Proceedings of the 42nd International Conference on Advanced Ceramics and Composites

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Valerie Wiesner
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The American Ceramic Society

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A Collection of Papers Presented at the 42nd International Conference on Advanced Ceramics and Composites
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7TH GLOBAL YOUNG INVESTIGATORS FORUM (GYIF)

INTRODUCTION

This collected proceedings consists of 22 papers that were submitted and approved for the proceedings of the 42nd International Conference on Advanced Ceramics and Composites (ICACC), held January 21-26, 2018 in Daytona Beach, Florida. ICACC is the most prominent international meeting in the area of advanced structural, functional, and nanoscopic ceramics, composites, and other emerging ceramic materials and technologies. This prestigious conference has been organized by the Engineering Ceramics Division (ECD) of The American Ceramic Society (ACerS) since 1977.

The 42nd ICACC hosted over 950 attendees from 37 countries. The topics ranged from ceramic nanomaterials to structural reliability of ceramic components, which demonstrated the linkage between materials science developments at the atomic level and macro level structural applications. Papers addressed material, model, and component development and investigated the interrelations between the processing, properties, and microstructure of ceramic materials. This year's conference included a special symposium in honor of Dr. Mrityunjay Singh titled: Advancing Frontiers of Ceramics for Sustainable Societal Development.

The 2018 conference was organized into the following 17 symposia, 3 Focused Sessions and 2 Special Symposia:

Symposium 1	Mechanical Behavior and Performance of Ceramics and Composites
Symposium 2	Advanced Ceramic Coatings for Structural, Environmental, and Functional Applications
Symposium 3	15th International Symposium on Solid Oxide Fuel Cells (SOFC): Materials, Science, and Technology
Symposium 4	Armor Ceramics: Challenges and New Developments
Symposium 5	Next Generation Bioceramics and Biocomposites
Symposium 6	Advanced Materials and Technologies for Direct Thermal Energy Conversion and Rechargeable Energy Storage
Symposium 7	12th International Symposium on Nanostructured Materials: Functional Nanomaterials and Thin Films for Sustainable Energy Harvesting, Environmental and Health Applications

Symposium 8	12th International Symposium on Advanced Processing & Manufacturing Technologies for Structural & Multifunctional Materials and Systems
Symposium 9	Porous Ceramics: Novel Developments and Applications
Symposium 10	Virtual Materials (Computational) Design and Ceramic Genome
Symposium 11	Advanced Materials and Innovative Processing ideas for the Production Root Technology
Symposium 12	Materials for Extreme Environments: Ultrahigh Temperature Ceramics (UHTCs) and Nano-laminated Ternary Carbides and Nitrides (MAX Phases)
Symposium 13	Advanced Ceramics and Composites for Nuclear Fission and Fusion Energy
Symposium 14	Crystalline Materials for Electrical, Optical and Medical Applications
Symposium 15	Additive Manufacturing and 3D Printing Technologies
Symposium 16	Geopolymers, Chemically Bonded Ceramics, Eco-friendly and Sustainable Materials
Symposium 17	Advanced Ceramic Materials and Processing for Photonics and Energy
Focused Session 1	Bio-Inspired Processing of Advanced Materials
Focused Session 2	Tomography and Microscopy Based Modeling of Ceramics
Focused Session 3	Chemical Processing of Functional Materials
Special Symposia	7th Global Young Investigators Forum (GYIF)
Special Symposia	Advancing Frontiers of Ceramics for Sustainable Societal Development – International symposium in honor of Dr. Mrityunjay Singh

The proceedings papers from this conference are published in the below two issues of the 2018 CESP; Volume 39, Issues 2-3, as listed below.

- CESP Volume 39, Issue 2: Includes 25 papers from Symposia 1, 2, 3, 4, 6, and 8
- CESP Volume 39, Issue 3: Includes 22 papers from Symposia 9, 10, 12, 13, 14, 15, 16, FS1, The 7th GYIF, and the Dr. Mrityunjay Singh Honorary Symposium

The organization of the Daytona Beach meeting and the publication of these proceedings were possible thanks to the professional staff of ACerS and the tireless dedication of many ECD members. We would especially like to express our sincere thanks to the symposia organizers, session chairs, presenters and conference attendees, for their efforts and enthusiastic participation in the vibrant and cutting-edge conference.

