Recent Developments in Neutron Measurements at EC-JRC-IRMM

CCRI(III) meeting of 01 to 03 April 2009

Stephan Oberstedt

Institute for Reference Materials and Measurements European Commission, Joint Research Centre IRMM Retieseweg 111, Geel Belgium

Neutron fluence measurements at IRMM are mainly carried out as a routine support to characterise and model neutron fields for experimental activities in neutron data measurements. Experimental projects are related to investigation of fundamental material properties and initiated by demands from international organisations (like JEF - Joint European File and the OECD) as well as policies of the European Commission. The experimental work is divided into two major actions; "Neutron data for waste transmutation and safety of different reactor systems" and "Basic Research in Nuclear Physics and Neutron Data Standards".

The objective of the first action is to produce neutron data needed for the assessment of partitioning and transmutation (P&T) strategies and the safety of different reactor systems. Data is also provided for scenarios that aim at reduced production of high level nuclear waste, such as the Th/U fuel cycle, and the various GenIV concepts. More specifically, experiments include:

- Cross-section measurements for the transmutation of minor actinides (fission, capture and total) and long-lived fission products (capture and total).
- Fission yield measurements and cross-sections for light charged particle production and ternary fission (tritium) for the safe handling, processing and disposal of nuclear waste.
- Measurements of cross-sections for Pb and Bi for neutron transport calculations in ADS (Accelerator Driven Systems): inelastic-, (n,xn)-, elastic-, capture- and total cross sections.
- Cross-section measurements for the assessment of the integrity of structural components, including inelastic cross-sections and, in particular, the (n,xp)- and (n,x- alpha) processes in structural materials.
- Measurements of cross sections for the Th/U fuel cycle.
- Measurements of cross sections relevant to multiple recycling of fuel.
- Measurements of cross sections relevant to high burn up fuel

The second action targets more basic research, including:

- Measurements of neutron data standards.
- Experimental and theoretical work for the understanding of the nuclear fission process.
- Development of improved measurement techniques using state-of-the-art digital signal processing technology.
- Development of new neutron detection concepts.
- Support of external users for development of new nuclear methods for non- destructive materials analysis.

Annual reports for the scientific and experimental work are available in PDF-format on:

http://www.irmm.jrc.be/html/publications/technical_reports/nuclear_research/index.htm

Neutron production

The IRMM operates two accelerators for neutron production, a 7 MV Van de Graaff accelerator for quasi mono-energetic neutron fields and a 150 MeV electron linac (GELINA) generating a pulsed white neutron spectrum. Both accelerators operate on a 24 h / day schedule.

The Van de Graaff produces either continuous or pulsed ion beams by means of two pulsing systems:

- 1. a fast beam pulsing generating a minimum ion beam pulse width of 1.5 ns and pulse repetition rates of 2.5, 1.25 or 0.625 MHz, and
- 2. a slow beam pulsing system giving a minimum pulsing width of 10 μ s at a continuously adjustable frequency up to 5 kHz.

Neutron fields with well defined energies are produced using the nuclear reactions Li(p,n), T(p,n), D(d,n) or T(d,n) giving neutrons within the energy regions 0.3 - 10.0 MeV and 14.5 - 24 MeV. More specified neutron fluence data at 10 cm from the source are given in the table below.

Reaction	< E _n >	ΔE_n	< E _p >	Ip	Target	Target thickness	neutron fluence rate
	MeV	MeV	MeV	μA		mg/cm ²	$cm^{-2} s^{-1}$
							(at 10 cm distance)
Li(p,n)	0.25	0.075	2.047	25	LiF	0.5	5.03E+05
Li(p,n)	0.565	0.061	2.321	20	LiF	0.5	2.00E+06
T(p,n)	1.2	0.05	2.022	25	TiT	2.0	3.23E+06
T(p,n)	2.5	0.137	3.351	15	TiT	2.0	3.89E+06
D(d,n)	5.0	0.356	1.943	25	TiD	2.0	2.82E+06
D(d,n)	8.0	0.171	4.841	10	TiD	2.0	1.97E+06
$T(d,n)^*$	14.8	0.428	0.964	50	TiT	2.0	4.22E+06
T(d,n)	16.2	1.049	0.966	50	TiT	2.0	4.62E+06
T(d,n)	19.0	0.338	2.679	20	TiT	2.0	7.58E+05

^{*}Neutron fluence in 74 deg. Angle.

The GELINA neutron source is dedicated for neutron time-of-flight measurements. A typical operating parameter setting is 100 MeV average electron energy, 10 ns pulse length, 800 Hz repetition rate, 12 A peak and 100 μ A average current. With a post-acceleration pulse compression system, the electron pulse width can be reduced to approximately 1 ns (FWHM) while preserving the current, resulting in a peak current of 120 A. The accelerated electrons produce Bremsstrahlung in a uranium target which in turn, by photonuclear reactions, produces neutrons. Within a 1 ns pulse a peak neutron production of 4.3 x 10¹⁰ neutrons is achieved (average flux of 3.4 x 10¹³ neutrons/s).

