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Short communication

Acoustic emission studies of low thermal expansion aluminum-titanate ceramics strengthened by compounding mullite

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Abstract

Mullite (Mu) with high strength was compounded into aluminum-titanate (AT) ceramics with low thermal expansion to increase their strength. For the AT–Mu system composites, thermal contraction and expansion and acoustic emission (AE) event count rate were measured during cooling using the AE technique and the characteristics of AT–Mu composites were evaluated. The expansion due to microcracking in the range of AE count peak temperatures to room temperature was obtained and the crack volume was estimated from the expansion by cracking. A linear relation with a very high correlation ($r = 0.993$) was observed between bending strength and crack volume at room temperature. From the linear plot, the bending strength at crack-free temperature in the best AT–Mu composite was shown to be 130 MP.

Keywords: Aluminum-titanate–mullite system composite; AE technique; Bending strength

1. Introduction

Materials that undergo repeated thermal shocks are required to show excellent thermal shock resistance. However, materials with low thermal expansion generally have low strength. Aluminum-titanate (AT) ceramics, which are typical materials with low thermal expansion, also have low strength because of many microcracks due to their large thermal expansion anisotropy (coefficient of thermal expansion (CTE) = 1.0×10^{-6}/K, bending strength = 10–40 MPa) [1-2]. Therefore, there have been a number of attempts to improve the properties of AT ceramics [3–5] and to compound ceramics with high strength into AT ceramics [6-7]. However, such composites with sufficient properties have yet to be developed. It is necessary to investigate microcracking temperature during cooling to evaluate the energy of microcracking due to thermal stress. For aluminium-titanate ceramics, Ohya et al. determined microcracking temperature based on measurement of thermal contraction and expansion during cooling, which was accompanied by acoustic emission (AE). They showed that the temperature difference, ΔT, between the sintering temperature and the microcracking temperature could be used as a measure of the energy of grain-boundary microcracking [8]. In composites of AT, the mechanism of microcracking is expected to be more complicated than that of AT ceramics and there have been few studies applying the AE technique to the evaluation of composite ceramics. However, AE and ΔT can also be used as measures of microcracking and microcracking energy in composites, respectively. A decrease in microcracking temperature, i.e., an increase in ΔT, indicates increases in both strength and thermal shock resistance. Therefore, we attempted to strengthen aluminum-titanate ceramics by compounding mullite (Mu) with high strength (300 MPa) and intermediate thermal expansion (4.0×10^{-6}/K) and to evaluate the characteristics of AT–Mu composites using the AE technique.

In the present study, AE event count rate and thermal expansion rate during cooling were measured for AT–Mu system composites using the AE measurement apparatus for high temperatures developed in our previous study [9]. Based on the results, the optimal fabrication conditions where strength was compatible with low thermal expansion were investigated, and also the strength of an optimal composite at crack-free temperature was estimated by performing bending tests at room temperature.

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2. Experimental

2.1. Samples

AT–Mu system composite specimens for measurement of AE and thermal expansion were made as follows. Powders of AT with a particle size of 2.7 μm and Mu with a particle size of 0.8 μm (KM-101 of Kyoritsu Yogyo Genryo Co. Ltd.) were weighed at AT:Mu ratios of 85:15, 80:20, 75:25, and 70:30, and then polycarbonate dispersant (Celuna D-305 of Chyukyo Yushi Co. Ltd.) was added fixing solid content at 75 wt.%. They were then ball-milled for 24 h, and green cylindrical composites were made by oscillating slip casting. The samples were fired for 2 h at three temperatures – 1450 °C, 1550 °C, and 1600 °C – and processed to be 30 mm in length and 15 mm in diameter using an accurate cutting and grinding machine (4020-SM of Tokyo Seiki Kousakusyo Co. Ltd.). A total of 12 specimens were prepared.

Specimens for measurement of bending strength at room temperature were made from AT–Mu system composites fired at 1550 °C using an accurate cutting and grinding machine and a plane grinding machine (SGM-52E2 of Nagase Index Co. Ltd.). The specimen size was 3(t) mm × 4(w) mm × 40(l) mm. A total of 12 specimens were prepared.

2.2. Measurement methods

As the microcracks in AT ceramics were healed at 1100 °C [5], the samples were heated to 1200 °C at a rate of 6 °C/min in a furnace. The samples were held at a temperature of 1200 °C for 30 min, then cooled to 600 °C at a rate of 6 °C/min, after which they were allowed to cool freely. The expansion rate was measured using a laser displacement meter and was calculated using the cooling starting temperature of 1200 °C (T_0) as the standard temperature. The AE event count rate and thermal expansion rate during cooling were measured in the range of 1200–100 °C. AE waves were detected with an AE sensor through the wave-guide of an alumina rod and were transmitted to a discriminator through a pre-amplifier. After amplifying and passing through a high-pass filter with a cutoff frequency of 100 kHz, the AE events were counted by the judgment system with a pair of discriminating levels. The measurement apparatus and the method used were described in our previous paper [9].

