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## DEVELOPMENT OF HIGH PERFORMANCE BS-PT BASED PIEZOELECTRIC TRANSDUCERS FOR HIGH-TEMPERATURE APPLICATIONS

Yu-Hung Li<sup>1</sup>, Sang Jong Kim<sup>2</sup>, Nathan Salowitz<sup>2</sup>, Fu-Kuo Chang<sup>2</sup>

<sup>1</sup> Department of Materials Science and Engineering, Stanford University, CA, USA

<sup>2</sup> Department of Aeronautics and Astronautics, Stanford University, CA, USA

liyuhung@stanford.edu

### ABSTRACT

This paper focuses on the development of the new BiScO<sub>3</sub>-PbTiO<sub>3</sub> (BS-PT) based piezoelectric ceramic transducers for high-temperature SHM applications. By controlling the PbO deficiency in the material system, we modify the lossy ferroelectric properties and enhance the piezoelectric responses from the intrinsic BS-PT. The new in-house fabricated piezoelectric transducers can maintain comparably high piezoelectric responses at temperatures up to 350°C continuously for at least 10 hours. The signals of the new BS-PT based piezoelectric transducers were tested and studied at different temperatures on various structures, and we observed a promising trend of obtaining stronger signals from the BS-PT transducers at higher temperatures. A new microfabrication method was developed to miniaturize and to mass fabricate the piezoelectric transducers for potential applications on complex structures in extreme environments.

**KEYWORDS :** *high-temperature piezoelectric transducer, microfabrication, polyimide*

### INTRODUCTION

The significance of structural health monitoring (SHM) has been emphasized in so many fields that more and more attention is being drawn to the implementation of SHM in extreme environments. Among other solutions, applying in-situ ultrasound-based SHM techniques to structures has been investigated extensively because of its promising potential. The advantage of using the in-situ ultrasound-based SHM techniques is that on-board SHM transducers can locate anomalies automatically and can help assess the integrity of the structures in real time to reduce the dependence on the labor-intensive post-operation inspections and maintenance processes in industries like aerospace. However, similar ultrasonic SHM techniques for high-temperature applications have been desirable yet challenging because most of the PZT-based ultrasonic transducers cannot survive the thermal depoling process when temperatures exceed 250°C. Researchers have been looking for new materials to replace the PZT-based piezoelectric transducers for adapting the advantages of SHM and for implementing a well-developed ultrasound-based system in harsh environments for weight-critical applications (engines and high-speed aircrafts) and non-accessible infrastructure (power plant pipelines and geothermal downhole casings).

Recent research in high-temperature piezoelectric materials has facilitated the advancement of high-temperature ultrasound-based SHM systems. To solve the thermal depoling issues of the piezoelectric elements used in the ultrasonic SHM techniques, we first studied the development of a high-temperature piezoelectric material system, BiScO<sub>3</sub>-PbTiO<sub>3</sub> (BS-PT), at its morphotropic phase boundary (MPB). The unique phase combination of BiScO<sub>3</sub> and PbTiO<sub>3</sub> at the MPB provides a good starting point with a sufficiently high depoling temperature and fair piezoelectric responses [1][2].

In this work, to explore the possibilities of how to enhance the piezoelectric responses and heat resistance of the BS-PT system, we conducted a series of studies on compositional effects around the MPB composition and developed an effective way to modify the original properties of the intrinsic BS-PT transducers. In addition to material development and characterization, we also investigated the temperature dependence of the transducer signals and demonstrated the SHM capabilities of our in-house fabricated BS-PT transducers on various structures. For embedded transducer applications where the dimensions of the transducers are critical, such as in bondline integrity monitoring, we developed a novel microfabrication process to fabricate miniaturized BS-PT thick-film transducer networks. With the thin and flexible polyimide substrate, the integrated BS-PT transducer network can potentially be used for SHM of more complex structures in high-temperature environments.

