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Conductive Coating on Structural Ceramics for Strain Detection Utilizing Electrical Measurements

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Abstract

Strain detection in Al_2O_3 ceramics and glass plates was investigated by coating them with an electrically conducting composite (epoxy resin and needle-like $SnO_2(Sb)$-coated TiO_2 filler) and by measuring surface resistance during and after loading. By adding more than 6 vol%-filler, the composite became electrically conductive. Surface electrical resistance increased with increasing strain during loading, and the degree of electrical resistance change versus strain was larger when the filler volume fraction was close to the percolation critical volume fraction. In addition, when the specimens were cyclically loaded, residual electrical resistance was observed even after removing load. The value of the residual electrical resistance was dependent on the maximum strain under the stress applied. These results suggest that estimation of maximum strain is possible by measuring resistance of the composite formed on structural ceramics. Based on the results of microfracture observation, the effect of applied stress on the electrical resistance change of electroconductive composites is discussed. © *1999 Elsevier Science Limited. All rights reserved*

Keywords: strain detection, composites, electrical, Al_2O_3, glass, sensors.

1 Introduction

Since ceramics generally exhibit brittle fracture behavior, a great deal of investigation has been conducted to improve their reliability as structural materials. A fundamental approach to improve reliability is the design of ceramics with high fracture toughness and strength,[1] and various methods have been carried out such as the use of stress-induced martensitic transformation of metastable tetragonal zirconia grains[2–4] and the fabrication of ceramic matrix composites reinforced with whiskers,[5,6] platelets,[7] rod-like grains,[8] and fine particles in nanometer scale.[9–11] However, brittleness is an intrinsic character of a ceramic material and hence, it is difficult to overcome this problem completely in ceramics for practical use. Accordingly, development of screening techniques and methods for damage detection and fracture forecasting are important subjects to ensure the reliability of structural ceramics. To prevent unstable fracture of a ceramics part, it is necessary and important to characterize the defect initializing the fatal fracture and strain in the part induced by loads. For example, in some ceramics showing subcritical crack growth (SCG), including alumina ceramics, a crack starts propagating as soon as the stress intensity factor (K) exceeds some threshold value, K_0, before reaching K_{IC}.[12–15] Because the stress intensity factor is determined by the stress near the crack tip and shape of the crack tip, it is very important to determine maximum stress or strain applied, crack shape and its location in a ceramics part. From the viewpoint of strain detection, a gauge method is usually applied to measure direct strain under conditions of practical use; however, this is not practical for determination of maximum strain applied after the load has been removed. In addition, non-destructive evaluation techniques using X-ray, ultrasonic and acoustic emission (AE) have been widely investigated and applied to evaluate cracks in materials. However, since those techniques demand large-scale equipment

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for evaluation of cracks, they are generally not available for materials with cracks under condition of practical use. Accordingly, a novel simple method for improving the reliability of brittle ceramics has been desired.

Recently, a method of fracture forecasting by electrical measurement has been proposed. Muto *et al.*[16,17] reported the possibility of forecasting of fatal fracture in carbon-fiber-glass-fiber-reinforced plastics (CFGFRP) composites by measuring electrical resistance. Electrical resistance of CFGFRP composites increases during loading and the increased resistance remains after removing the load. The residual change in resistance can be used as the parameter to indicate the maximum load applied to the material in the past. Ishida *et al.*[18] fabricated CaF_2/SiC-whisker composite ceramics in which an electric conduction path is formed by SiC whiskers. The phenomenon similar to that in CFGFRP was observed, and it was confirmed that the fracture prediction is possible even in ceramic composites, although the elastic moduli of the composite decreased with increasing SiC whisker content. They also reported the effect of tensile and compressive stress on the change in electrical resistance of the composites, and confirmed that the change of electrical resistance occurs by a change in connectivity of SiC whisker in the composites.[19] This self-diagnosis mechanism as an intelligent function in ceramic materials has been proposed by Yanagida.[20] Although it is promising to fabricate ceramic composites provided with such a self-diagnosis mechanism, available ceramic systems are limited and it seems difficult to maintain the mechanical properties at a level equivalent to those of the structural ceramics already used. In addition, because engineering ceramics usually show small strain until fracture, it is necessary for evaluating the strain that the electrical resistance change by strain must be much larger than has been obtained so far. Accordingly, we tried to introduce a thin percolation phase provided with electrical conduction onto structural ceramics, then carried out evaluation of the strain in ceramics coated with them from electrical measurements. In addition, there has been little research regarding electrical properties of electronic conductor/insulator composite in film morphology, especially its electrical resistance change under applied stress.

