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Experimental Verification of Neutron Phenomenology in Lead and Transmutation by Adiabatic Resonance Crossing in Accelerator Driven Systems: a Short Summary

The TARC Collaboration[*]

Abstract

The Transmutation by Adiabatic Resonance Crossing (TARC) experiment was carried out as PS211 at the CERN PS from 1996 to 1999. Energy and space distributions of spallation neutrons (from 2.5 and 3.57 GeV/c CERN proton beams) slowing down in a 3.3×3.3×3 m^3 lead volume and neutron capture rates on long-lived fission fragments ^{99}Tc and ^{129}I demonstrate that Adiabatic Resonance Crossing (ARC) can be used to eliminate efficiently such nuclear waste and validate innovative simulation.

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The TARC Collaboration

A. Abánades[n,(§)], J. Aleixandre[b], S. Andriamonje[c,a], A. Angelopoulos[j], A. Apostolakis[j(‡)], H. Arnould[a], E. Belle[g], C.A. Bompas[a], D. Brozzi[c], J. Bueno[b], S. Buono[c,h,(¥)], F. Carminati[c], F. Casagrande[c,e,(¶)], P. Cennini[c], J.I. Collar[c,(æ)], E. Cerro[b], R. Del Moral[a], S. Díez[m,(§)], L. Dumps[c], C. Eleftheriadis[l], M. Embid[i,d], R. Fernández[c,d], J. Gálvez[i], J. García[n,d], C. Gelès[c], A. Giorni[g], E. González[d], O. González[b], I. Goulas[c], D. Heuer[g], M. Hussonnois[f], Y. Kadi[c], P. Karaiskos[j], G. Kitis[l], R. Klapisch[c,(ﬁ)], P. Kokkas[k], V. Lacoste[a], C. Le Naour[f], C. López[i], J.M. Loiseaux[g], J.M. Martínez-Val[n], O. Méplan[g], H. Nifenecker[g], J. Oropesa[c], I. Papadopoulos[l], P. Pavlopoulos[k], E. Pérez-Enciso[i], A. Pérez-Navarro[m,(§)], M. Perlado[n], A. Placci[c], M. Poza[i], J.-P. Revol[c], C. Rubbia[c], J.A. Rubio[c], L. Sakelliou[j], F. Saldaña[c], E. Savvidis[l], F. Schussler[g], C. Sirvent[i], J. Tamarit[b], D. Trubert[f], A. Tzima[l], J. B. Viano[g], S. Vieira[i], V. Vlachoudis[a,l], K. Zioutas[l].[*]

(a) CEN, Bordeaux-Gradignan, France; (b) CEDEX, Madrid, Spain; (c) CERN, Geneva, Switzerland; (d) CIEMAT, Madrid, Spain; (e) INFN, Laboratori Nazionali di Frascati, Italy; (f) IPN, Orsay, France; (g) ISN, Grenoble, France; (h) Sincrotrone Trieste, Trieste, Italy; (i) Universidad Autónoma de Madrid, Madrid, Spain; (j) University of Athens, Athens, Greece; (k) University of Basel, Basel Switzerland; (l) University of Thessaloniki, Thessaloniki, Greece; (m) Universidad Alfonso X el Sabio, Madrid, Spain; (n) Universidad Politécnica de Madrid, Madrid, Spain.

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[*] (§) Present address LAESA, Zaragoza, Spain

(¥) Present address CRS4, Cagliari, Italy

(¶) Present address MIT, Cambridge, USA

(æ) Present address Université Paris VI, Paris, France

(ﬁ) Also at CSNSM, IN2P3, Orsay, France

(‡) Deceased

The TARC experiment was carried out at the CERN PS and received strong support from the European Union, within its Fourth Framework Programme. The first results of the TARC experiment have been published in Ref. [1], and a detailed description of the various experimental techniques and of the results can be found in Ref. [2]. We only present here a brief summary of the concepts and results.

