IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

"m=1" coatings for neutron guides

This content has been downloaded from IOPscience. Please scroll down to see the full text.

2014 J. Phys.: Conf. Ser. 528 012005

(http://iopscience.iop.org/1742-6596/528/1/012005)

View the table of contents for this issue, or go to the journal homepage for more

Download details:

IP Address: 157.26.72.161
This content was downloaded on 10/08/2015 at 08:34

Please note that terms and conditions apply.

"m=1" coatings for neutron guides

C P Cooper-Jensen[1,2], A Vorobiev[2], E Klinkby[3,1], V Kapaklis[2], H Wilkens[4], D Rats[5], B Hjörvarsson[2], O Kirstein[1,2,6], P M Bentley[1,2]

[1] European Spallation Source ESS AB, Lund, Sweden
[2] Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden
[3] DTU Nutech, Technical University of Denmark, Lyngby, Denmark
[4] General AtomicNes, San Diego,Californian, USA
[5] NeoCoat SA, La Chaux de Fonds, Switzerland
[6] School of Engineering, University of Newcastle, Australia

carsten.cooper-jensen@esss.se

Abstract. A substantial fraction of the price for a supermirror neutron guide system is the shielding, which is needed because of the gamma radiation produced as a result of neutron absorption in the supermirror layers. Traditional coatings have been made of nickel-titanium heterostructures, but Ni and Ti also have a fairly high absorption cross section for cold and thermal neutrons. We examine a number of alternatives to Ni as part of a study to reduce the gamma radiation from neutron guides. Materials such as diamond and Be have higher neutron scattering density than Ni, smaller absorption cross section, and when a neutron is absorbed they emit gamma photons with lower energies. We present reflectivity data comparing Ni with Be and preliminary results from diamond coatings showing there use as neutron guide coatings. Calculations show that Be and diamond coatings emit two orders of magnitude fewer gamma photons compared to Ni, mainly because of the lower absorption cross section.

1. Introduction

Neutron guides were introduced in 1963 [1] to increase the neutron flux at the sample position. The guides were evacuated to prevent the neutrons from scattering on the air molecules, and the low-divergent neutrons hitting the neutron guide walls at low grazing angles were reflected and transported more efficiently. Most materials have a refractive index that is slightly below 1 for cold and thermal neutrons. This means that there is a critical angle, θ_c, below which there will be total external reflection of the neutrons, as demonstrated by Fermi and co-workers [2,3] in the mid 1940s. The critical angle is given by

$$sin\theta_c = \lambda\sqrt{Nb/\pi}$$

where N is the atomic number density of the material and b is the average coherent neutron scattering length. Nb is referred to as the neutron scattering length density. Natural nickel (Ni) has one of the highest neutron scattering length densities of the elements in the periodic table. The critical angle for Ni is defined as m=1. To reflect neutrons at still higher angles than that corresponding to m=1, one could use the ^{58}Ni isotope. This has a higher neutron scattering length density compared to Ni, equivalent to m=1.18. Another option is to use supermirrors, proposed by Mezei [4] in 1976, and which today can be made with m-values above m=7. The cost of Ni isotope enrichment is very high,

