

THE BARILOCHE NEUTRON PHYSICS GROUP CURRENT ACTIVITIES

R.E. Mayer, PhD

N.M.B. D'Amico, Eng.

CNEA and IB, Argentina

J.R. Granada, PhD

J. Dawidowski, PhD

CNEA, CONICET and IB Argentina

J.R. Santisteban, PhD

J.J. Blostein, PhD

A. Tartaglione, PhD

L.A. Rodríguez Palomino, PhD

I. Marquez-Damian, PhD

CONICET and IB, Argentina

C. Sepúlveda Sosa. MEng.

CCHEN, Chile

CNEA : Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, Argentina

CONICET : Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina

IB : Instituto Balseiro, Universidad Nacional de Cuyo and CNEA, Argentina

CCHEN : Comisión Chilena de Energía Nuclear, Chile.

Abstract

Our group has evolved around a small accelerator-based neutron source (ABNS), the 25 million electron Volt (MeV) linear electron accelerator at the Bariloche Atomic Centre. It is dedicated to applications of neutronic methods to tackle problems of basic sciences and to technological applications. Among these, the determination of total cross section of a material as a function of neutron energy by means of transmission experiments for thermal and sub-thermal neutrons is very sensitive to the geometric arrangement and movement of the atoms, over distances ranging from the 'first-neighbour scale' up to the microstructural level or 'grain scale'. This also allowed to test theoretical models of calculated cross sections and scattering kernels. Interest has moved from pulsed neutron diffraction towards deep inelastic neutron scattering (DINS), a powerful tool for the determination of atomic momentum distribution in condensed matter and for non-destructive mass spectroscopy. In recent years non-intrusive techniques

aimed at the scanning of large cargo containers have started to be developed with this ABNS, testing the capacity and limitations to detect special nuclear material and dangerous substances in thick cargo arrangements. More recently, the use of the ever-present “bremsstrahlung” radiation has been recognized as a useful complement to instrumental neutron activation, as it permits to detect other nuclear species through high-energy photon activation. The facility is also used for graduate and undergraduate students experimental work within the frame of Instituto Balseiro Physics and Nuclear Engineering courses of study, and also MSc and PhD theses work.

Keywords: Neutron applications, pulsed neutrons

Introduction

The Neutron Physics Department at the Bariloche Atomic Centre has evolved its experimental activity, from its very beginning, around the already mentioned 25 MeV electron linear accelerator, the LINAC, which started operation in 1970. It is a travelling wave 25 MeV electron accelerator pulsed machine. Electron pulse may be extended up to 2 microseconds. This small machine reaches a maximum neutron production operating at 100 Hz and 25 microampere mean electron current. It belongs to the Argentinean Atomic Energy Commission, CNEA, and is situated in an atomic centre by the southern Patagonian Andes mountain range. In what follows, we shall briefly review the most recent interdisciplinary activities of this research group.

Total neutron cross section

Neutron transmission is a very simple technique that measures the reduction in intensity in a neutron beam after traversing a sample. The reduction in intensity depends on the energy of the incident neutron and on the nuclear cross section and microstructure of the material composing the sample. So, analysis of the transmitted neutron spectra provides information about the microscopic properties of an object. Such analysis is performed on the total cross section, an intrinsic property that does not depend on object thickness. Advances in instrumentation have provided such spectroscopic capability to neutron radiography (Lehmann, 2009).

The energy dependence of the total cross section for thermal neutrons is dictated by the structure and dynamics of the atoms composing the material. The effect of the *microscopic* ordering (between 0.1 and 100 Å) on the total cross section results in the appearance of discontinuities called Bragg edges (Fermi, 1947), which have been used to study and characterize materials since the 50's. On the other hand, the effect of the *mesoscopic* structure (between 0.1 and 100 µm) on the total cross section has received little attention. We are currently investigating the effect of such

microstructural features in the total cross section, with the aim of using neutron transmission for non-destructive characterization of nuclear materials.

