Nuclear Waste Transmutation

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Abstract

Accelerators can play a role in the disposal of long-lived radioactive waste: an alternative to the storage in deep underground repositories might be transmuting long-lived elements into stable or short-lived ones in subcritical systems driven by spallation neutrons. These neutrons would be produced by a high intensity, intermediate energy proton accelerator irradiating a heavy target. Similar systems have also been proposed to produce energy with a minimized waste inventory. Since a good knowledge of the spallation process is essential for designing and optimizing the target-blanket assembly, new programmes aimed at studying spallation reactions are in progress.

1 Long-lived nuclear waste

The disposal of radioactive waste arising from nuclear power production or from the dismantling of nuclear weapons is becoming a major public environmental concern. The spent fuel of a PWR (pressurized water reactor) is composed of 96% uranium, 1% plutonium and 3% minor actinides (MA) and fission products (FP). Some of the isotopes have a very long lifetime, potentially presenting very long term radiation hazards. As an example, Table 1 shows the half-lives and amounts of the principal long-lived isotopes (defined as those with a half-life longer than 30 years) found in the fuel unloaded every year in France from the 55 PWRs in operation. In some countries (France, GB, Japan) spent fuel is reprocessed to extract uranium and plutonium for further use while in others (like the USA or Switzerland) unloaded fuel is intended to be stored directly. Until recently, the only seriously considered solution for the disposal of long-lived waste was the storage in deep underground geologically stable repositories. However, this solution suffers from the uncertainties of the safety and impregnability of the storage over hundreds of thousands of years and also from negative public opinion. That is why the possibility of partitioning and transmuting long-lived radioactive waste into stable or short-lived isotopes which could then be surface-stored is now being investigated. Fig.1 shows how the long-term radiotoxicity, defined as the quantity of water required to dilute the material to reach safe drinking water standards, of the spent fuel can be reduced if most of the long-lived isotopes are extracted. It can be seen that the removal of all the actinides reduces the toxicity to

Element (quantity)	Isotope	Half-life (years)	Quantity (ton/year)
Plutonium (11.4 ton/year)	^{238}Pu	88	0.19
	^{239}Pu	$2.4.10^4$	6.53
	^{240}Pu	$6.5.10^3$	2.52
Minor actinides (1.1 ton/year)	^{237}Np	$2.1.10^6$	0.48
	^{241}Am	430	0.25
	^{243}Am	$7.4.10^3$	0.14
	^{245}Cm	$8.5.10^3$	0.001
Fission products (39 ton/year)	^{135}Cs	$2.3.10^6$	0.4
	^{99}Tc	$2.1.10^5$	1.0
	^{93}Zr	$1.5.10^6$	0.9
	^{129}I	$1.0.120^7$	0.2
	^{107}Pd	$6.5.10^6$	0.25

Table 1
Half-life and amount of the principal long-lived isotopes in the fuel unloaded every year from all the French reactors

the level of that of uranium ore after about 400 years. The additional removal of some of the FP reduces the long-term radiotoxicity further and contributes drastically to the lowering of the short-term hazards because of the 30 year half-life isotopes ^{90}Sr and ^{137}Cs. It should be stressed that this definition of the waste toxicity does not take into account the fact that some of the elements have higher chemical reactivity and mobility than others and consequently are expected to migrate more easily into the biosphere.

To transmute long-lived isotopes into stable or short-lived elements two nuclear reactions can be used: the capture of one neutron, or (for actinides) neutron induced fission, both processes occurring in a reactor-type neutron flux. Other options which were once suggested (direct spallation, (γ, n) reactions...), seem to be economically unrealistic [1]. Studies on the transmutation capabilities of reactors are being conducted in several countries, for instance in France in the SPIN program [2] or in Japan in the OMEGA program[3]. A recent analysis by Kusters et al [4] seems to indicate that classic thermal reactors have a very low transmutation potential because of a very tight neutron balance while fast neutron reactors are more promising because of the more favourable fission-to-capture ratio in a fast neutron spectrum. However, they seem to be able to achieve only a slow reduction of the MA inventory. None of them is efficient in transmuting FP because of the lack of extra available neutrons.

