EUROTRANS/NUDATRA: LOW AND INTERMEDIATE ENERGY NUCLEAR DATA MEASUREMENTS

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The Integrated Project on European transmutation (EUROTRANS) concerns research and development for the design of an accelerator driven system (ADS) dedicated to the transmutation of high-level nuclear waste. Domain 5 on nuclear data for transmutation (NUDATRA) consists of four workpackages. Work package 2 on "Low and intermediate energy nuclear data measurements" investigates neutron-induced inelastic and (n,xn) reactions of lead and bismuth, the neutron capture reaction of ^{243}Am and the neutron-induced fission reaction of ^{244}Cm. These reactions were selected on the basis of sensitivity studies performed by Aliberti et al., that indicated their importance for ADS design. An overview will be presented by the workpackage coordinator of measurements that were completed (lead and bismuth) and the status of the data analysis and evaluation for the remaining work..

I. INTRODUCTION

I.B. International collaboration

The European Commission (EC) consistently emphasises the importance of research and development for viable technical solutions that may reduce the amount of high-level nuclear waste. In Framework Program 5 this resulted in considerable emphasis on nuclear data measurement programs to improve the reliability of calculations for various waste minimisation scenarios and the envisioned innovative accelerator driven transmuters. The succesful HINDAS project was dedicated to high and intermediate energy nuclear data for accelerator driven systems and in particular the spallation target and its immediate vicinity.[1] [a] The N_TOF_ND_ADS project developed a CERN-based neutron time-of-flight facility for the study of neutron data for accelerator driven systems.[2] [b]

In Framework Program 6 (2003-2006), the integrated project EUROTRANS emphasises the development of a design for an accelerator driven transmutation (test) facility.[3] The nuclear data efforts are concentrated in the domain NUDATRA and aim at improved data and simulation tools for reliable performance estimates of the considered designs through new measurements, evaluations, and data validation.[c] The impact of nuclear data uncertainties on design- and fuel cycle parameters are investigated through sensitivity calculations. NUDATRA consists of four workpackages: 1) Sensitivity analysis and validation of nuclear data and simulation tools, 2) Low and intermediate nuclear data measurements, 3) Nuclear data libraries evaluation and low-intermediate energy models, 4) High energy experiments and modeling. Here, the status of the efforts related to Work Package 2 (WP2) is summarised.

Further to these nuclear data efforts, three additional EC sponsored projects are currently under way in the domain of nuclear waste. Transnational access is provided to the Insitute for Reference Materials and Measurements (IRMM) neutron data measurement facilities (NUDAME) at the Geel Electron Linear Accerator (GELINA) and the Van de Graaff laboratories.[d] Beam time is offered to external users who wish to perform measurements relevant to the domain of nuclear waste. Experiments that are approved by the program advisory committee (PAC) are in to the final year of this three years program which will end March 31, 2008. In the integrated infrastructure initiative European Facilities for Nuclear Data measurements (EFNUDAT) nine laboratories combine efforts to facilitate and improve nuclear data measurements in the low and intermediate energy range through networking, transnational access to nine facilities through one PAC and joint research activities.[4e] Finally the Coordination Action on Nuclear Data for Industrial Development in Europe (CANDIDE) aims at establishing a durable network on nuclear data efforts in the domain of minimising high-level nuclear waste.[5]

[a] Website: www.fynu.ucl.ac.be/collaborations/hindas/
[b] Website: pceet075.cern.ch
[c] Website: fachp1.ciemat.es/nudatra, username: nudatra, password: pbmahe
[d] Website: www.irmm.jrc.be
[e] Website: www.efnudat.eu

I.B. Low and Intermediate Energy Nuclear Data Measurements

Work package 2 on low and intermediate energy nuclear data measurements is a concentrated effort of new measurements for certain key nuclear data. The measurements were selected on the basis of an ADS sensitivity study by Aliberti et al., that analysed the impact of nuclear data uncertainties on key reactor parameters. In fact, this study established target uncertainties for nuclear data on the basis of target uncertainties for key parameters characterising the performance of a reference ADS dedicated to transmutation of waste. This elaborate study identifies isotopes, reaction channels and energy domains. The nuclear data target accuracies are identified by a minimisation process that guarantees the uncertainties on the target uncertainties for reactor parameters are met through constraints and at the same time weighs the relative difficulty of the various data improvements. Desired nuclear data uncertainties are confronted with estimated uncertainties based on estimated covariance matrices. [6]

The efforts are organised in three main tasks. The first concerns nuclear data for lead and bismuth, the second fission of actinides and the third the capture cross section of ^{243}Am.

