Development of thermal scattering kernels for sodium hydroxide

Esben Klinkby[1,*] , *José Ignacio* Marquez Damian[2], *Douglas* D. DiJulio[2], *Günter* Muhrer[2], *Matthew* Krzystyniak[3], *Ali* Mortazavi[3], *Chris* Goodway[3], and *Giovanni* Romanelli[4]

[1]Seaborg Technologies, Titangade 11, 2200 Copenhagen, Denmark
[2]European Spallation Source, ESS ERIC, Partikelgatan 2, 224 84 Lund, Sweden
[3]STFC UKRI, Rutherford Appleton Laboratory, Harwell Oxford, Didcot OX11 0QX, United Kingdom
[4]Physics Department, University of Rome Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy

Abstract. NaOH was recently proposed as a moderator for the compact molten salt reactors, under development by Seaborg Technologies. In order to reliably predict the thermalisation of neutrons in such a reactor, the simulations must be based on modelling which accurately accounts for the low-energy neutron interactions in NaOH. This information is contained within thermal scattering libraries that are used during Monte-Carlo simulations. In this paper, we present the results of neutron scattering experiments carried out at VESUVIO, ISIS in order to facilitate the development thermal scattering libraries for solid NaOH.

1 Introduction

In recent years, molten salt reactor (MSR) concepts have seen renewed interest both within academia and emerging industry. Several widespread reactor concepts are under development, sharing the feature that the fissile fuel is dissolved in a molten fluoride or chloride salt which remains liquid over a wide temperature range at atmospheric pressure. One of the attractive features of MSRs is precisely that high pressure is avoided, thereby eliminating a range of possible accident scenarios. Combining this with the fuel salt fission product retention properties and the ability to do online fission product processing paves the way for a completely different safety case for MSR concepts with respect to conventional reactors, potentially allowing significant savings on construction costs.

The MSR concept is not new. Two reactors were built and operated at ORNL in the 50's and 60's [1], but the technology never matured into commercial reactors. Though generally successful according to the subsequent assessment reports, the operation of these reactors did identify several challenges. Among the more concerning were (irradiation assisted) corrosion issues of the structural material, while another relates to the mechanical and moderation properties of the graphite moderator under irradiation [2]. Although significant efforts have been made, the issues relating to the graphite behaviour under irradiation have not been resolved during the half century that has passed. At present, some MSR developers are focusing their research on solving this issue [3], while others plan frequent replacements of the core, to limit irradiation damage [4]. At Seaborg Technologies, a Danish based MSR developer, a different path is chosen, namely to replace the graphite moderator with sodium hydroxide (NaOH) [5]. While this introduces several complications relating to corrosion and chemistry control, it does also promise several benefits. During operation of the reactor, the NaOH would be liquid, thus eliminating issues relating to radiation damage. Additionally, due to the presence of hydrogen, the moderation power is significant and allows for a compact core design, which is essential for the assembly line based deployment and central to Seaborg's business case. Another challenge following the use of sodium hydroxide relates to the fact that only limited past experience is available in the literature [6]. For energetic neutrons (>1eV), the interactions with materials can be reliably predicted using Monte-Carlo techniques using nuclear data per nuclide. However, in the low-energy regime, the interactions are instead dominated by the structure and dynamics on an atomic and molecular level.

2 Experimental setup

To facilitate the usage of NaOH as a moderator, the modelling must be supplemented with thermal scattering kernels describing interactions between neutrons and NaOH in the thermal regime. The development of such scattering kernels relies on experiments as well as modelling.

To serve as a benchmark for such a kernel, we carried out total cross section measurements using the pulsed neutron beam available at the ISIS Pulsed Neutron Muon Souce, at the Rutherford Appleton Laboratory (UK). The total cross section was determined over a wide range of energies using the time-of-flight (TOF) technique.

