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Reliability enhancement in nickel-particle-dispersed alkaline niobate piezoelectric composites and actuators

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Abstract

Piezoceramics and related devices are naturally fragile. Here we have produced piezoelectric composites of Ni-particle-dispersed Li-doped (K,Na)NbO_3 (LKNN/Ni), and used these composites to make actuators with functionally graded microstructure (FGM). The mechanical properties of the LKNN/Ni composites were greatly enhanced by the incorporation of Ni particles. An LKNN–20%Ni composite exhibited double the fracture toughness of the monolithic LKNN ceramic. An LKNN/Ni piezoelectric FGM actuator subjected to a large number of cycles of electrical loading still maintained acceptable driven performance. The enhancement of reliability in the LKNN/Ni composites and actuators is attributed to the uniformly dispersed Ni particles in the piezoceramic matrix. Deformation of these Ni particles absorbs the energy of crack propagation and thus greatly strengthens the composites and devices. These new FGMs contribute further to the reliability enhancement of piezoelectric actuators.

Keywords: Sintering; Composites; Toughness and toughening; Niobates; Actuators

1. Introduction

Piezoceramics have been widely used in piezoelectric actuators that undertake repeated bending at high frequency.[1] For actuator applications, the piezoceramics are required to have both high piezoelectric constants and good mechanical properties. Unfortunately, piezoceramics are inherently fragile, which is a problem for actuator applications. A bimorph-type actuator, consisting of two ceramic plates and a central metal shim bonded together with an organic glue, is one of most widely used piezoelectric actuators. During service, interface stresses and interface debonding are usually observed in this actuator due to the connection of two dissimilar materials – ceramic and metal.[2]

Recently, metal-particle-dispersed piezoceramic composites of Pb(Zr,Ti)O_3/Pt (PZT/Pt)[3] and PZT/Ag[4] have been developed to solve the above-mentioned problem in the bimorph actuator. When the metal loading in the composite alters gradually with distance, the piezoelectric properties are tailored to form a functionally graded microstructure (FGM)[5,6] to replace the bonding interfaces between the ceramic plate and metal shim. Due to the harmonious strains produced by the various FGM layers, interfacial stresses are greatly reduced. More importantly, the dispersed metal particles have been demonstrated to greatly increase the mechanical properties of the piezoceramic matrix.[3,7] It is clearly shown that this piezoceramic/metal FGM structure is able to enhance the reliability of bimorph-type actuators.[8,9]

One may argue that the piezoelectric properties will be sacrificed when metal particles are incorporated into a piezoceramic matrix,[3,4] an apparent disadvantage of metal-particle-dispersed piezoceramic composites. Actually, the inhomogeneous strains produced by the various FGM layers can lead to a considerable amount of out-of-plane displacement along the thickness of the actuator.[8,9] This structure is specifically applicable to some cantilever-type devices, such as piezoelectric relays or piezoelectric valves. Theoretical calculations have revealed that electric-field-induced displacements of a piezoelectric FGM actuator are comparable to a conventional bimorph-type actuator,[9] and the mechanical performance of the FGM-based device is greatly enhanced at the same time.

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In this study, we focus on mechanical property enhancement in the metal-particle-dispersed piezoceramic composites. A number of recent papers[10–12] have investigated domain switching toughening in ferroelectric ceramics. Stress concentration can induce switching of ferroelectric domains, leading to energy consumption and prevention of microcrack initiation in ferroelectric ceramics.[10] In a metal-particle-dispersed ferroelectric ceramic, once the inevitable microcracks are produced during cycling[13] the metal particles continue to absorb the energy of crack propagation. This mechanism can prevent the initiated microcracks from developing to macrocracks and hence enhance the reliability of ferroelectric ceramics and related devices.[7]

We have previously produced Ni-particle-dispersed Li-doped (K,Na)NbO_3 (LKNN/Ni) composites *via* sintering in a protective atmosphere, and identified the effect of Ni particles on the electrical properties.[14] Compared with reported PZT/Pt[3] and PZT/Ag,[4] this new material system is a combination of lead-free LKNN piezoceramics and base metal Ni. In this work, we investigate the mechanical property enhancement by Ni particles dispersed in the composites. A piezoelectric FGM actuator is then produced using the LKNN/Ni composites and is subjected to extensive cycles of electrical loading so as to evaluate the reliability of piezoelectric devices containing the composites.

