Design and performance of a compact subthermal neutron source for an Electron Linear Accelerator

2013 JINST 8 P11009

(http://iopscience.iop.org/1748-0221/8/11/P11009)

PUBLISHED BY IOP PUBLISHING FOR SISSA MEDIALAB

RECEIVED: *October 4, 2013*
REVISED: *October 23, 2013*
ACCEPTED: *October 28, 2013*
PUBLISHED: *November 12, 2013*

Design and performance of a compact subthermal neutron source for an Electron Linear Accelerator

A. Tartaglione,[a,b,c,1] **V. Galván,**[a,b] **J. Dawidowski,**[a,b,c,d] **F. Cantargi**[a,c,d] **and J.J. Blostein**[a,b,c]

[a]*Departamento de Física de Neutrones, Centro Atómico Bariloche, Av. Bustillo 9500, Bariloche, Argentina*
[b]*Consejo Nacional de Investigaciones Científicas y Técnicas, Av. Bustillo 9500, Bariloche, Argentina, Argentina*
[c]*Instituto Balseiro, Universidad Nacional de Cuyo, Av. Bustillo 9500, Bariloche, Argentina*
[d]*Comisión Nacional de Energía Atómica, Av. Bustillo 9500, Bariloche, Argentina, Argentina*

E-mail: tartagla@cab.cnea.gov.ar

ABSTRACT: We present the design, construction and performance of a compact subthermal neutron source to be operated at the Bariloche Electron LINAC. The design was based on the premise of keeping the moderator temperature stable at 77 K for at least seven hours without refilling liquid nitrogen. Two moderator materials in semi-cylindrical geometry were studied: polyethylene and mesitylene. The effective moderator volumes were optimized by Monte Carlo simulations. Experimental data and calculations corresponding to the neutron production as a function of the neutron energy for those materials at room temperature and at 77 K were compared. We observed that the rate of subthermal neutrons, compared to epithermal neutrons, increases up to five times for both moderators when they are cooled down.

KEYWORDS: Instrumentation for neutron sources; Neutron sources

[1]Corresponding author.

doi:10.1088/1748-0221/8/11/P11009

2013 JINST 8 P11009

Contents

1 Introduction

Scattering techniques based on subthermal and cold neutrons are used nowadays in a wide variety of basic and applied studies in condensed and soft matter. In the last years, the increase and internationalization of the neutron activity have produced a concentration of the experimental capacities around the big facilities (reactors and spallation sources). However, in small facilities, as those compounded by linear accelerators, it is possible to perform with some advantages an important part of the experimental activity. These are very versatile, allowing to perform experiments to test targets and moderators [1, 2], and to apply experimental techniques that are not normally available in the big facilities (such as the technique of neutron transmission [3]).

Neutron sources based on linear accelerators have traditionally been used to produce neutrons in the thermal and epithermal energy ranges [4]. They were employed as a very important source of nuclear data [1, 5] and have gradually been adapted to work at subthermal energies. Efforts to improve and adapt cold neutron sources in small facilities, found an important application in the implementation of the mini-focusing Small Angle Neutron Scattering instrument [6]. It is also worth mentioning the recent implementation of subthermal neutron transmission experiments to study small-angle scattering effects as shown in ref. [7].

The pulsed neutron source based on the Bariloche electron linear accelerator (LINAC) facility (Argentina) produces neutrons by the interaction of 25-MeV-electrons with a lead target. A moderator, placed next to the target is easily changed, allowing the production of different neutron spectra according to different requirements. A typical configuration to get thermal neutrons is a slab of polyethylene at room temperature with an area of 21 cm $\times$ 21 cm and a thickness of 4 cm. Given the versatility of the neutron source, that allows the test of moderators, a research line in this direction was developed investigating the optimization of geometries [8] and materials [9]. In this regard, particular attention, deserved the study of mesitylene. Due to its high protonic density, good resistance to radiation [10–12], easy handling and the existence of low-energy rotational modes, mesitylene has been recommended and also used as a moderator for a cold neutron source [13–16].

At room temperature it is a colourless liquid whereas at low temperatures and depending on the thermal procedure, solid mesitylene can exist in three different phases [17].

In this work we describe the characteristics of a compact cold neutron source designed to enhance the subthermal flux, which allows a faster determination of the total neutron cross section in that energy range. Polyethylene and mesitylene were the chosen materials to test and optimize the design.

