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Effect of post-deposition annealing on transverse piezoelectric coefficient and vibration sensing performance of ZnO thin films

Sudeep Joshi[a], M.M. Nayak[b], K. Rajanna[a,*]

[a] Department of Instrumentation and Applied Physics, Indian Institute of Science, Bangalore 560012, India
[b] Centre for Nano Science and Engineering, Indian Institute of Science, Bangalore 560012, India

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ABSTRACT

The present experimental study investigates the influence of post-deposition annealing on the transverse piezoelectric coefficient (d_{31}) value of ZnO thin films deposited on a flexible metal alloy substrate, and its relationship with the vibration sensing performance. Highly c-axis oriented and crystalline ZnO thin films were deposited on flexible Phynox alloy substrate via radio frequency (RF) reactive magnetron sputtering. ZnO thin film samples were annealed at different temperatures ranging from 100 °C to 500 °C, resulting in the temperature of 300 °C determined as the optimum annealing temperature. The crystallinity, morphology, microstructure, and rms surface roughness of annealed ZnO thin films were systematically investigated by X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Atomic Force Microscopy (AFM), respectively. The piezoelectric d_{31} coefficient value was measured by 4-point bending method. ZnO thin film annealed at 300 °C was highly c-axis oriented, crystalline, possesses fine surface morphology with uniformity in the grain size. This film showed higher d_{31} coefficient value of 7.2 pm V^{-1}. A suitable in-house designed and developed experimental set-up, for evaluating the vibration sensing performance of annealed ZnO thin films is discussed. As expected the ZnO thin film annealed at 300 °C showed relatively better result for vibration sensing studies. It generates comparatively higher peak output voltage of 147 mV, due to improved structural and morphological properties, and higher piezoelectric d_{31} coefficient value.

1. Introduction

Thin films deposited on flexible substrates opens a new area of research for wide range of applications in the fields like flexible and printed electronics, organic electronics, Thin Film Transistors (TFTs), smart skin, energy harvesting, sensors, and actuators technology. Recently, several research groups have demonstrated various application aspects of piezoelectric thin films deposited on flexible substrates, with numerous added advantages over conventionally employed substrates [1–3]. A few investigators have employed polymer type flexible substrates, such as polycarbonate [4], Poly Ethylene Terephthalate (PET) [5], Poly Propylene Adipate (PPA) [6], and Poly Tetra Flouro Ethylene (PTFE) [7] for thin film deposition. These polymer flexible substrates suffer disadvantages, such as a bottom metallic electrode layer has to be deposited to form an MIM (Metal Insulator Metal) type structure during sensing applications. Moreover, they cannot withstand higher temperatures during deposition. The metal alloy flexible substrates seems to be more advantageous than polymer flexible substrates due to, lower thermal expansion coefficient, higher impact resistance, and good mechanical strength with exceptional spring properties [8]. Therefore, for the present experimental study, a flexible metal alloy (Phynox) was selected as a substrate for the deposition of piezoelectric thin films.

The structural properties, surface morphology, microstructure, and piezoelectric coefficient value of deposited thin film shows strong dependence on the substrate material. Therefore, it is of immense importance to optimize the properties of thin film for the substrate material under consideration for the intended application. The evaluation of piezoelectric coefficient of thin film is of extreme importance for assessing its suitability for various applications in the field of sensors and actuators technology. There are a few reports, which investigate the dependence of piezoelectric coefficients on sputtering process parameters, such as substrate temperature [9], and Ar/O_2 gas ratio [10]. Till now there is no report on the effect of post-deposition annealing treatment on the piezoelectric coefficient value of thin films. The piezoelectric coefficient value of thin film will determine its performance characteristics, and the extent of its application. Therefore, considerable amount of research experimentation has to be carried out in order to study

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* Corresponding author. Tel.: +91 80 22933188;
fax: +91 80 23600135/+91 80 23600683.
E-mail addresses: kraj@isu.iisc.ernet.in, krajanna2011@gmail.com, krajanna2003@yahoo.com (K. Rajanna).

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the effect of annealing temperature on the piezoelectric coefficient value of thin films.

Generally, the vibration sensing is performed with sensors based on the piezoelectric principle. There are various materials which exhibit piezoelectric property in thin film form, such as ZnO, PZT, GaN, AlN, $LiNbO_3$, and $BaTiO_3$. Zinc Oxide (ZnO) has hexagonal wurtzite crystal structure, which is one of the most stable forms, and lacks the inversion symmetry, due to which it possesses piezoelectric property. The selection of ZnO thin film for the present experimental study was motivated due to its ease of deposition on wide variety of substrates, excellent substrate adherence [11], and it is a compositionally simple material [12]. ZnO thin film is examined to a greater extent because of its excellent piezoelectric, mechanical, and optical properties [13]. Moreover, due to its self poling ability the ZnO thin film does not require any thermal-electric poling, unlike PZT (Lead Zirconate Titanate), and PVDF (Polyvinylidene Fluoride) [14]. When compared to AlN (Aluminum Nitride) thin films, ZnO thin film possesses higher value of piezoelectric coupling coefficient [15]. In case of ZnO thin film deposition, various properties such as the stoichiometry, texture, and preferred orientation can be controlled with ease [16]. Generally, ZnO thin film possessing good piezoelectric property, exhibits higher value of piezoelectric coefficients. ZnO thin films have been applied in wide range of applications, such as Surface Acoustic Wave (SAW) devices [17], Film Bulk Acoustic Resonator (FBAR) [18], solar cells [19], and optoelectronics devices [20]. Previously, a few researchers have studied the effect of post-deposition annealing on the structural and optical properties of ZnO thin films [11,21–23]. However, the effect of post-deposition annealing on the transverse piezoelectric coefficient (d_{31}) value and its relationship with the vibration sensing performance has not been reported.

