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Manufacture of large displacement antiferroelectric ceramic and its properties

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Abstract: Reduced and internally biased oxide wafer (RAINBOW) Pb (Sn, Zr, Ti) O_3 (PSZT) antiferroelectric ceramics are fabricated by chemical reduction. It is a new kind of large displacement actuating materials composed of reduced and unreduced layers. It is found that PSZT is easily reduced and the optimal conditions for producing RAINBOW samples are determined to be 870℃ for 2 to 3 h, which results in a reduced layer composed of metallic lead and refractory oxides (PbO, ZrO_2 and $ZrTiO_4$). The phase transitions from antiferroelectric state to ferroelectric state occur at lower field strength in RAINBOW samples compared with normal PSZT ceramics. Larger axial displacement is also obtained from RAINBOW samples by application of electric fields exceeding the phase switching level. However, the actuating properties of RAINBOW samples are dependent on the manner of applying load on it.

Key words: reduction; antiferroelectric ceramic; phase switching

During the past several years, piezoelectric actuators have been subject to numerous investigation and undergone remarkable advance[1]. Traditional piezoelectric actuators offer many advantages including quick response, high-induced stress, low energy consumption and low cost which make them very attractive for a number of applications. However, the electric field-induced strains of piezoelectric ceramics are relatively small, which considerably limits their use. To achieve a higher displacement from the ceramics, a number of strain magnification mechanisms have been employed. Examples include the traditional unimorph and bimorph benders, and the "moonie" microstructures[2,3]. However, an increase of induced displacement is achieved at the expense of lowering generated stress significantly. The most recently developed strain amplifying method for piezoelectric actuators, which shows promise for meeting many applications, is known as the reduced and internally biased oxide wafer (RAINBOW) technologies[4,5]. In their most basic sense, RAINBOW can be thought of as pre-stressed, monolithic, axial-mode benders. Because of their unique dome or saddle-like configuration, RAINBOW ceramics are able to produce much higher displacements and sustain significantly greater loads compared with conventional piezoelectric actuators. It is fabricated by reducing one surface of ceramics on flat carbon block at an elevated temperature. Since the reduced layer and the remaining unreduced layer have different thermal expansion coefficients, internal thermal stresses and the dome structure will be generated when the ceramic is cooled during room temperature after reduction.

On the other hand, it has been found that the Pb(Sn, Zr, Ti)O_3(PSZT), one kind of antiferroelectrics with compositions in the vicinity between antiferroelectric (AFE) state and ferroelectric (FE) state boundary exhibit very large field-induced strains resulting from the transition from AFE to FE state. A strain of 1.1% that is the highest ever reported for ferroelectric ceramics was claimed in Ref.[6]. Furthermore, the strain characteristics of these ceramics can be modified through selection of appropriate compositions[7,8]. So, the strain property of the PSZT is very useful actuating mechanism for actuators; but the relatively high field strength needed for the phase switching makes it impractical.

The objective of this work is to combine the high-induced strains of PSZT with the RAINBOW technology to produce a new kind of ultra-high displacement actuator that can be operated in relatively low field and used in more applications, especially in smart structures. In this paper, the fabrication and properties of RAINBOW actuators made of PSZT are studied.

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1 Experimental Procedure

1.1 Sample preparations

The RAINBOW samples were prepared from PSZT ($Pb_{0.97}La_{0.02}(Zr_{64}Sn_{26}Ti_{10})O_3$) sintered ceramics. The ceramic wafers (ϕ27 mm × 0.5 mm) obtained from PSZT slugs were chemically reduced by placing them on a graphite block and introducing the assembly into a pre-heated furnace. A ziconica disk was placed on top of the PSZT wafer to prevent its thermal shock during processing. After reduction, samples were quenched during air, which make samples have a unique dome shape. Silver electrodes were cured on two surfaces of samples to measure their electrical properties.

1.2 Measurements

The crystalline phases of reduced PSZT were examined by an X-ray diffractormeter (D/Max-γC). The thickness of the reduced layer of the RAINBOW samples was measured from the sample cross-sections by means of an optical microscope. Conventional dc hysteresis loop equipment was employed to measure the relationship between polarization and electric field. Electric fields which were greater than the AFE-FE phase transition levels were gradually applied to the samples. A measuring device with linear voltage difference transmitter (LVDT) and PC as shown in Fig.1 was used to determine the change of the field-induced displacement with electric field. The RAINBOW sample was placed on a metal ring in a container that was filled with silicon oil for insulating purpose. The movable core of the LVDT was adjusted to contact the center of the RAINBOW sample. Mechanical load on the RAINBOW samples was accomplished by placing weights on top of the LVDT. The variations of polarization and axial displacement with electric field were simultaneously measured as the samples were loaded.

