New Results on CaH$_2$ Thermal Neutron Scattering Cross Sections

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Abstract. Calcium hydride (CaH$_2$) is a compound of interest in the frame of a current research program on the transmutation of long-lived nuclear wastes. Since CaH$_2$ is relatively stable in liquid sodium, it is one possible material that can be used for local moderation of the neutron spectrum in fast neutron reactors such as PHENIX. In order to describe the moderated region from Monte Carlo and/or deterministic calculations, thermal neutron scattering data are needed. In particular, an adequate treatment of the thermal inelastic scattering cross sections for bound hydrogen is requested. The present work aims at the determination of these data. The first step was the measurement of the phonon frequency spectrum, which was carried out on the three axis spectrometer of the Institut Laue Langevin in Grenoble (France). This phonon frequency spectrum has already been published and so only a brief description of this measurement will be given here. Then, from physical grounds, the acoustic mode has been weighted relative to the optical modes in order to treat Hydrogen atoms bound in CaH$_2$. The S(α,β) scattering laws have been generated for various temperatures using the NJOY code working in the incoherent approximation and the Gaussian approximation. The deduced incoherent elastic and incoherent inelastic cross sections are shown and discussed. These new thermal neutron scattering data will be proposed in the JEFF3.1 European library.

INTRODUCTION

In the frame of possible methods for transmutation of actinides and/or long lived fission products, several experimental programs have been proposed to investigate the neutronic characteristics of fast neutron reactor cores containing small quantities of various moderator materials. For instance, the ‘COSMO’ programs were carried out in the MASURCA research reactor (Cadarache, France) where B$_4$C, ZrH$_2$ and CaH$_2$ moderator materials were used [1, 2]. Also, the ECRIX-C (with B$_4$C) and ECRIX-H (with ZrH$_2$ and CaH$_2$) experiments are planned in the PHENIX fast neutron reactor (France).

In order to simulate these experimental programs from deterministic and/or stochastic calculations, thermal scattering cross sections related to the moderator materials are needed. Thermal data can be found for B$_4$C and ZrH$_2$ in the evaluation files, but no data is available for calcium hydride (CaH$_2$). In particular, thermal incoherent cross sections for H bound in Calcium must be determined. This is the objective of the present work.

The first step was to measure the CaH$_2$ phonon frequency spectrum. This measurement has already been presented in a previous paper [3] and therefore only a brief description is given here. From physical grounds, the phonon frequency spectrum for H bound in CaH$_2$ was deduced and the S(α,β) scattering laws were generated using the NJOY code. Lastly, the calculated thermal cross sections are shown and discussed.

CaH$_2$ PHONON FREQUENCY SPECTRUM MEASUREMENT

The measurement of the phonon frequency spectrum was performed by Morris et al. [3] at the high flux reactor of the Institut Laue Langevin (ILL) in Grenoble (France) using the three axis spectrometer IN1 Be-filter instrument. A powder CaH$_2$ sample (~95 % pure) unexposed to air was used. The principle of the measurement is to determine the number of scattered neutrons with a fixed energy (4 meV) for a given incident neutron energy. The incident neutron energy was increased from 15 meV up to 180 meV in steps of 1 meV. A monitor was placed in front of the neutron source in order to normalise the number of incident neutrons.

After having removed the background as well as the small Ca(OH)$_2$ contribution (impurities in the sample), the final CaH$_2$ phonon frequency spectrum at T=295 K could be obtained (see left part of Fig. 1). This spectrum has the following characteristics:

1. An acoustic mode (around 20 meV), which corresponds to the motions in phase of the H and Ca atoms,
2. A first optical mode (between 70 and 100 meV), which has been seen in the previous measurements performed by Maeland [4] and Bergsma [5],
3. A second optical mode (between 110 and
140 meV), which has been observed by Maeland [4], but with a poor energy resolution. The position of this optical mode is in agreement with the prediction made by Ross [6].

Fine structures are also visible in each optical mode. All these characteristics are consistent with the known crystal structure of CaH$_2$ (see [7, 8]).

**THERMAL NEUTRON SCATTERING CROSS SECTIONS FOR H IN CaH$_2$**

**H in CaH$_2$ Phonon Spectrum**

The thermal scattering cross sections for H bound in CaH$_2$ can be calculated knowing the partial phonon frequency distribution corresponding to the H atom vibrations. In principle, a rigorous way to get this partial phonon frequency distribution is to use a lattice dynamical model. Such a model has been applied by Slaggie [9] for describing H in ZrH$_2$. More recently, a similar formalism was performed by Al-Qasir et al. [10] for studying H in ThH$_2$.

In the present work, we did not follow this rigorous approach, but we used instead the following approximation: in his work on ZrH$_2$, Slaggie [9] has shown that the ratio between the acoustic and the optical modes for H in ZrH$_2$ was 1/242. In our case (H in CaH$_2$), we have assumed that this ratio must be increased by the $A_{Zr}/A_{Ca}$ factor, where $A_{Zr}$ and $A_{Ca}$ are the Zirconium and Calcium masses, respectively. In this way, the weight of the acoustic mode for H in CaH$_2$ becomes 1/106. The corresponding phonon spectrum is shown in Fig. 1 (right part) as well as the one for H in ZrH$_2$ calculated by Slaggie. A similar approximation has been applied in the past by Picton [11] for describing H in TiH$_2$.

