TRANSMUTATION OF NUCLEAR WASTES WITH GAS-COOLED PEBBLE-BED ADS

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Abstract

Transmutation of nuclear wastes is being explored for its application to waste management, a fundamental issue for nuclear industry. Several concepts are under consideration, mainly fast breeder reactors and accelerator driven systems (ADS). Inside this second category, we are analysing a helium-cooled graphite moderated sub-critical assembly, which uses as fuel units a small amount of transuranics diluted, in the form of TRISO coated particles, in graphite pebbles. This configuration (PBT) allows for neutron spectra that, taking advantage of the existence of huge capture resonances in the epithermal region, increase in a substantial factor the system transmutation efficiency.

Neutronic studies to determine transmutation performance and thermal behaviour are presented and discussed together with an analysis of the additional studies to address before going into detailed design activities.

1. Introduction

Accelerator driven transmutation is a promising method to alleviate the environmental impact associated with the final disposal of the spent nuclear fuel from Light Water Reactors (LWR). According to such method, nuclear cascades, initiated by spallation on heavy materials by medium energy (few hundred MeV) protons, are used in a sub-critical assembly to transmute the unwanted wastes into less harmful species.

Previous designs of these transmuters have been essentially derived from the Energy Amplifier concept [1] initially intended to produce energy by using the Thorium cycle. Here, we are instead focusing on a pure transmuter based on the use of the Adiabatic Resonance Crossing (ARC) method and specifically oriented to the most effective elimination of nuclear wastes, especially plutonium that is the most worrisome and abundant element in those wastes.

In this paper we are presenting the main results from the studies being carrying out by LAESA and collaborators on a Pebble Bed Transmuter (PBT) based on the above-mentioned philosophy. PBT is a gas-cooled sub-critical nuclear core filled with graphite-fuel pebbles and coupled to an accelerator. A small amount of transuranics is diluted inside the graphite pebbles in form of TRISO coated particles (a few grams of TRU in each 6-cm diameter pebble). The lethargy gain per scattering and the small TRU concentration make the neutron slowing-down to follow the ARC scheme. In addition, this system could take advantages of the technology already developed in the seventies for the High Temperature Gas Reactor (HTGR) in Germany and USA

2. Resonance enhanced transmutation

Nuclear waste transmutation reactions are based on neutrons as inducing particles. Neutron energy spectrum can be classified in three main types:

- Thermal spectrum, neutron energy in the range of 0.1-1 eV, depending on the moderator temperature.
- Fast spectrum, with energies in the order of 1-10 MeV.
- Iso-lethargic spectrum, intermediate to the previous ones with neutron energy over the nuclei resonance region.

ARC moderation is produced when high-energy neutrons are injected in a large, diffusive medium with negligible absorption and large elastic cross section values, such as lead or carbon. ARC moderation generates an iso-lethargic spectrum based on the tiny energy loss steps of the neutron in its way down to thermal energies. Hence, the neutron crosses in its moderation process all the energy range, having a great chance to find the cross-section resonances of the material in the medium. With regard to the elimination of actinides, this spectrum has the advantage, compared to the thermal case, of a bigger fission to capture ratio, while compared to the fast spectrum it is more efficient both for fission and capture. This method was tested and proved in the TARC experiment at CERN [2].

3. The pebble-bed transmuter

PBT, helium-cooled graphite moderated sub-critical assembly using as fuel small amounts of transuranics diluted in graphite pebbles, is a device optimised for nuclear waste transmutation, in particular TRU burning, although fission products elimination is also envisaged. Its main parameters are shown in Table 1. Figure 1 shows a schematic view of the system.