1.1 Operation requirements

The operation requirements of the cold source are strongly related with the operating conditions of the LINAC. They can be classified as:

- Accessibility: the accelerator, the lead target and the moderator are placed within a concrete bunker, that has prohibited access during the operation.
- Autonomy: at the moment of this work, no automatic liquid nitrogen refilling system is installed to keep the moderator cold, therefore this operation is manually performed before the accelerator becomes operational. This fact implies that the nitrogen bath must provide an autonomy time of seven hours (equal to the daily operating time of the accelerator).
- Size: the final size of the constructed neutron source is limited by the available space inside the biological shielding placed around the lead target of the accelerator. This shielding consists of a hollow steel cylinder with 48 cm of internal diameter with walls of 10 cm thick filled with water. The setup is completed with graphite reflectors and a cadmium plate to decouple the cold moderator from the slow neutrons produced by the water shielding, to minimize the mean emission time of neutrons emitted from the source, and the uncertainty in the neutron flight length.
- Interaction with radiation and neutrons: the materials employed must resist the high gamma radiation dose generated during the accelerator operation, and also satisfy the condition of neutronic transparency to avoid a high neutron absorption.
- Radiological security: as the materials that compose the source can be activated after the irradiation, the activation cross sections and the decay times must allow the human interaction in a reasonable time.

1.2 Design and construction

The best geometry for the compact cold source that fulfills the operational conditions mentioned in the previous section is basically composed of a cylindrical Dewar flask containing the neutron moderator under a permanent liquid nitrogen bath. The Dewar flask is formed by two coaxial cylinders in contact at the top through a cylindrical piece of Polyoxymethylene (Delrin ®). In its bottom, it has a hole in which the vacuum system is connected (see figure 1). The vacuum line connects the source with a mechanical pump placed outside the LINAC bunker.

The material employed for the cylinder walls and bases was Aluminum 6061 alloy [18]. This alloy, meets the requirements of good mechanical properties, low absorbance for subthermal neutrons and low neutron activation.

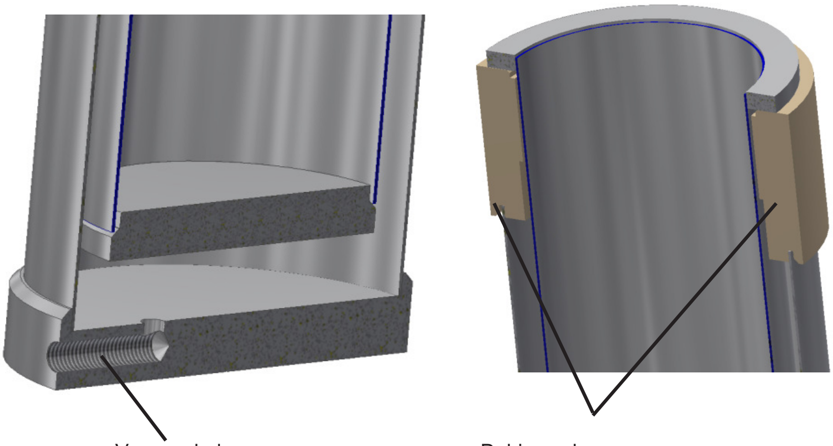

Figure 1. Bottom and top views of the Dewar flask transversal cut. The seal at the top was made of Delrin®. A hole at the bottom is used to evacuate the air from the Dewar flask wall with a mechanic pump.

The external diameters of the external and internal cylinders are, 127 and 101 mm respectively, and they have a corresponding wall thickness of 2 mm and 3 mm. A polyethylene foam cover of 10 cm thick, is placed at the top of the Dewar flask to help with the preservation of the nitrogen bath. The lengths of the internal and external cylinders are 67 cm and 65 cm respectively, so the maximum volume of liquid nitrogen that can be stored in the flask (without the moderator) is around 5 liters.

The mechanical resistance of the aluminum pieces was evaluated by simulating the first deformation mode with the software Autodesk Inventor™. Applied internal and external pressures of 137.9 kPa on each cylinder (1.4 times higher than atmospheric pressure) were simulated. The cylinders reached and exceeded the security factor established in ref [20]). Figure 2 shows the complete geometry of the cold neutron source on its mounting.

