EPJ Web of Conferences **298**, 01003 (2024) https://doi.org/10.1051/epjconf/202429801003
UCANS-10

Ethane as a Neutron Moderator at Cryogenic Temperatures

Monia El Barbari, *Ulrich* Rücker, *Alexander* Schwab, *Junyang* Chen, *Paul* Zakalek, *Jingjing* Li, *Thomas* Gutberlet, and *Thomas* Brückel

Jülich Centre for Neutron Science JCNS-HBS, Forschungszentrum Jülich GmbH,
52425 Jülich, Germany

Abstract: The study's objective is to examine the neutron temperature response, using liquid and solid ethane at various cryogenic temperatures to evaluate the effectiveness of ethane as a cold neutron moderator in compact moderator systems. The experimental measurements were carried out at temperatures of 170 K, 135 K, 100 K, 70 K, 50 K, 25 K, 15 K, and 10 K. Time-of-flight neutron spectra were measured with an instrument at the JULIC Neutron Platform. The effective temperatures of the neutron spectrum emitted are determined by fitting the Maxwellian distribution as a function of the wavelength.

1 Introduction

The cold neutron moderator is a vital component in neutron scattering instrumentation, in which it plays a key role in slowing down thermal neutrons, enhancing their suitability for studying atomic and molecular structures in materials with higher resolution compared to investigations with thermal neutrons. Over the past years, many neutron facilities opted for developing low-dimensional cold moderators which refer to finger-like moderators with lengths of the same magnitude as the neutron's mean free path, which depends on the materials that the moderator is filled with [1]. Specific design and moderator material choices are made to efficiently moderate neutrons while minimizing absorption. These designs conditions are crucial for maximizing the number of low-energy neutrons available for scientific investigations in condensed matter physics and materials science. For this reason, many studies researching different geometry optimizations and featuring diverse materials have been performed [2]. This paper is a contribution to these studies to characterize another material to develop low-dimensional cold moderators that can be used in an efficient way to help users of different fields to access high-performing while flexible instrumentation in the neutron research facilities.

The selection of a moderator material involves a delicate balance between efficiency, stability, and safety, particularly in environments prone to intense radiation fields. In this aspect, ethane emerges as a promising candidate offering advantages in terms of stability and safety. Ethane in the solid state exhibits different phases depending on temperature and pressure conditions. These phases include a crystalline form stable between 60 and 70 K, a metastable phase between 30 and 55 K, and an amorphous phase effective at temperatures below 30 K [3]. Methane hydrate can be an alternative too, but its reduced intensity of neutrons makes it less attractive for applications requiring high brightness [4].

2 Experimental setup

2.1 Moderator assembly

In Figure 1, the setup of the ethane cold moderator vessel is depicted. The volume of the moderator material is enclosed in a cylindrical aluminum vessel with 45 mm in length similar to the main free path of hydrogen and an outer diameter of 22 mm due to geometrical constraints of the moderator-reflector unit. Around the moderator volume there is a labyrinth structure through which cold helium gas flows to cool the moderator material.

This vessel is integrated into a beam extraction plug, wherein the four supply lines (two for the He-cooling circuit and two for ethane supply and exhaust) and the neutron extraction optics are accommodated. This plug is positioned within one of the extraction channels of the target station shielding of the JULIC Neutron Platform [5,6]. The moderator volume together with all supply lines are contained in a common vacuum vessel which is evacuated down to a pressure of 10^{-6} mbar for thermal isolation.

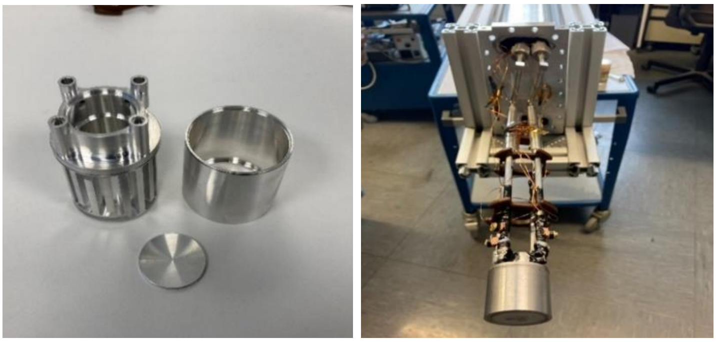

Fig. 1. Left: the moderator vessel with ethane and helium cryogenic supply lines. The helium cooling flows through the outer labyrinth. Ethane is condensed and frozen inside the inner volume. Right: a flexible mounting configuration with the moderator volume and four temperature sensors for adaptive experimental setups at the front of an extraction plug containing the neutron.

