

Interface Reaction between C/C composite and Tungsten Silicide

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The C/C composite produced by laying up carbon fibers was used as a starting material. After cutting the composites in either parallel or perpendicular direction to the carbon fibers, the surface of these samples obtained were reacted with WSi_2 at $1450^\circ C$ in high-purity argon gas. From XRD measurements, the four phases of graphite, WSi_2 , WC and α -SiC were detected on the surface of samples after heating, irrespective of the direction to fibers on C/C composite, and the α -SiC decreased gradually along the depth from surface of the samples, whereas no concentration gradient of WC was observed. In the results of EDX measurements, it was found that the interface reaction between C/C composites and WSi_2 occurred in the matrix portion of the samples. No reaction between fibers and WSi_2 was observed, that is, there is no formation of carbides on carbon fibers in C/C composites.

Key words: C/C composite, Tungsten silicide, Carbide, Reaction

1. INTRODUCTION

Plasma facing components for fusion reactors require excellent erosion properties on the surface against high heat loading from the plasma and high resistance to radiation damage by neutrons with high energy. Carbon fiber/carbon composites (C/C composite) with high thermal conductivity have been extensively used as plasma facing materials in nuclear fusion device [1]. High thermal conductivity and low atomic number are required for facing materials [2]. However, after radiation with the high energy neutrons generated by fusion reaction, the thermal conductivity of materials is lower than that before radiation. And also, the erosion by high heat loading is observed in the materials and results in quite low thermal conductivity at high temperature [3]. Therefore, it is necessary to develop carbon materials that have higher thermal conductivity at high temperature and more stable properties to radiation damage for the use in future fusion reactors.

In order to improve these problems in carbon materials described above, it has been proposed to add metallic powders with high thermal conductivity into carbon materials, such as copper, silver and titanium, etc [4-6]. However, the disadvantage of carbon containing their metals is that the sputtering rates of metal are higher than that of carbon materials, that is, it resulted in radiation damage by high-energy neutrons. In order to solve disadvantage, the use of alloys in which the sputtering rates are small is proposed, such as WSi_2 and $MoSi_2$, etc [7].

As the first step of the research in the present work for preparing the C/C composites coated with WSi_2 , the interface reaction between C/C composites and WSi_2 was carried out at $1450^\circ C$, and the formation of carbide near the interface of the C/C composites after heating was examined.

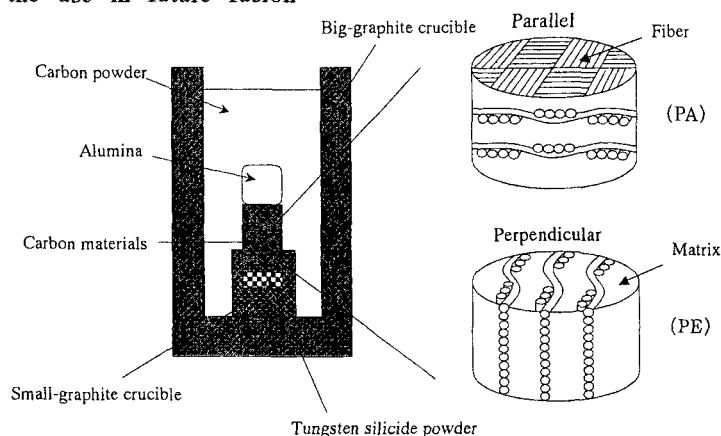


Fig. 1 The shape of carbon materials and the schematic diagram of apparatus used in this study.

2. EXPERIMENTAL

The shape of the carbon materials used in the present study and even a set of the material and the pressed WSi_2 powder in a graphite crucible were illustrated in Fig. 1. The carbon materials used were the carbon fiber/carbon composites (CX-2002U; Toyo Tanso, Co. Ltd, Japan) with a size of $\phi 10\text{mm} \times 20\text{mm}$. These materials cut parallel (PA) and perpendicular (PE) direction to fibers, and two C/C samples were produced. The surfaces of the samples were washed in ethanol with ultrasonic agitation for 5 min in order to remove impurities from its surface. An Al_2O_3 block of 200g was put on the sample in order to obtain a good contact between C/C sample and WSi_2 . The crucible containing this set was heated at 1450°C in high purity argon gas (purity:99.9999%) for 1, 3 and 6h.

In order to know the concentration gradient of SiC along the thickness of the samples, the surface of the samples after heating was polished by diamond paste and then the surfaces polished were measured by X-ray diffraction measurement (XRD). An energy-dispersive X-ray analysis (EDX) was carried out to examine the concentration of either tungsten or silicon along the depth of the samples.