Two moderators (shown in figure 3) with similar geometries made of polyethylene and mesitylene were produced. The Polyethylene moderator was a solid half cylinder 13 cm height, 3.5 cm thick and 9 cm of external diameter. Monte Carlo simulations (described in section 1.3) to optimize the neutron flux, helped to determine these dimensions. Being a liquid at room temperature, mesitylene was contained in an aluminum semi-cylinder can, of 2 mm wall-thickness and 9 cm diameter. This recipient has a Swagelok™ connector in the filling hole, where a small PT100 platinum resistance is inserted inside the mesitylene. This resistance allows us to check in real time the moderator temperature during the experiment. On the other hand, the polyethylene moderator temperature was not controlled inside the material. The resistance in this case was just over the moderator as a nitrogen minimum level sensor. Figure 3 shows both moderators, the polyethylene semi-cylinder and the semi-cylindrical aluminum can containing the mesitylene .

1.3 Monte Carlo simulations

Monte Carlo simulations were performed using MCNP code [22]. This code solves the integral transport equation by simulating particle histories in any geometry regardless its complexity and

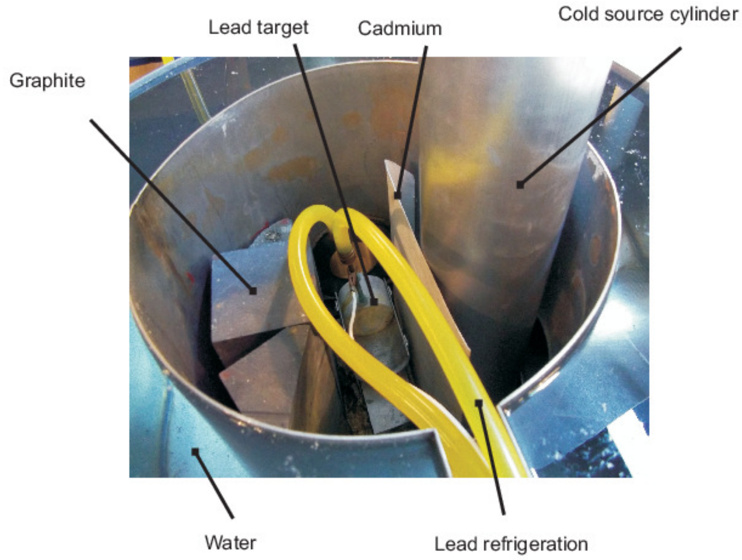

Figure 2. Geometry of the cold neutron source mounted inside the biological shielding.

Figure 3. Neutron moderators. Left: mesitylene aluminum container. Right: polyethylene semi-cylinder.

including a description of materials by means of cross section libraries. Calculations can be run in several transport modes. In particular in this work, “neutron only” mode was employed. Variance reduction techniques can also be used to improve efficiency. The geometry used for the calculations is shown in figure 4 . It describes the cylindrical Dewar flask containing a semi-cylindrical moderator placed in the biological shielding around the lead target were neutrons are produced. Parametric studies were performed by varying the parameter D of the mesitylene moderator at 77 K, shown in figure 4. To this end, a cross section library at this temperature was generated using the code NJOY [21] which was fed with the frequency spectra already validated for this material at a close temperature [23]. All simulations were run in a cluster facility at Centro Atómico Bariloche, which involves 16 Intel ® Hyper-Threading processors. Neutron flux was calculated at 800 cm from the moderator where real detectors are placed in an experiment. Growing importance along the flight tube and a source biasing were used in order to improve the calculation by reducing the calculation time. All the calculated spectra were expressed in units of counted neutron per source particle, as