Valerie Wiesner, NASA Glenn Research Center, USA

Manabu Fukushima, National Institute of Advanced Industrial Science and Technology (AIST), Japan

Volume Editors
August 2018

Advancing Frontiers of Ceramics for Sustainable Societal Development: International Symposium in Honor of Dr. Mrityunjay Singh

PROGRESS IN POLYMER-DERIVED SiC-BASED FIBERS: IMPROVEMENT OF SURFACE ROUGHNESS

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ABSTRACT

Polymer-derived SiC-polycrystalline fiber (Tyranno SA) shows excellent heat-resistance up to 2000ºC, and relatively high strength. However, to extend the application field, increase in the fiber's strength is eagerly required. Up to now, through our research, the relationship between the strength and residual defects of the fiber, which were formed during the heat-treatment processes (degradation of raw fiber and sintering), has been clarified. In this paper, we addressed the relationship between the heat-treatment condition and the surface roughness of the obtained SiC-polycrystalline fiber, using three different raw fibers (Elementary ratio: $Si_1Al_{0.01}C_{1.5}O_{0.4\sim0.5}$) and three types of carbon vessel (Open system, Partially closed system, and Closed system). With increase in the oxygen content in the raw fiber, the degradation during the heat-treatment process easily proceeded. This led to nearly stoichiometric composition of SiC crystal. And, higher oxygen content in the raw fiber and the closed system led to relatively high partial-pressure of SiO at the initial stage of the degradation process. In this case, the degradation reactions ($SiO+2C=SiC+CO$ and $SiO_2+3C=SiC+2CO$) in the inside of each filament became faster, and then the CO partial pressure at the surface region of each filament was found to be increased. In consequence, according to Le Chatelier's principle, the surface degradation reaction and grain growth of formed SiC crystals would be considered to become slower. That is to say, using the raw fiber with higher oxygen content and closed system (highest CO content in the reactor), much smoother surface of the SiC-polycrystalline fiber could be achieved.

INTRODUCTION

Since the first precursor ceramics using polycarbosilane was developed, lots of polymer-derived SiC-base fibers have been developed. Through these developments, the heat-resistances of the SiC-based fibers were remarkably increased from 1200ºC to 2000ºC. Of these fibers, SiC-polycrystalline fibers (Tyranno SA, Hi-Nicalon Type S, and Sylramic) show the highest heat-resistance up to 2000ºC, and then have been actively evaluated for aerospace applications as SiC/SiC composites [1-5]. However, to extend the application field, increase in the fiber's strength is eagerly required. Up to now, through our research, the relationship between the strength and the residual defects contained in the fiber, which were formed during the production processes, has been clarified [6-9]. In these researches, we have proposed several new methods for reducing the residual defects, and demonstrated them using the conversion process from amorphous Si-Al-C-O fiber to SiC-polycrystalline fiber (Tyranno SA). Tyranno SA is produced by heat-treatment processes of amorphous Si-Al-C-O fiber which is synthesized from polyaluminocarbosilane. During the heat-treatment processes, a degradation of the Si-Al-C-O fiber and the subsequent sintering of the degraded fiber proceed as well, accompanied by a release of CO gas and compositional changes, to finally obtain the dense structure. Since these structural changes proceed in each filament, a strict control should be needed to minimize residual defects. As mentioned above, to reduce the residual defects, we proposed new conversion processes and demonstrated them. And then, using these new processes, the inside defects were remarkably reduced. In this case, by controlling the advantageous degradation-reaction and preventing the disappearance of gaseous SiO from each filament, residual carbon (one of residual defects) was remarkably reduced along with prevention of abnormal surface grain growth. In consequence, the surface roughness was relatively improved compared with the obtained surface using previous processes. However, the surface roughness was very sensitive depending on the atmospheric condition during the degradation process, and then obtaining much smoother surface was a relatively difficult problem. Smoother surface of the fiber is very important for obtaining good fibrous fracture behavior of ceramic matrix composites (CMCs) [10]. Accordingly, it should be important to clarify the relationship between the process condition and the surface roughness of the obtained fiber.

Here, we describe the formation mechanism of the surface structure and the relationship between the process condition and the surface roughness of the obtained SiC-polycrystalline fiber.

