Attenuation Measurements of Transition Metal Hydrides formed by Self-Propagating High-Temperature Synthesis

I. P. Swainson[1]

[1] Canadian Neutron Beam Centre, National Research Council, Chalk River Laboratories, Ontario, Canada K0J 1J0

These measurements of the potential for metal hydrides to be neutron shielding materials were performed as part of ISTC Grant A-1249 with the Armenian National Academy of Sciences: Institute of Chemical Physics, who synthesized all samples.

Scattering consists of two-independent cross-sections of coherent (σ_c) and incoherent (σ_i) components, which for shielding purposes are summed to the scattering cross-section σ_s.

$$\sigma_s = \sigma_c + \sigma_i$$

In general, for energy less than 1 MeV, and therefore over all over thermal energies, the approximation is made that the scattering cross-section is independent of the incident neutron energy. Scattering depletes a directional beam; multiple scattering ultimately scattering the beam into 4π.

Absorption is the process by which a neutron is ultimately captured. Unlike scattering cross-sections, absorption cross-sections are generally strongly energy dependent. Except for the cases of isotopes that have a resonance near the thermal energy regime, absorption cross-sections, $\sigma_a(v)$, in the thermal regime follow the $1/v$ law, where v is the velocity of the neutron. Absorption cross-sections in the fast neutron regime are usually very small.

Beer-Lambert Attenuation

For optical radiation, the Beer-Lambert law is usually applied to the absorption of a monochromatic beam, assuming no scattering. The approximation is usually made with neutron radiation that the attenuation of a directional beam is the result of the depletion of that beam by absorption and scattering, and that the scattered neutrons are deflected from the main beam, and not ultimately rescattered into the detector. The total microscopic cross-section, σ_{tot}, can be defined as

$$\sigma_{tot} = \sigma_s + \sigma_a(v).$$

In the absence of any other phenomena, the Beer-Lambert law can be applied to the attenuation of thermal neutrons by a scatterer, where the measured intensity, I, is related to the incident intensity, I_0, by the formula

$$I - I_0 \exp\{-\Sigma_{tot} t\},$$

and

$$\Sigma_{tot} = N\sigma_{tot},$$

where t is the thickness, N is the number density of scatterers per unit volume, and Σ_{tot} is known as the macroscopic cross-section, having dimensions of L^{-1}, i.e., is the linear attenuation factor, and units of cm^{-1}, where σ is in barns and N in $Å^{-3}$.

The macroscopic cross-section for a compound is given by summing the component cross-sections for each element (isotope). For a crystalline compound, $N = Z/V$, where V is the unit cell volume, and the number of formula units per unit cell is Z. For example,

$$\Sigma_{TiH_2} = \frac{Z}{V}(\sigma_{Ti} + 2\sigma_H),$$

where the σ in the above equations are understood to be total microscopic cross-sections. The macroscopic cross-sections would be scaled linearly for solids that are not 100% dense. The mean free path length in the material $\lambda = 1/\Sigma_{tot}$. As a general rule, if the thickness of a given material is many times the mean free path, it should be an efficient attenuator.

Assumptions of the Beer-Lambert absorption include that the beam is perfectly parallel, with each ray traversing the same thickness, that the radiation is monochromatic, and that the radiation does not strongly interact with the sample; the latter assumptions are strongly violated for hydrogenous compounds.

Moderation

Materials composed with a significant proportion of light elements can moderate the neutron spectrum. Moderation occurs via elastic collision, where most of the energy of the neutron is transferred to the light nucleus. Moderating efficiencies can be derived by calculating two quantities α and ξ, where

$$\alpha = \frac{(A-1)^2}{(A+1)^2},$$

where A is the atomic mass. The number of collisions to slow a neutron from an initial energy, E_0, to a given final energy, E', is $\ln(E_0/E')/\xi$, where ξ is the average logarithmic energy decrement per collision, defined as:

$$\xi = 1 + \frac{\alpha \ln \alpha}{1-\alpha}.$$

The maximum energy loss is where the neutron is scattered backwards and $E' = \alpha E$, and the average loss is $(1-\alpha)E/2$. Therefore, for ^1H, where $A = 1$ and $\alpha = 0$ it is possible that all of the energy of a neutron is lost in a single collision. Heavier nuclei result in lower energy losses per collision.

