following three criteria:

1) *Exploitation of the full energy content of the actinides will reduce fuel costs and streams of waste from mining as well as from spent fuel.* A higher long-term toxicity in the fuel and a change in reactor parameters (delayed neutrons, temperature- and void- dependent effects) could be the price one has to pay for this benefit. If the corresponding risks would be low enough, one hopes to reach acceptable waste-storage strategies, in which the life-time of long-lived radioactive components is shortened and in which actinides would be harmless in any waste-disposal scenario even if this scenario would be falsified by human intrusion.

2) Some of the untreated actinide waste transforms itself into a manageable form of partly fissile matter by decay of short-lived products, and disposal sites might become attractive actinide mines. *Future proliferation risks could be eliminated by transmutation.* Actual proliferation risks

during the P & T process itself could be the price for this benefit, and P & T processes could only generate broad public acceptance if risks and costs are controllable.

3) *Dose-risks by leakage of mobile elements like Rn-222 and the metalloid fission products technetium and iodine could be reduced.* The price which should be paid relates to the avoidance of ecological risks of the P & T process itself [1], as one should not spread radiotoxicity by procedures such as machining or spilling of solvents. It seems a sensible strategy to distribute the efforts in such a way that the total dose-risk is minimised for a given amount of spent fuel.

Above mentioned criteria should be related to a cost-risk analysis. For oil and coal the loss of human lives to the present generation lies between 1 and 10 per year for a production of one GW(e). Casualties are lower for nuclear energy [2], and the long term risks ought to be also lower than long-term risks from the use of fossil fuel. As each scenario will have a price, it could be an approach to see this price expressed in an amount of dollars needed to save a life. To protect contemporary individual radiological workers from harmful overdoses, one might offer a price of for example about 100 000 US $ for each man Sv avoided [3]. If however risks for any individual are low enough, collective risks will only be handled according to a strategy called ALARA (As Low As Reasonably Achievable), after all it is considered to be "more important to avoid one man to be hit by a bag of barley than to avoid every one to be hit by some grains".

2. TRANSMUTATION of ACTINIDES in REACTORS and SUB-CRITICAL SYSTEMS

In thermal incinerators some actinides could be used as fuel. Commercial thermal reactors are however energy and plutonium producing entities, which are not dedicated to waste transmutation. These LWRs may of course transmute Pu-239, and application of plutonium in MOX fuel for electricity generating LWRs will clearly diminish its growth (see fig.1). However the amount of transuranium elements will increase anyway due to continuous capture of neutrons in U-238. It is difficult to leave out this U-238 because its capture process is essential to the economy and safety of LWRs: it increases the reactivity swing of the fuel by breeding fissile plutonium, and the Doppler broadening of capture resonances will keep temperature coefficients negative even for large systems. As long as U-238 is the main component in the fuel, the accumulated plutonium mass will increase in any recycling scenario. Due to the build-up of even plutonium isotopes the multi-recycled fuel will become much less fissile in thermal spectra, whereas it remains fissile in fast spectra. If no external neutron source would be applied, an LWR would require an increasingly higher fissile enrichment, and this would create a less economical situation. High concentrations of even-N transuranium isotopes in LWR fuel would also give safety problems due to the fact that the number of formed fission neutrons in such multi-recycled fuel increases with a hardening of the neutron spectrum (see table 1). This problem should be solved, either by integrating the fuel with the moderator or by applying an external neutron source to a sub-critical system, otherwise the reactor could become a prompt critical fast reactor after an accidental loss of its moderator! Thermal systems will have as a further disadvantage that the toxicity in the U/Pu cycle increases continuously due to the growing in of heavier actinides. One would end up with a toxic mixture, in which the long term radio-toxicity is almost entirely determined by americium (thousands of years) and neptunium (millions of years). It is shown by J.L. Kloosterman and W.J.M. de Kruijf in a paper for this meeting that an extremely high neutron fluence (of the order of 10^{22} cm^{-2}) is needed to reduce the long term toxicity in an americium sample.