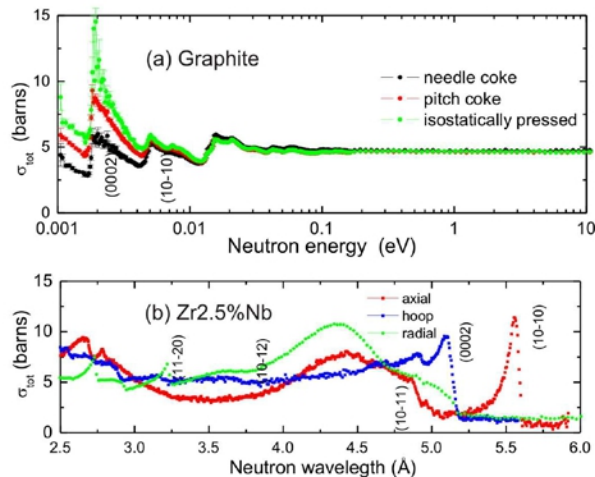


Fig.1: Upper graph shows the total cross section measured at our laboratory for nuclear-grade graphite produced by different manufacturing processes; first two Bragg edges indexed. Differences seen at neutron energies below 0.004 eV are due to different porosities. Lower graph shows the total cross section of a Zr2.5%Nb pressure tube from a CANDU power plant along the three principal directions, with Bragg edges indexed. Large differences between directions are due to crystallographic texture in the material.

Related with the examples depicted in Fig.1, for the first case a theoretical model was developed to extract the mean size of the pores from such measurements (Petriw, 2010) and for the second case, we are developing a model to measure the orientation distribution function of the crystallites from such experiments (Santisteban, 2011).

Scattering kernels

Calculations of moderators require scattering kernels that are accurate in both the energetic and angular distribution of outgoing neutrons. However, traditional neutron moderators like light and heavy water have scattering cross section libraries (Mattes,2005) in ENDF format (Chadwick, 2011) to be used in Monte Carlo and deterministic calculations that rely on water models that do not utilize the up-to-date knowledge of their structure and dynamics.

Very recently (Marquez Damian, 2013) we presented a new model to compute the scattering kernels for light and heavy water based on molecular dynamics simulations. Using the molecular dynamics program GROMACS and a flexible SPC (Single Point Charge) model (Toukan, 1985) we obtained the frequency spectra of hydrogen, deuterium and oxygen, bound in light and heavy water. With those spectra we computed the scattering laws using the

LEAPR module of NJOY (MacFarlane, 1999), which uses the incoherent approximation.

Scattering laws for D and O in D₂O are corrected using the Sköld approximation together with available partial structure factors. Using those kernels we computed double differential and integral cross sections over a wide range of neutron energies (1e-5 to 1 eV). These results represent an improvement over existing models when compared with experimental values, especially for low incident neutron energies. This is shown in Fig.2, where we can observe that our new scattering kernel is finally able to reproduce the total cross section that we measured 40 years ago (Kropff, 1974) using the (at that time new) Bariloche LINAC!

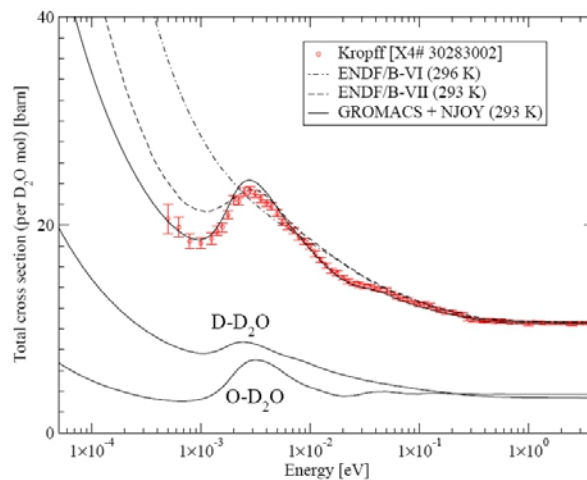


Fig.2: The total cross section of D₂O at room temperature, comparing the results from existing libraries (Mattes, 2005), our present model, and our own transmission experiment (Kropff, 1974).

