MINIATURE PIEZOCERAMIC ACTUATORS WITH IMPROVED FATIGUE RESISTANCE

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ABSTRACT

Piezoelectric ceramics are desirable actuator materials for many biomedical applications due to their ability to generate precise, controlled motion with applied voltage. Herein, we report the fabrication of miniature piezoelectric ceramic actuators and their subsequent performance testing under high electric fields. The actuators were produced by tape casting sub-micrometer lead zirconate titanate (PZT) powder, followed by electroding, laminating, and dicing to form multilayer devices. The resulting devices consisted of ten active (i.e., PZT) layers, each of which was approximately 50 μm thick. To evaluate the effect of microstructure on performance, the multilayer components were heated to temperatures ranging from 1175-1325 °C and held at temperature for 24 minutes. Fatigue resistance was then determined by monitoring the change in net polarization during continuous cyclic operation. Actuators sintered at lower temperatures possessed smaller-grained microstructures and exhibited improved fatigue resistance. In these specimens, polarization decreased by less than 15 % compared to more than 50 % for devices processed at higher temperatures. These results indicate that sintering control can be used to minimize degradation in performance, improve reliability, and promote long-term stability.

INTRODUCTION

Piezoelectric ceramics are desirable materials for many emerging actuator applications due to their fast response, wide operational bandwidth, high force, large power density, relatively compact size, and ease of motion control. These materials are already under development for adaptive optics, automobile fuel injectors, hard-disk-drive shock sensors, active flow-control devices, and vibration dampers. Additionally, there are opportunities to utilize piezoceramics in the biomedical field for miniature pumps, ultrasonic surgical tools, microneedle arrays, and micro- and nanorobotic systems. In the latter case, biomimetic locomotion is desired, mimicking natural motions, such as stretching, flexing, walking, or jumping, which often involve high strains and repeated activity. Actuator stability during these repetitive motions is particularly critical, representing a challenge for piezoelectric ceramics. The properties of these materials can degrade significantly during continuous operation, due to domain pinning, stress relaxation, and, in certain cases, microcracking[1,2]. This degradation (referred to as fatigue) can be even more pronounced when large electric fields are used to achieve high displacement.

In this work, microstructure control is investigated as a potential avenue to maximize fatigue resistance in lead zirconate titanate (PZT) actuators. Tailoring of the microstructure of PZT has been previously reported to optimize both mechanical and electrical behavior, with smaller grained ceramics typically demonstrating improved mechanical properties. For example, Shrout, *et al.*[3] reported an increase of 30 % in bending strength when grain size was confined below 1 μm, while Davis, *et al.*[4] reported increased blocked force, free displacement, and energy density for similar-sized grains. However, Karastamatis, *et al.*[5] demonstrated that larger-grained (5 μm) PZT ceramics displayed more ferroelastic strain than smaller-grained material (1.4 μm), indicating a slight improvement in fracture toughness with increasing grain size. Dos Santos, *et al.*[6] confirmed this increase in fracture toughness (15 % from 5.3 to 6.4 μm).

Regarding electrical properties, Randall, *et al.*[7] compared hot-pressed ceramics (0.2 μm grains) with conventionally sintered materials (~ 4 μm grains). Their experiments indicated that remanent polarization, piezoelectric properties, and electromechanical coupling coefficients decrease as grain size is reduced. This effect is likely due to a limitation in the number of domain orientation variants at ultra-fine grain sizes. This limitation effectively reduces poling efficiency, requiring added dopants to increase domain wall mobility.

Fatigue in PZT ceramics is related to both electrical and mechanical behavior, complicating the role that microstructure may play in optimizing fatigue resistance. Herein we address the fatigue-microstructure relationship, specifically for multilayer PZT devices. These components were prepared using a conventional fabrication route and sintered over a 150 °C temperature range (i.e., 1175-1325 °C), which was sufficient to produce considerable variation in the fired ceramic microstructures. Comparison of degradation in performance when the devices were continuously operated at high electric fields is described in the following sections.

