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Interfacial reactions in aluminum/SiC fibre composite electric power cable using low oxygen SiC fibre reinforcement

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We have evaluated the interfacial reactions of SiC fibre reinforced Al electrical power cable using low oxygen SiC fibre (Si : 62.4, C : 37.1, 0 : 0.5 mass%), and determined the relationship between the tensile strength and the amount of reaction products at the interface. The following are occurring at the SiC/Al interface: i) diffusion of Al atoms into the SiC fibre, ii) formation of needle–shape Al_4C_3 compounds, and iii) formation of Al_9Si compounds. Formation of Al_4C_3 and Al_9Si compounds at the interface causes the strength of SiC/Al composite electric power cable to deteriorate. *© 2000 Kluwer Academic Publishers*

1. Introduction

Electrical power demands are increasing every year, but it is difficult to increase sizes and numbers of the power distribution networks because there is little available ground to built electrical towers on. Therefore, the material for electric cables should be improved so it is lightweight and has a high transmission capacity, high thermal resistance and low sag [1]. We have been breaking new ground in the field of electric cables through the development of a SiC fibre reinforced aluminum conductor [2–5]. Many research reports are performed about SiC/Al composite regarding its interfacial reactions and mechanical characteristics [6–20]. However, there are few detailed reports concerning SiC fibre and interfacial reaction phenomena with Al matrix. We have already analyzed the interfacial reactions in the Al composite reinforced with the high oxygen SiC fibre (Nippon Carbon Co., Ltd. Nicalon^{TM}; Si : 63.7, C : 35.8, 0 : 12.3 mass%) [2, 3]. We have also clarified the reaction of SiC fibre and molten Al [4] and identified a suitable manufacturing process of composite electric cable reinforced with low oxygen SiC fibre (Nippon Carbon Co. Ltd., Hi–Nicalon^{TM}: Si : 62.4, C : 37.1, 0 : 0.5 mass%). Moreover, we have reported the difference between high oxygen SiC fibre and low oxygen SiC fibre in the diffusion phenomenon of Al in the SiC fibre during transmission of electricity, i.e., in the temperature range of the solid state reaction [5].

In this paper, we describe the interfacial reactions of low oxygen SiC fibre and solid state Al. We determine the relationship between reaction products and the tensile strength.

2. Experimental procedure

2.1. Production of SiC fibre/Al composite cable

The production process of the composite electric power cable is shown in Fig. 1. The fibre consists of β-SiC crystallized particles and contains 0.5 mass% oxygen. 1500 pieces of fibre approximately 15 μm in diameter and 500 m to 1000 m in length are formed into one bundle [17]. The “SiC/Al preformed wire” is prepared by dipping the SiC fibre bundle into molten Al (JIS A-1050) at 700°C and reeling it continuously. The preformed wire has a circular cross section with a diameter of 0.85 mm. Dipped SiC fibre/Al wire (diameter: 3.0 mm) is prepared by bundling and dipping seven preformed wires into molten Al, passing through a die,

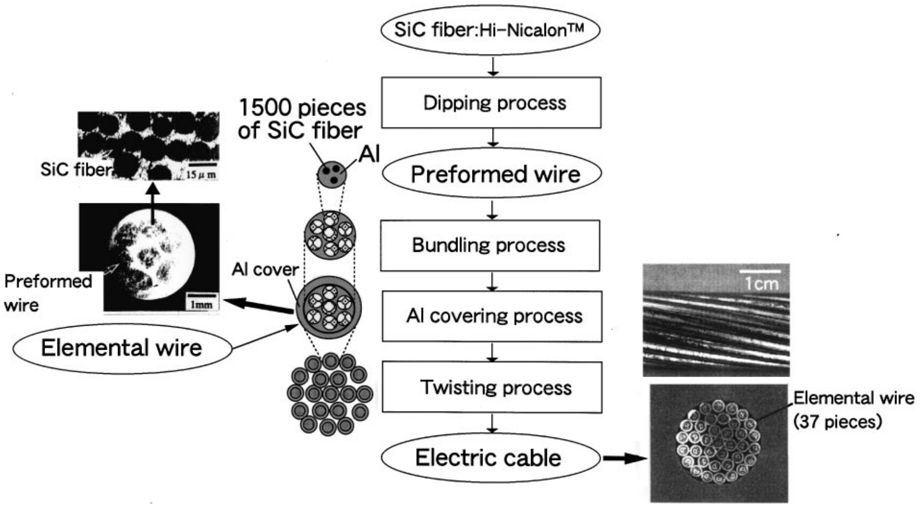


Figure 1 Production process of SiC fiber reinforced aluminum composite electric cable.

and rolling. SiC fibre/Al elemental wire, with a diameter of 3.5 mm and 18% volume fraction of fibre, is obtained by covering the dipped wire with Al by an extrusion process. The electric cable for practical use is produced by bundling 37 SiC fibre/Al composite elemental wires together.