Figure 1. **PBT conceptual view**

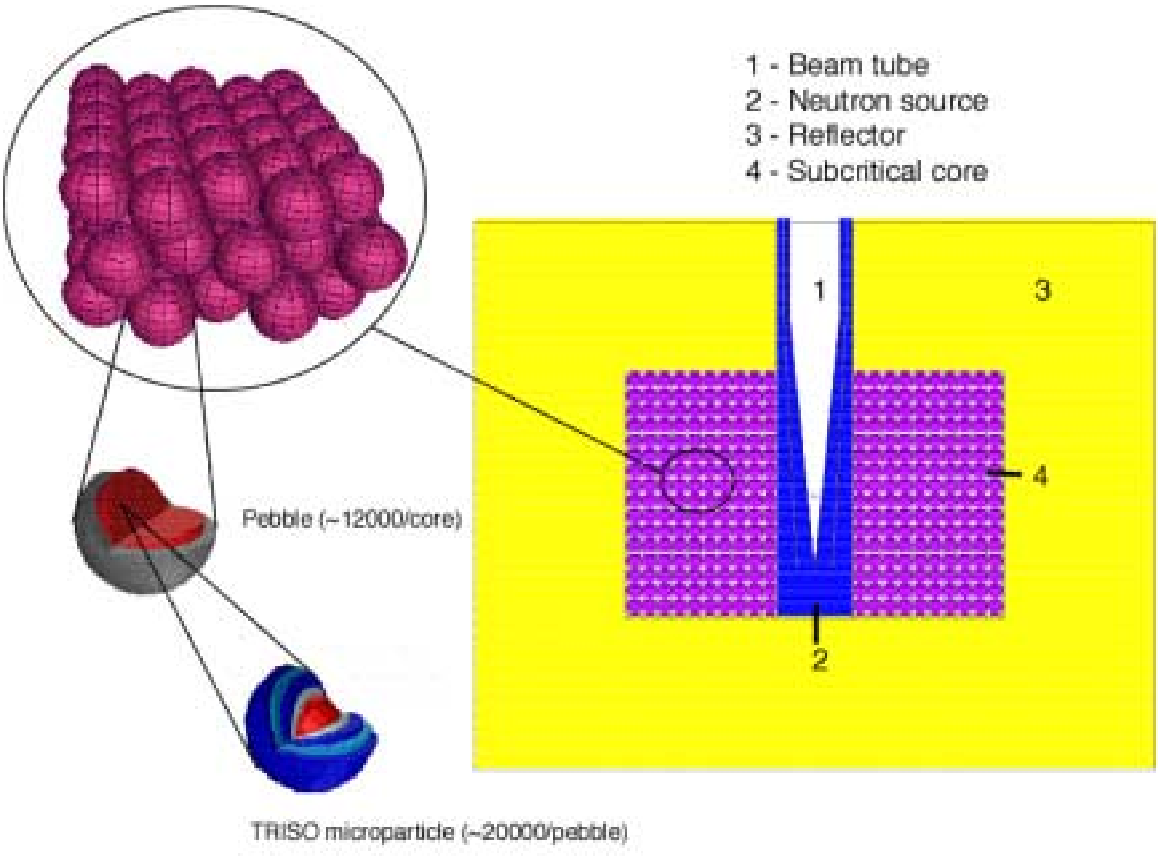


Table 1. **10 MW PBT parameters**

Parameter	Value	Unit
Thermal power	10	MW
Beam power	3.8	MW
Criticality constant	0.75	
Mean power density	4.7	W/g
Core mass	2.19	Tm
Initial TRU load	14.6	Kg
Fuel	Graphite pebbles + TRU	
Proton beam energy	380	MeV
Beam current	10	mA

Main PBT components are:

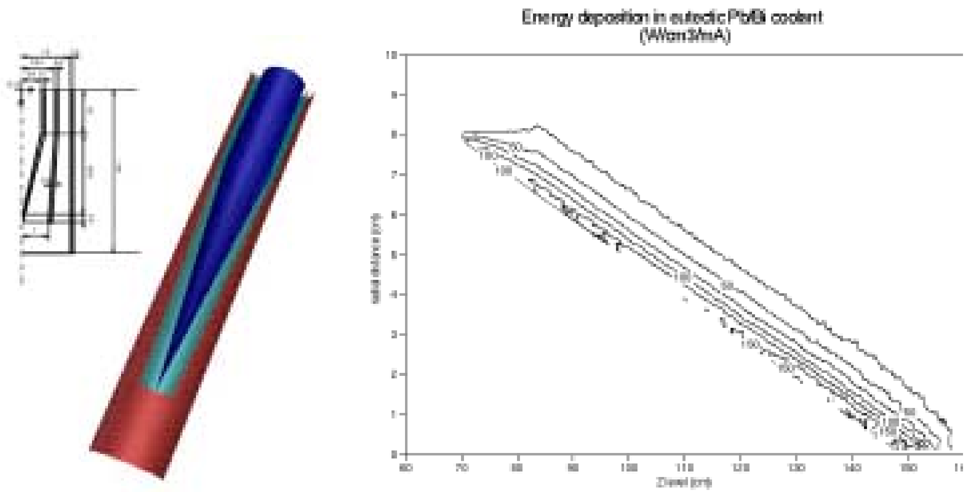
- The accelerator system, to provide a medium-energy high-intensity proton beam.
- The spallation target, which couples the accelerator proton beam to the nuclear system, providing the neutron source needed to sustain the sub-critical system.