Deep inelastic neutron scattering (dins)

Originally devised by Hohenberg and Platzmann in the 60's to investigate the Bose-Einstein condensation in superfluid ⁴He, Deep Inelastic Neutron Scattering (also known as Neutron Compton Scattering) developed as a customary research tool to investigate momentum distributions in Condensed Matter. In a DINS experiment scattered neutrons are detected at a given angle, and a mobile filter (with a neutron absorption resonance at a few eV energy) is placed in the path of the scattered neutrons, and alternative 'filer-out' and 'filter-in' spectra measurements are performed. The difference between these spectra as a function of the neutron time of flight is the basic magnitude determined in DINS experiments, and is related with the sought momentum distributions. Initially developed at Rutherford Appleton Laboratory, the technique proved to be applicable in our low-intensity

neutron source, especially in regard to the development of data processing methods.

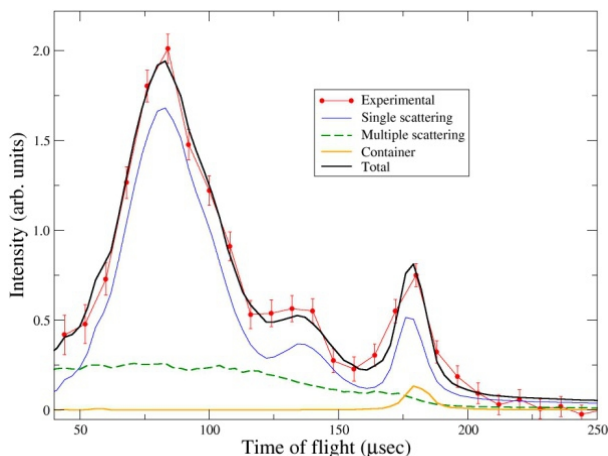


Fig.3: Measured neutron Compton profile for the light water/heavy water mixture, together with results of Monte Carlo simulations showing single, multiple and total scattering contributions to the observed spectra.

A remarkable result in the methodological aspect, was the determination of the Compton Profiles' absolute intensities in light water/heavy water mixtures, depicted in Fig.3 (Blostein, 2009), which showed no anomalies in the cross section of hydrogen at epithermal energies, which was the subject of a long scientific debate (Chatzidimitriou-Dreis mann, 1997).

At present we have two ^3He detector banks, which allow the simultaneous measurement at selectable angles. This combination aims at the study of the profiles of hydrogen and light atoms. A combination of forward and backward scattering angles, allow the separate study of the profiles of light and heavy atoms.

Cargo scanning complementary techniques

The use of slow neutrons for the purpose of investigating thick cargo arrays is viewed here as complementary to the widely tested fast neutron approach. Slow neutrons tend to diffuse into samples, with loss of time-of-flight (TOF) information, but inducing absorption reactions through higher interaction cross sections. Many related results have been presented at the AccApp2009-International Topical Meeting on Nuclear Research Applications and Utilization of Accelerators (Mayer a, 2009).

Detector set-ups were used to probe into realistic 2m thick cargo arrays with 3 mm steel plates in the incoming and outgoing sides of the simulated container. To mimic a lower intensity neutron source, the experimental set-up is positioned at least five meters away from the target of

the 25 MeV linear electron accelerator. The low intensity falling on the sample is in the order of 200 thermal n/cm^2sec and 90 near epithermal n/cm^2sec (above cadmium cut-off energy).

One detector array built consists of four commercial 2"x2" NaI(Tl) scintillators with photomultiplier tubes and *ad hoc* voltage dividers coupled to only one preamplifier, designed to reduce the initial dead time after each accelerator burst. Shielding made it possible to sort-out the gamma response to neutron interactions in the presence of a combined intense pulsed high energy X-ray and neutron field. Substances of interest tested for their gamma response are Cl, N, S, Ag, As, Cr, Cd, Hg.

