NEUTRON INDUCED REACTION STUDIES AT THE ACCELERATOR-BASED NEUTRON SOURCES OF IRMM

Arjan Plompen

European Commission, Joint Research Centre, Institute for Reference Materials and Measurements: Retieseweg 111, 2440 Geel, Belgium, Arjan.Plompen@ec.europa.eu

The Neutron Physics unit of the IRMM performs neutron-induced reaction studies at two accelerator-based neutron sources. At GELINA, the Geel Electron LINear Accelerator, a pulsed white neutron source for high-resolution neutron time-of-flight measurements is used for studies of neutron-induced total, capture, inelastic, fission and charged-particle-emission cross sections. At the 7MV CN Van de Graaff accelerator, quasi mono-energetic neutron fields are produced in the range from 0.1-20 MeV for studies of neutron-induced fission, activation and charged-particle-emission reactions. Reaction studies are performed in the interest of nuclear waste management, reactor safety, neutron standard cross sections and basic nuclear physics. This includes measurements in the interest of Generation-IV and accelerator driven systems. An overview of recent work is presented.

I. INTRODUCTION

I.A. General

The Institute for Reference Materials and Measurements (IRMM) located in Geel, Belgium is part of the Joint Research Centre, a Directorate General of the Commission of the European Communities of the European Union.[a] A multidisciplinary institute, the IRMM has five research units, Reference Materials, Isotopic Measurements, Food Safety and Quality, Scientific Quality and Strategy and Neutron Physics. The mission of the Neutron Physics unit is to provide European safety authorities and industry with neutron reaction data for the safety of nuclear installations and the nuclear fuel cycle and the feasibility studies and development of waste transmutation facilities.

The neutron physics unit is organized in two projects (actions). Action 51401 ND-STDS "Basic research in nuclear physics and nuclear data standards"[b] and Action 51402 ND-MINWASTE "Nuclear data for radioactive waste management and safety of new reactor developments".[b] These two projects involve neutron-induced reaction studies at two accelerator-based neutron sources at IRMM. At the Geel linear electron accelerator laboratory GELINA neutron-induced reactions are studied using the time-of-flight technique at a very high energy resolution for incident neutrons in the range from 10 meV to 20 MeV. At the 7 MV CN Van de Graaff accelerator quasi mono-energetic neutrons are produced with energies from 0.1 to 20 MeV with well-known binary source reactions ^{7}Li(p,n)^{7}Be, ^{3}H(p,n)^{3}He, ^{2}H(d,n)^{3}He, ^{3}H(d,n)^{4}He.

I.B. International collaboration

International collaboration in nuclear measurements is essential for the development of new initiatives of the unit that are in the interest of the users of nuclear data in Europe. It guarantees the timely integration of the new measurements in data libraries used in applications and ensures that the projects of the unit are timely and of interest. The Neutron Physics Unit participates actively in international collaborations at the European level through participation in nuclear data activities organized by the European Commission (DG-RTD). These include participation in the integrated project on European transmutation (EUROTRANS),[1] [c] the integrated infrastructure initiative European Facilities for nuclear data for nuclear waste transmutation (EFNUDAT),[2d] the coordination action on nuclear data for industrial development in Europe (CANDIDE),[3] and the transnational access scheme for nuclear data measurements (NUDAME).[a] The unit furthermore participates in the development of the Joint Evaluated Fission and Fusion file maintained at the OECD Nuclear Energy Agency (NEA) and contributes to international projects of the Working Party on Evaluation Cooperation

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[a] Website: www.irmm.jrc.be

[b] Website: projects-2007.jrc.ec.europa.eu (IRMM)

[c] Website: fachp1.ciemat.es/nudatra, username: nudatra, password: pbmahe

[d] Website: www.efnudat.eu

(WPEC) of the NEA.[e] Similarly the unit has contributed and continues to do so in coordinated research projects (CRPs) of the Nuclear Data Section of the International Atomic Energy Agency.[f]

Besides these broader international cooperative efforts several bilateral collaborations exist with e.g. CEA, CNRS, INFN, CIEMAT, FZK, IKI Budapest. Another example is the participation with several actions in the collaboration agreement between the JRC and the Department of Energy (DOE). These activities involve collaborations with LANL and ORNL on fission cross section measurements, measurements for criticality safety and for detector characterization.