2.2. Analytical methods

To analyze the interfacial reactions and their kinetics, the SiC/Al element wire samples were heat treated in air at temperatures from 300°C to 600°C. A fracture surface and a polished cross section of each sample were analyzed with an optical microscope, SEM (HITACHI S-800, JEOL JSM-6330F) and EDX (EDAX). TEM (HITACHI, H-9000NAR; 300 kV) and EDX (GATAN) analyses were also carried out. The samples for TEM analysis were prepared by the ion-milling method. TEM observations during milling confirmed these samples had a composite structure in which hardness varied greatly. The Al-matrix of the heat-treated composite wires was removed by soaking in 5% NaOH solution to allow direct observation of the surface of the SiC fibre.

3. Results and discussion

3.1. Strength characteristic by solid reaction

Fig. 2 shows the relationship between the survival rate of the tensile strength of the low oxygen SiC fibre/Al composite wires as a function of the heat treatment time with temperature as a parameter. The strength of the composite wire heat-treated above 400°C decreases drastically. To clarify the cause of this decrease, the SiC fibre/Al interface after heat-treating under each set of conditions was analyzed by TEM and SEM methods.

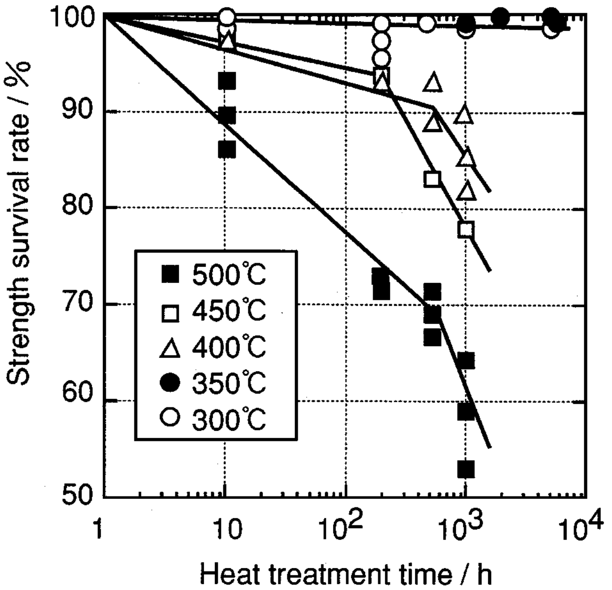


Figure 2 Relationship between the strength survival rate and the heat treatment time of the SiC fiber reinforced Al elemental wires.

The conditions at the interfaces after heat treatment are summarized in Table I. The following may occur at the interface during heat treatment: i) diffusion of Al atoms into SiC fibre, ii) formation of Al_4C_3 compound on the SiC fibre and iii) formation of Al_9Si compound in the SiC fibre. It seems the strength deterioration is caused by the formation of Al_4C_3 and Al_9Si compounds

TABLE I results of TEM analyses and tensile tests of heat-treated elemental wires

	Treatment conditions				
	300°C × 18 Ms	350°C × 21.6 Ms	400°C × 3.6 Ms	500°C × 3.6 Ms	600°C × 0.36 Ms
Strength deterioration	None	None	Yes	Yes	Yes
Al diffusion into SiC fiber	Yes	Yes	Yes	Yes	
Formation of Al_4C_3 compound	None	None	Yes	Yes	Yes
Formation of Al_9Si compound	None	None	Yes	Yes	Yes