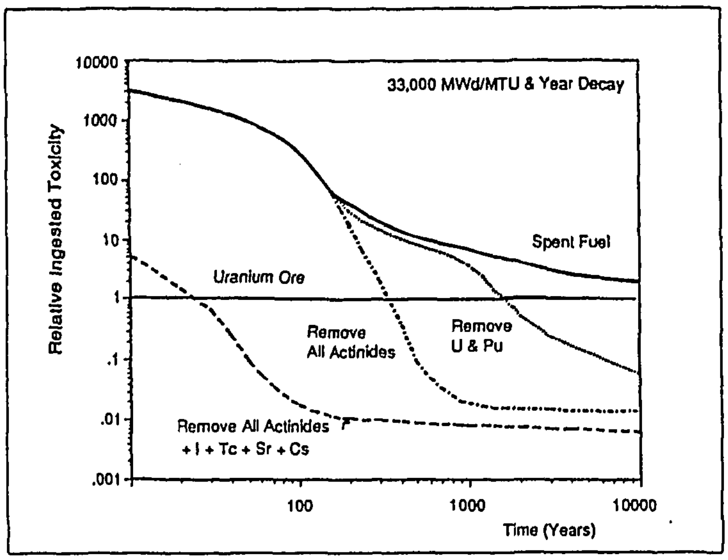


Fig. 1. *Ingested radiotoxicity of spent fuel relative to the toxicity of the natural uranium ore, assuming different waste processing scenarios. From ref.[1]*

2 Accelerator-driven systems

The need for a large excess of neutrons to transmute significant amounts of waste has led to proposals for the use of accelerator-driven reactors. In such systems, spallation reactions induced by a high-intensity beam (10 to 250 mA) of intermediate-energy (around 1 GeV) protons on a heavy target produce an intense neutron flux. These neutrons, after being more or less moderated, are used to drive a sub-critical blanket. The extra neutrons provided by the accelerator allow the maintenance of the chain reaction while burning the long-lived nuclear wastes. The system generates electricity, part of which is used to supply the accelerator. The idea of using an accelerator to feed a sub-critical reactor is not new but it has recently come back into favour because, with the progress in accelerator technology[5], the required high intensities seem no longer out of reach.

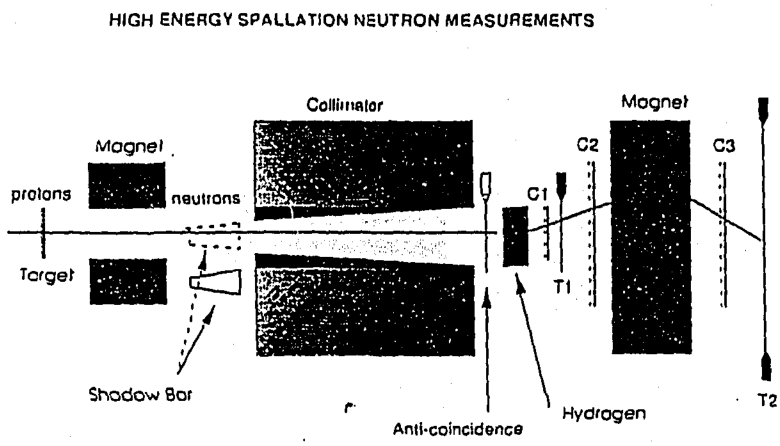
Various concepts of accelerator-driven systems have been proposed by several groups [3,6–9]. They differ in their goals: destruction of FP, MA or military Pu, or energy production with a minimized waste inventory. The technological choices regarding fuel, moderator, coolant, accelerator, etc., are also very different. In the ATW proposal from Los Alamos[6], for instance, a very high flux (up to 100 times the flux in a classic PWR) of thermal neutrons allows the burning of both MA and PF. In such a high flux, a MA like ^{237}Np, which is a poison in normal reactors, is expected to become a fuel because it can undergo two sequential neutron captures. The ATW system has a high burning efficiency and the advantage of a low actinide inventory, but it is based on molten salt reactor technology which has never been industrially implemented and is dependent on very complicated on-line chemical separation processes. Most of the other concepts are based on a fast neutron spectrum to take advantage of the higher fission-to-capture ratio to burn MA. The choice of the nuclear fuel also varies according to the concept. For the Energy Amplifier [8], the $^{232}Th/^{233}U$ fuel cycle is preferred (as in some ATW versions) because it is claimed that it generates less long-lived MA and is more proliferation resistant. The Energy Amplifier is one of the few examples where the accelerator is a cyclotron (of 10 mA). In most cases the proton beam is delivered by a linear accelerator (from 30 to 250 mA depending on the project).

