ACTIVITIES RELATED TO ACCELERATOR BASED TRANSMUTATION AT PSI

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1. General Background

In Switzerland, the radioactive waste generated by nuclear power reactors is limited (3 GWe installed capacity, no new projects currently in sight), and the official policy is to store this waste in repositories in suitable geological formations. The associated risk for the population is considered to be negligible. An interest in advanced technologies for reducing nuclear waste does exist, however, at the level of basic R&D. With its experience in reactor and accelerator-based physics, including the development of the SINQ spallation neutron source, the Paul Scherrer Institute (PSI) is in a good position to perform physics studies related to accelerator-based transmutation systems.

Since accelerator-based reactor systems are technically more complex and tend to be less economic than normal fission reactors, it is reasonable to design accelerator-based actinide transmutation systems specifically with the object of fissioning the even-neutron minor actinides (^{237}Np, ^{241}Am, etc.) which, due to the threshold in their fission cross section, cannot effectively be transmuted in normal reactors. This thinking leads to accelerator-based transmutation concepts based on fast neutrons. The PSI activities in the field of accelerator-based transmutation are aimed at establishing the scientific basis for assessing the effectiveness of such concepts and resolving related "data and methods" problems.

2. Potential of Fast-Neutron Based Systems

It is obvious that the transmutation effectiveness for actinides is related to the fission-to-capture ratio of the nuclides and, for even–neutron nuclides, therefore significantly increases with neutron energy. Accelerator–based systems offer the possibility of "hardening" the neutron spectrum beyond the limits of normal fission reactors (possibility of using pure minor actinide fuels, spectrum hardening due to evaporation neutrons which have a higher energy than the fission neutrons) and using the high-energy reactions themselves to fission actinides. Concepts with TRU targets incorporate all of these features and therefore appear to be particularly attractive. Examples are the Phoenix concept [1] and the molten salt concept proposed by JAERI [2].

[1] On attachment from CEA, CEN Cadarache, F-13108 St–Paul–lez–Durance

Fission–to-capture ratios for different systems, calculated using a scheme which incorporates the PSI version of the high-energy code HETC and JEF-2.2 data for the neutron transport below 15 MeV, are shown in Table 1. The "D_2O cell" values correspond to a well moderated thermal neutron spectrum, typical for the D_2O moderator of a continous spallation source, and the "FBR" values correspond to a Superphénix type spectrum. Very favorable fission-to-capture ratios are obtained for the Phoenix reference case, and these can be further improved by replacing the minor actinide oxide by metal fuel.

Another important aspect is the overall neutron balance of a system. For a closed, long–term system this should be such as to allow the complete conversion of the actinides to fission products. Salvatores et al. [3] have proposed to measure the overall neutron balance in terms of the "fuel neutron production" parameter, here denoted by p. Unlike other neutron balance parameters, p depends on the ratio of neutron induced reactions (fissions, captures, n,2n reactions) to radioactive decays and therefore on the neutron flux.

p values for different systems are shown in Table 2. For the "problem nuclides" (e.g. ^{237}Np), the overall neutron balance in an LWR is negative, indicating that the chain of successive transmutations does not provide enough neutrons to support itself. In thermal systems with a very high flux p becomes positive, but remains small compared with p values in fast systems. Again, the most favorable results are obtained for the Phoenix system (it should be noted that the tabulated values contain the evaporation source effect and the k_{eff} effect of the target, but do not account for the "external" neutron multiplication, i.e. the multiplication due to the spallation reactions).

3. Detailed Analyses of Concepts with TRU Targets

In view of their promising basic characteristics, the Phoenix system and the molten salt system of JAERI were analysed in detail (flux distribution, bumup, mass flow, toxicity reduction) using two-dimensional models of the targets. The analyses showed that, as regards toxicity reduction, the Phoenix reference system does not ***meet the expectations,*** if reprocessing losses of 1 % are conservatively assumed. A particularly poor effectiveness results, if the generated plutonium (mainly useless ^{238}Pu) is added to the waste. With the aim of remedying these deficiencies, an alternative fuel cycle, illustrated in Fig. 1, is being investigated. This fuel cycle uses metal fuel with an IFR type reprocessing and incorporates full TRU recycling.

