New prospects of VESUVIO applied to measurements in water mixtures

L. A. Rodríguez Palomino¹, J. Dawidowski¹, ³, J. J. Blostein¹, G. J. Cuello²

¹ Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Comisión Nacional de Energía Atómica-Universidad Nacional de Cuyo, Argentina
² Institut Laue Langevin, 71, Av. des Martyrs, 38042 Grenoble, France
E-mail: javier@cab.cnea.gov.ar

Abstract. We present new measurements on mixtures of light and heavy water in the spectrometer VESUVIO (Rutherford Appleton Laboratory, UK), and analyze them from the perspective of different kinds of applications. We perform a single detector analysis and show the multiple scattering and attenuation corrections with the aim to employ them in mass spectrometry. We also show the capabilities to perform transmission measurements to determine total cross sections of an acceptable quality by means of its transmission monitor.

1. Introduction
In this paper we discuss the results of new measurements of various mixtures of light water and heavy water at spectrometer VESUVIO (ISIS, Rutherford Appleton Laboratory, UK). The use of water as a system of interest, far from being considered as a new chapter in an old debate, is used here as an experience to highlight different capabilities of VESUVIO that had not been employed intensively so far. Although designed as an instrument to explore momentum distributions, VESUVIO provides information on scattered neutrons on a wide range of angles and energies. In fact it is one of the few available instruments that offers the possibility of reaching epithermal energies up to about 100 eV (exclusive property of accelerator-based sources), while having available a substantial flux of thermal neutrons [1]. Also, it has a transmission monitor, placed at a distance from the source that makes it capable to measure energy-dependent transmission spectra with very good resolution in energy. Given these possibilities, the feasibility of finding new uses for this facility has already begun to be explored. Such is the case of Mass-selective neutron spectroscopy recently explored by Krzystyniak et al. [2].

In this work we focus attention on electron-Volt spectroscopy applications that do not need to go through transformation from time-of-flight to the momentum transfer y-scale [3]. In those cases it is more convenient to work on individual detector signals in the time-of-flight scale [4]. We select the detectors that show the best signals and show the steps necessary to perform Monte Carlo simulations to calculate multiple scattering and attenuation corrections, a prerequisite aiming to employ single-detector information in mass spectrometry. In the description of the procedure, we summarize the steps of the analysis, that comprise the

³ Corresponding author
measurement of the incident neutron spectrum, and the definition of the detectors efficiency function, necessary to perform the Monte Carlo simulations to correct by multiple scattering and sample attenuation effects, which are briefly addressed.

We also pay particular attention to the use of the transmission monitor of VESUVIO to determine total cross sections, a feature which to the author’s knowledge has not been exploited to date. For this purpose, we examine the total cross sections of different H2O/D2O mixtures measured at VESUVIO compared with measurements performed at the Bariloche Electron LINAC, which has been employed in this field for decades [5]. We show that good quality total cross sections can be measured in several decades of energy.

2. Experimental

2.1. Samples

The measurements were performed on three different water mixtures at room temperature. The deuterium molar concentrations of the mixtures were XD = 0 (light water), 0.25, and 0.5. A coin-shaped aluminum can 1.4 mm thick and 5.01 cm diameter was employed as a container for all the samples. The sample covered the entire penumbra of the beam at the sample position, necessary condition to perform both transmission and standard Deep Inelastic Neutron Scattering (DINS) experiments at VESUVIO. The same mixtures (prepared at ISIS) were employed at the Bariloche Electron LINAC to perform transmission measurements.

2.2. VESUVIO setup

We will follow the nomenclature employed in Ref. [4], where a complete description of VESUVIO is found. YAP γ-ray detectors (S135 to S198) placed in forward scattering direction [6] were employed in the analysis of the present work. Detector S2 is a 6Li doped glass detector placed at 13.43 m from the neutron moderator was employed as transmission monitor, while detector S1 located 8.6 m from the moderator was used as the incident beam monitor, whose number of counts was used to normalize all the measured spectra. The sample is located inside an evacuated bell, at 11 m from the moderator. In addition to the measurements on water samples mentioned above, ancillary measurements were performed. A slab of width 2 mm lead was employed as a standard sample for calibration, and also measurements of empty-cell and empty instrument were performed.