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

and ^{58}Ni costs at present around 1000 USD per gram. As such, neutron guides with coatings above m=1 use supermirrors as the most economical option.
Neutrons that are not reflected by the coating will be transmitted, and will be absorbed in the coating or in the substrate. Most neutron absorption processes cause the emission of gamma radiation, and the quantity and energy of the photons depend on the absorbing atom.
The substrates for most neutron guides are made of glass that contains boron, and the products "borofloat" and "borkron" are common. Natural B contains 20% of the isotope ^{10}B, which has a very high absorption cross section for cold and thermal neutrons. When a neutron is absorbed in ^{10}B, the majority of the emitted gamma radiation is at a relatively low in energy (0.477 MeV). The other major components in the substrate are Si and O, which both have an absorption cross section that is many orders of magnitude below that of ^{10}B. Consequently, most of the neutrons absorbed in the glass will be due to absorption in ^{10}B. But before the neutrons can be absorbed by the ^{10}B, the neutrons have to pass through the Ni coating. The absorption cross section for Ni is lower than for B, but large enough to cause substantial absorption. When a neutron is absorbed in Ni, a large part of the emitted gamma radiation has energies exceeding 8 MeV.
The most common used shielding materials around neutron guides are iron, different versions of concrete, and sometimes lead. For all these materials, the attenuation coefficient for 0.5 MeV photons is higher then it is for 8 MeV photons. For lead the attenuation coefficient is 1.2 cm^{-1} for 0.5 MeV photons and 0.43 cm^{-1} for 8 MeV photons [5]. Therefore, to get the same decrease in gamma radiation, one needs nearly 3 times more material if the gamma radiation is at 8 MeV than for 0.5 MeV.
The material with the highest neutron scattering length density in the periodic table is carbon in the form of diamond. Thereafter come Ni and Be, which have nearly the same scattering length density. Both C and Be have a very low absorption cross section compared to Ni, and when a neutron is absorbed both emit gamma radiation at lower photon energies than Ni.
The coating for neutron guides is mainly produced by DC magnetron sputtering, since large areas of optical components can be sputtered efficiently with the quality needed. Be can also be coated using DC magnetron sputtering [6], but it requires special safety procedures. Be dust, which will appear in the coating facility during production, is a health hazard and can lead to a chronic lung disease called berylliosis.
Much effort has been spend developing the manufacture of carbon layers with high number of sp^3 bindings compared to number of sp^2 bindings, making coatings that are more like diamond than graphite. This corresponds to an increase in the mass density of the carbon coatings from 1.8 g/cm^3 for amorphous carbon towards the 3.5 g/cm^3 for diamond. This can be achieved by making coatings that are crystalline diamond or coatings that are amorphous but with high mass density, which are called diamond-like carbon (DLC). Current manufacturing processes for DLC coatings result in elevated concentrations of hydrogen in the coating, which reduce the total neutron scattering length density [7] and therefore make them less useful for coatings in neutron guides applications. DLC coatings without hydrogen for ultracold neutron guides have been demonstrated but these coatings have only had 60 % to 70 % sp^3 bindings and a mass density of 2.88 g/cm^3 [8]. This means that the neutron scattering length density is around the same as for Ni. Alternatively one can get close to 100% sp^3 bindings but these diamond coatings are crystalline [9].

In this paper we will compare Be, crystalline diamond, and Ni coatings to be used as m=1 coatings for neutron guides.

For the reflectivity calculation we have used IMD [10] with optical constants for neutrons from Neutron OPtics Scattering (NOP) [11]. To calculate the emitted gamma radiation when a neutron is absorbed we used MCNPX [12]. Three samples are shown to validate the concept that Ni in guides could be replaced by alternative materials, reducing the emitted gamma radiation as a result.

2. Samples and experimental setup

The Be coating was made by DC magnetron sputtering on a 100 mm Si wafer. The coating facility is normally used for coating thick (>150 μm) Be coatings [6]. The Be coating was 276 nm thick and the roughness of the Be surface was measured to be 0.3 nm using 8 keV (Cu kα) X-ray from a rotating anode.

The diamond coating was made by CVD [9] on a 100 mm x 20 mm fused silica substrate that was 10 mm thick. The diamond coating is on average 225 nm thick as measured by weight, but there is some thickness variation over the sample. The 2 mm from the edges of the samples have a thinner coating than the rest of the sample. The grain size was evaluated from SEM measurements to be 50 nm to 70 nm and the roughness of the coating was measured with 8 keV X-ray to be 6 nm. The deposition temperature of the diamond layer was 800°C. The content of sp^3 bindings is higher than 99% and the hydrogen content is around 2%, mainly concentrated at the grain boundaries.

The Ni sample was made using DC magnetron sputtering, on a system with base pressure of $2*10^{-10}$ Torr. The target was 99.995 % Ni and Ar was used as sputtering gas. The substrate was a 20 mm x 20 mm Si sample with a thermal SiO_2 layer.