3. RESULTS AND DISCUSSION

Figure 2 shows the changes of XRD patterns along the thickness of the PA sample heated at 1450°C for 6h. The surface of sample gives the diffraction peaks corresponding to the four phases of WSi_2 , WC and α -SiC in addition to graphite. The diffraction lines of WSi_2 are detected up to depth of 0.11mm, which disappear at the depth of 0.23mm. And also, no diffraction lines of WC are detected at the depth of 0.11mm. However, with several repetitions of polishing along the thickness, the diffraction lines of SiC are detected at the depth of 0.23mm, of which the intensity decreases with the increase of the thickness of the sample. The XRD measurement described above was also carried out on the PA samples heated for 1 and 3h.

The phases detected along the thickness of PA sample are summarized in Table 1. The three phases of graphite, WSi_2 and α -SiC are detected on the surface of the samples heated for 1,3 and 6h. On the sample heated for 6h, WC in addition to three phases described above is observed. The WC and the WSi_2 detected on the surface disappear along the depth from the surface of samples, but SiC exists within the PA samples, irrespective of the heating time. For the PE samples, *i.e.*, the samples with perpendicular direction to fibers, XRD measurement was also

carried out on the surface along the depth of samples, as shown in Table 2. In the case of PE samples, the detected phases are similar to that in the case of PA samples. From the XRD measurement, it is found that there is no difference on forming carbides in the interface between C/C samples and WSi_2 , in spite of the direction to fibers in the samples.

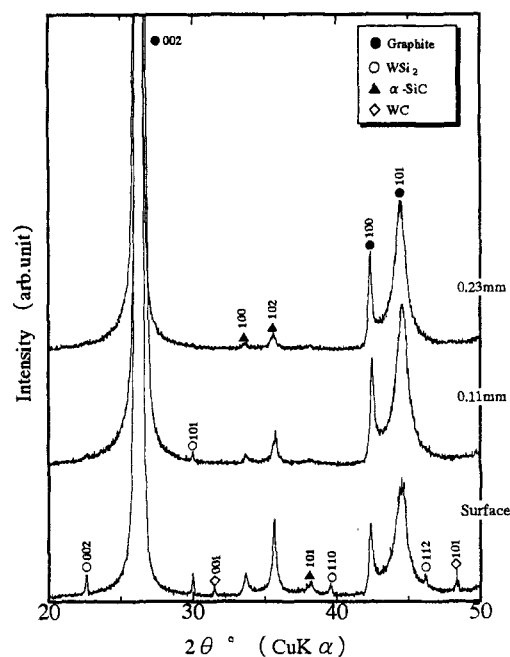


Fig. 2 XRD patterns along the thickness of the PA sample heated at 1450°C for 6h.

In order to examine the portion in the samples reacted with WSi_2 , *i.e.*, either matrix or fibers in the samples; the surface of the PA sample heated at 1450°C for 6h was measured by EDX. In the results, it was found that Si and W existed in the matrix portion of the sample, whereas no existence of these elements was detected in the fiber portion. This result indicates that the reaction between carbon and WSi_2 occurs in the matrix portion of C/C sample. The reason that no reaction between the fiber portion and WSi_2 was observed in C/C samples is not yet clear.

Next, we examined the concentration of W and Si along the depth from the surface of C/C samples by the line analysis of EDX. Fig. 3 shows the results of the PA sample (a) and the PE sample (b) heated at 1450°C for 6h. In the case of PA sample (see Fig. 3(a)), the overall concentration of Si, *i.e.*, concentration of SiC, tends to decrease from the surface to center of the sample even though there is scattering due to the presence of large carbon grains and pores. On

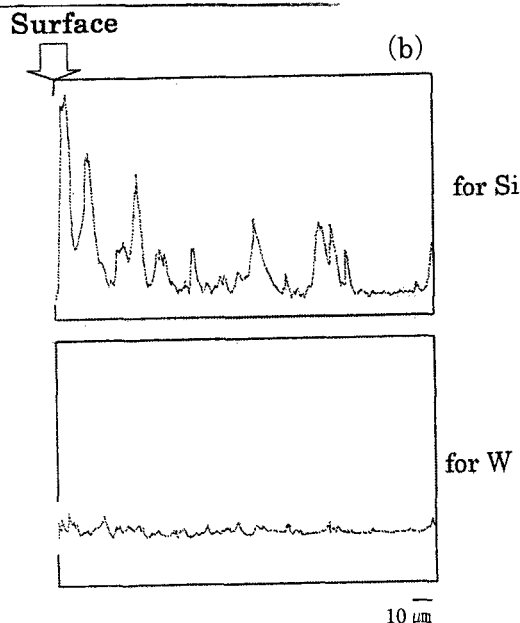
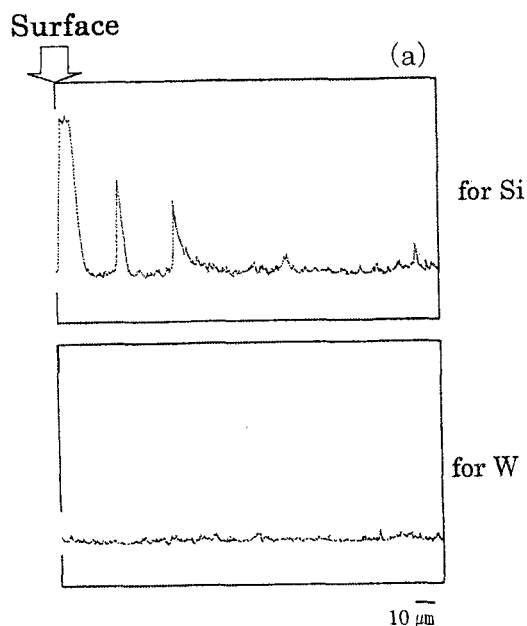
Table 1 The phases detected along the thickness of the PA samples heated at 1450°C.