2013 JINST 8 P11009

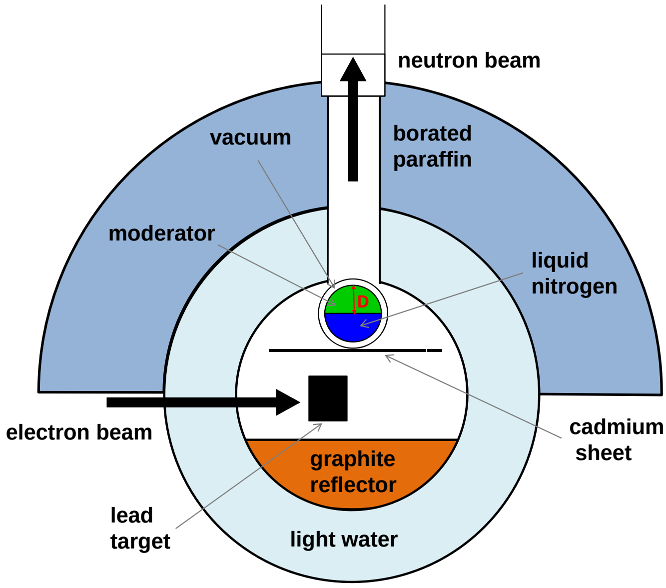

Figure 4. Geometry used as input for MCNP calculations, representing the moderator inside the Dewar flask and the biological shielding around the lead target. The dimension *D* that characterizes the moderator size is indicated.

usual in MCNP tallies. In figure 5 we observe that a semi-cylinder and a full cylinder of mesitylene at 77 K, (D = 4.15 and D = 8 cm respectively), produce a similar flux. In the inset we observe an improved subthermal flux in the moderator of D=4.15 cm compared to the fluxes of the thinner semicylinders with D$<$ 4.15 cm. The results of these simulations allowed to select an optimum size to construct the moderator.

2 Experimental setup

Neutrons were detected at 800 cm from the moderator position with a set of five ^{3}He detectors, 10 MPa of gas pressure, 2.54 cm diameter and 15 cm of active length. Neutron pulses (from the discriminator unit) were processed by a Canberra MCS unit through an AIM NIM module remotely connected by Ethernet with the Genie 2000 software in a PC. This electronic line allowed us to determine the neutron spectra as a function of time of flight, that were subsequently converted to neutron wavelength and/or energy. The start signal for the MCS unit was provided by a photo-multiplier (PM) tube located inside the bunker of the linear accelerator. This PM registers the gamma-flash produced at the precise moment when a packet of 25 MeV electrons hits the lead target, thus indicating the beginning of a LINAC pulse.

2.1 Experimental tests

Three main aspects were examined in the evaluation of the cold neutron source performance: the duration of the liquid nitrogen bath during the operation of the accelerator, a comparison between the neutron production of polyethylene and mesitylene moderators, and the necessary time to reach an equilibrium temperature in the moderator to ensure the stability of the neutron energy distribution.

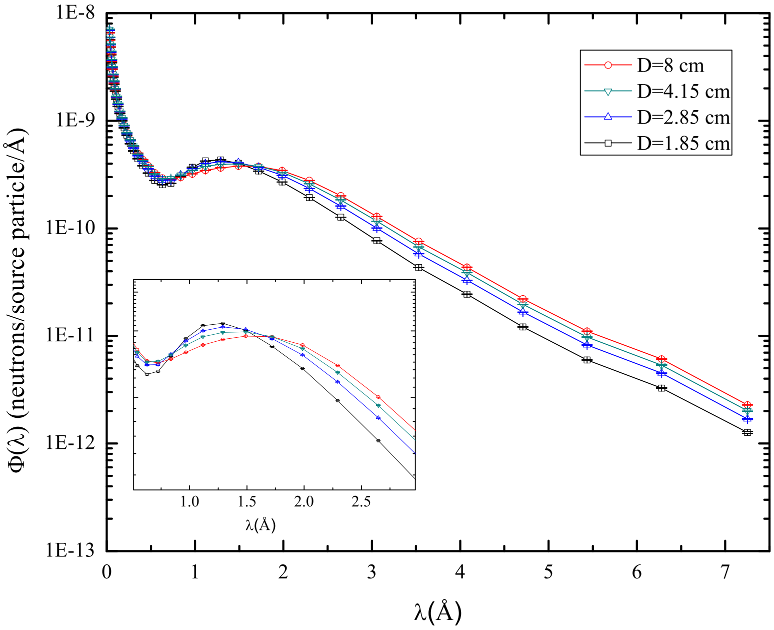

Figure 5. Mesitylene moderator spectra calculated for different moderator thicknesses *D*. In the inset, a detail of the thermal peaks is shown.

Autonomy. The autonomy or duration of the liquid nitrogen bath was studied with the polyethylene semi-cylinder as moderator. Once the vacuum inside the flask wall reached a value of 4 Pa, we made a pre-cooling bath of nitrogen during an hour before starting the irradiation, assuming that the polyethylene could reach an equilibrium temperature slowly. Then just before starting with the irradiation the Dewar was filled up to a safe level of 80% of its capacity. All the irradiations for this work were performed with the accelerator operating at 50 pulses per second and a mean electron current of 10 μA. In this condition we obtained 6h 50m of autonomy without refilling liquid nitrogen. The full capacity of the liquid nitrogen reservoir (4 liters), is enough to maintain the moderator at 77K for about 8h 30m.

