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Development of F82H composite materials with a high thermal conductivity

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A B S T R A C T

Composites of F82H and Fe added with W or Cu wires were sintered by spark plasma sintering in order to improve the thermal conductivity of F82H and pure Fe. W and Cu wires were two-dimensionally distributed on the plane perpendicular to pressure direction during sintering. The composites added with 10vol%Cu and 10vol %W were successfully sintered at 40 MPa at 800 °C and 1000 °C, respectively. The thermal diffusivity of the composites were improved compared to base materials, however, many pores were observed especially in the composites with 10vol%Cu. In order to improve the relative density of the composite of F82H added with 10vol %Cu, the use of smaller powder of both F82H and Cu would be needed. On the other hand, W-based compounds were observed in the composite of F82H added with 10vol%W. This would lead to not only less thermal conductivity but also mechanical property of the composite.

1. Introduction

F82H is one of candidate materials for fusion reactor component. F82H has a good mechanical property and lower thermal expansion compared with austenite steel, and also is expected to have a tolerance against neutron irradiation [1]. However, the thermal conductivity of F82H seems to be too low in consideration of the expected thermal load of 0.5 MW/m^2 [2]. On the other hand, W and Cu have higher thermal conductivity (173 W/m/K, 401 W/m/K, respectively). W has advantages of low coefficient of thermal expansion, high strength at elevated temperatures, high sputtering threshold and low tritium retention [3–6]. However, the difference in the coefficients of thermal expansion of W ($\sim 4.3 \times 10^{-6}$/K) and F82H ($\sim 12 \times 10^{-6}$/K) is relatively large, which would cause thermal mismatch and lead to failure for direct bonding of W to F82H. While Cu/Fe layers shows good irradiation recovering ability [7]. In this study, Fe or F82H composites added with Cu or W have been sintered by spark plasma sintering (SPS) in order to improve the thermal conductivity of F82H.

2. Experimental procedure

In this experiment, the composite of Fe added with W or Cu were firstly sintered using Spark Plasma Sintering (DR.SINTER.LAB SPS-510 L) in order to investigate the suitable sintering condition. Chemical composition of F82H used in this study was listed in Table 1. Pure Fe powder (3–5 μm, 99.99%) and F82H powder (~125 μm) were sintered with W (99.9%) or Cu (99.9%), which are of 0.5 μm in diameter and 10 μm in length. Fe or F82H powder and W or Cu wires were mixed by hand-mill with pestle and mortal. The mixed powder and the wires were filled into a graphite die with cylinder of 20 mm in diameter and 40 mm in height. According to Vincent et al., a pressure higher than 30 MPa and a sintering time over 10 min would remarkably decrease porosities in sintered body [8]. Therefore, Fe or F82H composites with W wires were sintered in the condition of the uniaxial pressure of 40 MPa for 10 min at 1000 °C, which was proved to be a property condition for matrix material in previous research [9]. With considering the melting point of Cu (1075 °C), the sintering temperature was decided to be 800 °C. The applied sintering condition is shown in the Fig. 1. Sintered specimens were cut into platelets with 10 mm in diameter, and then, discs of 2 ~ 3 mm in thick were wire-cut and provided for thermal conductivity measurement. W and Cu wires were two-dimensionally distributed on the plane perpendicular to pressure direction during SPS [10]. Thermal conductivity measurement was carried out at room temperature. To calculate thermal conductivity, density (ρ) of each sample was measured with Archimedes' method. Thermal diffusivity (α) and specific heat (C_p) were measured by Laser-flash method (ULVAC TC-7000). The thermal conductivity (λ) was calculated according to following formula [11].

$$\rho = \alpha C_p \lambda$$

Specimen surface was observed by using the Field-Emission Scanning Electron Microscope (2002 JEOL JSM-6500F). The Energy Dispersive X-ray Spectroscopy (JSM-6510LA) was used for quantitative analysis of solute concentration.

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Table 1
Chemical composition of F82H used in this study.