Mass changes for the investigated systems are given in Table 3. The tabulated values apply for an equilibrium cycle and indicate that all these systems could support a large number of light water reactors (the ^{237}Np production of a 1 GWe LWR is about 13 kg per year).

4. Sensitivity of Basic **Parameters to Data and Methods**

Tables 4 and 5 give fission-to-capture ratios and p values for different modelling assumptions and basic data sets. The comparison is carried out for the Phoenix reference system, i.e. the system with oxide fuel. "Critical" means that the calculation was carried out for a critical target, neglecting any source effect. The subcritical case simulates a fission neutron driven subcritical target with the same k_{eff} as that of the reference case, and "nofis" means that fissions in the

fission model of HETC are "switched off". It can be seen that the error introduced by the use of a fission neutron source rather than an evaporation neutron source is small but not negligible and that, for neutronic calculations, the use of a high-energy fission model is not essential. The differences arising from the use of different basic data sets are quite as significant as the effects of using different source approximations and hence also deserve attention.

5. Irradiation of Thin Samples of Actinides

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 [4]. In the context of transmutation, a correct prediction of the neutron source strength and power distribution in the target is not the only goal. 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, experimental verification is desirable, especially for actinides for which the experimental data are scarce (a recently completed international code comparison for intermediate energy nuclear data [5], in which PSI participated, did not address actinides).

To check the respective models, a basic validation experiment, *ATHENA*[2], in which thin samples of actinides are irradiated with 590 MeV protons from the PSI ring accelerator, is underway. Design and safety considerations for this experiment are summarised in Ref. 6. A uranium sample consisting of 258 mg of UO_2, encapsulated in a stainless steel tube with an inner diameter of 2.5 mm, has been irradiated using a relatively low beam current, and y-spectroscopy has been performed to confirm that the proton beam can properly be focused on the sample (cf. Fig. 2). First analyses have given promising results for ratios of calculated-to-measured activities. In the near future it is planned to irradiate a uranium and a thorium sample using a higher proton beam current. The utilised irradiation facility, Pirex, can handle currents up to 20 μA, which should make it possible to apply alternative measurement techniques such as inductively coupled plasma (ICP) mass spectrometry and total reflection X-ray fluorescence (TXRF) spectroscopy.

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[2] *ATHENA* = actinide transmutation using high energy accelerators

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Table 3: Mass Changes in kg/year for Different Systems

Nuclides	PHOENIX REF. 450 Mwth (per module)	PHOENIX METAL 450 Mwth (per module)	MOLTEN SALT 800 MWth
^{234}U	-0.44	1.03	2.60
^{237}Np	-115	-66.1	-102
^{238}Np	0.32	0.15	-0.01
^{238}Pu	90.7	13.0	-2.19
^{239}Pu	1.03	1.91	2.50
^{240}Pu	3.68	0.47	5.35
^{241}Pu	0.05	0.23	0.78
^{242}Pu	13.3	4.40	1.52
^{241}Am	-143	-82.7	-47.8
^{242}Am	0.13	0.06	-0.01
^{242m}Am	3.67	2.01	-0.39
^{243}Am	-22.6	-12.6	-21.6
^{242}Cm	22.3	10.9	-0.15
^{243}Cm	0.09	0.03	-0.03
^{244}Cm	+ 4.17	+ 1.26	-9.05
^{245}Cm	0.75	0.30	0.07
k_{boc}	0.844	0.811	0.925
Ak	0.011	-0.013	–
LWR Support Ratio	8.5	5.0	7.0