## 1 MATERIAL DEVELOPMENT

As reported by Eitel *et al.* in 2001,  $(1-x)$  BiScO<sub>3</sub> -  $x$  PbTiO<sub>3</sub> at its MPB ( $x \approx 0.64$ ) was first found to be a promising candidate for high temperature applications because of its good piezoelectric responses and its high Curie temperature (about 450°C) because of the small Perovskite tolerance factor of BiScO<sub>3</sub> in the solid-solution system with PbTiO<sub>3</sub> [1][2]. In order to further enhance the piezoelectric properties of this BS-PT based material system, other researchers have also been looking for different doping mechanisms to modify the intrinsic BS-PT material system [3][4][5]. To achieve a similar goal for better SHM applications in harsh environments, we started with the MPB composition of the intrinsic BS-PT and investigated the effects of crystal deficiency on its piezoelectric properties.

All the BS-PT test pellets in our experiments were fabricated using the conventional solid-state reaction method. Raw oxide powders (PbO, TiO<sub>2</sub>, Bi<sub>2</sub>O<sub>3</sub> and Sc<sub>2</sub>O<sub>3</sub>) were weighed accordingly with the target stoichiometric ratios shown in Figure 1(a) and ball-milled in the medium with alcohol and Yttrium-stabilized ZrO<sub>2</sub> balls for 24 hours. After fully drying the mixture at 90°C for another 24 hours followed by hand grinding and mixing in an alumina mortar, we calcinated the powders at 750°C for 3 hours in closed alumina crucibles to obtain uniform BS-PT powders. The BS-PT powders were then mixed with a 5 wt% polyvinyl alcohol (PVA) binder and pressed into green bodies followed by a sintering process at 1100°C. All the sintered ceramic pellets were ground to the desired thickness and electroded with the fire-on silver paste. The ferroelectric and piezoelectric properties were measured and characterized using an impedance analyzer (Agilent 4294A) and a ferroelectric tester (Radiant Technologies Inc., RT66B) with an external high-voltage amplifier (TREK 609E-6) after all the samples were fully polarized in an oil bath at 120°C with a DC poling electric field of 80 kV/cm applied across each sample.

We found that by controlling the PbO deficiency in the BS-PT material system, the piezoelectric responses were substantially enhanced because the current leakage characteristics were improved. This improvement is suggested by the higher remnant polarizations and smaller coercive fields of the PbO deficient samples (BS-PT#3 and BS-PT#4) in the polarization-electric field (P-E) curves shown in Figure 1(b).

To test the thermal tolerance of the PbO deficient samples, thermal depoling tests were conducted to compare commercial PZT based transducers (PZT-5A), intrinsic BS-PT transducers (BS-PT#2), and the newly developed BS-PT#3 transducers. Polarized samples were placed in a furnace and exposed to different thermal depoling environments for two hours. The  $d_{33}$  piezoelectric coefficients were measured with a YE2730  $d_{33}$  meter at room temperature before and after exposure to the depoling environments at each test temperature. As shown in Figure 2, the BS-PT#3 transducers exhibit a high  $d_{33}$  piezoelectric coefficient up to 350°C and remain over 80% of their original  $d_{33}$  values after being exposed at 350°C for two hours. These results show that the new BS-PT#3 transducers do have superior piezoelectric responses over a wide temperature range, and the thermal tolerance is not compromised by the enhancement of the piezoelectric responses.