In the present experimental study, ZnO thin films were deposited on flexible Phynox substrate by RF reactive magnetron sputtering technique. The as-deposited ZnO thin film samples were annealed at the temperatures ranging from 100 °C to 500 °C. This paper particularly investigates the effect of post-deposition annealing on the transverse piezoelectric coefficient (d_{31}) value of ZnO thin film. Furthermore, in order to experimentally verify piezoelectric properties of annealed ZnO thin films, an in-house designed and developed test set-up was employed for vibration sensing studies. It is to be noted that, this is the first attempt and a comprehensive study on the influence of post-deposition annealing on the transverse piezoelectric coefficient (d_{31}) value, and vibration sensing performance of ZnO thin films deposited on a flexible metal alloy substrate.

2. Experimental

2.1. Deposition and annealing of ZnO thin film on flexible Phynox substrate

In the present experimental study, Phynox (Lamineries, MATTHEY SA), which is an austenitic nickel-chromium-cobalt based alloy, was employed as a substrate material for ZnO thin film deposition. The selection of Phynox alloy was motivated from the facts that, it is electrically, and thermally conductive substrate. Moreover, it also has exceptional spring properties that make it a suitable candidate for its use as a sensing element [8]. Several other important properties of the Phynox alloy are listed in Table 1 [24]. Phynox is a biocompatible alloy hence it is widely used as an electrode material in pacemakers, and in implant components [25]. Recently, a novel gas flow sensor [24], an impact sensor [25], and micro actuation application [26] using piezoelectric ZnO thin films deposited on Phynox substrate has been demonstrated by our research group. (Please see Appendix A for the selection of Phynox alloy over other commonly used metal/metal alloy flexible substrates for thin film deposition, at the end of the manuscript.)

Table 1
Properties of the flexible Phynox alloy substrate.

Sr. no.	Properties	Values
1	Ultimate tensile strength (UTS) ($N\,mm^{-2}$)	2600
2	Modulus of elasticity ($kN\,mm^{-2}$)	220
3	Yield strength ($N\,mm^{-2}$)	2200
4	Poisson ratio	0.3
5	Thermal conductivity at 20 °C ($W\,m^{-1}\,K^{-1}$)	12.5
6	Electrical resistivity ($\mu\Omega$ cm)	95
7	Temperature range	−268.8 °C to 500 °C
8	Magnetic properties	Non-magnetic

Table 2
Optimized sputtering process parameters maintained for the deposition of good quality piezoelectric ZnO thin films.

Sr. no.	Sputtering process parameters	Values maintained
1	Ultimate pressure (mbar)	1×10^{-6}
2	Working pressure (mbar)	0.035
3	Target to substrate distance (mm)	55
4	Deposition time (min)	60
5	$Ar:O_2$ ratio (%)	90:10
6	Substrate temperature (°C)	R.T.[a]
7	Applied RF power (W)	100

[a] R.T. = Room temperature.

The Phynox alloy was properly cut into rectangular shaped substrates of dimensions—length 50 mm and width 5 mm using a wire-cut Electric Discharge Machine (EDM). The standard substrate cleaning procedure using organic solvents (acetone/iso-propyl alcohol) was followed prior to loading the Phynox substrates into the chamber for thin film deposition. Piezoelectric ZnO thin films were deposited by RF reactive magnetron sputtering technique. The optimized sputtering process parameters maintained for the deposition of good quality piezoelectric ZnO thin films are listed in Table 2. The as-deposited ZnO thin film samples were annealed in a conventional furnace within the temperature range of 100 °C to 500 °C, with an interval of 100 °C for the duration of 3 Hours. An accurate slow cooling rate has to be maintained during annealing studies, in order to avoid the possibility of any stress, and strain development in the thin film sample [15]. Therefore, in the present experimental work, cooling rate of about 2 $°C\,min^{-1}$ was precisely maintained for all samples.

2.2. In-house developed experimental set-up for vibration sensing studies

Fig. 1 shows the schematic diagram of the ZnO thin film deposited cantilever element employed for vibration sensing studies. It is an MIM structure, wherein the annealed ZnO thin film is sandwiched in between two thin metal electrodes. The Phynox substrate (thickness: 40 μm) itself acts as a bottom electrode. The annealed ZnO thin film of thickness (700 ± 30 nm) acts as a sensing layer. A top electrode of silver thin film (thickness: 100 nm) was deposited by RF magnetron sputtering on the annealed ZnO film. In order to have a comparative study between different annealed ZnO thin film samples, the ZnO film thicknesses were kept same for all the deposited samples.

Vibration sensing studies were performed in order to experimentally verify the structural, morphological, and piezoelectric properties of annealed ZnO thin films. Fig. 2 shows the schematic diagram of in-house designed, and developed experimental test set-up employed for vibration sensing studies. It consists of a piezoelectric actuator, function generator (Agilent 33220A), digital

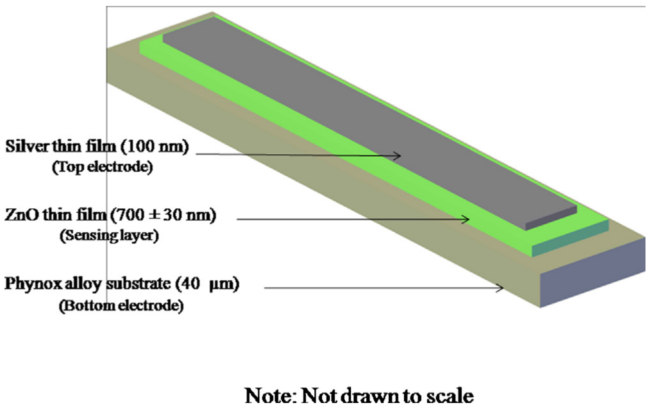

Fig. 1. Schematic diagram of the cantilever element for vibration sensing studies.