Fast incinerators have been proposed for the transmutation of actinides. Possibly the breeding mantles of fast incinerators could be replaced by moderated sub-assemblies, which could then also be used for the transmutation of fission products. From several studies it has been concluded that critical fast reactors could only have a minor-actinide fraction in the fuel up to 5%. If the transuranium concentrations in the fuel would be higher, some reactivity problems could appear, and the number of delayed neutrons per fission could become too small, especially for curium isotopes [5]. Reactivity problems could either be enhanced by positive voiding-feedback or by insufficient Doppler-damping of temperature excursions. If critical reactors are not considered safe enough one could consider systems with $k_{eff} < 0.95$. External neutrons could then be supplied by another reactor (possibly a fusion reactor), or by a spallation source with a GeV proton beam. Such external

sources could even provide enough excess neutrons for transmutation and generation of energy at the same time, and advanced reactor systems could be developed to reduce not only the amount of plutonium, but also to reduce the very long-term toxicity due to americium and neptunium [6].

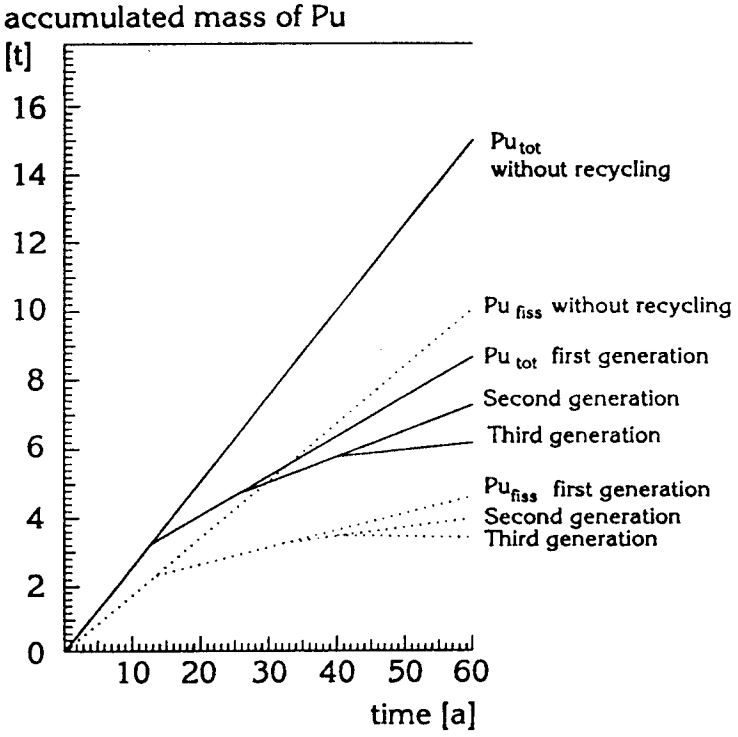

Fig.1: Evolution of the total mass Pu_{tot} and of the total fissile mass of plutonium Pu_{fiss} in a multi-recycling PWR scenario, 1300 MWe, 45 MWd/kg, and a recycling time of 12.5 years [4].

Table 1: Average amount of fission neutrons for a single neutron, which has been absorbed in fuel pins of Light Water Reactors or Liquid Metal Burner Reactors. This amount is given for several actinides.

Nuclide	LWR	LMFBR
U -235	1.98	1.92
U -236	0.11	0.43
U -238	0.29	0.39
Np-237	0.05	0.49
Pu-238	0.25	2.02
Pu-239	1.84	2.29
Pu-240	0.02	1.24
Pu-241	2.20	2.45
Pu-242	0.05	1.10
Am-241	0.04	0.46
Cm-242	0.64	1.33
Cm-243	2.94	2.99
Cm-244	0.20	1.58

Each proton of high energy (one GeV or more) will liberate dozens of spallation neutrons from a heavy-metal target [7]. Either directly or after moderation these neutrons can be used to transmute actinides or fission products. In a slightly subcritical booster setup the number of neutrons might be multiplied with a factor $k_{eff}/(1-k_{eff})$. Cost and safety aspects of neutron generation and the amount of excess neutrons both depend on the value of k_{eff}. For small values the safety of the system might be best; on the other hand costs for electricity and capital investment in the accelerator would be less if k_{eff} would be close to one. In comparing the different proposed systems, the safety-cost balance for actinides is quite different than for fission products. First of all actinides are a potential source of neutrons [8] and secondly for actinide-transmutation by fission fast neutrons are most suitable, whereas fission-products rather transmute by capture of moderated neutrons [9].

