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Fabrication and evaluation of PZT/Ag composites and functionally graded piezoelectric actuators

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Abstract Metallic Ag as the second phase was added into PZT ceramic matrix to fabricate piezoelectric composites and functionally graded actuators by gradually altering Ag concentration, aimed to improve mechanical properties and to solve possible interfacial debonding usually observed in conventional bimorph-type piezoelectric actuator. The PZT/Ag composites were obtained by directly co-firing PZT and Ag powders at 1200°C for 1 h. The fracture strength σ_f and fracture toughness K_{IC}, as well as the corresponding piezoelectric properties, were firstly evaluated upon the PZT/Ag composites for Ag concentrations of 0–30 vol%. The mechanical properties for the PZT/Ag composites were found to be greatly enhanced compared with pure PZT ceramics: from 69 to 129 MPa for σ_f and from 1.0 to 3.7 MPa.m$^{1/2}$ for K_{IC}. With increasing Ag concentration, the piezoelectric constant d_{33} of PZT/Ag composites was found to decrease from 419 to 86 pC/N. Then, a functionally graded actuator was fabricated and evaluated in terms of electric-induced curvature k. The PZT/Ag FGM actuator with size of 12 mm × 3 mm × 1 mm has a curvature k of 0.03–0.17 m^{-1} that corresponds to applied voltages of 100–500 V. A comprehensive comparison was made on the mechanical property enhancements by the metal particles dispersion, and the bending displacements produced by the FGM actuators between the PZT/Ag and previously fabricated PZT/Pt systems.

Keywords PZT/Ag · Functionally graded materials (FGMs) · Piezoelectric actuator · Electric-induced displacement

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1 Introduction

Piezoelectric actuators are mechanically and electrically loaded in their performance. It is important to note that the mechanical properties of actuators should be improved to sustain alternate bending induced by the application of a high-frequency electric field [1]. The dispersion of metallic phase is an effective way of toughening the piezoelectric ceramics. Furthermore, the corresponding electrical properties of the resultant piezoelectric/metal composites could be adjusted by the metallic-phase dispersion [2]. Piezoelectric actuators could be designed to bend harmoniously under an electric field applied through the fabrication of a functionally graded microstructure (FGM) that corresponds to graded electrical properties. As a result, interface debonding that is frequently observed in a conventional bimorph actuator could be avoided [3–5]. However, base metals are usually difficult to co-fire with piezoelectric ceramics, e.g. PZT. On the one hand, the metal is easy to oxidize during the sintering of the PZT/Metal composites in the atmosphere; On the other hand, the piezoelectric properties will lost much if the composites are sintered in low oxygen pressure in order to prevent metal oxidation. PZT/Pt composites were reported to fabricate successfully by co-firing inert metal Pt with PZT at 1200°C in the atmosphere [2]. PZT/Pt FGM actuators were further fabricated and evaluated, and these actuators possessed good electric-induced displacements comparable to conventional bimorph actuators [6].

It was reported that PZT/Ag composites could also be fabricated by a conventional sintering process owing to the decomposition of the Ag oxidation at higher temperatures (>200°C) [7–9]. Although those investigations [7–9] deal with the piezoelectric properties of the PZT/Ag composites, PZT/Ag FGM actuators have not been conducted yet. Taking into consideration the fact that Ag is much less expensive than Pt, we can fabricate the PZT/Ag FGM actuators and evaluate their electric-induced displacement based on the corresponding electrical and mechanical properties of the PZT/Ag composites in this paper. Further investigations should be devoted to the co-firing of base metals with piezoelectric ceramics, so as to reduce the fabrication costs while enhancing the performance reliability of the PZT/Metal FGM piezoelectric actuators.