2. Experimental

The mixed oxide method was employed to synthesize $[Li_{0.06}(K_{0.5}Na_{0.5})_{0.94}]NbO_3$ (LKNN) lead-free piezoceramics.[15] Ni powders (purity 99.0%, particle size 44 μm, J&K Chemical, China) were used as the metal filler to fabricate LKNN/Ni composites with Ni contents of 0–50 vol%. The powder compacts were sintered at 1100 °C for 2 h under industrial nitrogen gas (oxygen content ∼0.5%). Experimental details of the production of LKNN/Ni composites can be found elsewhere.[14]

Sample density of the LKNN/Ni composites was measured by the Archimedes method. The microstructure was observed by optical microscopy (OM, LEICA DMR, Germany) and scanning electron microscopy (SEM, Cambridge S-360, UK). The electrical conductivity of the LKNN/Ni composites was measured by the four-probe method. The fracture toughness of the LKNN/Ni composites was evaluated by the Vickers indentation fracture (VIF) method using a micro-hardness tester (MH-6, China). Five indents were made on each specimen, and three specimens were used for each Ni content. The indentation load applied was 49 N for each Ni content, so as to meet the prerequisite of $c/a > 2.5$, where a is the half length of the indentation and c is the length between the indentation center and crack tip. The dwell time was 15 s. The Young's modulus E values for the LKNN/Ni composites were calculated from simplified mixture laws,[16] using 207 GPa as the E value of metallic Ni and 76 GPa (the reported value for Li/Ta-codoped (K,Na)NbO_3 ceramics[17]) as the E value of monolithic LKNN ceramic. Two cases of mixture laws were considered to give upper (E_U) and lower (E_L) bounds for the predicted Young's modulus:

$$E_U = E_1 V_1 + E_2 V_2 \tag{1}$$

assuming that two phases in a biphasic composite have the same strain; and

$$\frac{1}{E_L} = \frac{V_1}{E_1} + \frac{V_2}{E_2} \tag{2}$$

assuming that the two phases have the same stress, where E_1 and E_2 are the Young's moduli of the two phases, and V_1 and V_2 are the volume fractions of the two phases.

The powder metallurgy route was employed to produce LKNN/Ni piezoelectric FGM actuators. Firstly LKNN/Ni composites with various Ni contents were stacked layer-by-layer in a Ø20 mm mold and sintered at a lower temperature of 1060 °C to give cylindrical samples. Then laminated beams with approximate dimensions of 16 × 3 × 2 mm were cut from the cylindrical samples and made into bimorph-type FGM actuators by applying silver electrodes and silver wire connections. Electric-field-induced displacements of the produced actuators were evaluated in terms of curvatures using a strain-gage method. The experimental details of the production of LKNN/Ni FGM actuators and the evaluation of electric-field-induced curvatures can be found elsewhere.[18]

3. Results and discussion

3.1. LKNN/Ni piezoelectric composites

We have previously investigated the effect of sintering atmosphere on the phase structure of Ni-particle-dispersed LKNN composites.[14] The results demonstrate that LKNN/Ni composites can be successfully produced in an industrial nitrogen gas with an oxygen content of 0.5%, corresponding to an oxygen partial pressure of $pO_2 = 5 \times 10^2$ Pa. The dispersed Ni particles are not oxidized and the LKNN phase maintains its perovskite structure. Fig. 1 shows representative microstructures of these LKNN/Ni composites. The Ni particles were found to uniformly disperse in the LKNN ceramic matrix (Fig. 1a). The size of Ni particles ranged from 30 to 60 μm, close to the starting size of 44 μm, which is consistent with good dispersion of the Ni second-phase in the LKNN matrix. The Ni particles were closely bonded with the LKNN matrix (Fig. 1b).