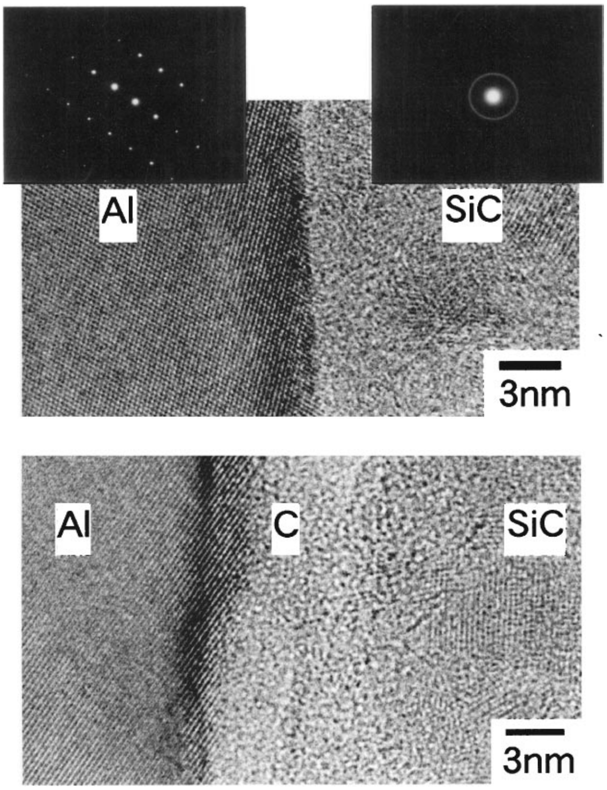


Figure 3 High resolution TEM image of SiC fiber/Al interface of the elemental wire heat-treated at 350°C for 21.6 Ms.

at the interface. The kinetic analysis of Al diffusion into SiC fibre has been described elsewhere [5]. The kinetic analysis of the formation of Al_4C_3 and Al_9Si compounds is discussed in the next section.

3.2. Formation of Al_4C_3 compound

No reaction products could be observed for the conditions of low oxygen SiC fibre/Al heat-treated at 300°C and 350°C. Fig. 3 shows a typical TEM image of the interface heat-treated at 350°C for 21.6 Ms. The low oxygen SiC fibre contained free carbon at the surface of the fibre [17]. On the other hand, the needle-shaped Al_4C_3 compound was formed at the surface of the SiC fibre when heat-treated at 400 to 600°C. As an example, the TEM image of the Al_4C_3 compound formed at the interface of the sample heat-treated at 500°C for 3.6 Ms is shown in Fig. 4. The maximum length of the Al_4C_3 compound was about 3 μm. However, there were also Al_4C_3-free interfaces, as shown in Fig. 5, indicating the reaction was not uniform. An Al_4C_3 compound (about 0.2 μm) was detected locally at the SiC/Al interface of the wire treated at 400°C for 3.6 Ms. Al_4C_3 compound possibly started to grow from the carbon layer on the surface of the fibre towards the Al side.

The growth rate of the Al_4C_3 compound is discussed next. The growth rate of Al_4C_3 compound increases