**NJOY Calculations**

The double differential scattering cross section takes the form:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\sigma_b}{4\pi kT} \sqrt{\frac{E'}{E}} S(\alpha, \beta)$$

where $\sigma_b$ is the bound scattering cross section for H-atom, $E$ and $E'$ the initial and final neutron energy, $T$ the temperature of the scattering medium and $S(\alpha, \beta)$ the so-called scattering laws which depend only on the dimensionless momentum transfer and energy transfer parameters, respectively: $\alpha = (E + E' - 2\mu E')/AkT$ and $\beta = (E - E')/kT$. Using the incoherent approximation and the Gaussian approximation, it can be shown (see [12] and references therein) that $S(\alpha, \beta)$ can be written as:

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\beta t} e^{-\gamma(t)} dt$$
where
\[ \gamma(t) = \alpha \int_{-\infty}^{\infty} P(\beta) [1 - e^{\beta t}] e^{-\beta/2} d\beta \] (3)
with
\[ P(\beta) = \frac{\rho(\beta)}{2\beta \sinh(\beta/2)} \] (4)

\( \rho(\beta) \) being the partial phonon frequency distribution normalised to one (right part of Fig. 1).

The LEAPR module implemented by McFarlane [12] in the NJOY code was used in order to calculate the \( S(\alpha, \beta) \) scattering laws. Then, from Eq. (1), the thermal incoherent elastic and inelastic cross sections for H in CaH\(_2\) could be deduced. This was done with the THERMR module of NJOY. The \( \alpha \) and \( \beta \) grids used for these calculations were similar as the one used by McFarlane for the H in ZrH\(_2\) case. In particular, it allows for energy transfer of around 2 eV at T=296 K. Calculations were performed for several temperatures and the results are shown in Fig. 2.

**Table 1.** Debye-Waller integrals and effective temperatures derived from the partial phonon frequency distribution for H in CaH\(_2\)

<table>
<thead>
<tr>
<th>Temp. (K)</th>
<th>Debye-Waller Integral (eV(^{-1}))</th>
<th>Effective Temp. (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>296</td>
<td>12.64</td>
<td>613.20</td>
</tr>
<tr>
<td>300</td>
<td>13.13</td>
<td>634.33</td>
</tr>
<tr>
<td>350</td>
<td>16.54</td>
<td>717.05</td>
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<tr>
<td>400</td>
<td>17.92</td>
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</tr>
<tr>
<td>450</td>
<td>19.40</td>
<td>862.60</td>
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<td>500</td>
<td>20.88</td>
<td>944.06</td>
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<td>600</td>
<td>24.12</td>
<td>1117.00</td>
</tr>
<tr>
<td>700</td>
<td>27.36</td>
<td>1298.33</td>
</tr>
<tr>
<td>800</td>
<td>30.60</td>
<td>1480.60</td>
</tr>
</tbody>
</table>

**Results and Discussion**

The thermal incoherent elastic part (top of Fig. 2) can be determined easily from the following equations:

\[ \sigma(E) = \frac{\sigma_0}{2} \left( 1 - e^{-4WE} \right) \] (5)

where \( W \) is the Debye-Waller integral given by \( W = \lambda/\Delta T \), with \( \lambda \) being the Debye-Waller coefficient:

\[ \lambda = \int_{0}^{\infty} \frac{\rho(\beta)}{\beta} \coth(\beta/2) d\beta \] (6)

The effective scattering temperature is obtained from:

\[ T_{\text{eff}} = \frac{1}{2} T \int_{0}^{\infty} \frac{\rho(\beta) \beta \coth(\beta/2)}{T} d\beta \] (7)

Values calculated from Eqs. (6) and (7) are given in Table 1. It is interesting to note that from the partial phonon frequency distribution, the root mean square displacement of the H-atom from its position in the metal lattice can also be calculated. We found: \( \sqrt{<u^2>} = 0.16 \) Å, in close agreement with the value obtained by Bergsma (0.17 Å) [5].

The thermal incoherent inelastic part (bottom of Fig. 2) corresponds to the case where creation or annihilation of one or more phonons occurs. It is illustrated in Fig. 3, where the scattered neutron energy distributions are plotted for four incident neutron energies (the arrows on the plot show the position of the incident neutron energy). For example, on both sides of the incident neutron energy, the gain and loss of the scattered neutron by one ‘acoustic’ phonon (around 20 meV) are clearly visible.

Lastly, thermal data for H in CaH\(_2\) were generated in ENDF format for inclusion in the JEFF3.1 evaluation file. The corresponding MAT number was taken as 8. The MT numbers for the incoherent elastic and inelastic cross sections were chosen as 237 and 238, respectively.
CONCLUSION

From a recent CaH$_2$ phonon frequency distribution measurement [3], the partial phonon frequency distribution for H in CaH$_2$ was deduced. Using the LEAPR and THERMR modules from NJOY, the thermal scattering laws and the incoherent elastic and inelastic cross sections were calculated. These new data are proposed as the JEFF3.1 evaluation file. Integral tests of these thermal data are planned from the COSMO3 experiment (performed at the MASURCA facility (CEA-Cadarache)) as well as from future experiments in the PHENIX fast neutron reactor.

It should be stressed that due to the high affinity for hydrogen binding with oxygen from the air-water vapour, the calcium hydride can easily be transformed into a hydroxide calcium material (Ca(OH)$_2$). Therefore, thermal data on this material could be useful.

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REFERENCES

10. Wehring and T. Zhou.