Adiabatic Resonance Crossing

An Accelerator Driven System (ADS) such as the Energy Amplifier (EA) [3], which is a fast neutron sub-critical device using natural thorium as fuel and lead as the neutron spallation target, moderator, heat-removal agent and neutron confinement medium, has the potential to destroy actinide elements by fission, thereby producing energy. In such a scenario, long-term (≥ 500 years) radiotoxicity of the waste is dominated by long-lived fission fragments (LLFF) which can, in practice, only be eliminated by nuclear decay to a stable state, following neutron capture.

In the field of medicine, radioactive elements are increasingly used for diagnoses, therapy and pain relief. These elements can be produced through neutron capture on stable elements in an accelerator-driven activator, as an alternative to nuclear reactor production, using the ‘inverse’ process invoked for the destruction of LLFF [4].

In both cases, it is important to optimize the efficiency of the neutron capture process. The specific neutron capture rate, R_{capt}, can be enhanced by maximizing each of the relevant factors in $R_{\mathrm{capt}} \equiv \int \phi(E)\sigma(E)\mathrm{d}E$ (E is the neutron energy) i.e.

(a) <u>the neutron flux</u> [$\phi(E)$] (Fig. 1) by the choice of a dense neutron ‘storage’ medium (lead) with high atomic mass, high neutron elastic cross-section (mean free path: λ_{el} ~3 cm) and high neutron transparency (the double magic nature of the ^{208}Pb nucleus makes natural lead one of the most transparent elements below 1 keV);

(b) <u>the effective capture cross-section</u> [$\sigma(E)$] (Fig. 1) of the element to be transmuted by an efficient use of resonances made possible by the very small lethargic steps of neutrons slowing down in lead, with the effect to increase the epithermal neutron flux.

The purpose of the Transmutation by Adiabatic Resonance Crossing experiment (TARC) was to test directly the concept of ARC to enhance significantly the neutron capture efficiency for LLFF, an idea put forward by C. Rubbia [4]. Indeed, neutrons have an interesting behaviour in lead:

(i) a small average lethargy ξ due to the high atomic mass of lead:

$$\xi \equiv 1 + \frac{\alpha}{1-\alpha} ln(\alpha) \approx 9.6 \times 10^{-3} \quad where\ \alpha \equiv \frac{\left(m_{Pb} - m_n\right)^2}{\left(m_{Pb} + m_n\right)^2} \approx 0.98;$$

(ii) a high and nearly energy-independent elastic scattering cross-section;

(iii) a long ‘storage’ time because, below the capture resonances ($E_{\mathrm{n}} \leq 1$ keV) and down to epithermal energies, the elastic scattering process is nearly isotropic and the lead transparency to neutrons is very high (it takes about 3 ms, 1800 scatterings and a path in lead of 60 m to thermalize a 1 MeV neutron).

As a consequence, neutrons produced by spallation at relatively high energy ($E_n \approx$ few MeV), after having been quickly moderated by (n,xn), (n,n') reactions down to energies of a few hundred keV will slow down quasi-adiabatically with small isolethargic steps and reach the capture resonance energy of an element to be transmuted where they have a high probability of being captured. The resonance width is usually larger than the average lethargic step. This is the case for instance for ^{99}Tc which has a strong neutron capture resonance at 5.6 eV (4000 barn) (Fig. 1) covering four average lethargy steps. Its resonance integral is 310 barn while the cross-section at thermal/epithermal neutron energies ($E_n \leq 1$ eV) is only about 20 barn. Neutron capture on ^{99}Tc ($t_{1/2} = 2.1\times10^5$ yr) produces ^{100}Tc ($t_{1/2}$ = 15.8 s) which then decays to ^{100}Ru, a stable element. Thus, the radiotoxicity can be eliminated in a single neutron capture and, since ^{100}Ru has a small neutron capture cross-section and both ^{101}Ru and ^{102}Ru are stable, essentially no new radioactive elements are produced. ARC should be most efficient for elements with strong capture resonances, such as ^{99}Tc and ^{129}I (which represent 95% of the total LLFF radiotoxicity inventory).