- The sub-critical nuclear core, whose fuel is mainly composed of the offending materials to transmute.

Two cyclotrons in cascade compose the accelerator system proposed for the PBT. The main one is a booster cyclotron with six separated sectors and four accelerating cavities capable to reach energies up to 380 MeV and beam intensities in the order of 10 mA. Four RF cavities operating at a frequency of 70.4 MHz are used in order to get sufficient turn separation at the extraction radius. A small injector provides the necessary beam, with energy of 20 MeV, at the entrance of the booster. This injector is a superconducting 40 MeV H_2^+ cyclotron that generates the required 20 MeV protons by stripping phenomena.

The spallation target, based on liquid lead-bismuth, has a geometrical design (Figure 2) oriented to optimise two main features: a) broadening of the source, and b) strengthening of the separation window. The system proposed for the PBT is conical in shape, with a length of nearly 1 meter, which will extend the neutron generation almost all over the core axis. Another subject to be carefully considered is the energy deposition along the target. As a result of the relatively low proton energy (380 MeV), this deposition is very close to the solid window, increasing the difficulties in the design of the cooling system. The ionisation losses in the structural material of the window are also relevant in our case. This target is under study by a LAESA/CRS4 working group [3] to establish material working conditions and structural damage, including neutronics, thermal-hydraulics and radiological hazard.

Figure 2. **PBT spallation target: conceptual view and energy deposition calculated with FLUKA [4] code**



The core is a cylinder containing the fuel pebbles. The central part is occupied by the spallation target, which acts as neutron source. The core is filled with graphite pellets containing the nuclear fuel. The proposed fuel pellet that is proposed for the PBT is similar to the one that has been developed for the HTGR reactors. The fuel is confined in 3-cm-radius pebbles. The external layer of the pebble is made of pyrolytic graphite with a thickness of 5 mm, while the inner 2.5-cm-radius volume is filled with 1-mm-diameter TRISO micro-spheres containing the fuel material. The main advantages for these pebble bed cores are the possibility of continuous refuelling and that pebbles are very tight holders for fission fragments and produced radioactivity for temperatures up to almost 2 000ºC. The reflector is made of a 60 cm carbon wall.

4. Transmutation performance

Evaluation of the efficiency of PBT in transmuting the actinides present in the PWR waste has been addressed by simulating the neutronic behaviour of the device with Monte Carlo techniques. We have used in these simulations the codes LAHET [5], NJOY, MCNP-4B [6] and ORIGEN-2.1 [7].

In those simulations, the core is divided in 10 horizontal layers. We have not considered the materials homogeneously distributed inside the core. By the contrary, our simulations respect the real distribution of materials. Homogeneous distribution is only assumed inside the pebble (except the coating) by using the corresponding fractions of carbon, silicon and fuel. We consider that the micro-particles are imbedded in a carbon matrix, and that all the volume not occupied by fuel or silicon carbide is filled by carbon with a density of 1.7 g/cm^3. The fuel is TRU-oxide, with the composition given by the TRU from a PWR discharge after 10 years of cooling-down. This fuel would include not only Pu, but also Np, Am and Cm. Cross sections for the most relevant isotopes (Carbon in particular) have been processed taking into account thermal working conditions and resonance broadening.

Initially, every layer of the core is filled with the same fuel. Afterwards, we make a series of cycles: every 99 days we reload the upper level with fresh fuel, extract the lower one, and shift one position down the rest. In this scheme, the fuel is exposed to a total burn-up of 990 days between the insertion up to the extraction times. The data here summarised are referred to the system under equilibrium conditions, namely 990 days after the beginning of operation of the machine.

In the equilibrium state, the 10 MW PBT gives an average mean power density of 6 W/cm^3 in the core, or 8 MW/cm^2 on the surface of the balls. However in the upper level that power density is more than twice that value while in the lower one it is less than one third of it. Then, the cooling will be enhanced by a downward flow. The power release in the core has been used for the thermal analysis of the PBT.