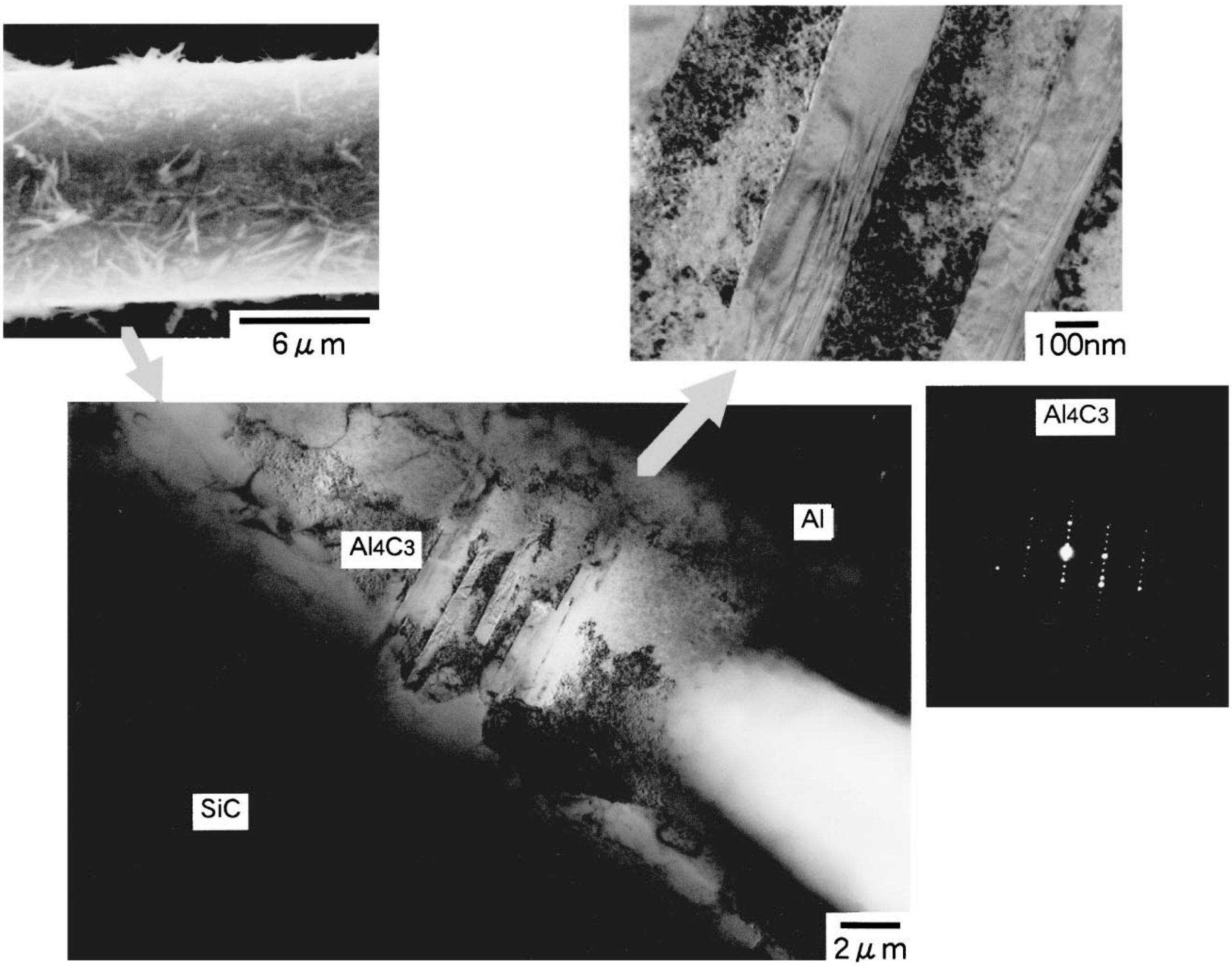


Figure 4 Al_4C_3 compound formed at the SiC fiber/Al interface of the elemental wire heat-treated at 500°C for 3.6 Ms.

parabolically as a function of time t. Here, the rate constant is assumed to follow the law of Arrhenius [3].

$$X^2 = k_p t, \quad \mathrm{kp} = A \exp\left(\frac{-Q}{RT}\right) \tag{1}$$

where X: length of Al_4C_3 compound, A: frequency factor, Q: activation energy, R: gas constant, T: temperature, t: time, k_p: reaction rate coefficient. According to Equation 1, A was calculated as 1.8×10^{-2} and Q as 234 kJ/mol. Thus, if the composite cable is heat-treated at 300°C for 36 years, the amount of Al_4C_3 formed on the SiC fibre can be estimated as 9.3×10^{-8} m.

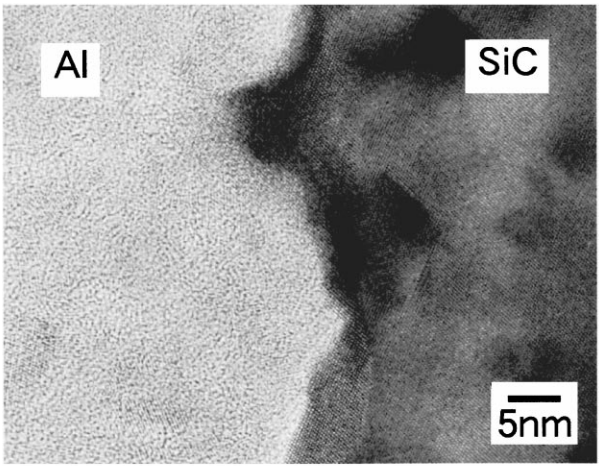


Figure 5 TEM image of the SiC fiber/Al interface of the elemental wire heat-treated at 500°C for 3.6 Ms.