The bending strength at room temperature (T_R) was measured by a three-point bending test using a universal testing machine (TENSILON UCT-5T, Orientec Co. Ltd.) according to the JIS bending test standard (JIS $R1601_{1995}$). The span was 30 mm and the crosshead speed was 0.5 mm/min.

3. Results and discussion

AE event count rate every 30 s and thermal expansion rate were measured during cooling for AT–Mu composite systems fired at 1450 °C, 1550 °C, and 1600 °C. Examples of the measurements for those at 1550 °C are shown in Fig. 1. T_P in the figures is the temperature at the maximum AE count peak. In

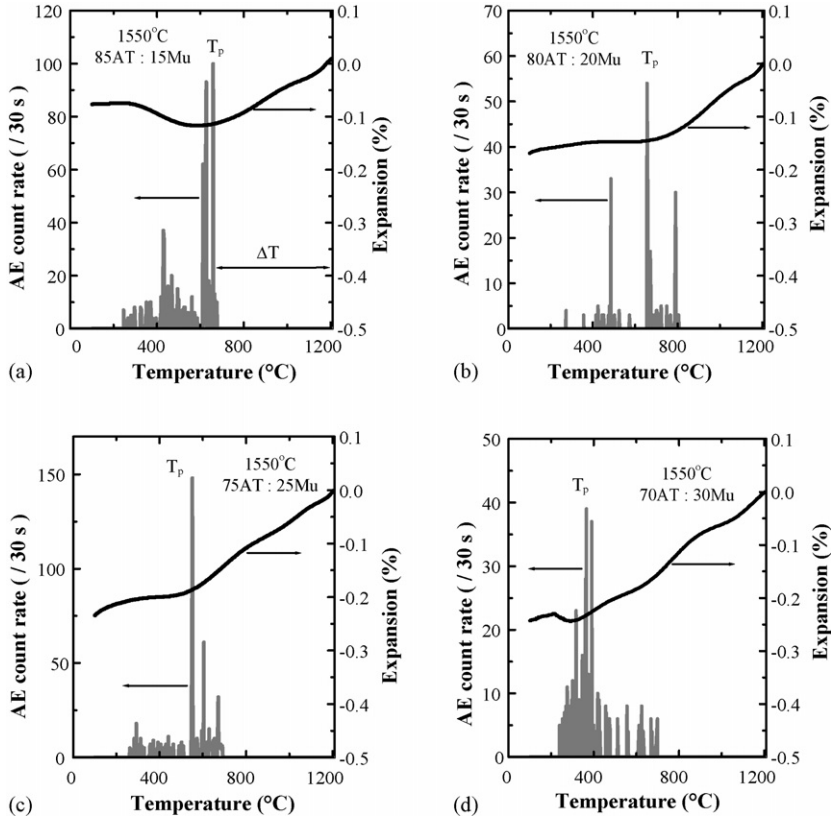

Fig. 1. AE count rate and thermal expansion curve in an AT–Mu composite system fired at 1550 °C. (a) AT:Mu = 85:15; (b) AT:Mu = 80:20; (c) AT:Mu = 75:25; and (d) AT:Mu = 70:30.

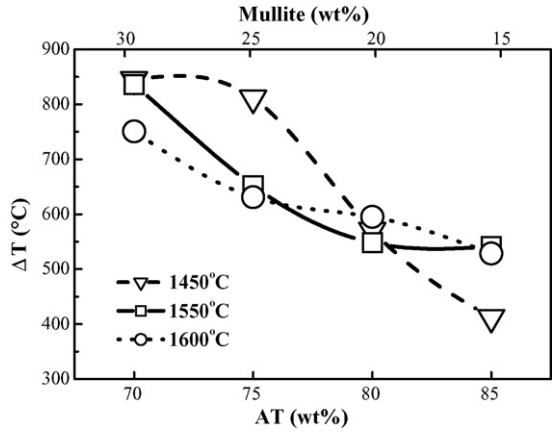

Fig. 2. Composition dependence of ΔT, the temperature difference between cooling start temperature and AE count peak temperature.

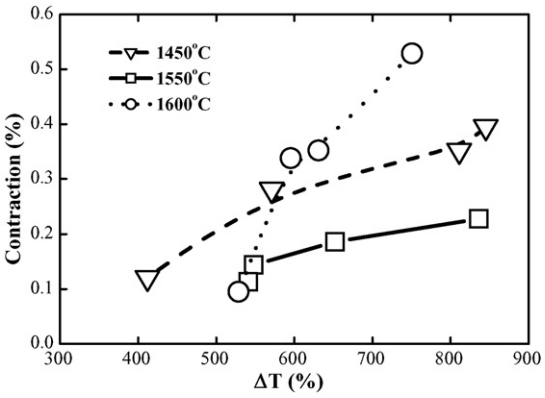

Fig. 4. Relationship between temperature difference, ΔT, and contraction rate in ΔT range in AT–Mu system composites.

each composite system, with decreasing AT ratio, T_P decreased and the thermal contraction at T_P increased and the expansion at temperatures under T_P decreased or increased.