3. Basic data analysis formalism

The basic expression for the difference filter-out minus filter-in spectra in the time of flight scale is based on the expression stated by Powles [7] for scattered spectra in the time of flight scale

\[ c(t, \theta) \Delta t = \int_{E_{inf}}^{\infty} dE_0 \Phi(E_0) \frac{d^2 \sigma(E_0, E, \theta)}{d\Omega dE} \varepsilon(E) \left[ 1 - e^{-nT \sigma_F(E)} \right] \left| \frac{\partial E}{\partial t} \right| \Delta \Omega \Delta t, \tag{1} \]

where \( \Phi(E_0) \) is the energy dependent incident neutron flux, \( \frac{d^2 \sigma(E_0, E, \theta)}{d\Omega dE} \) the double-differential cross section of the sample, \( \varepsilon(E) \) the detector efficiency, \( (1 - e^{-nT \sigma_F(E)}) \) the absorption probability of the resonant filter, characterized by a number density \( n \), a thickness \( T \), and a total cross section \( \sigma_F(E) \), and \( \Delta \Omega \) the solid angle subtended by the detectors. The lower limit of integration is determined by kinematic condition that in the second flight path the neutron has infinite velocity, so \( E_{inf} = \frac{1}{2} m L_0^2 \), where \( m \) is the neutron mass and \( L_0 \) the source-sample distance. In Reference [8] we show the link between the calculated spectrum (Eq. (1)), which is a microscopic magnitude, and that measured (comprising multiple scattering events, and container contributions). In the further sections we will show how to access to the required magnitudes employed in Eq. (1) from the experimental information taken at VESUVIO.
4. Results

In this section we present the results of two different types that can be obtained in VESUVIO. On one side we show the analysis of the data obtained from one of the selected YAP detectors in forward scattering direction, showing the multiple scattering and attenuation corrections, with the aim to evaluate its use as mass spectrometer. In addition we analyze the use of the transmission monitor signal to determine total cross sections. All the experimental data treatment we show in this work was performed with programs based on the Open Genie display and analysis of data suite, developed for the neutron scattering instruments at the ISIS facility [9].

4.1. YAP detector analysis

4.1.1. Detector selection As a first step of our analysis, we selected the detectors to work with. The selection of detectors was based on the criterion that the difference filter-out minus filter-in spectra showed a good baseline, free of the filter-dependent gamma background described by Mayers [10]. Thus, in our further work we will not apply the background calculations there described. Fig. 1 shows one of the selected spectra of H$_2$O by comparison with another in which the baseline shows a systematic behavior that affects the region of interest (between 0 and 500 µsec), and was thus discarded. With this criterion we selected for the analysis detectors S162 (located at a distance of 75.1 cm from the sample at a forward scattering angle of 32.76°) and S163 (located at a distance of 75.6 cm from the sample at a forward scattering angle of 42.84°).

4.1.2. Detector efficiency function The knowledge of the efficiency function of each detector is essential to perform the analysis based on Eq. (1). To determine it we applied a procedure similar to that described in [8], where knowledge of the incident spectrum is essential [11]. We firstly measured the incident spectrum with the transmission monitor S2 located at 13.44 m from the source, leaving empty the sample position. The measured spectra, corrected by detector efficiency as described below, is the incident spectrum $\Phi_0(E)$. We also measured the elastically scattered neutron spectrum by a lead sample $\Phi_{Pb}(E)$ at each detector, and also its background $\Phi_{Bg}(E)$ leaving empty the sample position. The efficiency can be then determined as

$$\varepsilon(E) = \frac{\Phi_{Pb}(E) - \Phi_{Bg}(E)}{\Phi_0(E)}.$$  (2)

As discussed in [8], this is a zero-order approach to the efficiency calculation. A more refined determination should include the subtraction of multiple scattering and inelastic effects. In this work we will omit such corrections since we proved that their contribution is secondary. Fig. 2 shows $\Phi_0(E)$, $\Phi_{Pb}(E)$ and $\Phi_{Bg}(E)$ (measured with detector S163, at a scattering angle of 42.8° in the forward direction, located at 75.6 cm of the sample). All the spectra were normalized by monitor counts measured by transmission monitor S1, located at 8.61 m from the source. In the inset we show the efficiency of detector S163 determined from Eq. (2). Leaving aside the thermal region where Bragg peaks appear and thus lead cannot be used as a mirror, the efficiency determined at the epithermal region shows peaks due to the resonances in the gold foils that cover the detectors. In particular, we note the resonance at 4.9 eV.