3.3. Formation of Al_9Si compound

EDX analysis was carried out in order to observe the interfacial reaction, and two kinds of interface layers were detected at the SiC fibre/Al interface. The inner layer consisted of Al and Si, and the outer layer consisted of Al and C. TEM analysis was also carried out which indicated that both of Al-Si and Al-C compounds were unity phases, respectively. The Al-C compound was identified as Al_4C_3 by EDX analysis. Though Al_9Si phase does not appear in the binary Al-Si system, the atomic ratio of Al : Si of the compound was identified as 9 : 1 by EDX analysis, thus we assumed this phase as Al_9Si. Al_9Si compounds was formed on the SiC fibre heat-treated at temperatures between 500 and 600°C . Fig. 6 shows the TEM image of the Al_9Si compound formed at the interface of the sample heat-treated at 500°C for 6 Ms. Thickness of the Al_9Si layer was about 0.1 μm for the sample heat-treated at 600°C for 0.36 Ms.

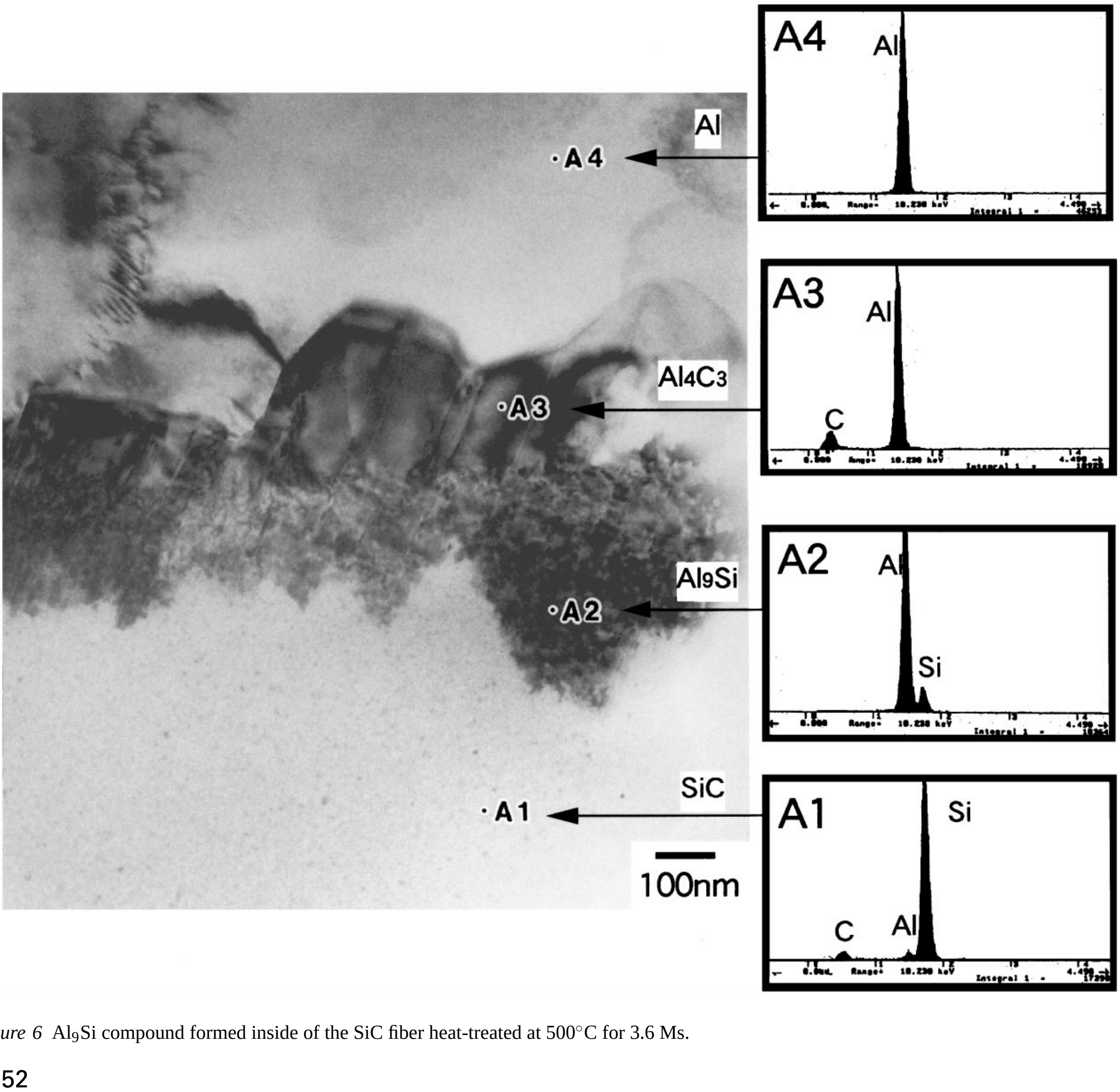


Figure 6 Al_9Si compound formed inside of the SiC fiber heat-treated at 500°C for 3.6 Ms.