Regarding transmutation performance of the system. Plutonium isotopes suffer a considerable mass reduction (Figure 3 and Table 2). In particular, the most abundant isotopes (^{239}Pu, ^{240}Pu and ^{241}Pu) are drastically reduced. ^{242}Pu and ^{238}Pu increase slightly their masses, but the radio-toxicity of ^{242}Pu is very small. The mass of ^{237}Np is also considerably reduced. ^{241}Am is eliminated while ^{243}Am mass increases. The quantity of curium also increases, but it remains much smaller than the initial mass of actinides.

Concerning the activity of the final waste stream in the long term (Figure 4), the creation of fission products implies an increment of the radioactivity during the first hundred years, but the smaller actinides mass produces a reduction in the long term. The minor actinides and their descendant (^{244}Cm, ^{243}Am) produce a very important part of the radioactivity. We are exploring possibilities to reduce these final minor actinides, either by reinsertion into the system waiting for a stabilisation of their masses (as proposed in [8]), or by using a system with multiple spallation targets.

Figure 3. **TRU elimination vs. irradiation time in the PBT**

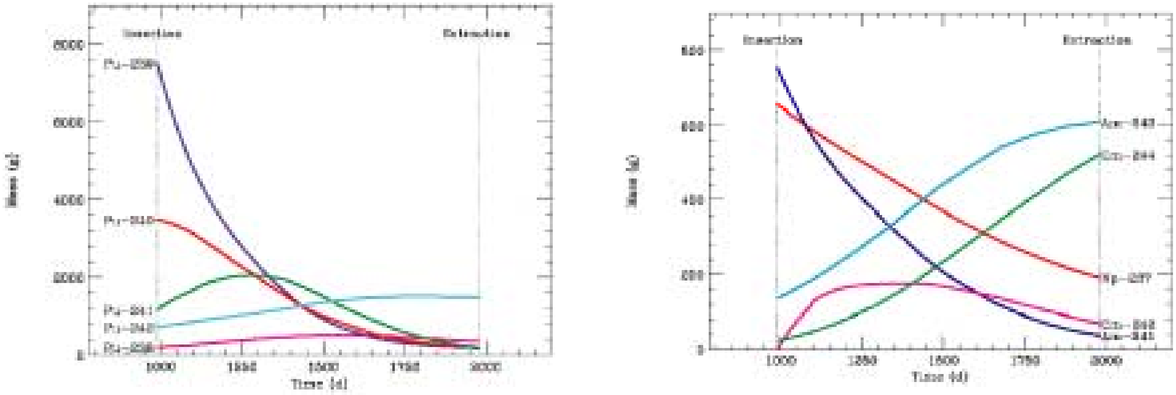


Figure 4. **Activity comparison of the radwaste stream before and after burning in the PBT**

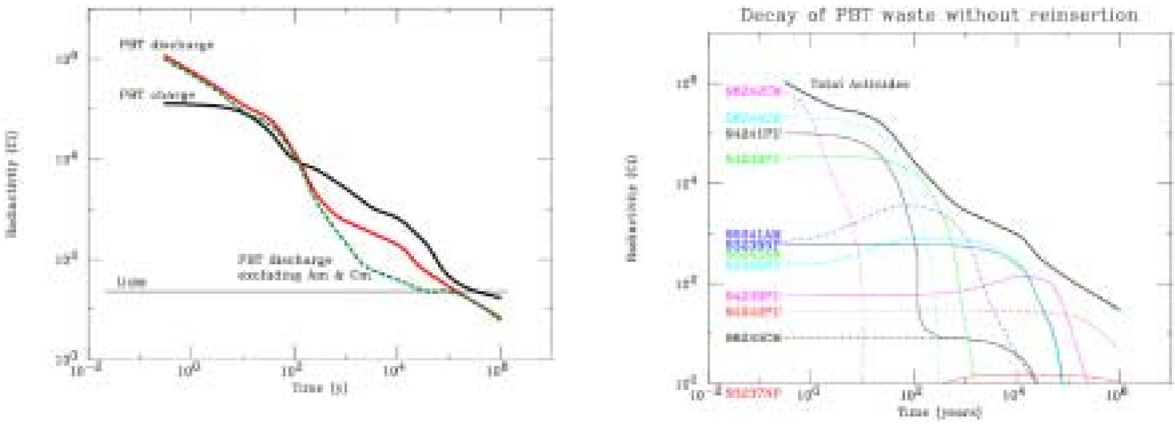


Table 2. **Initial and final TRU masses after irradiation in the 10 MW PBT**

Isotope	Initial mass (g)	Final mass (g)
^{237}NP	655	190
^{238}PU	203	346
^{239}PU	7 512	185
^{240}PU	3 481	201
^{241}PU	1 165	199
^{242}PU	713	1 484
^{241}AM	752	37
^{243}AM	134	610
^{242}CM	0	68
^{244}CM	24	522
^{245}CM	0	9

