NEUTRON DATA MEASUREMENTS FOR TRANSMUTATION AT EC-JRC-IRMM

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Abstract

Studies in waste transmutation require neutron data for conceptual and performance studies, as well as the study of ADS viability and the safety analysis of any of the involved new concepts. In Framework Program 6 the IRMM's Neutron Physics Unit has subdivided its activities in two actions, one of which concerns neutron data measurements for waste transmutation and new concepts in nuclear energy production. The ongoing and recently completed activities in this action include capture and total cross section measurements for ^{127,129}I, ^{206}Pb, ^{232}Th, precision total cross section measurements for ^{240,242}Pu, inelastic scattering measurements for ^{52}Cr, ^{58}Ni, ^{209}Bi and ^{207}Pb, (n,2n) measurements for ^{207}Pb, fission cross section measurements for ^{233}Pa, ^{234}U and ^{236}U, tritium yield measurements in ternary fission and (n,xp), (n,xα) and (n,xn) cross section measurements with the activation technique.

Introduction

The Neutron Physics unit of the IRMM performs neutron data measurements at two laboratories. The first, GELINA [1] (Fig. 1), is a pulsed white neutron source at which high resolution time-of-flight measurements are performed in the energy range from 1 meV to 20 MeV. Gelina uses a pulsed electron linear accelerator with repetition rates from 40 – 800 Hz and a unique pulse width of 0.7 ns full width at half maximum (fwhm). Electrons with energies from 70-140 MeV strike a rotating uranium target producing bremsstrahlung and subsequently neutrons by the (γ,n), (γ,2n) and (γ,F) reactions. Neutron-induced reactions may be studied at as many as twelve different neutron beams, simultaneously, with flight paths ranging from 8 to 400 m. Measurements either utilise the spectrum from 1 meV to 300 keV from water moderators placed above and below the uranium target or the spectrum from 50 keV to 20 MeV from the target itself. Installed measurement equipment includes setups for the measurement of total cross sections with the transmission technique, including the possibility of controlling the sample temperature from 15 K to room temperature, for the measurement of radiative capture cross sections, for the measurement of fission cross sections and charged particle emission cross sections, and for the measurement of gamma-production cross sections for gammas emitted in inelastic scattering and (n,2n) reactions. The second laboratory (Fig. 2) uses a 7 MV van de Graaff accelerator to produce quasi mono-energetic neutrons from 100 keV to 21 MeV with the Li(p,n), T(p,n), D(d,n) and T(d,n) reactions. Measurements performed include fission cross sections, fission product mass and kinetic energy distributions, activation cross sections and the ^{10}B(n,α) cross section.

Activities of the unit are grouped in two actions, "Basic research in nuclear physics and neutron data standards" and "Neutron data for waste transmutation and safety of different reactor systems". Here, an overview of the latter will be presented. For an overview of the former, see Refs. [4]. Measurements for waste transmutations are grouped under four headings that reflect the different issues and, to some extent, the different strategies. The first concerns data for the isotopes to be transmuted and the production of radioactive isotopes of concern for transmutation or for handling of nuclear waste (reprocessing, partitioning, and conditioning). The second concerns data specific to accelerator driven systems. The third concerns data for the thorium/uranium fuel cycle and the fourth concerns data for advanced concepts featuring high burn up and/or multiple Pu recycling.

The tasks pertaining to each of these headings are summarised below. As will be shown, many of these are carried out as collaborations with laboratories from Member States, Candidate Countries, West-Balkan States, the USA, Japan and Russia. Close contact with the Partitioning and Transmutation policy of the European Commission Directorate-General for Research, Technology and Development is maintained through participation in the Framework Program 5 (FP5) Shared Cost Action n_TOF-ND-ADS and the FP6 Integrated Project EUROTRANS. Measurement prioritisation is based on issues raised at the meetings of the Joint European Fusion and Fission (JEFF) library project, the annual meeting of the Working Party on Evaluation Cooperation (WPEC) – both coordinated by the OECD Nuclear Energy Agency, Nuclear Science Committee – and international conferences. The IRMM aims to widen access to its neutron facilities by external users through the EURATOM Research and Training program "Transnational Access to Large Infrastructures" scheme.