3.4. Cause of the strength deterioration

The strength deteriorations of the samples heat-treated at 500°C for 3.6 Ms and 600°C for 0.36 Ms, were caused by the formation of Al_9Si compound because the Al_9Si compound formed inside (not on the surface) of the fibre. However, there was no reaction product in the fibre for the sample heat-treated at 400°C for 3.6 Ms even though its strength decreased. Fig. 7 shows surfaces for Al-removed elemental wires heat-treated at 400°C for 3.6 Ms, 500°C for 3.6 Ms and 600°C for 1.8 Ms. A number of needle-shaped Al_4C_3 compounds were obtained in the samples heat-treated at 500°C and 600°C, while few Al_4C_3 were observed in the sample treated at 400°C.

The most possible fracture model is illustrated in Fig. 8. The fracture origin should be located at the Al_4C_3 for which Young's modulus is relatively high (430 Gpa), and the crack continues to the inside of the SiC fibre for which Young's modulus is low (270 Gpa). The Al matrix and the SiC fibre are strongly joined by the Al_4C_3 formation which suppresses the plastic deformation of the Al matrix and leads to brittle fracture of the composite cable. The sample heat-treated at 400°C for 3.6 Ms contained few Al_4C_3 on the SiC fibre and no reaction product inside of the SiC fibre. Thus its strength decrease indicated that the Al_4C_3 formation led to the strength deterioration. Fig. 9 shows SEM images of the fracture origins of SiC fibre in the tensile fracture surfaces of the as-received composite wire and the heat-treated one (350°C, 21.6 Ms). Fracture origins of almost all as-received fibres were voids, not at the interface, which indicated the suitable bonding between of the SiC fibre and Al matrix. However, fracture origins of the heat-treated sample were located at the interface, which indicated that the Al_4C_3 at the interface deteriorated the tensile strength.

Causes of the strength deterioration at each temperature are summarized in Fig. 10.

In the solid state reaction of low oxygen SiC fibre and Al, it is possible to arrange the amount of reaction

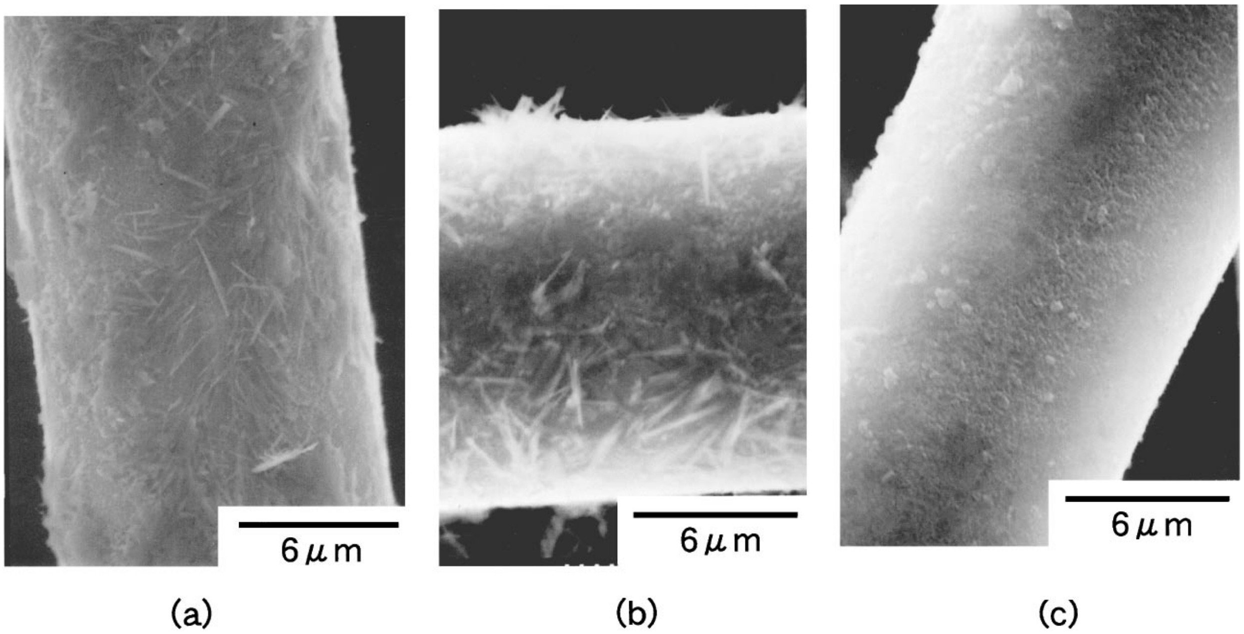


Figure 7 SEM images of Al removed composite cable (i.e. SiC fiber) heat-treated at the conditions of: (a) 600°C for 1.8 Ms, (b) 500°C for 3.6 Ms, (c) 400°C for 3.6 Ms.