Most attention was dedicated to chlorine, for which the only real case of interest is that of a container almost full of unwanted chlorinated contaminants. Fig.4 shows the result of plotting several 5 minutes irradiations, where the signal from solutions of different concentrations are not distinguishable, but clearly separated from the Cl in the empty container composition, and far from the background of the non chlorinated cargo array.

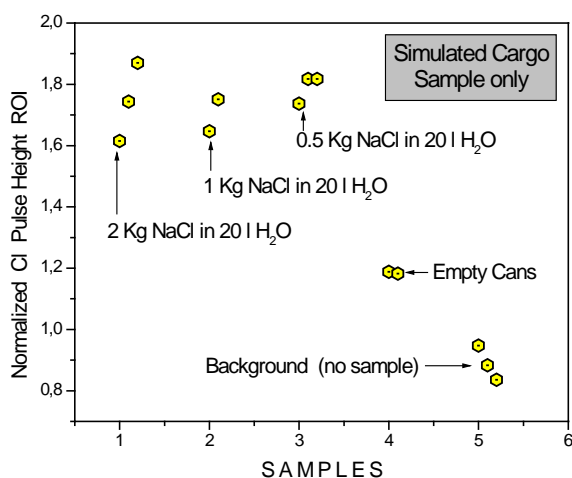


Fig.4: Simulated empty container. Later loaded with different NaCl solutions, 2, 1 and 0.5 kg in 20 l of water. 5 min recording time determinations. Appropriate regions of interest (ROI) of each gamma spectrum most representative of the substance, were normalized to a region of the spectrum not influenced by that substance

Fast neutron detection from uranium induced fission after each accelerator burst (Tartaglione, 2009), was carried out through 70 x100cm² active area detectors incorporating 10 He³ tubes (2.5cm diameter and 55cm active length, 4 atm gas pressure) inside Cd wrapped moderator. Uranium in simulated cargo arrays was materialized through an enriched uranium sample (HEU, U-235 mass= 27.5 g in Al) or a natural uranium fuel bundle (FB, U-235 mass= 38.38 g in 13 fuel pins), Fig.5. These results were presented at the

Third Research Coordination Meeting on Neutron Based Techniques for the Detection of Illicit Materials and Explosives, United Nations Internat. Atomic Energy Agency (Mayer b, 2009).

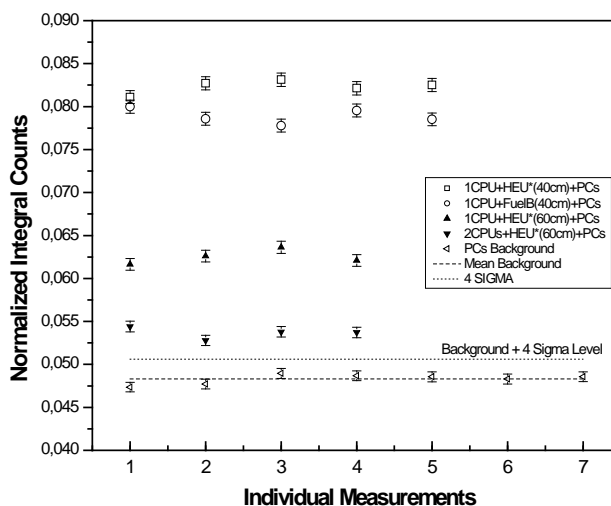


Fig.5: Integral fast neutron counts for each 5 min recording time determinations with HEU and FB, hidden behind different number of table-top computers and 3 mm steel plate. The mean background 4 sigma level is indicated for comparison

High energy photon activation

The exploitation of the high energy bremsstrahlung (high energy photons emitted by fast, relativistic, electrons being stopped by interaction with a heavy atomic nucleus) for photonuclear activation (PNA) has recently been explored. Several samples were activated and analysed. Fig.6 depicts two examples of initial interest. The upper graph is related to the need to find out if the pure quartz containers usually employed for neutron activation analysis would be apt for PNA. The result is that they are clearly not convenient for the considerable number of gamma “peaks“ that quartz produces and, consequently, if some existing samples were to be analysed with the new technique, they would have to be removed from the quartz capsules. The lower graph shows the result of activating a pure lead sample to test if lead could be easily detected and if impurities would be distinguished from it, because lead is not detectable through conventional instrumental neutron activation analysis.