Neutron measurements for transmutation of nuclear waste

Radio toxicity of high level nuclear waste as well as its longevity is dominated, first of all by the isotopes of Pu, and in next approximation by those of the minor actinides (MA). Long-lived fission products (LLFP) constitute a minor contribution to radio toxicity, but isotopes like ^{99}Tc and ^{129}I that are produced in reasonably large quantities show high mobility and may in the long run permeate to surface water, whereas this is not likely for the actinides. Thus, transmutation scenarios focus on reuse of Pu and MA, but also look for economic methods of dealing with the most important LLFPs. For a proper optimisation of transmutation scenarios, fission, capture and total cross sections are required in energy ranges from thermal to several MeV. At IRMM total and capture cross sections measurements were made for ^{237}Np [6] and ^{99}Tc [7] in collaboration with CEA Saclay and, most recently, ^{129}I [8] in collaboration with CEA Cadarache.

Measurements for ^{129}I and their analysis were recently completed (Figs. 3 and 4.). Measurements were performed on a PbI_2 sample containing fission product iodine from spent fuel obtained from La Hague. As the sample contained both ^{127}I and ^{129}I in a ratio of about 1 to 4, capture and transmission measurements for ^{127}I were made under the same conditions in order to disentangle the contributions from the two isotopes. Complicated chemistry for the separation of the iodine from the waste and the production of the sample resulted in a poor knowledge of the stoichiometry. A combination of analyses of the sample finally resulted in an adequate understanding of the elemental and isotopic composition. Resonance shape analyses of ^{127,129}I capture and transmission data was made up to 10 keV, whereas the average capture and transmission data were analysed with Sammy/Fitacs from 3.5 keV to 100 keV. Resonance parameters were obtained for 719 resonances of ^{127}I and for 400 resonances of ^{129}I. From the averaged data, s- and p-wave strength functions, potential scattering radii and mean radiative widths were derived. For the resonance region an ENDF formatted file was generated for both ^{127}I and ^{129}I, ready for inclusion in JEFF3.1. Preliminary application libraries were generated for MCNP and ERANOS in the interest of the Profil and Efftra projects. The ^{129}I transmutation half-life for Profil-M is very close to that obtained with JEFF3.0 whereas it is about 2 standard deviations larger for Profil-R. Transmutation rates for ^{127}I in the HFR/Efftra irradiations are about 3.5% larger than those of JEFF3.0 and 23% less than for ENDF/B-VI.

Neutron data for Accelerator Driven Systems

Several scenarios for transmutation of waste feature a number of accelerator driven systems (ADS), either as the primary means of elimination of the accumulated volume of high level waste (HLW), or as the closing stone in a multi-strata scenario for a closed fuel cycle to eliminate a fraction of the minor actinides that could not be taken care of by a foreseen park of fast reactors. ADS are actively studied in the EU, Japan, Russia and the US. Conceptual design studies for an experimental and a demo facility are the subject of the FP6 Integrated Project Eurotrans.

Inelastic scattering

In a recent paper Aliberti et al. [9] performed a sensitivity study propagating the desired final uncertainties on key reactor and ADS target/source parameters backwards to determine desired final uncertainties for cross sections. One of the conclusions of that work is that it will be necessary to improve on inelastic cross sections of lead and bismuth to obtain a final uncertainty on the level of 5-10%. Lead and bismuth inelastic cross sections impact reactor parameters such as K_{eff} through the so-called indirect effect: a modification of reaction rates as a result of a change in the neutron spectrum.

At IRMM a new setup was developed [10] for the measurement of gamma-ray production cross sections from inelastic scattering and (n,2n) reactions. Excitation curves are obtained from threshold to 20 MeV. Total inelastic and level inelastic cross sections are deduced relying on the knowledge of the level scheme as obtained from the ENSDF database. This formally limits the results to be exact up to a few MeV (3-5 MeV) after which we obtain a lower limit for the total inelastic cross section and an upper limit for the level inelastic cross sections. Measurements were previously made for ^{52}Cr [11] and ^{58}Ni [12] showing the lower limit to the inelastic cross section to be very close, i.e. very nearly coinciding with the actual cross section. On the contrary, for level cross sections the upper limit derived by our method clearly overestimates the level inelastic cross section above the excitation energy where the level scheme is known, a fact well established in the literature.