Neutron production: polyethylene and mesitylene. The experimental neutron spectra of both moderators at room temperature and at 77 K are compared in figure 6, together with the background. The acquisition time was twenty to fifty minutes per spectrum. The background spectrum was measured for twelve minutes, placing a polyethylene and cadmium cover in the hole from where the neutron beam emerges. All the neutron spectra were normalized by the number of LINAC pulses during the measurement, and the detector efficiency was taken into account [19] as well as the deterction area to determine the flux in neutrons per square centimeter per second and per angstrom. Calculations by MCNP shown in figure 7 exhibit the same relative behavior.

From the observation of figures 6 and 7 it is clear that for both moderators, the value of the most probable neutron wavelength increases as the moderator is cooled down. It is also noticeable that the neutron production increases for wavelengths larger than 1.8 Å when any of the moderators is cooled down up to liquid nitrogen temperature. Considering the range 1.8 Å to 8 Å the increments in neutron fluxes from room to liquid nitrogen temperatures are: $8.48\,10^{-4}\pm1\,10^{-6}$ to

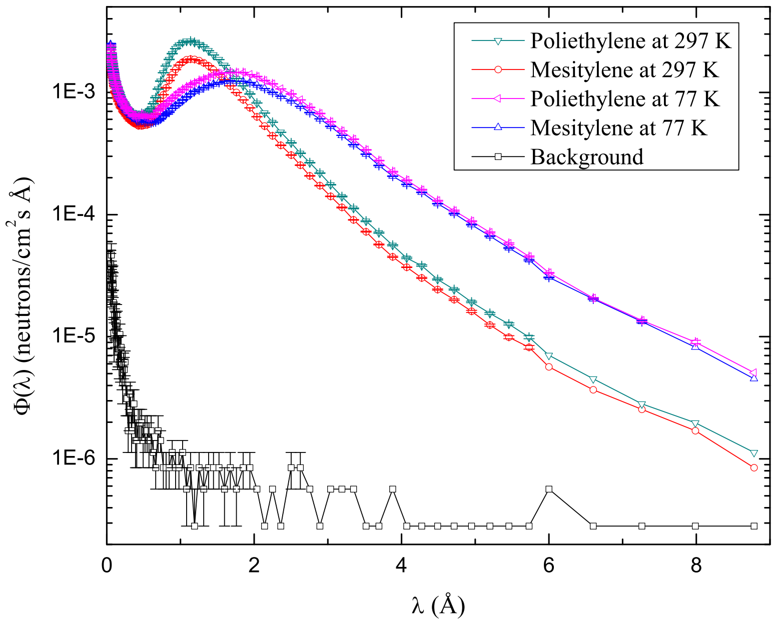

Figure 6. Measured neutron spectra of polyethylene and mesitylene moderators at room temperature and 77K, as a function of the neutron wavelength.

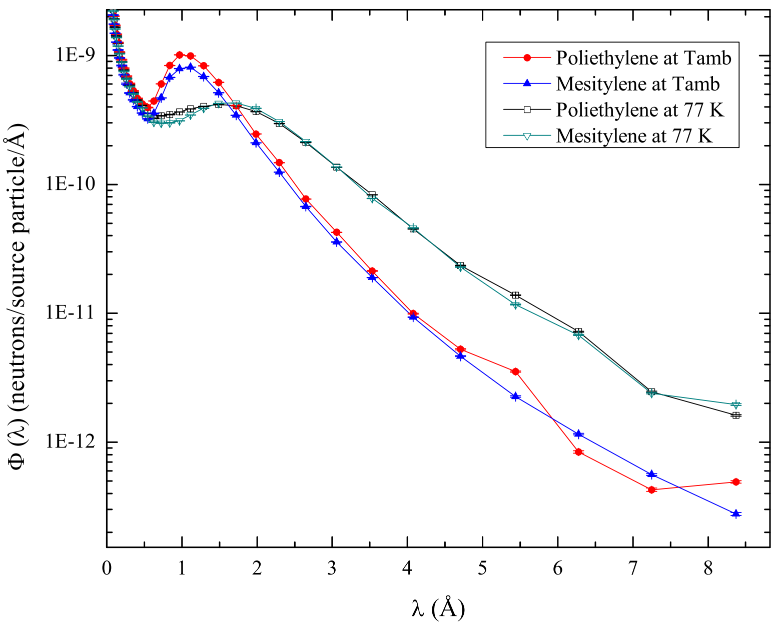

Figure 7. Calculated neutron spectra for the mesitylene and the polyethylene moderators at 77 K and room temperature. Neutron production for polyethylene is higher at room temperature. At 77 K both moderators show equivalent productions.