Fe	Cr	W	V	Ta	Mn	Si	C	N	B
Bal.	8	2	0.2	0.04	0.5	0.2	0.1	<0.01	0.003

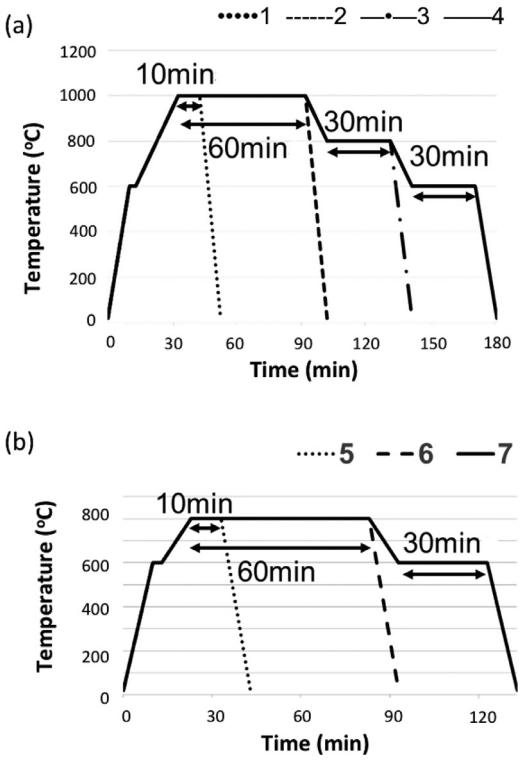

Fig. 1. Temperature and holding time for (a) Fe/F82H + 10vol%W and (b) Fe/F82H + 10vol%Cu. 1: 1000 °C, 10 min; 2: 1000 °C, 60 min; 3: 1000 °C, 60 min; 800 °C, 30 min; 4: 1000 °C, 60 min; 800 °C, 30 min; 600 °C, 30 min; 5: 800 °C, 10 min; 6: 800 °C, 60 min; 7: 800 °C, 60 min; 600 °C, 30 min.

Table 2
Thermal conductivities of Fe or F82H based composites sintered with and without W or Cu wires.

(W/m/K)	Bulk	Sintered without W or Cu	Sintered with 10vol%Cu	Sintered with 10vol%W
Fe	80	42.5	102.5	76. 7
F82H	28	22.4	50.1	31.1

3. Results

3.1. Thermal conductivities and relative densities

Thermal conductivities of Fe or F82H based composites sintered with and without W or Cu wires are listed in Table 2. The thermal conductivities and relative densities of Fe added with 10vol%W: Fe + 10vol%W (light) and Fe added with 10vol%Cu: Fe + 10vol%Cu (shadow) are shown in Fig. 2. According to Pietrak and Wisniewski [12], some equations for the prediction of composites are given. In this study, to simplify calculation, the ideal thermal conductivity was calculated by the equation: $\lambda_{composite} = \lambda_{matrix}V_{matrix} + \lambda_{additive}V_{additive}$, where λ and eans the thermal conductivity and the volume fraction, respectively. In Fig. 2, all the composites of Fe + 10vol%W show good relative densities. In addition, the sintering at 1000 °C for 60 min, followed by at 800 °C for 30 min, resulted in the best thermal conductivity of 76.7 W/m/K. The value of 76.7 W/m/K seems to be still low but acceptable compared with that of bulk Fe (89.7 W/m/K). The density of Fe + 10vol%Cu was increased with holding time from 10 to 60 min. The sintering at 40 MPa at 800 °C for 60 min resulted in the best thermal conductivity of 102.5 W/m/K, which is 28.1% greater comparing with bulk Fe. The thermal conductivities of F82H + 10vol%W and F82H + 10vol%Cu are shown in Fig. 3. The highest thermal conductivities of F82H + 10vol%W and F82H + 10vol%Cu were 31.1 W/m/K and 50.1 W/m/K, respectively. Those values were 99.4% and 160.0% of that in bulk F82H. The sintering condition of those samples was the same as that of Fe-based composite. It seems that the sintering for 10 min would result in the highest thermal conductivity. Furthermore, the relative density of F82H + 10vol%Cu was about 80% of that in bulk F82H. It means that a longer sintering time would not improve the relative density. One of possible reasons on this reduction could be the difference in size between Fe and F82H powders.