Besides the more favourable neutron economy, additional advantages of accelerator-driven systems are safety and versatility. The introduction of actinides into a reactor makes it more sensitive to reactivity changes and consequently more difficult to control. This is mainly due to the smaller fraction of delayed neutrons. Obviously the operation of the reactor in a sub-critical state with an accelerator as an external source that can be shut down in case of a sudden increase of reactivity is a major advantage. Also, accelerator-driven systems are more flexible than reactors since the intensity of the accelerator can be adjusted to counteract the growth of poisonous isotopes or when adding elements to be transmuted. Their main drawbacks are their complexity and the technological progress they imply for the accelerator, the target-blanket and the interface between them.

3 Physics studies for spallation-driven systems

The design and optimization of the target-blanket assembly requires a detailed knowledge of the spallation process. In particular, it will be important to know:
i) the number of spallation neutrons produced per incident proton which is related to the ratio of the power produced to the power delivered by the accelerator and is thus important for evaluating the profitability of the system;
ii) the energy spectrum and angular distribution of the spallation particles, which are necessary for the optimization of the target geometry. They are also essential for estimating radiation damage in target and structural materials which is expected to be very severe, due to the high energy of the escaping particles;
iii) the isotopic distribution of the residual nuclides produced in the spallation target in order to check that one does not create more undesirable radioactive isotopes than are destroyed.

Simulation codes describing elementary production of particles in nuclear reactions as well as the transport of these particles in thick targets are available. However, recent intercomparisons[10,11] of these codes have shown their limits of reliability especially above 800 MeV where experimental data are scarce. Since in most of the proposed concepts proton beam energies are between 0.8 and 1.6 GeV, new data are needed. Two types of measurement are necessary: the generation of fundamental nuclear data to improve basic nuclear models used in simulation codes; and thick target measurements to validate the transport part of the codes and determine the performance of spallation targets. These studies are also useful for other applications: spallation sources for solid state physics, tritium production with accelerators, radiation damage in space, etc...



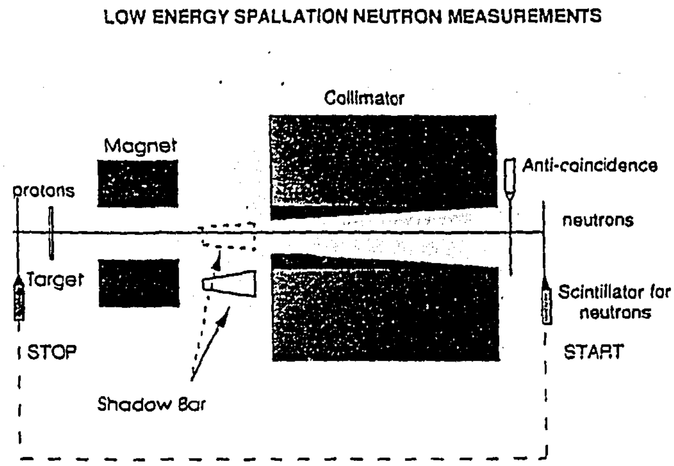


Fig. 2. *Experimental set-up for the measurement of double differential cross-sections for the production of spallation neutrons.*

3.1 Neutron production

-Energy and angular distributions.

