Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Advanced neutron shielding material using zirconium borohydride and zirconium hydride

T. Hayashi^{a,*}, K. Tobita^a, Y. Nakamori^b, S. Orimo^b

^a Division of Tokamak System Technology, Department of Fusion Facilities, Fusion Research and Development Directorate, Japan Atomic Energy Agency, 801-1 Mukouyama, Naka, Ibaraki 311-0193, Japan

^b Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Sendai 980-8577, Japan

ARTICLE INFO

PACS: 28.52.-s 28.20.Gd 67.63.-r 81.05.Je

ABSTRACT

Neutron transport calculations have been carried out to assess the capability of zirconium borohydride $(Zr(BH_4)_4)$ and zirconium hydride (ZrH_2) as advanced shield materials, because excellent shields can be used to protect outer structural materials from serious activation. The neutron shielding capability of $Zr(BH_4)_4$ is lower than ZrH_2 , even though the hydrogen density of $Zr(BH_4)_4$ is slightly higher than that of ZrH_2 . High-Z atoms are effective in neutron shielding as well as hydrogen atoms. The combination of steel and $Zr(BH_4)_4$ can improve the neutron shielding capability. The combinations of $(Zr(BH_4)_4 + F82H)$ and $(ZrH_2 + F82H)$ can reduce the thickness of the shield by 6.5% and 19% compared to (water + F82H), respectively. The neutron flux for $Zr(BH_4)_4$ is drastically reduced in the range of neutron energy below 100 eV compared to other materials, due to the effect of boron, which can lead to a reduction of radwaste from fusion reactors.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

The fusion DEMO studies at Japan Atomic Energy Agency (JAEA) focuses on low aspect ratio (A) tokamak, SlimCS, to demonstrate an economical power generation with a compact reactor [1–3]. The toroidal magnetic field sharply decreases with the major radius in low-A tokamak. It is important to reduce the nuclear heating arising from both neutron and gamma interactions in order to keep superconductive state of toroidal field (TF) coils. From the aspect of the plasma power, since the toroidal field is one of the most important parameters to achieve a high power density, it is significantly important to shorten the distance between the plasma and the inner leg of the TF coils by using a compact neutron shield. Besides, such an excellent shield can be also used to protect outer structural materials from serious activation, and can lead to a dramatic reduction of radwaste [4–6].

This paper presents neutron shielding capability of advanced neutron shielding materials, zirconium borohydride $(Zr(BH_4)_4)$ and zirconium hydride (ZrH_2) , which are promising materials to store more hydrogen. Fundamental researches have been conducted on complex hydrides to develop practical hydrogen storage materials with higher gravimetric hydrogen densities than those of conventional materials [7–9]. Neutronics calculations were carried

out in order to assess the capability of $Zr(BH_4)_4$ and ZrH_2 as advanced candidate shield materials.

2. Shielding design

2.1. Calculation model

The neutron and gamma-ray fluxes during plasma operation were calculated with the one dimensional Sn code, ANISN [10]. A transport group constant set of FUSION-40 [11], which consists of 42 neutron groups and 21 gamma-ray groups based on JENDL3.3 [12], were used for the calculations. Source neutron energy and wall loading were 14 MeV and 5 MW/m², respectively. The calculation model was a toroidal cylindrical geometry including blankets, shields, vacuum vessels and TF coils on both sides of a plasma, and a central solenoid (CS) coil. The thicknesses of the blanket and shield on outboard side are 0.4 m and 0.7 m, respectively.

2.2. Zirconium borohydride and zirconium hydride

In general, a hydrogen-rich material has the potential to effectively shield neutrons because the contained hydrogen nuclei work as a moderator of fast neutrons, reducing the fast neutron flux. This indicates that neutron shielding capability depends heavily on the hydrogen concentration. Table 1 indicates properties of the $Zr(BH_4)_4$ and ZrH_2 . The anticipated hydrogen concentration of





^{*} Corresponding author. Tel.: +81 29 270 7324; fax: +81 29 270 7419. *E-mail address*: hayashi.takao@jaea.go.jp (T. Hayashi).

^{0022-3115/\$ -} see front matter @ 2009 Elsevier B.V. All rights reserved. doi:10.1016/j.jnucmat.2008.12.073

Table 1	
Hydrogen density and properties of $Zr(BH_4)_4$	and ZrH ₂ .