In the present paper, a new technique for strain detection in structural ceramics is reported. A conducting composite, consisting of epoxy resin and conductive ceramic filler, was coated onto Al_2O_3 ceramics and glass, and electrical resistance changes in the composite caused by applying stress to the brittle substrate were investigated during loading and after unloading. In addition, from electron microscopy observation, microscopic fracture behavior in the electroconductive composite film was examined to elucidate the mechanism of the resistance change induced by the stress field.

2 Experimental Procedure

2.1 Sample preparation

Epoxy resin was selected for insulating matrix polymer. Electroconductive composite film was prepared by mixing low-viscosity epoxy resin (DGEBA, Epikote 828, Yuka Shell Epoxy K. K., Tokyo, Japan), an amine curing agent (ATU, Epomate B-002W, Yuka Shell Epoxy K. K., Tokyo, Japan), needle-like electrically conductive filler with a 0·2 μm average diameter and average initial length of 5·1 μm (SnO_2(Sb)-coated TiO_2, FT-3000, Ishihara Sangyo Kaisha Ltd., Mie, Japan) and 1,4-dioxane as solvent. To obtain films with a uniform thickness, the viscosity of solutions was controlled adequately by adjusting the amount of solvent. Glass with dimensions 75×26×1 mm and Al_2O_3 with dimensions 50×25·4×0·9 mm were used as testing plates. They were ultrasonically cleaned in acetone and the mixed coating suspension was spin-coated onto the plate in air at 1600 rpm for 20 s and then cured at 130°C for 2 h in air.

2.2 Microstructural and microfracture analysis

For characterization of the film morphology and determination of film thickness, film surface and fracture surface of these samples were examined with a scanning electron microscope (SEM) (Model JSM-T330A, JEOL Co. Ltd., Japan) using secondary electron imaging, operated at 15 kV. A transmission electron microscope (TEM) (Model JEM-2010, JEOL Co. Ltd., Japan) at an accelerating voltage of 200 kV was used to characterize morphology and microstructure of SnO_2(Sb)-coated TiO_2 filler and analyze microfracture of the composite film.

2.3 Electrical and mechanical measurements

Measurement of surface electrical resistivity of the films at room temperature was performed by two-probe direct-current method using a digital electrometer (Model TR8652, Advantest Co. Ltd., Tokyo, Japan) which has a constant voltage supply within. Silver electrodes were formed on the surface of the film for electrical measurements. A strain gauge was attached onto a film surface to measure strain using a digital strain amplifier card (Model PCD-100A, Kyowa Electronic Instruments Co. Ltd., Tokyo, Japan). Electrical resistance of the specimens under applied stress was measured by setting the composite film on the lower side of

specimens in a three-point bending test. The function of strain detection was evaluated by simultaneous measurement of load, strain and electrical resistance under three-point bending load at 25°C. The three-point bending test machine (Model AGS-5kND, Shimazu Co. Ltd., Kyoto, Japan), supplied with an airtight sample box, was used to apply bending load (40 mm span) to the specimen with crosshead speed of 0·5 mm min^{-1}. The load was applied to specimens in a single and cyclic mode until the specimens fractured. Since the surface resistivity of composite films is slightly influenced by relative humidity, electrical measurements under bending load were performed at constant relative humidity of 65%. Relative humidity was adjusted by controlling flow rates of dry air and wet air saturated with distilled water vapor, and monitored with a humidity meter (Model AY-21, Yamato Co. Ltd., Tokyo, Japan), with accuracy of 3%.

3 Results and Discussion

3.1 Microstructure of fillers and electroconductive composite films

Figure 1 shows the microstructure of SnO_2(Sb)-coated TiO_2 filler examined by TEM. The fillers have needle-like morphology [Fig. 1(a)], and are revealed to consist of fine SnO_2(Sb) particles ($\approx$10 nm) and TiO_2-whiskers (rutile structure) [Fig. 1(b)]. The fine SnO_2(Sb) particles are highly agglomerated on the surface. The surface of the filler is rough due to agglomerates of fine SnO_2(Sb) particles on the surface. These results make us expect that the contact condition of fillers in matrix is sensitive to applied stress.

Figure 2 shows SEM micrographs of surface morphology of the composite film and fracture surface of glass coated with composite. There are no voids but some agglomerates of filler were observed on the surface [Fig. 2(a)]. The film thickness is approximately $10 \pm 2\,\mu$m independent of filler concentration and substrate materials. Voids in composite film and interface debonding between the film and substrate are not observed on fracture surface [Fig. 2(b)]. Since interface debonding between the film and substrate was not observed even after cyclical loading, adhesive strength of the film to substrate is considered to be strong.

