Classification of Microfracture Process Type in Glass Matrix Composites by Quantitative Emission Method

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Particle dispersed glass matrix composites have been developed in order to increase the strength of glass, and microfracture before the final fracture during bend test has been observed in many ceramics and glass composites as acoustic emission (AE) signals. Stochastic process treatment for microfracture of these composites was performed to understand the mechanical properties of these materials. Bending strength of these materials was measured as various conditions in loading rate and atomosphere. AE behavior during these tests was also detected with two transducers and a two-channel waveform acquisition system to evaluate the microfracture location in the materials. Microfracture processes during bending tests were clearly classified into four major types from the results of source location of AE, that is, (i) unstable fracture type, (ii) crack propagation type, (iii) transition type from random microfracture to crack propagation and (iv) competition type between random microfracture and crack propagation. The effect of loading rate, atmosphere and volume fraction of reinforcing particles on microfracture process was discussed.

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1. Introduction

Fracture in ceramics and glass has been considered to be essentially a brittle and probabilistic phenomenon. Thus Weibull distribution, which is based on the weak link theory, has been used in order to analyze and explain the scatter of strength of these brittle materials, where only one crack is considered.¹ However, many possible sites of microfracture exist in real materials such as pores, grain boundary, interface between matrix, filler and so on.² Assumption of Weibull distribution may be satisfied in monolithic glass and ceramics without intergranular fracture, but many ceramics with weak grain boundary which demonstrate complicated fracture behavior with crack arrest and deflection would not seem to satisfy these assumptions completely. Glass or ceramics composites with reinforcement also do not demonstrate a simple fracture behavior in general. Weibull plot is very convenient technique to explain the scatter of strength of brittle materials, but this method cannot be easily applied to any materials without careful consideration of their microfracture process.

Acoustic emission (AE) is a phenomenon, which occurs due to microfracture before the final fracture, and many studies have been reported to understand the fracture process of various materials. AE events related to microfracture were also observed in many ceramics and glass composites under various stress conditions.³⁻⁵ Most of the microfracture in process zone near the crack tip of ceramics such as microcrackting, crack deflection and transformation releases energy in its generation, which is detectable as AE event through surface displacement. Many AE events before rupture were also observed during bending test in monolithic ceramics, ceramic composites and glass composites, and the distribution of AE source locations which was calculated from the difference of arrival time between AE sensors was found to be dependent on environmental parameters as well as materials properties.⁶⁻⁷ The results of measurements of AE source locations during four point bending test of PbO-borosilicate glass composites were reported,⁸ where the distribution of AE source locations during test in vacuum demonstrates that microfracture in these materials occurred randomly everywhere in specimen, on the other hand, the distribution in air shows that the concentrated microfracture was related to the slow crack growth in these materials. As these kinds of fracture behavior are not consistent with the assumptions of Weibull distribution obviously, the theory or models for the stochastic microfracture behavior should be developed in order to quantitatively characterize these behaviors.

In the previous paper,⁹ the analytical approach was proposed to explain two above mentioned microfracture behaviors of both independent and concentrated microfracture. Expressions of probability function of microfracture stress and location for independent microfracture were represented where it was assumed that there is no interaction between microcracks and microfracture stress obeys the Weibull distribution. Concentrated fracture was described using the step function where fracture is assumed to occur at a certain point. Plots resulting from these models were very useful to understand the whole behavior of stochastic microfracture in brittle materials. These models could quantitatively explain experimental data in simple cases such as random microfracture in very dry condition or crack growth in wet condition. In the present study, quantitative AE method was applied to the bending tests of glass matrix composites with various volume fractions under several loading rate and test atmosphere in order to understand the relationship between strength and particular type of microfracture process or the effect of environment on microfracture process.

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2. Microfracture Process Models

Analytical models for stochastic microfracture process were proposed in order to understand the scatter of strength of materials or the effect of testing environment and explain the AE behavior during bending test. As mentioned above, these models have a limitation to explain the whole experimental data. However, this approach gives an idea to characterize the stochastic microfracture process. Here two models are considered such as independent microcracking and crack propagation models.

