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Effects of the SiC/Al interface reaction on fracture behavior of a composite conductor using SiC fiber reinforced aluminum for next generation power equipment

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Electrical power demands are increasing every year, meaning that lightweight electric cable is needed which has high transmission capacity, high thermal resistance and low sag. Tokyo Electric Power Co., Chubu Electric Power Co. and Hitachi Cable Ltd. have been breaking new ground in the field of electric cable through the development of a SiC fiber reinforced aluminum conductor. In this work, the SiC/Al interface reaction during the manufacturing process and the electricity transmission temperature were studied by transmission electron microscopy (TEM), energy dispersive X-ray spectroscopy (EDX) and field emission-Auger electron spectroscopy (FE-AES) for long-term reliability assessment. No reaction products were detected at the SiC/Al interface of elemental wire consisting of 7 SiC/Al preformed wires, indicating that the wire manufacturing process was reliable. An Al_4C_3 product was detected locally at the SiC/Al interface of the wire which had been thermally treated in molten Al under unfavorable conditions. The activation energy, Q, of Al_4C_3 growth at the SiC/Al interface was about 190 kJ/mol. In the temperature range of electricity transmission, Al atoms diffused into SiC fiber during heat treatment, and the amount of the diffused Al increased with increasing treatment temperature and holding time. The activation energy of Al diffusion through the SiC/Al interface to SiC fiber was about 78 kJ/mol. Strength deterioration was not induced by Al diffusion into SiC fiber, but strength strongly depended on the formation of Al_2SiO_5 compound at the SiC/Al interface above 400 °C transmission temperatures. Kinetics calculations indicated that the rate of strength deterioration of the composite cable, held at 300 °C for 36 years, was about 5%, so that practical use of SiC/Al composite cable should not be far in the future.

1. Introduction

Electrical power demands increase yearly which leads to an increase in the carrying capacity. But it is difficult to erect more electric cable towers in large cities which have no more vacant space, so alternative means must be considered. The temperature limit of recently developed conductors, such as ultra thermo-resistant aluminum alloy conductors with steel reinforcing (UTACSR), is 200 °C owing to their thermal expansion properties. If the carrying capacity of present thermo-resistant aluminum power cables is doubled, thicker cables become necessary to decrease the Joule

heat generation. This means the present electric cable towers would have to be strengthened. Thus a lightweight electric cable which has high transmission capacity, high thermal resistance and low sag is highly desirable. Joint research on a SiC/Al composite cable has been conducted by Tokyo Electric Power Co., Chubu Electric Power Co. and Hitachi Cable, Ltd. [1, 2]. One of the problems in developing the SiC/Al electric cable is the reaction of SiC fiber and Al which causes deterioration of strength.

Many papers have already dealt with metal matrix composites such as SiC fiber reinforced Al composite [3–29], and the general mechanical properties of SiC fiber/Al composite have been detailed [3–20]. TEM was used to examine the formation of Al_4C_3 products in the interface of the carbon fiber/Al composites [21–23]. Also, X-ray diffraction analysis was used to confirm formation of Al_4C_3 in SiC fiber/Al composite treated at 900 °C [24]. Liu and Wei [25] reported the formation of Al_4C_3, but, theirs was only a guess based on a comparison of thermodynamic considerations and scanning electron microscopy (SEM) observations. To date, few reports have used TEM to study the interface reaction of the SiC fiber/Al composite and none have identified the reaction mechanism of SiC fiber/Al composite.

It is feared that the quality of SiC/Al power cable deteriorates in the production process (carried out at 700 °C) and during transmission of electricity (which occurs at 300 °C). In this report, the SiC/Al interface reaction during the manufacturing process and at the transmission temperature was studied for long-term reliability assessment.

2. Experimental procedure

2.1. Production of SiC fiber/Al composite cable

Production of the SiC fiber (Si: 63.7, C: 35.8, O: 12.3 mass%)/Al composite cable is shown in Fig. 1. The SiC fiber [30] is polycarbosilane fiber, which was oxidation cured at 1200 to 1500 °C. The fiber consists of β-SiC particles, about 3 to 5 nm in size, and contains 12% oxygen. 1500 pieces of fiber approximately 15 μm in diameter and 500 to 1000 m in length are formed into one bundle. SiC/Al preformed wire is prepared by dipping the SiC fibers bundle into molten Al (JIS A-1050) at 700 °C and reeling it continuously. Titanium (0.5 mass%) is added to the molten Al to improve the wettability with SiC fiber. The preformed wire has a circular cross section with a diameter of 0.5 mm. Al is well infiused among the SiC fibers, and few defects are observed. Volume fraction of the fiber is 40%. Dipped SiC fiber/Al wire (diameter: 2.6 mm) is prepared by bundling and dipping 7 preformed wires into molten Al, passing through a die, and rolling. SiC fiber/Al elemental wire, with a diameter of 3.2 mm and 18% volume fraction of fiber, is obtained by covering the dipped wire with Al by an extrusion process. Fig. 2 shows a cross section of the SiC/Al elemental wire. Electric cable for practical use is produced by bundling 27 SiC fiber/Al composite elemental wires together.

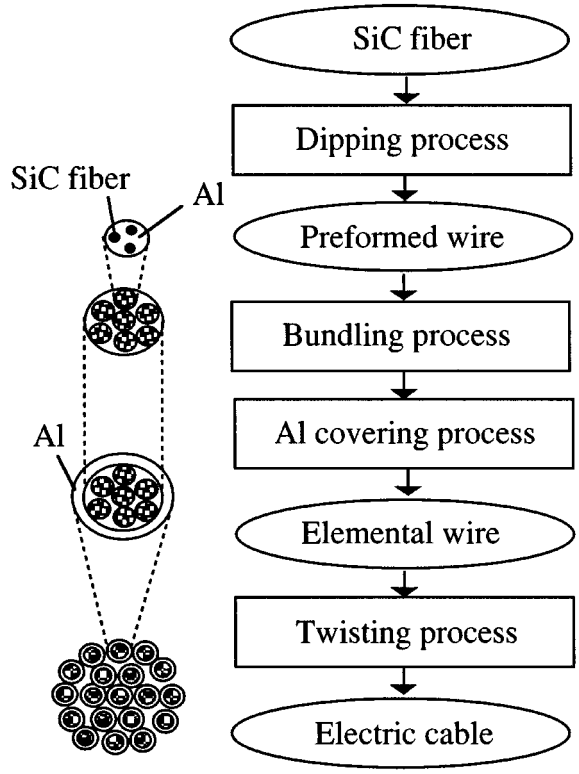

Figure 1 Production flow chart for SiC fiber reinforced aluminum composite electric cable.

