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Direct Integration of Thin Film Piezoelectric Sensors with Structural Materials for Structural Health Monitoring

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ABSTRACT

Structural health monitoring is a means for drastically decreasing the maintenance and logistical cost associated with vehicular platforms especially aircraft. A system of small piezoelectric sensors distributed throughout the vehicle will be capable of acting passively or actively to monitor the changes within a structure that presage a component failure, and they will be able to detect and localize all impacts on the structure and evaluate any damage. Piezoelectric thin films were directly integrated with structural titanium utilizing a metal-organic chemical solution approach. The optimum integration strategy yielded a process easily performed without a cleanroom and semiconductor fabrication tools.

Keywords: Ferroelectric thin films; integration; structural materials; BST; titanium

INTRODUCTION

Structural health monitoring (SHM) is a means for drastically decreasing the maintenance and logistical cost associated with the DoD's numerous vehicular platforms by optimizing maintenance schedules and isolating small problems before they become large and expensive [1]. The next generation of SHM, addressed in this research, distributes a system of small piezoelectric sensors through the vehicle. This network of sensors is not only capable of acting

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either passively or actively to monitor the subtle changes within a structure that presage a component failure but are also able to detect and localize all impacts on the structure and evaluate the damage [1–3].

SHM addresses an urgent need of our aging infrastructure. SHM sets out to determine the health of a structure by reading a network of sensors that are embedded within the structure and monitored over time. In recent years, piezoelectric wafers permanently attached to the structure have been employed to generate and detect guided waves for structural health monitoring. Piezoelectric wafer active sensors (PWAS) [2] are inexpensive, non-intrusive, un-obtrusive, and minimally invasive. They can be surface-mounted on existing structures, inserted between the layers of lap joints, or placed inside composite materials.

Most of the methods used in conventional NDE have been demonstrated experimentally with PWAS [3]. PWAS are essential elements in Lamb-wave SHM with acoustic emission, pitch-catch, pulse-echo, phased array and electromechanical (E/M) impedance methods as shown in Fig. 1. For embedded NDE applications, PWAS couple their in-plane motion, excited by the applied oscillatory voltage, through the piezoelectric effect with the Lamb-waves particle motion on the material surface. Therefore, the voltage and power required to operate a PWAS network are directly related to the thickness of the PWAS sensors to the first and the third power, respectively. For every order of magnitude the thickness decreases, the operating voltage decreases by one order of magnitude and the power decreases by three orders of magnitude [3].

Historically, PWAS are adhesively bonded to the substrate structure or incorporated between the layers of a laminated structure during fabrication. This adhesive bonding between the PWAS sensor and the structural substrate is the weak link in the sensory system as it deteriorates in time under environmental attacks (humidity, temperature cycles, etc.). Additionally, currently available piezoelectric wafers and PZT thick films have a porous ceramic structure with multi-orientation piezoelectric domains, which results in a poor piezoelectric

Figure 1. PWAS application in SHM (A) From top to bottom: Acoustic emission detection, Pitch-catch method, pulse-echo method (B) Impedance spectra signature for Lamb modes tuning peaks and structural resonances of the PWAS impedance measured in the 200–2600 kHz range on the specimen.

performance. Current PWAS require a large voltage (~ 150 V) for full activation because the electric field is the ration between applied voltage and wafer thickness which is still relatively large [3]. There is a need for thinner wafers that will work with low voltages (< 10 V). There is an acute need to develop a PWAS material that is 1) significantly thinner than existing materials, 2) substantially improve the piezoelectric coupling and response of the PWAS sensors, and 3) easily incorporated into the structural system without the need to for an environmentally susceptible bonding agent.

This research addresses the need to directly integrate the piezoelectric thin films with the structural materials (alpha titanium for this research) by developing and utilizing a metal-organic chemical solution approach to optimize the integration strategy [4]. During the integration process, it was critical to determine which methods were capable of being performed in a Depot and which were impractical. Fabrication protocols were developed during this research for the integration of the piezoelectric thin-film active sensor (PTAS). An initial investigation focused on the preparation of the surface prior to the PTAS deposition, and this was followed by a second more involved investigation concentrated on the PTAS deposition. For the first protocol, the surface roughness had to be minimized to ensure good adhesion and mechanical coupling. For this process, three polishing methods were examined: mechanical, electro-chemical and gas-cluster ion bean smoothing [5]. During the integration of PTAS three distinct processes were examined: a rapid thermal annealing (RTA) treatment, a conventional annealing treatment, and the use of a thermally grown oxide buffer layer prior to film deposition and integration.