For the present work, we carried out experiments in May 2020 using the VESUVIO+ beamline, which is a thermal-to-epithermal neutron station optimised for the routine measurement of the total cross sections over a broad energy range[21–23] - see figure 1. A sample of

*e-mail: Esben.Klinkby@seaborg.com

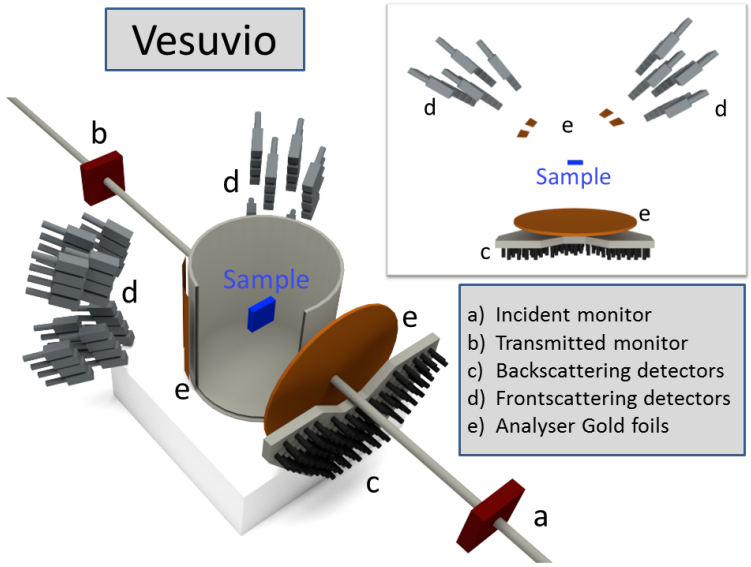

Figure 1. VESUVIO instrument at ISIS. Source: [8].

8.47g of NaOH powder at room temperature was prepared as shown in figure 2 [9], and placed centrally in the neutron beam.

The samples were pressed to 1.5 mm thickness inside a thin polyethylene bag in order to avoid direct contact with the aluminium container and corrosion effects. The sample, polyethylene bag and container had surfaces perpendicular to the neutron beam which where larger than the beam profile in order to avoid undesired scattering from the sample container. The thickness value was chosen to optimise the transmission spectrum across the entire energy range discussed in this work.

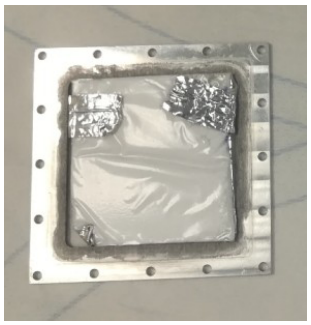

Figure 2. Sample of NaOH sealed in polyethylene.

3 Results

The incident as well as transmitted beams were monitored, allowing the TOF spectrum to be converted into a neutron energy spectrum, ranging over several orders of magnitude and covering the energy region of interest for thermal scattering kernel development (roughly: 1meV-0.1eV). In the post-processing, the measured data was normalised to the well-known epithermal free atom cross-section.

The resulting total cross-section measurement data is shown in figure 3, along with the results of two different modelling approaches. The differences in terms of physics modelling between the NJOY[10] and NCrystal[11] approaches is that the NCrystal model includes the (small) coherent component, while the NJOY model was computed in the incoherent approximation. Both models use the same phonon spectra for NaOH, which was calculated using ab-initio approaches taken from the Kyoto University database[12–15] and processed with Phonopy[16].

As seen in figure 3, the model predictions are very much aligned between the two approaches. Small deviations are visible in the meV region, but are covered by the uncertainty from the measurement results, illustrating that for NaOH, the coherent scattering contribution to the total cross-section is minor.

4 Towards measuring thermal scattering kernels for liquid NaOH

In the Compact Molten Salt Reactor under development by Seaborg Technologies [17], NaOH will serve as a moderator. During operations, liquid NaOH will fill up spaces between fuel tubes and will be circulated to ensure adequate spatial temperature distribution and to facilitate heat removal. Therefore, for the use-case considered here, liquid NaOH at elevated temperatures is of particular interest. From an experimental perspective, however, liquid NaOH at elevated temperatures is more difficult to assess, since it poses severe constraints on the sample environment. NaOH is corrosive, and more so at elevated temperatures, making it non-trivial to identify structural materials which are sufficiently corrosion and temperature