The neutron reflectivity measurements shown in this paper were preformed using 4.41 Å neutrons at the SuperADAM reflectometer at ILL [13]. The samples were measured using a non-polarizing periodic Bragg mirror that gives a divergence of the neutrons of 0.27 mrad. The detector was a 2D Position Sensitive Detector (PSD). The Be and diamond coatings were measured with a 0.15 mm horizontal slit in front of the sample. The Ni coating was measured with a 0.05 mm horizontal slit because of the smaller sample size. The size of the vertical slit in front of the sample remained constant at 50 mm in all experiments. All data are corrected for variations in the incoming neutron flux by scaling the data to a beam monitor.

3. Results and discussion

Figure 1 compares the reflectivity measurements of the Ni coating and the Be coating. It is apparent that the critical angles for the Ni and the Be coatings are practically the same, as would be expected since they have a neutron scattering length density that is almost equal, as shown in Table 1. It can also be seen that the measured reflectivity is in good agreement with the calculated reflectivity for Ni.

In Figure 2 we show the measured reflectivity curve for the Be and diamond coating together with the calculated reflectivity curve for Ni and ^{58}Ni. In the angular range 0-0.2 degrees the length of the sample and the horizontal slit size define the shape of the reflectivity curve, which is consistent with total external reflection. The diamond data have been scaled to the Be data so the shape in this region is the same since they the samples have the same length and are measured with the same slit settings.

Table 1. The neutron scattering length density is calculated using data from [14] and the absorption cross section is from [14]. The data is for thermal neutrons (2200 m/s).

	Neutron scattering length density ($1*10^{14}$ m^{-2})	Absorption cross section (barn)
Be	9.63	0.0088
C(amorphous)	6.00	0.0035
C(diamond)	11.70	0.0035
Ni	9.41	4.49
^{58}Ni	13.14	4.50

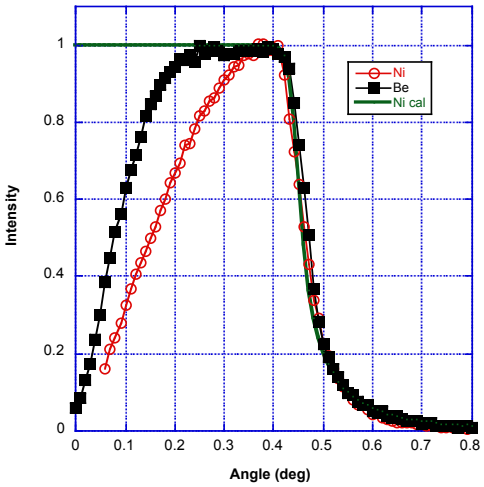

Figure 1. Reflectivity curves of the Ni and the Be samples measured with 4.41 Å neutrons. The thick line is the calculated reflectivity for Ni, the other lines are guides for the eye. The shapes of the experimental curves below 0.3 degrees are caused by the finite sample length and the size of the horizontal slit. Error bars are generally smaller than the marker size.

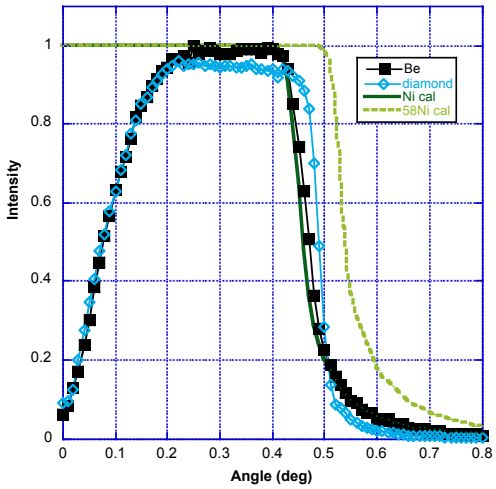

Fig. 2. Reflectivity curves for the Be and the diamond samples measured with 4.41 Å neutrons. The two thick lines are the calculated reflectivity for Ni (solid) and ^{58}Ni (dashed), the thin lines are guides for the eye. The intensity curve for the diamond data is scaled with a constant so the shape of the curve follows that of Be below 0.2 deg. Error bars are generally smaller than the marker size.

From 0.2 degree and to the critical angle of the diamond coating the reflectivity curve is flat as expected for total external reflection, but at a reduced reflectivity of 0.95. This reduction is most likely to be caused by the crystalline diamond coating, which has a high roughness of 6 nm. Furthermore, the sample contains about 2% hydrogen, which are mainly concentrated in the grain boundaries. Hydrogen has a very high incoherent scattering cross section that also contributes to the loss of reflectivity at the sample surface.