Table 1: Fission-to-Capture Ratios for TRU Nuclides with $T_{1/2} > 10$ a

	D_2O CELL	LWR (KKG)	FBR (SPX)	MOLTEN SALT	PH(REF.	ENIX METAL
^{237}Np	0.00	0.02	0.23	0.45	0.94	1.10
^{238}Pu	0.03	0.09	2.16	3.39	5.94	6.94
^{239}Pu	2.27	1.76	3.69	5.27	8.98	10.26
^{240}Pu	0.00	0.00	0.74	1.38	2.88	3.35
^{242}Pu	0.00	0.02	0.61	1.26	2.68	3.19
^{244}Pu	0.02	0.16	1.10	2.60	8.44	10.42
^{241}Am	0.01	0.01	0.15	0.30	0.57	0.63
^{242m}Am	4.88	4.93	6.55	7.03	7.83	8.22
^{243}Am	0.00	0.01	0.14	0.29	0.59	0.66
^{243}Cm	4.99	5.88	7.42	9.97	24.44	33.46
^{244}Cm	0.04	0.06	0.84	1.51	2.82	3.22
^{245}Cm	6.58	6.87	6.24	7.79	12.96	14.96
^{246}Cm	0.07	0.22	1.27	2.69	5.77	6.41
^{247}Cm	1.47	1.56	6.40	9.01	13.60	15.56
^{248}Cm	0.05	0.12	1.40	2.74	6.10	6.98

Table 2: 'Fuel Neutron Production' for TRU Nuclides with $T_{1/2} > 10$ a

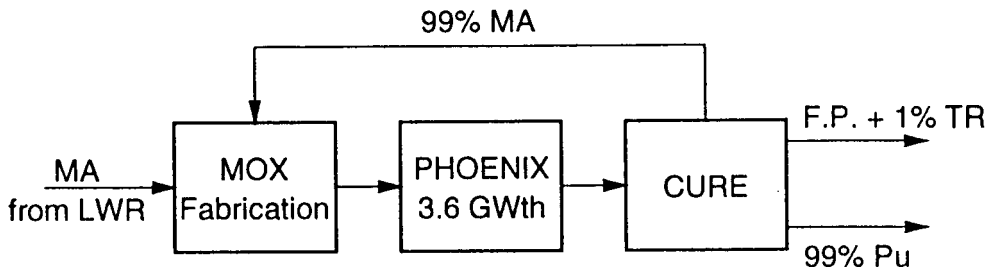
Flux	D_2O CELL 1E16	LWR (KKG) 1E14	FBR (SPX) 1E15	MOLTEN SALT 1E15	PH(REF. 1E15	ENIX METAL 1E15
^{237}Np	0.20	-1.05	0.67	1.03	1.32	1.43
^{238}Pu	0.07	-0.10	1.41	1.65	1.83	1.88
^{239}Pu	1,01	0.72	1.53	1.74	1.89	1.93
^{240}Pu	0.04	-0.30	1.00	1.41	1.73	1.82
^{242}Pu	-0.56	-1.16	0.60	1.27	1.70	1.83
^{244}Pu	1.38	1.55	1.94	2.14	2.25	2.26
^{241}Am	-0.43	-0.94	0.68	1.13	1.52	1.68
^{242m}Am	1.73	1,63	1.89	2.00	2.10	2.15
^{243}Am	0.39	-0.22	0.71	1.19	1.59	1.75
^{243}Cm	2.06	1.90	2.12	2.23	2.34	2.38
^{244}Cm	1.39	0.76	1.47	1.80	2.06	2.13
^{245}Cm	2.36	2.43	2.63	2.76	2.90	2.95
^{246}Cm	0.33	0.75	2.23	2.58	2.76	2.79
^{247}Cm	1.18	1.31	2.41	2.59	2.70	2.74
^{248}Cm	0.11	0.31	1.68	2.18	2.55	2.64

Table 4: PHOENIX System: Sensitivity of Fission-to-Capture Ratios for TRU Nuclides with $T_{1/2} > 10$ a

