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Detection of cracks and fracture in ceramics with the piezoelectric PZT thin films

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The piezoelectric lead zirconate titanate (PZT) thin films with the (100) preferred orientation were successfully deposited on various polycrystalline ceramic substrates, such as SiC, Si_3N_4 and Al_2O_3 using r.f.-sputtering method. The piezoelectric coefficients of these films were evaluated by the static method using the direct piezoelectric effect. Their piezoelectricities were also investigated from the maximum piezoelectric voltage (V_{max}) responses generated by the mechanical impacts (F_i). Results show that two peaks appear in the relation of V_{max} versus F_i, which just correspond to the fracture process of ceramics. The two peaks represent the crack initiation and fracture in the ceramic substrates, respectively. By means of this method, detection of cracks and fracture in ceramics with the PZT film can be realized. The relations of the oriented degree of the PZT films to the V_{max} are discussed in this paper. From this primary work, it can be induced that the PZT film deposited directly on the ceramics is a promising material to detect the cracks and fracture of engineering ceramic. Further research work on this application is in progress in our laboratory.

Key words: PZT film, Piezoelectricity, ceramics, detection

1. INTRODUCTION

As a typical piezoelectric material, the PZT is widely developed as sensors and actuators, especially in the microelectronic mechanical system (MEMS) research field. Usually, the PZT film is deposited on single crystal silicon substrate since the MEMS technique is based on the silicon planar process. However, we expect the PZT film to be deposited on various amorphous and polycrystalline substrates in order to detect or predict the mechanical behavior of bulk materials. As we known, the fracture of materials often induces serious accidents, especially in aviation and aerospace field, as well as the other heavy industrial fields. It becomes an inexorable trend to develop energetically the monitor system of materials or prepare synergetic materials. The carbon fiber reinforced plastics (CFRP) with embedded piezoelectric PZT fiber was investigated to realize the self-diagnosis and self-repairing of CFRP [1]. The growth of the PZT film needs a compatible environment, therefore, most of the research works have been approached in the composite bulk and the laminate materials so far.

In this paper, the PZT films were deposited on amorphous glass, polycrystalline SiC, Si_3N_4 and Al_2O_3 ceramics, in which, the piezoelectric PZT films look like the skins on substrates to detect and predict the mechanical behaviors of substrates. The piezoelectric voltage responses of the PZT films on static stress and impact load were investigated in detail.

2 EXPERIMENTAL

The PZT films were fabricated by r.f.-sputtering method. The composition of the target is the 0.4at% Nb-doped $Pb_{1.15}(Zr_{0.52}Ti_{0.48})O_3$. Table I shows the optimized process conditions of the PZT films on various substrates. After deposition, the PZT films were post thermally treated in situ in the chamber under the action of different atmospheres to obtain the pure perovskite phase and preferred orientation.

Some of the PZT films were deposited on the LNO films prepared by the aqueous technique. The perovskite structure of the LNO film is compatible with that of the PZT and may be helpful to promote the growth of the PZT film with a high preferred orientation. Moreover, the conductivity of the LNO film makes it possible as a bottom electrode.

The piezoelectric coefficient d_{33} of the PZT films was measured by the static method using the direct piezoelectric effect [2]. The stress was exerted perpendicularly on the surface of the sample, and could be measured by a pressure gauge. The diameter of the copper tip is about 1mm, which equals the diameter of the electrode in the surface of the sample. The surface of the tip was manufactured carefully to ensure the uniformity of the stress exerted on the sample. Herein, the d_{33} does not represent the true piezoelectric coefficient of the film since it is always clamped to the substrate. Using this method, the real piezoelectric effect was judged in the process of increasing or decreasing the stress that resulted in the reversible voltage response. The sample was composed of an Au/PZT/LNO/substrate structure, in which, the gold dot matrix with diameter of 1 mm was used as the upper electrode and the LNO as the bottom electrode.