2.2. Analytical methods

To analyze the interface reaction between SiC fiber and molten Al, samples were prepared by dipping the SiC/Al elemental wire into molten Al at 700 °C for 2 and 200 min, and at 750 °C for 20 min. To analyze the interface reaction between SiC fiber and solid Al, samples were prepared by heat treating the SiC/Al element wire in air at the conditions of 300 °C for 5000 h, 400 °C for 200 h, 500 °C for 200 h, 500 °C for 529 h, 600 °C for 10 h, and 600 °C for 107 h. Fracture surface and polished surface of each sample were analyzed by optical microscope, SEM (HITACHI S-800) and EDX (EDAX). TEM analysis (HITACHI, H-9000NAR; 300 kV) and EDX (GATAN) analysis were also carried out for detailed observations. Samples for TEM analysis were prepared by the usual ion-milling method. TEM observations while milling confirmed these samples had a composite structure in which the hardness varied greatly. The mechanism of the interface reaction was analyzed by FE-AES (PERKIN ELMER, PHI-670) under the following conditions: acceleration voltage = 20 kV; current = 2 nA; beam diameter = 15 nm at 0 deg angle of inclination and pressure = 6×10^{-8} Pa; removal of the sample surface contamination by Ar^+ iron etching prior analysis.

3. Results and discussion

3.1. Reaction of SiC fiber and molten Al

Fig. 3 shows sectional low magnification TEM images of elemental wire. The SiC fiber is cylindrical and Al fills spaces between the SiC fibers. Fig. 4 shows a high magnification TEM image obtained at the SiC/Al interface. Electron diffraction spots are shown in the insert. No reaction compound was found in the SiC/Al

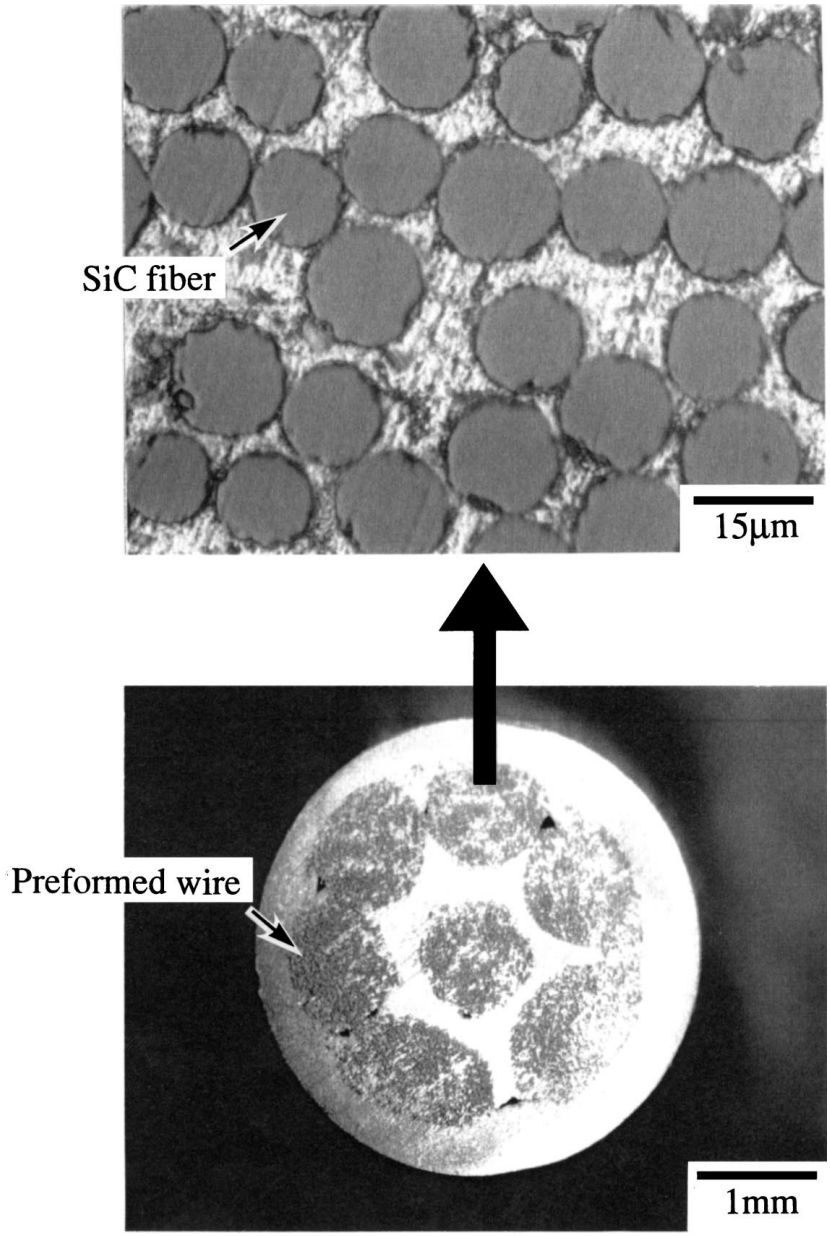

Figure 2 Cross section of the SiC fiber reinforced aluminum elemental wire.

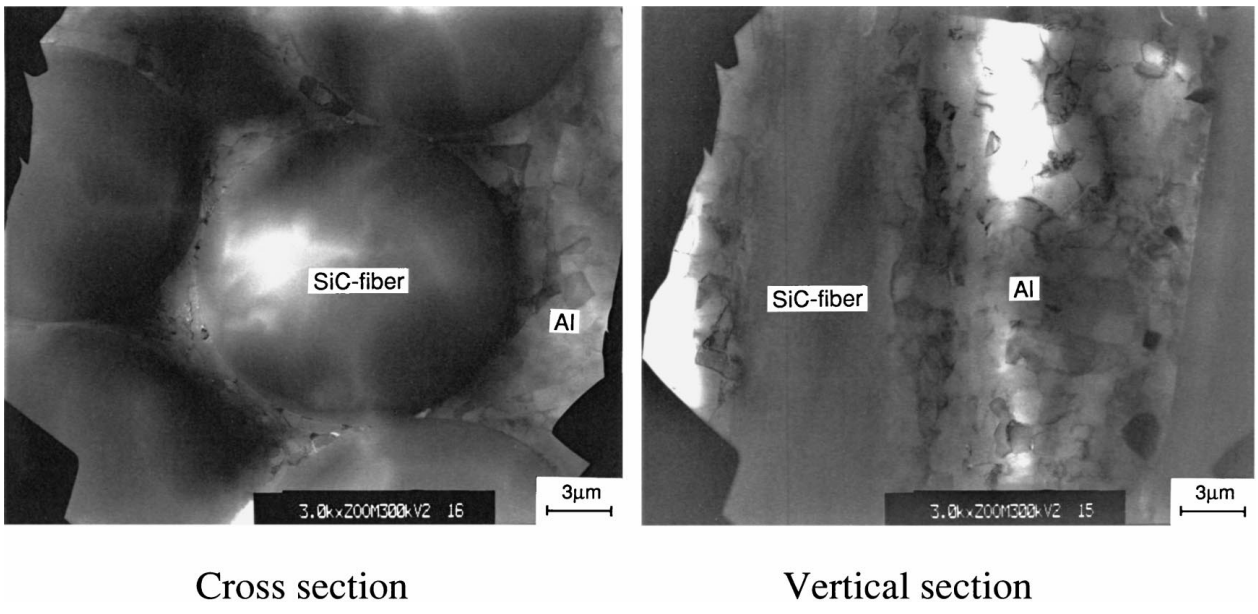

Figure 3 Low maginification TEM images of SiC/Al composite.

