RESEARCH ACTIVITIES RELATED TO ACCELERATOR-BASED TRANSMUTATION AT PSI

P. Wydler

Laboratory for Reactor Physics and Systems Technology

Abstract

Transmutation of actinides and fission products using reactors and other types of nuclear systems may play a role in future waste management schemes. Possible advantages of separation and transmutation are: volume reductions, the re-use of materials, the avoidance of a cumulative risk, and limiting the duration of the risk. With its experience in reactor physics, accelerator-based physics, and the development of the SINQ spallation neutron source, PSI is in a good position to perform basic theoretical and experimental studies relating to the accelerator-based transmutation of actinides.

Theoretical studies at PSI have been concentrated, so far, on systems in which protons are used directly to transmute actinides. With such systems and appropriate recycling schemes, the studies showed that, considerable reduction factors for long-term toxicity can be obtained.

With the aim of solving some specific data and methods problems related to these types of systems, a programme of differential and integral measurements at the PSI ring accelerator has been initiated. In a first phase of this programme, thin samples of actinides will be irradiated with 590 MeV protons, using an existing irradiation facility. The generated spallation and fission products will be analysed using different experimental techniques, and the results will be compared with theoretical predictions based on high-energy nucleon-meson transport calculations. The principal motivation for these experiments is to resolve discrepancies observed between calculations based on different high-energy fission models. In a second phase of the programme, it is proposed to study the neutronic behaviour of multiplying target-blanket assemblies with the help of zero-power experiments set up at a separate, dedicated beam line of the accelerator. These experiments are intended to provide integral checks on the nuclear data (models) and calculational methods for specific target-blanket configurations.

1 Introduction

In Switzerland, transmutation is currently not considered as a method for simplifying the country's radioactive waste management problems. Because of the institution of a 10-year nuclear moratorium, decisions for replacing existing, or launching new, nuclear power plants have to be deferred at least until the turn of the century. From a short- to medium-term point of view, the radioactive waste management problem is therefore limited: the National Cooperative for the Storage of Radioactive Waste assumes a 120 GWe-year scenario and has a firm policy to establish low-level and to plan medium- and high-level waste repositories in suitable geological formations. The associated risk for the population is considered to be negligible.

On the other hand, countries with larger nuclear power programmes, such as Japan and France, are taking an increasing interest in advanced reprocessing technologies and have meanwhile initiated major partitioning and transmutation programmes. In view of the long-term potential of nuclear fission energy, and since the Swiss Government has explicitly declared its will to keep the nuclear option open, there exists an incentive for investigating such technologies also in Switzerland. In the longer term, interest in these technologies will probably be strengthened, since the expected benefits, such as volume reductions of radiotoxic materials, re-use of materials, avoidance of a cumulative risk, and limiting the duration of the risk, may play an important role in the discussion of a 'sustainable development' of nuclear energy.

Transmutation of long-lived radiotoxic nuclides (actinides and fission products) into shorter-lived, less toxic species can be achieved with normal fast reactors, special burner reactors, and other types of nuclear systems. With its experience in reactor physics, accelerator-based physics, and the development of the SINQ spallation neutron source, PSI is in a good position to perform basic theoretical and experimental studies relating to *accelerator-based* transmutation systems. For a research institute such as PSI, work in this field is attractive, since accelerator-based systems are less developed and have a higher potential than reactor-based systems, and the associated research has a high scientific content. Studies of the theoretically achievable toxicity reduction for ^{237}Np and other long-lived minor actinides, which will be briefly described in Section 2, have confirmed the high potential of such systems.