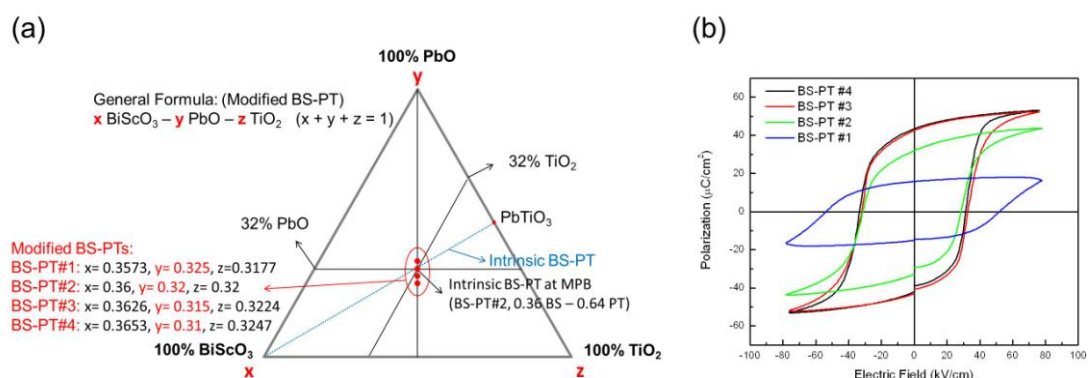


Figure 1. (a) BS-PT ternary phase diagram. The blue dotted line represents the intrinsic  $(1-x)$   $\text{BiScO}_3 - x$   $\text{PbTiO}_3$  with different composition ratios. In this work, we controlled different amounts of PbO in the solid-solution system to form BS-PT#1 to #4, which are marked in red dots in the diagram. (b) The P-E curves of BS-PT#1~BS-PT#4. Excess PbO in BS-PT#1 aggravates the leaky nature of the intrinsic BS-PT (BS-PT#2) while PbO deficiency in BS-PT#3 and #4 leads to a less leaky characteristic and a higher remnant polarization.

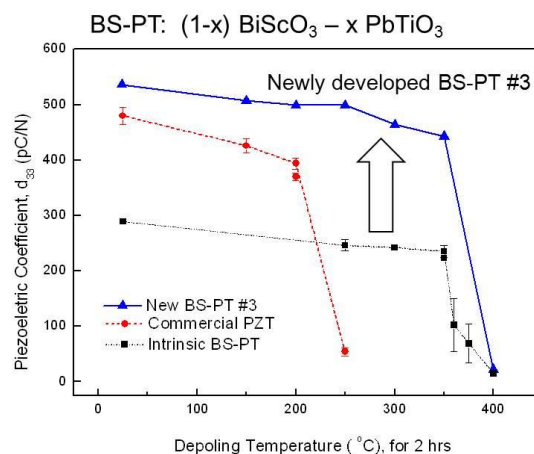


Figure 2. Results of thermal depoling tests for commercial PZTs (PZT-5A), intrinsic BS-PTs (BS-PT#2) and the newly developed BS-PTs (BS-PT#3)

## 2 SHM SIGNAL STUDY

In addition to material characterizations, we also tested the new BS-PT#3 transducers integrated with simple metallic structures to see whether typical SHM signals can be actuated, conveyed, and sensed properly at different temperatures. Four in-house fabricated BS-PT#3 transducers were mounted on a 1.3-mm-thick aluminum plate with a thin layer of high-temperature silicate-based conductive adhesive (Aremco Pyro-duct 597A [6]). The test plate geometry and sensor layout are described in Figure 3. As a reference, four commercial PZT-based transducers (PZT-5A) were installed in the same way on an identical aluminum plate so that we could compare signals from both setups at different temperatures.

In the test, we used the standard 5-peak Gaussian windowed tone burst from 150 kHz to 800 kHz as actuation signals. Commercial data acquisition hardware and software (Acellent Technologies Inc.) were used to generate and collect signals with the piezoelectric ceramic transducers. To see the temperature effects on the sensor signals, the whole test structure with surface-mounted transducers was placed in an oven, and all the signals were taken from room temperature to 300°C. To ensure a uniform temperature distribution, the oven temperatures were

held at each set point for 30 minutes before the signals were sent and collected. Figure 4(a) shows a typical signal generated by an actuator-sensor pair (Path 1-to-2) at room temperature. We applied the Hilbert transform to the raw signal data afterwards and extracted the time-of-flight and signal amplitudes of the first arriving wave packets generated by different transducers at different temperatures.