3.2 Electrical resistivity of composite films

The electrical resistivity of electroconductive composites has been the subject of much interest and many experimental investigations have been made on this problem for many years.[21–23] The percolation transition between the insulating and conducting states has been observed in many studies about composites of insulating polymer and conductive filler and some experimental results are compared

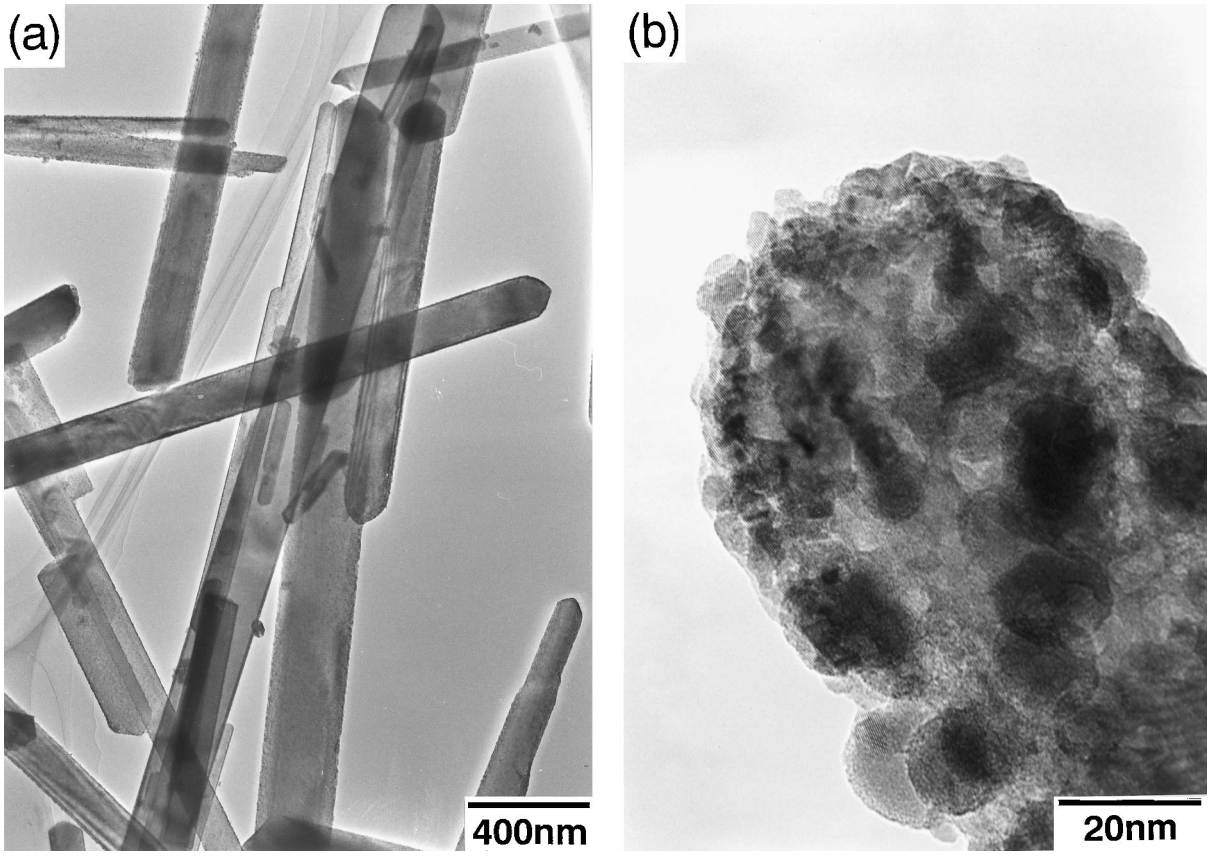

Fig. 1. Bright-field (a) low and (b) high magnification TEM images of SnO_2(Sb)-coated TiO_2 filler.

with theoretical models based on Kirkpatrick's percolation theory.[24]

Figure 3 shows the composition dependence of surface resistivity of the composite film on glass. As the volume fractions of filler is increased, a sharp drop of resistivity is observed at about 6 vol%-filler, and the resistivity further decreases at higher filler fractions. This result indicates that a network of conducting phase by agglomerates of the filler is formed above 6 vol% fraction of filler. Surface resistivity of the composite below percolation threshold is very high and beyond the measurable limit ($>10^{12}\Omega/\square$).

In the case of random fiber-polymer composites,[25,26] the network formation depends mostly on the geometrical effects, especially their aspect ratio of filler. In fact, it has been reported that the volume fraction of fiber to form a conduction path decreases with increase in fiber aspect ratio. Aspect ratio of filler (SnO_2(Sb)-coated TiO_2) used in the present study is approximately 25. On the other hand, in short carbon fiber (CF) filled polychloroprene composites,[26] Jana reported that the critical volume fraction of CF was observed to lie in the range 7–10·5 vol% of CF when the aspect ratio of CF was 25. Compared with this report, the present results show a lower critical volume fraction of filler. This disagreement seems to be due to a difference of dispersion condition of fillers in the matrix polymer on account of difference in wettability of polymer to filler, and the film morphology of composite in the present study.