We see also that the diamond coating has a higher critical angle than Ni and Be, corresponding to m=1.07, which is nonetheless below the theoretical value, m=1.12, for diamond. There are several reasons for this. The diamond film bindings are likely to not be 100% sp^3 bindings, so the mass

density of the coating is lower than for diamond. The hydrogen contamination and roughness would also be expected to contribute to this lower performance.

We have calculated the gamma radiation emitted from a 250 nm thick coating of the different materials using MCNPX [12]. For each coating 10^9 neutrons were calculated. Since the tabulated values for absorption cross sections are for thermal neutrons (λ = 1.80 Å) we used thermal neutrons with an incidence angle of 0.3 degree for these calculations. For thermal neutrons, m=1 corresponds to 0.18 degrees. Therefore, at 0.3 degrees there will be very little reflection and all the neutrons will be transmitted into the coating. In Figure 3, we show the calculated probability for emitted gamma rays per neutron from the various coatings. The emitted gamma rays from Be and C have fewer than 20 different discrete energies. For Ni, the numbers of discrete energies exceeds 500. In order to make the plot easier to read, the data are shown in 0.1 MeV bins. MCNPX does not include the 2 carbon gamma ray lines at energies of 6.1 MeV and 8.2 MeV, however both have a very small probability to occur.

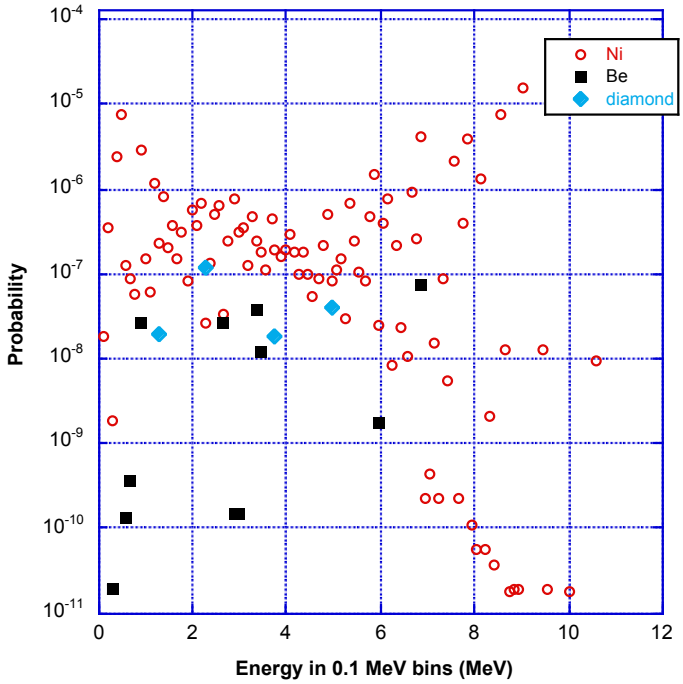

Figure 3. Probability of gamma photon emission from a 250 nm thick films from one grazing incidence neutron as a function of photon energy. The data were calculated using MCNPX as described in the text.

Table 2. The integrated probability for gamma photon emission from one absorbed neutron. The data were calculated using MCNPX as described in the text.

	Gamma ray emitted from one absorbed neutron (Probability)	Total gamma ray emitted from film (Probability)
Be	1.8	$1.8*10^{-7}$
Diamond	1.5	$2.0*10^{-7}$
Ni	1.5	$6.6*10^{-5}$
^{10}B	0.94	-

From Table 2, we see that when a neutron is absorbed the probability for gamma ray emission is the same for Ni and diamond, and for Be the total probability is a little higher. But because the absorption cross section for Ni is order of magnitude larger then for Be and diamond (Table 1), the total number of gamma ray photons from the Ni coating is much larger than for the Be and diamond coating, as shown in Table 2. From Figure 3, we see that the gamma radiation from Ni also has higher energies than that of Be and diamond.
The non-reflected neutrons, which are not absorbed in the coating, will still need to be absorbed, which is why borated substrates are generally used. When a neutron is absorbed in ^{10}B the number of emitted gamma photons is lower than when the neutron is absorbed in Ni, as shown in Table 2. Also the majority of the emitted gamma rays from ^{10}B are at the energy of 0.477 MeV. As a result, by replacing the Ni coating with Be or diamond, one will both reduce the total amount of gamma radiation and shift the radiation to lower energies. Both these two effects will help reduce the amount of shielding required around the guide.