3. TRANSMUTATION of FISSION PRODUCTS

In contrast to the situation for the actinides any thermal reactor could be used to diminish the toxicity of fission products. In principle even commercial LWRs could be used to get rid of the technetium and perhaps also of the iodine-129. An efficient transmutation at low flux would however require huge loadings of waste, additional fuel enrichment and extra recanning and reprocessing efforts. Reactors with a somewhat higher flux (HWRs like CANDU) and with possibilities for refuelling on-line, are therefore being studied. High-flux thermal reactors could be entirely dedicated to transmutation, and moderated sub-assemblies of high flux fast reactors might also have potentials, as is shown in a contribution of Kloosterman and Li to this meeting. For a large scale transmutation of long-lived fission products a feasible and economic technology is however not yet available, and there still are several limitations related to safety and cost aspects.

Fission products with low cross sections would require extra neutrons [7,8], and one considers application of high-energy proton accelerators as neutron boosters. Any accelerator-based system will however be less efficient than a corresponding reactor system without a booster, which can be seen as follows: One 1.5 GeV neutron produces 30 neutrons in a lead/tungsten target. Suppose the accelerator efficiency is about 50% (a very optimistic statement). In this case the price of one thermal neutron in terms of electrical energy is 1500/(30*0.5) = 100 MeV. As this electrical energy had to be derived from thermal energy one would have required about three times more energy. It is unrealistic to assume that each neutron will transmute a nucleus, but even then one would need an equivalent of at least 300 MeV thermal energy to transmute one nucleus. If this 300 MeV would have been generated by means of a nuclear reactor, this would mean that more than one fission in the reactor is needed to transmute one nucleus at the accelerator. A direct transmutation in the reactor seems more economic because then also one free neutron might become available for each fission in the reactor itself. Reactor-based transmutation of a technetium nucleus is clearly more direct because the 200 MeV, which is generated by fission, will still be useful to generate electricity, which is no longer needed for the accelerator. In hybrid accelerator-reactor combinations each neutron from the accelerator-target system might produce again up to ten new fission neutrons in a sub-critical assembly, and transmutation costs of such systems could be in between that of pure accelerators and that of critical reactors [9,10]. An advantage for accelerator scenarios could be the good neutron economy [8], which would allow for a better loading and lower handling costs.

4. REDUCTION OF DOSE-RISKS FROM LEAKAGE

As long as the integrity of a disposal site is guaranteed, *long-lived fission products will determine the leakage-risks* (products of toxicity and mobility). For an unperturbed granite repository [11] table 2 shows the risk due to spent LWR fuel in a once through scenario [9]. It is seen from this table that the unspent uranium would give the main residual actinide contribution, and that Tc and I-129 dominate dose-risks. Diffusion of anions of iodine and per-technate in ground water is more rapid than flow of the ground water itself, as is now again being realized.

Any collective dose rate could be compared with the natural rate from radon, and even for the most relevant long-lived fission product Tc-99 the collective dose-risk is only marginal. If it can be assumed that in due time the Tc will distribute itself evenly in time and space over the world, a value of 10^{-12} Sv/year would be the order of magnitude for the personal average dose-rate due to electro-nuclear production of one GW(e) year. This value corresponds to about one part per billion in terms of the natural radon dose-rate.

TABLE 2: TIME-INTEGRATED LEAKAGE-DOSE DUE TO SPENT LWR FUEL
(direct storage of spent fuel, due to nuclear generation of one GW(e) year)

Period :	One million years	Hundred million years
Nuclides :		
Tc-99	98 %	46 %
I -129	2 %	1 %
Cs-135		24 %
U -235		6 %
U -238		14 %
Np-237		5 %
Pu-239		4 %
man Sv	9000 *)	20 000 *)

*) Collective dose for the global population. The average yearly individual dose may be obtained by dividing by the affected number of people and the indicated period.

Arguments on the collective leakage-dose should be treated with some caution. First of all it is questionable whether the risk should be ranked as in table 1 (a collective integrated dose from leakage out of a repository). If one would for example rank according to the highest possible individual dose, the I-129 risk could dominate for repositories of clay [12] or rock salt [9]. Local dose-risks are almost entirely due to uptake of iodine in the thyroid. This gland usually contains about 10 mg of iodine, and risks could be made marginal by diluting the I-129 isotopically with natural iodine. In most transmutation scenarios collective leakage doses are mostly due to U-234.