3.3 Electrical resistance change under applied stress

Fig. 4 shows the relationship between strain and electrical resistance change for glass coated with composite containing various filler fractions under a three point bending load. For all specimens measured, surface electrical resistance increases with increasing strain. The changes in electrical

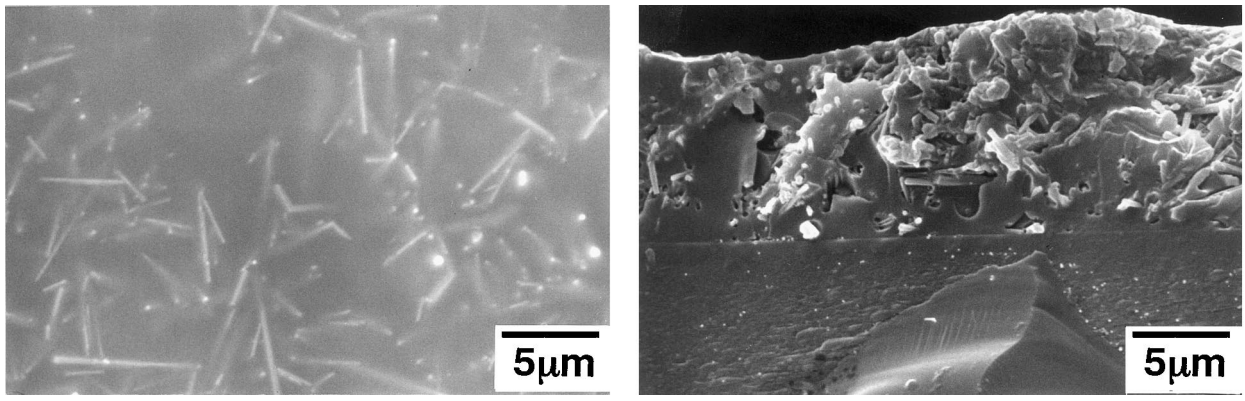

Fig. 2. Scanning electron micrographs for (a) surface morphology of the composite film containing 10·5 vol% of fillers and (b) fracture surface of glass coated with composite containing 10·5 vol% of fillers.

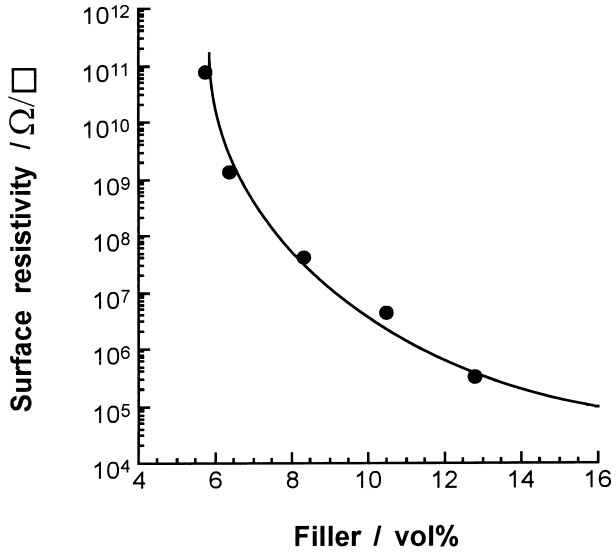

Fig. 3. Composition dependence of surface resistivity in the composite film formed on glass at 25°C.

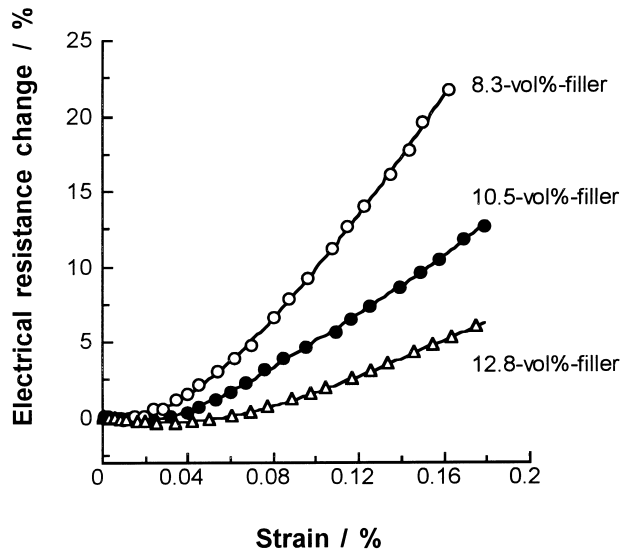

Fig. 4. Relationship between strain and electrical resistance change for glass coated with composite containing various filler fractions.

