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Deposition and Characterization of Thick $Pb(Zr,Ti)O_3$ Films on Optical Fibers

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In this work we report on the deposition of PZT thick film onto optical fibers and characterization of their microstructure, dielectric, and piezoelectric properties. PZT thick films were deposited onto standard single-mode optical communication fiber of a 0.125 mm diameter by dip-coating method. The final sintering temperature of the films was 600°C and the thickness was around 2 μm. Deposition procedure was developed for both bare fibers and for those with metallic coating (Pt). Bending vibrations of the fiber driven with the external electric field at different frequencies and under different applied voltages were studied.

Keywords Optical fiber; PZT thick film; piezoelectricity

Introduction

Integrated fiber-optic signal processing devices, which can control guided light directly within the communication line, are becoming increasingly important in telecommunication networks and sensors [1]. A piezoelectric cylinder tube wrapped around the optical fiber was used for the phase modulation in optical fiber interferometers [2]. Other applications of in-line fiber phase modulators, such as fibers with coaxial piezoelectric transducer and those with piezoelectric coating have been reported [3, 4]. Piezoelectric thin films deposited directly onto a short section of a fiber (a few millimetres) that can be used as high frequency phase modulators have been studied in the past [5, 6]. Piezoelectric thin films of oriented zinc oxide (ZnO) was used as a driving element for such modulators. However, the deposition of ZnO films with proper orientation on a long cylindrical surface is a very difficult technological task [6]. A concept of coaxial piezoelectric modulator based on thick $Pb(Zr,Ti)O_3$ (PZT) is used in this work not only to modulate the optical signal but also to cause the bending displacement of the fiber tip. Bending displacement of the fiber may provide mechanical scanning of the fiber tip, opening new possibilities for the scanning

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microscopes and fiber alignment systems. Such devices can be simple, inexpensive, and of extremely small size as compared to conventional modulators.

Experimental

The PZT (Zr/Ti 58/42) films were fabricated by a sol-gel method. Sol-gel precursor was prepared by dissolution Ti-isopropoxide and Zr- propoxide in methoxyethanol. Lead acetate tryhidrate was than added as a Pb-containing source. A 5% excess of lead was introduced in the solution for lead loss compensation. Details of the precursor solution preparation were described elsewhere [7]. Commercially available fine PZT powder (TRS 600FG, TRS Ceramics, Inc) was used as a ceramic filler dispersed in the solution (0.6 M PZT). Powder concentration in the slurry was 500 mg/ml. PZT thick films were deposited onto standard single-mode optical fiber of a 0.125 mm diameter by dip-coating method. Film deposition included three steps: (i) Deposition of 3 layers of thin film with sol-gel solution (0.4M PZT) without powder. Each layer was then dried at 300°C during 30 s, (ii) Deposition of 4 layers of thick film using a solution mixed with powder and drying each layer under the same conditions. Before deposition, the slurry was ultrasonicated during 1 min and the composite slurry was deposited immediately. (iii) Infiltration procedure. It was performed similar to step 1 with more layers. After each step, the film was sintered at temperatures 600°C for 10 min, 1 hour, and 30 min, respectively. Before the PZT film deposition a thin layer of platinum was sputtered onto the fiber. The layer was used as a bottom electrode for electrical measurements. The top electrode was sputtered with gold.

Phase analysis of the films was performed by x-ray diffraction (XRD). XRD data were collected at room temperature using a Rigaku D/MAX-B diffractometer with Cu Kα radiation. Film microstructure was examined by scanning electron microscopy (SEM, Hitachi S-4100). The dielectric permittivity and loss tangent of the film were measured with LCR meter (HP4284A) in a frequency range from 100 Hz to 1 MHz. Ferroelectric properties were studied by a TF Analyzer (AIXACT). Piezoelectric characteristics of the film were measured by a fiber-optic setup based on Fotonic Sensor (MTI 2000) [8].