$\xi\Sigma_s$ is known as the moderating power and $\xi\Sigma_s/\Sigma_a$ is termed the moderating ratio (MR).

For mixtures or compounds the moderating power can be calculated by linear addition, e.g.;

$$\xi\Sigma_{TiH2} = \xi_{Ti}\Sigma_{Ti} + 2\xi_{H}\Sigma_{H}.$$

General considerations

For thermal neutron shielding, materials that combine a large absorption cross-section with a large scattering cross-section are ideal. Without absorption a potential shielding material may be a very strongly scatterer and, therefore, attenuator, but would still emanate neutrons, although a directional beam would issue as a more diffuse neutron field from the shielding material.

Efficient shielding of high-energy neutrons is generally best achieved by moderating the energy, and absorbing the thermal neutrons with isotopes that have a high absorption cross-section at that energy; e.g., polyethylene and cadmium are typically used in conjunction around neutron detectors, as the H in polyethylene has a strong scattering cross-section and low mass and this makes it a good moderator. However, the thermal capture cross-section is not high. Therefore a thin sheet of cadmium is applied after the polyethylene.

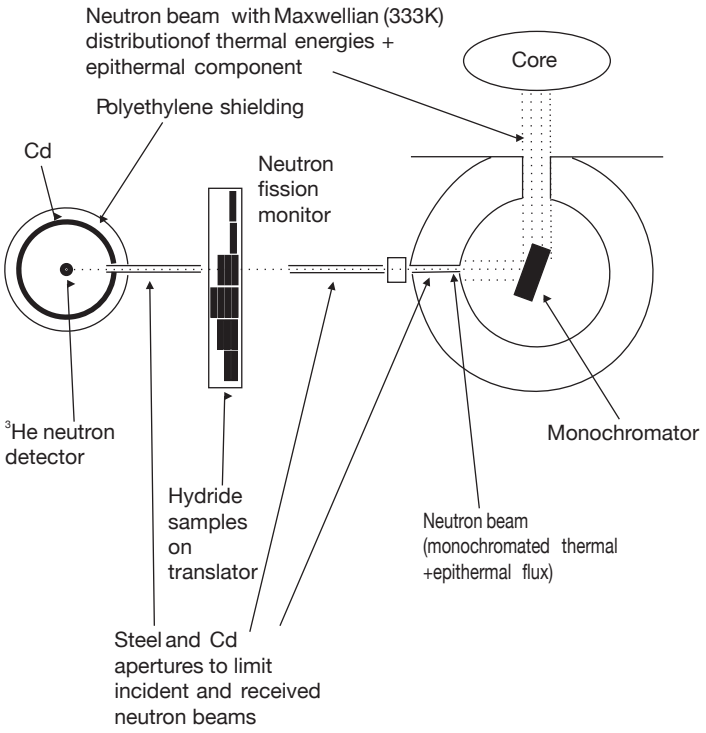

Fig 1. *Schematic diagram of the experimental set up of the attenuation measurements of the metal hydrides, produced by self-propagating high-temperature synthesis, on the E3 spectrometer, at NRU.*

The neutron attenuation measurements were performed on E3 (Fig. 1). Neutrons of wavelength 1.3997 Å were selected using a Ge monochromator. The take-off angle, $2\theta_m$, was fixed at 80°. The typical spread of wavelengths is $\Delta\lambda/\lambda \sim 1\%$ or better.

On the detector side, the beam channel was again shut down with a long steel plug with fine aperture to cut down the fast neutron background reaching the sample area. A long "snout" was extended towards the samples with a Cd slit 3.81 mm tall by 1mm wide defining the illuminated area of the hydride pellet. The Cd slit on the receiving snout was 3.82 mm tall by 1 mm wide. 20 cm long bars of steel were used to shut down the aperture to fast neutrons on the detector side. The slits and detector were adjusted were adjusted to ensure that the incident beam, slits and detector were collinear. A ^{3}He neutron detector was used to record the transmitted neutrons.

Determination of the epithermal Cd flux.

The epithermal neutron flux is usually derived by placing Cd in the beam. Cd is an efficient high-pass filter for thermal neutrons. The absorption cross-section increases by three orders of magnitude for Cd varies dramatically over the range 1-0.1 eV. The cut-off energy, below which Cd is considered to be nearly 100% effective, is usually taken as E~0.5 eV (λ~0.40Å). The remaining neutrons that pass through the filter are epithermal ('fast') neutrons (E > 0.5 eV). The ^{3}He detector is a high-efficiency detector for neutrons in the thermal regime. However, its response to neutrons is highly wavelength dependent for λ < 2.7Å. For epicadmium neutrons its efficiency is less than 20%.