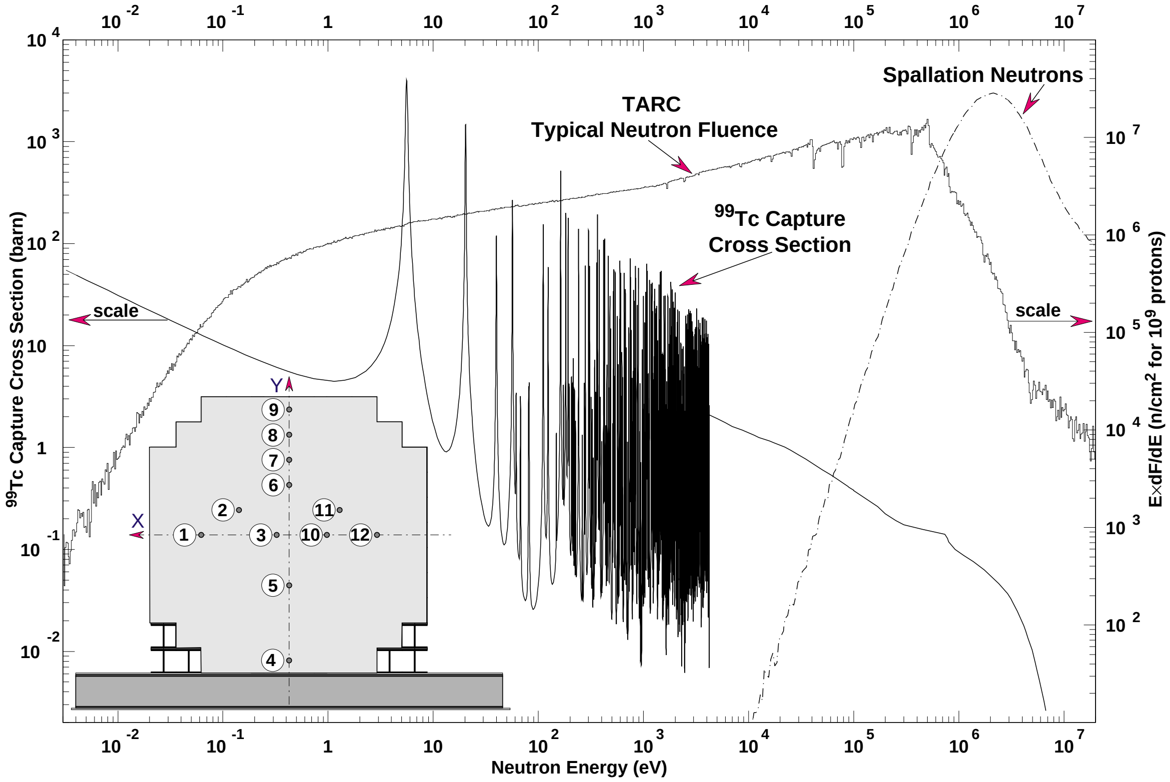

Figure 1: ^{99}Tc neutron capture cross section (JENDL-3.2 data base [5]), as a function of neutron energy (left-hand side scale); Typical neutron fluence energy distribution in TARC, as a function of neutron energy in isolethargic bins, for 3.57 GeV/c protons (right-hand side scale); Energy distribution of spallation neutrons as they were born in lead in arbitrary units. About 14% of these neutrons are above 20 MeV. Instrumentation hole positions are shown in a sketch of the detector seen along the beam line (Z axis).

TARC Experimental Set-up

Beams of protons from the CERN Proton Synchrotron (PS) hit a large lead assembly. The PS beam was provided in two different modes:

(a) fast extraction mode (20 ns long pulses; momentum of 2.5 or 3.57 GeV/*c*; intensity ranging from 10^7 to 10^{10} protons per pulse with a repetition time of 14.4 s) used either for measurements of neutron energy from timing, or for sample irradiation requiring high intensity.

(b) slow extraction mode magnetically selecting secondary particles produced on a target made of aluminium and tungsten (spill length of about 350 ms; momentum of 2.50 GeV/*c*). This mode was used for neutron fluence measurements with ^{3}He ionization counters which required a very low beam intensity (1000 protons per spill) not achievable in fast extraction mode.