A new programme[12] aimed at measuring the double differential cross-sections for the production of neutrons induced by protons and deuterons on various targets, has begun at the Saturne accelerator in Saclay. Since Saturne is not a pulsed machine, conventional time-of-flight measurements are not possible. Two different experimental techniques are thus used for these measurements: for the low energy part of the neutron spectrum (2 MeV to 400 MeV), time-of-flight between the incident tagged proton and a NE213-scintillator detecting the neutron is employed. At higher energies the flight path is too short to allow a sufficient energy resolution. Therefore, the neutron energy spectrum, from 200 MeV to the beam energy, is obtained through the detection of recoil protons in a magnetic spectrometer, after scattering in a liquid hydrogen

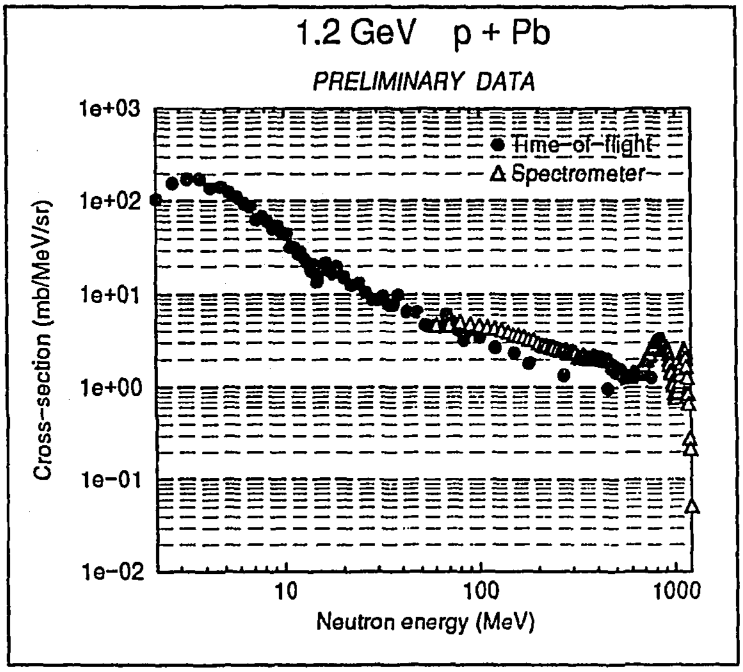


Fig. 3. *Preliminary results obtained for the production at* $0°$ *of neutrons in a thin Pb target induced by* 1.2 *GeV protons. Top: measurement of neutron energy from* 200 *MeV to the beam energy by (n,p) conversion. Bottom: measurement of neutron energy from* 2 *to* 400 *MeV by time of flight.*

convertor. The efficiency of the detectors is determined through the measurement of quasi-monoenergetic neutrons obtained by deuteron breakup on a Be target. Fig.2 shows both experimental set-ups. So far, measurements have been made of neutrons produced at 0° by 800 MeV to 1.6 GeV protons and deuterons on several targets. Analysis of the data is in progress and fig.3 shows preliminary results obtained for the reaction $p + Pb$ at 1.2 GeV. Once all the efficiency and background corrections have been made, the two curves are expected to overlap more closely. In the future, the experimental set-up will be moved to a larger area and modified to allow for the measurement of complete angular distributions. The programme will also be extended to the study of neutron energy spectra from thick targets. Measurements with different length, diameter and composition of targets will be made.

-Neutron multiplicity measurements

The ORION collaboration (GANIL-Berlin-SATURNE-Orsay-Liège) has measured at Saturne neutron multiplicity distributions obtained in reactions induced by protons or ^3He projectiles on a series of thin targets[13]. The apparatus used is the 4π liquid scintillator detector loaded with gadolinium, ORION, which can measure the number of emitted neutrons event by event. Results have shown that the multiplicity distributions obtained with protons or ^3He of the same total energy are very similar. A

