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RESEARCH ACTIVITIES RELATED TO ACCELERATOR-BASED TRANSMUTATION AT PSI

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ABSTRACT

The Paul Scherrer Institute (PSI) has experience with experimental reactor physics, accelerator-based experimental physics and the development of a spallation neutron source which can directly be applied to problems in the field of accelerator-based transmutation. Building on this experience and an experimental infrastructure which can easily be adapted to the respective needs, PSI 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.

In a first phase of this programme, thin samples of actinides will be irradiated with 590 MeV protons from the PSI ring accelerator 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 and to confirm the high potential of the high-energy fission reactions for transmutation.

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 PSI ring accelerator. The experiments are intended to provide integral checks on the nuclear data (models) and calculational methods. The experimental methods will mainly be those which have been developed for integral measurements in zero-power fission reactors. A preliminary study confirmed the feasibility of these experiments.

1. INTRODUCTION

In Switzerland, transmutation is presently not considered to be a realistic option for simplifying the country's radioactive waste management problems. As a result of the acceptance of a 10–year nuclear moratorium by the Swiss voters in 1990, 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 (NAGRA) 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, the Swiss Government has explicitly declared its will to keep the nuclear option open. Therefore, in view of the long–term potential of nuclear fission energy, there remains an incentive for investigating advanced reprocessing and waste management schemes. Under boundary conditions as described above, the respective research is best carried out by national research centres.

In Switzerland, the Paul Scherrer Institute (PSI) has experience with experimental reactor physics, accelerator-based experimental physics and the development of the SINQ spallation neutron source which can directly be applied to problems in the field of accelerator-based transmutation. Research in this field is attractive for PSI, since it meets the criteria for medium and long–tam research with high scientific content.

Since there is no Swiss concept for a transmutation system, PSI plans to focus its 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 stations with the necessary handling equipment, and a hot laboratory in which irradiated samples can be analysed using advanced examination methods.

Building on this experience and experimental infrastructure, PSI 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). This paper briefly describes the first two phases of this programme.

2. MOTIVATION FOR BASIC PHYSICS-ORIENTATED STUDIES

Actinide transmutation systems must be designed primarily to fission the actinides. Depending on the concept, this is achieved in different ways: for instance, in systems based on strong thermal neutron sources, 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, in fast reactors and accelerator–driven subcritical fast reactors the actinides are mainly fissioned by fast neutrons, and in molten-salt target systems actinides are fissioned directly by high energy spallation reactions.

In another paper for this meeting [1] it is shown that the fission probability in spallation reactions is much higher than that in fast reactors ($p_{fiss} \sim 0.8$ for 1 GeV protons impinging on a thick actinide target). There is thus an incentive for designing accelerator–driven subcritical fast reactor systems such as to enhance spallation reactions in the fuel. For spallation–based thermal neutron source systems, the use of actinides in the target is currently not considered, but spallation reactions with actinides due to high-energy particles leaking into the blanket region may have to be taken into account as "background" reactions in the analyses of the transmutation performance of these systems.

It therefore appears that, 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. 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.

Simple code comparisons for the irradiation of thin samples of actinides with high–energy protons have revealed considerable differences in the total yield and 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, an experimental verification is desirable, especially for actinides where the experimental data are scarce.

3. IRRADIATION OF THIN SAMPLES OF ACTINIDES

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-I, in* which thin samples of actinides will be irradiated with 590 MeV protons from the PSI ring accelerator, has been initiated. 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 Th-232, U–238, Np-237, Am-241 and possibly other minor actinides. U-238 will be used in the. first experiments because it is the easiest to handle. Th-232 is interesting from a theoretical viewpoint, because in the RAL fission model [2] its atomic number is close to the lower limit (Z = 89) of the region in which the systematic of Vandenbosh and Huizenga for the ratio of fission to neutron emission is applied.

Samples with a thickness of about 1 g/cm^2 will be irradiated in an existing facility, PIREX, which is designed for simulating displacement damage and impurity production in first wall materials of fusion reactors [3]. 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 to the hot laboratory in a shielded flask, 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 multielement and isotopic analyses to be made in 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 [4] has originally been 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 favorable conditions, at the Stanford Synchrotrons Radiation Laboratory (SSRL) using an electron storage ring to produce extremely intense X-ray beams [5].

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 the 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.

For accelerator–driven subcritical fast reactor 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 μA, depending on the multiplication factor of the assemblies, 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.

5. REFERENCES

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