$1.920\,10^{-3}\pm2\,10^{-6}$, and $6.65\,10^{-4}\pm7\,10^{-7}$ to $1.702\,10^{-3}\pm2\,10^{-6}$ for polyethylene and mesitylene respectively, in neutrons/cm^2/s. The lower integration limit was arbitrarily chosen as the crossover point of the room temperature and cold spectra.

2013 JINST 8 P11009

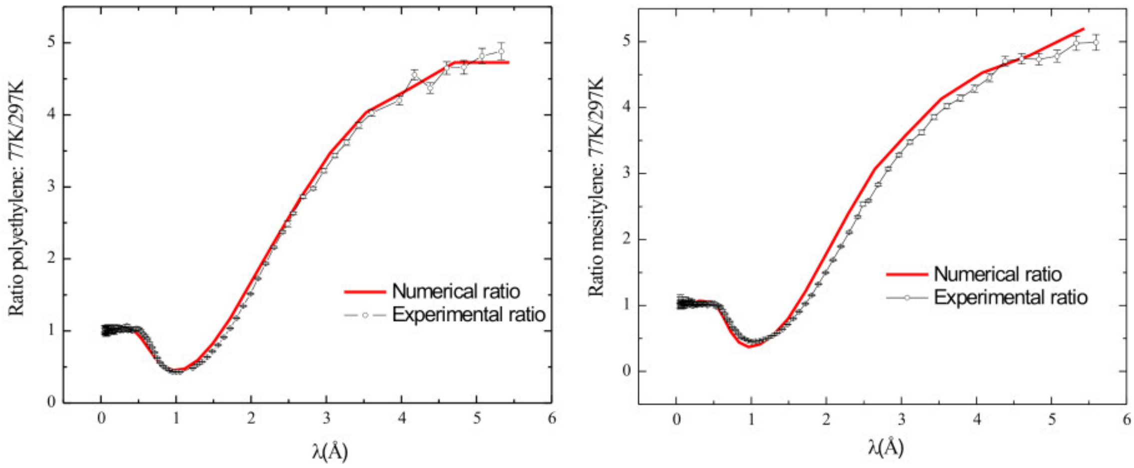

Figure 8. Production ratio of the mesitylene (left) and polyethylene (right) moderators at 77 K with respect to room temperature as a function of neutron wave length. In both, the neutron production at 77K increases starting from 2.5 Å, up to five times at 6 Å.

A figure of merit can be drawn to analyze the moderators performance, defined by the ratio of spectra at different temperatures (77 K vs. 297 K). Figure 8 shows such ratios for both moderators, determined from MCNP calculations and experiments. From that results, it follows that with the relatively simple cooling device shown in this report, we can achieve a gain up to five times in neutron production at wavelengths of 6 Å, with an appreciable increase starting from 2 Å.

Stability. Temperature stability during the experiment is an essential feature that the source must fulfill to be a useful device for neutron transmission and scattering experiments. When the moderator is cooled down, a transitory regime exists until the equilibrium temperature is achieved, in this case around 77 K. We employed two methods to control and characterize the stability of the moderator temperature and the neutron spectra. The first method is direct, and consisted in controlling the temperature of the moderator on-line during the irradiation. This was done for the mesitylene moderator with the platinum resistance inside the material. The values of temperature were read every second during the cooling down process, the irradiation period, and after the irradiation (when the liquid nitrogen level had dropped and the moderator temperature had warmed up naturally). The cooling down process up to about 77 K, with the moderator under a permanent liquid nitrogen bath, took around eighty minutes. Afterwards, we started an irradiation that lasted four hours with a moderator temperature of (76.74±0.17)K. The complete temperature evolution is shown in the figure 9.