2 Experimental procedure

2.1 Sample preparation

The starting materials were commercially available PZT powder (Zr/Ti atomic ratio = 0.516/0.484, average particle size: 0.97 μm, PZT-LQ, Sakai Chemical Industry, Japan) and Ag powder (average particle size: 2 μm, purity: 99.9%, High Purity Chemicals, Japan). In the range of 0–30 vol%, the Ag powders as the metal filler were added into the PZT matrix to fabricate PZT/Ag piezoelectric composites. The powder mixtures were die pressed in a Φ16 mm mold at 100 MPa, and then cold isostatically pressed (CIPed) at 200 MPa. The pure PZT, PZT-5 vol%Ag and PZT-15 vol%Ag materials were selected to fabricate PZT/Ag FGMs, by stacking the three materials layer-by-layer with an increasing Ag concentration in the mold. The thickness of every compositional layer was kept the same at approximately 0.33 mm. The resultant powder compacts for the PZT/Ag composites or FGMs were sintered at 1200°C for 1 h in a covered Al_2O_3 crucible containing $PbZrO_3$ powders to protect severe Pb loss. Batches of samples, 13 mm in diameter and 1 mm in height, were obtained.

2.2 Characterization

Scanning electron microscopy (SEM, JSM-6460LV, JEOL, Japan) was used to observe the microstructures of the PZT/Ag composites, and optical microscopy (OM, BX60, Olympus, Japan) was used to inspect the compositional profiles of the PZT/Ag FGMs. To evaluate electrical properties, both surfaces of a batch of composite samples were coated with Ag pastes and baked at 650°C for 30 min to form the Ag electrodes. The Ag-pasted samples were poled at 120°C for 10 min in a bath of silicone oil under dc electric fields of 10–20 kV/cm. The piezoelectric constant d_{33} was measured by using a quasi-static piezoelectric d_{33} meter (ZJ-3A, Institute of Acoustic, China). The fracture strength σ_f of the PZT/Ag composites was measured by a modified small punch (MSP) testing method [10]. A batch of composite samples were firstly sawed to the dimensions of Φ10 mm × 0.5 mm, with one surface well polished, and then loaded in a MSP jig. The fracture toughness K_{IC} of the PZT/Ag composites was evaluated by the Vickers indentation microstructure (IM) technique. A batch of composite samples were polished with 1 μm diamond paste to achieve a mirror-like finish, and then loaded with loadings from 9.8 to 196 N for different composites, ensuring that the ratio of the initiated crack length to the half-length of the indentation diagonal is larger than 1.5 [11].

To evaluate the electric-induced displacement of the PZT/Ag FGM actuators, the FGM samples were cut to beams with dimensions of 12 mm × 3 mm × 1 mm, and two strain gages were mounted onto the two 12 mm × 3 mm surfaces. The two strains of ε_1 and ε_2, produced under applied voltages (100–500 V), were converted into curvature k by using the following equation [6]:

$$k = \frac{\varepsilon_1 + \varepsilon_2}{1 + \varepsilon_1} d_t^{-1} \tag{1}$$

where d_t is the thickness of the actuator along the direction of the electric field applied, and d_t is equal to 1 mm in this study.

3 Results and discussion

Figure 1 shows the SEM observations of the PZT/Ag composites sintered at 1200°C in this study. Near-circular silver particles were distributed in the PZT matrix, and relatively dense microstructures were obtained. Some pull-outs produced during the preparation of the SEM samples were also observed in Fig. 1. It should be noted that a quantity of silver diffused to the outer surface of the samples during sintering, when Ag concentration was higher than 20 vol% [9]. The large diffusing rate of the Ag atoms accounts for the Ag loss in the PZT/Ag composites [7, 12]. The attempt to sinter the PZT/Ag composites at a lower temperature of 900°C, unfortunately, produced relatively porous microstructures [13].

