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SPECIFIC CONTRIBUTIONS OF THE DUTCH PROGRAMME "RAS" TOWARDS ACCELERATOR-BASED TRANSMUTATION

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SPECIFIC CONTRIBUTIONS of THE DUTCH PROGRAMME "RAS" TOWARDS ACCELERATOR-BASED TRANSMUTATION

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Abstract. Accelerator-based transmutation is being studied by ECN within its general nuclear waste transmutation programme RAS. In this paper the following contributions are presented:
1) Evaluation of cross sections at intermediate energies, within an international frame given by NEA,
2) Cell calculations on the equilibration of transuranium actinides in thermal molten-salt transmuters,
3) Irradiation facilities at the European research reactor HFR in Petten, which have been constructed with the purpose to demonstrate and investigate the transmutation of waste in a high neutron flux,
4) Studies of accelerator-based neutron generating systems to transmute neptunium and technetium,
5) Comparison of several systems on the basis of criteria for successful nuclear waste-management.

THE DUTCH PROGRAMME RAS

In contrast to an OECD average of 25 %, The Netherlands produces only 5% of its required electricity by means of nuclear power. Exploitation of the apparent growth potential would require a broad acceptance of an appropriate waste management strategy. Deep geological disposal of the high level waste is considered, but due to the long half-life of some of the radionuclides, integrity of the isolation-barriers would have to be guaranteed for a period of at least 100.000 years. In order to study complementary options a research programme (RAS) on the recycling actinides and on the transmutation of long-lived fission products has been defined by ECN. One of the aims of this programme is the compilation and evaluation of various schemes proposed for reducing environmental risks. Within this RAS programme there is a small but significant study on the subject of accelerator-based transmutation and on its competing demands of safety, economy, and aspects of proliferation. Ref. [1] considers evolutionary scenarios on low-waste producing systems, the thorium/uranium cycle, and end-scenarios in which very large amounts of actinide waste are offered. The present paper treats more specific contributions:

Cross Sections at Intermediate Energies

On behalf of the design of systems for accelerator-based transmutation, contributions are given in the form of evaluations of cross sections at intermediate energies. Lead-208 is taken as a target material for a "sample evaluation" (ref. [2] and table 1), which fits as a benchmark of evaluation methods in an international frame given by NEA (Working Party on Evaluation Coordination of the Nuclear Science Committee). In earlier work [3] the most urgent data needs have been prioritized and translated in terms of intermediate energy data libraries. This work is a contribution to the subgroup on Intermediate Energy Nuclear Data Evaluation of the above mentioned Working Party. In Fig. 1 the complicated set of relevant reactions are indicated. Some studies on nuclear data needs for ATW [4, 5, 6] have revealed that single- and double-differential neutron and proton cross sections of proton- and neutron- induced reactions up to 1500 MeV have a very high priority. Also important are total, elastic, gamma production, fission and activation/transmutation cross sections. Most relevant nuclides are actinides, target materials (Ta, W, Pb and Bi), and materials in structures, shielding, and coolants (H, C, N, O, Na, Mg, Al, Ar, K, Ca, Cr, Mn, Fe, Co, Ni, Zn, Zr). Extensions of the numerical data base EXFOR with high-priority intermediate-energy data are made and evaluation programmes on intermediate-energy nuclear data are being defined in the framework of the above mentioned NEA activity. In addition ECN evaluates the European Activation file (EAF), which consists of a huge set of activation and transmutation cross sections, so far up to 20 MeV [7].

MF	MT	Description
1	451	General information
3	2	Elastic cross section
3	5	(p,anything) cross section
6	2	Elastic angular distribution
6	5	(p,anything) yields and energy-angle distributions

Table 1: Directory of proton data up to 100 MeV for Pb-208 (ENDF-6 Format).