Sample densities measured for the LKNN/Ni composites varied from 4.21 to 4.81 g/cm^3 with increasing Ni content from 0 to 20 vol%. Relative densities of the LKNN/Ni composites were derived by assuming 4.51 g/cm^3 for the theoretical density (TD) of $(K_{0.5}Na_{0.5})NbO_3$[19] as the TD of LKNN and 8.90 g/cm^3 as the TD of Ni and by using the mixture law to calculate the TDs of LKNN/Ni composites. Fig. 2 shows the results of derived relative densities. It can be seen that all the LKNN/Ni composites possessed 90–95% of theoretical densities, giving rather dense microstructures.

An attractive aspect of metal-particle-dispersed piezoceramic composites is enhanced mechanical properties. Fig. 3 shows the fracture toughness of the LKNN/Ni composites as evaluated by the VIF method. Two sets of Young's moduli derived from Eqs. (1) and (2) respectively were used to calculate the fracture toughness, giving the upper and lower bounds of the K_R values. Ni addition markedly increased the fracture toughness in compari-

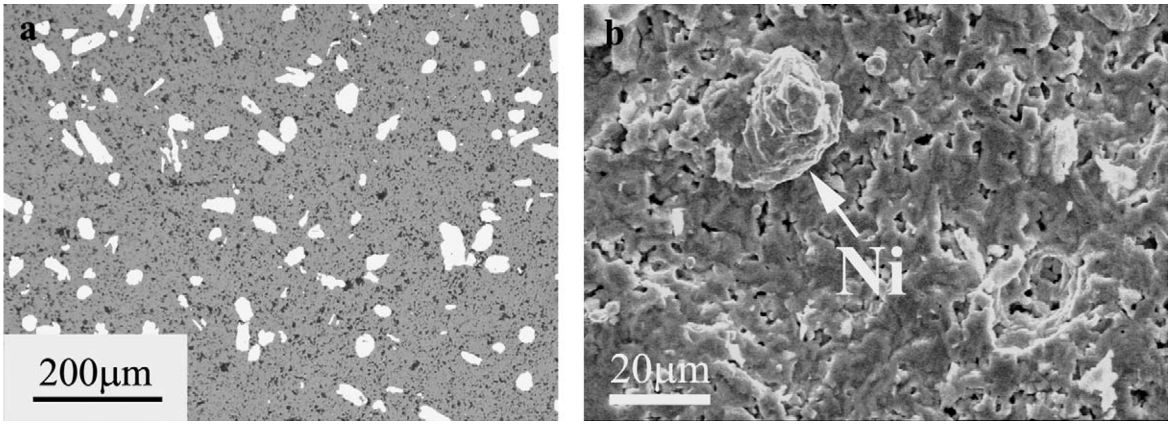

Fig. 1. Microstructures showing the distribution of Ni particles in an LKNN–10%Ni composite: (a) optical micrograph of a polished surface and (b) SEM image of a fractured surface.

son with monolithic LKNN ceramics. The results are consistent with those observed in Ag-particle-dispersed PZT composites.[7] The K_R value was characterized as a function of Ni content according to the Anstis model.[20] The LKNN–20%Ni composite

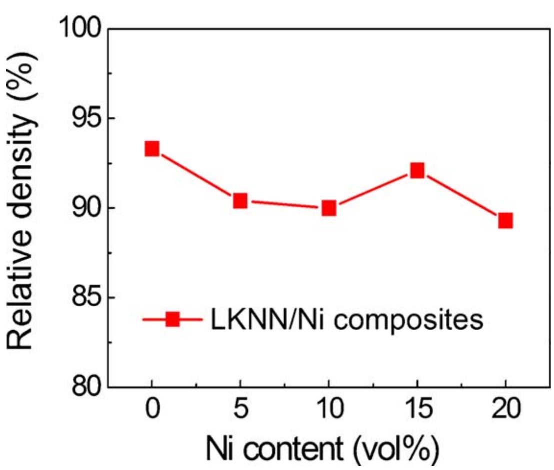

Fig. 2. Relative density as a function of Ni content for the LKNN/Ni composites.