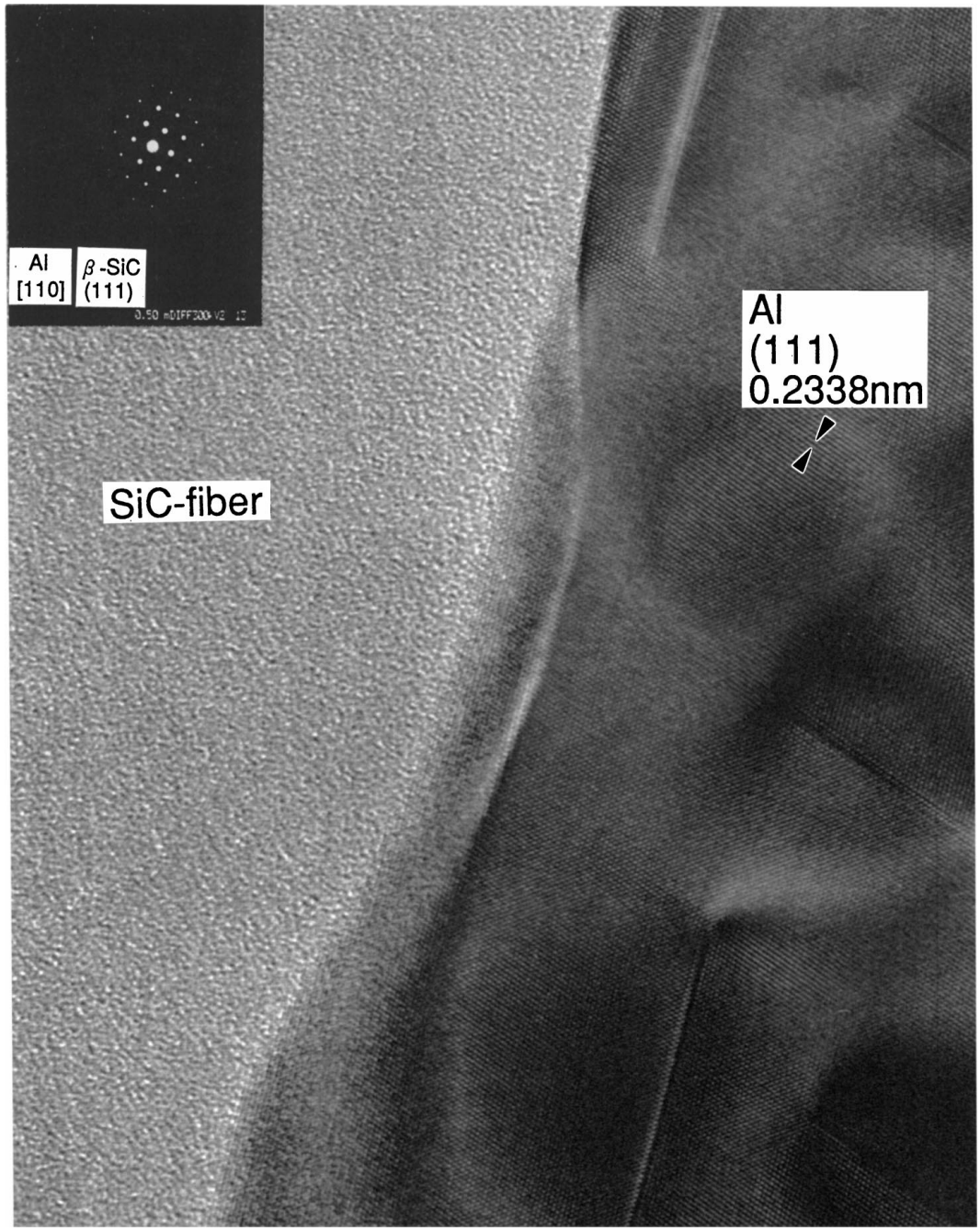

Figure 4 TEM image of SiC/Al interface of elemental wire.

interface nor was Al bonded with SiC fiber directly. Observation of the SiC/Al interface at high magnification TEM showed no reaction layer. No products were detected at the SiC/Al interface of the SiC/Al elemental wires after the last heat-treatment manufacturing process of SiC/Al electric cable, indicating that the wire manufacture process was satisfactory.

Next, samples were analyzed for SiC/Al elemental wire dipped into molten Al at 700 °C for 2 and 200 min, and at 750 °C for 20 min to confirm reaction with molten Al under unfavorable conditions. Al_4C_3 compound was detected locally depths of about 20 nm, 1 μm and 0.1 μm from the SiC/Al interface of the three respective elemental wires. Figs 5 and 6 show TEM images of Al_4C_3 which formed in the SiC/Al interface of the thermally treated elemental wire (molten Al, 700 °C for 200 min). Wedge-shaped Al_4C_3 compound eroded the SiC fiber. Fig. 7 shows SEM observation results of SiC fiber state after Al removal by acid from the elemental wire which had the same thermal treatment. SiC fiber surface was porous due to erosion by the Al_4C_3 compound. So, Al_4C_3 compound eroded the SiC fiber which had been thermally treated in molten Al for a long time.