Gamma-ray production cross section measurements for ^{209}Bi were undertaken and a preliminary analysis was recently completed [13] (Fig. 5). Excitation curves were determined for 19 transitions corresponding to 19 consecutive excited states up to 3.4 MeV excitation energy. The inelastic cross section was constructed up to 18 MeV and compared to existing data and a blind Talys calculation showing agreement at the level of 20% in the entire energy range studied.

These measurements are part of a campaign in which all isotopes of lead will be studied as well (except for ^{204}Pb). Data taking for ^{207}Pb was recently completed [14] and measurements for ^{206}Pb and ^{208}Pb are scheduled for 2005. The equipment will be upgraded from two HPGe detectors of 65 and 85% relative efficiency to seven HPGe detectors, six of which will have 95% relative efficiency. Data-acquisition will be upgraded from conventional electronics to a system based on 400 MSPS, 12 bit digitizers to minimize deadtime and remove slow rise time dependence of the time resolution.

The ^{207}Pb(n,2n)^{206}Pb reaction

The hard spectrum of an ADS reaches well over the 10 MeV conventionally needed for fast reactors and thus has led to an enhanced interest in (n,xn) reactions for incident energies as high as 200 MeV. Outside of the spallation target the spectrum is dominated by neutrons below 20-30 MeV and source term, multiplication factor and radiation damage estimates are sensitive to (n,2n) and (n,3n) reactions of the major isotopes in the system. In the context of the FP5 n_TOF-ND-ADS shared cost action such reactions are studied by a collaboration of IReS Strasbourg, U and TU of Vienna and the Vinca Institute in Belgrade.

At IRMM measurements were made for the ^{207}Pb(n,2n)^{206}Pb reaction using a 92.4% enriched ^{207}Pb sample of 89.9 g. Such measurements were facilitated by the use of a 65 MSPS 14 bit digitizer with on board signal processing capabilities implemented in the form of a Field Programmable Gate Array (FPGA). This device was developed at IReS in the interest of several measurements employing HPGe, such as AGATA and ISOLDE. Gamma-production cross sections were measured with the setup described above for inelastic scattering measurements. Three HPGe detectors were used at angles of 110, 125 and 150 degrees. Preliminary analysis has shown the clear alteration of the spectrum when the energy moves across the (n,2n) threshold from one dominated by the (n,n') reaction on ^{207}Pb to one dominated by the (n,2n) reaction, i.e. with the strongest gammas from ^{206}Pb. Data analysis is in progress [14].

The ^{209}Bi(n,γ)^{210m,g}Bi cross sections and isomer ratio

This reaction is of relevance for lead/bismuth eutectic cooled and moderated reactors as it leads to a short lived alpha emitter, ^{210}Po, and a long-lived one, ^{210m}Bi. The α-emitter ^{210}Po ($T_{1/2}$ = 138.4 d) results from β-decay of ^{210g}Bi ($T_{1/2}$ = 5.013 d). The isomeric state at 271.3 keV has a half-life $T_{1/2}$ of 3.04 x 10^6 years. IRMM, CEA Saclay and the KFKI Budapest initiated a collaboration to determine the ^{209}Bi(n,γ)^{210m}Bi and ^{209}Bi(n,γ)^{210g}Bi cross sections and corresponding isomer ratio in an energy region between thermal and 500 keV. Measurements at a reactor beam and at GELINA are scheduled. First experimental data obtained at ILL at thermal energy [15] showed good agreement with the compilation of Mughghab for the total cross section, but the isomer ratio deviates significantly. To confirm the ILL result measurements at the cold neutron beam of the Budapest Neutron Centre (BNC) were undertaken using the PGAA setup. The new measurements confirm the result for the isomer ratio obtained at ILL and also clarify the suitability and limitations of the method [16]. In a subsequent work the energy dependence of the cross sections and isomer ratios that is of relevance to fast reactors will be studied at the Gelina time-of-flight facility.