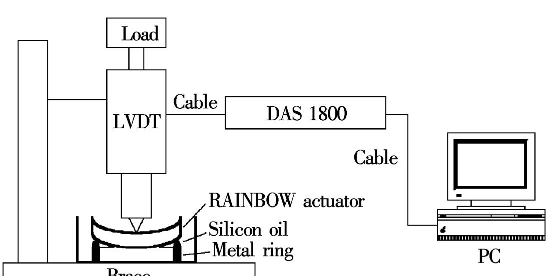

Fig.1 Schematic drawing of device for displacement measurement

2 Results and Discussions

2.1 Reduction of PSZT ceramics

Temperature and reduction time are important factors in controlling the reduction process during fabrication of RAINBOW actuators. The relations between reduced thickness and time in high temperature depend on the material itself. At a given temperature, the reduced thickness is approximately linear with time for Pb (La, Zr, Ti) O_3 (PLZT) ferroelectric ceramics (materials most frequently used to make RAINBOW actuators), however, the parabolic law is followed by Pb (Zr, Ti) O_3 (PZT)[9]. A significantly thicker reduced layer in PSZT than in PLZT was produced when they were reduced at the same temperature for a given time. Fig.2(a) shows the reduced layer thickness of the RAINBOW sample as an appropriately linear function of reduction temperature for 1 h. An approximately 600 μm thick reduced layer was created at about 975 ℃. In addition, Fig. 2 (b) shows the change of the reduced layer thickness with time at a constant temperature of 870 ℃ for the PSZT sample. A nearly linear relationship was observed.

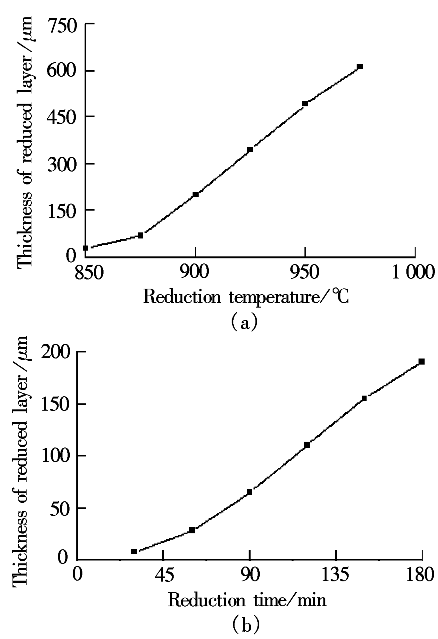

Fig.2 Variation of thickness of PSZT reduced layer with reduction conditions. (a) 1 h; (b) 870 ℃

Fig.3 shows the X-ray diffraction pattern from a sample reduced in 870 ℃ for 2 h. It can be found obviously that a significant amount of the original PSZT phase remained in addition to Pb, PbO, ZrO_2 and $ZrTiO_4$, which resulted from the reduction reaction. That is to say the reduction of the PSZT is incomplete though its reduction speed is very rapid, which is unlike the situation of the reduction of PLZT and PZT. Generally, at a higher temperature, the rapid reaction in PSZT ceramics leads to the loss of a large portion of lead phase from the reduced region. As a result, the reduced region has poor electrical conductivity, which is detrimental to the performance of RAINBOW actuators since the reduced layer must be electrically conductive in order for a RAINBOW to operate properly[9]. To prevent the heavy loss of the lead content from occurring, lower reduction temperature must be used. However, a very low temperature implies impractical and long reduction time. It is found that the useful temperature range for the production of PSZT RAINBOW is actually narrow, approximately 820 to 900 ℃. The optimal conditions for producing RAINBOW samples from PSZT ceramics are determined to be 870 ℃ for 2 to 3 h in this study.