resistance versus strain is obviously larger when the filler concentration is closer to the critical volume fraction. In CaF_2/SiC composites,[18] the changes in resistance as a function of deflection showed no clear dependence on the SiC whisker content, however, it is revealed in the present study that the obvious composition dependence of resistance change by strain existed in the electroconductive composite, as shown in Fig. 4. On the other hand, the degree of electrical resistance change versus strain is higher than that of CFGFRP composites, reported previously.[16,17] The present result is considered to be derived from breaking of the continuous conduction paths in the composite. On the other hand, in the case of CFGFRP composites, the resistance increase is derived from partial fracture of conductive fiber (carbon fiber).

Cyclic loading test of specimens was carried out. Figure 5 shows the relationship between load, strain, and electrical resistance change for glass coated with composite containing 8·3 vol%-filler. In this test, the specimen was loaded and unloaded cyclically with progressive increase in strain as indicated by arrows in this figure. The load in this material is proportional to the strain. In the same way as with the single mode test, electrical resistance increases with increasing strain. However, hysteresis is observed in the relationship between strain and electrical resistance, although the load in the specimen is proportional to the strain until fracture. In Fig. 5, electrical resistance increases nonlinearly with increasing strain initially (stage 1). However, the decrease in electrical resistance on removing the load (stage 2) is smaller than the resistance increase on loading, and hence a residual electrical resistance is observed (stage 3). This result suggests that the detection of strain in ceramics by electrical resistance change in composite coated on the ceramics during loading is possible.

Figure 6 shows the relationship between changes in residual electrical resistance ($(R_{re}-R_0)/R_0$) and the maximum strain applied for glass coated with composite containing various filler fractions. The residual electrical resistance of the composite films is systematically dependent on the maximum strain applied in the past, which is also the case for any other composite within the strain region measured. This means that the electrically conductive composite film formed on ceramics have the ability to memorize the maximum strain applied in the past as a residual electrical resistance. This can be used as a signal which indicates the possibility of damage (crack propagating) in brittle ceramics showing subcritical crack growth as mentioned in the introduction, i.e. when the K_I value induced by maximum stress calculated from the residual resistance is larger than K_0; the ceramic part can then be recognised to have suffered from some damage. To this point, it is thought that our method has a merit compared with acoustic emission (AE), since the AE method cannot determine damage condition of structure after unloading. Moreover, compared also with the conventional gauge method, since the coating itself can memorize the maximum strain as residual resistance, there is no necessity for sequence measurement of strain while the gauge method demands sequence measurement for assessing the maximum strain applied.

Figure 7 shows the relationship between strain and electrical resistance change for Al_2O_3 and glass

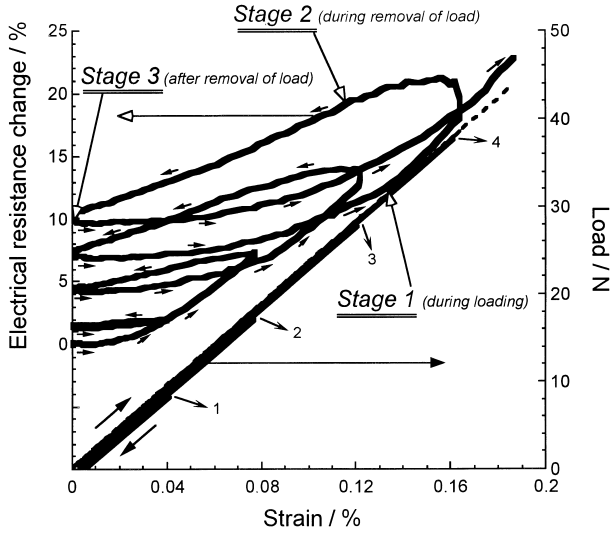

Fig. 5. Relationship between load, strain, and electrical resistance change for glass coated with composite containing 8·3 vol% of fillers. In this test, the specimen was loaded and unloaded cyclically with progressive increase in strain as indicated by arrows in this figure. Numbers in the load-strain curve indicate maximum strain (load) for each cycle.

