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Piezoelectric thick bismuth titanate/lead zirconate titanate composite film transducers for smart NDE of metals

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Abstract
Thick film piezoelectric ceramic sensors have been successfully deposited on different metallic substrates with different shapes by a sol–gel spray technique. The ball-milled bismuth titanate fine powders were dispersed into PZT solution to achieve the gel. The films with desired thickness up to 200 μm have been obtained through the multilayer coating approach. These thick films were also effectively coated onto thin sheet metals of thickness down to 25 μm. Self-support films with flat and shell geometries were made. Piezoelectricity was achieved using the corona discharge poling method. The area of the top silver paste electrode was also optimized. The center frequencies of ultrasonic signals generated by these films ranged from 3.6 to 30 MHz and their bandwidth was broad as well. The ultrasonic signals generated and received by these ultrasonic transducers (UTs) operated in the pulse/echo mode had a signal to noise ratio more than 30 dB. The main advantages of such sensors are that they (1) do not need couplant, (2) can serve as piezoelectric and UT, (3) can be coated onto curved surfaces and (4) can operate up to 440 °C. The capability of these thick film UTs for non-destructive evaluation of materials at 440 °C has been demonstrated.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Piezoelectric ceramic sensors and actuators are commonly used as key candidates for smart materials and structures. They have been used as structural vibration actuators and suppression or damping elements, structural health monitoring sensors, active shape control tools, non-destructive evaluation probes for materials and structures etc [1–7]. These piezoelectric devices can be surface bonded onto, embedded into and coated onto the host materials, which may be fiber composites such as graphite/epoxy, semiconductors and metals. Multi-element array, single-element, plate, laminated plate, rod, shell and thick and thin film configurations have been used for large and/or miniature materials or structures. Nearly all of these previous works focused on room temperature operations. In this investigation, we are proposing the use of thick film piezoelectric ceramic transducers for smart non-destructive evaluation of metals with flat and odd shapes at elevated temperatures up to 440 °C, which is currently the limitation of our measurement equipment.

Thick (>40 μm) piezoelectric ceramic films can be made by the technologies of jet printing [8], screen printing [9], dipping [10], tape casting [11], the hydrothermal method [12] etc. Here an alternative sol–gel spray technique is used. This fabrication process was first developed at Queen's [13]. The piezoelectric particles are dispersed in the sol–gel solution to produce a thick piezoelectric film [14–16]. The spray can be carried out by an air gun at room temperature and it is

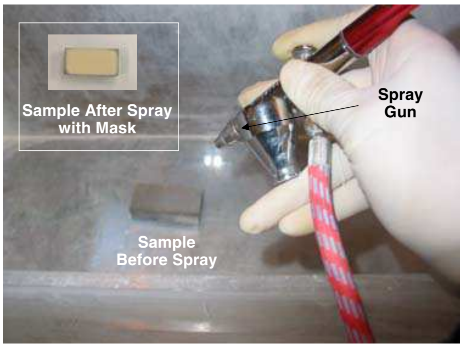

Figure 1. Set-up of sol–gel spray.

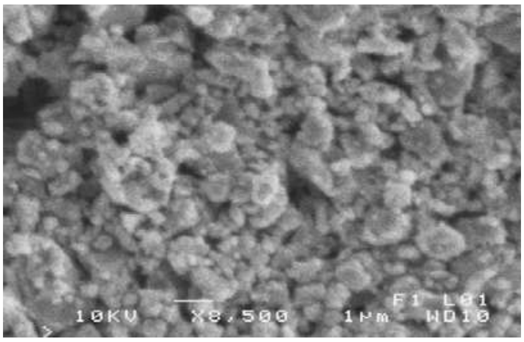

Figure 2. SEM image of one BIT/PZT film.