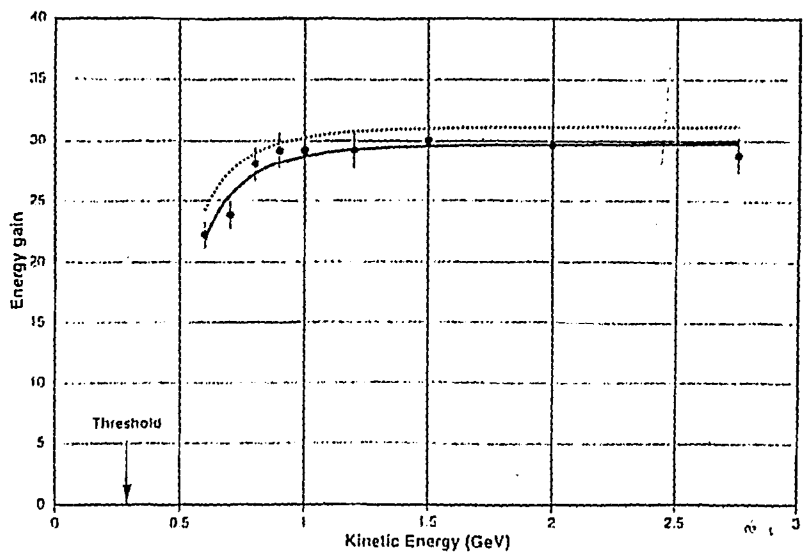


Fig. 4. *Average energy gain, i.e energy produced in the device to the energy delivered by the beam, in the sub-critical arrangement as a function of beam energy. From ref.[8].*

strong dependence on the target and on the projectile energy has also been observed. Similar measurements on thick targets are now being conducted at CERN.

-Integral neutron production on thick targets.

Recently an experimental determination of the energy generated by spallation by a high energy beam has been carried out at CERN-PS by Rubbia's group[14]. A sub-critical assembly made of natural U moderated by water was irradiated by a proton beam at several kinetic energies from 0.6 to 2.75 GeV and the energy gain was measured by the temperature rise and by counting fission reactions inside the device. Fig.4 shows the average energy gain obtained as a function of the beam energy, and a comparison with a simulation. It can be seen that the gain increases with beam energy up to 1 GeV above which it remains essentially constant. This indicates that a convenient choice for the beam energy in this case would be around 1 GeV.

Also, a group from Los Alamos[15] is planning to determine the absolute proton-to-neutron production rate by protons and deuterons on various spallation targets at Saturne. This will be done by measuring the number of neutrons thermalized in a manganese sulfate solution surrounding the target-blanket assembly. The purpose of this experiment is to extend previous experiments done at LAMPF (up to 800 MeV) to higher energies.

3.2 Residual nuclide production

Since, at present, models are not able to reliably predict the yield of particular nuclides, a large amount of data are needed. A collaboration led by Michel [16] has been studying the production of nuclides in spallation reactions for several years, and on several facilities, with the aim of interpreting the observed abundance of cosmo-