Material	Appearance	Mass number	Density (10 ³ kg/m ³)	Atomic density (10 ²⁸ /m ³)			
				Н	Zr	В	0
Zr(BH ₄) ₄	Powder	150.60	1.18	7.5	0.5	1.9	-
ZrH ₂	Powder	93.24	5.6	7.2	3.6	-	-
H ₂ O	Liquid	18.02	1.0	6.7	-	-	3.3

 $Zr(BH_4)_4$ is as high as 7.5×10^{28} H-atoms/m³, surpassing those of ZrH_2 and water.

Hydrogen in metal hydrides is generally released at high temperature. However, the dissociation pressure at 800 °C of ZrH_2 is lower than 1 atm [13,14]. The highest surface temperature of the shield is expected to be considerably less than 800 °C. The temperature dependence of the dissociation pressure in $Zr(BH_4)_4$ was not



Fig. 1. Attenuations of neutron fluxes in the outboard shield for various materials: (a) total neutron, and (b) fast neutron above 0.1 MeV.

investigated. Assuming that the dissociation property of $Zr(BH_4)_4$ is comparable to that of the ZrH_2 , $Zr(BH_4)_4$ and ZrH_2 are suitable for the shield of fusion reactors.

3. Result and discussions

3.1. Comparison of shield effect of $Zr(BH_4)_4$ and ZrH_2

Fig. 1(a) and (b) shows the calculated attenuation of neutron fluxes in the outboard shield of 0.7 m thickness for various materials: (a) total neutron, and (b) fast neutron above 0.1 MeV. In Fig. 1(a), ZrH_2 and $Zr(BH_4)_4$ show superior shielding capability than water. The total neutron flux of the Zr(BH₄)₄ is significantly lower than those of ZrH₂ and water in the shallow region (less than 0.2 m depth). This is due to both the presence of boron, whose effect is described in Section 3.3 for details, and the slightly-high H density (see Table 1). However the total neutron fluxes of Zr(BH₄)₄ and ZrH₂ replace each other at a depth of 0.2 m, and they are, respectively, 1.6 and 34 times less than that of water at the back surface of the shield. This is because the atomic density of zirconium of $Zr(BH_4)_4$ (0.5 × 10²⁸/m³) is lower than that of ZrH_2 (3.6 × 10²⁸/ m³). This indicates that neutron shielding mechanism is complex, and that high-Z atoms are effective in neutron shielding as well as hydrogen and boron atoms, which means that the combination of steel and $Zr(BH_4)_4$ can improve the neutron shielding capability.

In Fig. 1(b) and also Fig. 2(b), the attenuation of fast neutron fluxes of $Zr(BH_4)_4$ and water are comparable whereas the constituent elements and the atomic density of H are different each other. This indicates that the boron is not effective in shielding of fast



Fig. 2. Neutron fluxes as a function of the volume ratio of F82H: (a) total neutron, and (b) fast neutron above 0.1 MeV.



Fig. 3. Calculated neutron flux spectra for various materials. The volume ratios of F82H optimized in Section 3.2 were used for the calculations.

neutron as described in Section 3.3, and that the much oxygen of water can make up for both lack of zirconium and its low H density.

3.2. Shielding material mixed with hydride and F82H

The dependence of the shielding capability on the mixing ratio of hydride and steel has been investigated. The calculations were carried out for the hydride combined with F82H, which is one of the promising structural materials under research for fusion because of its good irradiation properties at high temperature and low activation compared with SS316. Fig. 2(a) and (b) shows the calculated neutron fluxes at the back surface of the outboard shield as a function of the volume ratio of F82H: (a) total neutron, and (b) fast neutron above 0.1 MeV. In the case of $Zr(BH_4)_4$ as well as water, the neutron shielding capability becomes higher as the ratio of F82H increases up to 0.7, while the hydrogen concentration decreases with the ratio. For the combination of $Zr(BH_4)_4$ (0.3) and F82H (0.7), the total neutron flux was 8.6 times smaller than that for pure $Zr(BH_4)_4$. On the other hand, the neutron flux for ZrH_2 is fairly constant when the ratio of F82H is less than 0.5, though the hydrogen concentrations decrease with the ratio. In the cases of optimized volume ratios of F82H, concretely the ratios for ZrH_2 , $Zr(BH_4)_4$ and water are 0.4, 0.7 and 0.7, respectively, the total neutron fluxes at the back surface for (ZrH₂ + F82H) and $(Zr(BH_4)_4 + F82H)$ are 4.7 and 1.4 times less than that for (water + F82H), respectively. Thus, ZrH₂ and Zr(BH₄)₄ can reduce the thickness of the shield by 19% and 6.5% compared to water, respectively.

