EFFECT OF SiC NANO SIZING ON SELF-CRACK-HEALING DURING SERVICE

Wataru Nakao*, Yasuyuki Tsutagawa**, Koji Takahashi* and Kotoji Ando*

* Faculty of Engineering, Yokohama National University

**Graduated school, Yokohama National University

Phone: +81-45-339-4026
Fax: +81-45-339-4024
wnakao@ynu.ac.jp
http://www.bsk.ynu.ac.jp/~andolab/

In the present study, the efficient temperature region for in-situ self-crack-healing was improved by SiC nano-sizing. The present study adapted the following self assemble reaction to synthesis method alumina composite containing nano-size silicon carbide particles $3(3\text{Al}_2\text{O}_3\cdot2\text{SiO}_2) + 8\text{Al} + 6\text{C} = 13\text{Al}_2\text{O}_3 + 6\text{SiC}$. The formed SiC particles have a particle size from 10 nm to 30 nm and exist in alumina grain and on alumina grain boundary. It was found that the new alumina/ SiC nano composite can heal the pre-crack of 0.1 mm at 1223 K for 10 h. The temperature is 250 K smaller than that of the previous alumina/ SiC composite (SiC particle size = 270 nm). Furthermore, the crack-healed specimen has no large strength decrease up to 1573 K. Therefore, new alumina/ silicon carbide nano-composite was found to exhibit the in-situ self-crack-healing efficient temperature region of 300 K.

Keywords: Self-Healing, Structural Ceramics, Crack Healing, Nanometer-sized material

1 Introduction

For energy plants, the elastic components, such as spring, to be able to operate at high temperature are absolutely required. Only ceramics are available to this application. Alternatively, huge structural reliability is also required for the component of energy plants. It is well known that ceramics are brittle material and exhibit low structural reliability. This feature is caused by a large strength decrease due to cracking. If ceramics can heal cracks, which are introduced by crush, thermal shock and fatigue during service, simultaneously with cracking, we can use ceramic to energy plants.

Self-crack-healing behavior of ceramic was firstly reported in 1966 [1]. Then, Lange and Gupta [2] reported the strengthening ZnO and MgO by heat-treatment, and used the term of “crack-healing” first in 1970. Now, we can find more than 200 reports on the strengthening effects by heat-treatment for cracked ceramics. Most of all reports are to investigate resintering [1, 2], i.e. diffusive crack-healing process. The phenomenon requires the too high crack-healing temperature to generate the grain growth, i.e. it is almost same temperature as sintering. Therefore, the crack-healing cannot occur during service.
The present authors succeeded to endow alumina [3, 4] and mullite [5- 7] with self-crack-healing ability by containing SiC of appropriate volume fraction. The crack-healing is driven by the oxidation of the contained SiC particles. The crack-healing would occur simultaneously with cracking. When these ceramics are worked in air at high temperature, SiC that exist on crack walls would start reacting with oxygen at the same time as cracking. Since the reaction includes 80.1 % volume increase in the condense phases, the space between the crack walls is completely filled with the formed oxide. Furthermore, the oxidation includes a large exothermic heat. The reaction heat makes the formed oxide and the base material to react or to once melt. This phenomenon leads to strong bonding between the reaction products and crack walls.

Unfortunately, the self-crack-healing driven by the oxidation of the commercial SiC particles can occur in only limited temperature region. Actually alumina containing 15 vol% commercial SiC particles1, which have means particle size of 270 nm, can heal the indented semi-elliptical crack having depth of 0.05 mm by heat-treatment at 1573 K for 1 h in air, but it exhibits creep deformation above 1573 K. Therefore, to overcome the object, the present authors tried by nano-sizing the contained SiC. Nano-sizing SiC makes the surface of SiC to be active. Thereby, the reaction would progress actively at lower temperature. Furthermore, existence of SiC particles in the alumina matrix grains due to nano-sizing caused large resistance to the glide deformations of alumina grain and alumina grain boundary.

In the present study, the following self-propagating high-temperature synthesis was adapted to produce alumina/ SiC nano-composite,

\[ 3(3Al_2O_3 \cdot 2SiO_2) + 8Al + 6C = 13Al_2O_3 + 6SiC \] (1)

By this method, Zhang et al. [8] succeeded fabricating alumina/ SiC nano-composite in which SiC particle has a means particle size of 50 nm. Based on this knowledge, alumina/ SiC nano-composite was prepared and the crack-healing behavior of the formed alumina/ SiC nano-composite was investigated.

## 2 Experimental

### 2.1 Sample preparation

In the present study, the self-propagating high-temperature synthesis expressed as Reaction (1) was adapted to produce alumina/ SiC nano-composite. The synthesized composite has the SiC contents of 18.4 vol.%. The alumina/ SiC composite is abbreviated as AS18NP in this paper.

The used mullite (KM101, Kyoritsu Materials Ltd, Japan), aluminum (600F, Minaruko Ltd., Japan) and carbon powders (#4000B, Mitsubishi Chemical Ltd., Japan) have means of the particles size of 780 nm, 5400 nm and 20 nm, respectively.