The first task of measurements for lead and bismuth has four subtasks. The first subtask is the measurement of lead and bismuth inelastic scattering cross sections with the (n,n'γ)-technique and an accuracy meeting the requirements of reference 6, which is better than 8-10% in the range from threshold to 6 MeV. Complementary measurements are performed on the same isotopes for the (n,2n)-channel using the (n,xnγ)-technique for energies up to 20 MeV. These measurements allow to benchmark model calculations, so that the desired accuracies of 15-20% for predictions of these cross sections can be checked. At higher energies little is known about (n,xn) reactions in the lead region. Within the HINDAS project the elastic scattering cross section was studied at 100 MeV neutron energy for natural lead using the SCANDAL facility.[7] The same setup extended with a second detector arrangement for the measurement of low energy neutrons was used to study the (n,xn) reactions at 100 MeV. At these energies mass and energy dependencies of cross sections are moderate and relatively easy to model, so that a single benchmark experiment is an important contribution to the general understanding of this process at intermediate energies.

In a lead or lead/bismuth cooled ADS, bismuth is present either as a main element of the lead-bismuth eutectic or as a minor element growing-in the lead coolant following neutron capture on ^{208}Pb. In either case, the neutron capture reaction on ^{209}Bi is an important contributor to the production of radioactivity in the coolant. The capture reaction produces ^{210}Bi either in its ground state or in an isomeric state. The ground state is short-lived and decays to ^{210}Po, a well known alpha-emitter of relatively short half life that may constitute a radiation hazard for opertors of an ADS. The long-lived isomer is an alpha-emitter that should be condidered as contributing to radioactive waste. In this project, the isomer ratio characterising the production of the ground state relative to that of the isomer is studied at the GELINA facility.

The second task concerns the determination of the cross section for neutron-induced fission on ^{244}Cm. To this end a measurement of the fission probabilty of ^{245}Cm is performed by means of the (^{3}He,pf) transfer reaction on ^{243}Am. The experimental work is carried out at the tandem accelerator of IPN-Orsay in France. Requirements for improved cross sections for neutron-induced fission on ^{241}Am, ^{243}Am, ^{244}Cm, and ^{245}Cm were identified in reference 6.

Of similar importance is the study of neutron-induced capture reactions on minor actinides, and in particular ^{241,243}Am. The third subtask concerns the measurement of ^{243}Am at N_TOF and more information can be found in reference 2.

Work package 2 is a collaborative effort of European nuclear data research laboratories with contributions from the IRMM in Belgium, CNRS and CEA in France, CIEMAT and CSIC in Spain, the University of Uppsala in Sweden, INFN in Italy, FZK in Germany and the Technical University Wien in Austria.

II. STATUS

II.A. Measurements for lead and bismuth

II.A.1. Inelastic scattering of lead and bismuth

Measurements for ^{206,207,208}Pb and ^{209}Bi neutron inelastic scattering with the (n,n'γ)-technique were carried out at a 200 m station of the neutron time-of-flight facility at GELINA. Neutrons were produced by (γ,xn) and (γ,F) reactions induced by bremsstrahlung photons on a metallic rotary uranium target. The bremsstrahlung results from the impact on the same target of an intense pulse of electrons of about 100 A peak current and less than 1 ns full width at half maximum pulse duration.. The useful neutron spectrum ranges from the threshold of the inelastic reactions up to about 20 MeV for strong reaction channels.

The measurements contributed to and supported by EUROTRANS/NUDATRA use the methodology that was described elsewhere and was applied to ^{58}Ni and ^{52}Cr[8,9]. Here the gamma-rays associated with the (n,n') process are detected and gamma-ray production cross sections are measured with high accuracy and very good neutron energy resolution. From these primary results the total inelastic cross section and the cross section for the excitation of specific levels are deduced. The achieved accuracy meets the goals set in the paper of Aliberti et al.[6], and should thus be adequate for the sought-for improvements in ADS modeling. Measurements were conducted with various upgrades of the detection system which involved the employment of additional high-purity germanium (HPGe) detectors of 100% relative efficiency up to a total number of four. Further upgrades concern the data-acquisition system. Here a new digitizer based system was compared in detail with a conventional data-acquisition system in case of the study of ^{206}Pb with favorable results for the former in terms of dead time and flexibility.[10] For more details consult references 11 and 12.