EXPERIMENTAL PROCEDURE

PZT powder was obtained from a commercial vendor. The as-received powder contained a binder, which was subsequently removed by heating at 20 °C/min to 400 °C and annealing for 30 minutes. The heat-treated powder had a specific surface area of 1.23 m^2/g (by nitrogen adsorption testing) and an estimated average particle diameter of 635 nm. The powder was then attrition milled for 6 hours to reduce the particles to approximately 140 nm, combined with a surfactant and modifier, dispersed in toluene, and milled at high speed for 10 minutes using a laboratory-scale shaker mill. After adding a binder and plasticizer, the mixture was ball-milled at low speed for 24 hours to ensure complete dispersion of the powder. Zirconia media were used throughout the milling process to minimize added impurities.

A conventional tape-casting technique[8,9] was employed to fabricate the actuators, as shown in Figure 1. A silicone-coated plastic carrier film served as the substrate onto which the cast tape was deposited, and the continuous tape was cut into 10 cm x 10 cm sheets to form the individual layers in the multilayer stack. A doctor blade designed for casting tape with a thickness of 0.17 mm was used throughout this work. Platinum ink was screen-printed onto individual PZT sheets to serve as the internal electrodes within the device. The printed electrodes were then dried in a belt oven at 80 °C.

Next, the multilayer assembly was prepared by stacking together the individual PZT sheets and applying temperature and pressure to form a cohesive pad. The pad contained 36 sheets of PZT tape, including eight cover (i.e., inactive) layers on both the top and bottom. To form the active section of the actuator across which the voltage is applied, ten blank sheets were interleaved with ten electroded sheets to form an interdigitated internal electrode structure similar to that shown in Figure 2a. The entire pad was then tacked together at 58 °C for 30 seconds with 0.4 MPa of pressure and laminated at 70 °C for 20 minutes at 12.4 MPa. Separation of discrete actuator components (0.30 cm x 0.15 cm; see Figure 2b) was achieved using a vision-assisted dicing saw.

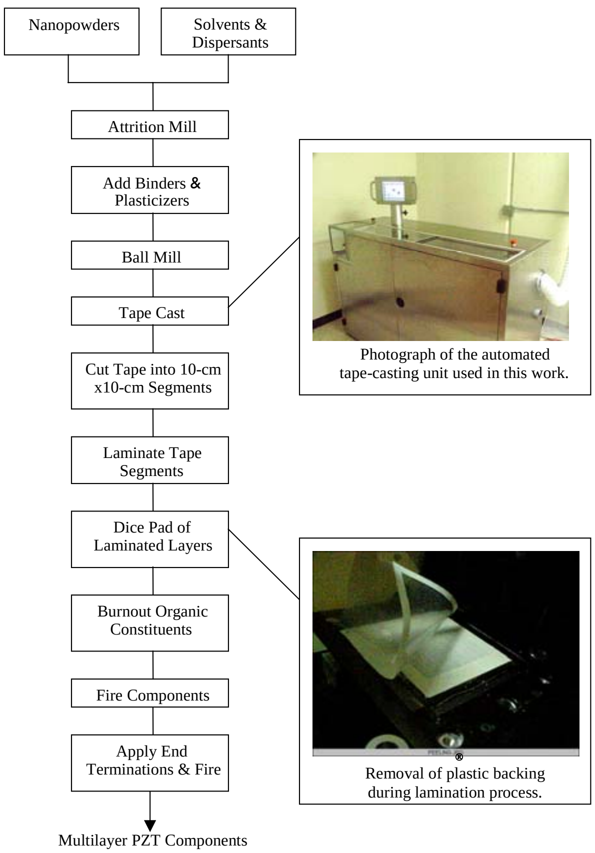
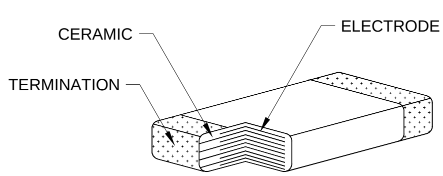


Figure 1. The actuator fabrication process.