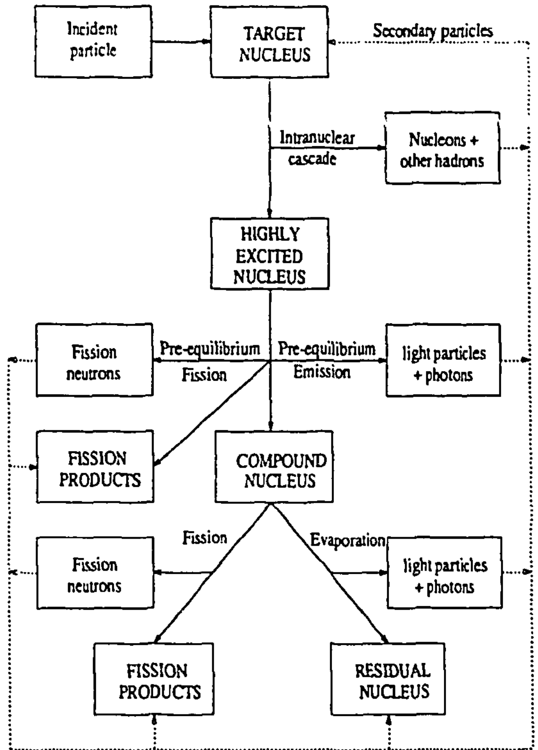


Fig.1: Intermediate-energy nuclear reactions. Solid arrows refer to an intranuclear cascade process (thin target and single proton nucleus reaction), whereas dotted arrows represent an internuclear cascade process (thick target and multiple transport processes).

Equilibration of actinides in a molten salt reactor

There are many advantages of actinide transmutation in a thermal reactor with molten salt as fuel: it allows for on-line reprocessing of the molten salt, continuous and easy refuelling, and for small neutron loss in fission products. Calculations show [19] that due to reactivity feedback, the operation of critical molten salt transmuters fuelled with transuranic LWR mix, is possible in only a very limited range of its design parameters. This could make it necessary to operate the system sub-critically like an ATW. For the various levels of the neutron flux in such a molten-salt reactor, fig. 2 presents equilibrium masses of the actinides. Heavier actinides, being products of shorter-lived trans-uranium isotopes, will increase with the flux-level, whereas the plutonium isotopes decrease. As is discussed later, this causes a lack of delayed neutrons in the system.

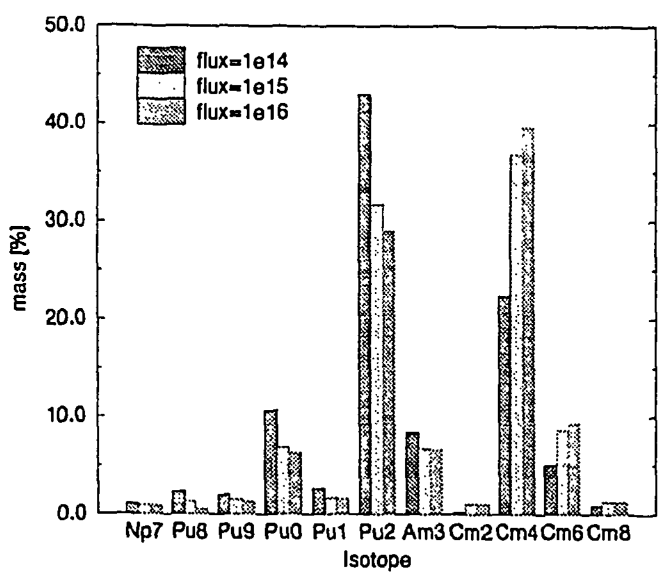


Fig. 2: Actinide masses in a molten salt transmuter.