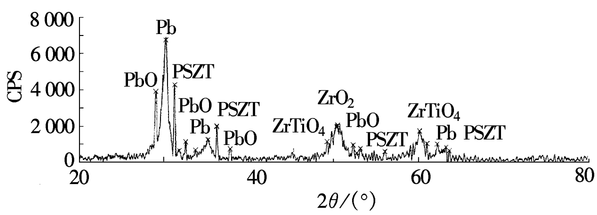

Fig.3 X-ray diffraction pattern of the reduced sample

2.2 Properties of RAINBOW PSZT ceramics

Tab. 1 summarizes the basic properties obtained from RAINBOW and normal PSZT ceramics. The PSZT RAINBOW, in general, possessed a lower dielectric constant and a higher loss factor than the normal PSZT. The phase switching fields, E_{AF} and E_{FA}, of the RAINBOW PSZT are much lower than those of normal PSZT. However, the saturated polarization was almost similar in two materials. The total field-induced axial displacement of the RAINBOW PSZT is much larger than that of the normal PSZT.

Tab.1 Properties of PSZT ceramics

Samples	Dielectric constant	$\tan\delta$/ %	E_{AF}/ $(MV \cdot m^{-1})$	E_{FA}/ $(MV \cdot m^{-1})$	P_S, P_R/ $(C \cdot m^{-2})$	Y_M/μm
RAINBOW	821	3.9	1.65	−0.3	3 000	187
Normal	913	1.9	2.8	0.1	3 100	120

Notes: E_{AF} is the antiferroelectric to ferroelectric switching field; P_S is the saturated polarization; E_{FA} is the ferroelectric to antiferroelectric switching field; P_R is the remnant polarization; Y_M is the maximum axial displacement with an applied electric field of 1.2 E_{AF}.

Although a larger axial displacement (about 190 μm) has been achieved in this study, it is also found that load directions have great influence on the actuating displacement of the PSZT RAINBOW actuators. As clearly illustrated in Fig.4, there is only a slight change in the displacement up to 570 g when load is placed on the unreduced layer; however, the displacement with load on the reduced layer decreased continuously with increased load. The different characteristics under the two load conditions can be explained by the behavior of ferroelastic domains under stress[10]. Ferroelastic domains tend to be in line with the directions in which stress is effectively relieved. When load is applied vertically to the unreduced layer surface of a RAINBOW, ferroelastic domains are preferably aligned parallel to the surface due to the compressive stress in the planar directions produced by the load. Similarly, when load is placed on the reduced layer, ferroelastic domains tend to be oriented vertical to the surface as a result of planar tensile stress. It is obvious that a PSZT RAINBOW actuator is more advantageous when operated with load on the unreduced side.

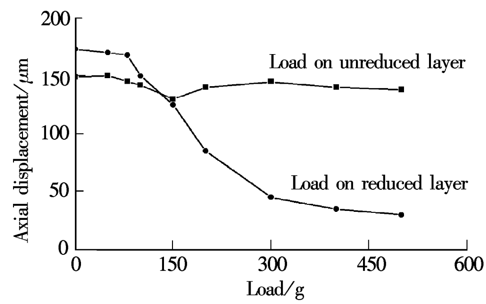

Fig.4 Variation of axial displacement with load

There are also other factors, which are important to the properties of the RAINBOW ceramics. For example, different thickness ratios of ceramic layer and reduced layer will generate different axial displacements of RAINBOW ceramics.

3 Conclusion

The fabrication and properties of PSZT RAINBOW actuators have been investigated. The optimal reduction condition for the manufacture of PSZT RAINBOW samples is 870 ℃ for 2 to 3 h. The AFE-FE phase transitions occur at lower field strength in RAINBOW actuators compared with normal ceramics. Larger axial displacement (about 190 μm) were obtained from the RAINBOW actuators by application of electric fields exceeding the phase switching level. The field-induced displacement of the RAINBOW actuators is dependent on the manner of applying mechanical load on the samples. There is only a slight change on the displacement for loads up to 570 g with load on the unreduced layer. However, when load is placed on the reduced layer, the displacement decreases markedly with the increase of load.

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大位移反铁电陶瓷的制备及其性能

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摘要: 通过化学还原的方法制备了 RAINBOW Pb(Sn, Zr, Ti)O_3(PSZT)反铁电陶瓷. 它是一种具有还原层和未还原层、驱动位移极大的新型驱动材料. 研究表明: PSZT 反铁电陶瓷较易还原, 理想的还原条件为 870 ℃保温 2 ~3 h; 还原层主要由金属铅以及 PbO, ZrO_2, $ZrTiO_4$ 等金属氧化物组成; 与普通的反铁电驱动器相比, RAINBOW 试样能在强度较低的电场下发生反铁电-铁电相变, 并能得到极大的轴向驱动位移, 但加载方式对其驱动性能有决定性的影响作用.

关键词: 还原; 反铁电陶瓷; 相变

中图分类号: O346.4