storage oscilloscope (Yokogawa DLM-2022), and annealed ZnO thin film deposited cantilever sample. The annealed cantilever samples were firmly mounted on to the piezoelectric actuator with an adhesive double sided tape. The piezoelectric actuator was the source of vibration for mounted cantilever samples. In order to allow the comparison, all the cantilever samples (as-deposited and annealed) were given same magnitude of vibration from the piezoelectric actuator (supply voltage of 10 V p–p at a frequency of 2 kHz). In order to achieve higher accuracy and repeatability during experiment, measurements were repeated for 5 times for a particular annealed cantilever sample.

3. Results and discussions

3.1. Crystallographic characteristics of annealed ZnO thin films on Phynox substrate

Fig. 3 shows the X-ray diffraction spectra of the as-deposited ZnO thin film, and of films annealed at different temperatures. The X-ray diffraction studies were performed by using Bruker D8 Advance X-ray diffractometer. The peak at about 34.4° corresponds to the diffraction from (0 0 2) plane of ZnO. The presence of (0 0 2) peak indicates that, the films have a strong *c*-axis orientation perpendicular to the substrate. The as-deposited ZnO film was having relatively low intensity of (0 0 2) peak, and higher value of Full Width at Half Maxima (FWHM), which makes it a comparatively poor crystalline film. When ZnO films were annealed at the temperatures of 100 °C, and then at 200 °C, an increasing trend in the (0 0 2) peak intensity was observed, and FWHM value starts decreasing. When the annealing temperature was increased to 300 °C, the (0 0 2) peak intensity becomes maximum, with lowest FWHM value. This is attributed to the better crystallinity of the film with larger grain size. This in turn indicates that, *c*-axis of the hexagonal phase of ZnO is perpendicular to the substrate, which is an indication of good quality piezoelectric thin film [13]. This high degree of *c*-axis orientation, and (0 0 2) preferred crystalline structure enhances piezoelectric properties of ZnO thin films [13]. At higher annealing temperatures of 400 °C, and 500 °C, the decreasing trend in (0 0 2) peak intensity was observed, whereas the FWHM value was relatively increased. This shows that, higher annealing temperatures deteriorate the piezoelectric properties of ZnO thin films deposited on Phynox substrate. The lattice parameters of annealed ZnO films were found to be $a = 3.35$ Å and $c = 5.22$ Å as denoted by ICDD (International Centre for Diffraction Data) (PDF No. 751533), which are comparable to the values reported for good quality piezoelectric ZnO films [27]. In the Fig. 3, peaks at 43.5° and 74.6° corresponding to bare Phynox substrate (i.e. without ZnO thin film deposition) were present in all the samples. These peaks

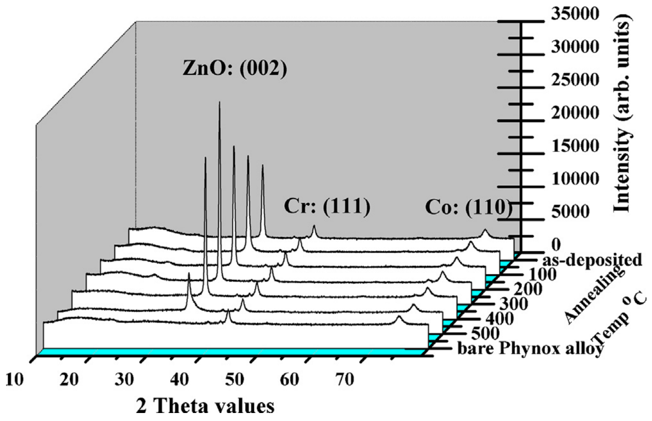

Fig. 3. X-ray diffraction spectra of the as-deposited ZnO thin film and of films annealed at different temperatures.

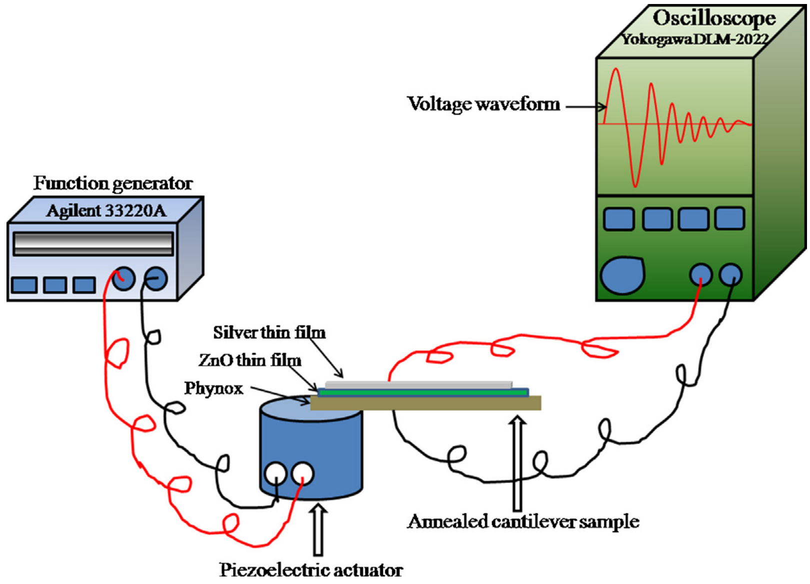

Fig. 2. Schematic diagram of the in-house designed and developed experimental set-up for vibration sensing studies.