2.1 Independent microcracking model

It is assumed that there is no interaction between microcracks generated during the test and microfracture stress is described by Weibull distribution. Also uniaxial tensile stress of four point bending test is supposed. Probability density function (PDF) for microfracture stress and location can be represented as

\[ f(\sigma_m, x) = \exp \left\{ - \int \left( \frac{\sigma}{\sigma_0} \right)^m d\sigma \right\} \frac{\partial}{\partial \sigma_0} \left( \frac{\sigma}{\sigma_0} \right)^m \]  (1)

where \( \sigma_m \) is the reference stress, \( x \) is a location, \( m \) and \( \sigma_0 \) is a scale and shape parameter of Weibull distribution, respectively. The probability density function for microfracture is defined as

\[ g(\sigma_m) = \int_L f(\sigma_m, x) dx \]  (2)

where \( L \) indicates the stressed region. By integrating the eq. (2) in location the probability distribution function for microfracture for four point bending can be represented as

\[ G(\sigma_m) = 1 - \exp \left\{ -2b \left( x_1 - \frac{m x_0}{m + 1} \right)^m \right\} \]  (3)

where \( \sigma_m \) is the maximum tensile stress at the stressed surface of bend specimen, \( x_0 \) means the half of the difference between upper and lower span and \( x_1 \) means the half of upper span, respectively. The plot of eq. (3) in the form of \( \ln \left( 1 / (1 - G(\sigma_m)) \right) \) versus \( \ln \sigma_m \) gives a declining line and the slope of line represents a scale parameter \( m \) of Weibull distribution for microfracture.

2.2 Crack propagation model

Crack propagation behavior is essentially a stochastic process and the model describing stochastic process is necessary to quantitatively evaluate it. It is not easy to establish a stochastic process model for crack propagation in order to quantitatively evaluate the experimental results. The following model was considered to qualitatively understand the difference between independent microfracture and crack propagation. Suppose that a crack propagates from an origin at certain stress very rapidly, and then the probability density function for microfracture stress is defined as

\[ g(\sigma_m) = \delta(\sigma_m - \sigma_f) \]  (4)

where \( \sigma_f \) is a fracture stress and \( \delta(x) \) means the delta function. By integrating the eq. (4) the probability distribution function can be represented as

\[ G(\sigma_m) = u(\sigma_m - \sigma_f) \]  (5)

where \( u(x) \) means the Heaviside unit function. The plot of eq. (5) gives a vertical line.

3. Experimental Procedure

3.1 Materials

SiC particle dispersed CaO–BaO–SiO_2 glass composites were used in experiments. The average size of SiC particle was about \( 8 \mu \text{m} \) and the volume fraction of particle was 10, 20 and 30\%, respectively. The glass powder and SiC particle were mixed by ball milling in methanol and were dried in air. The mixed powder was sintered using hot press. The hot pressing temperature was \( 870°C \) that were \( 20°C \) higher than the softening point of this glass and pressure was applied up to \( 30 \text{ MPa} \) to increase the density.

3.2 Four point bending test

Samples were cut to 3 by 4 by 40 mm and were polished using 1 \( \mu \text{m} \) diamond slurry. The bending strength was measured according to JIS-R1601 of four-point bending tests, with upper span of 10 mm and lower span of 30 mm. The cross head speed (CHS) was changed from \( 2 \times 10^{-7} \) to \( 1 \times 10^{-5} \text{ m/s} \) and tests were performed in both air and vacuum (\( 6.6 \times 10^{-3} \text{ Pa} \)). Fracture surfaces after four point bending tests were observed by scanning electron microscope (SEM).

3.3 AE measurement and analysis

AE measurements were performed during strength tests. AE sensor used in this study was wide range response type (M304A, Fuji Ceramics, Japan) and AE measuring system was DCM140E (JT, Japan), respectively. The AE sensors were attached to both ends of the specimens. Figure 1 shows the AE measuring system and measuring condition. The one-dimensional source location with two channels AE system was carried out. Probability of microfracture during bending test was calculated using microfracture stress and locations

![Fig. 1 Schematics of four point bending test and AE measurement.](image-url)
obtained from AE results. The median rank method was used to estimate the probability, which was plotted in the form of Weibull plot.

The location of each source event is determined by measuring the differences in the wave arrival time between two transducers. The general equation for source location can be solved using the least-square method. The detected signals of AE are not generally the displacement field because of the response function of the measuring system. The detected signals can be represented by the convolution integral of the transfer function of sensor, the Green’s function of media and the moment tensor of microfracture. In order to determine the moment tensor and characterize the AE sources, the multiple deconvolution method must be carried out in multiple convolution equation by using the recorded AE waveform with more than six channels. However, if the mode of microcracking is the tensile type, this equation can be reduced to a simple linear convolution equation, and then only the size of microcracking is the unknown parameter and the measurement with one channel can determine the size and generation velocity of microcracking using the convolution method. The response function, which includes both the transfer function of the measuring system and the Green’s function of specimen, can be experimentally calibrated by a breaking pencil lead.