EXPERIMENTAL

The SiC-polycrystalline fiber (Tyranno SA) was synthesized by heat-treatment up to 1900°C of an amorphous Si-Al-C-O fiber, which is synthesized from polyaluminocarbosilane. The polyaluminocarbosilane was synthesized by a reaction of polycarbosilane with tetra-butoxyaluminum at 300°C in nitrogen atmosphere. A spun fiber was obtained by melt-spinning of the polyaluminocarbosilane, and then the spun fiber was cured at around 200°C in air. The cured fiber was fired at around 1300°C in nitrogen atmosphere to obtain the amorphous Si-Al-C-O fiber. The Si-Al-C-O fiber was composed of SiC fine crystals, oxide phases (estimated forms: SiO_2, AlO_x), and excess carbons. By the way, as mentioned above, since in this synthesis we used polyaluminocarbosilane which was synthesized by the reaction of polycarbosilane and tetra-butoxyaluminum, we presumed that the aluminum existed as some oxide forms in the Si-Al-C-O fiber. In the next step, the amorphous Si-Al-C-O fiber was heat-treated up to around 1500°C in argon gas atmosphere. During the heat-treatment, by the existence of the oxide phase and excess carbon in the fiber, the amorphous Si-Al-C-O fiber was degraded accompanied by a release of CO gas to obtain a porous degraded fiber. The porous degraded fiber was composed of a nearly stoichiometric SiC composition containing small amount of aluminum. In this case, since a part of the aluminum contained in the amorphous Si-Al-C-O fiber was found to vaporize as some oxide materials during the heat-treatment process, consequently a very small amount of aluminum (less than 1wt%) was contained in the degraded fiber. By the existence of the small amount of aluminum, at the next step, an effective sintering proceeded in each degraded filament composed of the nearly stoichiometric SiC crystals during further heat-treatment up to 2000°C in argon atmosphere. The production scheme of the polymer-derived SiC polycrystalline fiber using the Si-Al-C-O fiber as the raw fiber is shown in Fig.1. As mentioned in our previous papers, degradation reactions of the amorphous Si-Al-C-O fiber enclosed in red frame in this figure (Fig.1) strongly affects the final fine-structure (Cross-section and Surface) of the SiC-polycrystalline fiber. Especially, reaction condition concerning CO gas content during the degradation process is most important [6, 9]. Accordingly, in this research we adopted three types of reaction vessel (Open system, Partially closed system, and Closed system) made of carbon shown in Fig.2. For the heat-treatment (degradation reaction and sintering) of the Si-Al-C-O fiber, we used “Super High Temperature Inert Gas Furnace (NEWTONIAN Pascal-40, Produced by NAGANO)” under argon gas flow (1 L/min). The size of the heating zone (made of graphite and C/C composites) is 35 mm in diameter and 40 mm in height. Several types of raw fibers (about 10 mg) (Elementary ratio: $Si_1Al_{0.01}C_{1.5}O_{0.4\sim0.5}$) were used and located in each vessel. The programing rate and the maximum temperature were 400°C/min and 2000°C, respectively.

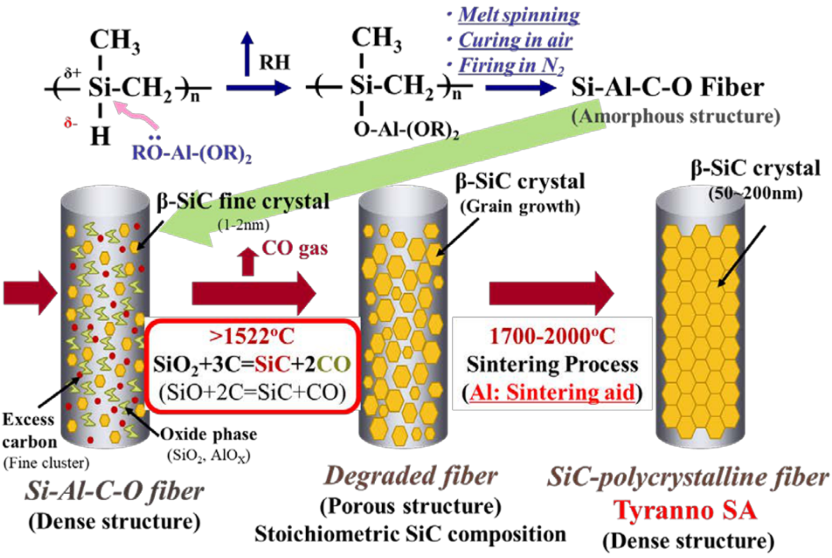

Fig.1 The production scheme of the SiC-polycrystalline fiber using a raw Si-Al-C-O fiber