3.3. Neutron spectrum at the back of shield

Neutron flux spectra have been compared in order to investigate the different tendencies between total and fast neutron fluxes for $Zr(BH_4)_4$ and water. In Figs. 1 and 2(a) and (b), the shielding capability of Zr(BH₄)₄ is higher than water for total neutron, but comparable with water for fast neutron. Fig. 3 shows calculated neutron flux spectra at the back surface of the outboard shield for various materials. The volume ratios of F82H optimized in Section 3.2 were used for the calculations. The neutron spectrum at the front of shield is also shown, which is the spectrum when there is no shielding material to avoid the influence of reflected neutron. In the cases of $Zr(BH_4)_4$ and $(Zr(BH_4)_4 + F82H)$, the neutron flux is drastically reduced in the range of neutron energy below 100 eV compared to other materials. Especially in the combination of Zr(BH₄)₄ and F82H, the neutron flux below 10 eV is lower than that of ZrH₂. This is due to the effect of boron, because the ${}^{10}B(n,\alpha)^7Li$ cross section is significantly large in the lower neutron energy region [15]. $Zr(BH_4)_4$ is effective to reduce the induced activity and radwaste arising from neutron capture reactions.

4. Conclusion

Neutron transport calculations have been carried out to assess the capability of zirconium borohydride and zirconium hydride as advanced shield materials. The main points are summarized below.

- (1) The neutron shielding capability of $Zr(BH_4)_4$ is lower than ZrH_2 , even though the hydrogen density of $Zr(BH_4)_4$ is slightly higher than that of ZrH_2 . Thus high-Z atoms are effective in neutron shielding as well as hydrogen atoms.
- (2) The combination of steel and $Zr(BH_4)_4$ can improve the neutron shielding capability. The combinations of $(Zr(BH_4)_4 + F82H)$ and $(ZrH_2 + F82H)$ can reduce the thickness of the shield by 6.5% and 19% compared to (water + F82H), respectively.
- (3) The neutron flux for $Zr(BH_4)_4$ is drastically reduced in the range of neutron energy below 100 eV compared to other materials, due to the effect of boron. $Zr(BH_4)_4$ is effective to reduce the induced activity and radwaste arising from neutron capture reactions, namely (n, γ) reactions.

References

- [1] K. Tobita, S. Nishio, M. Sato, et al., Nucl. Fus. 47 (2007) 892.
- [2] K. Tobita, S. Nishio, M. Enoeda, et al., Fus. Eng. Des. 81 (2006) 1151.
- [3] T. Hayashi, K. Tobita, S. Nishio, et al., Fus. Eng. Des. 81 (2006) 1285.
- [4] T. Hayashi, R. Kasada, K. Tobita, et al., Fus. Eng. Des. 82 (2007) 2850.
- [5] K. Tobita, S. Konishi, S. Nishio, et al., J. Plasma Fus. Res. 77 (10) (2001) 1035.
- [6] K. Tobita, S. Nishio, S. Konishi, et al., J. Nucl. Mater. 329–333 (2004) 1610.
 - [7] S. Orimo, Y. Nakamori, J.R. Eliseo, A. Zuttel, C.M. Jensen, Chem. Rev. 107 (2007) 4111.
 - [8] Y. Nakamori, K. Miwa, A. Ninomiya, H.-W. Li, N. Ohba, S. Towata, A. Zuttel, S. Orimo, Phys. Rev. B. 74 (2006) 045126(1).
 - [9] S. Orimo, Y. Nakamori, A. Züttel, Mater. Sci. Eng. B 108 (2004) 51.
 - [10] W.W. Engle Jr., A User's Manual for ANISN, A One–Dimensional Discrete Ordinates Transport Code with anisotropic Scattering, K–1693, 1967.
 - [11] K. Maki, K. Kosako, Y. Seki et al., Nuclear Group Constant Set FUSION–J3 for Fusion Reactor Nuclear Calculations Based on JENDL–3, JAERI–M 91–072, Japan Atomic Energy Research Institute, 1991.
 - [12] K. Shibata, T. Kawano, T. Nakagawa, J. Nucl. Sci. Technol. 39 (2002) 1125.
 - [13] G.G. Libowitz, The Solid State Chemistry of Binary Metal Hydrides, W.A. Benjamin, Inc., New York, 1965.
 - [14] Y. Yurum (Ed.), Hydrogen Energy System: Production and Utilization of Hydrogen and Future Aspects, NATO Asi Series, Kluwer Academic Publishers, The Netherlands, 1995.
 - [15] <http://www.nndc.bnl.gov/exfor/index.html>.