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Effects of Gadolinium in Fe based amorphous ribbons with high boron contents on the neutron shielding efficiency

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ABSTRACT

Fe based amorphous alloys with high boron and gadolinium contents have been developed through a systematic alloy design technique for neutron shielding applications. Amorphous ribbons with the Gd content up to 10 at.% were prepared by a melt-spinning method and used as reinforcements within polyester matrix and concrete matrix, respectively. Excellent neutron shielding efficiency about 75% was obtained from the polyester hexahedron composites (5 cm × 5 cm × 1 cm) including only 1 vol% ribbon with the composition of $Fe_{72}B_{15}Mo_3Gd_{10}$. Moreover, a concrete composite (φ 10 cm × 5 cm thick) in which the amorphous ribbons with the composition of $Fe_{72}B_{24.5}Mo_3Gd_{0.5}$ were uniformly distributed exhibited neutron shielding efficiency of around 86% demonstrating the applicability of amorphous ribbons as reinforcements in various composites for neutron shielding facility constructions.

1. Introduction

As global warming issues become serious, the electric production using nuclear power is considered to reduce the consumption of fossil fuel for the electric production. However, nuclear power plant inevitably arises issues on the safe and economic storage of spent nuclear fuel (Sovacool and Funk, 2013). For the construction of spent nuclear fuel storage facility, thermal neutron absorbing alloys such as borated stainless steels have been applied (Basturk et al., 2005). That is, alloys including elements with high thermal neutron absorption cross section (e.g., Boron or Gadolinium) are used to enhance thermal neutron absorption efficiency (Basturk et al., 2005; Piotrowskia et al., 2015). Moreover, high corrosion resistance and mechanical strength of the alloys are also required to secure reliability of the storage facility. Therefore, developments of new Fe-based alloys with high contents of B and Gd exhibiting high corrosion resistance and mechanical strength are important for cost-effective thermal neutron shielding applications.

In Fe-based crystalline alloys, the solubility of B and Gd is very limited and thus, excess B or Gd forms intermetallic compound(s) deteriorating corrosion resistance and ductility of the alloys (Okamoto, 2013). Therefore, Fe-based alloy with extended B and Gd solubility should be developed. Since B is known to act as a strong glass forming element in Fe-based alloys (Louzguine-Luzgin and Inoue, 2013), amorphous alloys with extended B and/or Gd solubility can be fabricated by a systematic alloy design technique. Farmer et al. reported the development of Fe-based amorphous alloys with high B content and high glass forming ability (e.g., SAM2X5: $Fe_{49.7}Cr_{17.7}Mn_{1.9}Mo_{7.4}W_{1.6}B_{15.2}C_{3.8}Si_{2.4}$). Due to high glass forming ability (GFA) of the alloy (SAM2X5), amorphous coatings can be formed on the canister through a commercial plasma spray coating process. The amorphous coatings exhibited high neutron absorption efficiency and corrosion resistance (Farmer et al., 2008).

As an alternative application example of Fe-based amorphous alloys with relatively low GFA, Fe-based amorphous ribbons with high corrosion resistance, strength and flexibility attributed to the amorphous nature of the ribbons have been attracted. Fe-based amorphous ribbons with the nominal composition of $(Fe, Cr)_{80}(P, C, Si)_{20}$ have been commercialized under the name of FIBRAFLEX and applied as reinforcements in concrete vessels for low level nuclear waste in France (Pech, 1992). However, in most cases of these applications, the Fe-based amorphous ribbons were used to enhance durability rather than thermal neutron absorption efficiency of concrete structures (Hudoba, 2007; Brandt, 2015; Saleh, 2013). Simultaneous improvement of durability and thermal neutron absorption efficiency using Fe based amorphous ribbon reinforcements containing high B and Gd has rarely been studied to date.

In this study, Fe-based amorphous ribbons with high B and Gd have been fabricated through a melt spinning process. The ribbons

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can be applied as reinforcements of various composites to enhance neutron shielding efficiency. Moreover, when the ribbon reinforcements with flexibility, i.e., ductility are added to brittle composites such as concrete, facture toughness of the concrete can be increased with enhanced neutron shielding efficiency. Effects of B and Gd contents on the neutron shielding efficiency, corrosion resistance and mechanical properties have been discussed. The Fe-based amorphous alloys with high B and Gd contents can be used for thermal neutron shielding applications in various types.

2. Experimental procedure

2.1. Preparation and characterization of amorphous ribbons

Master alloys (20 g) were prepared by an arc-melting method under a high-purity argon atmosphere using pure elements (Fe, Mo, Cr, Gd > 99.8%) and ferro-boron (>99%). Ribbons of 22 (±3) μm in thickness and 0.8 (±0.1) mm in width were prepared using a melt spinner equipped with a Cu wheel rotating at a speed of ∼35 m/s.

The phases in the ribbons were identified by X-ray diffraction (D/Max-2500, Rigaku) using CuKα radiation. Thermal properties of each amorphous ribbon were analyzed by The simultaneous TGA/DSC (SDT) (Q600, TA Instruments) at the heating rate of 10 °C/min.

The tensile strength of each alloy ribbon was measured by a universal testing machine (Instron 4482, Instron) operating at a speed of 1.04×10^{-4} s^{-1} with a gauge length of 38–40 mm.

Electrochemical properties of the ribbons were evaluated using a potentiostat (Parstat 2273, Princeton Applied Research) at a scan rate of 1 mV/s in a 3.5 wt% NaCl solution. Platinum counter electrode and saturated calomel electrode references (KCl) were used.