Next, the actuators were slowly heated in flowing nitrogen to 550 °C to remove the remaining organic materials. A large muffle furnace was then used to sinter the devices in air at temperatures ranging from 1175 to 1325°C (24 minute annealing period). All parts were buried in PZT powder and fired in sealed crucibles to minimize lead loss. Relatively fast ramp rates (20 °C/minute) were used for both heat-up and cool-down to minimize total time at or near the peak temperature. Termination pads were then applied to the device edges by means of a commercial silver-palladium paste and heated to 850 °C for 15 minutes to volatilize the organic components in the paste and ensure conductive contacts.

To determine microstructure features, a representative component from each temperature was evaluated using scanning electron microscopy (SEM). Dielectric properties were measured at 1 kHz for an additional 10 components, and a modified Sawyer-Tower circuit was used to obtain ferroelectric properties (i.e., remanent polarization (P_r), saturation polarization (P_{sat}), and coercive field (E_c)) for these devices. In the latter measurement, a 1 Hz sinusoidal wave served as input to the circuit, and P-E hysteresis loops were acquired with a digital oscilloscope.

To determine the effect of long-term use, three actuators from each temperature were operated for up to 1 million cycles, using a test frequency of 35 Hz. Data were acquired at regular intervals, and P_r, P_{sat}, and E_c were recorded as a function of cumulative exposure. To compare the



(a)

(b)

Figure 2. Schematic showing internal layers (a), and green parts diced from the pad (b).

properties as a function of sintering temperature, the values for P_r were normalized, with P_r at zero exposure representing a value of one. One specimen for each temperature was then cycled for extended periods to assess whether further degradation occurred with longer-term operation. The results from these examinations are described below.

RESULTS AND DISCUSSION

Table 1 summarizes the dielectric and ferroelectric properties of the as-produced multilayer actuators as a function of temperature. In particular, capacitance and polarization reached maxima when devices were processed between 1250 and 1275 °C (a typical temperature range for sintering PZT). However, SEM images of fractured components (Figure 3) showed that considerable grain growth occurred at these temperatures, with individual grains becoming less distinct by 1275 °C.

Table 1. Effect of sintering temperature on dielectric and ferroelectric properties.

Temperature	Capacitance (nF)	Dielectric loss factor, tan δ	P_{sat} ($\mu C/cm^2$)	P_r ($\mu C/cm^2$)	E_c (kV/cm)
1175 °C	6.48	0.023	17.65	12.82	14.06
1200 °C	6.38	0.022	20.35	15.32	14.70
1225 °C	6.67	0.021	25.79	20.95	15.73
1250 °C	6.79	0.020	30.20	24.30	16.38
1275 °C	6.65	0.019	31.99	26.17	17.19
1300 °C	6.32	0.019	31.55	26.09	18.02
1325 °C	5.17	0.017	27.13	22.40	19.97