4.2. Experimental and Monte Carlo results

In Figure 3 we show the experimental spectra in time-of-flight scale for detectors S162 and S163 and the samples of H$_2$O and the water mixtures H$_2$O/D$_2$O for $x_D = 0.25$ and 0.5. Since we use single detectors for our analysis, the time of flight data were rebinned to 5 µsec intervals in order to improve statistics shown in each channel. Also in the same figure we show the Monte Carlo results commented below. For the particular case of $x_D = 0.25$ at a scattering angle of
Figure 1. Comparison between two filter difference spectra (filter-out minus filter-in) in H$_2$O showing the stability of the baseline. Spectrum S163 shows no background and was selected for the analysis.

42.8° we show the contributions of H (light shaded area), D (medium shaded area), and O (dark shaded area) involved in the calculation.

The basic principles on which the Monte Carlo code is based, are exhaustively described in Ref. [12], so only a brief description will be given here. For the purposes of this work, in Monte Carlo calculations the samples were modeled considering a free-atom model as in Ref. [13], with effective temperatures calculated employing Granada’s Synthetic model [14]. Thus for hydrogen we considered \((k_B T)_H = 115.2\) meV, for deuterium \((k_B T)_D = 80.11\) meV and for oxygen \((k_B T)_O = 36.86\) meV. HDO molecules were not considered. They have slightly different vibrational modes that produce minor changes in those effective temperatures. Thus the double differential cross section of a mixture is described by

\[
\sigma(E_0, E, \theta) = \frac{1}{4\pi} \sqrt{\frac{E}{E_0}} \left[ 2(1 - x_D)\sigma_H(E_0, E, \theta) + 2x_D\sigma_D(E_0, E, \theta) + \sigma_O(E_0, E, \theta) \right]
\]  

where \(E_0\) and \(E\) are respectively the initial and final energy of the interaction, and \(\sigma_A(E_0, E, \theta)\) is the free gas scattering function of atom \(A\).

The input data necessary to perform Monte Carlo simulations require prior knowledge of detector and material parameters. In the following we will describe them, and how to obtain the desired information.
Figure 2. Incident neutron spectrum measured with detector S2 shown together with a lead sample spectrum measured with detector S163, located at 75.6 cm of the sample, at an angle of 42.8° in the forward direction. Also, a background spectrum measured without sample. Inset: the efficiency of detector S163 determined from Eq. (2).

(a) Incident spectrum corrected by detector efficiency. We already described the measurement of the incident spectrum. To get $\Phi(E_0)$ employed in Eq. (1) the measured spectrum must be divided by the detector efficiency. The efficiency curve of the $^6$Li-doped glass detector S2, based on calibrations previously done at VESUVIO is $\varepsilon_{S2}(E) = 1 - \exp \left[ -0.051 \sqrt{E} \right]$ where the energy $E$ must be expressed in eV.

(b) Efficiency of the analyzed detector. We determined it as described in §4.1.2. In the inset of Fig. 2 we show the efficiency of detector S163.

(c) Sample and filter geometry and materials. Sample inner and outer diameters and lids thickness are required parameters. Also the tabulated total cross sections as a function of energy of the sample and the container must be given as input.

(d) Instrument parameters. These parameters include the incident beam energy and its transverse section dimensions. The gold filter analyzer thicknesses are also required.
Figure 3. Experimental results for H$_2$O and the water mixtures H$_2$O/D$_2$O for $x_D = 0.25$ and 0.5 (blue circles) at two different angles in forward scattering, compared with the Monte Carlo simulation results. Red line: single scattering, green line: multiple scattering, black line: total scattering. Also shown contributions of H (light shaded area), D (medium shaded area), and O (dark shaded area), for $x_D = 0.25$ at 42.8°.
(e) Filter analyzer absorption cross section as a function of energy. Absorption cross section tables can be obtained from Ref. [15]. As analyzed in Ref. [16] a good description of the time-of-flight profiles requires a complete table of the filter analyzer absorption.