4. Results and Discussion

4.1 Strength and microfracture behavior

Figure 2 shows the effect of loading rate on strength in the 20 vol%SiC glass matrix composite. The bending strength in air was increased with the increase of loading rate. Bending strength in vacuum was slightly increased with the increase of loading rate. The strength in vacuum was higher than that in air at the whole loading rate. This behavior demonstrates the effect of water in atmosphere on bending strength. Figure 3 shows the effect of loading rate on the total number of AE events in the 20 vol%SiC glass matrix composite during four point bending test. Total number of AE during bending test in air was decreased with the increase of loading rate. Number of AE in vacuum was smaller than that in air and it was almost independent of loading rate. This AE behavior also suggests the effect of water on microfracture process and strength. Since microfracture process in vacuum does not depend on the loading rate, strength is also independent. On the other hand, microfracture process in air is strongly affected by loading rate because water in environment induces many microcracks and also reduces the strength of materials. The increase of AE events with the increase of testing time also supports that the stress corrosion cracking by water produces a lot of microcracks. However, the reduction of strength is saturated with the increase of microcracks. As discussed later, a critical microcrack size and a reduced crack propagation resistance would control the strength of these materials.

4.2 Source location and microfracture process type

It is possible to estimate a location of each microfracture with applied stress from the detected AE waveforms using source location technique. These results were used to classify the types of microfracture process and the following four types were identified. Figure 4 shows an example of source location of AE during bending test in 10 vol%SiC glass matrix composite in vacuum at cross head speed of $2 \times 10^{-7}$ m/s.

![Fig. 2 Effect of loading rate on four point bending strength in 20 vol%SiC glass matrix composite in both air and vacuum environment.](image1)

![Fig. 3 Effect of loading rate on total number of AE events during four point bending test in 20 vol%SiC glass matrix composite in both air and vacuum environment.](image2)

![Fig. 4 An example of source location result for “unstable fracture type” in 10 vol%SiC glass matrix composite in vacuum at cross head speed of $2 \times 10^{-7}$ m/s.](image3)
In this "unstable fracture" type, only one AE event was generated with the final fracture of specimen. The microfracture process in this type is considered as follows. Microfracture is generated and subsequent crack proceeds without arrest, because the local stress intensity factor due to microcrack size and applied stress on surface of specimen is larger than the critical stress intensity factor for crack growth.

Figure 5(a) shows another example of AE source location in 20 vol%SiC glass matrix composite in vacuum at cross head speed of $2 \times 10^{-7}$ m/s, where horizontal axis means a logarithm of stress and vertical axis is a probability of microfracture in the form of Weibull plot. The mean rank method was used to plot experimental data. Location of AE is also shown in Fig. 5(b), which demonstrates that AE events were generated at the same position. In this "crack propagation" type, AE was generated in the restricted area and stress. This behavior corresponds to the crack propagation model described in the above chapter. Some AE events occurred in this specimen because each microcrack is restricted or arrested by microstructure of material and the first microcrack size is not enough for unstable crack propagation.

Figure 6 shows a location result in 30 vol%SiC glass matrix composite in vacuum at cross head speed of $1 \times 10^{-5}$ m/s, where vertical axis means the order of AE events. This is a typical example of "transition" type from random microfracture to crack propagation, that is, the first ten AE events were generated almost randomly within short span and then AE events occurred in the restricted area. Probability of this microfracture is plotted in the form of Weibull plot, shown in Fig. 7(a). This plot clearly demonstrates the transition of microfracture from independent microcracking type to crack propagation type, described in the above chapter.

Some specimens showed a slightly different type of microfracture process. Figure 8 shows a location result in 30 vol%SiC glass matrix composite in air at cross head speed of $4 \times 10^{-6}$ m/s. Microfracture process in the beginning was very similar to the previous transition type. However, the transition to crack propagation is not clear and AE events were observed in several locations of specimen. The final fracture occurred at one of the microfracture sites, that is, the ruptured location was determined after the "competition" of several microfractures.

Microfracture processes during bending tests were clearly classified into above four types from the results of source location of AE, that is, (i) unstable fracture type, (ii) crack propagation type, (iii) transition type from random microfracture
to crack propagation and (iv) competition type between random microfracture and crack propagation. The effect of loading rate, atmosphere and volume fraction of reinforced particle on microfracture process was summarized in Fig. 9. All the specimens with 10 vol% SiC demonstrate “unstable fracture” like monolithic glass because there is little effect of reinforcement on microfracture process. Four microfracture types were observed in 20 vol% material, but most of fracture type was “competition” type. In this material vacuum environment increased the ratio of “unstable fracture” type and there was no effect of loading rate in vacuum on microfracture process type. On the other hand, the increase of loading rate induced “unstable fracture” in air environment. As there were many microfracture sites due to reinforcement by particles in 30 vol% material, microcracks were easily generated and arrested, that is, “competition” type microfracture might be mainly generated. However, “transition type” microfracture process in 30 vol% material was frequently observed in vacuum environment nevertheless “competition” type was dominant. That is, microfracture sites were restricted due to the reduction of stress corrosion effect and it became difficult that cracks grew at several locations.