From the size of Al_4C_3 reaction compound obtained in the above analysis the growth rate of Al_4C_3 compound was calculated. The relation between size of the reaction compound and heat treatment conditions can be expressed in the next equation.

$$X^2 = k \cdot t \tag{1}$$

X is size of reaction compound Al_4C_3, t is heat treatment time and T is heat treatment temperature. K is the

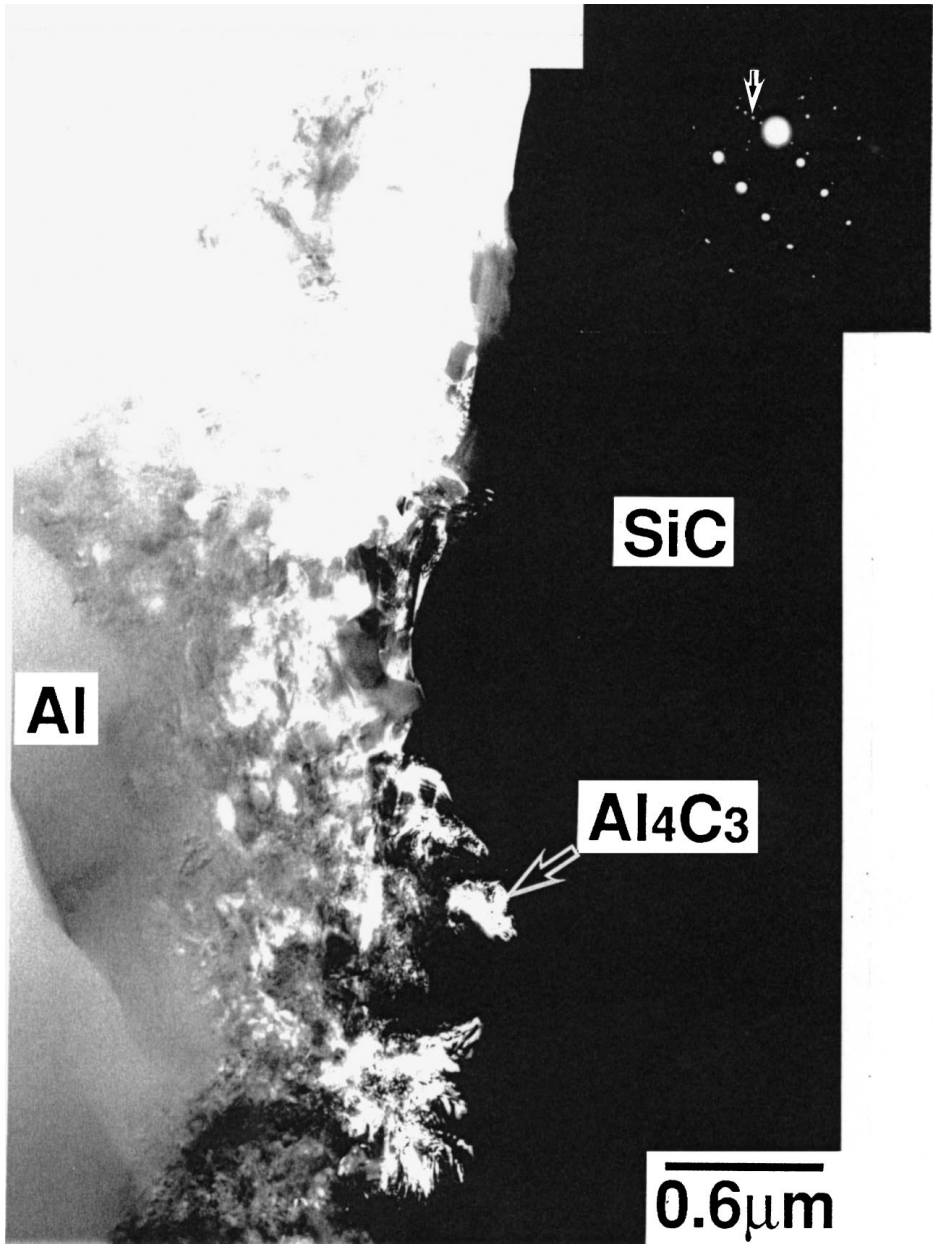

Figure 5 Dark field TEM image of Al_4C_3 compound in the SiC/Al interface of thermally treated elemental wire (molten Al, 700 °C for 200 min).

reaction coefficient given as

$$k = A \cdot \exp(-Q/R \cdot T) \qquad (2)$$

where A is a constant and Q is activation energy,

Constant $A = 1.0 \times 10^{-7}$ and activation energy $Q = 190$ kJ/mol were calculated. The latter was almost the same activation energy as for Al_4C_3 growth in the SiC fiber/Al interface reaction obtained by differential thermal analysis [22]. As mentioned above, no products were detected at the SiC/Al interface of the elemental wires, indicating that the wire manufacture process is satisfactory.

3.2. Diffusion of Al into SiC fiber during the reaction between solid Al and SiC fiber

The SiC/Al interface of elemental wires which had been thermally treated from 300 to 600 °C was analyzed to confirm long term reliability at 300 °C for 36 years. Fig. 8 shows analysis results by FE-AES. Mutual diffusion occurred, so Al atoms diffused into the SiC fiber while Si and C atoms diffused to the Al side for elemental wires which had been thermally treated at 600 °C for 10 and 107 h. Fig. 9 shows Al concentrations obtained by TEM-EDX analysis, which were used to calculate diffusion rate of Al into SiC fiber. In the interface there were no reaction products. Table I lists diffusion coefficients calculated than Fick's law using Al concentration measurement results of Fig. 9. The activation energy of Al diffusion through the SiC/Al interface to SiC fiber was about 78 kJ/mol. And, the quantity of Al diffusion into SiC fiber at 300 °C after 36 years was 5.6×10^{-7} m.

Fig. 10 shows the relationship between strength survival rate and the quantity of the diffusion calculation value of Al in samples which had been thermally treated under different conditions. There was no strength deterioration at 300 °C or 350 °C for the long treatment. Above 400 °C, strength deteriorated suddenly with the