The above mentioned NUDAME project[g] offered support to external users of the IRMM neutron sources under a transnational access scheme of DG RTD. It ran for three years in the form of annual call for proposals which was evaluated by a program advisory committee. The large interest in the use of the IRMM facilities manifested itself in a three times higher demand on the available beam time than could be allocated under this scheme.

I.C. The neutron facilities

At GELINA, neutrons are produced by (γ,xn) and (γ,F) reactions on depleted uranium.[4,5,6]. The gamma-rays result from bremsstrahlung produced by an electron beam impacting the uranium target. Under typical operating conditions a pulsed beam is used with 800 Hz repetition rate, pulses of less than 1 ns full width at half maximum, an average current of 75 μA and mean electron energy of 100 MeV. To extend the time-base lower repetition rates may be used, e.g. 50 Hz. For power dissipation the uranium target is a slowly rotating annular disk cooled by mercury. The spectrum of neutrons emitted by the uranium (direct spectrum) consists of evaporation and fission neutrons peaking between 1 and 2 MeV and falling of exponentially, above. To extend the useful energy range down to about 10 meV, Be water tanks are placed above and below the uranium target to moderate the neutrons (moderated spectrum). Flight paths with lengths varying from 8 to 400 m view the neutron producing target. Typically the spectrum in a particular flight path is limited to either the moderated or the direct spectrum by appropriate shadow bars and collimators. Dedicated equipment is available for transmission measurements (two flight paths w. Li-glass detectors), capture measurements (three flight paths, C_6D_6 and HPGe detectors), neutron inelastic and (n,2n) cross sections (two flight paths, HPGe detectors), light charged particle measurements (2 flight paths, ionization chambers and Si telescopes) and fission measurements (2 flight paths, ionization chambers, telescopes and scintillators). Typically, these measurements are running in parallel on different flight paths.

At the Van de Graaff laboratory, six beam lines are available for different measurement arrangements. Four of these may be used for any of the above mentioned source reactions while a fifth is reserved for use with non radioactive targets, only. Solid targets are used for all reactions, but for the $^{2}H(d,n)^{3}He$ reaction a deuterium gas-cell may be used as well. Solid tritium and deuterium targets consist of an 0.5 mm silver backing with an evaporated layer of titanium (frequently 2 mg/cm^2) that is impregnated with tritium or deuterium gas to an atom ratio of about 1.7 (^{3}H or ^{2}H)/Ti. Proton and deuteron currents are in the range of a few to a few tens of microamperes. Current measurement projects involve studies of neutron-induced fission, light charged-particle emission and activation, as well as neutron dosimetry and dosimeter characterization. Fission studies concern the search for a fission isomer in ^{235}U, the determination of the ^{235}U fission neutron spectrum at 0.5 MeV and the study of subthreshold fission resonances for ^{238}U. For light charged particles results were obtained for the ^{10}B(n,α) reaction and work is ongoing for the ^{16}O(n,α) reaction. Activation cross sections were obtained for isotopes of Cr, Ni, Cu, Zr, Ta and W.

Below a selection of recent results is presented.

II. SELECTED RESULTS

II.A. Neutron inelastic scattering

Accurate results for neutron inelastic scattering on important reactor materials are important for accurate determinations of reaction rates and in particular criticality estimates. For fast reactors such as accelerator driven systems and Generation-IV concepts, the uncertainties of reactor parameters such as k_{eff} and the void coefficient show large contributions from the uncertainties in the data for inelastic scattering.[7,8] This is almost entirely due to the fact that the neutron spectrum hardens or softens when the probability for inelastic scattering lowers or increases. In a fast system, elastic scattering lowers a neutron's energy from a fraction of to a few percent, while inelastic scattering removes between about 50 and 1000 keV. Therefore, almost any neutron reaching the slowing down range scattered inelastically, at least once. Since several sensitivity studies pointed towards the importance of inelastic scattering for reactor concepts relevant to waste minimization, at IRMM a setup was developed to determine such cross sections accurately for non-deformed isotopes.