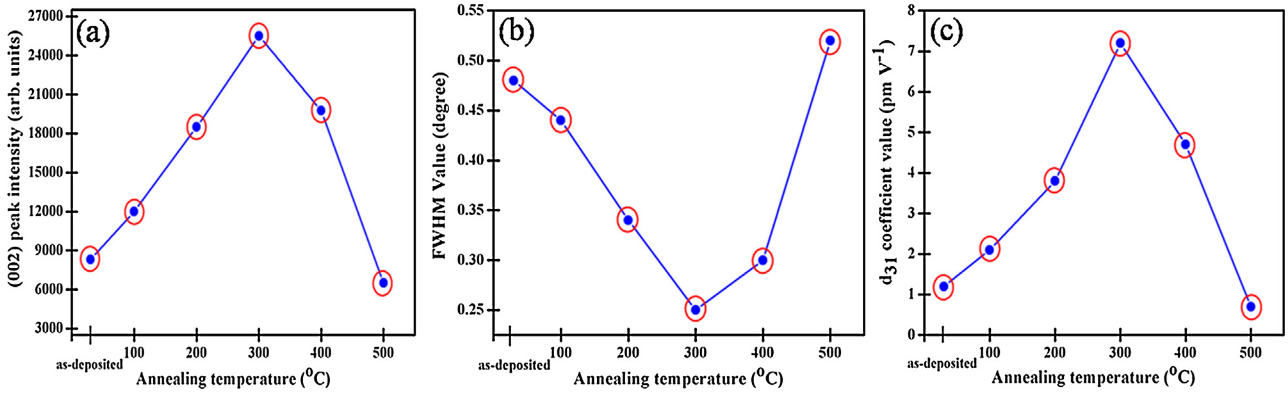

Fig. 4. (a) Variation of X-ray diffraction intensity of (0 0 2) peak, (b) FWHM value of (0 0 2) peak, and (c) d_{31} coefficient value as a function of annealing temperature.

corresponds to (1 1 1), and (1 1 0) planes of chromium, and cobalt, respectively, (ICDD, PDF No. 882323, and 011278, respectively), as they are the major constituents of Phynox alloy [24,25].

Fig. 4(a)–(c) shows the variation of X-ray diffraction intensity of (0 0 2) peak (Fig. 4(a)), FWHM value (Fig. 4(b)), and piezoelectric d_{31} coefficient value (Fig. 4(c)) as a function of annealing temperature. The piezoelectric d_{31} coefficient value was measured by 4-point bending method (aixACCT 4-point bending system, model: aix4PB). The as-deposited ZnO film was having lower value of (0 0 2) peak intensity, with comparatively higher FWHM value, and lesser d_{31} coefficient value. When the annealing temperature was increased to 100 °C, and then to 200 °C, the (0 0 2) peak intensity, and d_{31} coefficient value showed an increasing trend, whereas the FWHM value decreased considerably. When the annealing temperature was further increased to 300 °C, the (0 0 2) peak intensity, d_{31} coefficient value were maximum, and FWHM gets reduced to minimum value.

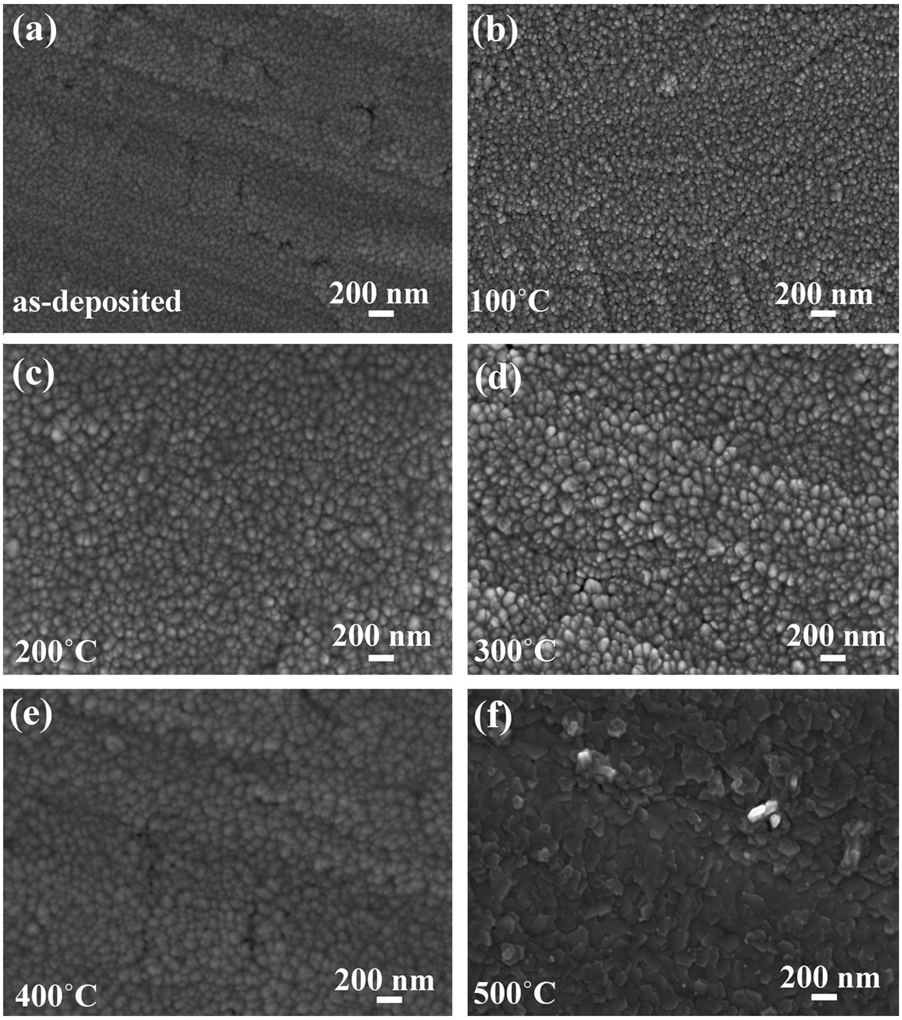

Fig. 5. (a)–(f) FESEM images of ZnO thin films deposited on Phynox substrate at different annealing temperatures: (a) as-deposited, (b) 100 °C, (c) 200 °C, (d) 300 °C, (e) 400 °C, and (f) 500 °C.