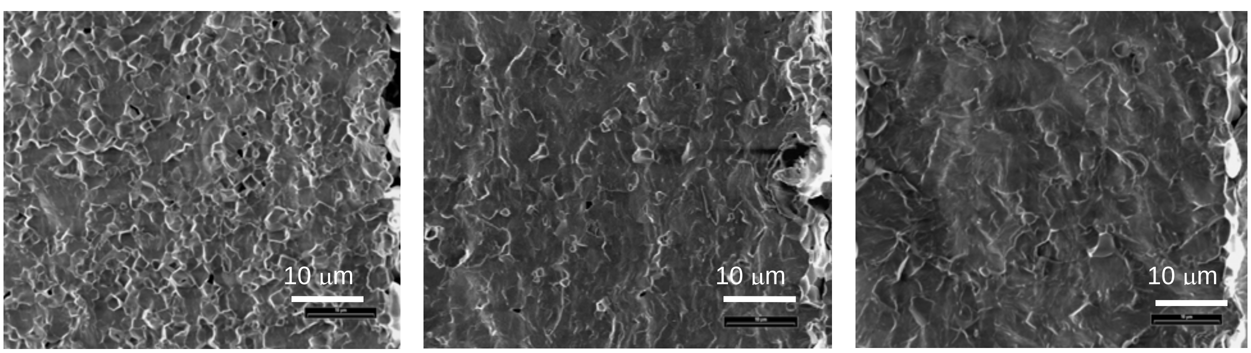


Figure 3. SEM micrographs of fractured cross-sections for devices processed at 1175, 1250, and 1325 °C (from left to right), illustrating the evolution of the microstructure.

Polarization degraded rapidly for all parts during continuous cycling. As shown in Figure 4, significant changes in the shape of the hysteresis loops were evident after only 45 minutes of testing (i.e., 100,000 cumulative cycles). However, it should be noted that the switching field used for these measurements was quite large (i.e., at or near the material's coercive field) and was selected in order to ensure measurable property degradation. As a result, the degradation in ferroelectric response observed during these tests is likely more rapid and extensive than that anticipated during conventional operation. Within each temperature condition evaluated, part-to-part variability was relatively low, with a maximum standard deviation in P_r of ± 6.06 % for specimens processed at 1225 °C. The best agreement was found for the 1250 °C specimens, exhibiting a standard deviation of only 0.83 % among the three parts tested.

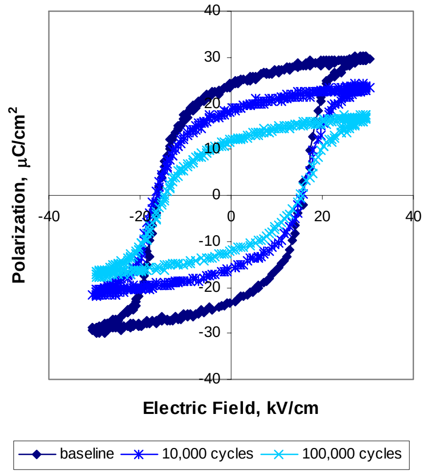
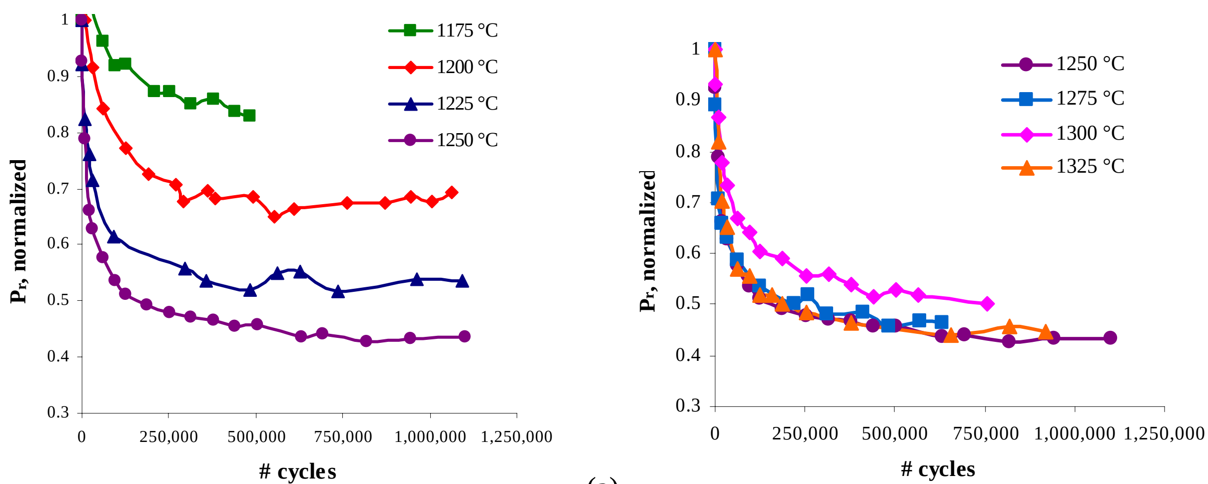


Figure 4. Change in P-E loops during the first 45 minutes of testing for a typical specimen processed at 1250 °C.