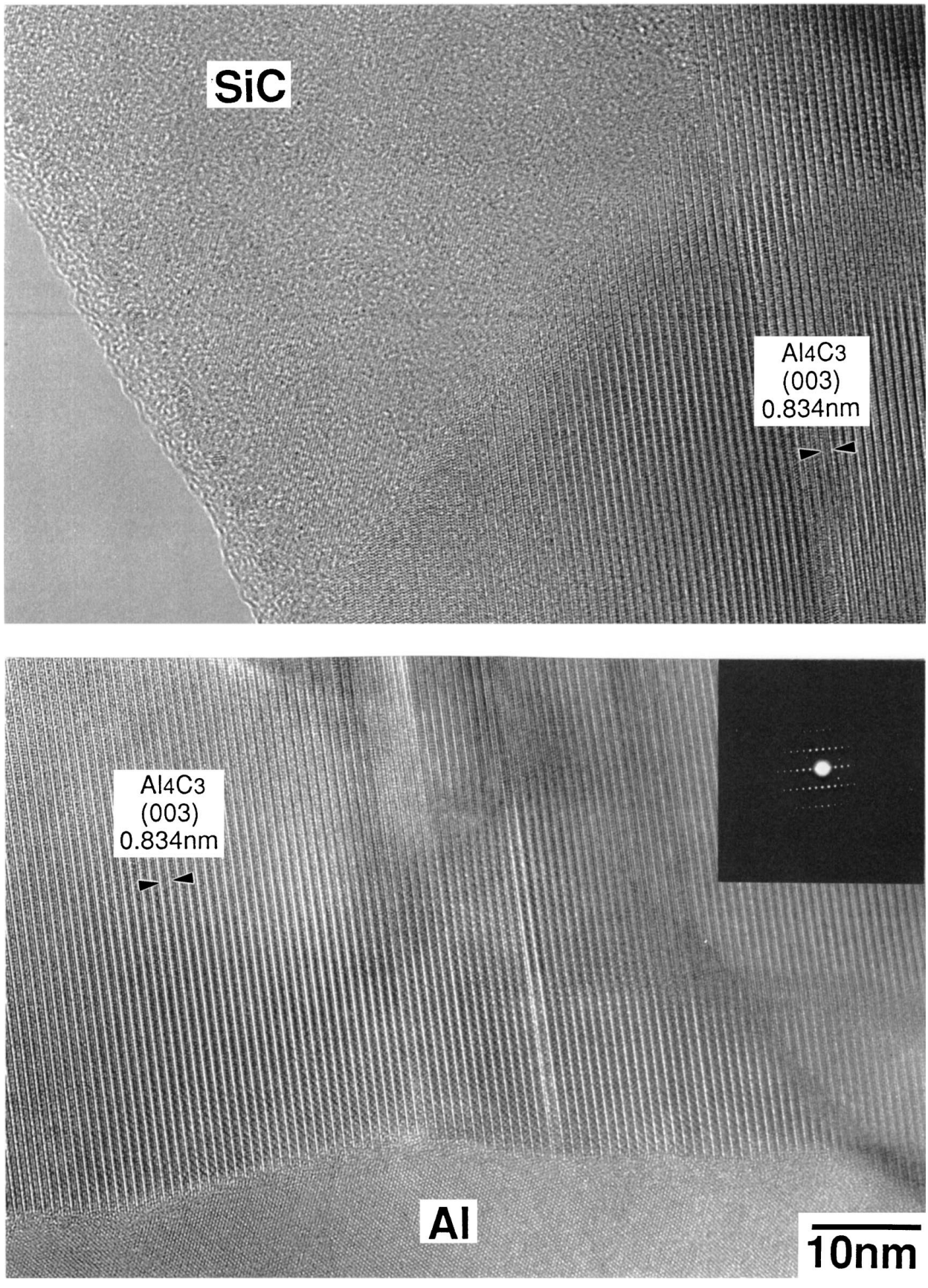

Figure 6 High magnification TEM images of Al_4C_3 compound in the SiC/Al interface of thermally treated elemental wire (molten Al, 700 °C for 200 min).

TABLE I Diffusion coefficients of Al in SiC fiber calculated than Fick's law using Al concentration measurements by TEM-EDX

Treatment conditions	600 °C × 10 h	500 °C × 200 h	500 °C × 529 h	300 °C × 5000 h
Al concentration at 0.1 μm in SiC fiber from interface (at %)	4.73	2.90	8.95	2.15
Diffusion coefficient (m^2/s)	4.9×10^{-20}	1.9×10^{-21}	1.5×10^{-21}	6.8×10^{-23}

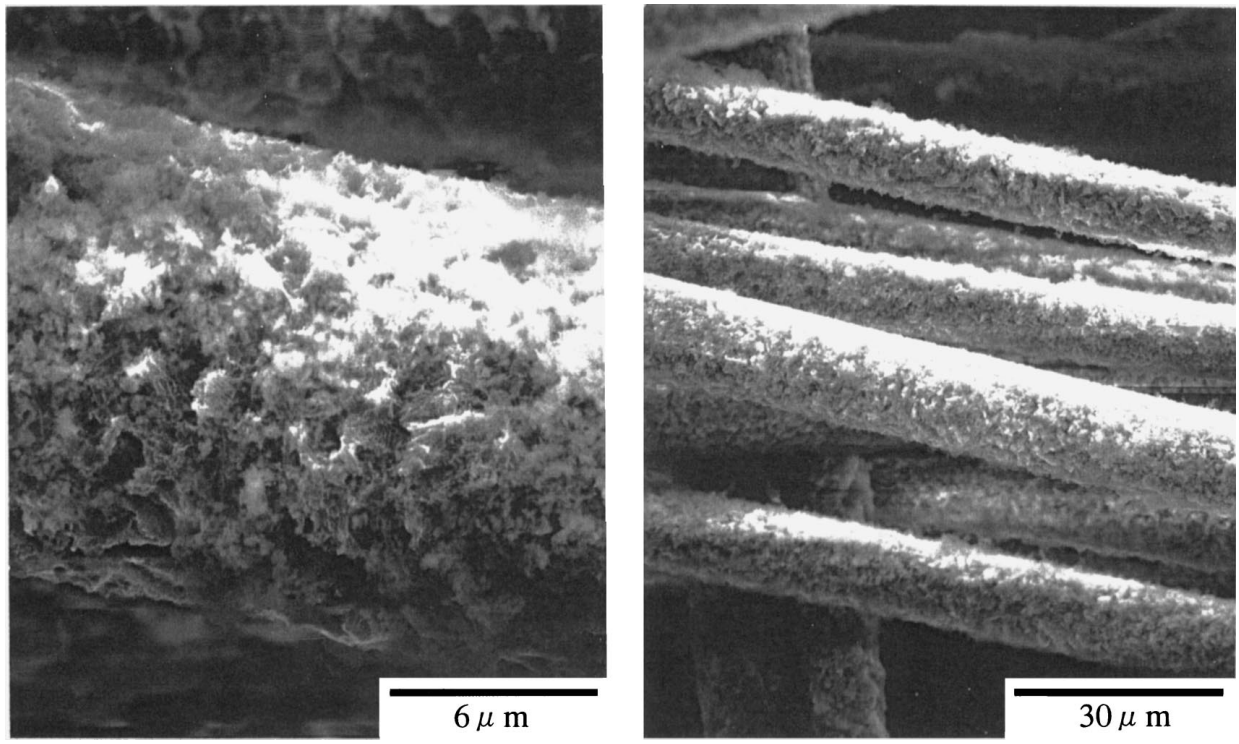

Figure 7 SEM image of SiC fiber after Al removal from thermally treated elemental wire (molten Al, 700 °C for 200 min).