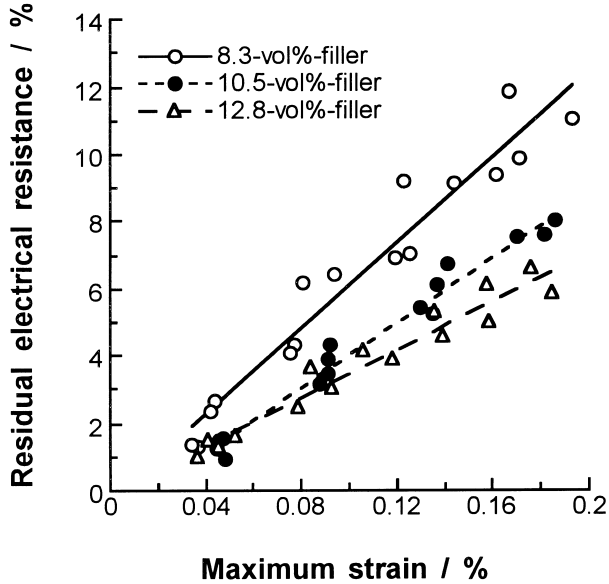

Fig. 6. Relationship between maximum strain applied in the past and changes in residual electrical resistance after removing load for glass coated with composite containing various filler fractions.

coated with composite containing 8·3 and 10·5 vol%-filler. The electrical resistance increases with increasing strain. The behavior of electrical resistance change of composite coated on Al_2O_3 almost coincides with that on glass when filler concentration is the same. Figure 8 shows the relationship between changes in residual electrical resistance and the maximum strain applied for Al_2O_3 and glass coated with composite containing 8·3 and 10·5 vol% filler. The behavior of residual resistance change of composite coated on Al_2O_3 is also identical to that on glass when filler concentration is the same. These results suggests that strain detection of structural ceramics coated with the composite is possible regardless of the kind of ceramics. At the same time, considering the results that a resistance change is observed even in the small strain region ($< 0{\cdot}1\%$), the strain detection and application for damage estimation in other structural ceramics would be possible by coating them with the composite and by measuring its electrical resistance.

3.4 Microfracture observations

The electrical resistivity of electroconductive composites is strongly related to the geometric arrangement of the conductive second phase restrained by the insulating matrix; hence, the resistance change induced by applied stress should be reflected by the arrangement change of filler resulting from matrix deformation and/or fracture, and by the breaking of the fillers which form the continuous conduction path. Accordingly, characterization of fracture behavior of the composite and the interface of filler/matrix is considered to be very important to elucidate the mechanism of resistance change. Therefore, we carried out electron microscopic observation, including SEM and TEM, on the composite films to characterize microfracture behavior and know some information of the filler/matrix interface.

Figure 9 shows a SEM micrograph of a fracture surface of the composite film. Filler pull-out behavior is observed in all fracture surfaces. Microcracks of matrix polymer near the filler pullout are observed as indicated by arrows in this figure. However, filler break was not observed in fracture surface. Figure 10 shows a TEM micrograph of fracture surface for the composite film. Pull-out filler is also observed, and the remarkable plastic

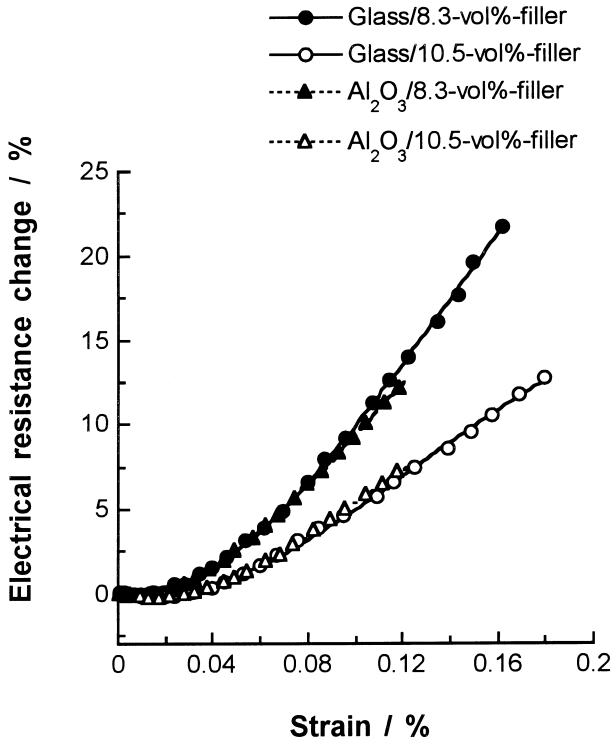

Fig. 7. Relationship between strain and electrical resistance change for Al_2O_3 and glass coated with composite containing 8·3 and 10·5 vol% of fillers.