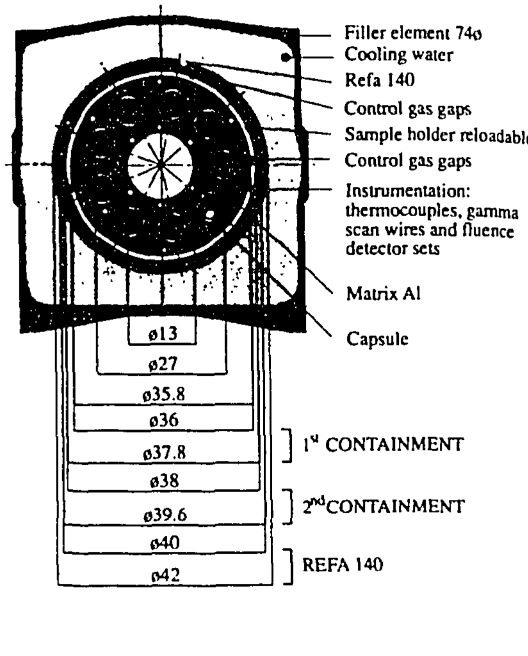
Demonstrations and Experimental Work

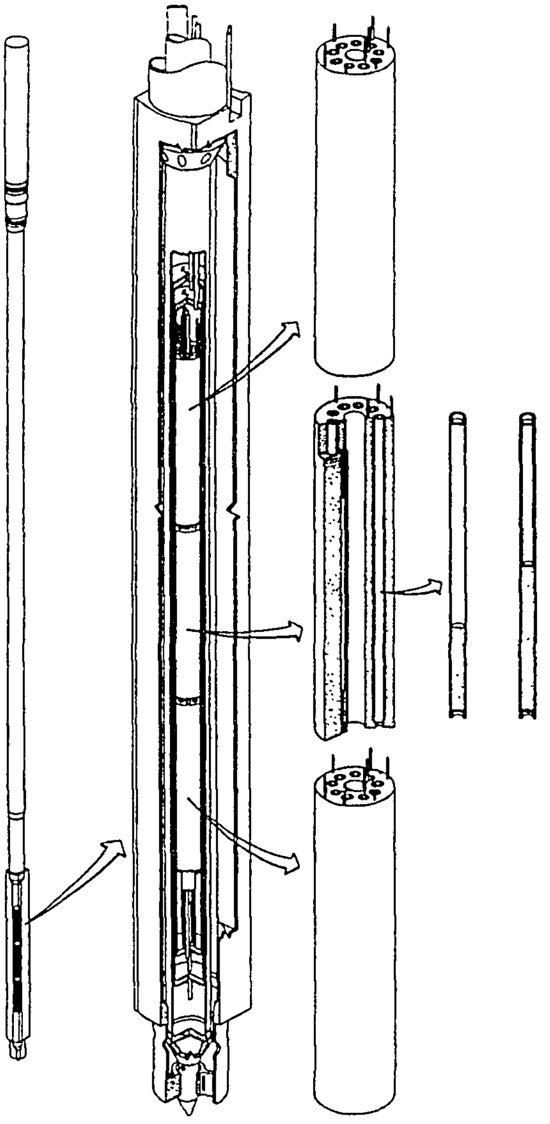
For experimental and technological research on transmutation a European network EFTTRA (Experimental Feasibility of Targets for TRAnsmutation) has been founded. In this network ECN participates together with the French organisations CEA and EdF, the German KfK and the EC-establishment TUI. Targets are prepared in the laboratories of TUI, CEA and ECN, irradiations are being performed in the Petten HFR by ECN (see fig. 3 and ref. [8] for a description of the setup), and will later also be performed in the French Phénix reactor. After finishing the irradiations, targets will be examined to study transmutation efficiency and material aspects. The EFTTRA programme is focused on irradiation of the actinides (especially the americium isotopes), but also the fission products iodine and technetium are being studied. Chemical and material properties of candidate target materials will be studied before and after irradiation of about one year.

From the shielded one-group cross section of the fission products I-129 and Tc-99 (about 12 and 7 b, respectively), one might conclude that in a high flux research reactor such as the Petten HFR, about 50% transmutation is feasible within about three years [9]. This 45 MW(th) reactor is excellent for R&D on high-flux (10^{14}-10^{15}) systems, but it is not designed to cope with the huge amount of fission product waste from power reactors. Drastic changes in reactor parameters would be needed for transmutation of such amounts.