3.2. Wire distribution and interfaces

Microstructures of F82H + 10vol%W and F82H + 10vol%Cu were investigated by scanning electron microscopy. As shown in Fig. 4, F82H powder seemed to be well sintered, but some fractures occurred during sintering. This is probably due to a high pressure during sintering. In addition, a high W concentration area was observed on the surface of W wires. Sintered F82H + 10vol%Cu exhibited many pores as shown in Fig. 5. This would be a reason why the relative density was about 80% of that in bulk F82H. EDS analysis was carried out around the interface between matrix and wire in F82H + 10vol%W and F82H + 10vol%Cu (Fig. 6). It seemed that F82H + 10vol%Cu exhibited less diffusion of Cu, while F82H + 10vol%W did a little diffusion of W (20 micro meter from the interface) during SPS.

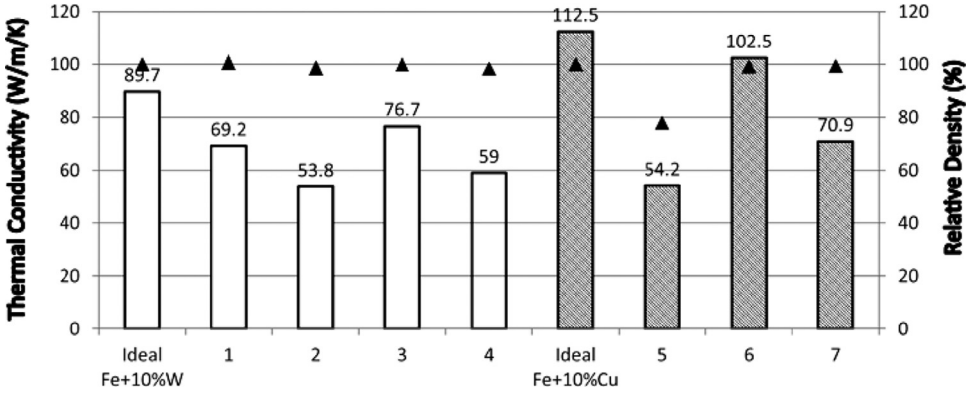

Fig. 2. Thermal conductivities (bars) and relative densities (▲) for Fe + 10vol%W (white bars) and Fe + 10vol%Cu (gray bars) sintered at designated sintering conditions. 1: 1000 °C, 10 min; 2: 1000 °C, 60 min; 3: 1000 °C, 60 min; 800 °C, 30 min; 4: 1000 °C, 60 min; 800 °C, 30 min; 600 °C, 30 min; 5: 800 °C, 10 min; 6: 800 °C, 60 min; 7: 800 °C, 60 min; 600 °C, 30 min.