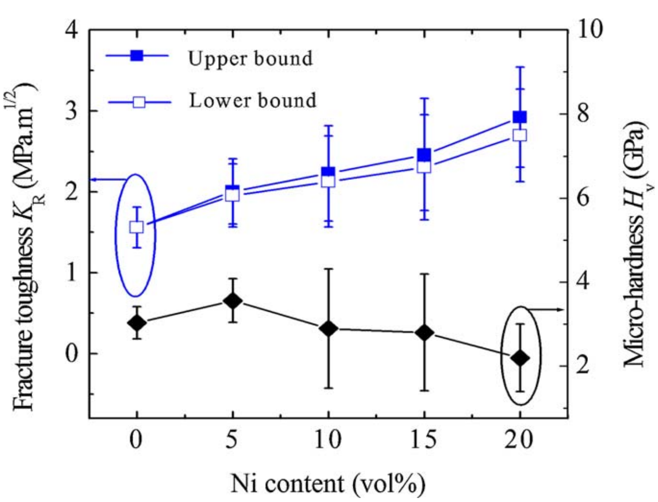

Fig. 3. Dependence of Vickers micro-hardness and fracture toughness of LKNN/Ni composites upon Ni content in the range of 0–20 vol%. The Anstis[20] model has been used to calculate the fracture toughness.

had a fracture toughness almost twice that of monolithic LKNN ceramic, yielding a K_R value of 2.92 MPa.m$^{1/2}$. As shown in Fig. 3, the micro-hardness H_v of the composites decreased with Ni addition, owing to the softness of metallic Ni (H_v = 0.63 GPa). Although there is argument about the validity of applying the VIF method to measure fracture toughness,[21] this method has been widely utilized to evaluate quantitatively the fracture toughness of brittle materials. The VIF-evaluated fracture toughness indeed reflects a resistance to crack propagation for materials. Here we emphasize this meaning by denoting the VIF-evaluated fracture toughness as K_R, where "R" represents "resistance".

The enhancement of fracture toughness in the LKNN/Ni composites should originate from the dispersion of metallic Ni particles. The Ni particles and added interfaces will absorb the energy that is required for crack propagation, thus toughening the LKNN ceramic matrix. This is clearly supported by the observed microstructures. Fig. 4 shows the VIF-induced crack profiles observed in monolithic LKNN ceramic and LKNN/Ni composite. For comparison, the crack in the LKNN–20%Ni composite (Fig. 4b) was shorter than that in monolithic LKNN ceramic (Fig. 4a), although the indentation loads applied to the composite and the monolithic ceramic were the same. This finding implies that some fracture energy has been absorbed by deformation of Ni particles. The crack paths were also different – the crack extended in a straight manner in monolithic LKNN ceramics, while crack deflection was observed several times when the crack tip met an Ni particle in the LKNN–20%Ni composite. In LKNN/Ni composites, the increase in crack length due to crack deflection means that more fracture energy is consumed before ultimate failure.

Quantitative evidence for the enhancement of fracture toughness is provided by the dependence of VIF-induced crack length on Ni content, where the same indentation load of 49 N was applied for each Ni content, shown as in Fig. 5. The length of cracks used for K_R characterization decreased gradually with increasing Ni content. This clearly demonstrates that Ni particles prevent the propagation of cracks and toughen the composites. Characterization of R-curve behavior[10] will be helpful for further understanding of reliability of LKNN/Ni composites, and will be the subject of future study.