4. Conclusion and future work

Today many new neutron guides have sophisticated shapes compared to traditional straight neutron guides. Especially for long guides there are still parts of the guide where only m=1 coatings are required or where the benefit of using higher m-value coatings do not outweigh the increase in price for the supermirrors. Gamma radiation that is emitted when a neutron is absorbed in the guide has to be attenuated for the safety of the people working in the area and because the gamma radiation can be problematic for some experiments at the end of the neutron guide. Both by reducing the amount of gamma radiation and/or by reducing the energy of the emitted gamma radiation, one will reduce the amount of shielding needed.
With this work we have demonstrated that Be could be a promising coating for neutron guides to replace the traditional Ni coatings. This would be expected to reduce the total emitted gamma ray from the guide, and shift the gamma ray spectrum towards lower energies. Coating the guide optic with Be will be more expensive than coating with Ni, because of the extra precautions that have to be taken during the coating process, but are expected to result in lower overall prices due to the fact that less shielding is needed around the neutron guide. A cost-benefit analysis of this will be made in the near future.
On the other hand, with the high roughness of our diamond sample coating it would not compete as a coating for neutron guides. Instead, we conclude that diamond coatings with a high level of sp^3 bindings will have a m-value that is higher than 1. More work is needed on the diamond-like carbon (DLC) coatings and nano-crystalline diamond coatings, but these could provide performance advantages over both Ni and Be and without the toxicity drawbacks that Be carries.

Acknowledgement

This work was supported under the Cluster of Research Infrastructures for Synergies in Physics (CRISP) program, which is co-funded, by the partners and the European Commission under the 7th Framework Programme Grant Agreement 283745.

References

[1] Maier-Leibnitz H and Springer T 1963 *Reactor Science and Technology* **17** 217
[2] Fermi E and Zinn W H 1946 *Phys.Rev*. **70** 103
[3] Fermi E and Marshall L 1947 *Phys. Rev*. **71** 666
[4] Mezei 1976 *Communications on Physics* **1** 81
[5] Dianoux A J and Lander G 2003 *Neutron Data Booklet, Secound Edition* (Institit Laue-Langevin) p 4.2-1 and 4.2-2
[6] Xu H, Alford C, Chason E, Detor A J, Fuller T, Hamza A V, Hayes J, Moreno K A, Nikroo A, Buuren T, Wang Y, Wu J, Wilkens H and Youngblood K P 2012 *J. Mater. Res*. **27** 822
[7] Grundy M J, Richardson R M, Roser S J, Beamson G, Brennan W J, Howard J, O'Neil M,

Penfold J, Shackleton C and Ward R C 1989 *Thin Solid Films* **172** 269
[8] Heule S, Atchison F, Daum M, Foelske A, Henneck R, Kasprzak M, Kirch K, Knecht A, Kuzniak M, Lippert T, Meier M, Pichlmaier A and Straumann U 2007 *Applied Surface Science* **253** 8245
[9] Mueller C, Haenni W, Binggeli M and Hintermann H E 1993 *Diamond and Related Materials* **2** 1211
[10] Windt D L 1998 *Computers in Physics* **12** 360
[11] Alianelli L, Sánchez del Río M and Felici R 2004 *Physica B* **350** e737
[12] Waters L S, McKinney G W, Durkee J W, Fensin M L, Hendricks J S, James M R, Johns R C and Pelowitz D B 2007 *AIP conference Proceedings* **896** 81
[13] Devishvili A, Zhernenkov K, Dennison A J C, Toperverg B P, Wolff M, Hjörvarsson B and Zabel H 2013 *Review of Scientific Instruments* **84** 025112
[14] www.ncnr.nist.gov/resources/n-lengths/