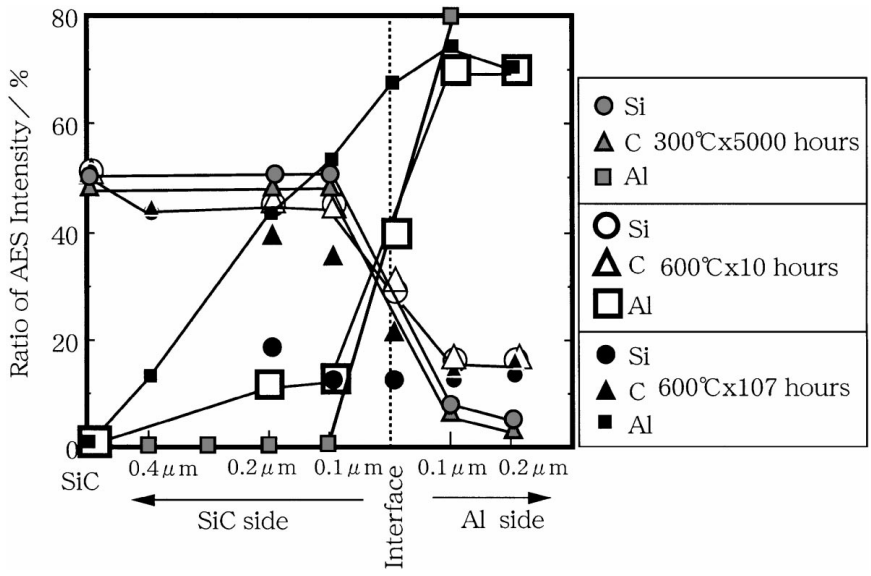

Figure 8 FE-AES analysis of SiC fiber/Al interface of heat treated elemental wires.

increase of Al diffusion. Curves were different at 300 °C and above 400 °C in Fig. 10. Fig. 11 compares the fracture surface after the tensile test of elemental wires which had been thermally treated at 300 °C for 5000 h and 500 °C for 200 h. For the treated at 300 °C, traces of pull out of SiC fibers were found after the tensile test. On the other hand, for the 500 °C treatment, the fracture surface was flat, and SiC Fibers and Al were strongly

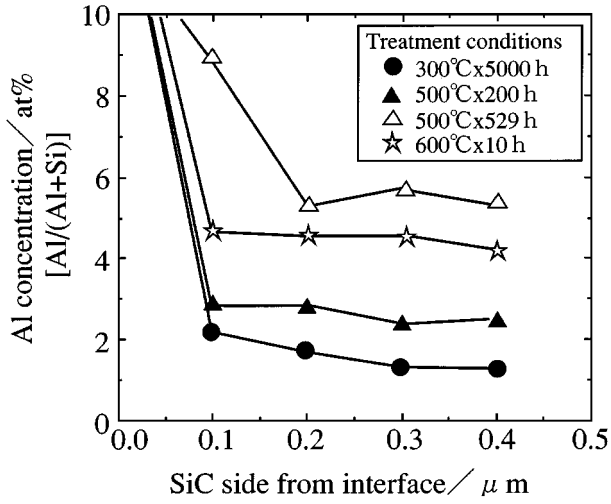

Figure 9 Al concentration by TEM-EDX analysis in SiC fiber of thermally treated elemental wires.

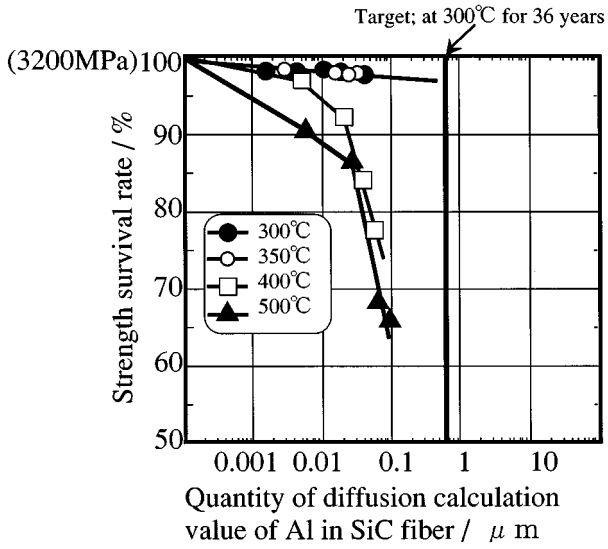

Figure 10 Relation between strength survival rate and quantity of diffusion calculation value of Al.

held together. At 300 °C and above 400 °C the interface reaction phenomenon differed.

3.3. Reaction products from the SiC fiber/Al interface from reaction of solid Al and SiC fiber

Table II lists reaction products obtained from the SiC/Al interface when treated at 300 to 600 °C. No compound was confirmed in elemental wire which was treated at 300 °C for 5000 h, as shown in the high resolution TEM image of Fig. 12. On the other hand, Al_2SiO_5 compound formed in the interface when heat-treated above 400 °C. An Al_2SiO_5 compound layer about 15 nm thick was detected locally at the SiC/Al interface for wire treated at 400 °C for 200 h. Al_2SiO_5 compound formed throughout the whole interface with wires treated at 500 and 600 °C. Fig. 13 shows TEM analysis of Al_2SiO_5 compound from the SiC/Al interface in a treated wire

TABLE II Reaction products from the SiC/Al interface in thermally treated elemental wires as identified by TEM

Treatment conditions of elemental wire	Product
300 °C for 5000 h	None
400 °C for 200 h	Al_2SiO_5/15 nm
500 °C for 200 h	Al_2SiO_5/50 nm
600 °C for 107 h	Al_2SiO_5/200 nm

(500 °C for 200 h). Fig. 14 shows a proposed formation mechanism for Al_2SiO_5 compound at the SiC/Al interface of thermally treated elemental wires. Al_2SiO_5 compound reacted with oxygen in SiC fiber and formed the SiC/Al interface when heat treated above 400 °C. The activation energy of Al_2SiO_5 growth at the SiC/Al interface was about 142 kJ/mol and the constant A was 3.1×10^{-11} from Equations 1 and 2, in the reaction with solid Al and SiC fiber. Fig. 15 relates the quantity of Al_2SiO_5 compound and tensile strength survival rate. As mentioned above, for SiC/Al elemental wire, strength deteriorated directly with the quantity of Al_2SiO_5 compound above 400 °C. But no product was observed at the SiC/Al interface at 300 °C. The rate of strength deterioration of the composite cable held at 300 °C for 36 years would be about 5%, so practical use of SiC/Al composite cable should not be far in the future.

4. Conclusions

To evaluate the reliability of SiC/Al electrical cable, we analyzed the interface reaction between SiC fiber and Al and obtained the following results.