Fig. 3a: The facility (shown on the right) consists of a thimble, which forms an irradiation chamber for specific instrument-insets. Primary reactor-cooling water is being used for cooling.

Fig. 3b: A sample holder (shown below) consists of ring-shaped aluminium blocks with peripheral holes for positioning test capsules. Stainless steel 100 mm long capsules are used, and targets are 50 mm long and have a diameter of 5 mm. The instrumentation consists of thermocouples and of neutron fluency detectors. The sample holders are surrounded by three stainless steel containments. Containments have gaps for inert gas (He or Ne) used for temperature control near about 450 °C.





A Study of the Stability of an ATW System

A study has been made [13] of the high-flux ATW system of ref. [12], which transmutes Np-237 and Tc-99. Although the Los Alamos group has recently proposed another ATW system [10], the first system had quite some interesting aspects due to the fact that it is non-linear. A surprising nature of some solutions of differential equations for flux ϕ, concentrations N, and rates of transmutation R, originates from the likelihood that in a very high flux Np-238 will fission before it decays with a half-life of about two days. This fission process will convert neptunium at high flux [14] from an absorber into a fuel. Because of the retarded build up and the small decay constant λ_8 of Np-238, instabilities will not have a run-away behaviour, but will rather be of a controllable and creeping kind (time constants are of the order of days). A feature related to this fuel/ absorber conversion is the occurrence of several branches in the flux-concentration diagram for the steady state (see fig. 4). If the fractional load N_7 of Np is far below a critical value of about $2 * 10^{-4}$, only the fixed amount N_{Tc} of technetium acts as an absorber of neutrons [12], and the neutron loss n is equal to the transmutation rate R_{Tc} of technetium, and the source strength is given by: $n = \phi * N_{Tc} * \sigma_{Tc}$. In approaching the above-mentioned critical value of the fractional neptunium load N_7, the neptunium starts to act as a fuel and the flux ϕ will slowly rise until the value of N_7 has decreased again. In an intermediary region (up to a second critical value of the fractional load of about $1.5 * 10^{-3}$) a "chaotic" behaviour of the system is found, where the flux will change unpredictably but slowly to either lower or higher values. For selected values of N_{Tc} a third region starts above a second critical point with a solution at high flux, for which the Np acts mostly as a fuel, and a solution at low flux, for which the Np acts more as an absorber. This "post-critical" region will be stable in its "absorber" branch into which the "fuel" branch will decay.

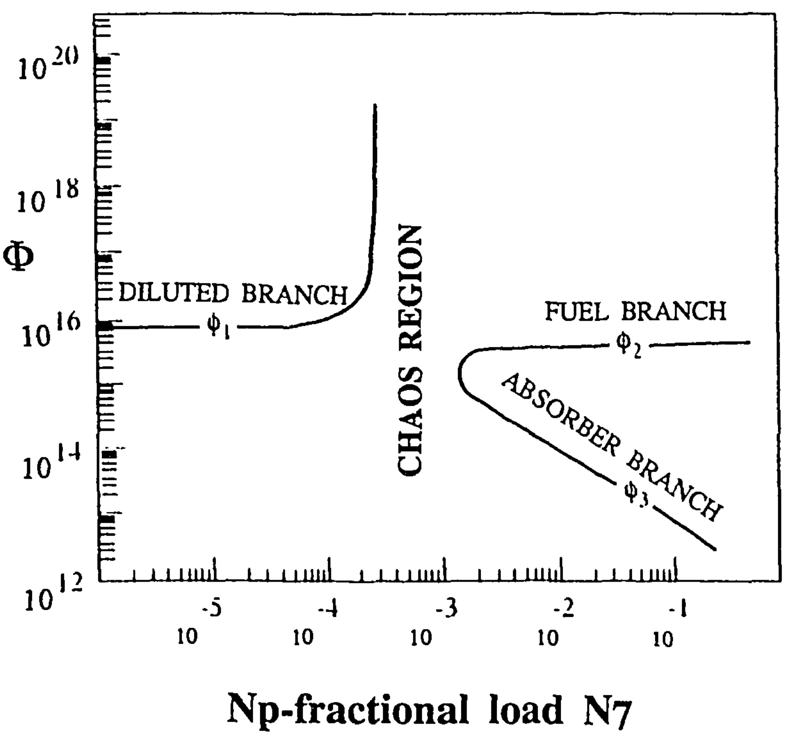


Fig. 4: Steady flux states in a Np-Tc transmuter [13]