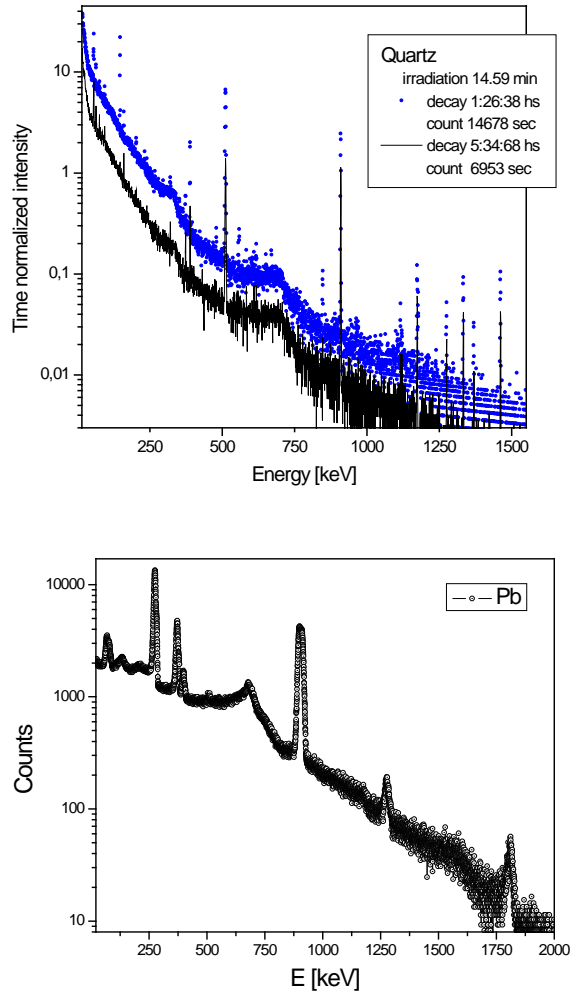


Fig.6: Gamma response from samples activated through photonuclear reactions. Upper graph: gamma spectra from quartz sample after two initial decay times. Lower graph: pure lead sample.

Undergraduate and postgraduate students experimental activities

Students of Nuclear Engineering and some students pursuing Master or Doctoral degrees, perform experimental determinations of neutron field profile in moderators by foil activation and measurements of die-away time in moderators (Fig.7), as a function of geometrical buckling and of macroscopic absorption cross section, using miniature fission chamber neutron detectors, taking advantage of the pulsed nature of this versatile neutron source.

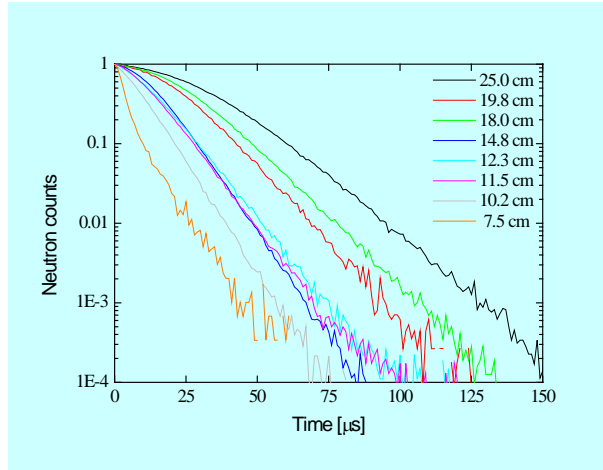


Fig.7: Neutron population die-away for cubic moderators of the lateral dimensions indicated

LOCA (Loss Of Coolant Accident) simulation experiments are available for students, in which the pulsed neutron field is studied around a natural uranium fuel element in the presence of surrounding moderator, with and without coolant (light water). The fuel pins pitch can also be changed.