1. No products were detected at the SiC/Al interface of the electrical cable, indicating that the wire manufacture process was satisfactory.
2. An Al_4C_3 compound was detected locally at the SiC/Al interface of the wire which had been thermally treated in molten Al under unfavorable conditions. The activation energy, Q, of Al_4C_3 growth at the SiC/Al interface was about 190 kJ/mol.
3. Al atoms diffused into SiC fiber while Si and C atoms diffused to the Al side during heat treatment within the temperature range of electricity transmission. The activation energy of Al diffusion through the SiC/Al interface to SiC fiber was about 78 kJ/mol.
4. Formation of Al_2SiO_5 compound at the SiC/Al interface strongly depended on the treatment temperature condition. The activation energy of Al_2SiO_5 growth at the SiC/Al interface was about 142 kJ/mol.
5. No products were observed at the SiC/Al interface for wire heat treated at 300 °C. The rate of strength deterioration of the composite cable held at 300 °C for 36 years, was about 5%. Therefore practical use of SiC/Al composite cable should not be far in the future.

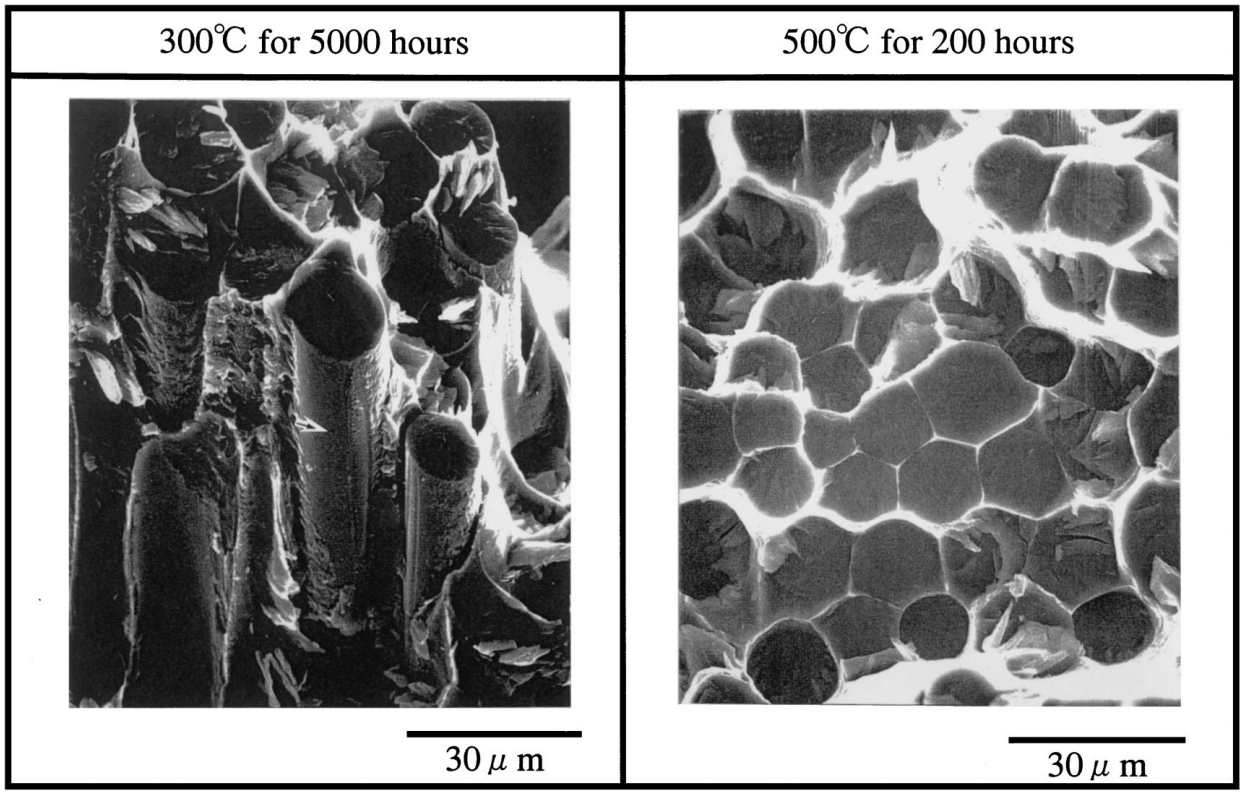

Figure 11 Comparison of fracture surface after tensile test of thermally treated elemental wires.