The piezoelectric voltage response was generated by the effect of the impact load on the sample when a falling ball is released from a certain height. The relation of maximum piezoelectric voltage response to the impact load could be obtained. The measuring system has been described elsewhere [3]. The whole measurement system was screened very well to avoid the noise. The Al/PZT/substrate samples were used in this impact test system. The width of interdigital transducer (IDT) Al electrode/distance between two electrodes=1mm/1mm, and the pair numbers of the IDT electrodes =7.

Table I Optimized process conditions of the PZT films on various substrates

Substrate / Parameter	Al_2O_3	Si_3N_4	SiC	LNO/ Al_2O_3	LNO/Si_3N_4	LNO/SiC	Glass
Substrate temp. (°C)	690	600	690	400	690	690	650
Sputtering time (h)	3	3	6	3	3	6	3
Atmosphere	Ar/O_2=3/0.5	Ar	Ar	Ar	Ar	Ar	Ar
Pressure (Pa)	2	2	2	2	2	2	2
Post treated atmosphere	Air	Vacuum	Vacuum	Vacuum	Vacuum	Vacuum	Vacuum
Treated temp. (°C)	600	600	600	600	600	600	600
Treated time (h)	3	3	6	3	3	6	3
Microstructure	Perovskite	Perovskite	Perovskite	Perovskite	Perovskite	Perovskite	Perovskite
Orientation	No	Preferred	Preferred	Orientation	Preferred	Preferred	Preferred
Oriented degree N*		N(100)=3.3 N(111)=4	N(100)=8.7	N(100)>12.5	N(100)=8.2	N(100)=9.1 N(111)=10.5	N(100)=5.1

*Note: $N(hkl) = [I(hkl)/(I(hkl)+I(110))]/[I_0(hkl)/(I_0(hkl)+I(110))]$, Io is the peak intensity of standard powder. Wherein, N=1, no orientation; 1<N<12.5, preferred orientation; N≥12.5, orientation [4,5].

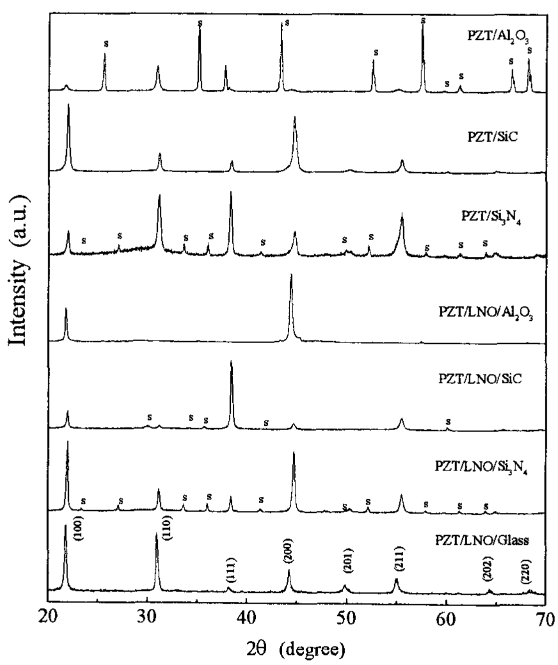

Fig.1 XRD patterns of the PZT films on various substrates (s: substrate)

3. RESULTS AND DISCUSSION

Table I shows that the substrate temperature is the most sensible process factor of crystallization of the PZT films because the surface and microstructure of various substrates decide the nuclear status and the growth of the PZT films under different substrate temperatures. Usually, the higher substrate temperature, the better the crystallization of the PZT film. In our study, the Si_3N_4 and LNO/Al_2O_3 substrates show lower sputtering temperature. Especially for LNO/Al_2O_3 substrate, the sputtering temperature is only about 400°C, the reason is being analyzed by TEM, SEM and XRD techniques. However, for Al_2O_3 substrate, we can not obtain the pure perovskite phase at all under the pure argon atmosphere. It may be caused by the uncoordinated structure and composition with the nitride and the carbide. Experimental results also show that a short sputtering time could not get the PZT film with the pure perovskite phase for SiC substrate. Prolonging the sputtering time would effectively improve the crystallinity of the PZT film. Meanwhile, the post thermally treated time was also prolonged to make the extra lead evaporate enough.