Figure 1 shows the setup currently available on Flight Path 3 at 200 m from the neutron producing source of the GELINA laboratory. Eight high purity germanium

[e] Website: www.nea.fr
[f] Website: www-nds.iaea.org
[g] Website: www.irmm.jrc.be/html/nudame

(HPGe) detectors are used to determine gamma-production cross sections with high incident energy resolution and selectivity for transitions from levels excited by inelastic scatering or (n,2n) reactions. Seven of these detectors have a relative efficiency of 100%. The detectors are placed at 110 and 150 degrees to allow a very good angle integration for gamma-ray multipolarities up to 3 hbar. Data acquisition is based on the DC440 digitizer card of ACQIRIS with 2.4 ns sampling time and 12 bits amplitude resolution. The present state of the setup was reached earlier this year and a first set of measurements was recently completed for natural silicon. Half the number of the detectors shown were used for the measurements on iron described below.

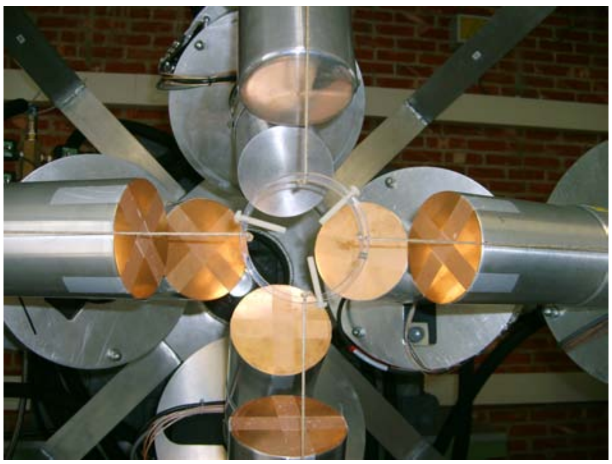

Fig. 1. The new setup of eight HPGe detectors at Flight Path 3, 200 m from the neutron source.[9]

In 2006 considerable time was spent on a second measurement of ^{206}Pb with a radiogenic enriched sample on loan from Oak Ridge National Laboratory (ORNL) using a predecessor of the present setup. The sample differed in size, thickness and isotopic composition from that used in an earlier measurement. In particular, the first sample (50 mm Ø, thickness 2 mm, enrichment 99.82%) had a diameter smaller than the neutron beam (60 mm), while the latter (70 mm Ø, thickness 5 mm, enrichment 88.5%) had a diameter larger than the beam. It may be noted that this seemingly trivial consistency check, involved substantial differences for flux normalization, neutron self attenuation and multiple scattering corrections, as well as gamma-ray self attenuation, angular and spatial distribution corrections. In addition a conventional data acquisition system was replaced by a system based on the above mentioned digitizers, implying differences in dead time and the energy dependent gamma-ray efficiency. Remarkably, the agreement between the two measurements was within 1 to 2%, aside from the observed statistical bin-to-bin fluctuations above E_n=2 MeV, confirming to a high degree the reproducibility of the results (Fig. 2).

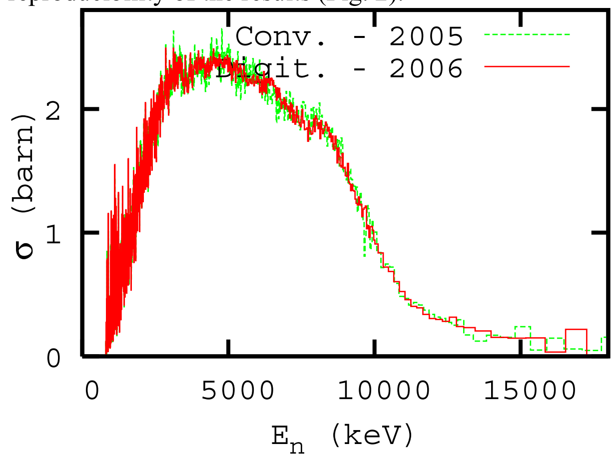

Fig. 2. Gamma production cross section for the 803 keV transition following neutron inelastic scattering on ^{206}Pb. Results are shown for two different experiments.