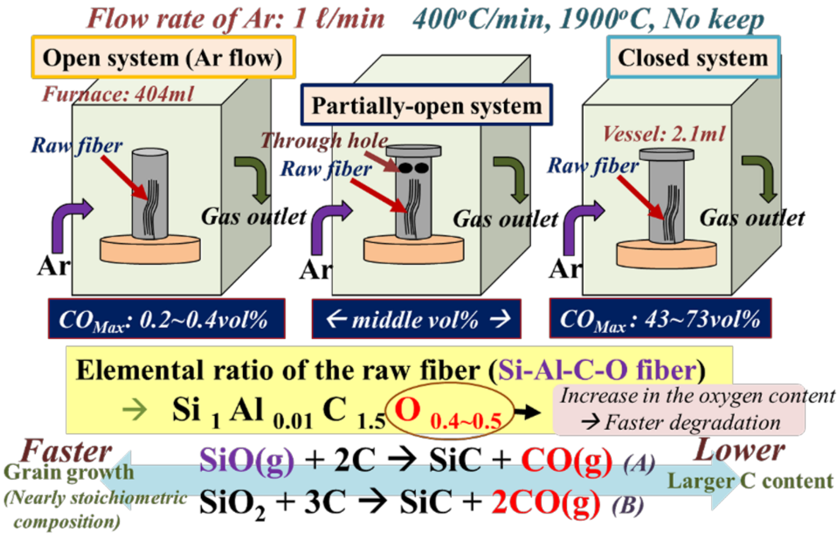

Fig.2 Experimental condition for research on fiber’s surface roughness

The surfaces and cross sections of the obtained fibers were observed using a field emission scanning electron microscope (FE-SEM), model JSM-700F (JEOL, Ltd.). Parts of surface region and inside of the several samples were sharpened by an etching machine using focused ion beam (FIB), and then the fine structures were observed by the transmission electron microscope (TEM), model JEM-2100F (JEOL, Ltd.). Surface roughness was observed using Atomic Force Microscope (AFM), model AFM 5000II (Hitachi, Ltd.).

RESULTS AND DISCUSSION

Morphological changes during the degradation process

As mentioned before, for obtaining the SiC-polycrystalline fiber, at the first step, the amorphous Si-Al-C-O fiber was heat-treated up to 1500°C in Ar gas atmosphere. During the heat-treatment process, by the existence of the oxide phase and excess carbon in the fiber, the amorphous Si-Al-C-O fiber was degraded accompanied by a release of CO gas to obtain a porous degraded fiber. This degradation of the Si-Al-C-O fiber proceeds mainly by the following two types of reactions.

(1) $SiO_2 + 3C = SiC + 2CO$ ($⊿G < 0$ over 1522°C)
(2) $SiO + 2C = SiC + CO$ ($⊿G < 0$ at all temperatures range)

The porous degraded fiber was composed of a nearly stoichiometric SiC composition containing small amount of aluminum (less than 1 wt%). By the existence of the small amount of aluminum, at the next step, an effective sintering proceeded in each degraded filament composed of the nearly stoichiometric SiC crystals during further heat-treatment up to 2000°C in Ar gas atmosphere. And then, the dense SiC-polycrystalline fiber was obtained. The morphological changes of each filament during the further heat-treatment are shown in Fig.3.