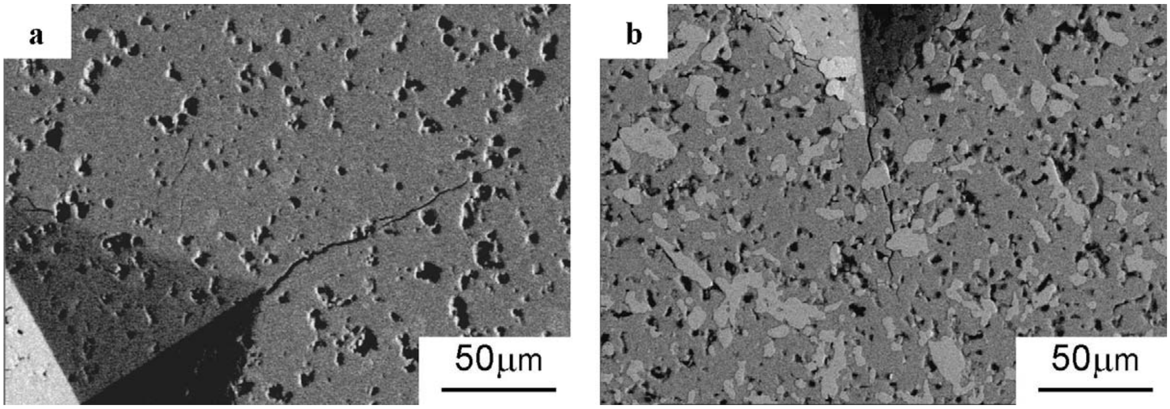

Fig. 4. SEM images showing the propagation of VIF-induced cracks in (a) monolithic LKNN ceramic and (b) LKNN–20%Ni composite.

3.2. LKNN/Ni piezoelectric FGM actuators

To produce a bimorph-type actuator, a composition with properties appropriate for a central electrode must be chosen. Hence, the electrical conductivities of the LKNN/Ni composites were characterized, and the results are shown in Fig. 6. The conductivities were found to increase dramatically with increasing Ni content and reach a level of 0.1 S/m above 40 vol% added Ni. In this study, the LKNN–40%Ni composite was selected as the inner electrode for multilayered piezoelectric actuators.

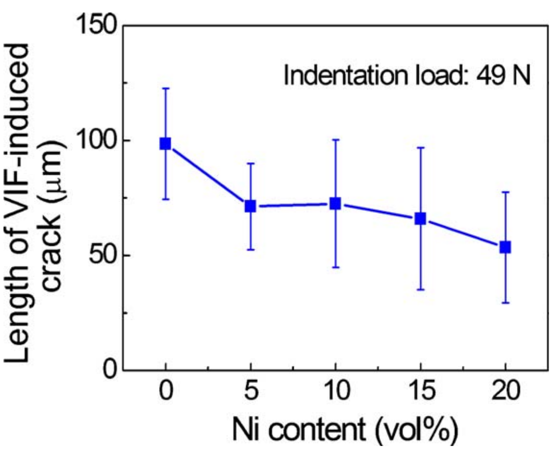

Fig. 5. Length of VIF-induced cracks (measured from indenter corner to crack tip) for the various LKNN/Ni composites applied by the same indentation load of 49 N.

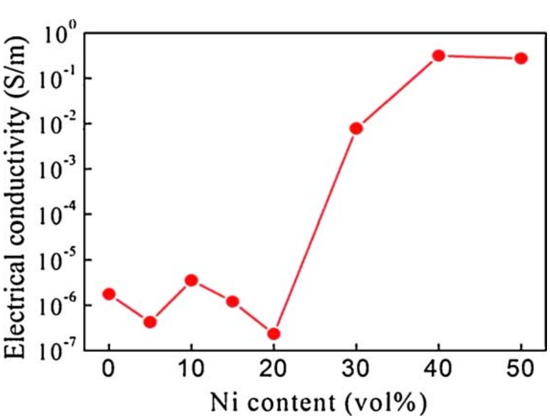

Fig. 6. Dependence of electrical conductivity of the LKNN/Ni composites upon Ni content in the range of 0–50 vol%.

Fig. 7 shows the microstructure of a LKNN/Ni piezoelectric FGM actuator produced by the powder metallurgy method. This bimorph-type actuator consisted of five layers, with the LKNN–40%Ni composite acting as the central electrode. The Ni content increased towards the central layer, giving a symmetrically graded compositional profile. The LKNN–40%Ni layer was about 0.4 mm thick, the LKNN–10%Ni was 0.3 mm thick and the thickness of the monolithic LKNN layers was 0.5 mm. Sound bonding between the layers could clearly be seen at the interfaces.