(f) Total cross section of the sample materials as a function of energy. These data must be tabulated in an energy range wide enough to cover not only the energies corresponding to the incident spectrum (epithermal energies), but also to consider the energy transfers after a number of multiple scattering steps (typically thermal energies). As shown in the next section these data can be provided by the information generated by the transmission monitor.

Fig. 3 shows the Monte Carlo results compared with the experimental data for the two selected angles. With red line we show singly scattered neutron contribution, with green line the multiple scattering and with black line the total scattering.

The Monte Carlo results are used to define the attenuation and multiple scattering factors that must be used to correct the experimental data. The attenuation factor \( H(t) \) is defined as ratio of detected singly-scattered neutrons to the total singly-scattered neutrons, and the multiple scattering factor \( M(t) \) is defined as the ratio of detected singly-scattered neutrons to the total scattered neutrons[12]. The experimental data are then corrected by

\[
E_{\text{corr}}(t) = \frac{M(t)E(t)}{H(t)}
\]

Fig. 4 shows the uncorrected and corrected data that result after applying Eq. (4). In the inset we show the attenuation factors, and the multiple scattering factors. The variation in peak intensities after corrections will influence the final quantitative results. A detailed study of the observed intensities will be presented in a future publication.

4.3. Transmission monitor
We explored the possibility to measure total cross sections with VESUVIO transmission monitor S2. To this end we measured the transmitted beam of each sample \( S(E) \), and the empty cell \( F(E) \). As in the former measurements the spectra were normalized by monitor counts of detector S1. To assess the magnitude of the background counts (i.e. the number of neutrons counted by S2 that do not come from the incident beam), we measured the transmitted beam of a Cadmium sheet 1mm thick. In the thermal region, the magnitude of this spectrum was between 2 and 3 orders of magnitude lower than the open beam, so we neglected its contribution.

The transmission is determined from

\[
T(E) = \frac{S(E)}{F(E)}
\]

and the total cross section \( \sigma_{\text{tot}}(E) \) results from [17]

\[
T(E) = \exp(-nT\sigma_{\text{tot}}(E))
\]

where is \( n \) the number density of the sample and \( T \) its thickness. The same samples prepared at ISIS were taken to the Bariloche LINAC laboratory, where control measurements were carried out, following the same procedure described in Ref. [17].

The results are shown in Fig. 5, where the total cross sections of the three mixtures measured in both laboratories are compared. For illustrative purposes, in the inset we show the ‘open beam’ (empty cell) and the ‘sample transmitted’ spectra measured at VESUVIO, for the H\(_2\)O sample. The agreement is remarkable, and the comparison serves to validate the measurements at VESUVIO since the Bariloche facility is customarily employed in this kind of experiments and its setup optimized for this purpose.
Figure 4. Uncorrected (dashed line) and corrected data (solid line) for H$_2$O (black lines) and the water mixtures H$_2$O/D$_2$O for $x_D = 0.25$ (red lines) and 0.5 (blue lines). Inset, the attenuation factors (dashed lines) and the multiple scattering factors (solid lines), with the same color convention.

5. Conclusions
In this paper we have shown two different aspects in the use and the analysis of the experimental information provided by VESUVIO. First, the use of information provided by single detectors in the time-of-flight scale is useful for quantitative analysis of atoms present in the sample. The steps that comprise analysis shown include multiple scattering attenuation and detector efficiency corrections, lead to a quantitative analysis in the way shown in Ref. [18]. In the second example, we showed that the transmission monitor S2 can be used to determine total cross sections of good quality, an application that until today had not been reported.

Future work under current development, shows the possibility to determine in a single experiment the composition of a sample and the total cross section of an element. The possibility of developing this type of experiments, will be of interest for those fields requiring the use of updated total cross sections, particularly Nuclear Engineering, that often use outdated information due to the lack of experimental facilities that provide it. In this sense, we envisage the possibility of future demand of information at different conditions of pressure and temperature.
Figure 5. Total cross sections of the three water mixtures measured at VESUVIO (open symbols) compared with measurements made on the same mixtures made at the Bariloche LINAC. Inset: Open beam and transmitted sample spectra at detector S2 of VESUVIO.

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