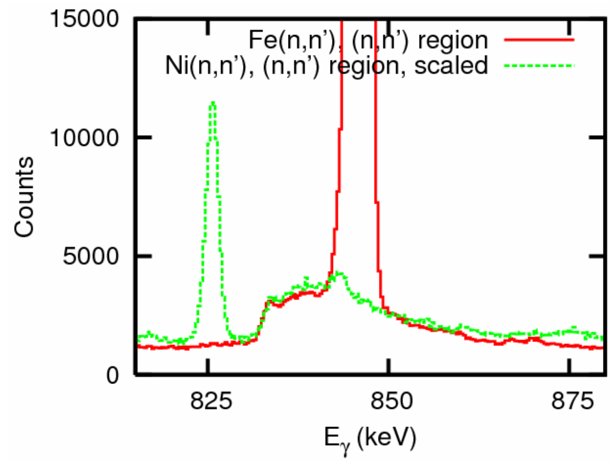

Fig. 3. Gamma-ray energy spectrum. The full line is for a sample of natural iron. The dashed line is for a sample of natural nickel.

Results were obtained for ^{56}Fe inelastic scattering using a natural sample of 80 mm diameter, 3 mm thickness and 99.5% elemental purity. The four detectors were mounted in the frame shown in Fig. 1, two at each of the two angles. Data were taken for five weeks. As previously, the neutron flux was determined using a ^{235}U fission chamber and the cross section from the new standards evaluation. The main transition excited in inelastic scattering is the decay of the first excited level at 847 keV. It carries nearly all of the inelastic scattering cross section and was observed in the range from threshold to 18 MeV. Above 7.8 MeV the (n,2n) reaction on ^{57}Fe (natural abundance 2.12(1)%) contributes to the intensity of this transition. A potentially important correction to the gamma production cross section of the 847 keV transition is the gamma-ray of 844 keV from inelastic scattering on ^{27}Al. The end caps of the Ge detectors and part of the support structures are aluminum and neutrons scattered on the sample could produce a contribution that would be difficult to resolve from the transition of interest. Fig. 3 shows that the problem is well under control. An independent measurement using a natural nickel sample as a scatterer, normalized for flux and number of atoms shows that the background from aluminum is nearly completely negligible. The high resolution of the present measurements is reflected by the resonance structure just above threshold (Fig. 4).

It may be noted that (n,2nγ) cross sections may be obtained using the same setup with very similar data analysis procedures and whenever possible such results are extracted from the data. Currently, a second setup is under development on Flight Path 16 at 30 m from the neutron target by a group from IN2P3/CNRS in Strasbourg, France. The intention is to use this setup for the study of ^{235}U and ^{233}U.

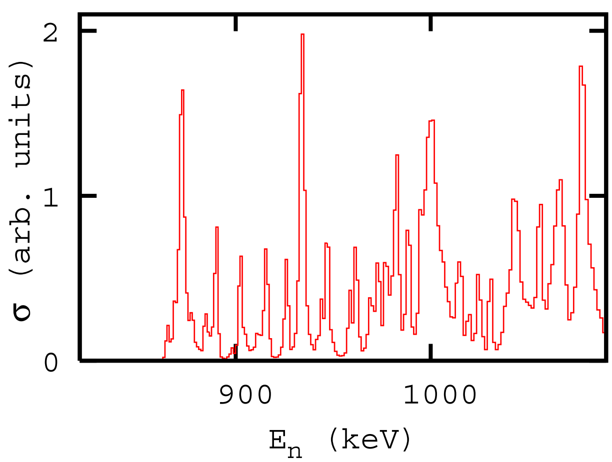

Fig. 4. Gamma production cross section for the 847 keV transition following neutron inelastic scattering on ^{56}Fe just above threshold.

II.B. Neutron capture by bismuth

Natural bismuth is a mono-isotopic element that currently receives a great deal of attention as a potential (major or trace) component of the coolant of an accelerator driven system. Neutron capture by ^{209}Bi leads to the unstable ^{210}Bi either in the ground state (5d) or in an excited state (3 10^6 y). The ground state decays to ^{210}Po (138 d), which subsequently emits an alpha and constitutes a radiation hazard. The isomer decays by alpha emission and contributes to radioactive waste. At IRMM the isomer ratio of radiative neutron capture by ^{209}Bi is studied from 0.8 to 20 keV at a short flight path using the moderated neutron spectrum and HPGe detectors to distinguish the population of the ground state from that of the isomer.