5. Cooling system

A preliminary design has been done to determine the pressure losses and working temperature at each part of the core during steady-state operation. He and N_2 have been taken into account as coolant candidates. The reference configuration that we have adopted after a parametric study is shown in Table 3. The increase in the mean gas temperature when passing through the core has been set up to 250ºC, resulting in a required pumping power between ~110 (He) and ~230 kW (N_2) at a gas working pressure of 20 bar. In these conditions, the maximum temperature reached in the fuel pellet is 982ºC, very well within the safety requirements for this kind of fuel.

A closed gas cycle has been considered for energy production in a PBT system. The cycle has the usual main components: high-pressure and low-pressure turbines, recuperator, compressor and condenser. The high-pressure turbine feeds the compressor to maintain the compression ratio between the working values. The low-pressure turbine is coupled to a generator connected to the external grid (or to the accelerator electrical power system). In Table 4 we summarise the general features of the system, for which we have chosen He as more favourable coolant. The required coolant flow to carry the 10 MW thermal power is 7.7 kg/s. The total efficiency in the cycle makes achievable to produce almost 4 MW of electrical power, what will be enough to supply the accelerator needs in steady-state conditions and under the established PBT design parameters. The scheme of the gas cycle and its TS diagram is shown in Figure 5.

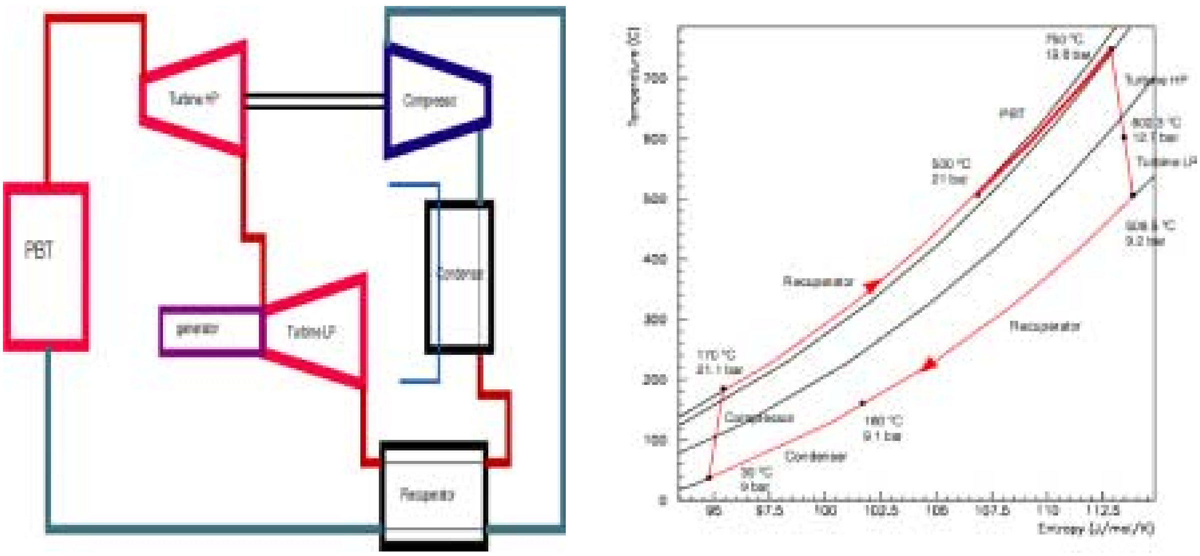
Table 3. **Cooling system reference parameters**

Parameter	**Value**	
Coolant	He	N_2
Pumping	Forced	Forced
Inlet temperature (°C)	500	500
Average outlet temperature (°C)	750	750
Pressure (bar)	20	20
Flow area/ball (cm^2)	0.5	0.5
Mass flow (kg/s)	7.7	34
Average velocity (m/s)	13.4	9
Pressure drop (Pa)	16072	48899
Pumping power (kW)	112	230
Mean mass density (kg/m^3)	1.1	7.2
Thermal power (MW)	9.7	9.7