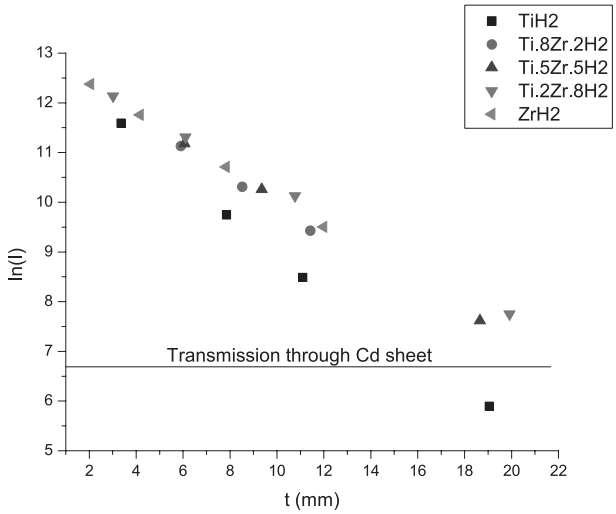

Fig 2. *Plot of $\log_e$(uncorrected counts) versus thickness of metal hydrides in the Ti-Hf series. The transmission through Cd is given as a straight line.*

Typically 1 mm of Cd in the beam is sufficient to remove all thermal neutrons. The number of neutrons recorded on the ^{3}He detector on passing through a sheet of Cd 2.64 mm thick, larger than the sample size of the hydrides, was 805 ± 24. For high thicknesses of metal hydrides, the number of neutrons transmitted is substantially lower than transmitted through the Cd sheet; e.g., 20.3 mm of $Ti_{0.8}Hf_{0.2}H_2$ transmits only 279 ± 22 and 19.05 mm of TiH_2 transmits only 264 ± 15 neutrons. Therefore, all the thermal neutrons and some of the epicadmium neutrons have been attenuated by these hydrides, which could only happen if there was significant moderation. The H atoms in the hydrides act as moderators of neutrons, so that

neutrons are more effectively attenuated by the sample than they otherwise would have been. Remoderation affects both the primary, monochromatic thermal beam, and the epicadmium component. Typical attenuation measurements are shown in Fig. 2.

Sources of systematic error

1. Although the incident and transmitted beams were shut down to be very small, the steel aperture was slightly larger than the final Cd mask, so that the area of hydride illuminated with fast neutrons (an aperture defined by long steel bars and plugs) may be slightly larger than the area illuminated by both thermal and fast neutrons (defined with the final Cd mask).

2. The Cd masks were placed close to the sample, to reduce the effects of multiple scattering of neutrons into the detector. However, the hydrides interact very strongly with neutrons and it is possible that the detector records some fraction of multiply scattered neutrons.

3. Moderation: of those neutrons that do undergo interactions with the hydride prior to entering the detector (rather than passing through with no interactions) it is likely that the energy with which they strike the detector is not the energy with which they entered the hydride. This can yield two opposing biases.

a. A systematic tendency to overcount the transmitted neutrons for those epicadmium neutrons that are moderated. A thermalized fast neutron is many times more likely to be recorded by the ^{3}He detector than a fast neutron.

b. A systematic undercounting of neutrons for those epicadmium neutrons that are moderated and then attenuated by the hydride more efficiently than they would otherwise have been.

The values of the effective linear attenuation factors (Fig. 3) are lower than the calculated values (Table 1), and the density of the pellets is not likely to account for the difference. Instead,

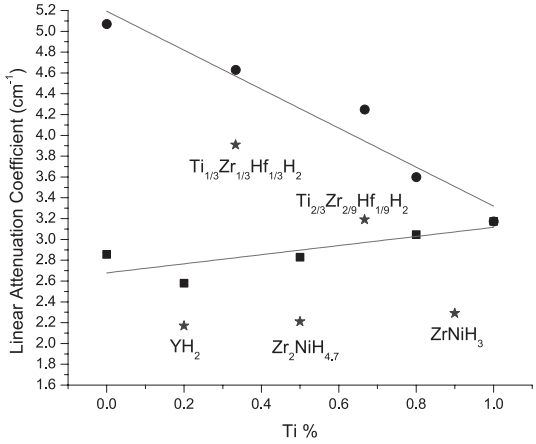

Fig 3. Effective linear attenuation coefficient, Σ_{tot}^{eff}, in cm^{-1}. The circles represent the Ti-Hf composition line, and the squares the Ti-Zr composition line. Lines are a linear least squares fit to the two composition lines.