Complementary to the work carried out for the isomer, capture using C_6D_6 detectors and total cross section measurements are carried out to improve the parameters describing these resonances in ^{209}Bi. These measurements are both needed as the resonance area in the total cross section is proportional to $g\Gamma_n$, where g is the statistical factor and Γ_n the neutron width, while the resonance area in the capture cross section is proportional to $g\Gamma_n\Gamma_g/\Gamma$, where Γ_g is the gamma width and Γ the total width ($\Gamma=\Gamma_n+\Gamma_g$). Since for bismuth the neutron width is much larger than the gamma-width the capture area is proportional to $g\Gamma_g$. Two transmission measurements were carried out with 0.3 mm (4 mm) thick samples at the 26 m (50 m) flight paths of GELINA. The energy range from 0-10 (0-40) keV was covered. Two capture measurements were carried out at 28.7 m (58.5 m) with a sample thickness of 1 mm (3 mm). Important corrections concern internal conversion and resonance dependent attenuation correction to the gamma-ray efficiency (weighting function) due to resonance self-shielding. The latter was determined using MCNP. MCNP was also used to determine the corrections due the neutron sensitivity of the detectors, which are non-negligible as a result of the large ratio of the neutron to the gamma width. Taking these effects into account excellent fits were obtained in a simultaneous analysis of the capture and transmission results using the REFIT code. Deviations were found in the capture areas compared to earlier work as well as in the $g\Gamma_n$ values. Further work is ongoing to understand these deviations.[10]

It may be noted that a considerable number of non-fuel isotopes (e.g. ^{55}Mn, ^{103}Rh, ^{133}Cs) are under study in collaboration with Oak Ridge National Laboratory in the interest of criticality safety. These are presented in more detail in reference 11.

II.C. Measurements for ^{241}Am

^{241}Am is not only an important constituent of high level radioactive waste. Its cross sections and in particular the branching ratio in radiative neutron capture are of key importance for the production of the other isotopes of Am and those of Cm. Therefore better cross sections for ^{241}Am will lead to better predictions for waste minimization scenarios. In a collaboration with CEA, IKI and FZK a number of measurements are planned and in progress. At IRMM total cross section and capture cross sections are being carried out at GELINA and (n,2n) cross sections with the activation technique are being determined at the Van de Graaff accelerator. Samples were prepared at the Institute for Trans Uranium elements (ITU, JRC-Karlsruhe) using the solgel bead technique.

First experiments were made to determine the total cross section below 20 eV and the (n,2n) cross section.

Sample preparation was completed at ITU in the fall of 2006. Samples consist of AmO_2 dissolved in a matrix of either Y_2O_3 (yttria) or Al_2O_3 (alumina). One sample of 20 mm diameter and about 300 mg Am was prepared for the study of low energy transmission and capture measurements at GELINA. Ten samples of about 40 mg Am in alumina matrix were prepared for the activation studies.

The choice for the solgel bead technique was made as it was a promising method to obtain a very homogeneous sample thickness distribution on the micro and mesoscopic scale. Such a homogeneous distribution is essential to avoid spurious apparent broadening of resonances with very high peak cross sections. The first results from transmission measurements were analyzed with the REFIT code and showed no need for any additional broadening mechanism besides that offered by the natural line width, the Doppler effect and the resolution of the setup. Further analysis and new measurements are scheduled for the near future.

The first data from the activation measurements for the (n,2n) reaction were taken at 8.2 and 9.0 MeV. At 9 MeV good agreement was obtained with earlier work, while the point at 8.2 MeV extends the earlier result to lower energies. Further measurements are planned at energies above 14 MeV.

II.D. Fission studies

Several experiments are ongoing that address neutron-induced fission.