Since there is no Swiss concept for a transmutation system, PSI has decided to focus its experimental work on basic or generic problems in areas where it can make the best use of its special experimental facilities. These comprise a 590 MeV proton accelerator, which is currently being upgraded to provide a current of $1 - 2\,mA$, different types of irradiation facilities with the necessary handling equipment, and a hot laboratory in which irradiated samples can be analysed using advanced examination methods. Building on this infrastructure, the Laboratory for Reactor Physics and Systems Technology is about to initiate a programme of measurements called *ATHENA* which is aimed at solving some specific 'data and methods' problems relating to the accelerator-based transmutation of actinides (*ATHENA* = actinide transmutation using high energy accelerators). A brief description of the first two phases of this programme *will be given in Sections 3 and 4.*

2 Transmutation Effectiveness of Spallation Reactions

Actinide transmutation systems must be designed primarily to fission the actinides. In the case of accelerator-based systems, this is achieved in two ways: in accelerator-driven 'thermal' systems (systems based on an intense thermal neutron source) nuclides which cannot be directly fissioned with thermal neutrons are converted to fissile nuclides by means of neutron capture reactions and the latter are fissioned before they can decay to other non-fissile nuclides; whereas in accelerator-driven 'fast' systems (driven subcritical systems with a fast neutron spectrum) the actinides are fissioned by fast neutrons, including evaporation neutrons from the spallation reactions, and by the high-energy reactions themselves.

Important advantages of utilising the high-energy reactions themselves as transmutation processes are the high fission probability (cf. Table 1) and the tendency of the high-energy reactions to directly reduce the mass of the bombarded nuclei and thus to mitigate the build-up of undesirable heavier reaction products. On the other hand, Table 1 shows that the high-energy reactions give only a modest energy multiplication factor, indicating the need for a powerful accelerator. The overall energy balance of transmutation systems with a high proportion of high-energy induced transmutations can, however, be improved by using the many free lower energy neutrons (about 30 per proton at a proton energy of 1 GeV) to generate additional fission power in a surrounding 'breeding' blanket [1].

To assess quantitatively the transmutation effectiveness of the spallation reactions, the toxicity of the remaining waste following transmutation of a minor actinide mixture with a composition corresponding to typical LWR spent fuel was calculated for four different recycling schemes. A (hypothetical) ideal transmutation facility with a quasi-infinite, non-multiplying target of pure minor actinides and negligible reprocessing losses were assumed. Recycling schemes 1.a to 1.c, illustrated in Fig. 1, are minor-actinide recycling schemes with and without partitioning of newly generated uranium and plutonium; scheme 2 is a full recycling scheme with recycling of all spallation products.

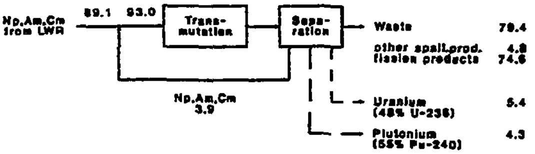

Figure 1: Minor-actinide recycling schemes without further partitioning (1.a), with partitioning of the newly generated plutonium (1.b), and with partitioning of plutonium and uranium (1.c). The numbers give the mass balance in kg/yr for a 300 MW proton beam and a load factor of 0.8. The partitioned uranium and plutonium could be recycled in LWRs together with other uranium and plutonium.

Figure 2 illustrates that the reduction of the radiological toxicity is highest for recycling scheme 2: beyond 200 years, the overall toxicity is reduced, theoretically, by more than 3 orders of magnitude compared with the toxicity of the original minor actinide mixture. It can be seen that the potential toxicity reduction strongly depends on the recycling scheme (newly generated uranium and plutonium should be separated from the waste). More comprehensive conclusions from this study are given in [2].