EXPERIMENTAL

Minimal difference in results was observed based upon the pretreatment polishing schedule employed. The electrochemical polishing did not result in significantly smoother surfaces yet incurred substantially more cost, and gas-cluster ion beam surface smoothing resulted in moderately smoother surfaces but at substantially increased costs. As the roughness of 1.64 nm was reasonable for a starting material, it was decided to utilize a traditional polishing treatment. The optimum polishing schedule began with a 240 grit sandpaper and proceeded incrementally to a 1200 grit. This was followed by polishing with $9\text{ }\mu\text{m}$ slurry and finishing with a $0.03\text{ }\mu\text{m}$ slurry. This process resulted in a surface with a roughness of 1.64 nm as measured by AFM.

The $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ (BST) piezoelectric thin films with Ba/Sr compositions of 80/20 were fabricated on structural alpha titanium stubs via the metal-organic solution deposition (MOSD) film fabrication techniques [4]. Pre-deposition preparation of the substrates involved a standard multi-step cleaning process. The substrates were dipped in acetone and methanol; the bare Ti stubs were also exposed to a weak HF solution to remove native oxide, and then rinsed in

de-ionized (DI) water. A subset of titanium stubs ($1\text{ cm} \times 1\text{ cm} \times 0.25\text{ cm}$) was thermally oxidized at 500°C to grow a TiO_2 oxide buffer layer on the surface in an attempt to minimize the reaction between the PTAS and the titanium during the annealing step [6]. A trace layer of platinum was deposited on all stubs to monitor the rate and extent of reaction between the PTAS and titanium. For the MOSD BST film fabrication, barium acetate, strontium acetate, and titanium isopropoxide were used as precursors to form BST. Acetic acid and 2-methoxyethanol were used as solvents. The precursor films were spin coated onto the substrates. Particulates were removed from the solution by filtering through 0.2 mm syringe filters. Subsequent to each coat, the films were pyrolyzed at 350°C for 10 min in order to evaporate solvents and organic addenda and form an inorganic amorphous film. The spin coat pyrolyzation process was repeated until a nominal film thickness of 300 nm was achieved. Crystallinity was achieved via post-deposition annealing in an oxygen ambience. The samples with the thermal oxide and a reference set of BST/Ti stubs were annealed under standard conditions at 650, 700, 750, 800°C for 60 minutes under flowing oxygen. A final set of BST/Ti stubs without a thermal oxide layer was exposed to a rapid thermal annealing at 650, 700, 750, 800°C for 5 minutes under flowing oxygen.

The active thin films were characterized for structural, microstructural, compositional, and surface morphological properties. Specifically, the surface roughness and grain size of the constituent films and the composite heterostructure was assessed by a Digital Instrument's Dimension 3000 atomic force microscope (AFM) employing tapping mode. Rutherford backscattering spectrometry (RBS) was utilized to assess the film elemental composition, areal thickness, interdiffusion, and film-substrate interface quality. The RBS measurements were obtained using 1.2 MeV He^+ ion beams from an NEC 5SDH-2 tandem positive ion accelerator. All spectra were fit and interpreted using the program RUMP [7, 8]. Glancing angle x-ray diffraction (GAXRD) with Bruker D5005 diffractometer with thin film attachment and $\text{CuK}\alpha$ radiation at 40 kV was employed to assess film crystallinity, phase formation, and reaction rate and extent. A Hitachi S4500 field emission scanning electron microscopy (FESEM) was employed to detail the films' plan-view microstructure, compositional uniformity, phase formation, and surface morphology.

RESULTS AND DISCUSSION

Atomic Force Microscopy (AFM) was employed to monitor the surface evolution during heat treatment. It is well known that annealing thin film oxide materials has both advantageous and disadvantageous results [9]. Higher heat treatment temperatures will substantially increase the degree of film crystallinity, which increases the piezoelectric properties, but it will also lead to grain growth. Though larger grains also improve the piezoelectric properties,

Figure 2. The atomic force microscopy (AFM) results detailing the roughness and grain size as a function of the annealing temperature for samples that underwent a standard annealing treatment, had a thermal oxide layer deposited before under going a standard annealing treatment, and underwent a Rapid Thermal Annealing (RTA) treatment.