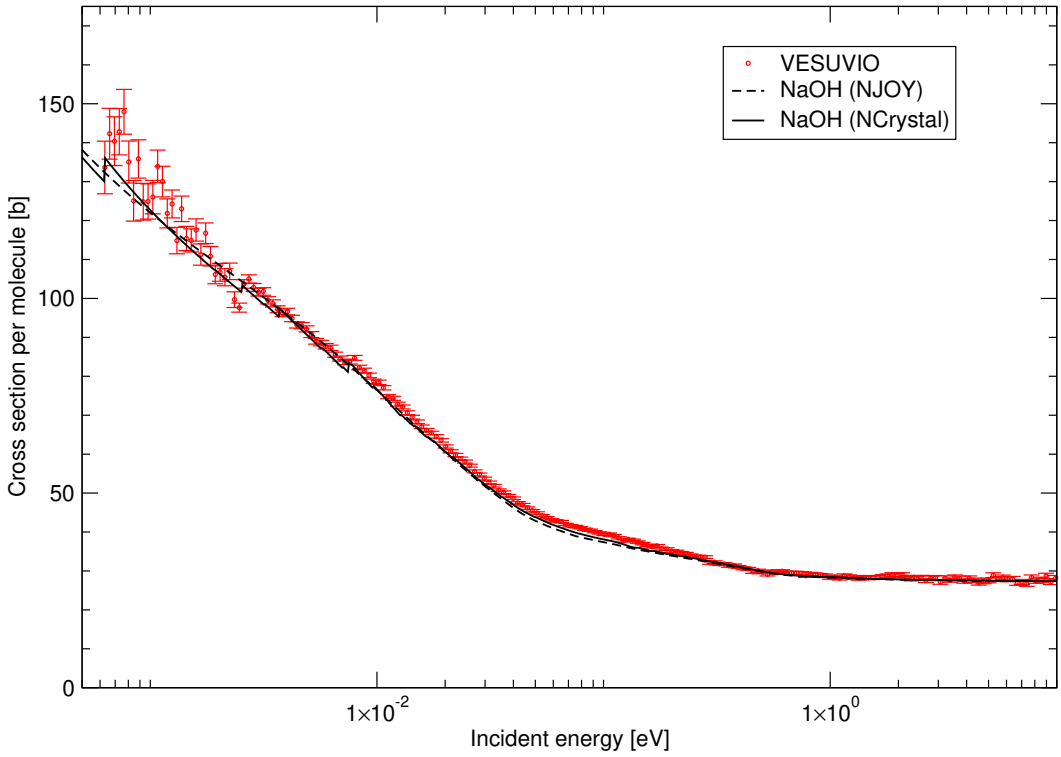

Figure 3. Measured and modelled total cross-section of solid NaOH at ambient temperatures.

resistant, while at the same time minimizing the scattering of the neutron beam and thus impacing the tranmission measurements.

None of the existing cells at the ISIS were able to cope with the requirements, so a new cell needed to be developed. After thorough investigations, Inconel alloy 625 was chosen as the structural material. In figure 4 the transmission probability for the developed cell is shown, while a picture of the completed cell is seen in figure 5.

5 Conclusions and outlook

Total transmission measurements of solid NaOH at ambient temperature were carried out at the VESUVIO+ instrument at ISIS. Using standard techniques, the total transmission measurements were converted into the energy dependent total cross-section, and compared to results from modelling approaches based on NJOY and NCrystal respectively. Given that satisfactory agreement between measurements and modelling was observed, the data was used to prepare thermal scattering libraries which, using NJOY or NJOY-NCrystal[19] can be used in Monte-Carlo codes such as MCNP and OpenMC. It is the ambition of the authors to facilitate that the data be made publicly available in general purpose databases such as e.g. JEFF[20]. In addition, preparations for transmission experiments of liquid NaOH at the VESUVIO beam-line

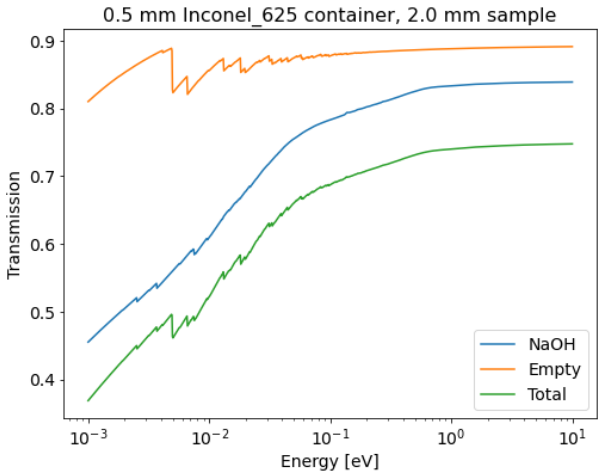

Figure 4. Transmission propability for the developed cell (orange), NaOH (blue) and total (green)

have been carried out and such experiments are presently being prepared.