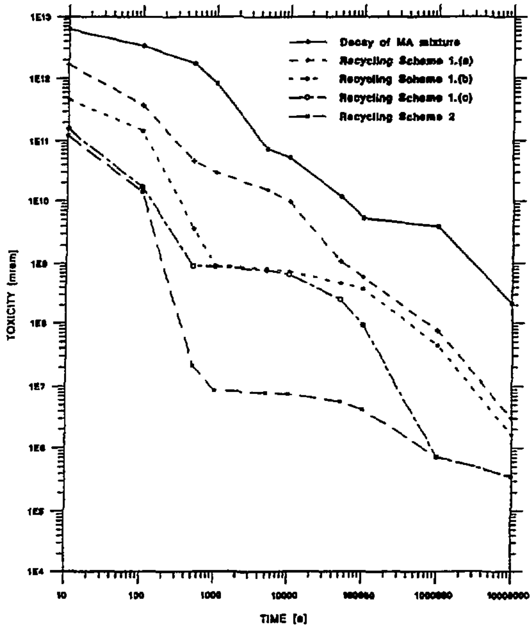

Figure 2: Long-term toxicity for ingestion of the remaining waste using different recycling schemes. The solid line gives the toxicity of the original minor actinide mixture and its decay products.

3 Irradiation of Thin Samples of Actinides

For all spallation-based transmutation systems, there is a greater or lesser need for a good understanding and an appropriate modelling of spallation processes involving not only medium-weight but also heavy nuclei. High-energy nucleon-meson transport codes, such as HETC, have usually been validated with a view to their use in the design of spallation neutron sources for solid-state physics applications [3]. In the context of transmutation, a correct prediction of the neutron source strength and power distribution in the target is not the only goal, but the code has also to be capable of correctly predicting the mass distribution of spallation and fission products, since the individual nuclides are associated with widely differing toxicities and half-lives [4].

Simple code comparisons for the irradiation of thin samples of actinides with high-energy protons have revealed considerable differences in the total yield and

Table 1: Yields per incident particle in a quasi-infinite target bombarded with 1 GeV protons.

Target material	Pure Np-237	Minor actinide mixture	Actinide mixture
No. of spallation products	1.1	1.0	1.0
No. of high-energy fissions	4.2	4.0	4.1
No. of transmuted nuclei	5.3	5.0	5.1
High-energy fission probability	0.79	0.80	0.80
Total energy deposited (GeV)	1.60	1.56	1.58
No. of neutrons below 15 MeV	30.9	30.5	31.0

the shape of the mass distribution for both spallation and fission products. Since pure theoretical models are being compared with experimentally adjusted models, these differences are partly understood. However, in view of the more stringent requirements indicated above, experimental verification is desirable, especially for actinides, where the experimental data are scarce.

Table 2: The ATHENA Programme

PHASE 0

- Analytical studies relating to spallation-based transmutation systems (e.g. calculation of theoretically possible toxicity reductions using different recycling schemes).
- Identification of gaps in our knowledge (relating, for example, to the nuclear models used in high-energy nucleon-meson transport codes).

PHASE 1 (1993–1995)

- Verification of the models by irradiation of thin Th, U, Np and Am samples with 590 MeV protons and analysis of the spallation and fission products using different experimental techniques.
- Improvement of the theoretical models.

PHASE 2 (from about 1995)

- Neutronic studies relating to multiplying target-blanket systems.
- Verification of data (models) and calculational methods by means of zero-power experiments at the PSI ring accelerator (measurement of integral parameters such as fission rate distributions and reaction rate ratios).

To check the models used in the calculation of the high-energy nuclear cascades and the de-excitation of the residual nuclides by fission and evaporation processes, a basic validation experiment, *ATHENA-1*, has been initiated (Table 2), in which thin samples of actinides will be irradiated with 590 MeV protons from the PSI ring accelerator. Mass spectrometry will be used to measure the mass distribution of the generated reaction products, and the results will be compared with nuclide concentrations measured independently using the total reflection X-ray fluorescence (TXRF) technique.