Results and Discussion

The main problem with the deposition of PZT onto a fiber is due to a significant difference between the temperature expansion coefficient (TEC) of silica and of PZT film. This effect leads to a fracture of the fiber during cooling down when a film achieves certain thickness. This problem was alleviated by the introducing of intermediate thin layer of PZT film (step 1). After cooling down the layer was inevitably cracked [Fig. 1(a,b)] but a relief of the high stress was achieved in this way. Deposition of additional layers was not influenced by the stress, and, since their thickness was much greater (sol-gel with powder) total properties did not degrade.

As described above, PZT thick films were deposited onto a fiber via dip-coating procedure. Perovskite phase formation was confirmed by XRD studies where no additional phases were detected. Microstructure of the films was tested with a SEM and is presented in Fig. 1(c, d). The thickness of the film with 4 layers of slurry deposition was about 2 μm. The disadvantage of the as-deposited film was due to its high porosity and surface roughness. In order to decrease the porosity and roughness of the films, 5 layers of infiltration with the same solution was used. After infiltration procedure the quality of the film was considerably improved [Fig. 1(e, f)].

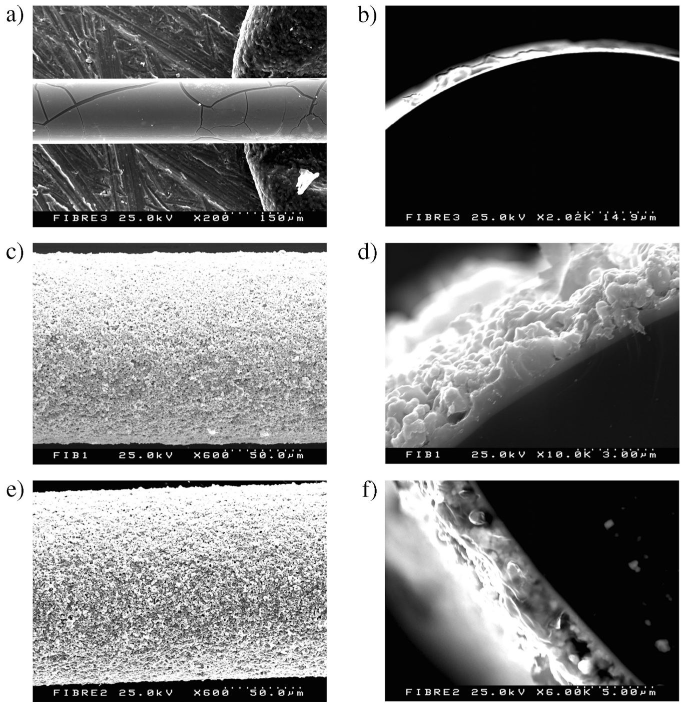

Figure 1. Surface microstructure (a, c, e) and cross-section (b, d, f) of PZT films deposited on optical fiber. (a, b) Intermediate layer after cooling down, (c, d) thick film without infiltration, (e, f) after 5 layers of infiltration.

A bottom electrode was sputtered with platinum onto one side of the bare fiber (see Fig. 2). The thickness of the electrode was close to 80 nm. After the film deposition a top electrode was sputtered with gold onto the same side of fiber forming a capacitor structure. Schematic of piezoelectric coating and bending stress of the fiber is presented in Fig. 2 (a). The length of the fiber was 6mm and its diameter was 0.125 mm. The length of the top electrode was 2 mm, and distance between the top electrode and the clamping point was 2 mm.

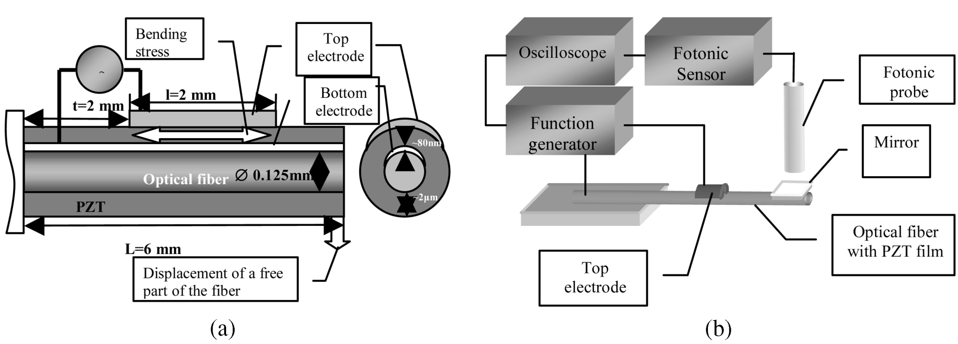

Figure 2. Schematic of piezoelectric coating and bending stress of the fiber (a) and measurement setup of voltage-induced bending displacements (b).