Conclusion

The capacity to detect neutrons as a function of neutron energy over a wide range of energy decades is possible when employing a pulsed neutron source, which, in our case, relatively simple spectrometers and associated instrumentation, has allowed obtaining sound statistical data even using a small accelerator-based neutron source as the 25 MeV linear electron accelerator at Bariloche. The possibility to employ the bremsstrahlung high-energy photons has arisen the capacity to carry out elemental analysis. And, although not mentioned herein, the direct irradiation with the lower energy electron beam has been used in other applications such as characterization of electronics for space satellites use.

References:

- Lehmann, E.H. et al, Nucl. Instr. Meth. A603 (2009) 429-438.
 Fermi, E., W.J. Sturm and R.G. Sachs, Phys. Rev. 71 (1947) 589-594.
 Petriw, S., J. Dawidowski, J.R. Santisteban, J Nuclear Materials 396 (2010) 181.
 Santisteban, J.R., et al, J Nuclear Materials 425 (2011) 218.
 Mattes M., J. Keinert. Thermal neutron scattering data for the moderator materials H₂O, D₂O and ZrHx in ENDF-6 format and as ACE library for MCNP (X) codes. INDC (NDS)-0470. IAEA (2005).

Chadwick, M. B., et al. "ENDF/B-VII.1. Nuclear data for science and technology: cross sections, covariances, fission product yields and decay data." *Nuclear Data Sheets* 112.12 (2011): 2887-2996.

Marquez Damian J.I., D.C. Malaspina, J.R. Granada."Vibrational spectra of light and heavy water with application to neutron cross section calculations". *J. Chem. Phys.*, 139 (2013) 024504. <http://dx.doi.org/10.1063/1.4812828>

Toukan, K., Rahman, A. Molecular-dynamics study of atomic motions in water. *Phys. Rev. B: Condens. Matter*, 31 (5), 1985.

MacFarlane, R., Muir, D. The NJOY Nuclear Data Processing System, Version 99. Tec. Report., Los Alamos National Laboratory, 1999.

Kropff, F., J.R. Latorre, J.R. Granada, C. Castro Madero. "Total Neutron Cross-Section of D2O at 20 °C between 0.0005 and 10 eV", EXFOR 30283.001 (1974).

Blostein, J.J., L.A. Rodríguez Palomino, J. Dawidowski, *Phys.Rev.Lett.*, 102 (2009) 097401.

Chatzidimitriou-Dreismann, C.A., T. Abdul Redah, R.M.F. Streffer, J. Mayers, *Phys.Rev.Lett.*, 79 (1997) 2839.

Mayer(a), R.E., A. Tartaglione, F. Di Lorenzo, C. Sepulveda Soza, M. Schneebeili, P. D'Avanzo, L. Capararo. Active Neutron Interrogation Approach to Detect Special Nuclear Material in Containers. AccApp2009-International Topical Meeting on Nuclear Research Applications and Utilization of Accelerators; *Satellite Meeting V Neutron Based Techniques for the Detection of Illicit Materials and Explosives*. Vienna, Austria, 4 - 8 may 2009.

<http://www-pub.iaea.org/MTCD/meetings/Announcements.asp?ConfID=173>
Tartaglione, A., F.Di Lorenzo, R.E.Mayer. Detection of Thermal Induced Prompt Fission Neutrons of Highly-enriched Uranium: A Position Sensitive Technique. *Nucl.Inst. and Meth. in Physics Research, B* 267 (2009) 2453-2456

Mayer(b), R. E., A. Tartaglione, J.J. Blostein, C. Sepúlveda Soza, M. Schneebeili, P. D'Avanzo, L. Capararo. Slow Neutron Interrogation for Detection of Concealed Substances. Third Research Coordination Meeting on Neutron Based Techniques for the Detection of Illicit Materials and Explosives, United Nations Internat. Atomic Energy Agency (IAEA) Coordinated Research Project. Johannesburg, South Africa 16 – 20 November 2009.

<http://cra.iaea.org/cra/documents/2009-annual-report.pdf>
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