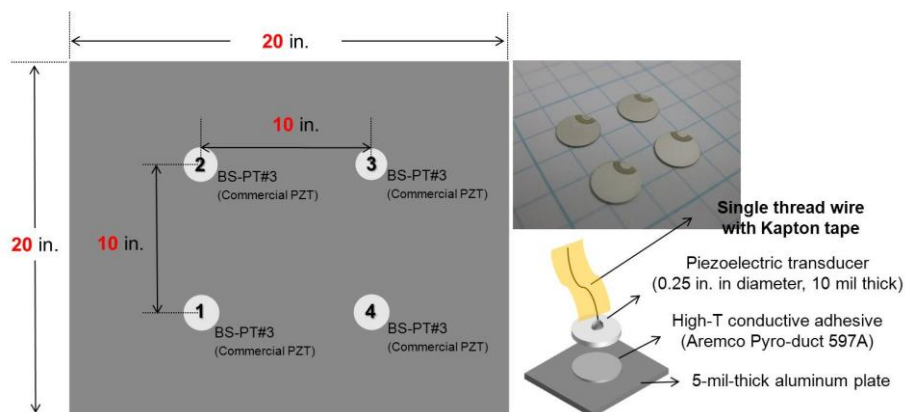


Figure 3. The test geometry and transducer dimensions of the SHM signal study

As shown in Figure 4(b) and 4(c), the signal strength of the BS-PT#3 transducers exhibits an increasing trend at higher temperatures while the commercial PZT transducers show a decreasing trend as the temperature increases. The pitch-and-catch test results not only confirm the good thermal and piezoelectric properties of our in-house fabricated BS-PT#3 transducers but they also demonstrate that SHM signals can be fully conveyed by the newly developed transducers and the signal strength can be even stronger at high temperatures.

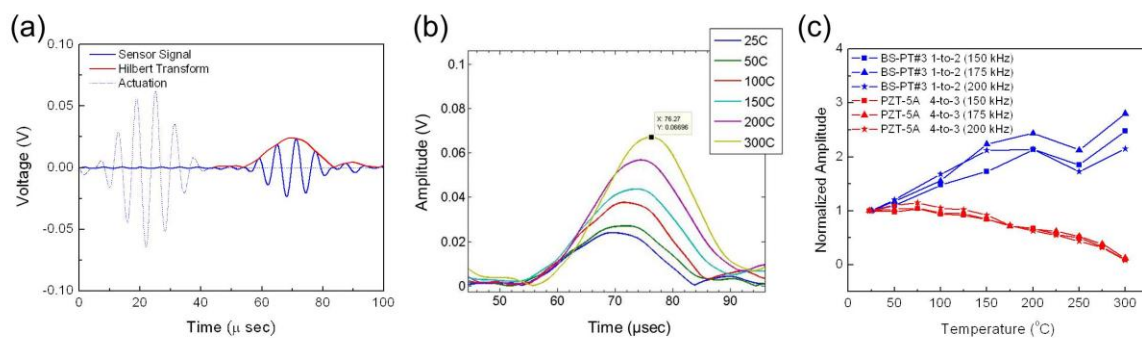


Figure 4. (a) The raw signal of BS-PT#3 transducers at room temperature (b) Signal amplitudes of the first arriving wave packets at different temperatures (c) The comparison of signal strength between BS-PT#3 and commercial PZT (PZT-5A) transducers using different actuation frequencies at different temperatures.

Normalized amplitude values are obtained by comparing the maximum signal amplitude measured in the Hilbert transform of the first wave packet at room temperature with the ones measured at different testing point from the same actuator-sensor pair.

### 3 SENSOR MINIATURIZATION

To fully exploit the good properties of the BS-PT based piezoelectric transducers in more SHM applications like embedded SHM systems, we have developed a microfabrication process with screen-printing techniques to fabricate thick-film BS-PT transducer arrays. The main advantage of using the screen-printing techniques is that these techniques enable mass-production of thinner piezoelectric transducers with more complex shapes at a lower cost, compared to the traditional

solid-state reaction method. Although smaller and thinner transducers are preferable especially in embedded SHM applications, as we try to shrink the dimensions of the transducers to minimize the structural influence of the embedded SHM transducers, the challenge is how to ensure the SHM capabilities of these miniaturized transducers. The trade-off between transducer dimensions and sensitivity/actuation strength is the key, but it is difficult to define what the optimal design is because different applications may have different requirements and tolerance. Therefore, our approach is to optimize the material properties within a targeted transducer thickness of 50  $\mu\text{m}$ , which is a reasonable thickness to be included either within an adhesive layer in a bondline or within composite plies for embedded SHM applications.

### 3.1 Development of Screen-Printed BS-PT Transducers

The BS-PT#3 powders were prepared by the same process mentioned previously, in Section 1, except instead of using PVA binders to form pressed green bodies, we used a three-roller miller to mix and grind the BS-PT#3 powders with a 20 wt% liquid polyvinyl butyral binder (FERRO Binder 75001B) to form printable ceramic paste. This ratio between the powders and the binder was optimized to obtain an ideal viscosity for good printing quality and to achieve the desired transducer thickness.