4.3 Criterion for different microfracture process

Stress intensity factor $K_1$ for a semi-elliptical surface flaw in a flat plate can be represented as:

$$K_1 = (\sigma_m + H\sigma_b) \sqrt{\frac{\pi a}{Q}} F\left(\frac{a}{r}, \frac{a}{c}, \frac{c}{w}, \phi\right)$$

where $\sigma_m$ is a tensile stress, $\sigma_b$ is a bending stress, $a$ is a depth of crack, $2c$ is a width of crack, $\phi$ is an elliptical angle, $2w$ is width of specimen, $r$ is a thickness of specimen. Complete form of equation is described in Ref. 13. It is assumed for calculation that $a$ equals to $c$ and the value at $\phi = 0$ is used for later discussion.

The 10 vol% material demonstrates “unstable fracture” with only one AE event. As AE measurement system has some limitations such as sensitivity, threshold and non-
sensitive time, in general there is a possibility not to detect a microcrack as AE event. However, a crack cannot stop if a distance between ceramic particles is considerably far, because possible microcrack sources are matrix cracking and interfacial debonding in the ceramic particle reinforced glass matrix composites. On the other hand, a generated crack will be arrested if this distance is enough short. Microstructure of the 10 vol% material showed that a distance between particles was frequently over 100 μm, and a calculated surface flaw size from the strength of 150 MPa and toughness of 1.2 MPa√m by eq. (6) is about 80 μm, that is, a crack is not arrested and microcrack process demonstrates “unstable fracture” type.

Figure 5(b) shows the location and estimated microcracking size. Since the size of first microcrack is about 140 μm and the applied stress is 166 MPa, the calculated stress intensity factor by eq. (6) is 1.78 MPa√m, which is just lower than the toughness of 1.8 MPa√m in this material. That is, a crack was arrested after the first microcrack until a crack satisfies the criterion for unstable crack growth and microcrack process represents “crack propagation” type. The first microcrack stress is reduced by the effect of stress corrosion cracking because the toughness for stress corrosion cracking is decreased by the existence of water.

The difference between “transition” and “competition” types is due to the frequency of existence of microcrack sites. That is, microcracks occur randomly before crack growth as a microcrack is arrested a ceramic particle, and then the stress intensity factor of a microcrack reach the critical stress intensity factor for stress corrosion cracking and this microcrack starts propagation. However, if the material is significantly affected by water, cracks easily grow at several locations simultaneously as microcracks are generated in a low stress and a plenty of microcracks occur everywhere in the specimen. Figure 8(b) shows that at least two cracks grew in the specimen and this sample demonstrates “competition” type microcrack process. The estimated crack size before crack propagation by AE source characterization is shown in Fig. 7(b), where the maximum size of microcrack is about 80 μm and the corresponding stress intensity factor from the stress of 92 MPa is estimated as 0.75 MPa√m, and also the location of maximum microcrack is same as the position of crack propagation. The stress intensity factor for stress corrosion cracking is evaluated from the stress of crack propagation of 110 MPa as 0.89 MPa√m using eq. (6). Thus, the quantitative AE method also enables to evaluate the properties of slow crack growth.

5. Conclusions

Microcrack process in SiC particle dispersed glass matrix composites was analyzed by quantitative AE method. The following conclusions are obtained.

(1) The loading rate and environment affected both strength and total number of AE. Strength in vacuum was higher than that in air. On the other hand, number of AE in vacuum was smaller than that in air. This behavior was related to the stress corrosion crack growth by water.

(2) Source location of AE using two sensors attached to the edge of bending specimen demonstrated the difference between microcrack processes. Microcrack process in these materials was clearly classified into four major types from the results of source location and two stochastic microcrack models. This technique was very useful to understand the microcrack process and strength of glass matrix composite materials.

(3) Air environment induced both the increase of number of microcracks and the decrease of the stress for generation, and the increase of volume fraction of particles reduced the size of microcracks. These effects on microcrack process could be explained by the results of each microcrack size by AE source characterization and toughness of materials.

REFERENCES