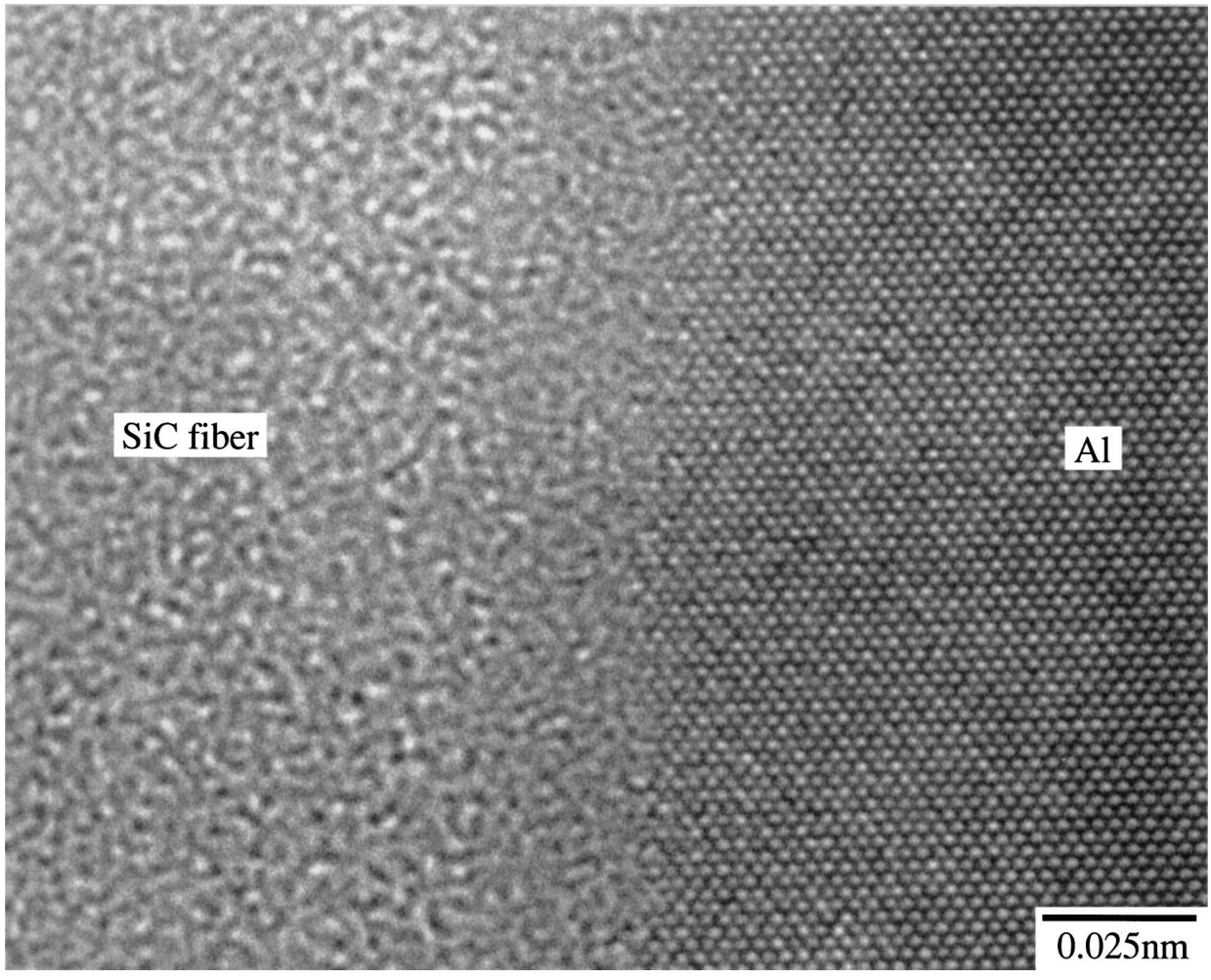

Figure 12 TEM image of SiC/Al interface of thermaly treated elemental wire (300 °C for 500 h).

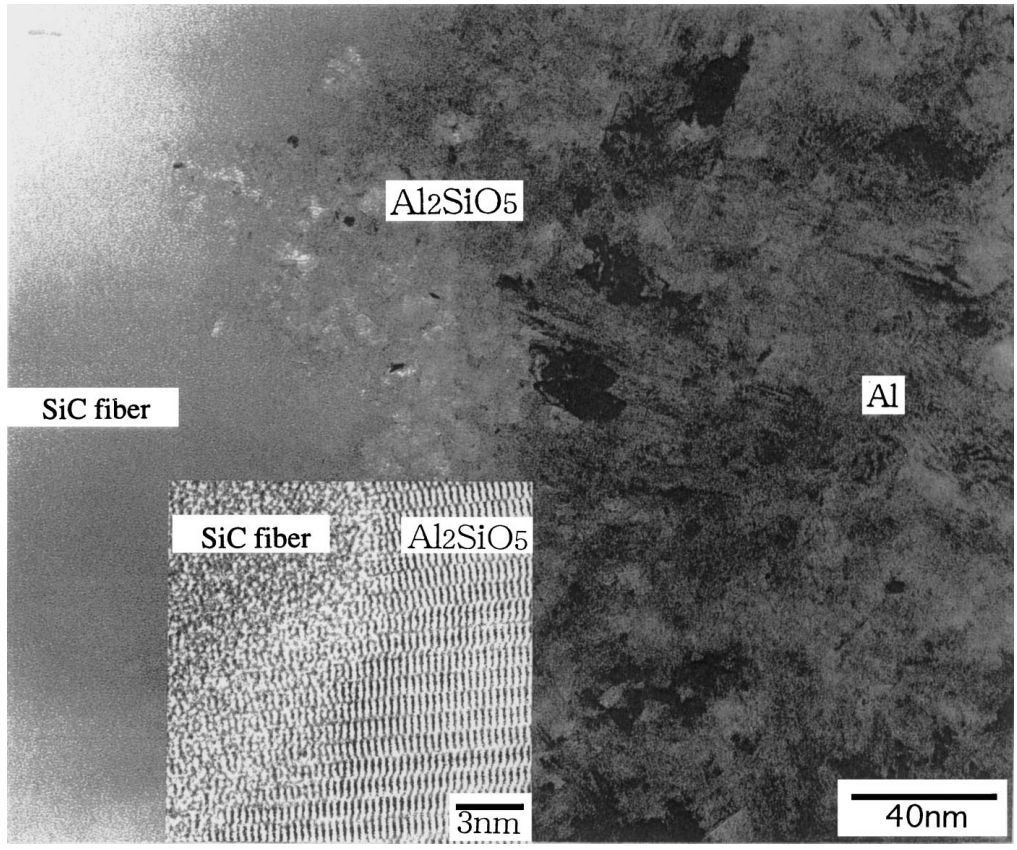

Figure 13 TEM image of Al_2SiO_5 compound from the SiC/Al interface of thermally treated elemental wire (500 °C for 200 h).

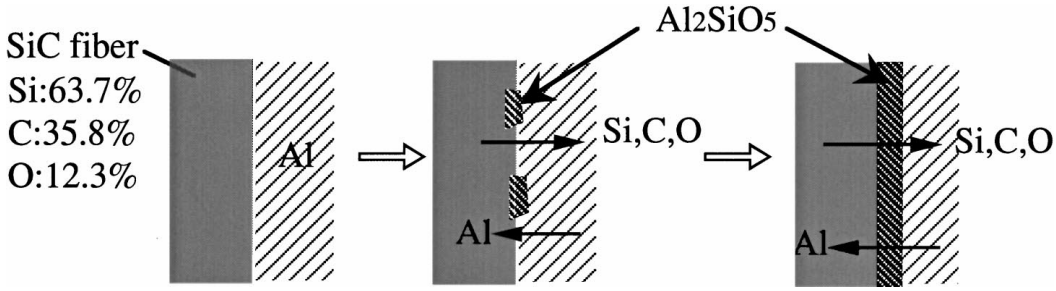

Figure 14 Formation mechanism of Al_2SiO_5 compound of SiC/Al interface of thermal treated elemental wire.

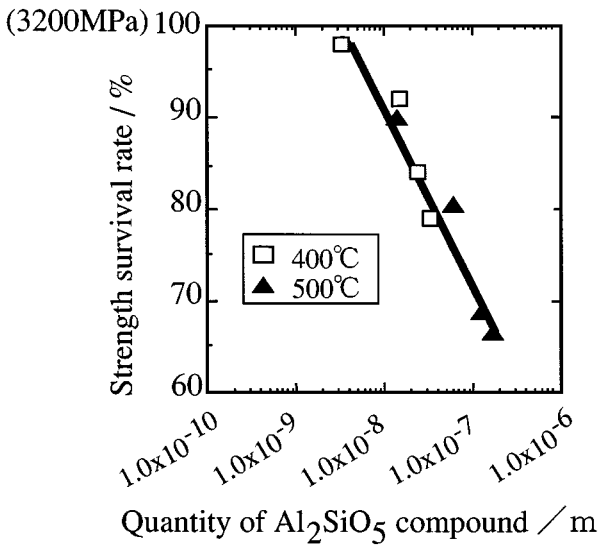

Figure 15 Relationship between tensile strength survival rate of thermally treated elemental wires and the quantity of Al_2SiO_5 compound in the SiC/Al interface.

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