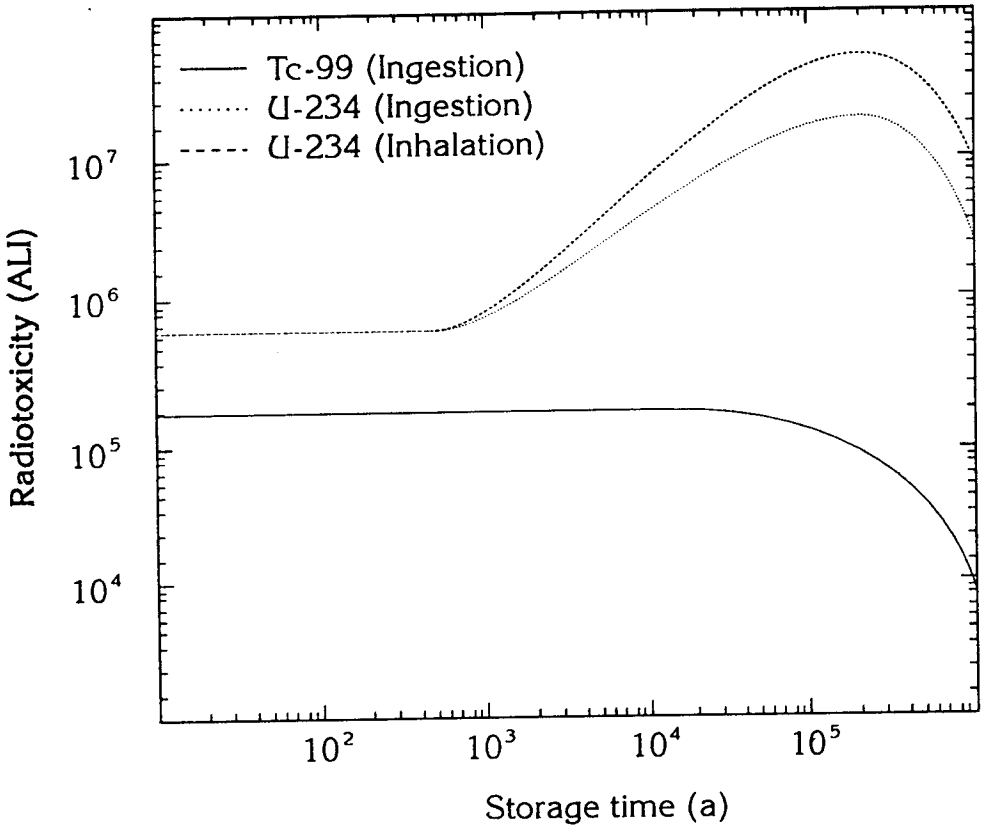

Fig.2: The radio-toxicity of Tc compared to that of U-234 and its daughters. The amount of material (9.6 kg of Tc-99 and 2 kg of U-234) is related to the yearly release of the Dutch reactor at Borssele (0.5 GW(e)).

Figure 2 shows for comparison the radio-toxicity of the Tc and the U-234, as these are released yearly by the Dutch reactor at Borssele (0.5 GW(e)). It should be noted that the dose-risk follows the radio-toxicity as given in the figure 2 if it is multiplied by the fraction of the material, which will reach the human biosphere. This might amplify the relative hazard of the U-234 considerably, as the geo-chemical mobility of radon is higher than that of technetium, and the reprocessed uranium (REPU) will only be subjected to shallow land burial in present scenarios. Therefore it is clear that the U-234 and its radon emanation leads to a much higher dose-risk than the corresponding amount of technetium, especially after a few thousand years when the extremely mobile radon-gas will be liberated by the radium, which has been formed in the mean time.

Due to the fact that U-234 will in the long run lead to mobile Rn-222 one could for example double the local dose due to emanations for a few hundred thousand years by spilling only a few hundred milligrams of U-234 or one of its precursors into the soil over a surface of only a few square kilometres. This isotope of uranium, which also occurs naturally (with an isotopic abundance of 0.0055 %) is responsible for most of the present radiation dose to mankind. Its most dangerous daughter Ra-226 has a half life of 1600 year, and about 60 t of radium in the

soil emanates almost 6 litre of radon each day, which builds up an equilibrium value of about 20 litre of radon in the total biosphere of our planet. This tiny amount nevertheless contributes for about 60 % to the total radiation dose, which averages to about 10^{-3} Sv per person yearly and this leads to an estimate of the collective dose of the order of $5 * 10^6$ man Sv/year. It would therefore be environmentally very unwanted if the present amount of U-234 would increase, and it might very well be that build-up of U-234 in the soil gives a much higher dose than the so much feared build-up of heavier actinides.