The vessel is then filled with ethane gas (it can also be replaced with different gaseous materials, provided that they possess comparable properties) to be condensed to liquid and later frozen to the solid state. The temperature control of the He cooling is used to determine the temperature of the moderator.

The extraction plug with the cold moderator at its front is placed in a moderator-reflector assembly made from polyethylene as thermal moderator and lead as reflector.

2.2 Instrumentation

A diffractometer instrument at the JULIC Neutron Platform is used to measure the spectra of the ethane moderator. Its main components are the extraction plug with the moderator vessel connected to an evacuated neutron guide of 6.5 m length, coated with ^{58}Ni. The cross-section of the guide is 30 x 45 mm² (width x height). At the end of the guide, ^{3}He detectors (4 bar gas pressure, 25 mm outer diameter) are placed. The detectors are situated inside a shielding box (polyethylene blended with B_2O_3) to reduce the background effect from neutrons that do not pass through the neutron guide.

The spectra are recorded in Time-of-Flight (ToF) mode with a time resolution of 200 μs. Primary neutron pulses are produced mainly by the (p,n) interaction of 45 MeV protons with a tantalum target in front of the moderator-reflector assembly. The proton pulse length during the experiments described here is 400 μs. After the reference measurement with an empty moderator vessel ("Thermal" in Figure 2), the experimental procedure involved the initial insertion of ethane gas into the moderator vessel, followed by a controlled condensation process achieved through the cooling of the gas using helium to reach the desired temperature. Moderator temperatures set during the measurements included: 170 K, 135 K, and 100 K representing the liquid state, 70 K for the solid state and crystalline phase, 50 K for the second phase characterized by a metastable state, and subsequently 25 K, 15 K, and 10 K for the amorphous phase of solid ethane.

3 Measurements

The neutron spectra were obtained using the Time-of-Flight method using a ^{3}He detector immediately at the exit of the neutron guide, with a measurement duration of 30 minutes for each temperature. This approach ensured detailed data collection and a consistent analysis of the ethane's behavior at different temperatures. Fig. 2 represents the data collected.

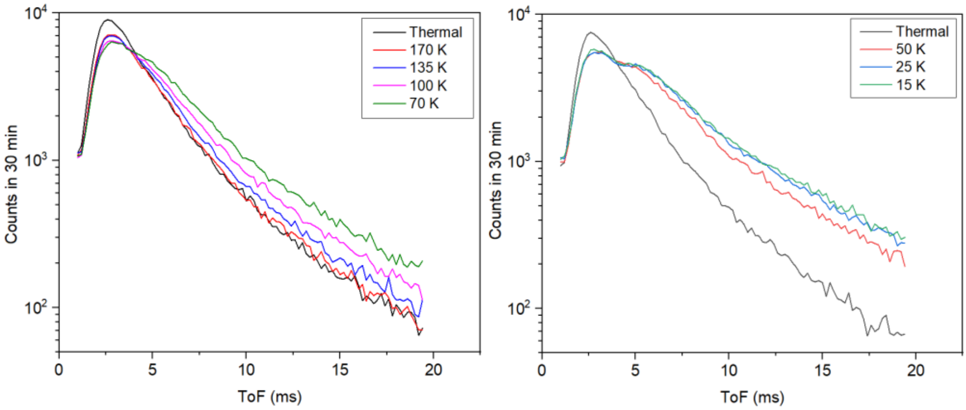

Fig. 2. Neutron spectra emitted from the ethane cold neutron moderator at different temperatures measured in Time-of-Flight at the end of a 6.5 m long neutron guide.

The two graphs (Fig. 2) depict the data collected at distinct times, signifying a different thermal contribution to consider for each group of temperatures included in the same test. The thermal contribution to each spectrum is evident through a small peak at t = 2.5 ms consistently present at every temperature measured.