Time (h)	Depth (mm)	Detected phase
1	Surface	Graphite, WSi_2 , α -SiC
	0.01-0.17	Graphite, WSi_2
	0.20	Graphite
3	Surface	Graphite, WSi_2 , α -SiC
	0.01-0.17	Graphite, α -SiC
	0.20	Graphite, α -SiC
6	Surface	Graphite, WSi_2 , α -SiC, WC
	0.01-0.14	Graphite, WSi_2 , α -SiC
	0.23	Graphite, α -SiC

Table 2 The phases detected along the thickness of the PE samples heated at 1450°C.

Time (h)	Depth (mm)	Detected phase
1	Surface	Graphite, WSi_2 , α -SiC
	0.01-0.15	Graphite, WSi_2
	0.20	Graphite
3	Surface	Graphite, WSi_2 , α -SiC
	0.01-0.15	Graphite, α -SiC
	0.20	Graphite, α -SiC
6	Surface	Graphite, WSi_2 , α -SiC, WC
	0.01-0.08	Graphite, WSi_2 , α -SiC
	0.13-0.21	Graphite, α -SiC

the other hand, W does not exist within PA sample. These results suggest that the concentration of SiC along the thickness decrease gradually, that is, the concentration gradient of SiC is formed within C/C sample. The results in the line analysis of PE sample are comparable with those of PA sample (see Fig. 3(b)). However, a small amount of W is observed within PE sample. In the comparison in SiC-concentration within C/C sample between PE sample and PA sample, it is found that the concentration of SiC on PA sample is lower than that on PE sample.

**Fig. 3** Line analysis for Si and W on the cross-section of the sample heated at 1450°C for 6h: (a) PA sample, (b) PE sample.

From the results of XRD and EDX measurements described above, it is supposed that the concentration gradient of SiC on C/C sample is formed after occurring the dissociation of WSi_2 in the interface between carbon and WSi_2 . On the C/C composites with high porosity, a concentration gradient of SiC was easily formed by reacting with

silicon at 1450°C [8]. These results suggest that it is highly possible for the open pores on the surface of C/C composites to participate in the formation of SiC-gradient. On the C/C composite used in this study, many pores with the size below 50 µm exist on its surface. Yamamoto *et al.* [9] carried out the preparation of SiC within the pores in carbon materials and reported that SiC was formed due to the reaction with SiO gas generated by the reduction of SiO₂ within pores. In this study, it is anticipated that a small amount of O₂ is enclosed in the open pores, because the contact between C/C samples and WSi₂ were carried out in air. Therefore, the formation of a SiC-gradient in C/C composite can be assumed; the oxygen enclosed in the open pores initially oxidizes silicon in WSi₂, and SiO gas is generated. SiO gas thus generated is diffused into the C/C sample through within open pores of matrix. The SiO gas reacts with carbon on the wall of pores and results in SiC on the surface of pores. The W obtained by the dissociation of WSi₂ seems to remain near the interface between C/C sample and WSi₂, and it results in WC by reacting with the carbon of the matrix portion in C/C composite. The reason that the concentration of SiC on PA sample is lower than that on PE sample is supposed to be due to the different in the amount of matrix portion.

4.CONCLUSION

The C/C composites consisting of parallel and perpendicular direction to carbon fibers were reacted with WSi₂ at 1450°C in an argon gas. The interface reaction between C/C composites and WSi₂ was examined. In the case of the composite heated for 6h, the four phases of graphite, WSi₂, α-SiC and WC were detected on its interface, but no formation of WC was observed in the case of composites heated for 1 and 3h, irrespective of the direction to fibers in C/C composites. The formation of these carbides occurred at the matrix portion in C/C composites. The concentration gradient of SiC was formed along the depth from the surface to center of C/C composite. The concentration of SiC in the composite with parallel direction to fibers is lower than that with perpendicular direction to fibers.

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