II.A.2. Lead and bismuth (n,xnγ) reactions below 20 MeV

As part of the N_TOF_ND_ADS project mentioned above, test measurements were carried out at IRMM by the n_TOF collaboration to optimise the data-acquisition system for a HPGe based setup. Digitizers developed in-house at IReS Strasbourg were used in combination with the setup above and it was demonstrated that (n,2nγ) cross sections could be measured at IRMM. This was shown for ^{207}Pb.[13] Subsequently in NUDATRA further measurements were carried out for the other isotopes and more elaborate results were obtained for ^{207}Pb. In view of the rather limited neutron intensity in the energy range concerned, results were obtained for a fair number of gamma-rays for each nucleus: 2 transitions for ^{206}Pb, 4 for ^{207}Pb, 5 for ^{208}Pb and 8 for ^{209}Bi. In addition results were obtained for one transition of the ^{208}Pb(n,3n) reaction. For more details consult reference 11.

II.A.3.The Pb(n,xnγ) reaction at 100 MeV

Measurements have been performed at the Svedberg laboratory (TSL) of the University of Uppsala using quasi mono-energetic neutrons of 96 MeV produced by the ^{7}Li(p,n) reaction. Secondary neutrons were detected in a two-armed setup. The first arm detects neutrons with energies between 30 and 100 MeV using the CLODIA and SCANDAL detectors, while the second detects neutrons in the range from 2 to 50 MeV with a plastic and a liquid scintillator called DECOI and DEMON. Clodia is a proportional counter loaded with polyethylene converters. Recoil protons are generated in the converter by elastic scattering for which Clodia determines the point of origin and initial direction. The full energy and further direction information of these protons is determined by the SCANDAL detector that consists of plastic ΔE detectors and CsI full energy detectors. This measurement includes both elastically and inelastically scattered neutrons. DECOI is a plastic scintillator that detects protons recoiling from small angle neutron scattering. The incident neutron proceeds to the DEMON liquid scintillator and the neutron's energy is determined from the time-of-flight between the DECOI and the DEMON detector. Measurements with the two arms have a range of overlap allowing a continuous range of measurement for emitted neutrons from 2~MeV to 100~MeV. For both detector arms a considerable amount of work concerns the elimination of background.

Double differential neutron emission cross sections were determined for five angles in the range from 15^{o} to 98^{o}. The contribution from the evaporation process was determined at 98^{o} and was found to be consistent with the Weisskopf-Ewing estimate. It was verified that this contribution is essentially angle independent. Contributions of elastic scattering were estimated and found to be in good agreement with earlier work. Subtracting elastic scattering and evaporation from the double differential data gave an estimate of pre-equilibrium contributions to the (n,xn) process. These contributions were integrated over the angles by fitting them to a functional form proposed by Kalbach. For the angle integrated result comparisons were made with various model calculations using the GEANT3 and MCNPX Monte Carlo codes for particle transport in combination with the reaction model codes GHEISHA, FLUKA, GNASH, INCL4-ABLA, TALYS, and DYWAN. All calculations are below the experimental data.[14]

II.A.4.The isomer ratio for the ^{209}Bi(n,γ) reaction

Figure 1 schematically shows the two decay paths following neutron capture on ^{209}Bi which were mentioned in the introduction. In view of the long half life of the isomer a measurement of the isomer ratio by the activation technique is extremely difficult.

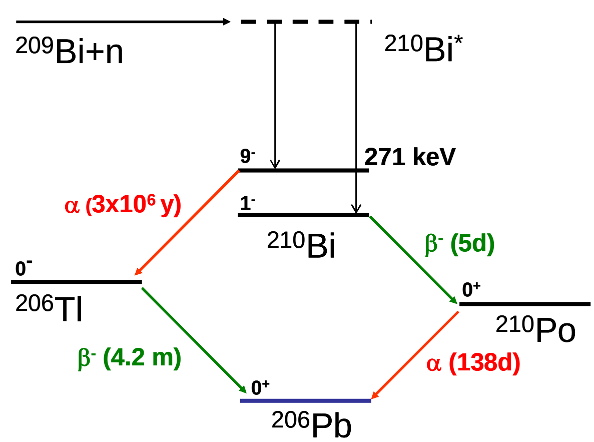

Fig. 1. Schematic decay scheme for neutron capture by ^{209}Bi.