Carbon powder was dispersed well in ethanol using ultra sonic vibration, prior to the mixing. Mullite and aluminum powders were added to carbon ultra-fine powder dispersed ethanol and the powders were mixed well via a Teflon pot and alumina balls for 24 h.
To finish reaction (1), the dried mixed powder was heated at 1673 K for 5 h in a carbon crucible and Ar atmosphere. After that, the formed alumina/ SiC powder was sintered via hot-press at 2073 K for 1 h in Ar. The sintered plate was cut into rectangular test specimens (Width = 4 mm, Length = 22 mm, Height = 3 mm).

2.2 Experimental procedure

A semi-elliptical surface crack was made at center of the specimen surface by Vickers indenter. The formed crack has surface length of 0.1 mm and aspect ratio of 0.9. The cracked specimens were crack-healed at temperatures from 1073 K to 1573 K for 1 h - 100 h. These specimens were called as “crack-healed specimen” in this paper.

All fracture tests were conducted on a three-point bending. The span was adapted to be 16 mm in order that the fracture is as possible as to occur from the crack-healed pre-crack. The test temperatures were room temperature and high temperatures from 1073 K to 1673 K. Microstructure and fracture initiation were investigated using a scanning electron microscopy (SEM).

Same experiments were conducted on the alumina containing 15 vol% commercial SiC particles composite, abbreviated as AS15P.

3 Results and discussions

3.1 Microstructure

Figure 1 shows the microstructure of the formed alumina/ SiC nano-composite. The grains size of the alumina matrix ranges from 1 μm to 10 μm. The formed nano-sized SiC particles were mainly entrapped inside alumina grains, as pointed by white arrows in Fig. 1. Alternatively, the submicron-sized SiC particles are located at the grain boundaries of alumina grains, as pointed by black arrows in Fig. 1. The grains size of the intra SiC particles ranges from 10 nm to 30 nm, corresponding to one tenth that of commercial SiC particles. The nano-sized SiC particles are beneficial for improving crack-healing ability.

Figure 1: SEM image of the fracture surface of the formed alumina/ SiC composite (AS18NP)
3.2 Crack-healing behavior

Figure 2 shows the bending strength of the crack-healed AS18NP as a function of crack-healing temperature. The crack-healing time is 10 h. Moreover, the left side column shows the bending strength of as-cracked specimens and the center-lined symbols indicate the specimens fractured from the pre-crack crack-healed. As crack-healing temperature increases, the bending strengths increase. As the crack-healing temperature is more than 1223 K, the strength recovery is found to be saturated. Moreover, half of all specimens exhibiting the saturated strength recovery fractured from the point outside the pre-crack crack-healed. This implies that crack-healing makes the pre-crack to erase completely and the strength is confirmed to be the strength of the complete surface smooth specimen. Therefore, the complete strength recovery is attained by heat-treatment at 1223 K for 10 h. In similarly, the complete strength recovery is attained by heat-treatment at 1373 K for 1 h.

For comparison, the data on AS15P is also shown as the open triangle in Fig. 2. The cracked strength and crack-healed strength of AS15P were found to be approximately equal to those of AS18NP. However, the complete strength recovery in AS15P was attained by crack-healing at the temperature of more than 1473 K for 10h. It was, therefore, confirmed that nano-sized SiC formed by SHS causes 250 K decrease to the temperature that self-crack-healing become active.

![Figure 2: Bending strength of the AS18NP crack-healed for 10 h at several temperatures, with that of the crack-healed AS15P, in which the center-lined symbols indicate the specimens fractured from the pre-crack crack-healed.](image)

3.3 High temperature strength

Figure 3 shows the temperature dependence of the bending strength of AS18NP crack-healed at 1473 K for 1 h in air. The strength decreases gradually as temperature increases. The means of the bending strengths are 850 MPa at RT, 650 MPa at 1273 K, 600 MPa at 1473 K, 500 MPa at 1573 K and 300 MPa at 1673 K. Furthermore, there are no outstanding plastic deformation behaviors from the all stress-strain curve. Therefore, the limit temperature for the bending strength of AS18NP would be at the least 1573 K.
The similar behavior of high temperature was occurred in the crack-healed AS15P, which is shown as the open triangle in Fig. 3. Therefore, it was confirmed that the nano-sized SiC formed by SHS cannot give further improvement in the limit temperature for the bending strength.

4 Summary

Using the following self-propagating high-temperature synthesis,

\[ 3(3\text{Al}_2\text{O}_3\text{SiO}_2) + 8\text{Al} + 6\text{C} = 13\text{Al}_2\text{O}_3 + 6\text{SiC}, \]

we can prepare alumina/SiC composite containing nano-sized SiC particles whose grains size corresponds to one tenth that of commercial SiC particles. The formed nano-sized SiC caused 250 K decrease to the temperature at which complete strength recovery can be attained by self-crack-healing for 10h. Furthermore, the limit temperature for the bending strength of the alumina/SiC nano-composite was found to be equal to that of the alumina containing commercial SiC particles. As a result, new alumina/SiC nano-composite can be applied to structural components with self-crack-healing from 1223 K to 1573 K.

ACKNOWLEDGEMENTS

This study was supported by Grant-in-Aid for scientific research associated to Japan Society for the Promotion Science (JSPS), Young Scientist Research Category (B) No. 17760562, from 2005 to 2007.
REFERENCES