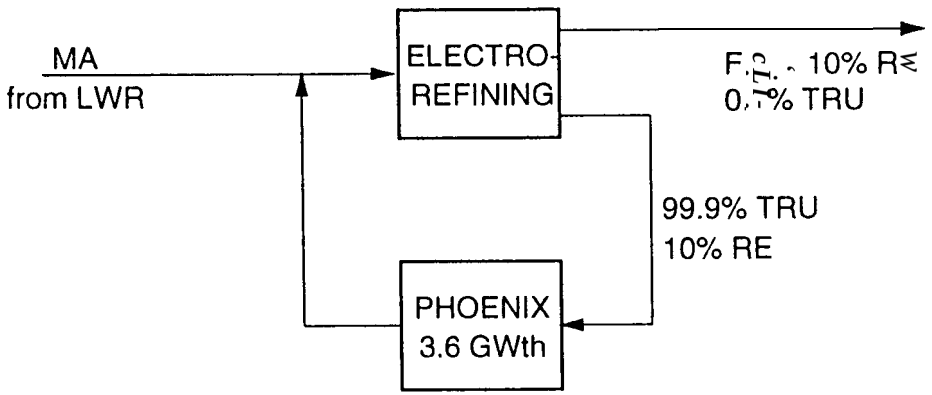
	CRIT.	k=0.8	NOFIS.	REF.	ENDFB-6	JENDL-3
^{237}Np	0.61	0.88	0.92	0.94	1.00	0.98
^{238}Pu	4.46	5.71	5.88	5.94	6.14	5.97
^{239}Pu	7.32	8.74	8.91	8.98	9.16	9.02
^{240}Pu	1.95	2.71	2.84	2.88	3.02	2.91
^{242}Pu	1.76	2.51	2.64	2.68	2.82	2.72
^{244}Pu	5.26	7.87	8.29	8.44	8.89	8.58
^{241}Am	0.37	0.53	0.56	0.57	0.61	0.58
^{242m}Am	7.00	7.67	7.79	7.83	7.98	7.85
^{243}Am	0.37	0.54	0.58	0.59	0.62	0.60
^{243}Cm	19.19	23.99	24.28	24.44	24.74	24.75
^{244}Cm	1.90	2.65	2.78	2.82	2.97	2.86
^{245}Cm	10.87	12.68	12.88	12.96	13.18	13.06
^{246}Cm	3.76	5.35	5.67	5.77	6.10	5.88
^{247}Cm	11.08	13.26	13.51	13.60	13.89	13.64
^{248}Cm	3.99	5.66	6.00	6.10	6.41	6.19

Table 5: PHOENIX System: Sensitivity y of 'Fuel Neutron Production' for TRU Nuclides with $T_{1/2} > 10$ a

Flux	CNT. 1E15	k=0.8 1E15	NOFIS. 1E15	REF. 1E15	ENDFB-6 1E15	JENDL-3 1E15
^{237}Np	1.12	1.27	1.31	1.32	1.39	1.32
^{238}Pu	1.71	1.79	1.82	1.83	1.84	1.83
^{239}Pu	1.81	1.86	1.89	1.89	1.90	1.89
^{240}Pu	1.52	1.67	1.72	1.73	1.75	1.73
^{242}Pu	1.42	1.63	1.69	1.70	1.73	1.71
^{244}Pu	2.16	2.20	2.24	2.25	2.26	2.25
^{241}Am	1,24	1.45	1.51	1.52	1,56	1.54
^{242m}Am	2.01	2.08	2,10	2.10	2.11	2.11
^{243}Am	1.29	1.51	1.58	1.59	1.63	1.61
^{243}Cm	2.28	2.32	2.34	2.34	2.35	2.35
^{244}Cm	1.87	2.00	2.05	2.06	2.08	2.06
^{245}Cm	2.82	2.87	2.89	2.90	2.91	2.90
^{246}Cm	2.60	2.69	2.75	2.76	2.77	2.77
^{247}Cm	2.61	2.67	2.70	2.70	2.71	2.71
^{248}Cm	2.31	2.47	2.54	2.55	2.57	2.56