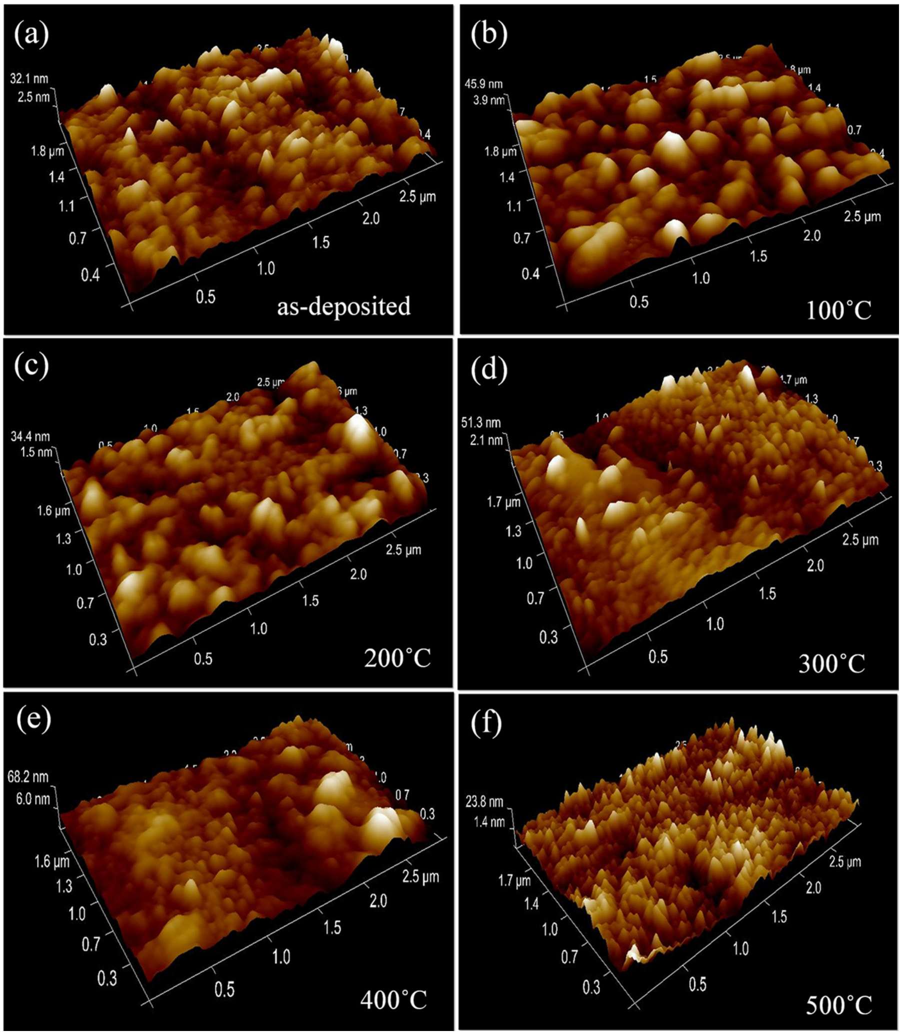

Fig. 6. (a)–(f) 3D AFM images of ZnO thin films deposited on Phynox substrate at different annealing temperatures: (a) as-deposited, (b) 100 °C, (c) 200 °C, (d) 300 °C, (e) 400 °C, and (f) 500 °C.

The reason for above observation is attributed to the higher crystallinity, and better *c*-axis orientation. Moreover, the lower FWHM value at 300 °C (Fig. 4(b)), indicates the increase in grain size of annealed ZnO film resulting in the higher piezoelectric d_{31} coefficient value [28]. However, at much higher annealing temperature of 500 °C, the (0 0 2) peak intensity, and d_{31} coefficient value were minimum, and FWHM value was maximum (it is clearly evident in Fig. 3 also). This decrease in d_{31} coefficient value was the result of increased FWHM value that indicates smaller grain size, deterioration in the degree *c*-axis orientation, and poor crystallinity.