The actinides to be studied include ^{232}Th, ^{238}U, ^{237}Np, ^{241}Am and possibly other minor actinides. ^{238}U will be used in the first experiments because it is the easiest to handle. ^{232}Th is interesting from a theoretical viewpoint, because in the RAL fission model [5] its atomic number is close to the lower limit ($Z = 89$) of the region in which the systematics of Vandenbosh and Huizenga for the ratio of fission to neutron emission is applied. Thin samples, with a mass of about 1 g/cm^2, will be irradiated in the existing PIREX facility, which is designed for simulating displacement damage and impurity production in first-wall materials of fusion reactors [6]. The maximum proton beam current is 20 μA, allowing production rates for fission and spallation products of the order of 0.1 ng/s. The samples will be encapsulated in aluminium and mounted in a special irradiation head which provides cooling with a temperature-stabilised helium circuit. After the irradiations, the head assembly will be transported in a shielded flask to the hot laboratory, where the samples will be dismounted and prepared for the analyses.

The mass spectrometric analysis will be carried out using an inductively coupled plasma mass spectrometer (ICP-MS). The ICP-MS method allows multi-element and isotopic analyses to be made over a large range of concentrations, with the option of performing quantitative measurements by utilising calibration standards. For high-accuracy isotopic ratio measurements, thermoionic mass spectrometry is also considered. The necessary sample preparation technique, which may involve separation steps, is currently being developed.

The total reflection X-ray fluorescence technique [7] was originally developed for measuring impurities in the 'first-wall' materials irradiated in PIREX, but has also been applied successfully for low-level iodine detection in a reactor safety experiment. The measurements can be carried out either at PSI using normal X-ray tubes or, under more favourable conditions, at the Stanford Synchrotron Radiation Laboratory (SSRL) using an electron storage ring to produce extremely intense X-ray beams [8].

4 Neutronic Studies Related to Target-Blanket Systems

As regards neutronics, accelerator-driven thermal transmutation systems are not very different from continuous spallation neutron sources for solid-state physics applications (the main difference between the two systems is that the moderator is optimised, in the former case, for maximum transmutation rates and, in the latter case, for maximum neutron beam intensity). Therefore, in the neutronic design of accelerator-driven thermal transmutation systems, one can rely on experience with the design of spallation neutron sources, and there appears to be no immediate need for specific verification experiments. The SINQ spallation neutron source, which should become operational at PSI in 1995, will provide a good benchmark for checking the calculational methods more thoroughly.

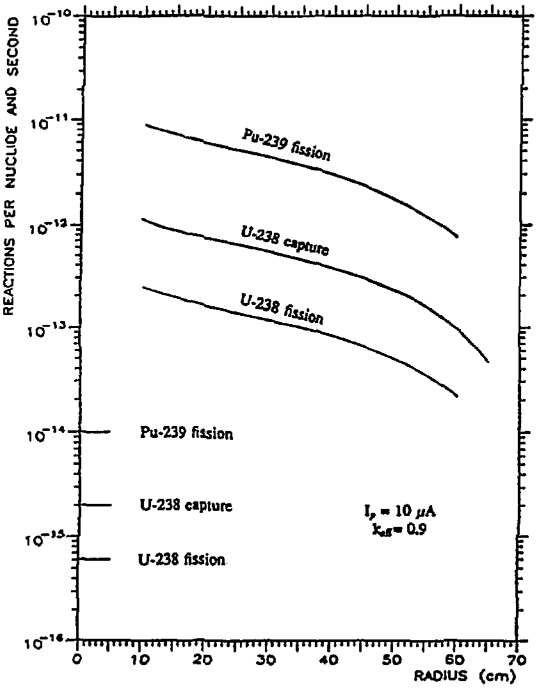

Figure 3: Predicted reaction rates in an accelerator-driven spherical blanket with mixed-oxide fuel. The marks indicate the lower limit of the applicability of measurement techniques. It can be seen that the important reaction rates can be measured easily with a proton beam current of $10\,\mu A$.

For accelerator-driven 'fast' systems, however, information on the adequacy of neutronic design methods is scarce. This applies particularly to systems in which protons are used directly to transmute actinides, and/or actinides are exposed to a neutron flux with a significant spallation neutron component. In a second phase of the *ATHENA* programme, it is therefore proposed to study the neutronic behaviour of multiplying target-blanket assemblies for different geometries and compositions with the help of zero-power experiments.