Before printing the transducer layer, we first grew a thin layer of silicon nitride (0.5 $\mu\text{m}$ ) on the silicon substrate as a bottom insulating layer. A platinum-based paste (FERRO 4092) was printed on the nitride layer and annealed at 1100 $^{\circ}\text{C}$  for 15 minutes. This annealed platinum layer not only forms the bottom electrodes of the transducer arrays but also acts as a diffusion barrier layer to prevent potential inter-diffusion between the substrate and the ceramic layer in the later steps. Then, the BS-PT#3 ceramic paste was printed on top of the electrodes and sintered using various heating conditions. Finally, the top electrode was evaporated on the ceramic layer to complete the parallel-capacitor-like transducer design. Because of the higher binder percentage in the paste and the lack of pressure in green body forming, the sintering conditions of the screen-printed piezoelectric transducers are very crucial to the densification of the ceramic layer while inter-diffusion also plays an important role in transducer performance. Therefore, as shown in Figure 5, we designed numerous test specimens to go through a sintering condition optimization process to determine the optimal parameters. Microstructure observation using a scanning electron microscope (SEM) and characterization of ferroelectric properties from the P-E curves confirmed that using a rapid thermal annealer (RTA) with high ramping and cooling rates can effectively sinter the ceramic body and hinder the inter-diffusion at the same time. The total thickness of the transducer layer was measured to be around 45  $\mu\text{m}$ , and the dense microstructure of the well-sintered ceramic layer with saturating P-E curves suggests promising transducer functionality.

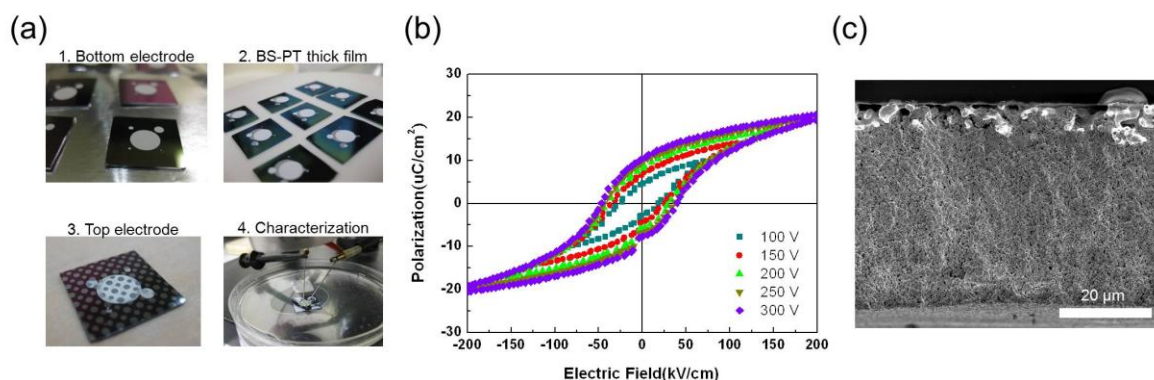


Figure 5. The optimization process of sintering conditions. (a) Process flow to fabricate the test specimens (b) The P-E curve of the optimized condition – a saturating hysteresis curve with the highest remnant polarization. (c) The cross-section SEM image of the screen-printed BS-PT#3 transducer showing a bottom electrode of around 5 $\mu\text{m}$  and a dense sintered ceramic layer of around 40  $\mu\text{m}$

### 3.2 Verification of SHM Capabilities

For testing the SHM capabilities of the screen-printed BS-PT#3 transducers, similarly, we used the 5-peak Gaussian windowed tone burst as the input actuation signal to see whether the screen-printed transducer was able to have both sensing and actuating capabilities on a simple structure. We used the supporting silicon substrate (0.5 mm) of the screen-printed transducer as the test structure and attached a commercial bulk PZT-5A transducer on the other end of the beam as illustrated in Figure 6. Different levels of input voltages were applied to the bulk commercial transducer and the screen-printed transducer to verify the sensing and actuating of Lamb waves with different amplitudes.