The $^{206}Pb(n,\gamma)^{207}Pb$ cross section

In a lead or lead/bismuth cooled and moderated fast reactor a considerable number of neutrons will be lost through capture on isotopes of lead even if such cross sections are rather small. Earlier studies of the lead isotopes focussed on astrophysics or basic nuclear physics and therefore sometimes lacked the focus on applications. Furthermore they did not employ all the presently available expertise on best measurement practices. Measurements with low neutron sensitive detectors (C_6D_6 scintillators) at 125 degrees were carried out at GELINA at the 60 m flight path with a highly enriched (99.82%) ^{206}Pb sample (Fig. 6). Capture yields resulted with 1 ns resolution up to 600 keV and capture areas were determined up to the maximum energy. Comparison with earlier work could only be made up to 150 keV and revealed systematic discrepancies that amounted to as much as 30%. The difference is understood as the combined effect of improper correction for neutron sensitivity and angular distribution and the neglect of sample dependent weighting function in earlier work [17].

Activation cross sections

Measurements of threshold reaction cross sections with the activation technique give access to (n,p), (n,a) and (n,2n) cross sections. Thus, for half lives from several seconds to many days, data are obtained, in an efficient manner, that are of relevance to hydrogen and helium production in structural materials, as well as radioactive isotope contributions to activity levels and decay heats in structural parts. The energy range between 14 and 20 MeV is of particular relevance, since there data are scarce whereas it is an important range for ADS [18]. At IRMM, many reactions have been studied recently in collaboration with ANL, FZJ, INRNE, U. Debrecen, KRI, NIPNE, NRG and NNDC. The systematics of $\Delta Z=1$ reactions was studied for isotopes of Cr, Fe, Ni, Mo, Pb and most recently Zr [19]. Such systematic studies are of importance in the benchmarking of model calculations. In addition to such studies, measurements are done for specific reactions of the NEA-NSC High Priority Request List and for reactions leading to well known important radioactive products such as in the case of recently finalised work for the ^{58}Ni(n,t)^{56}Co, ^{59}Co(n,p)^{59}Fe and ^{63}Cu(n,α)^{60m,g}Co reactions [20]. The IRMM Activation cross sections measurements constitute an important part of the work of Subgroup 19 of the WPEC [21].

Neutron data measurements for the Th/U fuel cycle

The Th/U fuel cycle has been proposed as the most effective means for elimination of minor actinides and for weapons grade plutonium. Furthermore, its use in energy generation is considered by countries with large resources of thorium ore. Large scale use of thorium in a reactor requires neutron data with accuracies similar to those for the conventional fuel cycle. This requirement has led to activities in the n_TOF-ND-ADS project, the French GEDEON program, and in Japan. Activities at ORELA/ORNL in the interest of this fuel cycle are motivated by the criticality safety program. To effectively channel these efforts to the user community the IAEA-NDS has started a Co-ordinated Research Project to provide a new library of up to date evaluations. At IRMM the ^{232}Th(n,γ) capture cross section [22] (Figs. 7 and 8) and the ^{231}Pa(n,f) [4,23], ^{233}Pa(n,f) [4,24], ^{233}U(n,f) [4], ^{234}U(n,f) [25] and the ^{236}U(n,f) fission cross sections are being studied or were recently completed. In addition, measurements were performed for the ^{236}U(n,γ) capture cross section in support of the n_TOF-ND-ADS project. This work was extensively reviewed at the Int. Conf. on Nuclear Data for Science and Technology, Santa Fe, NM, USA (2004) and at the Workshop on Neutron Measurements Evaluations and Applications, NEMEA-2, Bucharest, Romania (2004). See the above references.

Neutron data measurements for advanced concepts

Current trends for light water reactors and the study of Gen-IV concepts have in common the increasing emphasis on higher burnup. This results in requirements for improved fission product data accuracies (capture and transmission) and increased emphasis on the Pu isotopes. The latter feature is augmented by scenarios that foresee multiple recycling of Pu.

Improving the data for the most important fission products, is an exercise in high precision measurements. Recently, ^{103}Rh capture and transmission measurements were performed at IRMM in collaboration with CEA. The thermal capture cross section was measured to better than a percent and an improved capture resonance integral was determined from measurements for the lower portion of the resolved resonance range. The new results improve agreement with the benchmarks and the new resonance region evaluation based on these measurements will be joined with a new evaluation above 100 keV for the future JEFF3.1 library [26].