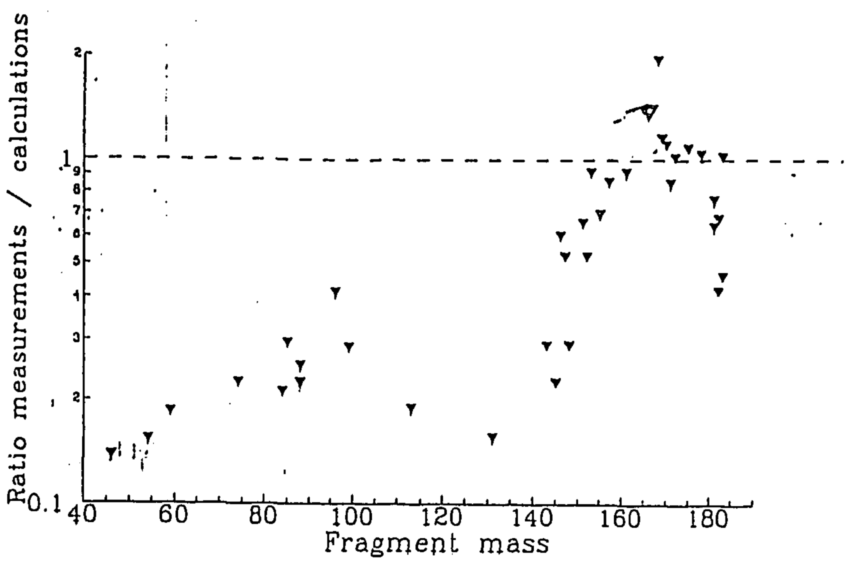


Fig. 5. *Ratio of the measured to calculated residual nuclide production in a thin W target irradiated by 800 MeV protons from ref.[16]. Calculations are done with the HETC code from Bruyères-le-Châtel[17].*

genic nuclides. This programme has now been extended to measurements on targets relevant to accelerator-based systems[17]. Results obtained by another group[18] at Saturne by γ-spectrometry on thin W targets are shown in fig.3, in which the ratio of the experimental data to a simulation code [19] is presented. It can be seen that while direct spallation product yields are more or less reproduced by the code, fission product yields are systematically overestimated by the calculation. The same group is conducting thick target experiments in which the production of the residual-nuclides is studied by analyzing activation products in thin foils placed at different longitudinal and radial positions inside a thick lead target.

At PSI irradiation of thin samples of actinides are being done in the ATHENA experiment [20]. The purpose of this experiment is to provide experimental data on spallation and fission product yields with a view to validating the fission models entering into the simulation codes.

Finally, there is a project to measure the complete reaction product distribution by an inverse-kinematic experiment at GSI [21].

4 Conclusion

Accelerator-driven sub-critical systems might offer attractive possibilities in the transmutation of long-lived radioactive waste in terms of larger transmutation capabilities than reactors, safety and flexibility. However, their technical feasibility is not yet proven and their cost might appear prohibitive. At present, accelerators are needed to provide new experimental data for evaluating the feasibility and then designing these systems.

References

[1] H.Takahashi and H.Rief, Proceedings of Specialists' Meeting on Accelerator-Based Transmutation, PSI Zurich, March 1992.

[2] M. Salvatores et al., GLOBAL'93, Int. Conf. on Future Nuclear Systems, Seattle, Washington, USA, September 93.

[3] T.Takizuka, Proceedings of the 8th Journees Saturne, Saclay May 5-6, 1994.

[4] H. Kusters and H.W. Weise, Trans. Am. Nucl. Soc. **64** (1991) 544.

[5] T. Stammbach, invited paper to this conference.

[6] C.D.Bowman et al., Nucl. Instr. and Meth. **A320** (1992) 336.

[7] G.J. Van Tuyle et al., Nucl. Technol. **101** (1993) 1.

[8] F. Carminati et al., preprint CERN/AT/93-47/ET (1993).

[9] I. Slessarev et al., Int. Conf. on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, NV, USA, August 1994.

[10] M. Blann et al., International Code Comparison for Intermediate Energy Nuclear Data, OECD/NEA (1994).

[11] D. Filges et al., OECD thick target benchmark, Report NSC/DOC(95)2 (1995).

[12] F. Brochard et al., Proceedings of the 8th Journees Saturne, Saclay, May 5-6, 1994.

[13] L. Pienkowski et al., Phys. Lett. **B336** (1994) 147.

[14] S. Andriamonje et al., Phys. Lett. **B348** (1995) 697.

[15] G. Morgan et al., Reasearch proposal at Saturne.

[16] R. Michel, Proceedings of Int. Conf. on Nuclear Data for Science and Technology, Gatlinburg, Tenessee, USA, May 9-13 (1994), and invited paper to this conference.

[17] D. Gloris et al., communication to this conference.

[18] J.L. Flament and J. Laurec, CEA - Bruyères-le-Châtel, private communication.

[19] Code TIERCE, CEA - Bruyères-le-Châtel.

[20] P. Wydler et al., Proceedings of the 8th Journees Saturne, Saclay, May 5-6, 1994.

[21] K.H. Schmidt et al., Proceedings of the 8th Journees Saturne, Saclay, May 5-6, 1994.