Figure 2 shows the measured piezoelectric constant d_{33} for the PZT/Ag composites sintered at 1200°C for 1 h. The d_{33} value decreases with increasing Ag concentration up to 22.5 vol%; above this the d_{33} value begins to increase gradually. The decrease in the d_{33} value with increasing Ag concentration is expected because Ag is nonpiezoelectric. However, the

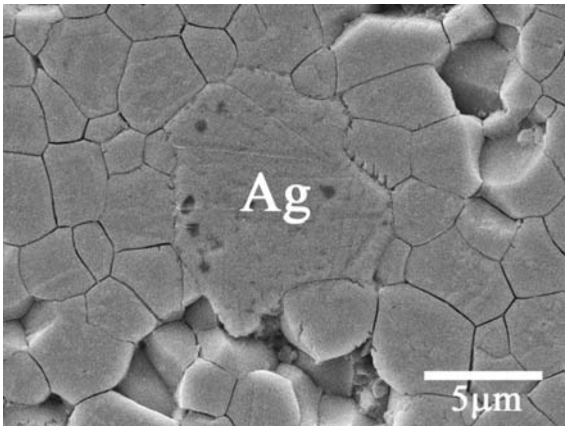

Fig. 1 SEM image showing microstructures of the PZT/Ag composites

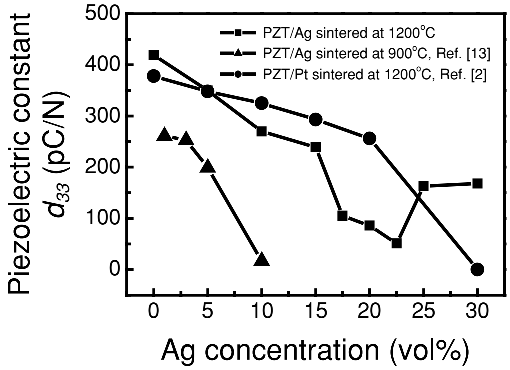

Fig. 2 The piezoelectric constant d_{33} as a function of Ag concentration for the PZT/Ag composites sintered at 1200°C for 1 h. The reported d_{33} for the PZT/Ag composites sintered at 900°C for 4 h [13] and for the PZT/Pt composites sintered at 1200°C for 1 h [2] are also given for comparison

quantitative influence of the Ag particle dispersion on piezoelectric properties of the PZT/Ag composites is still difficult to determine. The increase in d_{33} value after 22.5 vol% could be explained as follows: As previously mentioned, the loss of Ag becomes pronounced when Ag concentration is higher than 20 vol%. The actual Ag content in the 25 vol% or 30 vol% samples could be much less than their corresponding nominal compositions and further less than the Ag content in the 22.5 vol% sample. Because the d_{33} value increases with decreasing Ag concentration, the 22.5 vol% composite has a minimum d_{33} value. Since the Pt is stable in PZT at the same sintering temperature of 1200°C, this two-region characteristic is not observed in PZT/Pt composites. A comparison of the d_{33} value was made among the present materials, the PZT/Ag composites formerly sintered at 900°C [13], and the PZT/Pt composites sintered at 1200°C for 1 h [2]. When Ag concentration is 10 vol%, the d_{33} value is nearly zero for the PZT/Ag composites sintered at 900°C, whereas the d_{33} value is still as high as 270 pC/N for the PZT/Ag composites sintered at 1200°C. Compared with the PZT/Pt system, the d_{33} value of the PZT/Ag composites decreases more rapidly with increasing metal concentration. The dielectric and ferroelectric properties for the PZT/Ag composites in this study can be seen in a previous publication [9].

Figure 3 shows the fracture strength σ_f of the PZT/Ag composites. The σ_f value increases with increasing Ag concentration up to 15 vol%. Compared with pure PZT ceramics, the fracture strength obtained for the PZT-15 vol%Ag composite is almost doubled. It indicates that the metallic Ag dispersion has enhanced the fracture strength of the PZT/Ag composites. Although the Ag loss exists in the composites with a higher Ag concentration (>15 vol%), there is no strong dependence of fracture strength on Ag concentration. For comparison, the fracture strength of the PZT/Pt composites was also given in the same concentration range [2]. At the same metal concentration, the σ_f enhancement of the PZT/Ag composites is larger than that of the PZT/Pt composites. The explanation could be related to the difference in metallic microstructures or essential mechanical properties between the Ag and Pt.