Therefore, a capture measurement was carried out at the GELINA facility with the added advantage that data may be obtained for the resonance dependence of the ratio. The low excitation energy of the isomer of 271 keV demands the use of high resolution gamma-ray detectors to distinguish the feeding of the isomer from the feeding of the ground state. It is easily understood that a resonance dependence may occur as a result of the difference in resonance spins which affects the preferred decay paths of which there are several both feeding the ground state and the isomer. For the current measurements moderated neutrons were used which were obtained at a short flight path focussed at two water moderators placed above and below the above mentioned rotary target at GELINA. Despite the excellent gamma-ray energy resolution for HPGe considerable efforts were required to achieve a low enough background on account of the high neutron sensitivity of germanium. In addition it was shown that resonance self shielding in combination with gamma-ray self-shielding of the target leads to significant corrections to the detection efficiency that are resonance dependent. Preliminary results were obtained for six resonances and three groups of resonances in the incident neutron energy range from 0.8 to 20 keV. These results show that the isomer ratio, defined as the ratio of the cross section for the production of the ground state to that of the isomer, varies from about 0.5 to about 7. The observed behavior changes from resonance to resonance. This contrasts the smooth behavior predicted by certain model calculations. The resonance dependence is illustrated by the gamma-ray spectra shown in Fig. 2. Two resonces are shown for which the isomer ratio differs by a factor of five. For more details see reference 15.

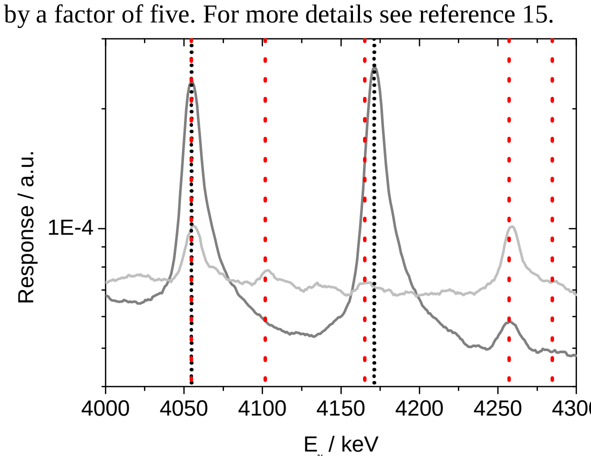

Fig. 2. Selected portion of the gamma-ray spectrum for two neutron resonances of ^{209}Bi. The light shaded line is for the 2.3 keV resonance with $J^{\pi}=5^{-}$, while the dark shaded line is for the 0.8 keV resonance with $J^{\pi}=4^{-}$.

II.B. Measurements for minor actinides

II.B.1. Fission studies

Accurate direct neutron-induced fission cross section measurements for Cm isotopes are very difficult. Good quality fission deposits are hard to come by and for short-lived isotopes like ^{242}Cm (163 d) and ^{244}Cm (18y) activities are beyond current day measurement capabilities. The group at CENBG, Bordeaux and collaborators have developed a method to measure the fission probability of fissioning nuclei by transfer reactions of the type $^{A}Z(^{3}He,xf)$.[16] Here x (=p, d, t, ^{3}He, or ^{4}He) is a light charged particle that is emitted prior to fission. If we denote x as ^{a}z, the light particle-tagged fission rate divided by the light particle singles rate is the fission probability P_f of the nucleus $^{A-a}(Z-z)$. Now all that remains is an accurate prediction for compound nucleus formation for the n+$^{A-a-1}(Z-z)$ system to obtain the corresponding fission cross section. The formation cross section is primarily determined by the optical model for neutrons on the target nucleus. This optical model is well developed by colleagues at CEA.

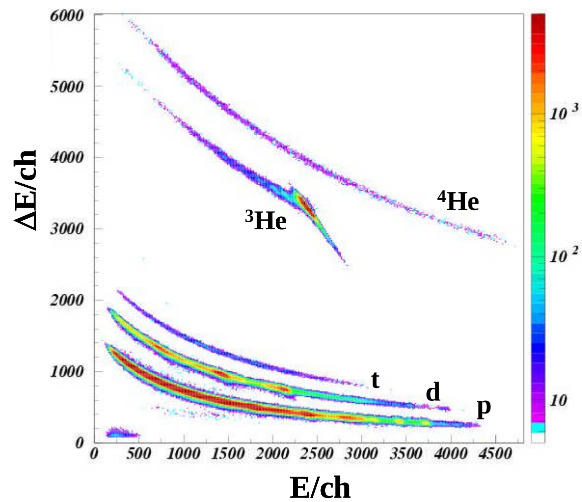

Fig. 3. Light charged particle identification.