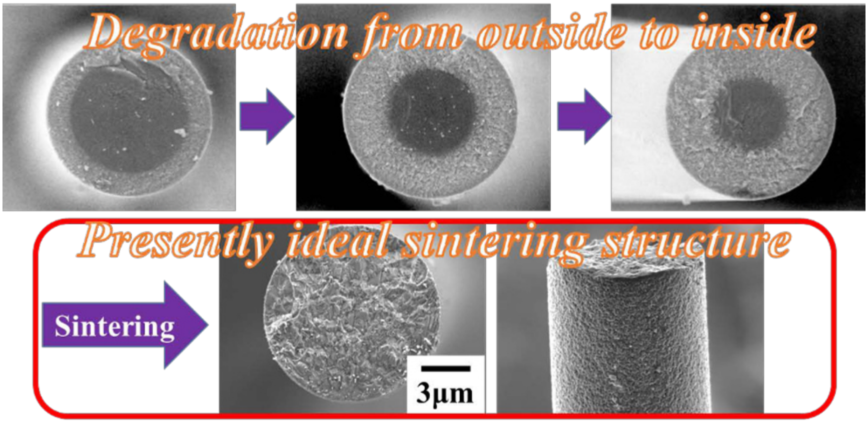

Fig.3 Morphological changes during the degradation and sintering processes

As can be seen from this figure (Fig.3), the degradation proceeded from outside to inside of the amorphous Si-Al-C-O fiber. And, regarding the SiC-crystalline size of the obtained sintered fiber, the surface SiC-crystals were relatively small compared with the inside crystals. This phenomenon was considered to be caused by the atmospheric condition (Especially: CO gas content) during the degradation process. That is to say, the degradation reactions (SiO_2 + 3C = SiC + 2CO and SiO + 2C = SiC + CO) are strongly dominated by the CO gas content in the reactor. According to Le Chatelier's principle, the higher the CO content becomes, the slower the reaction becomes. Anyway, the abovementioned degradation proceeds in the inside of each filament accompanied by a release of CO gas. So, the inside of each filament is saturated by the formed CO gas, and the surplus CO gas is ejected from the surface region to the outside. Furthermore, on the surface region of each filament, some boundary layer composed of CO gas must be formed. By these changes, in consequence, some CO gas distribution would be formed from the inside to the surface region of each filament. Accordingly, the degradation in the inside of each Si-Al-C-O filament was considered to proceed as shown in Fig.4.

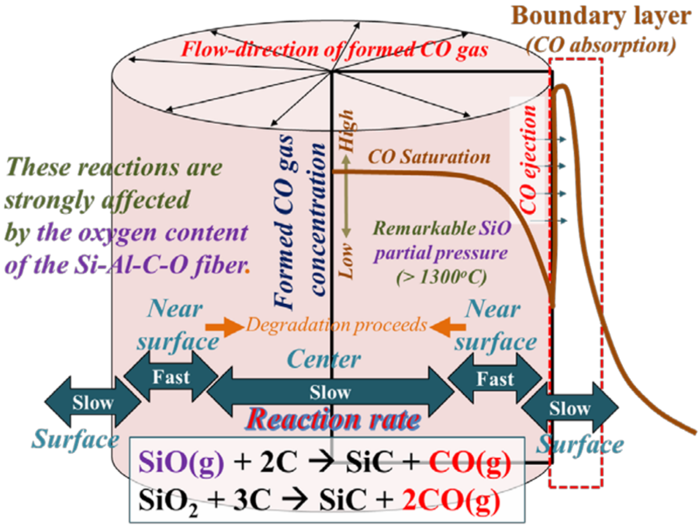

Fig.4 General degradation in the inside of each Si-Al-C-O filament

As can be seen from this figure (Fig.4), the first degradation reaction smoothly proceeds near the surface by relatively low CO content compared with that in the inside. So, the degradation proceeds from outside to inside. By the way, it is considered that the CO content at the surface region is relatively high compared with that in the inside of each filament. Accordingly, it is estimated that at the surface region, the degradation slowly proceeds to consequently prevent the SiC-crystalline grain growth. In the next section, we will address the relationship between the surface degradation reaction and the surface roughness of the obtained SiC-polycrystalline fiber.

Change in the surface roughness of the SiC-polycrystalline fiber

As mentioned before, both the degradation reaction and grain growth of the formed SiC crystals are strongly affected by the CO content in the reactor during the degradation reaction. As shown in Fig.2, we used three different types of vessel (Open system, Partially-open system, and Closed system) and several types of raw fiber (Elementary ratio: $Si_1Al_{0.01}C_{1.5}O_{0.4\sim0.5}$) for changing actual CO content in the reactor. Under our reaction condition, we calculated that the maximum CO content in the reactor was 73vol% when we used both the closed system and the raw fiber with highest oxygen content ($Si_1Al_{0.01}C_{1.5}O_{0.5}$), whereas the minimum CO content was 0.2vol% when we used both the open system and the other raw fiber with lowest oxygen content ($Si_1Al_{0.01}C_{1.5}O_{0.4}$). The surface structures (SEM images) of the obtained SiC-polycrystalline fibers are shown in Fig.5. These fibers were obtained by heat-treatment of the different types of

Si-Al-C-O fiber at 1900^{o}C in Ar using the three different types of vessels. As can be seen from this figure (Fig.5), higher oxygen content of the raw fiber and closed system led to the much smoother surface.