Both accelerators are subject to continuous upgrading. In 2009 the radio frequency ion source in the Van de Graaff will be replaced by an ECR ion source with the aim of improving the long term stability of the accelerated ion beam and reducing the amount of annual maintenance time.

Neutron spectrometry

IRMM uses a Bonner sphere system of type PTB-C for neutron spectrometry. The system includes 9 polyethylene spheres (3, 4, 5, 6, 7, 8, 10, 12 and 18 inches) with a spherical ³He proportional counter as neutron detector in the centre. The spheres are hold by a moveable crane during measurements and the precise height and distance are adjusted and measured using laser positioners.

The response functions of the spheres have been obtained from precise geometrical drawings and measurements from PTB and Monte Carlo calculations using MCNP, also from PTB. The measurements, including the volumes and the densities of the spheres, and MCNP calculations have been repeated at IRMM giving almost identical results. Few channel spectrum unfolding is done using the MAXED or GRAVEL codes, however, a new Windows style software has been developed at IRMM to replace the previously used U.M.G. package from PTB (see Fig. 1). The software produces an unfolded spectrum in real-time whenever an input parameter is changed. So far only the SAND-II procedure, including option for Gravel, has been included but the implementation of other unfolding procedures, e.g. maximum entropy, is foreseen.

Exit Comment Influence for DONA project.AmBe source, 50 n/cm2, 21/02/2005 Highest neutron energy (keV): Input and calculated data Files handling 10000 Reaction Measurement Unc. (%) (c-m)/m, def. (c-m)/m, fin. % tot fin used Use? Load new input file: 111000 Ti46(n,p)Sc46 0.271E-01 43.06 -0.479 0.141 13.0 Imput file: Use energy bin structure from Fe54(n,p)Mn54 0.904E-01 48.30 -0.254 -0.092 8.4 Imput Save unfolding Save unfolding Save unfolding Save unfolding Save unfolding	ONA, ver.2								
Highest neutron energy (keV): Input and calculated data Heasturement Unc. (%) (c-m)/m, def. (c-m)/m, fin. % tot fin used Use? Files handling 10000 Imput and calculated data Reaction Measurement Unc. (%) (c-m)/m, def. (c-m)/m, fin. % tot fin used Use? Load new input file: input Imput and calculated data Ti46(n,p)Sc46 0.271E-01 43.06 -0.473 0.141 13.0 Imput Imput and calculated data Ti48(n,p)Sc48 0.136E-02 23.92 -0.842 -0.335 30.9 Imput Use energy bin structure from Fe54(n,p)Mn54 0.904E-01 48.30 -0.254 -0.092 8.4 Imput Save unfolding Fe56(n,p)Mn56 0.371E-02 32.63 -0.667 0.151 13.9 Imput	Exit Com								
Image: Construction of the structure from Ti46(n,p)Sc46 0.271E-01 43.06 -0.479 0.141 13.0 Image: Construction of the structure from Use energy bin structure from Fe54(n,p)Mn54 0.904E-01 48.30 -0.254 -0.092 8.4 Image: Constructure from Save unfolding	nest neutron energy (keV):	out and calculated data Reaction	Measurement	Unc. (%)	(c-m)/m, def.	(c-m)/m, fin.	% tot fin used	Use?	Files handling
Use energy bin structure from Fe54(n,p)Mn54 0.904E-01 48.30 -0.254 -0.092 8.4 IV Save unfolding Save unfolding		Ti46(n,p)Sc46 Ti48(n,p)Sc48	0.271E-01	43.06	-0.479	-0.335	30.9	<u>र</u>	input
Fe56(n.p)Mn56 0.371E-02 32.63 -0.667 0.151 13.9 🗸	e energy bin structure from	Fe54(n,p)Mn54	0.904E-01	48.30	-0.254	-0.092	8.4	2	Save unfolding
C Fine bin structure Ni58(n,p)Co58 0.100E+00 11.38 -0.135 0.065 6.0 ▼ DONA	C Fine bin structure	Fe56(n,p)Mn56 Ni58(n,p)Co58	0.371E-02	32.63	-0.667	0.151	6.0	া ব ব ব	DDNA
C Default spectrum In115(n,n)In115 0.641E-01 12.34 0.206 0.250 23.0 ✓	Default spectrum	In115(n,n')In115	0.641E-01	12.34	0.206	0.250	23.0		
C Respons function In115(n,g)In116m 0.110E+01 12.59 -0.051 4.7 ✓ Used interrations: 20	C Respons function	In115(n,g)In116m	0.110E+01	12.59	-0.987	-0.051	4.7		Used interrations: 20
Solution spectrum as Max interrations: 500	dF/dE							Г	Max interrations: 500
C E*dF/dE Chi-2, default spc: 11.970	○ E*dF/dE		-					Г	Chi-2, default spc: 11.970
Default Use Gravel	ault							Г	Use Gravel 🗖
Default/slask.TXT Process message:	ault/slask.TXT				·		<u> </u>		Process message:
Spectrum: #39: No message	ctrum: XSUN 115In(n,n')								#99: No message
Scale default spectrum: 🔽	le default spectrum: 🔽		-		·		<u> </u>	Г	
Scaling factor: 0.523E-08	ling factor: 0.523E-08							Г	
Search default	arch default		- [Г	
List default Dptimize Standard deviation of used: 0.420 0.196	ist default	Optimize	Standard devia	ation of used:	0.420	0.196			