Pure lead (99.99%) was chosen to ensure that impurities have a negligible effect on the neutron flux. The 334 ton lead assembly with approximate cylindrical symmetry about the beam axis, was the result of an optimization between sufficient neutron containment (about 70% of all neutrons remain inside the volume), acceptable background conditions [within 1 m radius from the beam axis, neutron background from the environment is negligible ($\leq 2\%$)], and affordable cost.

Calibration of the Energy–Time Relation

Through the elastic scattering process in lead, a strong correlation develops between the time at which a neutron is observed and its kinetic energy:

$$E = \frac{K}{(t + t_0)^2}$$

This correlation, already noted by Feinberg in 1944, has been used to measure neutron energies in so-called slowing down spectrometers and is easily explained by Fermi’s theory of continuous slowing down. In TARC, two of the neutron fluence detectors (^{6}Li/^{233}U and ^{3}He/scintillation) use this technique to obtain the neutron energy from the measurement of the interaction time. The parameters K and t_0 of the energy-time relation were measured with the help of a $\mathrm{CeF_3}$ crystal mounted in front of a photomultiplier equipped with a quartz window to record the time distribution of the fast $\mathrm{CeF_3}$ scintillation UV light produced by prompt γs associated with radiative neutron capture on thin samples of elements with known capture resonance energies [^{181}Ta (4.28 eV), ^{197}Au (4.91 eV), ^{109}Ag (5.19 eV), ^{99}Tc (5.58 eV), ^{115}In (9.07 eV), ^{107}Ag (16.30 eV), ^{55}Mn (337 eV)]. We find that $K = (172 \pm 2)\ \mathrm{keV} \times \mu\mathrm{s}^2$ and $t_0 = 0.37 \pm 0.20\ \mu\mathrm{s}$, in good agreement with the Monte Carlo simulation. The observed resonance widths are consistent with the expected resolution of a slowing down spectrometer, given by:

$$\frac{\sigma_E}{E} \approx 11.3\sqrt{1 + \frac{2}{E(eV)}}\ (\%) \quad \left(valid\ for\ E_n \leq 1\ keV\right).$$

TARC Experimental Programme

In order to test the TARC effect, we have performed

(a) neutron fluence measurements with several complementary techniques (^{6}Li/^{233}U detectors, ^{3}He scintillation detectors, triple-foil activation, ^{3}He ionization detectors, TLD detectors, ^{232}Th fission detection in Lexan, activation measurements of $^{12}\mathrm{C(n,2n)C}^{11}$ and $^{12}\mathrm{C(n,3n)}^{10}\mathrm{C}$, as well as thermometer measurements of the heat deposited by neutrons in the lead medium), providing redundancy of information.

We have collected a large coherent set of neutron fluence data, over eight orders of magnitude in neutron energies (from thermal to 2 MeV), throughout the entire lead volume. In the energy region below 10 keV, this was done with three different detector techniques each with an absolute precision of the order of 15%.

At low energies, we have verified that $dF/d[\ln(E)]$, where F is the neutron fluence, decreases very slowly with neutron energy, only by one order of magnitude in the neutron energy range from 10 keV to 1 eV. This is the first important element in the demonstration of the efficiency of ARC. Furthermore, within a radius of 1.5 m, the neutron fluence, in good approximation, has spherical symmetry and behaves approximately as for a point-like source, even though neutron production through the spallation process is neither point-like nor isotropic. This is of course precisely what is expected from the small elastic collision length in lead and also from the fact that the lead volume is sufficiently large that edge effects do not constrain the shape of the neutron cloud.

On the contrary, at high energies (≥ 500 keV), the spatial distribution shows a significant forward-backward asymmetry (fission measurements in ^{232}Th, ^{12}C(n,2n)^{11}C, ^{12}C(n,3n)^{10}C and thermometers), and is limited to a relatively small region (~ 50 cm) from the centre of the cascade.