The extent of degradation in P_r as a function of cumulative exposure cycles is shown in Figure 5. In these graphs, the polarization data have been normalized in order to show the relative change across the different sintering temperatures. As shown in Figure 5a, a significant difference exists in the fatigue behavior of devices sintered from 1175-1250 °C. For example, components heated to 1175 °C retained an average of 91.9 % of their initial remanent polarization after 100,000 cycles, compared to only 51.8 % for devices processed at 1250 °C. Similar results were found for 250,000 (84.9 % versus 47.6 %) and 500,000 (82.8 % versus 45.6 %) cycles. However, for devices sintered from 1250-1325 °C, no significant differences in fatigue behavior were observed (see Figure 5b). These components all retained approximately 50 % of their initial polarization after 500,000 cycles.



(a) (b)

Figure 5. Degradation in P_r as a function of cumulative cycles for specimens processed from 1175 to 1250 °C (a) and 1250 to 1325 °C (b). Specimens sintered at 1175°C were tested to only 500,000 cycles. Data shown are for a typical specimen at each temperature. Standard deviations ranged from ±0.38 to ±6.06 %.

The data in Figure 5 illustrate that sintering temperature plays a role in determining an actuator's resistance to fatigue, defined herein as the change in ferroelectric response during operation. However, these data represent changes in performance when devices were tested using a low-frequency sinusoidal wave (35 Hz) over relatively short periods (8 hours for 1 million cycles). In conventional use, actuators can be exposed to considerably more cycles, higher switching frequencies, and periodic power-down-and-restart scenarios. Of particular interest for biomedical applications is the effect of longer-term operation.

In Figure 5, the majority of degradation is seen to occur during the first 100,000 cycles. Figure 6 shows the change in P_r and P_{sat} over 12 million cycles for a device processed at 1300 °C. P_r decreased 51.6 %, 52.7 %, and 59.2 % by 1, 5, and 10 million cycles respectively. Similar effects were seen for specimens processed at 1200 and 1250 °C. These results indicate that the degradation levels off over time. As a result, system designers may need to schedule a burn-in period to overcome the initial changes in performance.

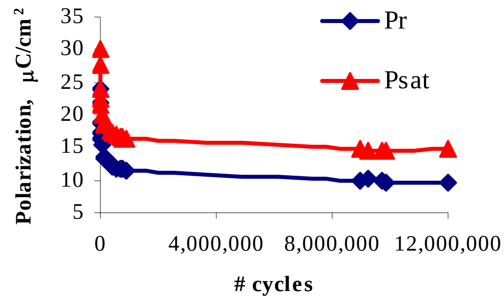


Figure 6. Change in polarization for a component processed at 1300 °C.

CONCLUSIONS

Miniature multilayer actuators were fabricated from 140 nm PZT powder using a conventional tape-casting and lamination procedure. The components were sintered at temperatures ranging from 1175 to 1325 °C to obtain different ceramic microstructures, with grain size increasing with increasing temperature. Components processed at the lowest temperature (1175 °C) showed significant improvement in fatigue resistance, retaining more than 90 % of their initial P_r after 100,000 cycles. By comparison, actuators sintered at 1250 °C retained only 50 % of their initial P_r after the same cumulative exposure. These results indicate that control of microstructure may be a viable avenue to maximize fatigue resistance in PZT ceramics for high-reliability applications. Additional studies are planned with finer-grained microstructures to further investigate this effect.

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