In these experiments, as in the case of zero-power experiments for normal fission reactors, the idea is to check the adequacy of the nuclear data (models) and the numerical methods by comparing calculated and measured integral parameters such as fission rate distributions and reaction rate ratios. The experimental methods will mainly be those which have been developed for integral measurements in zero-power fission reactors. First estimations show that adequate flux levels in the assemblies can be achieved with a proton beam current of 1 to $10\,\mu A$, depending on the multiplication factor of the assemblies (cf. Fig. 3), and that there will be no difficult cooling and activation problems. The possibility of setting up these experiments at a separate, dedicated beam line of the PSI ring accelerator is now being investigated more thoroughly.

Acknowledgements

This overview is based on two PSI contributions to a recent specialist's meeting (OECD-NEA Specialist's Meeting on Accelerator-Based Transmutation, PSI Würenlingen/Villigen, 24–26 March 1992) describing toxicity reduction studies for transmutation systems using high-energy spallation reactions [2] and planned experiments in support of the validation of nuclear data and calculational methods [4]. The experiments are carried out in the framework of the Transmutation Physics Project of the Laboratory for Reactor Physics and Systems Technology. This project receives valuable support from the actinide chemistry and irradiation technology groups within the Laboratory for Materials Technology and Nuclear Processes, and from the SINQ and Pirex Projects in the PSI Departments F3A and F3B.

References

[1] WYDLER P., "Reduction of the long-term toxicity of neptunium and other minor actinides by nuclear spallation", Proc. Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation, Mito City, Japan, 6–8 November 1990.

[2] WENGER H. U., WYDLER P. AND ATCHISON F., "The influence of different recycling schemes on toxicity reduction for transmutation systems using high-energy spallation reactions", Proc. OECD-NEA Specialist's Meeting on Accelerator-Based Transmutation, PSI Würenlingen/Villigen, 24–26 March 1992.

[3] ATCHISON F., "Data and methods for the design of accelerator-based transmutation systems (overview paper)", Proc. OECD-NEA Specialist's Meeting on Accelerator-Based Transmutation, PSI Würenlingen/Villigen, 24–26 March 1992.

[4] WYDLER P., "Research activities related to accelerator-based transmutation at PSI", Proc. OECD-NEA Specialist's Meeting on Accelerator-Based Transmutation, PSI Würenlingen/Villigen, 24–26 March 1992.

[5] ATCHISON F., "Spallation and Fission in Heavy Metal Nuclei under Medium Energy Proton Bombardment", Meeting on Targets for Neutron Beam Spallation Sources, KFA-Jülich, FRG, 11–12 June 1979, Jül-conf-34 (Jan. 1980).

[6] MARMY P., DAUM M., GAVILLET D., GREEN S., GREEN W. V., HEGEDÜS F., PROENNECKE S., ROHRER U., STIEFEL U. AND VICTORIA M., "PIREX II – A New Irradiation Facility for Testing Fusion First Wall Materials", Nuclear Instrumentation Methods in Physics Research **B47** (1990) 37–47.

[7] HEGEDÜS F., WINKLER P., WOBRAUSCHEK P. AND STRELI CH., "TXRF Spectrometer for Trace Element Detection", Advances in X-Ray Analysis **33** (1990) 581–583.

[8] HEGEDÜS F., WOBRAUSCHEK P., SOMMER W. F., RYON R. W., STRELI CH., WINKLER P., FERGUSON P., KREGSAMER P., RIEDER R., VICTORIA M. AND HORSEWELL A., "Total Reflection X-Ray Fluorescence Spectrometry of Metal Samples using Synchrotron Radiation at SSRL", European Conference on Energy Dispersive X-Ray Spectrometry, Mykonos, Greece, 30 May – 6 June 1992.