(b) neutron capture rate measurements on ^{99}Tc, ^{129}I and ^{127}I

For ^{99}Tc we used a ‘rabbit’ pneumatic system allowing a ^{99}Tc sample contained in a small low-activation carbon fibre shuttle to be positioned precisely at a given location within the lead volume.

The experimental precision in the measurement of the γ rate from ^{100}Tc decay is 8.2%, but the resulting uncertainty on ^{99}Tc transmutation rate is 16%, of which a global error of 14% comes from the uncertainty in the fraction of ^{100}Tc decays producing γ's (Br ~ 7%). The excellent agreement observed between predicted and measured transmutation rates confirm that ARC is under control.

With the CeF_3 crystal used to calibrate the energy–time relation, we measured the ^{99}Tc transmutation rate as a function of neutron energy. These measurements also agree well with our simulation and provide, after dividing by the neutron fluence, a high statistics measurement of the ^{99}Tc apparent neutron capture cross-section up to about 1 keV, with a precision of 20%.

For iodine, a sample of 64.7 mg of ^{129}I in the form of PbI_2 was irradiated at five different positions in the TARC volume. We used the 13.9% of ^{127}I present in the PbI_2 sample to measure at the same time the capture rate on natural iodine, relevant for the production of radioactive isotopes (i.e. ^{128}I) for medical applications.

We find a production ratio of ^{130m}I over ^{130g}I of 1.58 ± 0.14 in agreement with a previous measurement at MIT [6]. The total uncertainty on the ^{129}I and ^{127}I capture rates is 10%. Both for ^{129}I and for ^{127}I, the rates of transmutation predicted by our simulation is in excellent agreement with our measurements.

The many measurements performed with ^{99}Tc, ^{129}I, and ^{127}I throughout the lead volume provide self-consistency checks, including reproducibility of the measurements and control of the systematics, and confirm the validity of ^{99}Tc and ^{129}I cross-section data.

The excellent agreement with the simulation demonstrates without ambiguity the efficiency of the ARC effect.

Many other capture or fission measurements relevant to the design parameters of the EA or to various other applications were performed (neutron capture on ^{232}Th, ^{238}U, etc.; fission rates for ^{235}U, ^{nat}U, ^{237}Np, ^{239}Pu, ^{232}Th, etc.; ^{232}Th(n,2n)^{231}Th and production of ^{99m}Tc from natural molybdenum and of ^{128}I from ^{127}I in view of medical applications).

TARC Monte Carlo Simulation and Computing

An appropriate formalism and appropriate computational tools essential for the analysis and understanding of the data were developed and validated. Adequate statistics were obtained by using both a parallel computer CONVEX SPP 1200 with eight processors and a SPP2000 with sixteen processors.

A special effort was made, in the framework of the EA programme, to develop an innovative simulation which has taken up the most challenging approach to the problem of neutron transport and burnup simulation by integrating transport of the neutrons generated in the spallation process and evolution of material composition under the combined effects of neutron activation and nuclear decay. We now have a unique tool which combines the full precision of the Monte Carlo method with high simulation speed and an integrated time-evolution during burnup. In particular, the complex structure of cross-sections in the resonance region requires careful treatment to describe correctly self-shielding effects. The spallation process itself was simulated separately with FLUKA-96 [7], one of the best Monte Carlo codes available for hadronic interactions at intermediate energies.

Both the energy and space distributions and the absolute magnitude of the neutron fluence, throughout the entire lead volume, are well described by the simulation. For instance, neutron fluence data (Fig. 2) from thermal energies up to 2 MeV (over eight orders of magnitude) are very well reproduced, at both proton beam energies. We find that about 70% of the spallation neutrons survive the lead capture resonances. The ratio of fluences (between 0.1 eV and 10 keV) measured at two proton momenta (3.5 and 2.5 GeV/*c*) is found to be 1.52 ± 0.10 consistent with the expectation from the simulation: 1.57 ± 0.01 (stat.) which is essentially the ratio of kinetic energies of the beams. The global uncertainty in the Monte Carlo prediction comes mainly from the uncertainties in the spallation process (10%) and in the neutron transport in lead (mainly the uncertainty on the lead cross-section of 10%) and amounts to a total of about 15%. The impurity content is small enough, with sufficiently well-measured concentrations, that the corresponding uncertainty on the neutron fluence is negligible.