MA Oxide Fuel (Reference Case)



MA Metal Fuel

Fig. 1 : Fuel Cycles for PHOENIX System

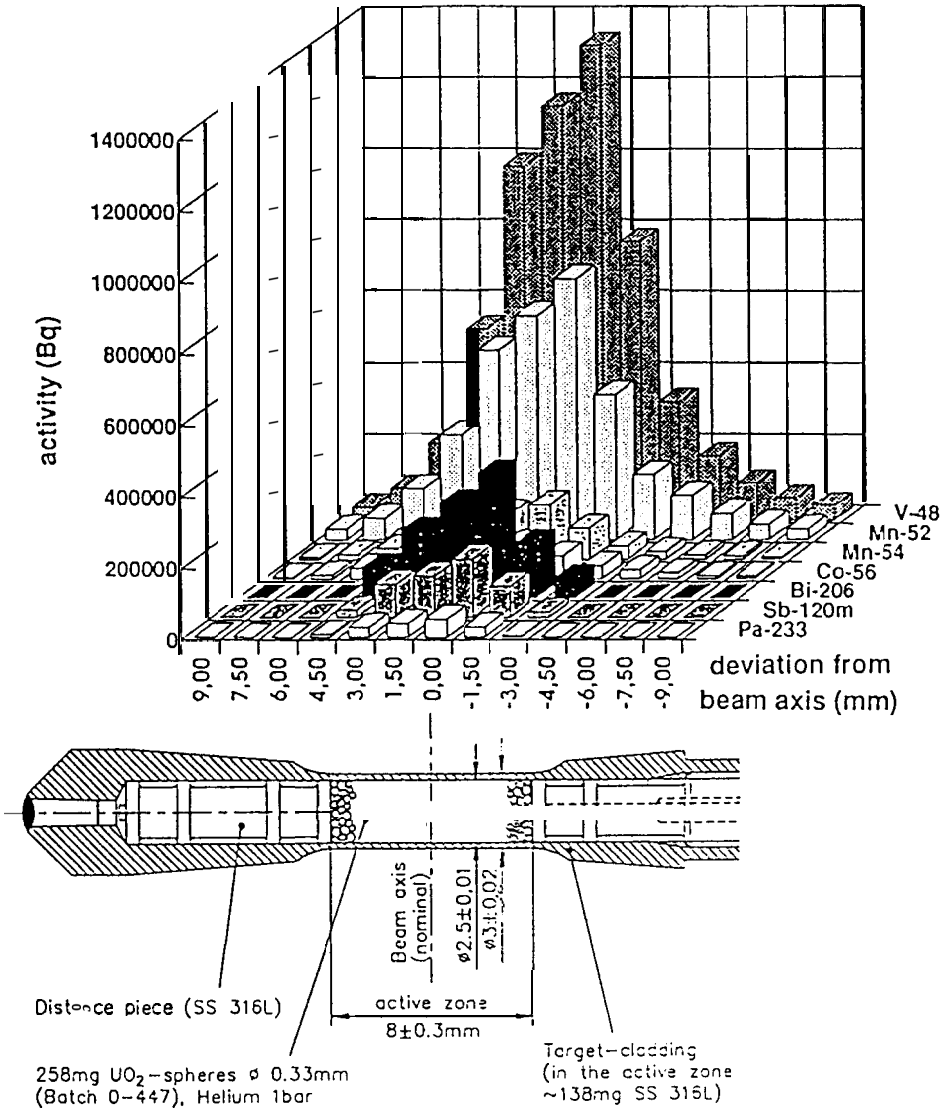


Fig. 2: γ–Activity Distributions for the Irradiated *ATHENA* Target–1