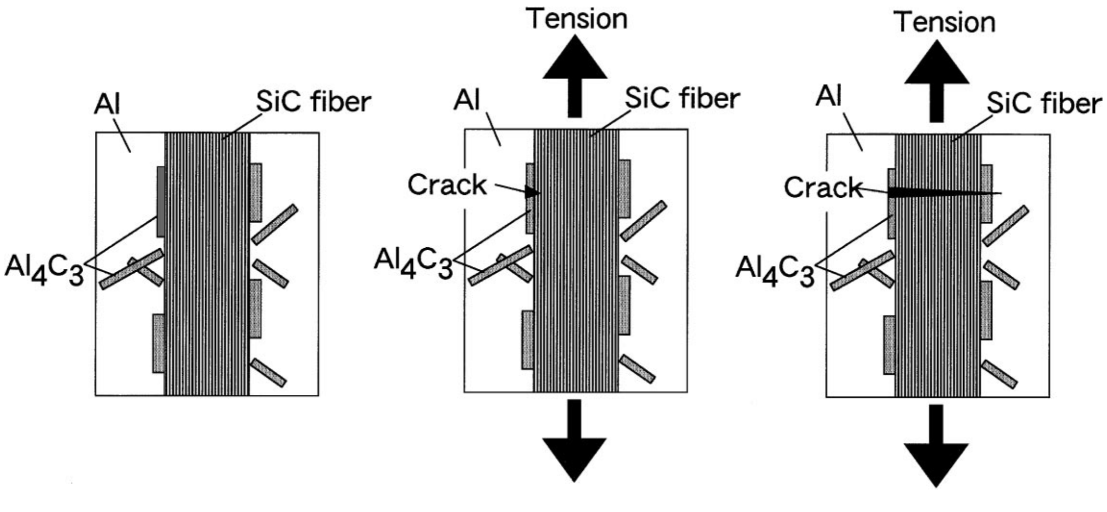
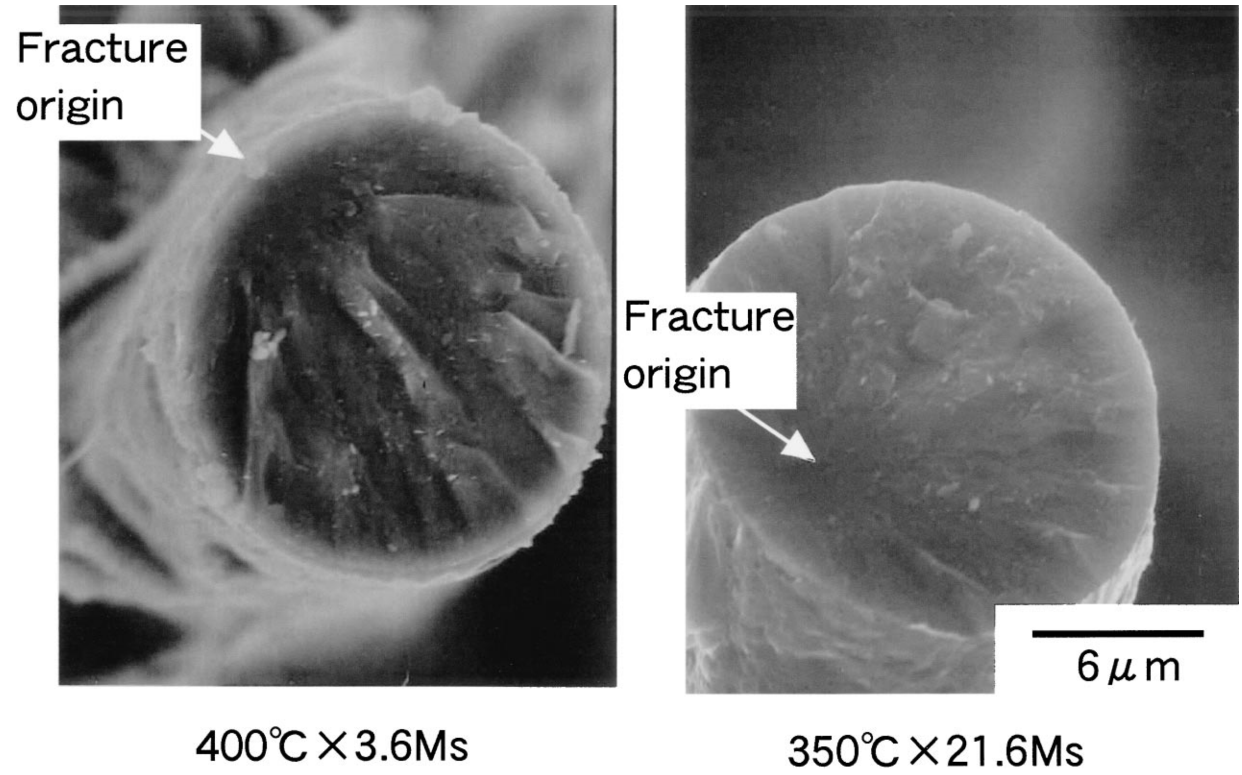