II.D.1. Search for a fission isomer in ^{235}U

At the Van de Graaff laboratory a fast beam chopper was developed to study fast neutron induced activities with a very short half life. The chopper has a maximum repetition rate of 5 kHz with a typical switching time from beam on to beam off of about 1 μs. The existence of a double-humped fission barrier for ^{235}U is well known, but the question whether the second well has an isomeric state was still open. Using an ionization chamber with high purity ^{234}U two experiments were carried out at E_n=0.95 and 1.27 MeV, corresponding to energies where the fission cross section shows a resonance-like structure. During a large number of beam-on, beam-off cycles delayed fission events were collected that allowed to confirm that delayed fission occurs in the n+^{234}U reaction and that the isomer involved has a half life of 3.6(1.8) ms.[12]

II.D.2.Fission neutron spectrum of ^{235}U

The fission neutron spectrum of neutron-induced fission on ^{235}U is an important quantity in reactor calculations. It is rather well known at thermal neutron energies but its incident energy dependence has been subject of debate. A detailed model of this spectrum was developed at Los Alamos by Madland and Nix.

At the Van de Graaff new measurements were made for incident neutrons of 0.5 MeV and for emitted neutrons of 0.8 to 10 MeV. The measured spectra are almost perfectly Maxwellian below 6 MeV in shape and agree with six earlier data sets. This is not consistent with the model of Madland Nix but is well described by a three source model. Measurements use the time-of-flight technique with liquid scintillators allowing for pulse shape analysis. The efficiency is characterized by a second time-of-flight measurement using a ^{252}Cf fission chamber. Further studies are under way.[13]

II.D.3. Subthreshold cross section for the ^{236}U(n,f) reaction

The cross section for the ^{236}U(n,f) reaction is important for the production of heavier actinides in the thorium-uranium fuel cycle and for the production of ^{237}Np in the conventional fuel cycle. An accurate determination of the cross section in the subthreshold region depends heavily on a very high sample purity. Results obtained at IRMM by Wagemans and co-workers from the U. of Gent and IRMM show that the cross sections currently found in evaluated libraries strongly overestimate the fission cross sections. In particular, in the energy range up to 2 keV only two resonances were found with an appreciable fission width, where the evaluated files have nonzero fission widths for many additional resonances on the basis of an earlier measurement. As a consequence the resonance integral is strongly reduced.[14]

II.E. Activation and light charged particles

II.E.1. Activation cross sections

The measurement of cross sections with the activation technique has been a large effort at the IRMM Van de Graaff facility. A large number of cross sections were measured in the range from 14 to 20 MeV and contributed to Subgroup 19 of the Working Party on Evaluation Cooperation of the OECD Nuclear Energy Agency. Such cross sections are of interest for estimates of dose rates, handling limits, decay heat, and damage to structural parts as a result from hydrogen and helium production. Since then several new measurement campaigns were carried out. These involve studies of the isotopes of Cr, Ni, Cu, Zr, Ta and W. Results contribute to the development of the European Activation File.[15]

II.E.2. Light charged particle emission.

The $^{16}O(n,\alpha)^{13}C$ reaction recently received considerable attention through a subgroup on the criticality under prediction of light-water reactors at the Working Party for Evaluation Co-operation of the OECD Nuclear Energy Agency. The detailed understanding of the criticality of light-water reactors has important economic impact and neutron removal through this reaction is one of the less well understood smaller contributions to the criticality estimate.

At IRMM, new measurements are being performed using a one dimensional Time-Projection-Chamber (TPC) that was developed in the context of the study of the $^{10}B(n,\alpha)^{7}Li$ reaction. For this work the technique is further developed in going from the study of a thin layer target to a gaseous (CO_2) target. A large number of preliminary data are available. These data differ from earlier measurements agreeing better with evaluated results (JEF2.2 and JENDL3.3). Measurements and data analysis to improve the accounting of spurious neutrons and other neutron source and detector characteristics are underway.[16]

III. CONCLUSIONS

An overview was presented of recent and ongoing work at the IRMM neutron physics unit. The measurements at IRMM are part of European efforts to minimize high level nuclear waste and increase the safety of nuclear energy. The IRMM neutron laboratories are important European facilities for the measurement of neutron data. A large number of international collaborations ensure that the unit's program is of interest to external users and furthermore provides important stimuli to develop equipment and methodologies to remain at the forefront of neutron data research for applications.

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