Figure 1: Screen dump of the IRMM few channel unfolding programme. The programme includes today the SAND-II procedure with a Gravel option.

A neutron spectrometer based on neutron activation of metal disks has recently been tested successfully. The detector consists of eight metal disks (5 mm in thickness and 20 mm in diameter) of carefully selected different materials arranged in a circle as depicted in Fig. 2. The induced activity is measured for every disk in a low background gamma measurement station, followed by spectrum unfolding to obtain a complete neutron spectrum. Thus, the method is similar to the well known multiple foil activation technique, however, detections

limits as low as a few neutrons per cm^2 and second has been achieved which facilitates the use of the detector for measurements of environmental fluencies.



Figure 2: Neutron spectrometer based on neutron activations. The disk materials are indicated in the schematic to the left. For this particular detector a stack of Au / Cd / Au / Cd foils has been placed in the centre to monitor low energy neutrons. The diameter of the complete detector is less than 10 cm.

As an example, the detector was tested for the measurement of neutron spectra outside a MOX fuel storage container at the MOX fuel manufacturing plant Belgonucleaire in Mol Belgium (see Fig. 3). The unfolded spectrum corresponds well to the IAEA library spectrum, which here also was used as default spectrum for the unfolding routine. A higher thermal component is, however, measured which is explained by in scattering from surrounding materials and the MOX container.



Figure 3: Left: neutron spectrum obtained using disk activation. Right: IAEA library spectrum for MOX fuel at storage place.

Extensive work has also been put on the development of a novel dedicated set-up for neutron spectrometry by means of time-of-flight measurements in the Van de Graaff neutron laboratory using a pulsed ion beam. The original accelerator beam chopping and bunching electronics from High Voltage Engineering have been fine tuned to obtain pulses of a minimum fwhm of 1.5 ns at beam target with an average current of about 300 nA at a pulse frequency of 2.5 MHz.

Neutron fluence measurements

IRMM uses two Los Alamos style recoil proton telescope (RPT) as key instruments for neutron fluence measurements. The instruments have proven to give accurate results also in no-coincidence mode using only radiator-in / radiator-out measurements and subtraction. In figure 4 below measurement data from 5 MeV neutrons are shown for both 3-coincidence mode (left) and radiator-in / radiator-out (right) measurements. A maximum deviation of 3 % is calculated for the two techniques. However, the uncertainty is considerably increased using radiator-in / radiator-out measurements due to the higher background that enters the calculations Even so, the technique can be useful when, for example, technical problems appear with the 3-coincidence acquisition or as a routine verification of the functioning of the 3-coincidence electronics.

Both IRMM RPT instruments were subject for careful geometrical measurements and evaluations as the CCRI(III)-K10 competitions from 2000 showed discrepancies up to 9.5 % for one of the RPT to the very similar PTB RPT instrument. However, no geometrical measure error was noted. A novel in-house production of hydrogen rich radiator foils of tristearin and polyethylene is being implemented to reduce uncertainties related to possible degradation of the used foils.

Laboratory routine neutron fluence measurement devices, in addition to the Bonner spheres and the RPTs, include long counters, stationary as well as moveable, fission chambers and activation foils.



Figure 4: Measurement data from 5 MeV neutrons for 3-coincidence mode (left) and radiator-in / radiator-out and subtraction (right) measurements.

NEPTUNE isomer spectrometer

Recently a new slow pulsing system has been installed at the Van de Graaff mono-energetic neutron source for investigating short-lived activation products, spin isomer population and fission shape isomer decay. *NEPTUNE* (NEutron Production with TUNEable pulse structure) permits to flexibly adjust pulse frequency and duty cycle according to the specific requirements of the experiment.

The maximum pulse frequency of *NEPTUNE* is 5 kHz. The minimum pulse width is in the order of 2 μ s. The instrument is particularly useful to monitor the neutron field in reaction cross-section measurements, where down-scattered or thermalized neutron background is critical.