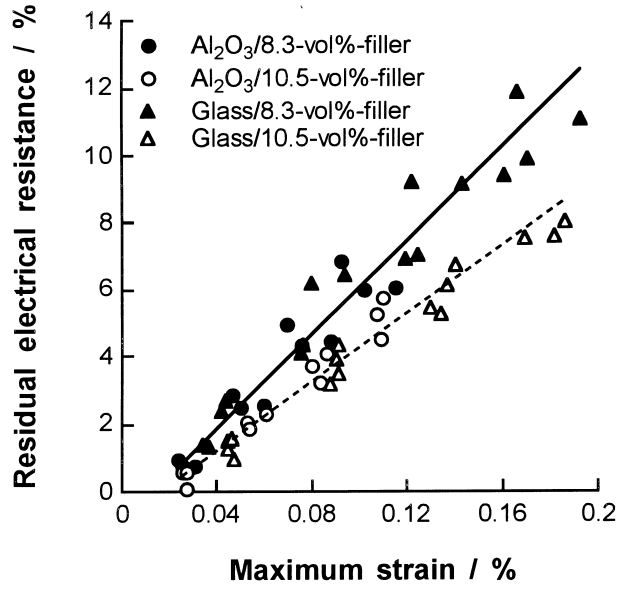

Fig. 8. Relationship between maximum strain applied in the past and changes in residual electrical resistance after removing load for Al_2O_3 and glass coated with composite containing 8·3 and 10·5 vol% of fillers.

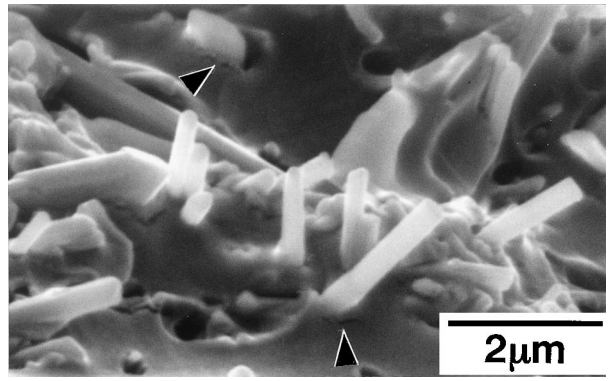

Fig. 9. Scanning electron micrograph for fracture surface of the composite film containing 10.5 vol% of fillers. The arrows indicate a microcrack in the matrix polymer near the pull-out filler.

deformation of matrix which holds the pull-out filler is clearly observed as indicated by arrows in this figure. These results indicate that relatively strong mechanical interactions between matrix and fillers exist in this system, and the interfacial shear strength is larger than the matrix shear strength. This is probably due to high adhesive strength of epoxy and large friction force at the polymer/filler interfaces resulting from the rough surface of filler as shown in Fig. 1(b). On the other hand, filler break, microcracks and interface debonding between matrix and fillers were not observed in the surface of composite film in samples to which the cyclic load was applied. It is considered that these results agree with the above results of microfracture observation.

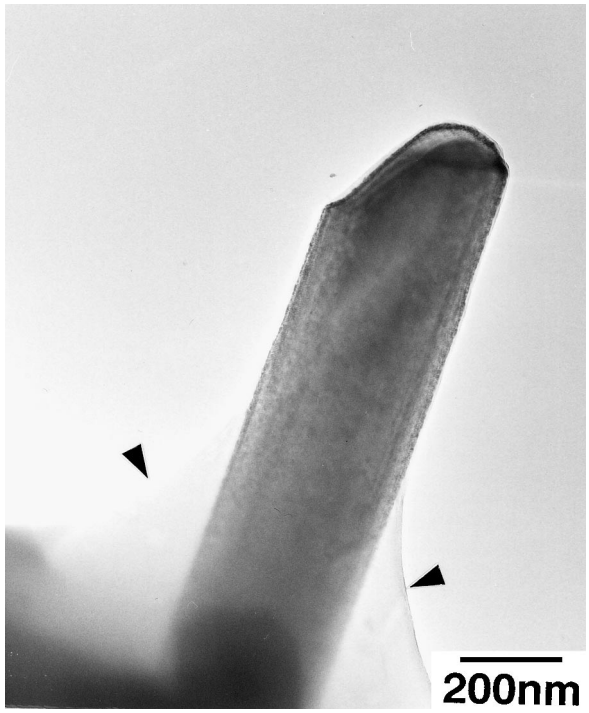

Fig. 10. Bright-field transmission electron micrograph for fracture surface of the composite film. The arrows indicate plastic deformation of matrix polymer near the pull-out filler.

3.5 Mechanism of resistance increase on tensile strain

From the experimental results and microscopic observations we can conclude that the resistance change by applied stress is related to microscopic deformation of polymer matrix near the contact point of fillers, rather than to filler breaking and/or the arrangement change of filler resulting from fracture of filler/matrix interface. Accordingly, the mechanism of the resistance increase of the composite by applied stress is considered to be explained by the following, as shown in Fig. 11. As mentioned in the experimental procedure, since the composite film was set on the lower side of specimens, tensile stress to films should be induced by bending load. At first, under tensile stress field, a microscopic arrangement change in the fillers occurs resulting from deformation of the matrix (See stage 1 in Fig. 5) and some of the continuous conduction path is disrupted at this time. Accordingly, electrical resistance of composites increases.