For electric-field-induced curvature measurements, the actuators were poled along the whole thickness. The right-half FGM was supplied with an electric field parallel to the poling direction, and the left-half FGM with an opposite electric field. As a result of these applied electric fields, the right-half FGM will expand and the left-half FGM will shrink. This will lead directly to a bending deformation along the thickness of the actuator, which can be measured in terms of out-of-plane curvature. Previous studies[14] show that the piezoelectric constant d_{33} of LKNN/Ni composites decreases gradually from 165 to 23 pC/N as the Ni content increases from 0 to 20 vol%. For a given applied electric field, the LKNN/Ni composites will expand or shrink in accordance with their individual piezoelectric properties. Therefore, the strains produced will be distributed in a graded profile along the thickness of an FGM. As shown in Fig. 7, this will lead

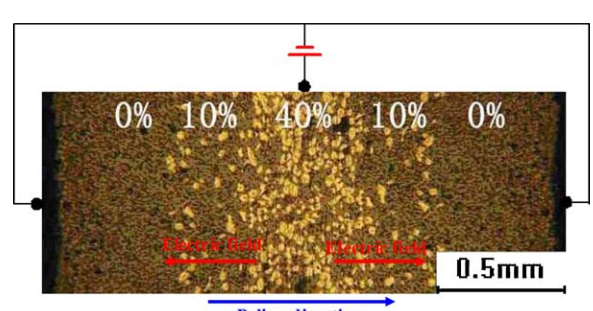

Fig. 7. Optical micrograph showing the microstructure of an LKNN/Ni piezoelectric FGM actuator.

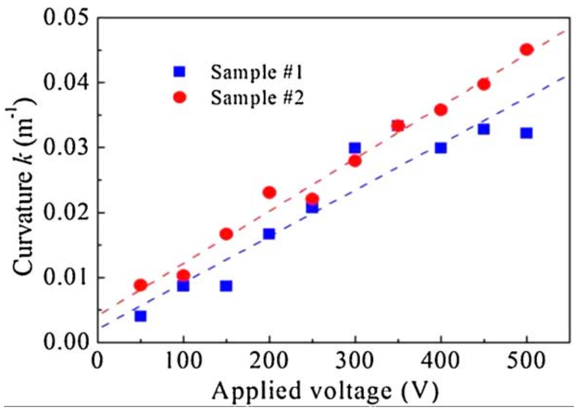

Fig. 8. Electric-field-induced curvature of two LKNN/Ni piezoelectric FGM actuators with Ni composition profile of 0%/10%/40%/10%/0%. The lines are guides for the eye.

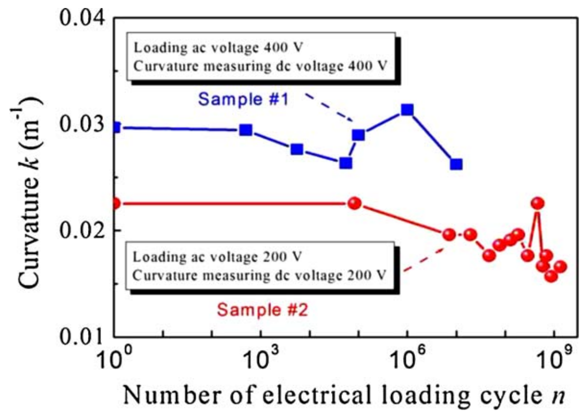

Fig. 9. Electric-field-induced curvature as a function of electrical loading cycle for two LKNN/Ni piezoelectric FGM actuators with dimensions of $16 \times 3 \times 2$ mm.

to a total out-of-plane curvature for the right-half or left-half FGM, owing to strain mismatch in the thickness direction. A modified classical lamination theory (CLT) has been applied to calculate electric-field-induced curvatures for piezoelectric FGM actuators,[9] and the results are consistent with experimental measurement.[22] This method is helpful for the design of piezoelectric FGM actuators with optimized driven performance.