The second method is indirect in terms of the temperature but direct in terms of the neutron distribution. We acquired neutron spectra and determined the wavelength at the maximum of each spectrum (λ_{max}) as a control parameter indicative of the temperature. The stability of this number indicated that, from a neutronic point of view, the moderator was in a stable condition. For mesitylene we obtained a mean λ_{max} of (1.79±0.15) Å, and for polyethylene (1.77±0.15) Å.

2013 JINST 8 P11009

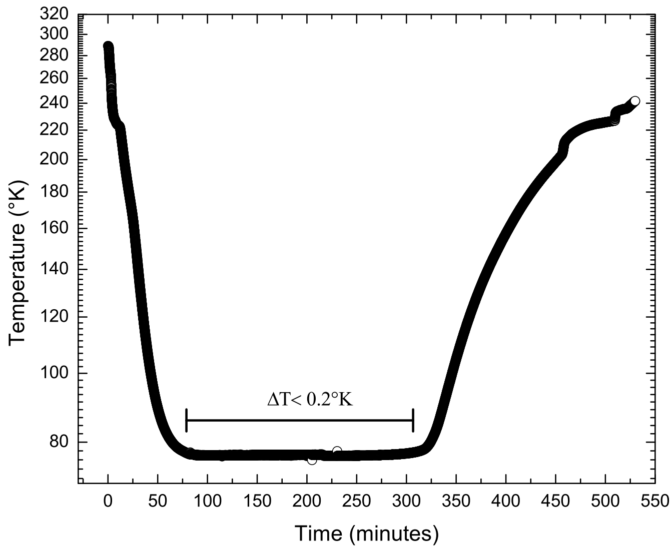

Figure 9. Temperature evolution of the mesitylene, showing the cooling-down period, and the temperature stability during the irradiation. The last period is the natural warming-up after irradiation.

3 Conclusions

We described a simple compact cold neutron source, designed to work in a small accelerator, based on a simple concept from the mechanical point of view. Two commercial cylinders of aluminum 6061 were enough to construct under security standards the main structure of the device. Although the Delrin® seal requires more design and test efforts, it is a commercial and relatively cheap material.

The main premise was to keep the neutron moderator under a liquid nitrogen bath as long as possible. To do this it was necessary to evacuate the air between the cylinders. With a mechanical vacuum pump we maintained a vacuum level of 4 Pa during the irradiations which was enough to keep at 77 K the 0.5 liter moderators for about seven hours with the accelerator operating at 50 pulses per second and starting with 80% of the nitrogen reservoir full, without refilling during the irradiation.

We evaluated the performance of two similar moderators, polyethylene and mesitylene. The results demonstrated the convenience of cooling down any of both moderators to increase the long wavelength neutron production up to five times compared with the same geometry at room temperature. Although similar spectra were obtained by cooling down both moderators, the mesitylene performance can be improved either by mixing it with toluene or by controlling the cooling down process in order to reach solid phase II instead of phase III as was analyzed in ref. [23]. In both cases, a cooler spectrum is achieved due to the larger population of low-frequency modes.

The subthermal neutron source opens the possibility to perform experiments that can be competitive in small-scale neutron sources. It is noteworthy in this regard, the determination of total cross-sections by the transmission technique, that proved to be useful to study small angle scattering phenomena, as an alternative to the well-known techniques based on large facilities.

Acknowledgments

This work was partially supported by the Universidad Nacional de Cuyo, ANPCyT (PICT 2011-534) and CONICET.

References

[1] J.R. Granada, J.R. Santisteban, J. Dawidowski and R.E. Mayer, *Neutron transmission: a powerful technique for small accelerator-based neutron sources*, *Phys. Procedia* **26** (2012) 108.

[2] International Atomic Energy Agency, *Development opportunities for small and medium scale accelerator driven neutron sources*, IAEA-TECDOC-1439 (2005) [ISBN 92-0-101705-7].

[3] J.R. Santisteban, L. Edwards, A. Steuwer and P.J. Withers, *Time-of-flight neutron transmission diffraction*, *J. Appl. Cryst.* **34** (2001) 289.

[4] M.R. Hawkesworth, *Neutron radiography: equipment and Methods*, *Atomic Energy Review* **15** (1977) 169.

[5] F. Cantargi, J.J. Blostein, L. Torres and J.R. Granada, *Thermal Neutron Cross section of Liquid and Solid Mesitylene*, *Nucl. Instrum. Meth.* **B 248** (2006) 340.