3.1 Geometric and correction processing

In the experimental setup, the moderator vessel has a cylindrical form in front of a bigger rectangular guide. For this reason, the guide is fed by the cold neutrons emitted from the cold moderator volume as well as by the thermal neutrons accepted from the area around the cold source. To mitigate this thermal contribution, it can be subtracted taking into account the fraction of the thermal neutrons left after adding the moderator cylinder, which is equal to

S_g-S_c/S_g = 0.747, where S_g is the guide surface and S_c the cylinder surface of the cold moderator.

The subsequent normalization takes into account the total intensity measured at the monitor detector to receive the proper relation to the total number of neutrons produced.

Additionally, the impact of the wavelength-dependent transmission of the guide must be addressed as a factor dependent on the square of the wavelength. Fig. 3 presents the conclusive results of the cold neutron spectra as a function of wavelength of all the measured temperatures.

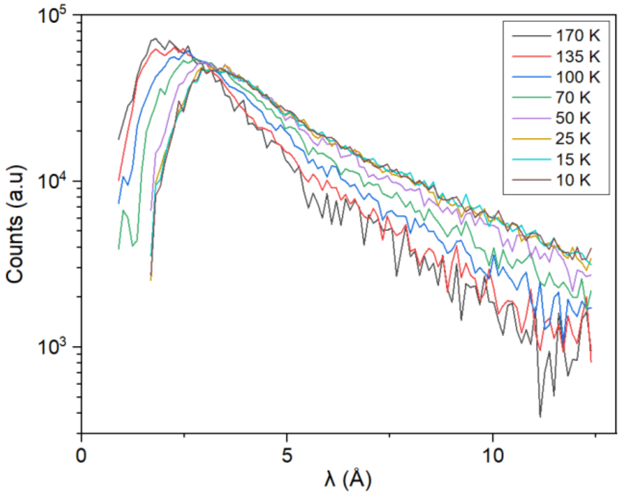

Fig. 3. Neutron spectra emitted by the ethane cold neutron moderator as a function of the wavelength for different moderator temperatures.

As expected, the spectrum's peak undergoes a shift towards higher wavelengths in colder neutron regimes as the temperature decreases. This is in accordance with expectations and indicates an increase in moderation efficiency. However, this shift becomes less evident as we approach 25 K and below. This will be reflected in the neutron temperature that is discussed in the following section.

3.2 Neutron temperature extraction

To extract neutron temperatures from the neutron spectra presented in the previous section, we employ a fitting process wherein we fit the neutron data to the normalized Maxwell wavelength distribution characterized by the following function:

$$M(\lambda,T) = 2\, a^2\, \lambda^{-5} \exp(-a/\lambda^2) \qquad (1)$$

where λ represents the wavelength in units of Å, T denotes the spectral temperature in K, and a is a parameter calculated as $950.52/T$, determined by the wavelength and temperature of a 25 meV neutron [7]. It may also be necessary to introduce a fitting parameter to appropriately adjust the spectrum amplitude to align with the provided spectrum. Fig. 4 illustrates an example of the Maxwellian distribution fitting corresponding to the ethane temperature of 170 K.

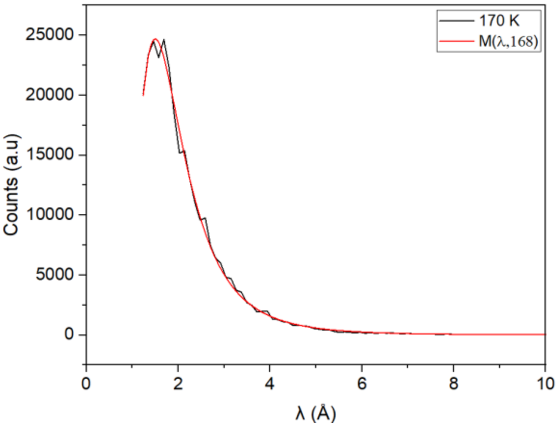

Fig. 4. The Maxwellian distribution in wavelength fitted to the neutron spectrum for the ethane moderator at 170 K.

The same method is used to fit the other spectra of different temperatures to determine the neutron flux real temperature, which is a parameter depending on the shift of the spectra along the wavelength values. As can be observed in Fig.3, these shifts become smaller as the temperature decreases until there is an almost clear superposition of the spectra positions at very low temperatures. This is more evident in Fig.5, which illustrates the variation of neutron temperatures noted T_n with respect to the ethane moderator material temperature noted T_m.