3.2. Evaluation of surface morphology and microstructure of annealed ZnO thin films

Fig. 5(a)–(f) shows FESEM images of the as-deposited ZnO thin film, and of films annealed at different temperatures. The surface morphology and microstructure of annealed ZnO thin films were examined with Field Emission Scanning Electron Microscope (FESEM), (Ultra 55 Karl Zeiss). As can be seen from images, the surface morphology, and microstructure of ZnO thin films annealed at different temperatures were quite different. The surface of as-deposited ZnO film (Fig. 5(a)) appears to be considerably rough, and lumpy. The film possesses grains of very small size, and without proper structural development, which is evident due to the presence of microvoids. This is attributed to the inadequate supply of kinetic energy to ad-atoms, which is required for surface mobility resulting in the proper diffusion. Such types of films are not desirable for device applications. As can be seen in Fig. 5(b) and (c), when the films were annealed at 100 °C, and 200 °C, there was a relative improvement in the surface morphology. There was an apparent growth in the grain size, which fills up the residual space and results in the proper structural development. In case of the film annealed at a temperature of 300 °C (Fig. 5(d)), the surface of film was free from defects and it appears to be excellently smooth as compared to films annealed at relatively lower temperatures. The grains were well oriented with more uniform and larger grain size of about 75–80 nm. The film has dense microstructure with clearly defined grain boundaries. ZnO thin film annealed at 300 °C also possesses higher piezoelectric d_{31} coefficient value of 7.2 pm V^{-1}. The possible reason for improvement in the structural properties is the supply of suitable amount of thermal energy, which in turn enhances the mobility of ad-atoms on the substrate surface. This thermal energy in turn accelerates the relocation of ad-atoms to the suitable site resulting in the proper structural development of the film. Hence, there was an enhancement in the grain size, which results in the increase of piezoelectric d_{31} coefficient value [28]. At much higher annealing temperature of 500 °C (Fig. 5(f)), the film surface started developing cracks. The deposited film started peeling off from the substrate surface. As a result, it was having poor adherence with the substrate and possesses extremely higher

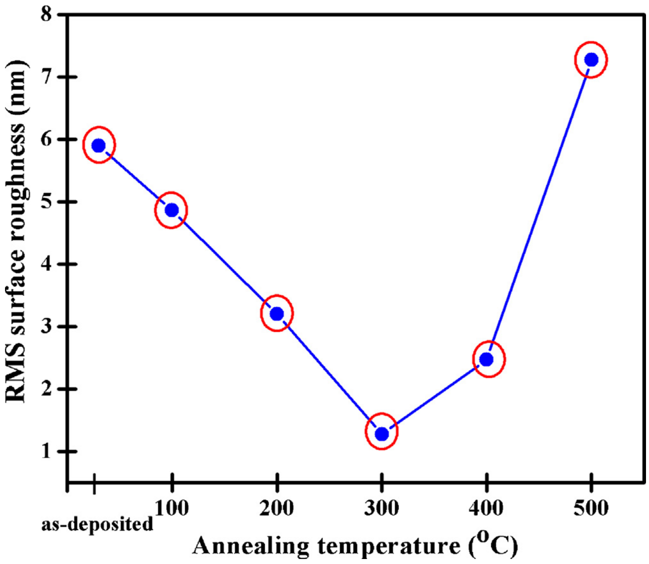

Fig. 7. Variation of the rms surface roughness value (nm) of ZnO thin films with respect to the annealing temperature (°C).

roughness (it is also evident from the AFM image, see Fig. 6(f)). ZnO film annealed at 500 °C was having very low d_{31} coefficient value of 0.7 pm V^{-1}.

3.3. *Evaluation of surface roughness of annealed ZnO thin films using AFM*

Fig. 6(a)-(f) shows 3D AFM images of the as-deposited ZnO thin film, and of films annealed at different temperatures. The surface roughness was measured using AFM (Dimension ICON with ScanAsyst2). As can be seen from images, the films with different surface roughness were obtained at different annealing temperatures. The surface roughness is a dominant factor that influences piezoelectric d_{31} coefficient value, and therefore the sensing properties of ZnO thin film. The as-deposited ZnO film (Fig. 6(a)) was having rms surface roughness value of about 5.9 nm due to improper grain growth that contributes toward porosity, and hence increases the surface roughness. As the annealing temperature increases, the surface roughness value decreases, and reaches its minimum value of 1.28 nm at the temperature of 300 °C (Fig. 6(d)). This is attributed to the optimum supply of thermal energy, which results in the grain growth, and hence contributes toward proper structural development of the ZnO film. The film with highest surface roughness value of 7.28 nm was obtained at the annealing temperature of 500 °C (Fig. 6(f)). This is attributed to the peeling off, of the deposited ZnO film at such high values of temperature, as discussed previously during SEM image analysis. Fig. 7 shows the variation of rms surface roughness value as a function of annealing temperature. As can be seen for the entire range of annealing temperature variation, the nature of surface roughness curve is exactly opposite to the d_{31} coefficient value curve (see Fig. 4(c)). Therefore, lower the rms surface roughness value, higher is the piezoelectric d_{31} coefficient value and the ZnO thin film possess better sensing properties.

Material characterization studies reveals that, the film annealed at a temperature of 300 °C possesses high crystallinity, lower value of FWHM and surface roughness, uniform and well oriented grains with higher piezoelectric d_{31} coefficient value. Higher the value of piezoelectric coefficient of ZnO film better is the sensing properties. Therefore, in the present experimental study, the quality of ZnO thin film deposited on Phynox substrate improves, and reaches its finest quality at the optimum annealing temperature of 300 °C. At much higher annealing temperature of 500 °C, the film quality degrades due to poor crystallinity, development of cracks, higher FWHM and surface roughness value, and lower d_{31} coefficient value.

3.4. *Vibration sensing studies on annealed ZnO thin films*

In the present experimental work, five cantilever samples annealed at same temperature were tested for the vibration sensing studies. The samples annealed at a particular temperature have generated almost similar amplitude for the first peak of output voltage. Therefore, the representative output voltage waveform generated from one of the samples is reported in the paper. Fig. 8(a)–(d) shows the output voltage responses of annealed ZnO thin film deposited cantilever samples due to vibrations from the piezoelectric actuator. Initially, as the actuator was turned on the cantilever sample vibrates freely. When the actuator was suddenly de-energized, the cantilever element comes to rest position due to the elastic restoring force. The mechanical vibrations of the cantilever sensing element get damped out after the actuator was turned off. As a result of these mechanical vibrations, a piezoelectric potential difference is generated in between top electrode (silver thin film), and bottom electrode (Phynox substrate). This piezoelectric potential difference will cause flow of electrons from the top electrode to the bottom electrode, and generates an output voltage signal. The output voltage from the ZnO film cantilever element is generated due to the bending stress induced piezoelectric effect [24]. When the actuator was turned off the cantilever vibrations get damped out, the similar trend was also followed by the output voltage signal. The output voltage amplitude gets damped out in an exponential manner (see Fig. 8(a)–(d)). Hence, the generated electrical output voltage signal faithfully follows the mechanical vibrations of the cantilever sensing element. The generated output voltage signal is an exponentially decaying sinusoidal waveform, which can be described by the following relation:

$$V = A\exp(-t)\sin(\omega t) \tag{1}$$

where A = amplitude of the peak, t = time on x-axis, and ω = frequency. The ZnO thin film cantilever annealed at 100 °C generates relatively lower amplitude of the first peak of 42 mV (Fig. 8(a)). This is due to the lower d_{31} coefficient value of 2.1 pm V^{-1}, which indicates poor piezoelectric property. The ZnO film annealed at a temperature of 200 °C (Fig. 8(b)) generates relatively higher amplitude of the first peak of 92 mV. As expected the ZnO thin film deposited cantilever sample annealed at 300 °C, showed relatively better result for the vibration sensing. It has generated highest amplitude of the first peak of 147 mV (Fig. 8(c)). This is attributed to the higher d_{31} coefficient value of 7.2 pm V^{-1}, due to the improved structural, morphological, and piezoelectric properties. The ZnO thin film annealed at a relatively higher temperature of 400 °C (Fig. 8(d)), generates comparatively lower amplitude of the first peak of 107 mV. When the annealing temperature was much higher of about 500 °C (figure not shown), the cantilever sample exhibited very poor vibration sensing performance, as the amplitude of first peak was drastically reduced. Hence, higher the amplitude of the first peak of output voltage waveform better is the vibration sensing performance of annealed ZnO thin film due to better piezoelectric property.

The annealed ZnO thin film cantilevers, when subjected to the bending due to vibrations from the actuator, a longitudinal tensile stress is generated in the deposited ZnO thin film. This longitudinal tensile stress in turn generates an electric field, and output voltage along the vertical direction. Hence, in the present case, d_{31} piezoelectric coefficient is responsible for the vibration sensing. The amplitude of first peaks of output voltage waveforms was averaged out over the results obtained from five samples for a

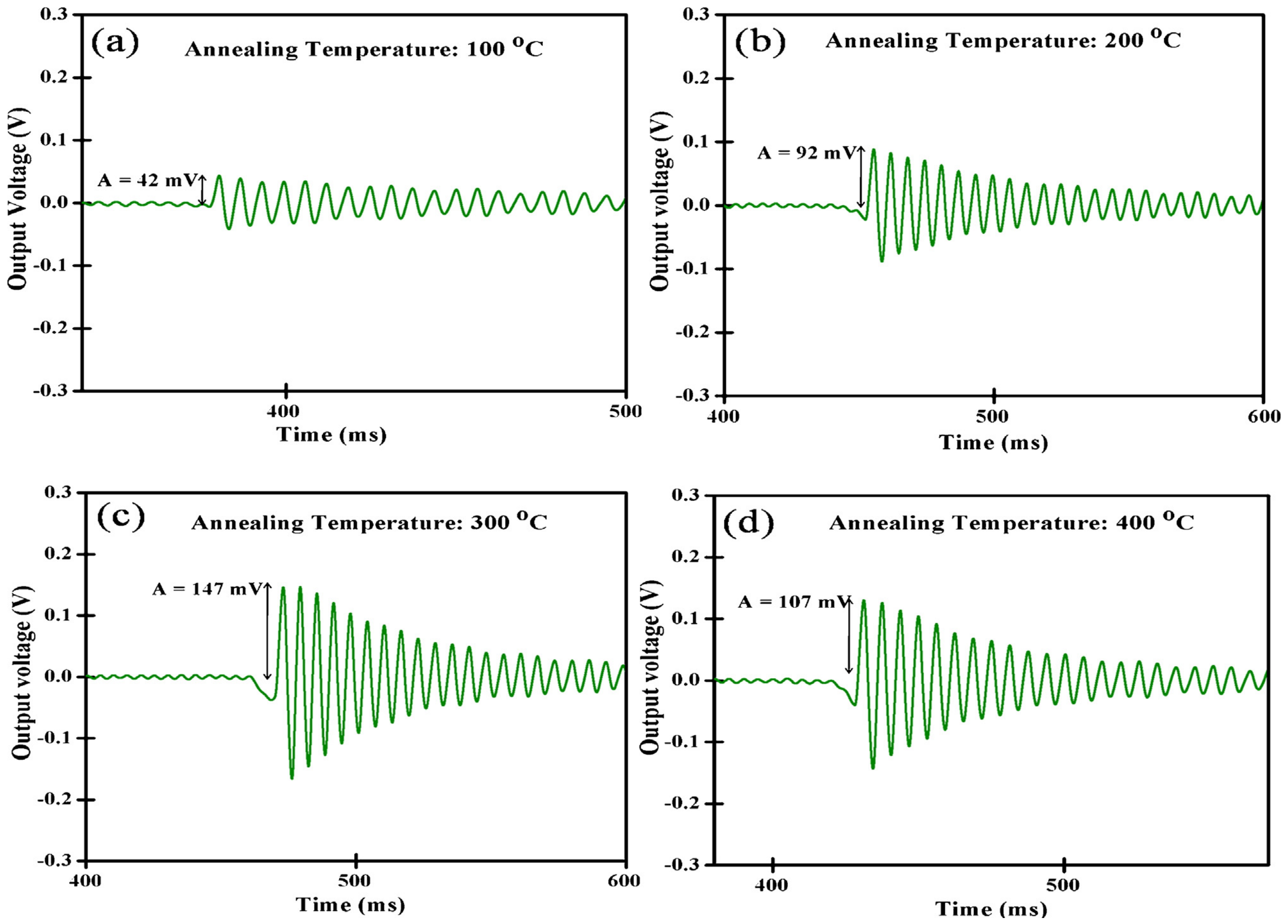

Fig. 8. (a)–(d) Output voltage responses of annealed ZnO thin film cantilever samples as obtained from vibration sensing studies: (a) 100 °C, (b) 200 °C, (c) 300 °C, and (d) 400 °C.