it is likely that the systematic errors in the experimental set up (slight difference in the size of the masks for fast and thermal neutrons) affect the magnitude and that a Beer-Lambert law is a poor description of the interactions of neutrons, due to the potential for multiple scattering, and especially the strong moderating efficiency of the hydrides: The H in hydrides makes them efficient moderators and turns an initially monochromatic beam into a remoderated spectrum: e.g., t_{mod} in table 1 gives the number of scattering interactions with each hydride required to reduce a 2 MeV fission neutron down to 0.1 eV. For most of the metal hydrides this distance is ca. 20-40 mm. In practice t_{mod} is not a physical thickness, since the interactions result in a random walk of the neutron, so the thickness of the pellet required to achieve this should be much lower.

Conclusions

Beer-Lambert style of analysis can only ever yield approximations to the processes going on inside moderator-attenuators. Analysis based around codes such as MCNP would be required to fully model experimental results. Hydrides based around Hf, Gd or Cd have the potential to be very efficient neutron moderator-attenuators.

Compound	Σ_s/N	Σ_a/N	ξ	MP	MR	Σ_{tot}/N	N	Σ_{tot}	λ	t_{mod}
TiH_2	168.4	5.25	0.975	7.50	31.26	173.6	4.57E-02	7.94	1.26	22.39
$Ti_{0.8}Zr_{0.2}H_2$	168.8	4.33	0.973	7.16	37.88	173.1	4.36E-02	7.55	1.32	25.38
$Ti_{0.67}Zr_{0.33}H_2$	169.1	3.74	0.971	6.79	43.93	172.8	4.14E-02	7.15	1.40	27.31
$Ti_{0.5}Zr_{0.5}H_2$	169.4	2.96	0.969	6.79	55.52	172.4	4.14E-02	7.13	1.40	36.07
$Ti_{0.2}Zr_{0.8}H_2$	170.1	1.58	0.965	6.23	103.94	171.6	3.79E-02	6.51	1.54	27.00
$Ti_{0.8}Zr_{0.2}H_2$	168.8	4.33	0.973	7.22	37.88	173.1	4.40E-02	7.62	1.31	20.67
$Ti_{0.66}Zr_{0.34}H_2$	169.1	3.69	0.971	6.99	44.47	172.8	4.26E-02	7.36	1.36	22.88
$Ti_{0.3}Zr_{0.7}H_2$	169.9	2.04	0.967	3.21	80.53	171.9	1.96E-02	3.36	2.97	46.41
$Ti_{0.66}Zr_{0.22}Hf_{0.11}H_2$	169.6	13.39	0.968	6.99	12.26	182.9	4.26E-02	7.79	1.28	22.56
$Ti_{0.33}Zr_{0.33}Hf_{0.33}H_2$	171.0	28.89	0.960	6.45	5.68	199.9	3.93E-02	7.85	1.27	24.46
ZrH_2	170.5	0.66	0.963	5.92	248.30	171.1	3.60E-02	6.17	1.62	28.45
$ZrNiH_3$	271.0	4.41	0.911	6.21	55.95	275.4	2.51E-02	6.93	1.44	27.11
Zr_2NiH_5	441.5	5.07	0.931	6.15	81.02	446.6	1.50E-02	6.68	1.50	27.25
HfH_2	174.2	81.47	0.942	6.22	2.01	255.7	3.79E-02	9.70	1.03	27.01
$GdH_{2.41}$	378.0	38651	0.529	5.47	0.01	39029	2.74E-02	1068	0.01	30.66
$GdH_{2.88}$	416.5	38651	0.573	7.21	0.01	39068	3.03E-02	1182	0.01	23.23
YH_2	171.7	1.51	0.956	4.66	108.55	173.2	2.84E-02	4.92	2.03	36.04
H_2O	250.5	1.76	0.970	6.32	137.95	252.2	2.60E-02	6.56	1.52	30.47

Table 1: Macroscopic cross-sections and moderating properties of particular hydrides.