they are also responsible for the substantial roughening of the surface, making it very difficult to deposit further layer, such as electrodes, on top of the PTAS. The effects of annealing and thermal oxide layer are evident in the data shown in Fig. 2. The surface roughness as measured by AFM indicates that the growth of a thermal oxide substantially roughens the surface prior to any PTAS deposition. This is verified by the RBS data shown in Fig. 3. The final PTAS surface roughness increases substantially with increasing annealing temperature for the standard annealing conditions, but there is no observable increase in surface roughness with annealing temperature for the RTA samples. The grain size, however, increased with increasing annealing temperature for all treatment methods. The largest grain size was observed in the PTAS films grown on the titanium without a thermal oxide buffer layer. However, the rate of increase in grain size between the PTAS samples grown on a thermal oxide and those without is comparable, indicating that the observed increase in grain size under the standard annealing conditions without a thermal oxide buffer layer is largely due to the increase in grain size within the titanium substrate. The PTAS samples grown on titanium stubs via an RTA process show increased grain size but at a markedly slower growth rate than either of the other methods.

Rutherford backscattering spectrometry (RBS) was utilized to examine the atomic level interactions of the PTAS layer with the underlying substrate for all treatment conditions, and the results are shown in Fig. 3. From the RBS it is possible to directly measure the BST thicknesses at around 500 nm in all cases. The “preoxidation” of the Ti created a TiO_2 layer around 280 nm $\pm$ 100 nm thick, indicating an oxide of somewhat non-uniform thickness. The 2 nm Pt marker layer was detectable after deposition, but it was undetectable after every anneal, standard or RTA, indicating that it must have diffused easily; it may have even reacted with the Ti during the pyrolyzation step of the

Figure 3. Rutherford backscattering spectrometry results for the titanium stubs with A) BST, trace Pt layer, no thermal oxide, and standard anneal; B) BST, trace Pt layer, no thermal oxide layer, and RTA; C) BST, trace Pt layer, thermal oxide, and standard anneal; and, D) unannealed Ti stub and unannealed Ti stub with trace Pt layer.

MOSD process. For standard anneals, the BST reacted with the underlying Ti or TiO₂ in every case. The Ti samples that underwent the standard annealing treatment show a monotonically increasing reaction rate, as measured by examining the slope of the low energy side of the BST peak in Fig. 3. It appears that the thermally grown TiO₂ slightly delayed the reaction onset (from 700°C in the standard anneal case to 750°C in the case of the samples with a thermal oxide). Somewhere between 700 and 750°C the BST started to significantly react with the underlying TiO₂. There was significantly less BST/substrate reaction in the case of RTA; however, there was still evidence of interaction, which seems to be independent of the RTA temperature. For all three sets of samples, except for the 800°C standard anneals, all of the samples remained still substantially “BST” on top of an interaction layer (i.e., they weren’t fully consumed by the substrate).

Figure 4. Glancing Angle X-Ray Diffraction patterns for Ti stubs with BST for A) standard annealing conditions and no thermal oxide, B) standard annealing conditions with a thermal oxide, and C) RTA with no thermal oxide. The diffraction peaks for BST are the lower set of numbers, while the diffraction peaks for rutile are shown in *italics* as the higher set of numbers. D) Shows a representative planar FE-SEM image of the grain structure of the BST thin films on the Ti stubs.

Glancing angle x-ray diffraction (GA-XRD) was utilized to measure the crystallinity of the PTAS layer, the presence of a piezoelectric phase, and to verify the measurements made by RBS regarding the interlocation of the oxide and PTAS layers. Figure 4A–C shows the results of the GA-XRD, where the peaks represented by a normal script (100) are for the BST, while peaks with italic script (*100*) are for the rutile phase of titania. The most striking item, when first examining the GA-XRD spectrum, is that for all annealing temperatures, even as low as 650°C, there are substantial BST peaks apparent. Previous research on semiconductor substrates had indicated that an annealing temperature of at least 700°C was necessary to generate reasonable crystallinity and 750°C was preferred. Examining Fig. 4A, there are several items of interest for the samples that underwent a standard annealing treatment with no thermally grown buffer layer. First, the titania and the BST peaks become narrower with increasing annealing temperature indicating improved crystallinity. Secondly, the area beneath the titania peaks grows at the expense of the BST peaks, verifying the RBS results that indicated that the BST was reacting with the underlying titanium. Finally, the BST peaks show noticeable peak broadening and splitting, which is indicative of the piezoelectric, tetragonal phase of BST. Figure 4D

shows a field emission scanning electron microscope (FE-SEM) of the surface and grain structure sample that underwent the standard annealing process at 650°C without a thermally grown oxide buffer layer. The grains are uniform and well developed and do not demonstrate any voiding or pin holes. This BST thin film on the structural titanium stub is representative of the PTAS films.