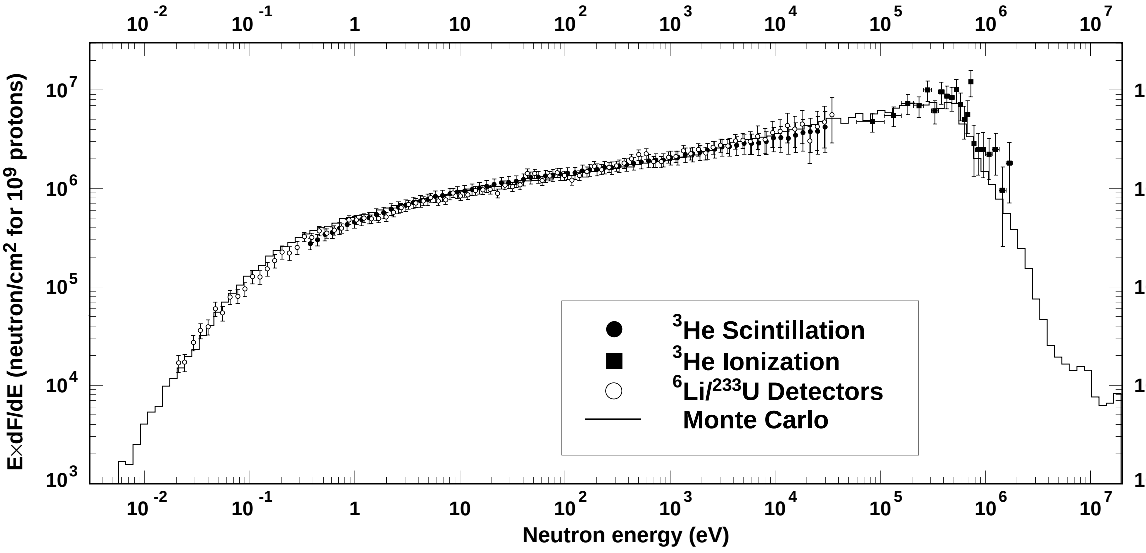

Figure 2: Distribution of the neutron fluence versus neutron energy in hole 10 at z = +7.5 cm, as obtained from ^{3}He scintillation, ^{3}He ionization and ^{6}Li/^{233}U detectors. The histogram is from the TARC Monte Carlo simulation. These data were taken with a proton beam momentum of 2.5 GeV/c.

The excellent agreement with the entire TARC data set validates in detail our innovative simulation. It confirms in particular that the spallation process is correctly predicted within the errors mentioned above. Typically, we expect 100 neutrons per 3.57 GeV/*c* proton (FLUKA neutrons with transport down to 19.6 MeV). The dependence of the neutron fluence on energy and space validates the neutron transport code and tests the reliability of the lead cross-sections. The correct prediction of integral and differential transmutation rates for ^{99}Tc and ^{129}I validates the efficiency of the TARC method to transmute LLFF.

Practical Scheme for the Transmutation of LLFF

The validated TARC simulation was then used to predict the transmutation rate for ^{99}Tc, in a dedicated lead volume outside the EA core where the neutron fluence is quasi isolethargic, precisely as in TARC.

For instance, in a 1 GW$_{th}$ EA, after a burnup of 100 GWatt$_{th}$ × day/ton and with an initial load of 270 kg of ^{99}Tc uniformly distributed in lead at a mass concentration of 10^{-3}, the TARC simulation predicts a transmutation rate of 12.3 kg/GW$_{th}$ × year to be compared to the production rate of 7.5 kg/GW$_{th}$ × year in the EA [4]. With a mass concentration of 1.8×10^{-3} for ^{129}I and an initial load of 430 kg, one obtains a ^{129}I burning rate of 8.8 kg/GW$_{th}$ × year to be compared to a production rate of 3.5 kg/GW$_{th}$ × year in the EA. The fact that this is happening outside the EA core, in a region where neutrons have a small probability to return to the fuel and therefore, little chance to contribute to fissions, makes it possible to envisage LLFF transmutation in a parasitic mode, thus minimizing the cost of the process. We find it a very attractive idea to profit from un-used neutrons outside the EA core to reduce the overall radiotoxicity.