2.2. Thermal neutron transmission test

Thermal neutrons were produced by a polyethylene moderator with dimensions of $20 \times 10 \times 10$ cm^3. The moderator has a hole in the center for the insertion of an ^{241}Am-Be source. Once neutrons are emitted from the source, they are moderated by scattering hydrogen and carbon in the polyethylene during their path in the polyethylene and ∼30% of them become thermal neutrons outside the polyethylene. Fig. 1 shows the neutron energy spectrum per single neutron emission of the exterior of the 5 cm-thick polyethylene moderator in lethargy unit. The spectrum was calculated using a simulation code MCNPX (Monte-Carlo N-Particle Extended)

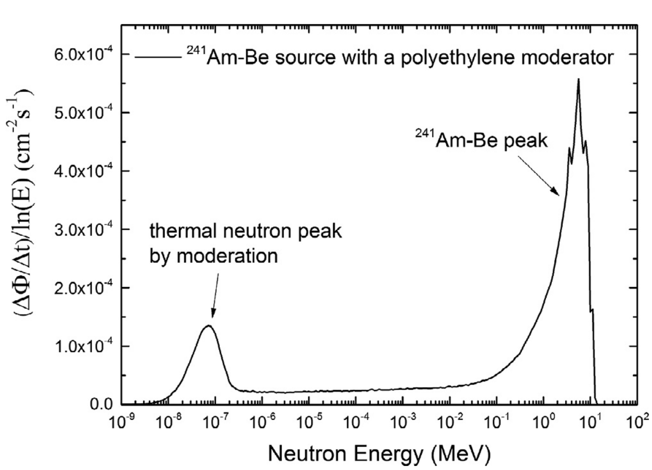

Fig. 1. Neutron energy spectrum of the exterior of the 5 cm-thick polyethylene moderator using a ^{241}Am-Be source.

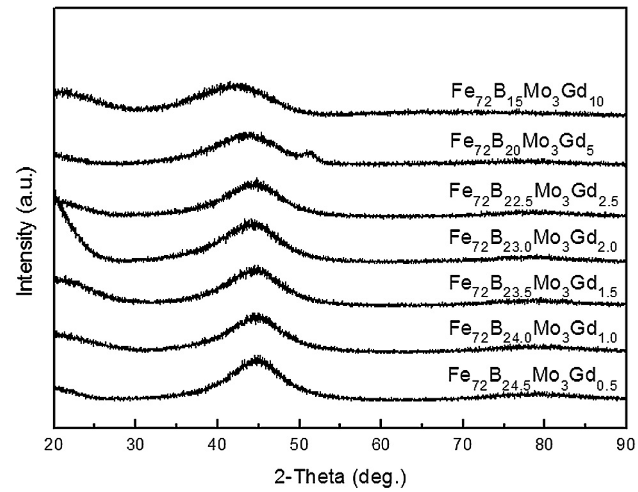

Fig. 2. X-ray diffraction patterns of the melt-spun ribbons $Fe_{72}B_{25-x}Mo_3Gd_x$ (x = 0.5–10 at.%).

(Hughes et al., 1997) (Fig. 1). The left peak is the thermal neutrons by moderation and right peak is the energy distribution of the ^{241}Am-Be source. Specimens were placed in this neutron field and thermal neutron shield effects were measured using a thermal neutron detector which is only sensitive to thermal neutrons. The emission rate of the ^{241}Am-Be source was measured by the KRISS manganese sulphate bath system (Park et al., 2005) and is 2.334×10^5 s^{-1} (1.1%, $k = 1$). The thermal neutron detecting system consists of a ^{3}He-filled spherical proportional counter (type SP9 2 atm, Centronic Ltd., UK), an AIOSAP-02 module and a multichannel analyzer (Easy-MCA 2 k, Ortec). The SP9 detector was covered with cadmium sheets except the front face of the detector in order to shield background thermal neutrons produced in the room. Signals of the SP9 were fed into the all-in-one module AIOSAP-02 which has a high voltage supplier, a pre-amplifier and a shaping amplifier and outputs of the AIOSP-02 were analyzed using the Easy-MCA and the corresponding software. The shielding effects can be calculated by comparing thermal neutron count rates of the SP9 detector with or without test specimens. A thermal neutron count rate was measured without a test specimen in the air and then, the test specimen was installed between the moderator and the SP9 detector one after another and the thermal neutron count rate was measured. The measurement in the air was carried out again to check the stability of the measurement. All measurements were done within 2% uncertainty.

3. Results and discussion

3.1. Properties of ribbons

Fig. 2 shows the XRD patterns obtained from the as-melt spun ribbons $Fe_{72}B_{25-x}Mo_3Gd_x$ (x = 0.5–10 at.%). The XRD patterns exhibited hallow patterns indicating the formation of amorphous phase within the ribbons. As the Gd content increases, two theta value corresponding to the maximum point of the halo pattern in Fig. 2 tends to decrease indicating a dense atomic packing nature of amorphous structure with the Gd additions.

Fig. 3 shows the DSC heating curves of the amorphous ribbons $Fe_{72}B_{25-x}Mo_3Gd_x$ (x = 0.5–10 at.%). As the Gd content increases, the crystallization temperature monotonically increases from 541 °C for the ribbon with 0.5 at.% Gd to 700 °C for the ribbon with 10 at.% Gd, respectively. Single exothermic event was observed during heating of the ribbons with Gd less than 5 at.%, while two separate exothermic events were observed from the ribbons with