For ^{240}Pu and ^{242}Pu, high precision transmission measurements were performed at 15, 77 and 300 K for the first few resonances with the aim to study the Doppler broadening mechanism. These measurements are part of the n_TOF-ND-ADS shared cost action [27].

Conclusion

A considerable number of measurements have been performed or are underway at IRMM with the aim to support studies in nuclear waste transmutation and advanced concepts that aim at sustainability of nuclear power. An overview was presented with a number of illustrative examples. A considerable part of the work is performed in close collaboration with laboratories from the EU, Candidate Countries, USA, Japan and Russia. From 2005 onwards, use by external parties of the IRMM neutron sources will be facilitated through a grant in the EURATOM "Transnational Access to Large Scale Facilities" program. The IRMM program will continue to look for possibilities to support the nuclear energy program in areas of waste minimisation, safety and sustainability. The direction of future activities will be addressed in the "International Workshop on Nuclear Data Needs for Generation IV Nuclear Energy Systems", Antwerp, Belgium, April 5-7, 2005 (www.jrc.cec.eu.int/gen4-workshop).

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Fig. 1 The Gelina flight path area.

Fig. 2 The Van de Graaff laboratory.

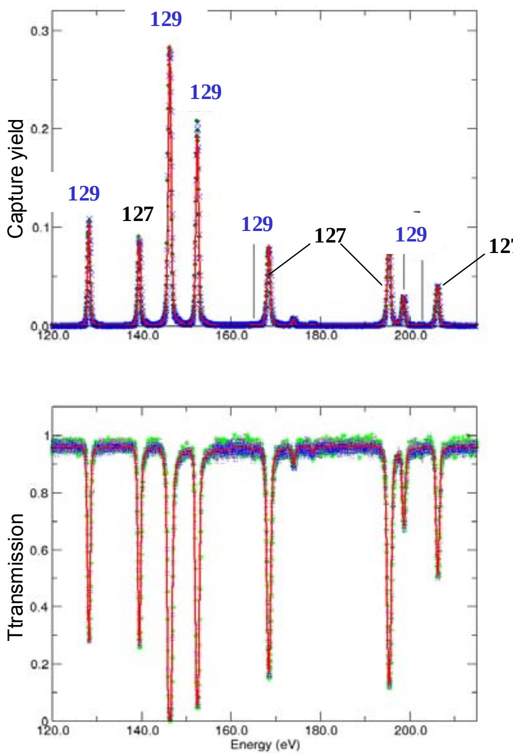

Fig. 3. Detail of the resolved range capture and transmission measurements for ^{129}I.

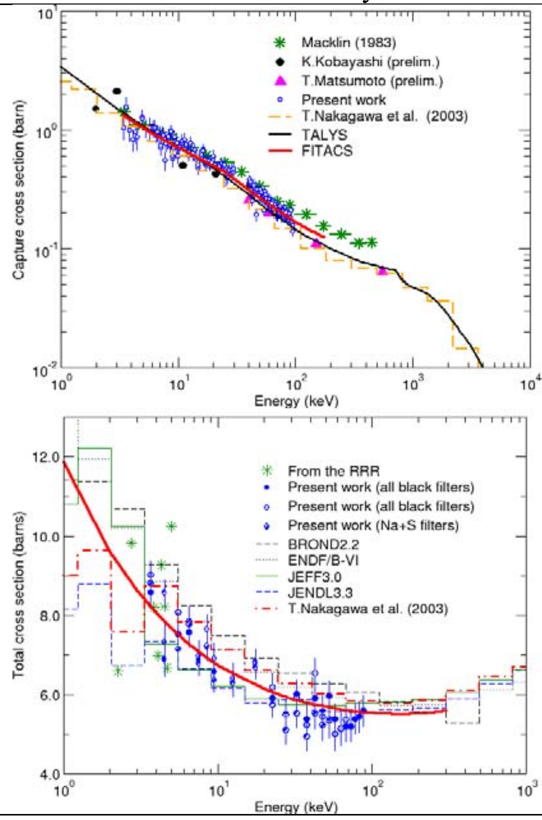

Fig. 4. Unresolved resonance range capture and transmission measurements for ^{129}I. Comparison with existing data, evaluations and the new analysis with the FITACS code.