[6] M. Furusaka et al., *First results from a mini- focusing Small-Angle Neutron Scattering Instrument (mfSANS) with an ellipsoidal mirror*, in proceedings of *International Conference on Advanced Neutron Sources ICANS XVIII*, Dongguan, China, 26–29 April 2007.

[7] S. Petriw, J. Dawidowski and J.R. Santisteban, *Porosity effects on the neutron total cross section of graphite*, *J. Nucl. Mater.* **396** (2010) 181.

[8] R.E. Mayer, J.R. Granada, V.H. Gillette and J. Dawidowski, *High efficiency moderator for pulsed neutron diffraction*, *Nucl. Instrum. Meth.* **A 288** (1990) 467.

[9] L. Torres and J.R. Granada, *Design of a cold-neutron source for the Bariloche LINAC with solid mesitylene as moderator material*, *Physica* **B 385-386** (2006) 1306.

[10] E. Shabalin, E. Kulagin, S. Kulikov and V. Melikhov, *Experimental study of spontaneous release of accumulated energy in irradiated ices*, *Radiat. Phys. Chem.* **67** (2003) 315.

[11] E. Kulagin, S. Kulikov, V. Melikhov and E. Shabalin, *Radiation effects in cold moderator materials: experimental study of accumulation and release of chemical energy*, *Nucl. Instrum. Meth.* **B 215** (2004) 181.

[12] E. Shabalin, S. Kulikov, V. Melikhov and E. Kulagin, *Some Radiation Effects In Cold Moderator Materials, Experimental Study*, in proceedings of *16th Meeting of the International Collaboration on Advanced Neutron Sources*, volume II, Düsseldorf-Neuss, Germany, 12–15 May 2003, G. Mank and H. Conrad eds. (2003) pg. 911.

[13] K. Unlu, C. Rios-Martinez and B.W. Wehring, *Prompt Gamma Activation Analysis with the Texas Cold Neutron Source*, *J. Radioanal. Nucl. Chem.* **193** (1995) 145.

[14] L. Cser and P. Vértes, *Optimization study of a simple geometry spallation source target design*, *J. Neut. Res.* **6** (1997) 185.

[15] F. Cantargi, J.R. Granada, S. N. Petriw and M.M. Sbaffoni, *New scattering kernels for some materials of interest as advanced cold neutron moderator*, *Physica* **B 385-386** (2006) 1312.

[16] S. Kulikov, A. Belyakov, M. Bulavin, K. Mukhun, E. Shabalin and A. Verhoglyadov, *Current Status of Advanced Pelletized Cold Moderators Development for IBR-2M Research Reactor*, *Phys. Part. Nuclei Lett.* **10** (2013) 147.

[17] I. Natkaniec and K. Holderna-Natkaniec, *Structural phase transitions and dynamics of solid mesitylene investigated by diffraction and inelastic incoherent neutron scattering methods*, in proceedings of *AcoM-6 Meeting*, Juelich, Germany (2003) [ISSN 1433-5506].

[18] T.V. Christy, N. Murugan and S. Kumar, *A Comparative Study on the Microstructures and Mechanical Properties of Al 6061 Alloy and the MMC Al 6061/TiB_2/12_p , J. Minerals & Materials Characterization & Engineering* **9** (2010) 57.

[19] L.A. Rodríguez Palomino, J.J. Blostein and J. Dawidowski, *Calibration and absolute normalization procedure of a new Deep Inelastic Neutron Scattering spectrometer*, *Nucl. Instrum. Meth.* **A 646** (2011) 142.

[20] J.H. Moore, C.C. Davis and M.A. Coplan, *Building Scientific Apparatus*, 4th edition, Cambridge University Press, New York, U.S.A. (2009).

[21] R.E. MacFarlane and D.W. Muir, *The NJOY Nuclear Data processing System*, Los Alamos National Laboratory Report LA-12740-M (1994).

[22] J.F. Briesmeister, *MCNP, a General Monte Carlo Code for Neutron and Photon Transport*, Los Alamos National Laboratory (1986).

[23] F. Cantargi and J.R. Granada, *Thermal neutron cross-section libraries for aromatic hydrocarbons*, *Nucl. Instrum. Meth.* **B 268** (2010) 2487.

2013 JINST 8 P11009