Figure 6(a) and 6(b) show the sensed signals from the screen-printed BS-PT#3 transducer and the bulk commercial transducer respectively. From both signals, obvious wave packets are observed; the screen-printed BS-PT#3 transducer can differentiate different levels of vibrations initiated by the bulk commercial transducers and can create different levels of vibrations itself. This indicates that the screen-printed BS-PT#3 transducers can not only sense the vibrations of different magnitudes sent from the other transducer but also actuate Lamb waves with different amplitudes. Lower input voltages were used to actuate the screen-printed BS-PT#3 transducer to compensate the thickness factor for creating similar electric fields. In the meantime, using lower voltages can prevent potential degradation of the screen-printed transducer because during actuation, the undesired depoling fields caused by the input AC signal can be high at the thinner region of the transducers.

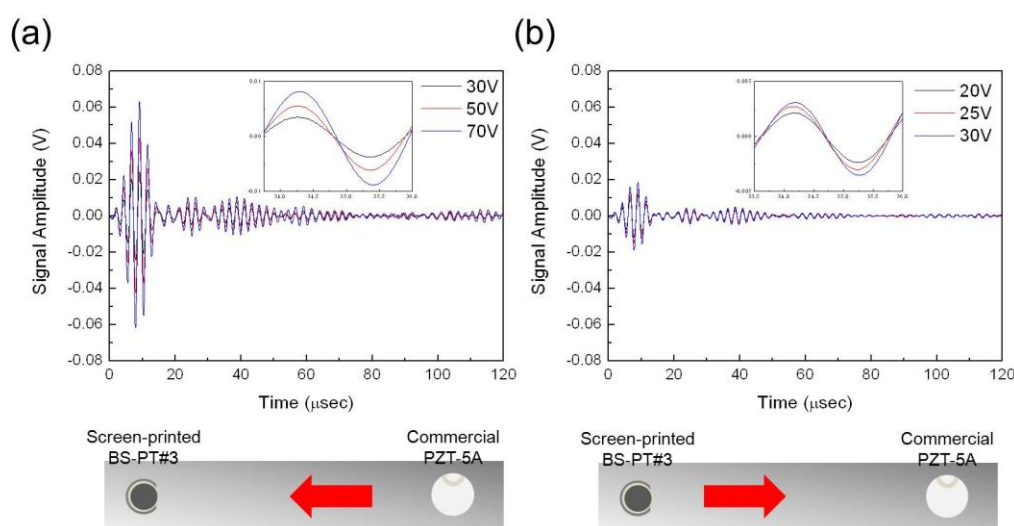


Figure 6. Demonstration of SHM capabilities of the screen-printed BS-PT#3 transducers – raw signal data at 400 kHz for (a) sensing and (b) actuation

### 3.3 Fabrication Integration with Flexible Substrate

A novel microfabrication process was developed to integrate the screen-printed piezoelectric transducers with thin polyimide substrates. Polyimide is a high-temperature polymer with a glass transition temperature range close to 400°C. The inert and robust nature of polyimide makes it a perfect candidate to be integrated with screen-printed BS-PT transducers for high-temperature SHM applications. With our flexible and stretchable designs, we can fabricate a piezoelectric transducer array from a small wafer and deploy the transducer arrays to cover a larger area or a complex-shaped structure [7].

We first started with the screen-printing process mentioned in the previous section but used photolithography techniques to pattern a different top electrode layer to create the transducer array. Two layers of liquid polyimide were then spin-coated on the stack and cured at 350°C for 30 minutes. Metal evaporation deposition and another photolithography process were applied on the

polyimide layer to pattern the aluminium etch mask with the stretchable network design. Lastly, an oxygen plasma etch was used to remove the extra polyimide and define the final polyimide-based transducer array, followed by a wet potassium hydroxide (KOH) etch to etch away the supporting silicon substrate and release the transducer layer. An overview of the integrated process is described in Figure 7. Our final goal for this thin transducer array is to achieve embedded SHM for application in harsh environments. Characterization of the released transducers and the interaction between the embedded transducer layer and the host structure are still under investigation. Ongoing work focuses on the structural influence of the transducer layer and the temperature dependence of the performance of the screen-printed BS-PT#3 transducers.