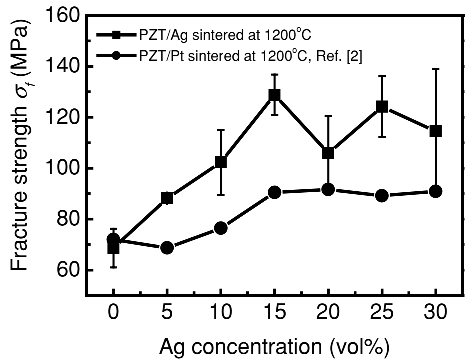

Fig. 3 The fracture strength σ_f as a function of Ag concentration for the PZT/Ag composites sintered at 1200°C for 1 h

Figure 4 shows the fracture toughness K_{IC} of the PZT/Ag composites. The variation tendency of fracture toughness is similar to that of the fracture strength of the PZT/Ag composites. The maximum K_{IC} is 3.7 MPa.m$^{1/2}$ for the PZT-20 vol%Ag composite, which is nearly 4 times that of the K_{IC} of pure PZT ceramics. The decrease in K_{IC} after 20 vol% Ag concentration is also attributed to the Ag loss due to Ag diffusion, because the Ag loss becomes more noticeable when Ag concentration is higher than 20 vol% and leads to the decrease in actual Ag content. The fracture toughness of the PZT/Ag composites is higher than that of the PZT/Pt composites [2] at the same metal concentration, which could be explained by the fact that Ag is more ductile than Pt.

Figure 5 shows the compositional profile of the PZT/Ag FGM with the three Ag concentrations of 0 vol%, 5 vol% and 15 vol%. The thickness of each compositional layer is about 0.33 mm, and the total thickness of the FGM is nearly 1 mm. It can be found that the Ag particles are uniformly

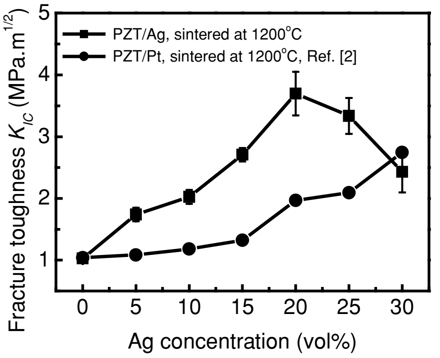

Fig. 4 The fracture toughness K_{IC} as a function of Ag concentration for the PZT/Ag composites sintered at 1200°C for 1 h

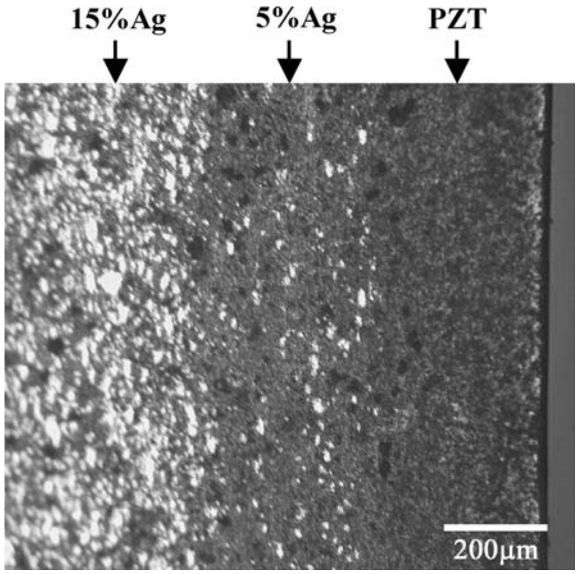

Fig. 5 Optical micrograph of a single FGM for the PZT/Ag piezoelectric actuator

distributed in the PZT matrix, and no defects such as cracks are observed in the interfaces between two neighboring layers. We attempted to fabricate the symmetrical PZT/Ag FGM actuators by using the PZT-30 vol%Ag composite as a central electrode. However, it was found that the PZT-30 vol%Ag layer became nonconductive due to Ag loss during sintering. Therefore, only the electric-induced displacement of the single FGM actuator shown in Fig. 5 was evaluated in the present study.

