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The design, fabrication and safety evaluation of a novel spent fuel storage basket material



Peng Wang^{a,b}, Xiaobin Tang^{a,b,*}, Feida Chen^{a,b}, Da Chen^{a,b}

^a Department of Nuclear Science and Engineering, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China ^b Jiangsu Key Laboratory of Nuclear Energy Equipment Materials Engineering, China

HIGHLIGHTS

- Neutron absorption performance of the novel neutron absorbing material was improved by adding three different kinds of neutron absorbers (LiF, Sm₂O₃, and Gd₂O₃).
- The percentage of the three kinds of neutron absorbers was optimised by Monte Carlo method.
- Carbon fibre and polyimide were used to enhance its mechanical behaviour and thermal behaviour.
- The radiation effect of the neutron absorbing material had been studied under Co-60 irradiation, and its irradiation-resistance performance was evaluated.

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ABSTRACT

A novel LiF, Sm_2O_3 , $Gd_2O_3/carbon$ fibre/polyimide material was designed in order to improve the neutron absorbing performance of the spent fuel storage basket in this paper. The volume fraction of three kinds of neutron absorbers (LiF, Sm_2O_3 and Gd_2O_3) in polyimide was optimised by Monte Carlo method. Calculation results showed that the novel neutron-absorbing material, in which the volume ratio of LiF, Sm_2O_3 and Gd_2O_3 was 1:2:13, can achieve the best absorption capacity. Based on the calculated results, the basket material was fabricated by compression moulding, and its mechanical behaviour, thermal behaviour, and irradiation resistant behaviour were evaluated, respectively. The experimental results proved that the tensile strength of the novel neutron-absorbing material was between 195 and 346 MPa and the maximum service temperature was up to 300 °C. Gamma irradiation dose was limited to 160 kGy, the bending strength of the material kept increasing from 10 to 19 MPa.

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1. Introduction

Spent fuel is defined as fuel that has been burned in a reactor. Compared with new fuel, the spent fuel has high radioactivity and can generate spallation neutrons by nuclear disintegration. Since the spallation neutrons may cause a nuclear criticality accident; using neutron absorbing materials, such as cadmium materials, boron plastics, B₄C/Al composites and boron steel, in its transportation and storage is an effective way of safely preventing

E-mail addresses: wp739130046@nuaa.edu.cn (P. Wang),

tangxiaobin@nuaa.edu.cn (X. Tang), fdchen@nuaa.edu.cn (F. Chen), chenda@nuaa.edu.cn (D. Chen).

http://dx.doi.org/10.1016/j.nucengdes.2014.12.010 0029-5493/© 2014 Elsevier B.V. All rights reserved. criticality accidents. However, the aforementioned materials have their disadvantages: cadmium plate is a good absorber of thermal neutrons, but its absorption of epithermal neutrons is very weak (Abrefah et al., 2011). Boron plastic plays an important role as a neutron shielding material; for example, lead boron polyethylene and $B_4C-PbO-Al(OH)_3$ -epoxy composite have extensive applications as neutron absorbing material (Ei-Sayed Abdo et al., 2003; Hu et al., 2008; Okuno, 2005; Sakurai et al., 2004). The tensile strength of B₄C-PbO-Al(OH)₃-epoxy composite is about 50 MPa and the maximum service temperature of lead boron polyethylene is 150 °C. However, compared with metal and metal matrix composites, there are still major gaps in mechanical and thermal performances. The manufacturing process of B₄C/Al composite is very complex (Halverson et al., 1989; Jung and Kang, 2004) and boron steel's boron content is insufficiently high for it to be applicable in this role (Bastürk et al., 2005; Zhang et al., 2013). Therefore, a novel neutron absorbing material needs to be researched

^{*} Corresponding author. Present address: Department of Nuclear Science and Engineering, Nanjing University of Aeronautics and Astronautics, 29 Yudao St., Nanjing 210016, China. Tel.: +86 13601582233; fax: +86 02552112906 80407.

to cope with the problem of neutron absorption from spent fuel.

With the purpose for improving the performance of neutron absorbing materials, this research studied a novel neutron shielding material for the use in spent fuel storage basket. Through adding different neutron absorbers, the basket material can absorb neutrons of different energy. A continuous carbon fibre reinforced polyimide resin was selected as the matrix for the new basket composite material to improve its heat resistance and mechanical properties. Because carbon fibre reinforced polyimide resin has high specific strength, high specific modulus, excellent temperature resistance, and excellent corrosion resistance; it is one type of high-performance material widely applied across various engineering applications. In addition, the neutron absorbing material can also be used in the spaceflight and for radiation protection.

2. Experimental methods

2.1. Calculation model

Compared with protons and electrons, neutrons have no charge and are strongly penetrability. The process of any neutron reaction with matter is divided into two steps: scattering and absorption. Generally, neutrons decrease their energy by scattering and their absorption by nucleus reduces its total number. The polymer is conceptually an excellent neutron-moderator and shielding material (*e.g.* polyimide resin). Substances with large absorption cross-section are chosen as neutron absorbers, *e.g.* ¹⁰B, ⁶Li, or ¹⁵⁷Gd. Traditional basket materials for spent fuel storage use boron carbide as a neutron absorber. Even though boron carbide is good neutron absorption cross-section (Cao et al., 2010). Therefore we chose the rare earth elements (*e.g.* Gd and Sm) for improving neutron absorptivity instead of boron.

A type of neutron absorber only absorbs neutrons with a specific range of energies, but the spent fuel's neutron energy range was too wide for a type of neutron absorber. A type of neutron absorber might not absorb all energy neutrons, so three different neutron absorbers were used in attempts to solve this problem (see Fig. 1(a)). In this work, the neutron energy spectrum of spent fuel assemblies with an initial uranium-235 enrichment of 4%, discharge burn-up of 45 GWD/MTU, and a 5-year cooling time was used (see Fig. 1(b)) (Zhang, 2010). The absorption behaviour of 10 mm thick neutron absorbing material was calculated to find the optimum volume fraction of three different neutron absorbers.

MCNP is particle transport simulation software and has a higher precision in radiation calculations (Ranft, 1967). Many shielding materials were designed by MCNP code (Khan et al., 2011; Miri-Hakimabad et al., 2007). In this work, MCNP-4C was used to design the novel neutron absorbing material.

In order to improve neutron absorption capability of the novel neutron absorbing material, the higher volume fraction of the overall neutron absorbers is better. However, the novel neutron absorbing material could not been formed if the volume fraction of the overall neutron absorbers was higher than 15%. Therefore the maximum volume fraction of the overall neutron absorbers was 15%.

A spherical shell model was established to determine the content for each neutron absorber. With the volume fraction of overall neutron absorbers being constant, the relative proportion of three absorbers was changed in calculations to obtain their neutron transmissivity. From the centre of each spherical shell to the outermost range; the point source was placed at the centre, followed by air and the neutron absorbing material. The outermost part of the model was the count surface and the F2 card (record of average



Fig. 1. (a) Neutron cross section of three kinds of neutron absorbers (LiF, Sm_2O_3 , and Gd_2O_3). (b) Neutron spectrum of spent fuel (4 wt% ²³⁵U, 45 GWD/MTU, cooling time 5 years).

surface flux) was used to assess the number of neutrons transmitted (see Fig. 2).

2.2. Fabrication of polymer materials

A polyacrylonitrile (PAN)-based carbon fibre cloth (with a thickness of 0.3 mm) and a thermosetting polyimide resin (Model:



Fig. 2. The spherical shell model used for calculation.

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