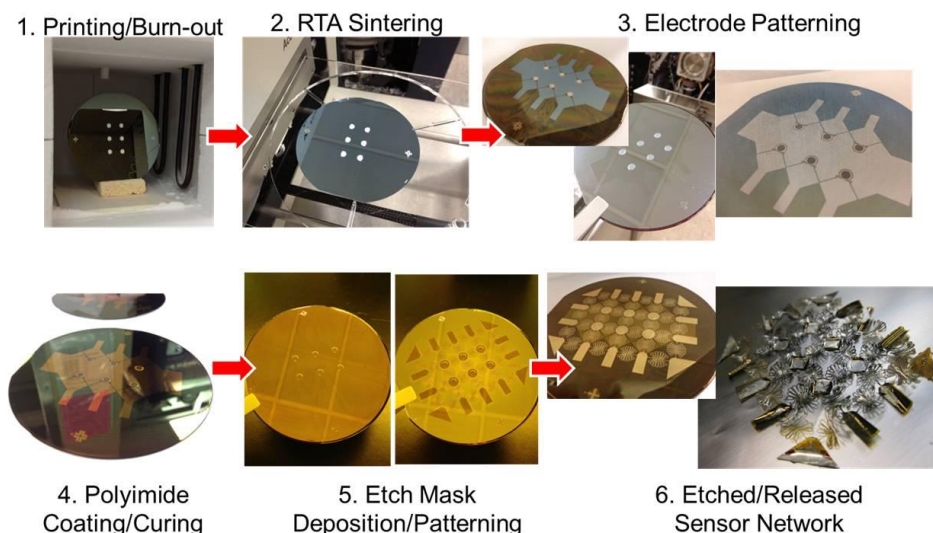


Figure 7. An overview of the integrated microfabrication process to transfer the screen-printed piezoelectric transducers on a polyimide-based flexible substrate

## CONCLUSION

In this work, we have demonstrated that by controlling the amount of PbO in the BS-PT material system, we can effectively enhance the piezoelectric responses while not compromising the thermal tolerance. The newly developed BS-PT based piezoelectric transducers maintain a high piezoelectric coefficient after exposure to high temperature environments and show an increasing trend in SHM signals at higher temperatures. To miniaturize the transducers for embedded SHM applications, a screen-printing-based microfabrication process has been developed. The preliminary results confirm that the screen-printed BS-PT#3 transducers can sense and actuate standard pulsed SHM signals. With our novel transferring method, it is very promising that the thin screen-printed BS-PT#3 transducer arrays may be integrated into complex structures with minimal structural influences for high-temperature SHM applications.

## ACKNOWLEDGEMENT

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## REFERENCES

- [1] R. Eitel *et al.* "New High Temperature Morphotropic Phase Boundary Piezoelectrics Based on Bi(Me)O<sub>3</sub>-PbTiO<sub>3</sub> Ceramics" *Jpn. J. Appl. Phys.* Vol. 40, No. 10, pp 5999-6002 (2001)
- [2] R. Eitel *et al.* "Preparation and Characterization of High Temperature Perovskite Ferroelectrics in the Solid-Solution (1-x) BiScO<sub>3</sub>-x PbTiO<sub>3</sub>" *Jpn. J. Appl. Phys.* Vol. 41, No. 4A, pp 2099-2104 (2002)
- [3] S. Zhang *et al.* "Manganese-Modified BiScO<sub>3</sub>-PbTiO<sub>3</sub> Piezoelectric Ceramic for High-temperature Shear Mode Sensor," *Appl. Phys. Lett.* Vol. 86, 262904 (2005)
- [4] A. Sehirlioglu, A. Sayir, F. Dynys "High Temperature Properties of BiScO<sub>3</sub>-PbTiO<sub>3</sub> Piezoelectric Ceramics," *J. Appl. Phys.* Vol. 106, 014102 (2009)
- [5] A. Sehirlioglu, A. Sayir, F. Dynys "Doping of BiScO<sub>3</sub>PbTiO<sub>3</sub> Ceramics for Enhanced Properties," *J. Am. Ceram. Soc.*, 93 [6] 1718-1724 (2010)
- [6] V. Giurgiutiu, B. Xu, W. Liu "Development and Testing of High-temperature Piezoelectric Wafer Active Sensors for Extreme Environments," *Structural Health Monitoring*, Vol. 9, No. 6, pp. 513-525 (2010)
- [7] G. Lanzara, N. Salowitz, Z. Guo, F.-K. Chang, "A Spider Web-Like Highly Expandable Sensor Network," *Advanced Materials*, Vol. 22, 41, pp 44643-4648 (2010)