3.6 Mechanism of residual resistance according to the maximum strain

In this composite, no macroscopic permanent strain was observed after unloading. If the filler arrangement came back to the initial one, residual resistance should not be observed. Then, the

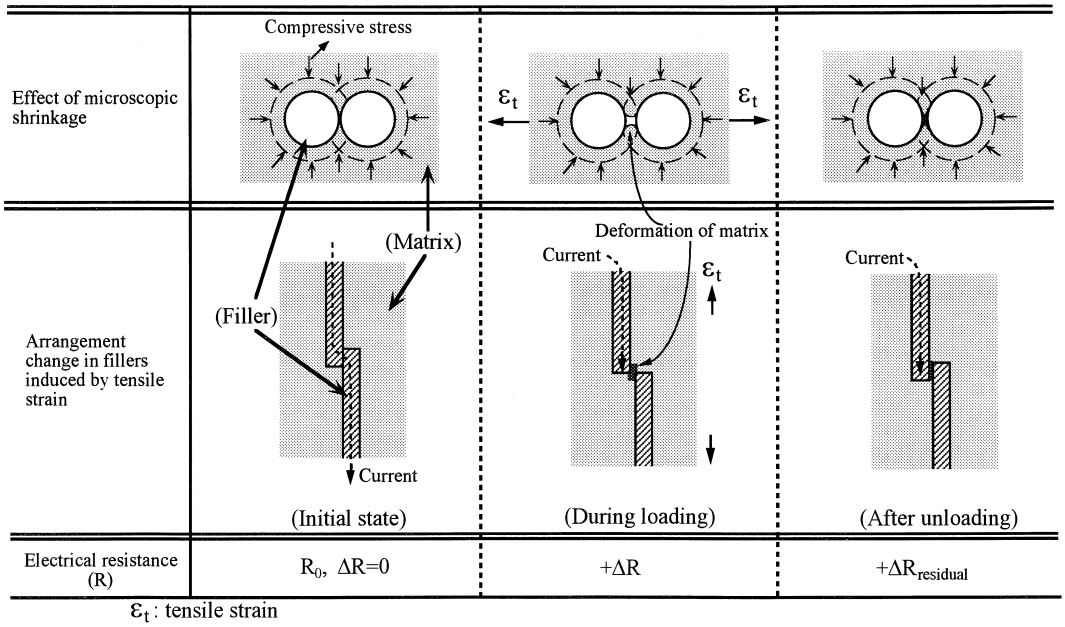

Fig. 11. Schematic model of mechanism for the resistance change by applied stress in electroconductive polymer composites.

contact resistance between fillers increased according to the rearrangement of fillers in the present case. Filler rearrangement without residual macroscopic strain can be explained by a residual microscopic strain as follows. At the first stage of composite preparation, the fillers are soaked in liquid resin, which then polymerizes around the fillers to form a rigid solid. During this curing process the resin sticks to the fillers, at the same time, it also shrinks during curing and cooling processes. Hence, we must consider the effect of the shrinkage of matrix during curing on the individual fillers; the compressive stress from matrix to fillers has been induced by the shrinkage.[27] Namely, when the contact of fillers is disrupted under tensile stress, release of the compressive stress toward its equilibrium state takes place and this state change leads to the microscopic deformation of the matrix near the contact point of fillers as shown in Fig. 11. During removal of the load, resistance decreases with decreasing strain because of some filler contacting within the matrix (See stage 2 in Fig. 5). However, because of microscopic residual deformation of the matrix near some contact point of fillers, geometrical arrangement of filler restrained by matrix has changed from its initial state (See stage 3 in Fig. 5). Accordingly, residual resistance is observed after removing load.

4 Conclusions

In the present study, strain detection in structural ceramics coated with polymer composite containing needle-like electrically conductive filler has been investigated by measuring electrical resistance during and after loading. Electrical resistance increased with increasing strain. The degree of electrical resistance change versus strain became large when the filler volume fraction was close to the critical percolation fraction. Residual electrical resistance was observed after removing load, and these changes were dependent on the maximum strain once applied. The behavior of residual resistance change of composite coated on Al_2O_3 was also identical to that on glass when the filler concentration was the same. These results suggests that estimation of maximum strain applied in the past could be applicable to a wide variety of ceramics. The rearrangement of fillers resulting from microscopic deformation of matrix induced by releasing of residual compressive stress is considered to be a predominant mechanism of residual resistance. The measurement of electrical resistance in structural ceramics coated with electroconductive composite is considered to be useful for strain detection.

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