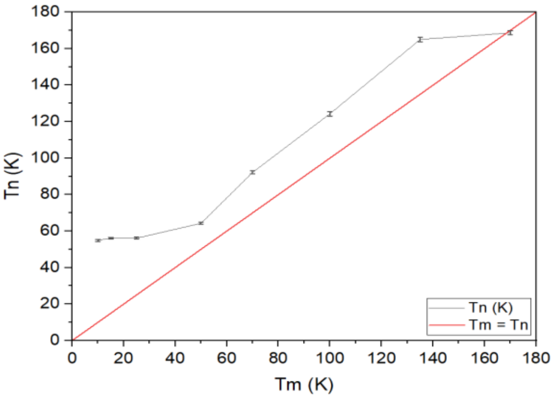

Fig. 5. The temperature of the emitted neutron spectrum compared to the moderator temperature.

As neutrons traverse through the moderator material, interactions with its constituent elements lead to energy transfer and absorption. These interactions result in a thermalization (cooling) process where neutrons lose kinetic energy and, consequently, experience a decrease in temperature, which basically is the role of such moderator materials. The neutron temperature in general seems to be higher than the moderator temperature, also taking a statistical error of around 1 K into account. This can be attributed to the small volume of the cryogenic moderator resulting in under moderated neutrons. At moderator temperatures below 50 K (i.e. in the amorphous phase) the neutron temperature does not decrease further, but the neutron temperature shows an asymptotic behavior towards 57 K. This indicates that ethane might not be perfectly suitable to produce very cold neutrons or other geometries with larger volumes need to be considered. Further investigations are being conducted.

4 Conclusion

In this study, we investigated the performance of ethane as a cold neutron moderator material in both its liquid and solid states across a range of temperatures. By fitting the experimental data to the Maxwell distribution, the effective temperature of the neutron flux was estimated. The results obtained demonstrated the ability of ethane to efficiently cool the neutron flux to low temperatures down to 60 K.

As ethane exhibits superior safety and stability compared to other cold moderator materials, it is a suitable choice for future endeavors in this field. Previous research has been conducted on ethane material as a cold neutron moderator [8,9]. However, it is essential to acknowledge that certain aspects of ethane's behavior as a cold neutron moderator still require more detailed and comprehensive examination which will be addressed in forthcoming studies, aiming to enhance our understanding and optimize the utilization of ethane as a neutron moderator material in future applications.

“This project has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant No 101034255.”

References

1. F. Mezei, L. Zanini, K. Batkov, E. Klinkby, A. Takibayev, J. Phys.Conf. Ser. **1021**, 012009 (2018)
2. F. Mezei, L. Zanini, A. Takibayev, K. Batkov, E. Klinkby, E. Pitcher, T. Schönfeldt, J. Neutron Res. **17**, 2 (2014)
3. R.L. Hudson, P.A. Gerakines, M.H. Moore, J. Icarus. **243** (2014)
4. M. H. Parajon, E. Abad, F.J. Bermejo, Phys. Procedia **60** (2014)
5. P. Zakalek, J. Baggemann, J. Li, U. Rücker, T. Gutberlet, T. Brückel, “The JULIC Neutron Platform, a testbed for HBS”, to be published in these proceedings
6. T. Brückel, T. Gutberlet, J. Baggemann, J. Chen, T. C. Weber, Q. Ding, M. El Barbari, J. Li, K. Lieutenant, E. Mauerhofer, U. Rücker, N. Schmidt, A. Schwa, J. Voigt, P. Zakalek, Y. Bessler, R. Hanslik, R. Achten, F. Löchte, M. Strothmann, O. Felden, R. Gebel, A. Lehrach, M. Rimmler, H. Podlech, O. Meusel, F. Ott, A. Menelle, M. A. Paulin, EPJ Web Conf. **286**, 02003 (2023)
7. S. Tasaki, Y. Idobata, Y. Adachi, F. Funama, Y. Abe, EPJ Web Conf. **231**, 04005 (2020)
8. F. Cantargi, J.R. Granada, J.I. Márquez Damian, EPJ Web Conf. **146**, 13003 (2017)
9. J.I. Robledo, J. Dawidowski, F. Cantargi, J.R. Granada, L.A. Rodríguez Palomino, G. Romanelli, M. Krzystyniak, G.J. Cuello, C.S. Helman, G. Škoro, j.nima. **1055**, 168501 (2023)