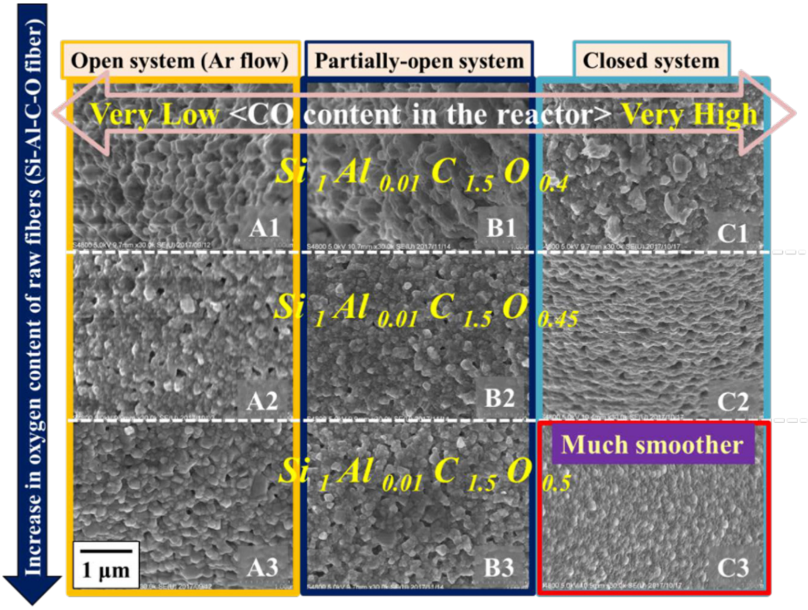

Fig.5 Changes in the surface structures (SEM images) using different types of raw fiber and vessels

These results are closely related to the CO content in the reactor during the degradation reaction. The much smoother surface was obtained using the raw fiber with highest oxygen content ($Si_1Al_{0.01}C_{1.5}O_{0.5}$) and the closed system. As can be seen from these results, the surface roughness is effectively controllable by changing the degradation conditions (Especially; CO content in the reactor). The most important factors for change in the CO content in the reactor are (1) Oxygen content of the raw Si-Al-C-O fiber, and (2) Reactor system (Open system, Partially-open system, and Closed system). By change in the combination of these factors, different degrees of surface roughness could be obtained as can be seen from Fig.5. Some phenomena caused by increase in the oxygen content of the raw Si-Al-C-O fiber are shown as follows along with some differences caused by change in the system (Open system, Partially-open system, and Closed system). As the oxygen content in the law fiber increases, the degradation reaction easily occurs. It leads to nearly stoichiometric composition of the degraded fiber. In this case, the partial pressure of CO gas at the surface region of the fiber becomes higher compared with that of the inside. This leads to lower reaction rate at the surface region of the fiber. Consequently, increase in the oxygen content of the raw Si-Al-C-O fiber led to decrease in the SiC crystalline size at the surface region. This means getting smooth surface. That is to say, higher oxygen content of the raw Si-Al-C-O fiber and closed system cause relatively higher CO content in the reactor during the degradation reaction, and then the consequent higher CO partial pressure at the surface region reduces the degradation reaction, that results in getting smooth surface.

Changes in actual surface roughness of the obtained SiC-polycrystalline fibers, which were synthesized by heat-treatment at 1900ºC in argon gas atmosphere using different raw fibers and different vessels, are shown in Fig.6. As can be seen from this figure (Fig.6), the surface roughness could be controlled by change in both oxygen content of the raw Si-Al-C-O fiber and the reaction vessel. In this case, for obtaining A1, B1, C1 in Fig.6, the raw fiber composed of $Si_1Al_{0.01}C_{1.5}O_{0.4}$ was used for the synthesis. And, for obtaining A2, B2, C2 and for obtaining A3, B3, C3, the raw fibers composed of $Si_1Al_{0.01}C_{1.5}O_{0.45}$ and $Si_1Al_{0.01}C_{1.5}O_{0.5}$ were used, respectively. As can be seen from Fig.6, we could control the surface roughness from 67.99nm (maximum value) to 9.05nm (minimum value).