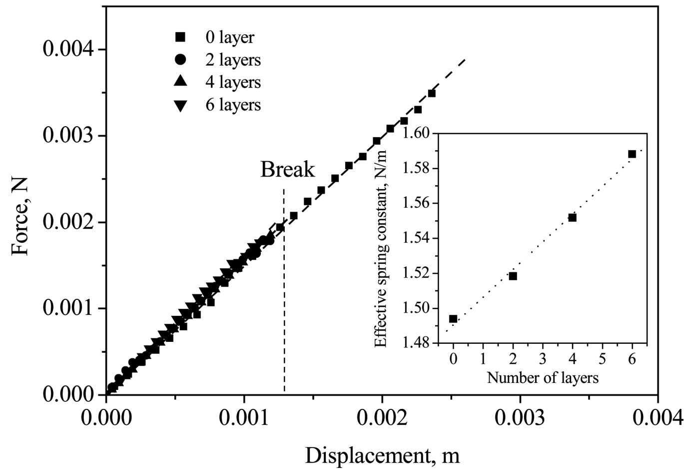

Figure 3. Displacement vs. force measurement on coated fibers with different number of PZT layers (Inset - effective spring constant vs. number of layers).

An experimental set up for the measurement of the voltage-induced bending displacements is shown in Fig. 2(b). The studied sample was clamped at one side by the sample holder, whereas the other edge was free allowing piezoelectrically induced deformation. An ac voltage of different amplitudes and frequencies was applied to the sample with the function generator. The displacement of the free end of the fiber was controlled by the Fotonic Sensor. In order to measure the displacement a small mirror was glued to the very end of the fiber. Since bottom and top electrodes were sputtered along axis of a fiber but covered only one side of the fiber a bending stress should cause significant displacements of the fiber's end [Fig. 2(a)] .

To study the mechanical properties of the coated fiber, a controlled force was applied to its free end in the direction perpendicular to axis of the fiber and the displacement of the fiber's end was measured. Figure 3 shows measured displacement as a function of applied force for fibers with different number of the deposited PZT layers. The dependence is close to linear up to breaking of the sample. An effective spring constant of the fiber with different number of PZT layers was calculated from deformation curves. Increasing number of PZT layers deposited onto a fiber led to a linear increase of the spring constant (Inset to Fig. 3). It allows calculating an effective elastic modulus of each PZT layer. However, the spring constant of a silica fiber is much greater than that of thin layer of PZT film and resonance frequency is mainly determined by the properties of the fiber.

Ferroelectric characteristics were measured at the frequency 400 Hz. Coercive field and remanent polarization of the film is 50 kV/cm and 18 $\mu C/cm^2$, respectively. Dielectric properties were measured within a wide frequency range from 100 Hz to 1 MHz with ac voltage of 1 V amplitude. This voltage was below the voltage at which ferroelectric hysteresis was observed. In principle, the system film + electrodes represents a part of cylindrical capacitor. However, in our case (distance between electrodes is much lower then radius of their curvature) the capacitor can be considered as the plane one.

Dielectric permittivity was found to decrease with increasing frequency of ac voltage (Fig. 4). This dispersion is typical for PZT films and can be explained within existing models [9]. For thick PZT film prepared by hybrid sol-gel route this dispersion was also observed by Barrow et al. [10].