NUDAME (NUtron DAta MEasurements)

During 2005-2008 the IRMM accelerator infrastructure has been opened for external users within the framework of the Euratom Transnational Access programme. Neutron measurement projects in the areas of radioactive waste management, radiation protection and other activities in the field of nuclear technologies and safety, was accepted provided the IRMM experimental infrastructure offered a significant added value to the project. The support included, in addition to the access to the facilities, also financial and scientific support. A total of 18 experiments from laboratories in 8 European countries were accepted.

NUDAME is followed up by the EUFRAT (EUropean Facility for innovative Reactor And Transmutation neutron data, <u>http://irmm.jrc.ec.europa.eu/html/activities/eufrat/index.htm</u>) programme EUFRAT started on November 1, 2008 and runs 4 years. A total of 4500 supplementary data-taking hours will be made available to external users: 3600 hours at GELINA and 900 hours he VdG.

Recent publications:

- 1. Total characterization of neutron detectors with a ²⁵²Cf source and a new light output determination, N. Kornilov et al., *Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 599 (2009) 226-233
- 2. Properties of the reaction 238U(n, f) at the vibrational resonances, E. Birgersson et al., *Nuclear Physics A* 817 (2009) 1-34
- 3. Prompt fission neutron multiplicity and spectrum calculations for thermal and fast neutron induced reactions on 233,231Pa nuclei, A. Tudora et al., *Annals of Nuclear Energy* 35 (2008) 1131-1139
- 4. Experimental study of the Zr91(n,γ) reaction up to 26 keV, G. Tagliente et al., *Physical Review C Nuclear Physics* 78, 045804 (2008)
- 5. Neutron resonance capture and neutron diffraction analysis of Roman bronze water taps, P. A. C. Schut, *Journal of Radioanalytical and Nuclear Chemistry* 278 (2008) 151-164
- 6. The new PADC based fast neutron dosimetry system of the INFN-LNF, R. Bedogni et al., *Radiation Measurements* 43 (2008) S491-S494
- 7. Comment on "neutron multiplicity in the fission of U238 and U235 with neutrons up to 200MeV", N. Kornilov, F.-J. Hambsch, *Physical Review Letters* 101, 039201 (2008)
- 8. An intercomparison of Monte Carlo codes used in gamma-ray spectrometry , T. Vidmar et al., *Applied Radiation and Isotopes* 66 (2008) 764-768
- 9. Neutron capture cross section of Zr90: Bottleneck in the s-process reaction flow, G. Tagliente et al., *Physical Review C Nuclear Physics* 77, 035802 (2008)
- 10. Thin liquid sample fabrication for neutron resonance spectroscopy, G. Noguere et al., Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 587 (2008) 52-59
- The measurement of the 206Pb(n, γ) cross section and stellar implications, C. Domingo-Pardo et al., *Journal of Physics G: Nuclear and Particle Physics* 35 (2008) 014020
- 12. Nuclear physics for the Re/Os clock , M. Mosconi et al., *Journal of Physics G: Nuclear* and Particle Physics 35 (2008) 014015
- 13. Identification of a shape isomer in U235 , A. Oberstedt et al., *Physical Review Letters* 99, 042502 (2007)

- 14. Light fission-fragment mass distribution from the reaction 251Cf(nth, f), E. Birgersson et al., *Nuclear Physics A* 791 (2007) 1-23
- 15. Energy degrader technique for light-charged particle spectroscopy at LOHENGRIN, A. Oberstedt, S. Oberstedt, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 570 (2007) 51-54
- 16. Measurement of the radiative neutron capture cross section of Pb206 and its astrophysical implications, C. Domingo-Pardo et al., *Physical Review C Nuclear Physics* 76, 045805 (2007)
- 17. Measurement of neutron excitation functions using wide energy neutron beams, G. Lövestam et al., Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 580 (2007) 1400-1409
- 18. The use of C6D6 detectors for neutron induced capture cross-section measurements in the resonance region, A. Borella et al., *Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 577 (2007) 626-640
- 19. High-resolution neutron transmission and capture measurements of the nucleus Pb206, A. Borella et al., *Physical Review C Nuclear Physics* 76, 014605 (2007)
- 20. The 10B(n,a0)/10B(n,a1γ) branching ratio, F.-J. Hambsch, I. Ruskov, *Nuclear Science and Engineering* 156 (2007) 103-114
- 21. Neutron resonance capture applied to some prehistoric bronze axes, H. Postma et al., *Nuovo Cimento della Societa Italiana di Fisica C* 30 (2007) 105-112
- 22. The recoil proton telescope in non-coincidence mode for neutron fluence measurements, G. Lövestam, *Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 566 (2006) 609-614