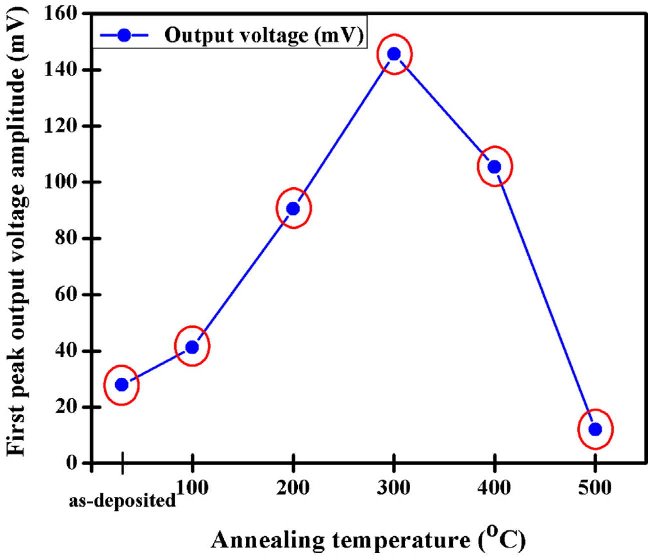

Fig. 9. Variation of the averaged amplitude of first peak of output voltage (mV) with respect to the annealing temperature (°C).

particular annealing temperature. Fig. 9 shows the variation of the averaged amplitude of the first peaks of output voltage waveforms with respect to the annealing temperature. As can be seen over the entire range of annealing temperature variation, the nature of curve of the output voltage, and d_{31} coefficient value (Fig. 4(c)) follows exactly similar trend. Therefore, higher the d_{31} coefficient value, higher is the amplitude of the first peak. As the annealing temperature approaches toward the temperature of 300 °C, maximum d_{31} coefficient value of 7.2 pm V^{-1} results in the highest peak output voltage of 147 mV. Therefore, results from the vibration sensing studies are in good agreement with the observations from material characterization studies. Hence, 300 °C is the optimized annealing temperature for obtaining good quality piezoelectric ZnO thin films on flexible Phynox substrate.

4. Conclusion

Piezoelectric ZnO thin films were deposited by RF reactive magnetron sputtering on flexible Phynox alloy substrate. The ZnO thin films were annealed in the temperature range of 100 °C to 500 °C for the duration of 3 h. The annealed films were characterized by X-ray diffraction, SEM, AFM, and piezoelectric d_{31} coefficient value measurement. ZnO film sample annealed at a temperature of 300 °C was highly crystalline having their *c*-axis perpendicular to substrate. Additionally, it was having lower surface roughness value of 1.28 nm, fine surface morphology with uniform, and larger grain size. This film sample also exhibited higher d_{31} coefficient value of 7.2 pm V^{-1}. In-house designed, and developed experimental set-up, was used for analyzing the effect of post-deposition annealing on the vibration sensing performance. The film annealed at the optimized temperature of 300 °C generated relatively higher first peak output voltage of 147 mV. At much higher annealing temperature of 500 °C, the structural, morphological, and piezoelectric properties of ZnO thin film were deteriorated, and hence results in very

Table A.1
Comparison of properties of the Phynox alloy with other commonly used flexible metal, and metal alloy substrates.

Sr. no.	Properties	Phynox	Aluminum	SS-304L
1.	Ultimate tensile strength (UTS) ($N\,mm^{-2}$)	2600	310	689
2.	Young's modulus ($kN\,mm^{-2}$)	220	68.9	190
3.	Yield strength ($N\,mm^{-2}$)	2200	276	275
4.	Melting temperature range (°C)	1450–1460	582–650	1350–1400

low d_{31} coefficient value of 0.7 pm V^{-1}. As a result this film also showed very poor vibration sensing performance. The variation of d_{31} coefficient value and amplitude of the first peak of output voltage waveform followed similar trend over the entire range of annealing temperature. The present experimental study indicates that, piezoelectric d_{31} coefficient value and vibration sensing performance of ZnO thin films are strongly affected by the variation in the annealing temperature.

Appendix A. Selection of Phynox alloy over other commonly used metal/metal alloy flexible substrates:

The reason for selecting the Phynox as a metallic alloy flexible substrate was its excellent spring properties. This property makes it a suitable candidate for the realization of various types of sensing elements, such as cantilevers and diaphragms. It is a non-magnetic alloy, hence can be easily used in the FESEM, and AFM systems for material characterization studies.

Furthermore, the comparison of the Phynox alloy with other commonly used flexible metal/metal alloy substrates like aluminum, and Stainless Steel (SS-304L) is as follows (Table A.1):

As can be seen from the above table that, in each and every aspect properties of the Phynox alloy scores over the commonly used flexible metal (aluminum), and metal alloy (Stainless Steel-304L) substrates. Therefore, Phynox is a better choice over the other flexible metal alloy substrates for the sensing element.

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