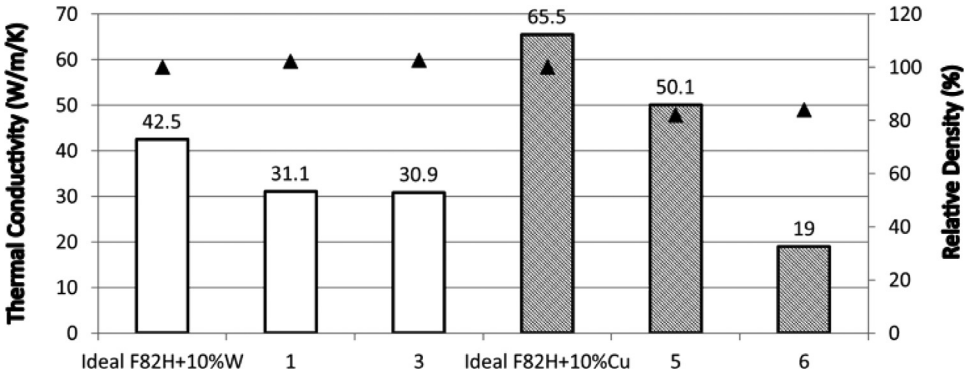

Fig. 3. Thermal conductivities (bars) and relative densities (▲) for F82H + 10vol%W (white bars) and F82H + 10vol%Cu (gray bars) sintered at designated sintering conditions. 1: 1000 °C, 10 min; 3: 1000 °C, 60 min; 800 °C, 30 min; 5: 800 °C, 10 min; 6: 800 °C, 60 min.

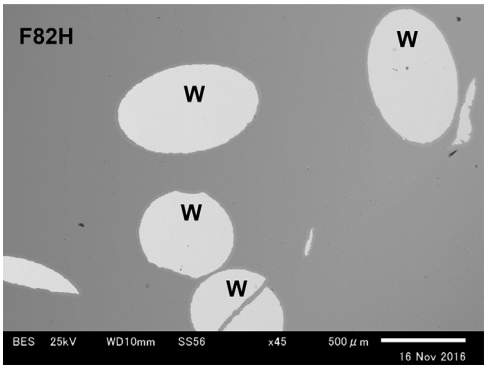

Fig. 4. Typical SEM image of F82H + 10vol%W composite.

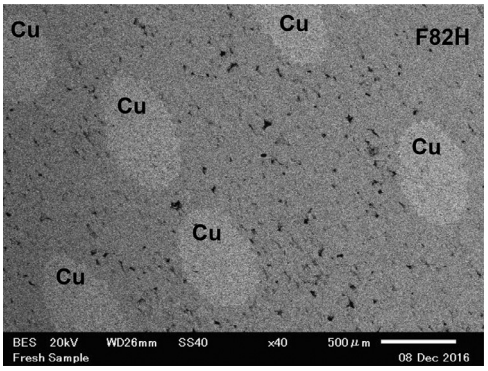

Fig. 5. Typical SEM image of F82H + 10vol%Cu composite.

Furthermore, it appears that W-rich phase was formed around the interface between matrix and W wire.

4. Discussion

W and ferritic steel have differences in their coefficients of thermal expansion [13], melting temperature, and elasticity modulus [14]. Temperature changes induced during cooling from the joining temperature and during subsequent service can generate high internal stresses caused by the mismatch and lead to poor joint strength or failure [15]. Tan et al. reported that the mechanically alloyed 316L-W powders was successfully SPSed at 45.5 MPa at 1050 °C for 5 min, and showed the formation of Fe_7W_6, Fe_3W_3C and Fe_2W phases between W and the 316 L matrix [16]. In this study, W-rich phase was observed in the composite of F82H + 10vol%W after SPS at 40 MPa at 1000 °C for 10 min. With considering the result of EDS analysis, the sintering condition, and Fe-W phase diagram [17], it is suggested that Fe_2W intermetallic compound formation would occur in this condition. The formation of compound at the interface between matrix and W wires would affect not only thermal conductivity but also mechanical property of the composite. Further experiments on the mechanical properties, such as hardness and tensile strength, of this composite would be needed.

F82H + 10vol%Cu composite exhibited lower relative density compared to F82H + 10vol%W. This is probably due to the existence of many pores [18]. It is noted that Fe + 10vol%Cu composite sintered at 40 MPa at 800 °C for 10 min had less pores (Fig. 7) compared with that in Fig. 5. This could be explained by difference in powder size between Fe and F82H. According to Diouf and Molinari [19], the coarse powder could result in a low relative density sintering and the relative density. For instance, the sintering with Cu powders of <25, 25–45, and 45–90 μm in size results in the relative density of 100%, 98.6%, and 97.8%, respectively [7]. The smaller in size would lead to the higher relative density. On the other hand, the potholes on Cu surface could be due to the excess heat. According to Chen et al [7], sintering with larger powders could cause the excess heat on their surface compared with smaller ones, so that the interface between F82H powder and Cu wire could be the source of the excess heat. This would lead to the thermal softening at interface between Cu wire and F82H powders.