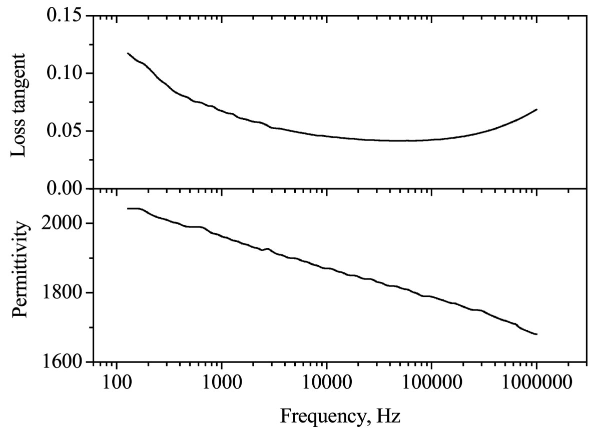

Figure 4. Frequency dispersion of dielectric permittivity and loss tangent of PZT film on a fiber.

Before the piezoelectric measurements the samples were poled with a dc voltage of 30 V, which is higher then the coercive field. No breakdown of the PZT was observed even for long poling time (10 min).

If an electric field is applied perpendicular to the film surface, the piezoelectric film layer will shrink or expand, depending on the direction of the field with respect to that of the net polarization in the film. This mechanical strain leads to bending of the fiber. The mechanical influence of the electrodes and piezoelectric layer can be neglected since their thickness is much less then the total thickness.

A coated fiber with the clamped end can be considered as a cylindrical cantilever. The resonance frequencies of a clamped cantilever with a homogeneous cross-section can be, in principle, calculated based on the equation of motion for flexural vibrations [11]. The fundamental resonance frequency is determined by the dimensions of the cantilever, its modulus of elasticity, and boundary conditions. However, due to the presence of a mirror (additional mass that adds to the distributed mass of the cantilever) the resonance frequency is shifted toward low frequencies and it cannot be calculated based on the equation of motion and thorough numerical simulations are needed. The measurements of the amplitude and phase of ac displacements vs. frequency of driving voltage ($V_{ac} = 5$ V) are presented in Fig. 5. A strong peak of amplitude and phase change is observed at 950 Hz (resonance frequency). The displacement of free fiber end at resonance frequency achieves 150 nm at applied ac voltage 5 V.

Figure 6 shows the effective $(d_{33})_{eff}$ hysteresis loops of studied PZT thick film deposited onto a fiber. In this measurement, the consecutive voltage pulses (5 s duration) were swept in the range -30 V $< U < +30$ V by the computer and the deformation under a small ac voltage (amplitude 5 V) was measured in between. The $(d_{33})_{eff}$ coefficient is coupled to the polarization, therefore it reflects saturation of polarization for both poling directions. Changing the $(d_{33})_{eff}$ sign (180° phase shift between applied voltage and deformation) corresponds to the polarization switching. This type of hysteresis provides full information on the piezoelectric behaviour of the sample. The measurements were carried out at the frequency 400 Hz. The maximum value of effective piezoelectric coefficient was found to be around 5500 pm/V. In principle, $(d_{33})_{eff}$ is much larger than that of the PZT film itself. It is related to d_{31} coefficient of piezoelectric film and geometrical dimensions of the device.

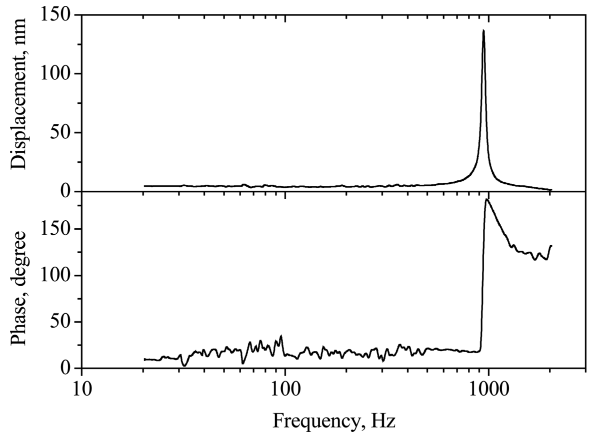

Figure 5. Amplitude and phase of ac displacements vs. frequency of driving voltage (Vac = 5V).