The measurement proposed in the context of EUROTRANS concerns the neutron-induced fission cross section on ^{244}Cm. The corresponding fission probability of ^{245}Cm is measured using the $^{243}Am(^{3}He,pf)$ reaction. The IPN Orsay tandem Van de Graaff was used to provide ^{3}He beam at 30 MeV bombarding energy. ^{243}Am targets have been used for two series of measurements. They were approximatively 100µg/cm^{2} thick deposited on to a 75µg/cm^{2} carbon foil. A single experiment results in data for four different reaction channels. The identification of the four reaction channels is done with four ΔE-E telescopes placed respectively at 90° and 130° relative to the ^{3}He beam axis (Fig. 3). The fission fragment detector system was designed to achieve a large geometrical efficiency (48%) and good granularity for fission fragment angular distribution. The arrangement consists of 15 photovoltaic cells (20x40 mm^2) distributed among 5 units, each unit consisting of 3 cells, the central cells are located at 5 cm from the target. Four units were placed in the forward direction with a covering angle from 14° to 125°. The last unit was placed at 180° relative to the foremost one, it adds one more point to the backward angular distribution measurements.

The method is illustrated in Fig. 4 where the singles and fission coincidence data are shown for the ^{243}Am(^{3}He, tf) reaction, that corresponds to the ^{242}Cm(n,f) reaction. It should be noted that although the data for four channels are obtained simultaneously, the analysis and complications for each channel may be different. In particular, for the reaction proposed as a contribution to NUDATRA an initial experiment showed an anomalously large proton singles rate. This was attributed to a possible contamination of the target and a somewhat too high incident ^{3}He energy. A second experiment was carried out in November 2006 with a new ^{243}Am target. The analysis of this experiment is in progress.

An active contributor to the study of fission cross sections on minor actinides, the group at CENBG is also involved in direct neutron-induced cross sections measurements for the 243Am(n,f) reaction using the NUDAME transnational access scheme to perform measurements at IRMM.[17]

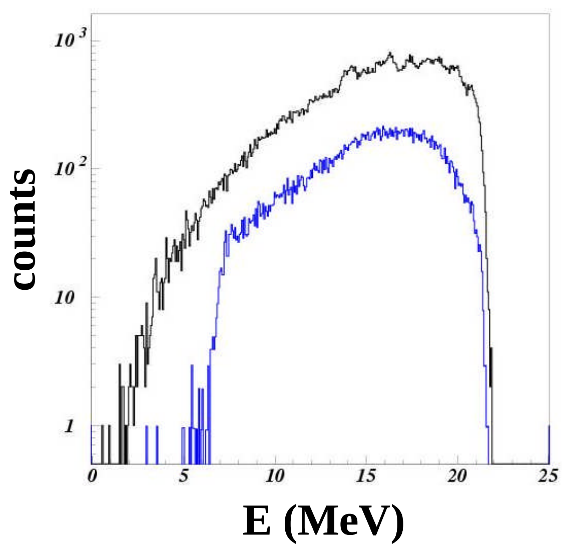

Fig. 4. Tritium particle singles yields (upper curve) and fission coincidence yields (lower curve) for the ^{3}He+^{243}Am reaction.

II. CONCLUSIONS

In the reference section below, Refs. 2, 3, and 4 provide examples of the formats for books, journal papers, and proceedings papers, respectively. Listing paper titles is not mandatory; however, it is encouraged as an additional help to readers.

II.B.2. The 243Am(n,g)244Am reaction

Measurements are planned at the n_TOF facility which is based at the CERN Proton-synchrotron. These measurements are required to improve upon the results obtained by the n-TOF collaboration in the above mentioned n_TOF_ND_ADS project of Framework Program 5. In particular, the energy range should be extended by means of a reduction of the background at the facility. To this end a new sealed sample design has been prepared. Measurements depend on a refurbishment of the neutron-producing target at CERN. These works are currently being planned and may be completed in 2007. In any event no new measurements are expected prior to the spring of 2008. For further details see reference 2.

III. SUMMARY

The activities of workpackage 2 of the domain NUDATRA of IP EUROTRANS are well under way. Planned measurements of the cross sections for lead and bismuth are nearing completion and many results have already been transferred to the evaluators of NUDATRA workpackage 3. The situation for the study of neutron-induced fission cross sections on Cm isotopes is progressing very well. For the study of the ^{243}Am(n,γ) reaction the collaboration awaits the refurbishment of the n_TOF facility at CERN.

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