Table 4. **General characteristics of the PBT gas cycle**

Compression ratio	2.3
He mass flow (for 10 MWth) (kg/s)	7.7
Polytropic efficiency of the turbine	0.89
Polytropic efficiency of the compressor	0.88
Efficiency coupling HP-compressor	0.95
Recuperator efficiency	0.95
Installation efficiency	0.38

Figure 5. **He gas cycle for the PBT and TS diagram**



6. Future R&D

Activities in progress are oriented to complete the above-mentioned studies and close a conceptual concept for PBT before going into design work. In particular, neutronics calculations are addressing: a) the feasibility to burn long-life fission products (as ^{99}Tc); b) the possibility to improve minor actinides elimination, and c) the optimisation of the power level in order to achieve transmutation rates suitable for industrial application.

Stability studies include reactivity coefficients analysis and transient studies and control methods, in special sub-criticality. Current work is in progress in two areas of research: the determination of the coolant gas void reactivity coefficient and on the effect of temperature and pressure transients on the system operating conditions. To measure the multiplication constant of the Pebble system we are addressing two methods have been successfully tested in Monte Carlo simulations performed with LAHET and MCNP-DSP [8,9]. The first one can be used with the proton source on and allows to perform an on line determination of the CPSD (w) function between the proton source and one neutron detector located in the system. This CPSD can be easily obtained experimentally, and Monte Carlo simulations with one detector in the system show that this method gives good values of the multiplication constant. Second method is based on the Mihalczo ratio of spectral densities [10,11] and can be used with the source turned off and a Californium source to excite the system.

PBT power level in these studies has been selected looking at the minimum required for a meaningful experimental device. Next step in our studies is the optimisation of that power level from an industrial point of view.

7. Conclusion

PBT device is devoted to nuclear waste transmutation, in particular Pu burning, although fission products elimination is also envisaged. Its transmutation efficiency is very high for most of the Pu isotopes in a LWR discharge. It is possible, in a single pass through PBT an almost complete elimination of ^{239}Pu, ^{240}Pu and ^{241}Pu with some residual long-lived and much less radiotoxic ^{242}Pu and some modest Cm and Am production. Such single pass-“once through” procedure is fast and can be accomplished in a time which is comparable to a single fuel cycle of a standard LWR. Additional technical and optimisation studies are in progress.

REFERENCES

[1] Rubbia C. *et al.*, *Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier,* (1995) CERN/AT/95-44 (ET).

[2] Arnould H. *et al.*, *Experimental Verification of Neutron Phenomenology in Lead and Transmutation by Adiabatic Resonance Crossing in Accelerator Driven Systems,* Phys. Lett. B 458 (1999) 167-180.

[3] Abánades A., Buono S., Castro A., Maciocco A., Moreau V., *Status Report of the Preliminary Design of the Proton Target Window for a Low-power ADS Prototype,* (1999) Internal report, LAESA-UI/99-16/23-VI.

[4] A. Fassò *et al.*, *FLUKA92*, Proc. of the Workshop on Simulating Accelerator Radiation Environments, Santa Fe, 11-15 January 1993.

[5] Prael R.E., Lichtenstein H., *User Guide to LCS: The LAHET Code System,* 1989. Group X-6. MS B226, Los Alamos National Laboratory.

[6] Briesmeister J.F., Editor, *MCNP. A General Monte Carlo N-Particle Transport Code,* 1997. LA-12625-M, Version B.

[7] Croff A.G., *A User’s Manual for the ORIGEN2 Computer Code,* 1980, ORNL/TM-7175, July 1980.

[8] Rubbia C., *Resonance Enhanced Nuclear Waste Transmutation,* University of Pavia, May 1999.

[9] Rugama Y., Muñoz-Cobo J.L., Valentine T. (2000), Proceedings of the MC2000 (Monte Carlo 2000, Lisbon), in press.

[10] Muñoz-Cobo J.L., Rugama Y., Valentine T.E., Mihalczo J.T., Pérez R.B., *Sub-critical Reactivity Monitoring in Accelerator Driven Systems*, submitted for publication to Annals of Nuclear Energy (2000).

[11] Mihalczo J.T., *Nuclear Science and Engineering*, Vol. 53, pp. 393-414 (1974).