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Scattering law of a magnesium hydride moderator

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ABSTRACT

Metal hydrides have long been considered possible moderator and pre-moderator materials for neutron sources. These materials have hydrogen density comparable to liquid hydrogen or light water. They usually do not undergo phase transitions in the desired operating range of 0–300 K, and display reasonable resistance to radiation damage. Magnesium hydride is such a simple, robust hydride system. To assess its neutronic usefulness as a moderator material, we determined experimentally the total scattering cross-section of the material. We compared our theoretical results to the experimental total neutron cross-section and to the data from quasi-elastic neutron scattering experiments, and produced a scattering kernel suitable for neutron transport calculations.

1. Introduction

Over the past 40 years thermal and cold neutron production by spallation sources was limited to a few materials: light water, liquid/solid methane, liquid/solid deuterium, and liquid hydrogen. While these materials have served their purpose well, they have reached their physical and neutronic limitations in current neutron source designs. Clever neutronic designs have exploited these materials to the limit, and there is little room left for further optimization. The increasing demand for brighter, next-generation neutron sources requires new materials.

Temperature is perhaps the most significant restriction in the use of traditional moderator materials. It limits severely the shape of the neutron energy spectrum. Many neutron scattering instruments that currently use thermal neutrons would benefit from a neutron energy spectrum closer to 150 K. This cannot be achieved with light water because ice is textured and suffers from radiation damage in intense radiation fields. This problem triggered the interest of the spallation physics community in metal hydrides because most of them are structurally stable in the desired temperature range. The original focus was on zirconium hydride, which has been used in reactor applications, and it is also one of the most stable metal hydrides. Several studies [1,2] have been performed on zirconium hydride, but the experimental results were discouraging. Indeed, the slowing down power and the moderation ratio for zirconium hydride with a realistic packing density are below those of light water.

By contrast, the values for magnesium hydride (MgH_2), even at a packing density of 50%, are comparable with those of light water. Santisteban et al. [3] considered MgH_2 as a potential moderator material. Unfortunately, in this work the total neutron cross-section was not measured below 3 meV, and some of the model parameters have a fairly wide range of validity in their predicting power. It is the purpose of this study to extend these data to the cold neutron regime and to narrow the flexibility of the model parameters. To accomplish this it will be necessary to develop a new model scattering kernel by including quasi-elastic neutron scattering data which will provide additional information on the inelastic scattering up to a few meV. It is expected that such a scattering kernel will allow to evaluate the standard LEAPR model [7] for energies below 3 meV.

In Section 2 we discuss sample preparation and the experimental setup used to measure the total neutron cross-section of MgH_2 down to 0.5 meV. In Section 3 we give a brief discussion of the theory of the neutron scattering cross-section, which is the basis of our numerical evaluation. The latter is presented in Section 4. Finally, in Section 5 conclusions are drawn.

2. Experiment

2.1. Sample preparation

Magnesium hydride was prepared according to Bodganovic et al. [8]. The following steps were carried out in an inert argon atmosphere: Ethyl bromide (75 μl) was dripped into a suspension of magnesium powder (750 mmol, 18.2 g) in anhydrous

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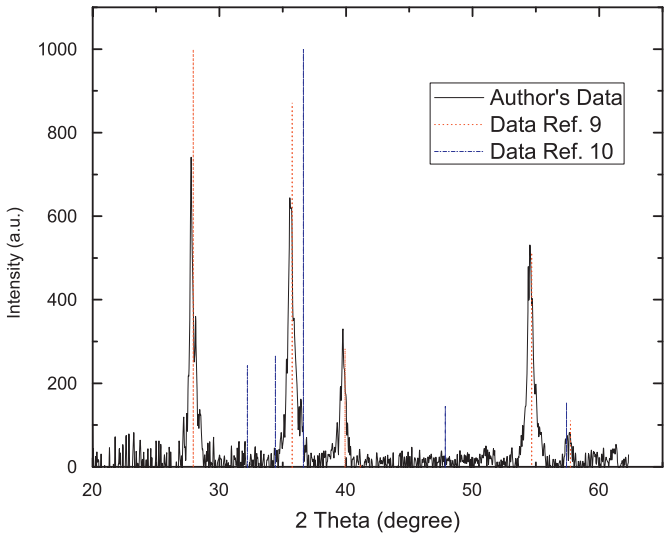

Fig. 1. Comparison of the X-ray powder diffraction patterns of magnesium hydride sample used for this study (black solid line) with historical data for magnesium hydride [9] (red dotted line) and for the magnesium [10] (blue dashed-dotted line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

tetrahydrofuran (THF) (90 ml). The mixture was stirred for 30 min before adding anthracene (7.5 mmol, 1.34 g). The suspension was then heated to 60 °C and stirred for 2 h to form the organo-magnesium complex. After cooling to room temperature, the catalyst chromium chloride (7.5 mmol, 1.19 g) was added. The suspension turns olive-green. It is stirred over night (8 h) and is then transferred to a 150 ml teflon lined autoclave fitted with a stirrer and temperature control. The autoclave was purged with Ar and pressurized with H_2 at 80 bar. It was kept at 60 °C for two days. After cooling to room temperature the light-grey suspension was filtered. The resulting MgH_2 powder was washed twice with anhydrous THF and twice with dry pentane. It was finally dried under vacuum at room temperature. We obtained 17.9 g (0.71 mol, 95% yield) of a light-grey powder. The structure and purity were confirmed by X-ray powder diffraction. This is possible because of the ionic nature of MgH_2 which does not allow for non-stoichiometric compounds (MgH_{2-x}). A MgH_{2-x} sample can only be produced by mixing MgH_2 powder with Mg powder at a ratio of $1 : x/(2-x)$. If the sample still contained Mg powder, Mg peaks would be detectable by X-ray powder diffraction. As it can be seen in Fig. 1 the magnesium peaks are no longer visible, which confirms the purity of the sample.