Figure 8 Tensile fracture process of heat-treated composite wire.



400℃×3.6Ms

350℃×21.6Ms

Figure 9 Fracture origins of the heat-treated SIC fiber reinforced Al composites.

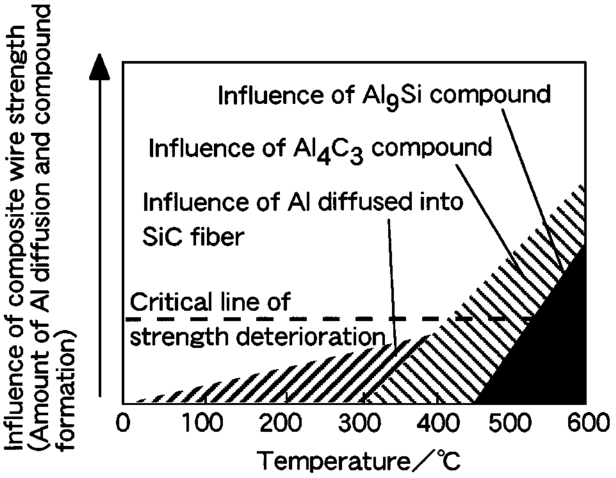


Figure 10 Three factors of strength deterioration of composite wire due to the solid state reaction.

product or Al diffusion regarding their influence on strength at each reaction temperature. It is a borderline (shown with the broken line) above which the strength of composite wire decreases.

(1) Al diffusion at 300°C does not result in strength deterioration.

(2) The formation of the Al_4C_3 compound occurs at around 400°C which results in strength deterioration.

(3) The additional $A1_9Si$ compound forms in low oxygen SiC fibre at 500°C and above, and the strength decreases drastically.

4. Conclusions

We evaluated the interfacial reactions of the SiC fibre reinforced Al electrical power cable using low oxygen SiC fibre (Si : 62.4, C : 37.1, 0 : 0.5 mass%), and determined the relationship between the strength of the cable and the reaction products at the interface. The outcome of this study can be summarized as follows.

(1) The following are occurring at the SiC/Al interface at temperatures between 300°C and 600°C : i) diffusion of Al atoms into the SiC fibre, ii) formation of Al_4C_3 compounds at the interface, and iii) formation of Al_9Si compounds inside the SiC fibre.

(2) Formation of Al_4C_3 and Al_9Si compounds at the interface causes strength deterioration of the SiC/Al composite cable.

(3) Needle-shaped Al_4C_3 compounds are formed on the fibre surface at temperatures between 400°C and 600°C. The growth of Al_4C_3 compound starts from the carbon layer on the SiC fibre.

(4) Activation energy, Q, and frequency constant, A, with respect to the growth rate of Al_4C_3 compound were calculated as 234 kJ/mo1and 1.8×10^{-2}, respectively.

(5) Al_9Si compound is formed at temperatures between 400 and 600°C. There are two kinds of interface layers between the SiC fibre and the Al matrix; the inner layer consists of Al_9Si, and the outer layer is Al_4C_3.

(6) Activation energy, Q, and frequency constant, A, with respect to the growth rate of Al_9Si compound were calculated as 129 kJ/mol and 1.5×10^{-12}, respectively.

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