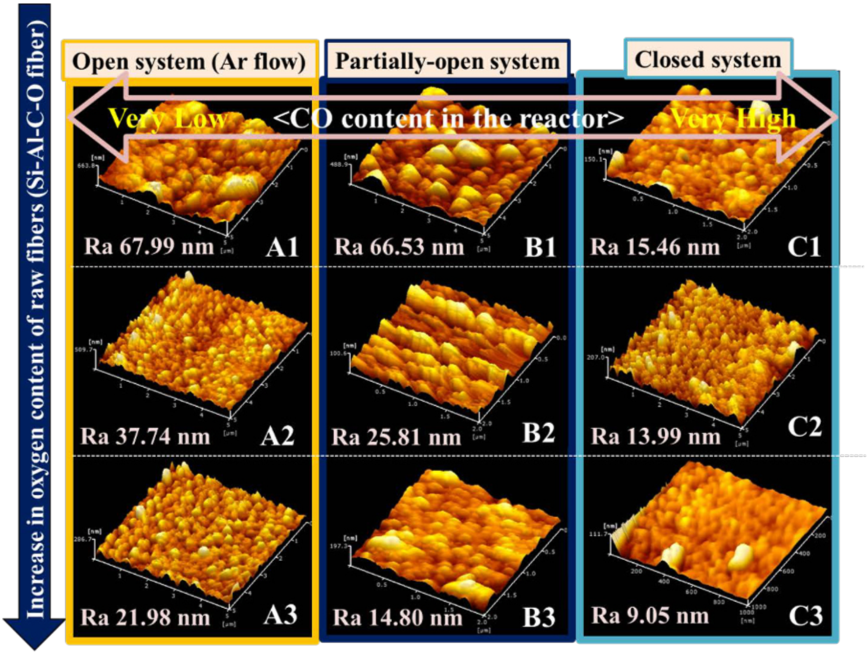

Fig.6 Change in the surface roughness of the SiC-polycrystalline fiber synthesized from different raw Si-Al-C-O fiber with different oxygen content at 1900ºC in argon atmosphere. For obtaining A1, B1, C1, the raw fiber composed of $Si_1Al_{0.01}C_{1.5}O_{0.4}$ was used. For obtaining A2, B2, C2 and for obtaining A3, B3, C3, the raw fibers composed of

$Si_1Al_{0.01}C_{1.5}O_{0.45}$ and $Si_1Al_{0.01}C_{1.5}O_{0.5}$ were used, respectively.

In this research, we used degradation process of the amorphous raw fiber (Si-Al-C-O fiber) accompanied by a release of CO gas and the subsequent sintering process, and showed the controllable SiC crystalline size constructing the obtained SiC-polycrystalline fiber by changing the CO gas partial pressure in the reactor. In consequence, we could control the surface roughness of the SiC-polycrystalline fiber using CO gas released from the raw fiber. However, this means that an intentional change in CO gas partial pressure in the reaction vessel can lead to preferable crystalline structure.

Fig.7 shows an improvement result regarding the surface roughness by achieving the adjustment of the CO gas partial pressure in the reactor during the degradation reaction. In this case, using Le Chatelier's principle, we accelerated the following reaction ($SiO_2+3C=SiC+2CO$) to consequently increase the CO gas partial pressure at the surface region of the fiber at initial degradation process. Regarding this degradation condition, we reported the detailed content in the previous paper [6].

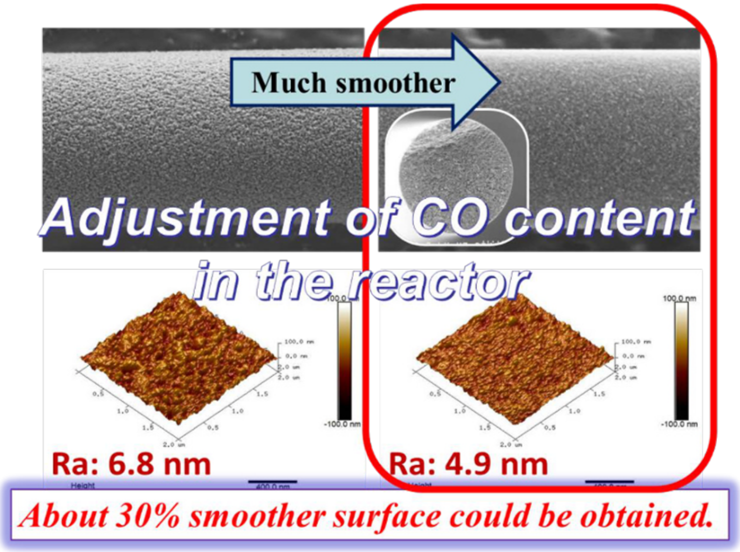

Fig.7 Improvement of the surface roughness by adjustment of CO content in the reactor