Fig. 8 shows the electric-field-induced curvatures measured for the actuator in Fig. 7. Two actuator specimens with the same composition profile of 0%/10%/40%/10%/0% Ni and the same dimensions of $16 \times 3 \times 2$ mm were evaluated. The results show acceptable consistency between the two specimens. The electric-field-induced curvature was found to be dependent on applied voltage. The k value of approximately $0.04\,\mathrm{m}^{-1}$ was obtained for an applied voltage of 500 V, corresponding to an electric field of 0.6 kV/mm derived from the dimensions in Fig. 7. For comparison, the curvatures produced by these LKNN/Ni FGM actuators are somewhat lower than those for PZT/Pt[22] or PZT/Ag[18] FGM actuators with similar dimensions at these applied voltages. There are several possible explanations for this difference. Firstly, the piezoelectric constant of LKNN ceramics ($d_{33} = 165$ pC/N) is lower than that of PZT ceramics ($d_{33} \sim 400$ pC/N). Secondly, the electric-field-induced curvatures of the actuators should also be related to the composition profile of the functionally graded microstructure. The composition profiles of the reported PZT/Pt and PZT/Ag FGM actuators differ from the profile used in this study. Thirdly, the different metal fillers (Pt, Ag, and Ni) are likely to deform differently.

During service, a piezoelectric actuator needs to bend repeatedly with applied electric fields. To evaluate the reliability of the LKNN/Ni FGM actuators produced in this work, an electrical loading experiment was conducted. Two actuators (similar to the one shown in Fig. 7) were electrically loaded under ac voltages of 200 and 400 V, at a fixed frequency of 50 Hz. After a number of cycles, the loadings were interrupted and the electric-field-induced curvatures of the actuators were measured under dc voltages of 200 or 400 V. The measured curvatures serve as indicators of the reliability of the actuators. Fig. 9 shows the results of reliability evaluations for the actuators. For the 200 V applied voltage, there was an abrupt decrease in curvature at the beginning of loading, from $0.023\,\mathrm{m}^{-1}$ initially to $0.018\,\mathrm{m}^{-1}$ after about 2×10^8 cycles. The curvature then maintained a roughly constant value of $0.018\,\mathrm{m}^{-1}$ up to 1.3×10^9 cycles. Reliable cycling for over 10^9 cycles fulfils the requirement for long-term service of devices such as piezoelectric relays used in communication equipment[23] or piezoelectric valves used in the fuel injection systems of diesel engines.[24] Evaluations with the higher voltage of 400 V were only performed for 10^7 cycles, but gave similar results to the 200 V evaluations, with the actuator maintaining good performance.

The LKNN/Ni piezoelectric FGM actuator has exhibited good endurance under the electric fields applied. This result is the consequence of the metal particles dispersed in the piezoceramic matrix. As shown in Fig. 3, the dispersed Ni particles greatly enhance the fracture toughness of the LKNN matrix. The deformation of metal particles can absorb the propagation energy of cracks once initiated, thus arresting the fast growth of cracks and delaying the failure of devices. In addition, functionally graded microstructure is also responsible for the reliability enhancement of actuators. The formed FGM can reduce the difference in mechanical deformation ability between the various layers.[8] The interfacial stresses that are usually produced in conventional bimorph-type actuators have thus been greatly lowered in the FGM actuators.

4. Conclusions

Ni-particle-dispersed $[Li_{0.06}(K_{0.5}Na_{0.5})_{0.94}]NbO_3$ composites and functionally graded piezoelectric actuators were produced in this study. The resistance to crack propagation in the LKNN/Ni composites was significantly enhanced compared with monolithic LKNN ceramics. The mechanical property enhancement in the composites is closely related to the added metallic Ni particles. Functionally graded actuators containing dispersed metal particles were able to sustain a large number ($>10^9$) of electrical loading cycles under a particular electrical field. The results demonstrate that metal-particle dispersion is an effective approach for enhancing reliability of piezoceramics and related driven devices.

Acknowledgments

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