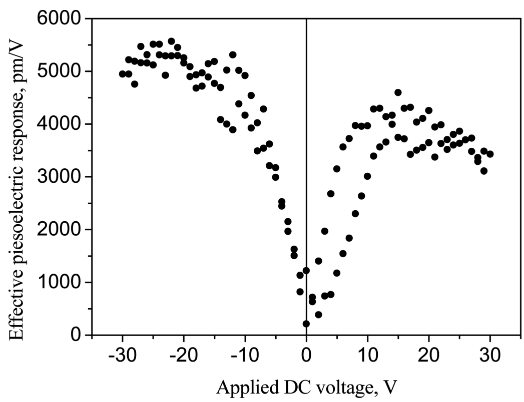

Figure 6. $(d_{33})_{eff}$ hysteresis at low frequency (400 Hz, 5 V) for both positive and negative bias.

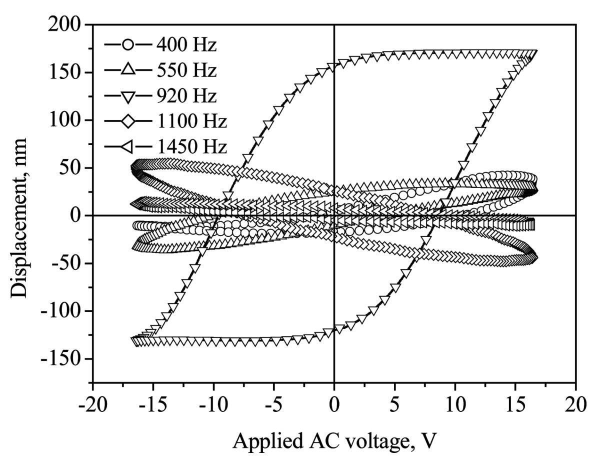

Figure 7. Strain-field hysteresis at different frequencies both above and below resonance (Vac = 16 V).

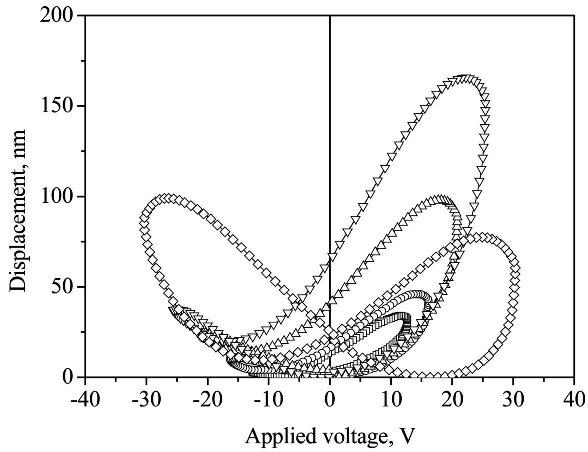

Figure 8. Strain-field hysteresis of a fiber tip under different maximum voltages (400 Hz). (Round tips are due to inertia mass of the mirror)

The deformation loops of the film were measured under both low (lower than coercive voltage) and high ac voltage. First, bipolar deformation loops were measured under a relatively low voltage both below and above the resonance frequency (Fig. 7). Using this data, the effective d_{33} coefficient can be evaluated by the slope of the loops at different frequencies.

With increasing applied voltage at low (at least two times lower then resonance frequency) frequency (400 Hz) the switching of polarization and typical butterfly loops were observed (Fig. 8). Some asymmetry observed in the loops taken at intermediate voltages is due to contribution of linear part (piezoelectric effect), and incomplete switching for one of the polarization directions. It was found that the asymmetry of the strain loops depends on initial poling of the sample. Increasing the amplitude of driving voltage leads to symmetrical butterfly loops with the shape expected from this composition. The butterfly-type hysteresis is a higher order electromechanical effect and is very important for the evaluation of the performance of ferroelectric films in microelectromechanical applications.

Conclusion

Hybrid PZT films (2 μm thickness) were successfully deposited onto standard telecommunication optical fibers by modified sol-gel method. The films exhibited high dielectric and ferroelectric properties similar to those of PZT films on planar substrates. Maximum displacement of ~150 nm under 5V driving field was achieved at resonance (950 Hz). Effective piezoelectric coefficient $(d_{33})_{eff}$ ~5500 pm/V was measured under a weak ac-field. The $(d_{33})_{eff}$ value could be tuned by the dc field.

Acknowledgments

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