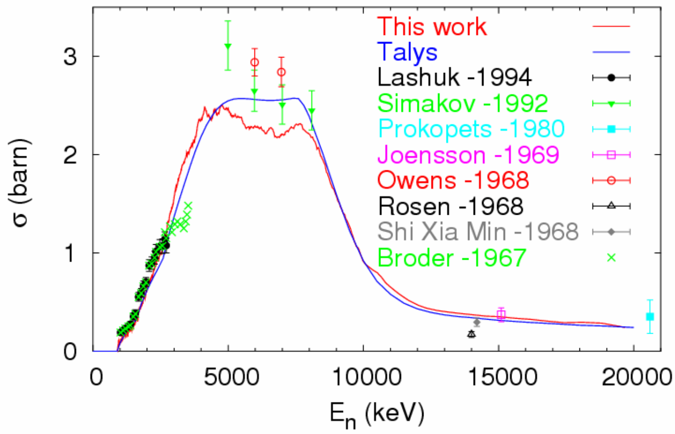

Fig 5. Results of measurements of the ^{209}Bi(n,n’) cross section with the (n,n’g) technique. Comparisons with earlier measurements and Talys are presented.

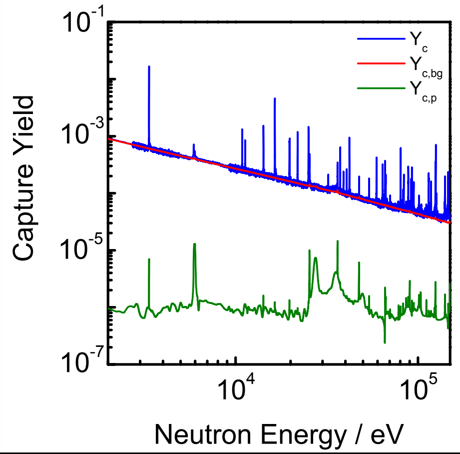

Fig. 6. Measurements for the ^{206}Pb(n,γ) cross section. Y_c shows the measured capture yield, $Y_{c,bg}$ the background and $Y_{c,p}$ the prompt background from by scattered neutrons.

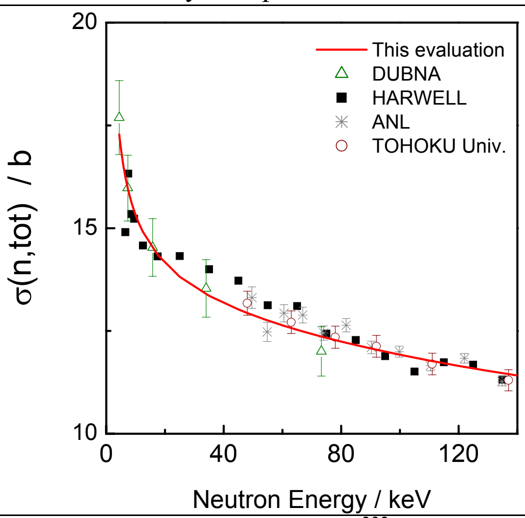

Fig. 7. URR evaluation for the ^{232}Th total cross section.

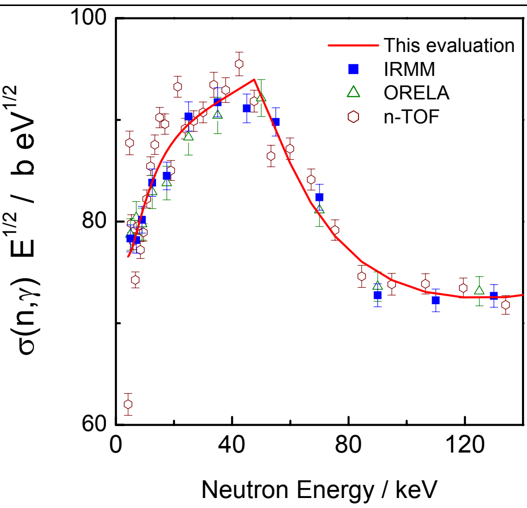

Fig. 8. URR evaluation for the capture cross section on ^{232}Th.