5. RISKS IN DIFFERENT TRANSMUTATION SCENARIOS

There are long-term dose-risks in regular transmutation scenarios, but also in end-scenarios, in which production of nuclear energy would be ended and large quantities of fissile waste are to be disposed of at once. Similar situations as in end-scenarios would occur if large quantities of weapon plutonium have to be transmuted [13]. Procedures would depend on the criteria to be adopted. In case of ultimate safeguarding requirements one could pollute the Pu-239 with less fissile material by irradiating it with thermal neutrons. This renders the plutonium less useful as a weapons material, and makes it hard to divert. This procedure would however increase the toxicity and the Pu-238 content considerably, and one would need to store the irradiated plutonium for a long time, and possibly in an irreversible and geologically secured way. As this storage procedure would hardly comply with present public demands for waste disposal, any acceptable end-scenario should be terminated by the fission of its actinide waste. This could only be achieved by means of an extended use of fast reactors, which however also yield increasing amounts of potential radon-emitters in the waste such as U-234, Pu-238, or Cm-242. It seems therefore very likely that non of the present scenarios for reduction of the actinides fulfils the third criterium mentioned upfront of this paper. If this situation cannot be improved, there might be very little motivation left to transmute Tc-99 and I-129 in end-scenarios. Release of potential radon emitters in the waste should be guarded against in any scenario, and it might be conceivable to recycle the spent uranium (REPU) together with the other actinides. The scenario in which it is most likely that all the uranium will be recycled, is the Th/U breeding cycle, which therefore will be treated below:

Relative dose risks and proliferation risks for the Th/U Cycle

Competing demands of safety, economy and non-proliferation also apply to the thorium cycle, in which capture breeds Pa-233 and the fissile U-233 is formed by the β-decay thereof. By parasitic capture in Pa-233 and in U-233 some U-234 will be formed, and after many cycles the isotopic dilution of the U-233 would reduce the risk for proliferation [14]. In this respect the Th/U cycle would be safer than the U/Pu cycle, in which the recycled plutonium is always considered to be weapons grade as soon as it can be purified chemically and traces of for example americium can be removed. On the other hand the U-233 as produced in the first few Th/U cycles is also weapons grade. Further it is possible to obtain isotopically pure U-233 by decay of Pa-233 in an external cycle. In the equilibrium fuel the rate of formation of Pa-233 by neutron capture is equal to its rate of decay into U-233 ($T_{1/2}$ = 27 d). Therefore the Pa-233 concentration will be an increasing function of the neutron flux in the fuel, and especially for high flux systems parasitic capture will remove many neutrons. Therefore it has been proposed to apply either an external neutron source or an extra initial enrichment of the fuel. Especially high-flux systems would need these fissile additives and/or a powerful accelerator to provide for the extra neutrons [13], and even then these systems are to be refuelled frequently and on-line partitioning of the Pa-233 is needed in order to reduce parasitic absorbtion. As was discussed above partitioning of Pa-233 leads to pure weapons grade U-233, and this procedure would require special controls at the facility. Apart from the proliferation problems one has the problem of the high radiation field at the reprocessing factory. Unless regular clean-ups can be performed, high radiation levels at reprocessing facilities result from the hard (2.6 MeV) gamma radiation from Tl-208, as produced at the end of a chain of α and β-decay according to: U-232 -> Th-228 -> ····· -> Tl-208.

In a low-flux system only a small external neutron source [14] or the addition of little extra plutonium to the fuel could compensate for capture losses in Pa-233, and the above mentioned on-line reprocessing would no longer be required. This is important, because as was discussed above, spilling of U-234 should be minimized. Application of an extremely long burn-up, possibly with a regular recanning of the fuel and an increased accelerator power, could limit the U-234 spilling. Nevertheless it should be realized that the isotopic contents of U-233,234 mixtures could range from 30-50 % in the equilibrium fuel. Because dose-risks due to Rn-222 might dominate over dose-risks from any actinide in the long term, *the Th/U cycle might give a very high long-time dose-risk due to radon emanation especially if one ever decides to stop the cycle. Any scenario in which the waste from the back-end of the Th/U cycle would come to the surface either by intrusion or by accident, would for example dwarf the technetium risk, unless gas tight disposal of the remains of U-234 is considered to prevent the radon to reach the atmosphere.*

6. CONCLUDING REMARKS and RECOMMENDATIONS

Transmutation of the existing plutonium is priority number one. After all proliferation risks are most clear for plutonium, and possibilities for future mining for this element should be eliminated to prevent very long lasting proliferation risks. No exclusive LWR-scenario has yet been found, which entirely solves the plutonium problem by transmutation.