Fig. 1 gives the XRD patterns of the PZT films on various substrates. All of them show that the PZT films possess the pure perovskite phase. The preferred orientation of the PZT films depends on the preparation process and the substrate. Most of the optimized films have the preferred orientation. The PZT films with the LNO bottom electrode exhibit stronger preferred orientation. It illustrates that LNO bottom electrode promotes the oriented growth of the PZT film on various substrates since the formation of perovskite phase at the LNO surface acts as a nucleation site for PZT crystallization [6].

The piezoelectric coefficient d_{33} of the unpoled PZT film was determined to be 15 pC/N for glass substrate and 24 pC/N for SiC ceramic substrate. Since the PZT film was clamped to the stiffness substrate, its piezoelectric coefficient is much lower than that of bulk and film without clamping. The poled treatment will increase the d_{33} values. After poling at room temperature using 5V DC voltage for 3 minutes, the d_{33}

of the film becomes 23 pC/N for glass substrate and 33 pC/N for SiC substrate, respectively.

Table II Bend strengths of substrates

Substrate	Glass	SiC	Si_3N_4	Al_2O_3
Bend strength(MPa)	91	314	337	375

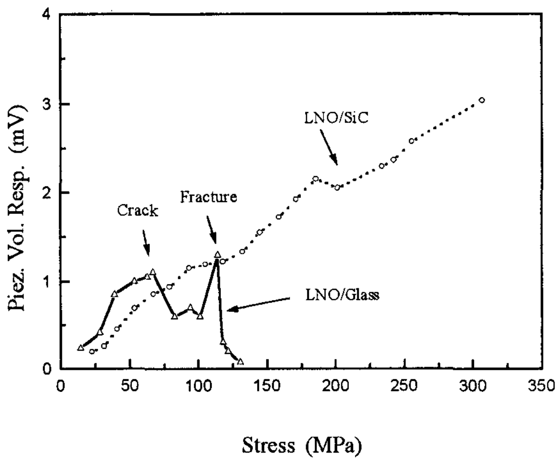

Fig 2 The piezoelectric voltage response V_{max} of the PZT films excited by the static stress