5. Summary

Fe and F82H based composite added with 10wt%W or 10wt%Cu wire were sintered by SPS in order to improve thermal conductivity. W and Cu wires were two-dimensionally distributed on the plane perpendicular to pressure direction during SPS, resulted in the improvement of thermal conductivity of Fe and F82H in a certain condition. The thermal conductivities of F82H + 10vol%W and F82H + 10vol%Cu were 99.4% and 160.0% of that in bulk F82H. It seemed that the addition of Cu wire would be more effective on the improvement of thermal conductivity of F82H compared to W wire. The composite of F82H + 10vol%W includes W-rich phase, suggested to be Fe_2W intermetallic compound, which could lead to less thermal conductivity. On the other hand, F82H + 10vol%Cu composite exhibited lower relative density compared to F82H + 10vol%W. In order to improve the relative density of the composite of F82H + 10vol%Cu, the use of smaller powder of both F82H and Cu would be needed.

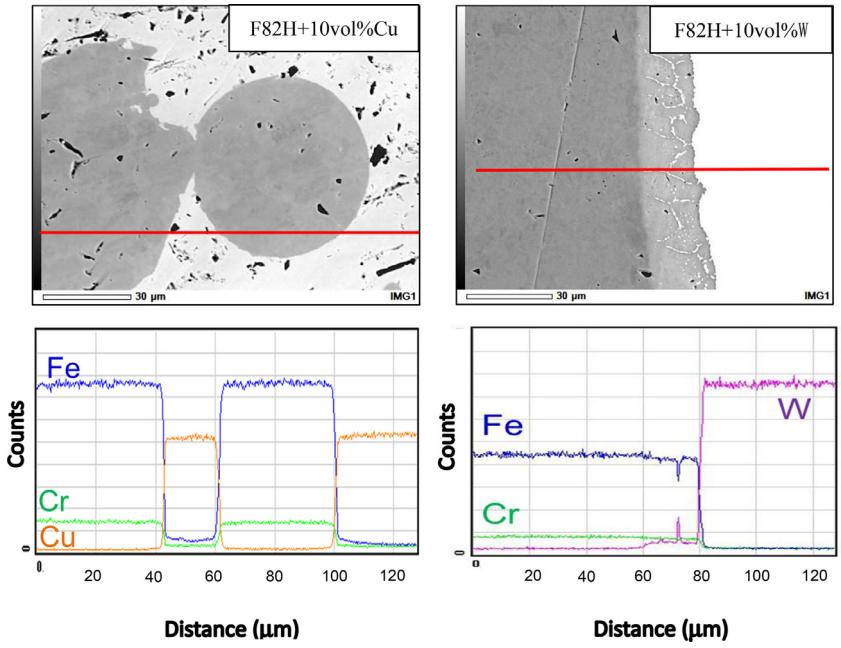

Fig. 6. SEM image and corresponding solute concentration in F82H + 10vol%Cu and F82H + 10vol%W.

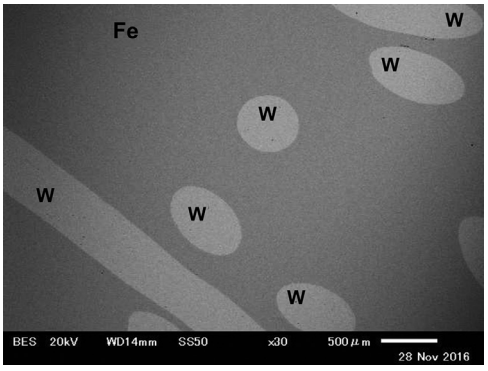

Fig. 7. SEM image of Fe + 10vol%Cu composite sintered at 40 MPa at 800 °C for 10 min.

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