Reduction of minor-actinides is priority number two. After all these actinides contribute for about 10 % to the total long-term toxicity of once used spent nuclear fuel. Recycling in PWRs will only increase the minor actinides. Fast reactors will probably be most suited to reach a high fission rate in a high neutron flux. Safety aspects of fast reactors, which are loaded with minor actinides could possibly be controlled by means of external neutron sources.

Criteria on the reduction of dose risks disfavour transmutation scenarios in which the waste will be contaminated with U-234 or with one of its precursors, unless a disposal method is applied that prevents radon emanation from the waste to reach the atmosphere.

Reductions of collective dose-risks, which are far below the natural dose-risks seem at first sight to be of less relevance. In proper disposal scenarios the world- and time-integrated collective dose from fission products is less than 10 000 man Sv for each GWe year, and thereby less than a fraction 10^{-9} of the natural dose risk. Resistance against the dumping of low-level nuclear waste into the ocean has shown that there are incentives to reduce the dose even below such marginally small values. These incentives seem hardly motivated by the wish to reduce dose risks to human beings, and in this respect it is illuminating to recall the argument on the dose risk from the fission product I-129, for which other people argue that "isotopic dilution with natural iodine should reduce the highest individual doses". As long as no agreement exists on existence of safe thresholds for dose-risks, priorities are a matter of taste, and are therefore subject to changes.

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REFERENCES

1. T.H. Pigford, Trans. of the ANS 62 (1990) 97, and 63 (1991) 80.

2. A.F. Fritzsche, "The Health Risk of Energy Production", Risk Anal. 9 565-577, 1992.

3. E. Schmidt, E. Zamorani, W. Hage, and S. Guardini, "Assessment Studies on Nuclear Transmutation of by-product Actinides". Commission of the European Communities, Joint Centre ISPRA Establishment, S.A./1.05.03.83.13.

4. G.J. Schlosser, S. Winnik, IEAE-SM-294/33 (1987), and draft status report from H. Küsters

5. K. Abrahams et al, Specific contributions of the Dutch programme RAS towards accelerator-based transmutation, Contribution to the International Conference on Accelerator-Based Transmutation Technologies and Applications, Las Vegas, July, 1994.

6. Hiroshi Takahashi and Herbert Rief, Concepts of Accelerator Based Transmutation Systems, OECD Specialists' Meeting on Accelerator Based Transmutation (Wuerenlingen/Villigen 1992),

   and H. Takada et al, Proc. Int. Conf. Emerg. Nucl. Energy systems (1991).
   and G. Schlosser to the NSC on Pu utilization.

7. L.H. Baetsle, "Role and Influence of Partitioning and Transmutation on the Management of Nuclear Waste Streams" NEA/P&T REPORT No 3 1992".

8. M. Salvatores, I. Slessarev, and M. Uematsu, "A Global Physics Approach to Transmutation of Radioactive Nuclei", Nuclear Science an Engineering, 116 (1994), 1.

9. K. Abrahams, "Transmutation of Long-Lived Fission Products", ECN-RX-94-003, IAEA Technical Committee Meeting on: Safety and environmental aspects of partitioning and transmutation of actinides and fission products, Vienna, 29-11-93.

10. A.J. Janssen, Transmutation of Fission Products in Reactors and Accelerator-driven Systems: Some Critical Remarks, Report to the CEC, and ECN--I-94-001 (1994).

11. S.F. Mobbs et al, "Comparison of Waste Management Aspects of Spent Fuel Disposal and Reprocessing: Post-Disposal Radiological Impact", EUR 13561 EN (1991).

12. L.H. Baetsle, Proc. of the Information Exchange Meeting on Act. and Fiss. Product Separation and Transmutation, Mito City, Japan, 7-11-90, OECD 37290 (1991) 299.

13. C.D. Bowman et al, "Nuclear Energy Generation and Waste Transmutation using an Accelerator-driven Intense Thermal Neutron Source", Nuclear Instruments and Methods in Physics Research A 320 (1992) 336-367.

14. F. Carmati, R. Klapisch, J.P. Revol, Ch. Roche, J.A. Rubio, and C. Rubia, "An Energy Amplifier for Cleaner and Inexhaustible Nuclear Energy Production by a Particle Beam Accelerator", CERN/AT/93-47.