The changes of T_P have the same significance as those in the temperature difference, ΔT, between T_0 and T_P. Therefore, the relation between ΔT and AT ratio was investigated. The results are shown in Fig. 2. This figure indicates that with decreasing AT ratio, ΔT increased because of the decrease in T_P and the change was greater in the composite system fired at lower temperature. ΔT was the greatest in composites with 70% AT, fired at 1450 °C and 1550 °C with values of 845 °C in the former and 840 °C in the latter. Ohya and Nakagawa reported that in AT ceramics, an effective way to obtain crack-free or less-cracked specimens was to sinter the specimen at low temperatures [8]. Our experimental results agreed with their report.

The relation between thermal contraction rate in ΔT range and AT ratio was investigated and the results are shown in Fig. 3. The thermal contraction rate in ΔT range increased with decreasing AT ratio. The rate of increase was markedly dependent on firing temperature and was the smallest in the composite system fired at 1550 °C, of which the contraction rate was the lowest in 70% AT with a value of 0.23%.

Based on these results, the relation between thermal contraction rate and ΔT in AT–Mu composites was investigated and the results are shown in Fig. 4. The figure shows that the contraction rate increased with increasing ΔT, which was a characteristic of this composite system, and only the composite system fired at 1550 °C reached the highest ΔT retaining low thermal expansion. Consequently, in the range examined in this experiment, the optimal fabrication conditions were firing temperature of 1550 °C and composition ratio of AT:Mu = 70:30, with values of ΔT = 840 °C and expansion rate = 0.23%, i.e., CTE = 2.7×10^{-6}/K.

It was difficult to find the strength value at T_P directly in the best specimen with 70% AT, fired at 1550 °C. Then, we attempted to estimate it. First, for the composite system fired at 1550 °C, the bending strength was measured at T_R by a three-point bending test and the best specimen was 101 MPa. Second, from Fig. 1, in the composite system fired at 1550 °C, as the expansion rate decreased to T_P in almost a linear manner with decreasing temperatures, the straight line was extended to T_R, and then the expansion due to microcracking in the temperature range of $T_P - T_R$ was obtained by taking the difference between two expansion rates at T_R. The crack volume was assumed by trebling the expansion difference [10] and that of the best composite was 0.23%. The relation between bending strength and crack volume at T_R in the composites fired at 1550 °C was investigated by plotting those values in Fig. 5. As shown in the

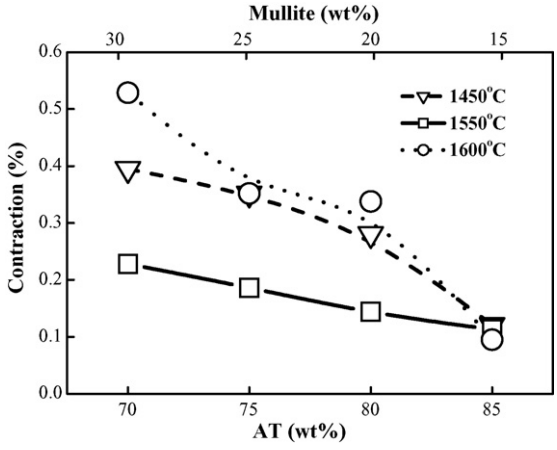

Fig. 3. Composition dependence of contraction rate in ΔT range.

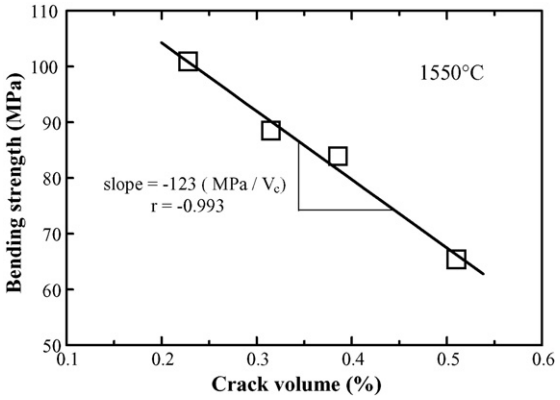

Fig. 5. Effects of crack volume on bending strength in an AT–Mu composite system fired at 1550 °C.

figure, there was a linear relation between the two with a very high negative correlation, $r = -0.993$. Therefore, from this straight line, it was estimated that the bending strength of the best composite at the crack-free temperature of 360 °C was 130 MPa. Consequently, AT ceramic composites strengthened by compounding with Mu could be fabricated and their characteristics could be evaluated by the AE technique for high temperatures.

This composite system showed some AE count peaks during cooling. It is expected that they suggest complex microcracking mechanisms in this composite system and the AE wave gives the information. Further studies are in progress to clarify the microcracking mechanism of this composite system by AE waveform analysis.

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