2.2. Cross-section measurement

To validate a calculated scattering kernel experimental data are needed. The first check is to compare the (calculated) total neutron cross-section with experiment. Note that this comparison alone is insufficient to prove that the theoretical model reproduces the physics of the problem.

The total MgH_2 neutron cross-section was measured on flight path 5 at the Manuel Lujan, Jr. Neutron Scattering Center at Los Alamos National Laboratory. Fig. 2 shows the experimental setup. An aluminum sample holder filled with MgH_2 was placed in the beam between the shutter and the collimator as shown in Fig. 2. The thickness of the MgH_2 sample in the direction of the neutron beam was 20 mm. As a reference measurement the sample was replaced by an empty sample holder with the same geometry. In both cases the transmission spectrum was measured with a fission

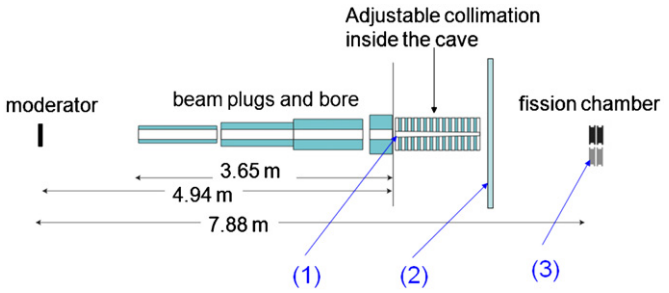

Fig. 2. Layout of Flight Path 5 (aperture: 20 mm) at the Manuel Lujan Jr. Neutron Scattering Center at Los Alamos National Laboratory. (1) Indicates the position of the MgH_2 sample which is placed in front of the collimation. (2) Points to the polyethylene wall which shields the hutch from neutrons scattering off the target and the collimation. Finally, (3) indicates the detector position.

chamber downstream of the collimator. This detector measures the time-dependent neutron flux after passing through the transmission sample by means of the $^{235}U(n, f)$ reaction. The parallel-plate ionization chamber with a 200 $\mu g/cm^2$ deposit of ^{235}U (99.9% isotopic purity) was placed 2.96 m downstream of the transmission sample. The data acquisition system recorded the detector pulse-height response and the neutron time-of-flight for each event. Fission events can thus be validated and the incident neutron energy is determined. The number of fission events as a function of neutron energy was then used to calculate the differential neutron flux. The resulting spectra were normalized by the integrated proton beam current on target, and the macroscopic total cross-section was calculated using the attenuation equation

$$N(x) = N(0)e^{-\Sigma_t x} \quad (1)$$

where Σ_t is the macroscopic total cross-section and x the thickness of the sample. $N(x)$ and $N(0)$ are the number of neutrons detected by the fission detector with an x-cm thick sample in the beam and no sample in beam, respectively. The total microscopic cross-section is defined as

$$\sigma_t = \Sigma_t/\rho \quad (2)$$

where ρ is the density of the material. This relation is valid only if multiple scattering is negligible. To ensure this, the sample was moved upstream [see position (2) in Fig. 2] of a 0.7 m long collimation system. The aperture was limited to 20 mm in diameter. The total microscopic neutron cross-section from these measurements is shown as the solid line in Fig. 3. The light (black) dotted and dashed lines indicate the 2-σ uncertainty on the data. In determining this uncertainty we estimated the error on the sample thickness to be 0.5% (0.1 mm of 20 mm thickness), the error on the sample density to be $\sim 2\%$, and, finally, we added the standard statistical counting error. The incoherent cross-section of MgH_2 was measured earlier by Santisteban et al. [3]. We used these data to estimate the total neutron cross-section under the assumption that the total neutron cross-section can be approximated by the bound cross-section of magnesium. The result is presented by the solid (red) squares in Fig. 3 and experiment agrees within 5% with this estimate. Furthermore it is expected that the total cross-section of the material converges to the total cross-section of the free gas model at high enough energies. Commonly 1 eV is assumed to be a high enough energy. However, Wick [11] has shown that the rate of convergence to the free gas model is a function of the reduced mass, with the convergence rate being the lower the reduced mass is. This makes H^1 the lowest converging isotope. In addition it can be seen from Fig. 4, which will be presented later on in Section 4, that the phonon energy cut-off for MgH_2 is at about 180 meV. This means that a 5-phonon term as defined in Eq. (8) has a maximum transfer energy of almost 1 eV. Since a 5-phonon term