CONCLUSIONS

We clarified the relationship between the heat-treatment condition and the surface roughness of the obtained SiC-polycrystalline fiber, using three different raw fibers (Elementary ratio: $Si_1Al_{0.01}C_{1.5}O_{0.4\sim0.5}$) and three types of carbon vessel (Open system, Partially closed system, and Closed system). With increase in the oxygen content in the raw fiber, the degradation during the heat-treatment process easily proceeded accompanied by a release of relatively high concentration of CO gas. When we used the raw fiber composed of $Si_1Al_{0.01}C_{1.5}O_{0.5}$ and closed system, much smoother surface of the obtained SiC-polycrystalline fiber could be achieved. In this case, the degradation reactions (SiO+2C=SiC+CO and SiO_2+3C=SiC+2CO) at the first stage in the inside of each filament became faster, and then the CO partial pressure at the surface region of each filament was found to be increased. In consequence, according to Le Chatelier's principle, the surface degradation reaction and grain growth of formed SiC crystals would be considered to become slower.

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THE INFLUENCE OF CASTING-CALENDERING PROCESS ON THE MICROSTRUCTURE OF PURE Al_2O_3 CERAMIC SUBSTRATE

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ABSTRACT

The forming process of ceramic green tape has heavy influence on the performance of ceramic substrate. Aimed on the application for electric package, a new technology of casting-calendering process for preparing pure Al_2O_3 substrate was proposed in this paper. Micro and nano multi-scale mixed ceramic powders were used as raw material. The ceramic green tape with micro-nano hierarchical structure was prepared by casting-calendering process. Finally, the pure Al_2O_3 ceramic substrates were sintered under 1300 to 1600℃, and the microstructure of the ceramic substrates prepared by casting-calendering process and casting process was compared. Experimental results showed that the pure Al_2O_3 ceramic substrate sintered at 1500℃ with no additives exhibits good densification degree of 92.5%. The thermal conductivity of the pure Al_2O_3 ceramic substrate prepared by casting-calendering process and sintered at 1500°C was 17.5W/(m·K), which was much higher than that by tape casting process under the same sintering condition.

INTRODUCTION

Due to the advantages of high thermal conductivity, good insulation, compatible thermal expansion coefficient with the chip, ceramic substrate has been widely used in electronic components, integrated circuits and electronic packaging fields. In recent years, with the rapid development of electronic packaging technology, packaging devices is becoming smaller and smaller and the packing density is becoming higher and higher. Thus, the heat density in the integrated circuit device is increasing. If the heat in the electronic device couldn't be dissipated quickly, it will not only reduce the reliability and performance of electronic devices, but also cause operation failure. According to the statistics[1], more than half of the failure of electronic device was caused by heat. Overheating has become the bottleneck that restricts the development of electronic device technology. Improving the thermal performance of the substrate has become the focus of the electronic packaging field[2].

Traditionally, the electronic packaging substrate mainly includes plastic substrate, metal substrate and ceramic substrate. Plastic substrate is the most commonly used substrate at present, but the thermal conductivity and reliability of which is too low. Metal substrate has high thermal conductivity, but its CET is several times higher than the chip, which will produce high thermal stress between the metal substrate and the chip during operation. As a result, it is easy to cause the chip peeled off or fracture failure[3]. Ceramic substrate has high thermal conductivity, compatible CET with chips and good insulation, which is a preeminent packaging substrate material with excellent comprehensive performance. However, ceramic materials are usually difficult to melt because they are compounds that have a combination of ionic and covalent bonds, thus the sintering temperature is usually very high. Besides the production process is complex, and the production equipment is expensive[4]. At present, the cost of traditional high performance ceramic substrate by hot press process is very high. The thermal conductivity of LTCC (Low temperature co-fired ceramics) substrate decreased sharply due to the great amount