On the basis of the TARC results, it appears possible to destroy outside the EA core, in a parasitic mode, large amounts of ^{99}Tc, ^{129}I or other LLFF at a rate exceeding the production rate, thereby making it practical to reduce correspondingly the existing stockpile of LLFF.

The TARC effect can be extended to other domains to provide an alternative to the production of radioactive isotopes with nuclear reactors (medical and industrial applications), to provide high neutron fluxes for research applications, for instance, high-precision measurements of neutron cross-sections by the time-of-flight method in the time-of-flight facility (TOF [8]) at CERN, to perhaps provide a way to construct a highly efficient engine for deep space travel [9], etc.

References

[1] H. Arnould et al., Phys. Lett. B 458 (1999) 167-180.

[2] H. Arnould et al., "Neutron-driven nuclear transmutation by adiabatic resonance crossing", DG XII / D.II.3 R & T Specific programme "Nuclear fission and safety", EUR 19117 EN;
H. Arnould et al., "The TARC Experiment (PS211): Neutron-driven nuclear transmutation by adiabatic resonance crossing, CERN Yellow Report 99-11, 15 December 1999.

[3] C. Rubbia et al., "Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier", CERN / AT / 95-44 (ET), Sept. 29, 1995;
see also C. Rubbia, "A High Gain Energy Amplifier Operated with fast Neutrons", AIP Conference Proc. 346, Int. Conf. on ADT Technologies and Applications, Las Vegas, 1994.

[4] C. Rubbia, "Resonance Enhanced Neutron Captures for Element Activation and Waste Transmutation", CERN / LHC / 97-04 (EET), June 22, 1997.

[5] JENDL-3.2 (OECD NEA Data Bank, 1996).

[6] P.K. Hopke et al., Phys. Rev. C 8 (1973) 745.

[7] A. Fassó et al., in *"Intermediate Energy Nuclear Data: Models and Codes"*, Proceedings of a Specialists', Meeting, Issy les Moulineaux (France) 30 May-1-June 1994, p. 271, published by OECD, 1994, and references therein; A. Fassó, A. Ferrari, J. Ranft, P.R. Sala, G.R. Stevenson and J.M. Zazula, Nuclear Instruments and Methods A, 332, 459 (1993); CERN report CERN / TIS-RP / 93-2 / PP (1993); A. Ferrari et al., “FLUKA-96 Manual”, INFN, Milan, 1996; A. Ferrari and P.R. Sala, "The physics of high energy reactions", Lecture given at the Workshop on Nuclear Reaction Data and Nuclear Reactor Physics and Safety, International Centre for Theoretical Physics, Trieste, 15 April - 17 May 1996.

[8] C. Rubbia et al., "A High Resolution Spallation Driven Facility at the CERN-PS to Measure Neutron Cross Sections in the Interval from 1 eV to 250 MeV", CERN / LHC / 98-02 (EET), May 30, 1998; "A High Resolution Spallation Driven Facility at the CERN-PS to Measure Neutron Cross Sections in the Interval from 1 eV to 250 MeV: A Relative Performance Assessment", CERN / LHC / 98-02 (EET)-add. 1, June 15, 1998;
S. Abramovich et al., "Proposal for a Neutron Time of Flight Facility", CERN / SPSC 99-8, SPSC / P 310, 17th March 1999.

[9] C. Rubbia, "Report of the Working Group on a Preliminary Assessment of a New Fission Fragment Heated Propulsion Concept and its Applicability to Manned Missions to the Planet Mars", Italian Space Agency, Rome, April 1, 1999 and private communication.