References

[1] P. N. Haubenreich J. R. Engel, "Experience with the Molten-Salt Reactor Experiment", Nucl. App. and Tech. **8**, Issue 2, 118-136 (1970)

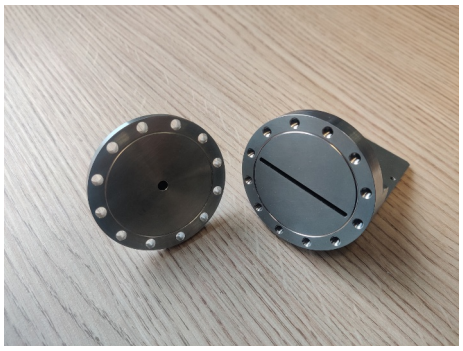

Figure 5. The Inconel 625 cell developed at ISIS to facilitate measurements of liquid NaOH at elevated temperatures.

[2] P. R.Kasten et al, "Graphite behavior and its effects on MSBR performance", Nucl. Eng. and Design, **9**, Issue 2, 157-195 (1969)

[3] G., Zhu et al, "Neutronic effect of graphite dimensional change in a small modular molten salt reactor",Int.J of Energy Research, **45**, Issue 8, 11976-11991 (2020)

[4] Examples include: https://www.terrestrialenergy.com/, https://www.terrapower.com/, https://thorconpower.com/

[5] Patent WO 2018/229265 Al - "Molten Salt Reactors"

[6] C.E. Teeter, et al.. Catalog Of Nuclear Reactor Concepts. Part I. Homogeneous and Quasi-Homogenous Reactors. Section III. Reactors Fueled With Molten-Salt Solutions. No. ANL-7092. Argonne National Lab., Ill. (1965)

[7] J.I. Robledo, et al, "Measurement of neutron total cross section at the VESUVIO spectrometer", Nucl. Inst. Meth. A, **A 971** 164096 (2020)

[8] https://www.isis.stfc.ac.uk/Pages/Vesuvio.aspx

[9] Sodium hydroxide white powder 757527. 25g ultra dry from Sigma Aldrich. https://www.sigmaaldrich.com/GB/en/product/aldrich/757527

[10] R. Macfarlane, et al. "The NJOY nuclear data processing system", version 2016. No. LA-UR-17-20093. Los Alamos National Lab.(LANL), Los Alamos, NM (United States), 2017.

[11] XX. Cai, and T. Kittelmann. "NCrystal: A library for thermal neutron transport." Computer Physics Communications **246** 106851, (2020)

[12] Togo A. Phonon database at Kyoto University (2015), [Online] http://phonondb.mtl.kyoto-u.ac.jp/

[13] A. Jain et al, The Materials Project: A materials genome approach to accelerating materials innovation APL Materials, 2013, **1(1)**, 011002.

[14] S. P. Ong, Python Materials Genomics (pymatgen) : A Robust, Open-Source Python Library for Materials Analysis. Computational Materials Science, 2013, **68**, 314-319.

[15] S. P. Ong et al, The Materials Application Programming Interface (API): A simple, flexible and efficient API for materials data based on REpresentational State Transfer (REST) principles. Computational Materials Science, 2015, **97**, 209–215.

[16] Togo A., Tanaka I., First principles phonon calculations in materials science Scr. Mater., **108** (2015), pp. 1-5

[17] https://www.seaborg.com/

[18] https://www.specialmetals.com/documents/technical-bulletins/

[19] K. Ramic et al., Nuclear Inst. and Methods in Physics Research, A **1027** 166227 (2022)

[20] https://www.oecd-nea.org/dbdata/jeff/

[21] G. Romanelli, et al, "The road to a station for epithermal and thermal neutron analysis". VII International Workshop on Electron-Volt Neutron Spectroscopy, (2018) 1055,

[22] M. Krzystyniak et al, "VESUVIO plus : The Current Testbed for a Next-generation Epithermal Neutron Spectrometer". 22nd Meeting of the International Collaboration on Advanced Neutron Sources, ICANS XXII, 1021, (2018)

[23] C. Andreani et al, "Electron-volt neutron spectroscopy: beyond fundamental systems". Advances in Physics, **266**, 1-73, (2017)