Addendum to fig.4: Values for the parameters N_{Tc} and for n are from [12]. If the flux ϕ and fractional load N_7 of neptunium are constant, the rates of change R_7, R_8 of Np-237 and Np-238 will be connected by:

$R_7 = \lambda_8 * N_8 + R_8$,
$n = R_7 + R_8 * \{H\} + R_{Tc}$,
with $\{H\} = (1+\alpha-\nu)/(1+\alpha)$.

If N_7 approaches zero, then $R_8=0$, $n=R_{Tc}$ and:
$\phi_1 = n/(N_{Tc} * \sigma_{Tc})$.

If $N_7 >> (n,R_{Tc})$,
the asymptotic solutions are:
$\phi_2 = (\lambda_8/\sigma_8) * [-1/(1+\{H\}]$,
and $\phi_3 = n / (N_7 * \sigma_7)$.

COMPARISON OF ACCELERATOR-BASED SYSTEMS

Large energy flows should be dealt with, during any effective incineration of fissile material. For accelerator-based hybrids [10-12], the accelerator power P_A can be expressed as a function of the hybrid power P_R according to: $P_A = C * P_R * (1 - k_{eff}) / k_{eff}$. Here the hybrid power P_R depends on the accelerator power P_A, the reactor variable k_{eff} and on a conversion factor C; it can be deduced that this design parameter C lies between one and two for competing proposals. Economy of incineration might require an approach to $k_{eff}=1$ to allow for a reduction of the electric power P_A in the accelerator beam [17]. Approach towards a critical system will lead to a more homogeneous neutron flux and less flux-peaking in the window of the GeV proton beam. This window should be cooled by non-pressurized liquids like sodium or molten salt, as it is the weakest spot of the system.

Accelerator Transmutation of Waste (ATW) systems, in which a very high powered (100 MWe) high-energy proton beam plays a key role, were proposed by Los Alamos as special hybrid accelerator-reactor systems [10,12] that operate at $k_{eff} < 0.95$. If a very high flux and diluted fission material inventories are used, the transmutation rate will be very high. This actinide-transmuter can be equipped with a molten salt reactor [12], in which thorium fuel can be used as well. Because liquid reactor fuel (D_2O moderated slurries or molten salts) could lead to safety problems, an ATW system with solid fuel was proposed. In this proposal [10] the solid fuel is cooled with low cross-section liquid metals like Li-7. Further studies are needed to see how efficient this system can transmute fission products.

A fast reactor setup as a sub-critical booster has been proposed in Ispra, Brookhaven and JAERI. Relative to ATWs, these systems require only a small accelerator [15]. In fact these slightly sub-critical fast reactors are partly regulated with an accelerator beam, which also acts as an "electronic safety rod". Such fast reactor-boosters have very attractive features to fission minor-actinides and even-isotopes of plutonium. Because these systems have only a small surplus of thermal neutrons, transmutation of long-lived fission products would require application of moderated sub-assemblies. However combined transmutation of fission products and actinides simultaneously has not yet been foreseen for these systems.

A thermal Th/U breeder, with external neutrons has been proposed by CERN [11]; it will have intrinsic safety parameters, and some degree of protection against diversion of weapons-grade material. The fissile U-233 will always be diluted isotopically with less fissile U-234 (table 2 shows the fuel composition as derived from [11]). However there are two drawbacks, both of which are related to a non-compliance with the need to reduce future dose-risks: first of all the simultaneous transmutation of the long-lived fission products and actinides is not foreseen, and secondly the amount of U-234 in the waste might be rather high, which could lead to long-term risks due to radon-emanation [1].