The curvature k was selected as the expression of the electric-induced displacements. In fact, the measured curvature k can be generally converted into the end displacement of an actuator, when the actuator is installed into a cantilever. The evaluation of the end displacement is left for future study. Figure 6 shows the electric-induced displacements of the PZT/Ag FGM actuators based on Eq. (1), and experimental results for the PZT/Pt FGM actuators [6]. In order to compare the electric-induced displacements of conventional bimorph actuators with those of the FGM actuators, an analytical calculation, based on a modified classical lamination theory (CLT), was conducted on a conventional bimorph actuator that consists of two PZT layers of 0.9 mm and a central electrode layer of 0.2 mm in thickness [6]. As shown in Fig. 6, the k value increases with increasing applied voltage. This curvature is similar to that produced by conventional bimorph actuators, especially at the applied voltages lower than 400 V. However, much performance reliability of the FGM actuator is achieved due to mechanical property enhancements by metal dispersion. In comparison, the curvature produced by the PZT/Ag FGM actuators [6] is very close to that produced by the PZT/Pt FGM actuators at given applied voltages, despite the fact that the PZT/Ag actuator is a single FGM of 1 mm in thickness, while the PZT/Pt actuator is a symmetric FGM of 2 mm in thickness. In fact, the bending principle is different for the single and symmetric FGM actuators. The PZT/Ag composites would produce expansion or shrinkage in relation to their piezoelectric properties. Therefore, the produced strains will distribute in a graded profile along the thickness of a single FGM, and thus a bending occurs, owing to the strain mismatch in the thickness direction. The bending of a symmetric FGM actuator is similar to a conventional bimorph actuator. The poling direction of the top FGM is opposite to that of the bottom FGM. When the same electric field is applied, the top FGM shrinks (or expands), whereas the bottom FGM expands (or shrinks). Therefore, a bending in the thickness direction occurs in the symmetric FGM actuator. To acquire the PZT/Ag symmetric FGM actuators, a central electrode layer should be incorporated with the top and bottom FGMs through other methods, and this will be further explored later.

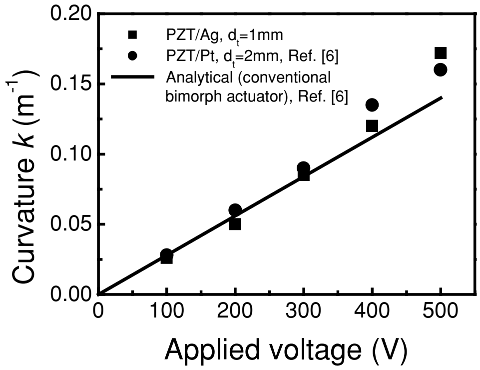

Fig. 6 The curvature produced by the FGM actuator as a function of applied voltage

4 Conclusions

The PZT/Ag composites were fabricated by directly co-firing PZT and pure Ag powders at 1200°C for 1 h. As expected, the mechanical properties of the PZT/Ag composites were greatly enhanced through Ag particle dispersion, and the addition of Ag into PZT could adjust the piezoelectric properties of the PZT/Ag composites to a graded profile. These two property aspects are both important to the application of the PZT/Ag composites for the FGM actuators. A PZT/Ag single FGM actuator was fabricated and the corresponding electric-induced displacements were evaluated in terms of curvature k. The curvature produced by the PZT/Ag single FGM actuator was found to be comparable to that produced by conventional bimorph actuators, and was also comparable to that produced by PZT/Pt symmetric FGM actuators. The PZT/Ag FGM actuators suffered somewhat from the Ag diffusion during sintering.

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