When the samples with a thermally grown oxide buffer layer that underwent a standard annealing process are examined, in Fig. 4B, many similarities are seen to the samples that did not have a thermal buffer layer but also underwent the standard annealing process. Both materials become more crystalline with increasing annealing temperature. The titania layer grows at the expense of the BST layer, but it appears to do so at a slower rate than the samples without a thermal buffer layer. The peak splitting associated with the piezoelectric tetragonal phase is especially apparent in the (200) and (211) peaks. When the samples that underwent a RTA treatment without a thermally grown oxide were examined, many of the same features are observed. The materials become more crystalline with increasing annealing temperature; the oxide material increases with increased annealing temperature, and the presence of the piezoelectric tetragonal phase is observed for all annealing temperatures. Additionally, there are several interesting aspects seen in the RTA samples. The * in Fig. 4C indicates the presence of additional phases. When the peaks centered around 39° are examined, it is apparent that there are three distinct peaks, where Fig. 4A and 4B had only a single dominant peak. In Fig. 3C the peak at 38° and 40° are for barium titanate and strontium titanate respectively, while the peak centered at 39° is for the atomistic mixture BST. Similar segregations can be seen for the 650°C anneals observed in Fig. 4A and 4B. When the annealing temperature is increased the BST peak becomes more dominant and the barium titanate and strontium titanate gradually disappear. However in the RTA samples, even at 800°C there is a residual strontium titanate peak. This indicates that the atomistic mixing typically observed for MOSD samples, even at the pyrolyzation step, is inhibited. While crystallinity appears to occur at substantially lower annealing temperatures for the MOSD samples on structural titanium substrates, a much higher activation energy for atomistic mobility on the surface is required. This is indicative a substrate surface is substantially rougher than the atomically smooth semiconductor substrates.

CONCLUSIONS

The PTAS deposition treatments were all successfully applied to the polished structural titanium stubs. Glancing angle X-ray diffraction illustrated that all of the PTAS films were crystalline, possessed BST and exhibited peak splitting of the (110) and (200) peaks, which is indicative of the piezoelectric phase, even at the lowest annealing temperature of 650°C. However, maximum crystallinity was achieved at 800°C annealing for all samples. For the RTA samples

in addition to BST, there were also barium titanate and strontium titanate phases present. The surface, which was rougher than semiconductor substrates, substantially lowered the activation energy necessary for nucleation and growth of the crystalline phases; however, the same roughness also raised the activation energy for surface hopping of the various atoms, impeding atomic level mixing.

The AFM results indicated that grain size increased with annealing temperature, indicative of increasing piezoelectric properties. The difference between grain size of the oxide buffer layer samples and the standard anneal samples indicates that the larger grain size observed in the standard anneal samples is largely due to a growth of the substrates grains. The roughness substantially increased for the standard and the oxide buffer layer samples, but remained nearly constant for the RTA samples as the annealing temperature increased. RBS indicated that only minimal reaction occurred for the RTA samples at all annealing temperatures, but all of the standard and the oxide buffer layer samples experience some intermixing. The oxide buffer layer, though, delayed the intermixing as compared to the standard annealing process. The presence of a reaction or interaction layer is not necessarily adverse from a performance perspective. The interaction layer will substantially increase the mechanical and the piezoelectric coupling between the sensor and the substrate. The higher heat treatment temperatures responsible for the large interaction zone also caused a substantially improved piezoelectric and crystallographic material to be formed. It may even be beneficial to intentionally sacrifice a portion of the thin film to create a bonding/coupling layer, so the interaction may not be a bad thing per se. Therefore, the optimum treatment was a mechanical polishing treatment followed by deposition of the PTAS thin films and an RTA annealing at 750–800°C. The use of an oxide buffer layer would substantially roughen the surface, but would also inhibit the intermixing between the PTAS and the substrate. For the RTA annealing there is only minimal intermixing, so the use of an oxide buffer layer would not be necessary. All of these processes are also the most readily adaptable to a Depot.

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