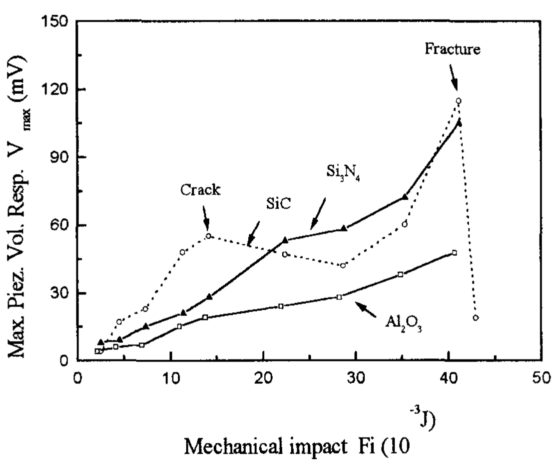

Fig. 3 The V_{max} excited by the impact load Fi

Table II gives the bend strength of various substrates measured using the three-point bend strength. Fig. 2 shows the piezoelectric voltage responses of the samples under the action of static stress. Fig. 3 presents the maximum piezoelectric voltage responses (V_{max}) of various samples excited by the impact load (F_I). Regardless of what kind of external load, two typical response curves were obtained in the measurement. They depended on the mechanical properties of substrates and measuring conditions. It may be addressed as four stages: firstly, within the elastic limit of the substrate materials, the piezoelectric voltage response increases almost linearly with the increasing of the stress or the impact load. In this case, we cannot observe any macro cracks in the substrate materials. It can be explained by the linear piezoelectric equation. Of course, we are not sure if the micro cracks exist, but at least, it still remains in the range of quantitative change. Above the elastic limit, the qualitative change takes place and the cracks appeared in the substrate (which can be observed by eyes), which corresponds to the first peak in the response curve. This is the second stage and then the cracks would propagate with the increasing of external load or impact load. In the stage of crack propagation, the growth of the cracks cause the reduction of the effective electrode area, therefore the response displays a random change. Finally, the fracture of the substrate occurs, which produces a very strong response, corresponding to the second peak in the curve. After this, the response reduces rapidly. For the crack and fracture to occur in the substrates two peaks are always formed in the response curve.

Under the action of static stress, the piezoelectric voltage response increases linearly with the increasing of the stress. Due to smaller bend strength in the glass substrates, they are easily to crack and fracture than the ceramic substrates. When a 115 MPa stress is applied to the glass substrate, the glass fractures and the relative response reaches the maximum value and then drops it to a small value. The PZT film deposited on SiC substrate basically increase linearly with the increasing of the stress on account of no cracks.

The same trend is obtained when the sample is subjected to the mechanical impact (Fig. 3). In these three kinds of substrates, the bend strength of the SiC ceramics is the smallest, therefore the SiC cracks and fractures firstly. The bend strength of Al_2O_3 ceramics is the biggest one, it still remains linear within the measured range. The bend strength of the Si_3N_4 ceramics is between both of them, it appeared to be increasing nonlinearly, but it still does not reach the peak of the crack. This result is identical with their bend strength shown in table II.

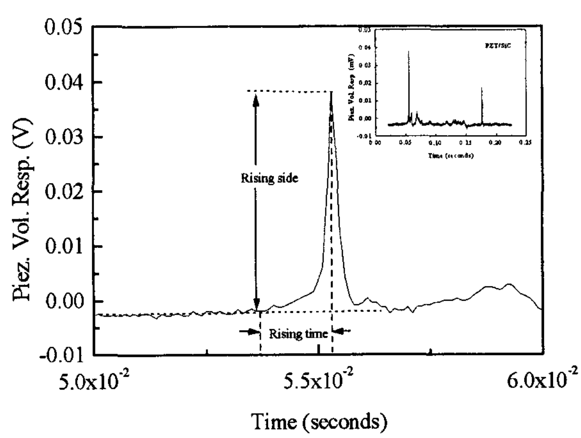

Fig. 4 Typical piezoelectric voltage response curve of the PZT/SiC excited by the impact load

Table I shows that the oriented degree of the PZT films on various substrates is Al_2O_3<Si_3N_4<SiC. Corresponding to the Fig. 3, the higher orientation, the

stronger the piezoelectric effect in the linear range, therefore, the PZT film with a high preferred orientation has the higher piezoelectric response.

Fig. 4 shows a typical response curve of the PZT film excited by the impact load. The time of the piezoelectric response is about in the order of millisecond.

4. CONCLUSION

The PZT films were successfully deposited on various substrates. Their piezoelectric coefficient d_{33} reaches about 33 PC/N and the relative piezoelectricity was discussed in detail under the static stress and the impact load. With the increasing of the load, the piezoelectric voltage response curves represent the mechanical state of substrate excited by the external load, the crack initiation, the crack propagation and fracture. The higher the orientation of the PZT films the higher the piezoelectric response. This means that the piezoelectric response of the PZT film reflects the fracture process of ceramic substrate, therefore, the detection of cracks and fracture in ceramics with PZT thin film has been realized. We believe that the PZT film, depositing directly on ceramics, is a promising material to detect cracks and fracture of engineering ceramic components. Further research work on this application is in progress in our laboratory.

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