Table 2 presents data relevant to safety and efficiency, and also compares the mass balance for some accelerator-based hybrids with an LWR (once-through). The delayed neutron fraction ν_d is an important safety parameter in reactor kinetics. For even-odd fissile nuclides it is fitted empirically by: $\nu_d = \exp\{16.46 - 0.5\ (3Z - A)\}$. For actinides with an even number of neutrons this value of ν_d should be divided by: $\{ 6.4 - (3Z - A)/8 \}$. Too small values of ν_d are found for thermal transmuters of transuranic waste, especially for high-flux thermal molten-salt systems (see earlier discussions on molten salt systems). Low values of ν_d could be compensated for by a reduction in k_{eff}, and by a corresponding increase in P_A; this however would not be economical nor be gentle towards the window.

Table 2: Actinide mass Balance EOC and delayed neutron fractions (both in percents)

		Energy producing systems		Waste burners	
	Type of transmuting system:	LWR (U/Pu) once through typical case	(Th/U) Energy-Amplifier	LWR waste molten-salt system	Metal fuelled system
	Delayed neutron fraction (ν_d in %)	k_{eff} =1	k_{eff} =0.92	k_{eff} =0.92	k_{eff} =0.89
		Efficiencies β *) of the four transmutation-systems are:			
		β = 1	β = 0.67	β = 0.67	β = 0.53
Nuclide:	Exp. [16] /Systematics	Mass balance EOC of the four transmuting systems			
Th-232	5.27(40) / 4.78	-	97.3	-	-
U-232	0.44(3) / 0.44	pm	pm	pm	pm
U-233	0.74(4) / 0.65	-	1.1	-	-
U-234	- / 0.93	$2*10^{-2}$	0.8	$2*10^{-2}$	$< 10^{-2}$
U-235	1.67(7) / 1.76	0.9	0.1	$5*10^{-2}$	-
U-236	- / 2.07	$2*10^{-2}$	0.6	-	$< 10^{-2}$
U-238	4.60(25) / 4.78	98.1	< 0.1	-	-
Np-237	1.07(10) / 0.93	$3*10^{-2}$	< 0.01	1	50
Pu-238	0.46(7) / 0.44	$2*10^{-2}$	< 0.01	2	10
Pu-239	0.65(5) / 0.65	0.5	< 0.01	2	11
Pu-240	0.90(9) / 0.93	0.2	< 0.01	8	10
Pu-241	1.57(15) / 1.76	0.1	< 0.01	3	2
Pu-242	1.86(9) / 2.07	$3*10^{-2}$	< 0.01	34	2
Am-241	0.44(5) / 0.44	$2*10^{-2}$	-	-	8
Am-242m	0.69(5) / 0.65	-	-	-	0.2
Am-243	- / 0.93	$1*10^{-2}$	-	7	4
Cm-242	- / 0.22	-	-	0.2	-
Cm-243	- / 0.24	-	-	$< 10^{-2}$	-
Cm-244	- / 0.44	$3*10^{-3}$	-	32	2
Cm-245	0.59(4) / 0.65	-	-	0.7	0.3
Cm-246	- / 0.93	-	-	8	-
Cm-248	- / 2.07	-	-	2	-
Cf-249	0.27(2) / 0.24	-	-	-	-

*) System-efficiency is defined by: $\beta = (P_R - 2P_A) / P_R = 1 - 2C (1 - k_{eff})/ k_{eff}$. For a critical reactor β is obviously equal to 1; for proposed energy producing systems the design parameter C turns out to be about 2, keff < 0.9, and β should be close to 2/3.

Systems which have been treated in this chapter were compared by means of earlier defined criteria [1]. From point of view of delayed neutrons LWRs and Energy-Amplifiers are stable and safe energy-producing systems, the latter being more secure with respect to safe-guarding than high-flux Th/U systems. The temperature- and voiding- behaviour should be evaluated for all systems. Metal fuel systems are most efficient in end-scenarios [18].

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