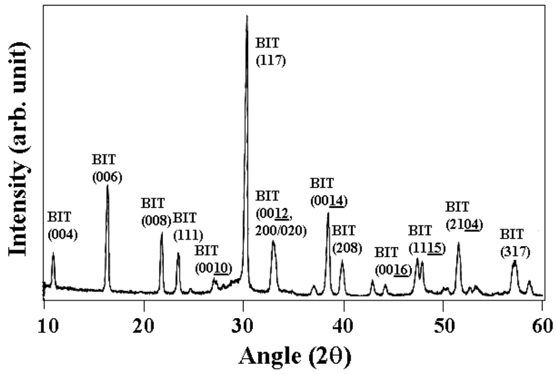

Figure 3. XRD pattern of one BIT/PZT film.

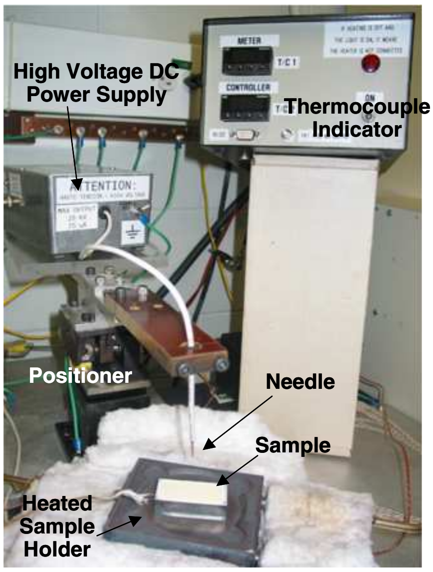

Figure 4. Set-up of corona poling.

simple and inexpensive. In our previous works [14–16] it was demonstrated that piezoelectric powders such as lead zirconate titanate (PZT) ceramics or lithium titanate crystals could be dispersed into PZT or alumina (Al_2O_3) solutions to achieve piezoelectricity, and steel and aluminum substrates were used. In this presentation we focus on the bismuth titanate (BIT) powder dispersed into PZT solutions because of the high Curie temperature, 675 °C, and reasonable piezoelectric strength of BIT. A new but convenient electric poling and top electrode deposition method will be employed and the poling conditions and electrode size will be optimized. Different substrates such as titanium, nickel and copper will be investigated as well.

2. Fabrication and characterization

The piezoelectric BIT powder was purchased with an average dimension of 50 μm and dispersed into the PZT solution by the ball milling method to achieve the gel. The final dimension of the BIT powder is estimated to be less than 1 μm. An air gun was then used to spray the sol–gel composite directly onto the samples as shown in figure 1 even with curved surfaces and odd shapes. With this technique, the BIT/PZT films can easily be produced at desired locations through a shadow mask made of even paperboard. After spray coating, thermal treatments such as drying, firing and annealing were carried out at temperatures of 90, 430 and 650 °C, respectively, with the optimal time duration. The volume ratio between BIT powder and PZT solution after annealing is about 3:1. Multiple layers were made in order to reach the desired thickness. During thermal treatments, drying, firing and annealing were used for the fabrication of each layer. The 40–200 μm thick films, to provide the desired center frequency in the range of 3.6–30 MHz, were produced. This frequency range is commonly preferred for NDE of metals because of its sufficient ranging resolution and acceptable ultrasonic attenuation in metals.

The measured relative dielectric constant was around 90 and dielectric $\tan\delta$ was near 0.01. These calculations were based on the measurement results obtained by a 4192A LF impedance analyzer (Hewlett Packard). The frequency constant along the thickness direction was around 1100. Figures 2 and 3 show the SEM image and the x-ray diffraction (XRD) pattern of the film, respectively. It is indicated in figure 2 that the grain size ranged between sub-micron and 1 μm and the film was not dense. XRD in figure 3 only shows that the crystallization was found for BIT but not PZT because there was no peak around 31°, where there should be the strongest peak for bulk PZT without orientation. It is speculated that the annealing condition used was not sufficient to obtain PZT crystallization.

The films were then electrically poled using the corona discharging technique as shown in figure 4. During poling the temperature of the substrate was between 200 and 400 °C; a high positive voltage supplied from a high voltage DC power supply was fed into a needle which was located several centimeters above the film coated on the metal substrate which

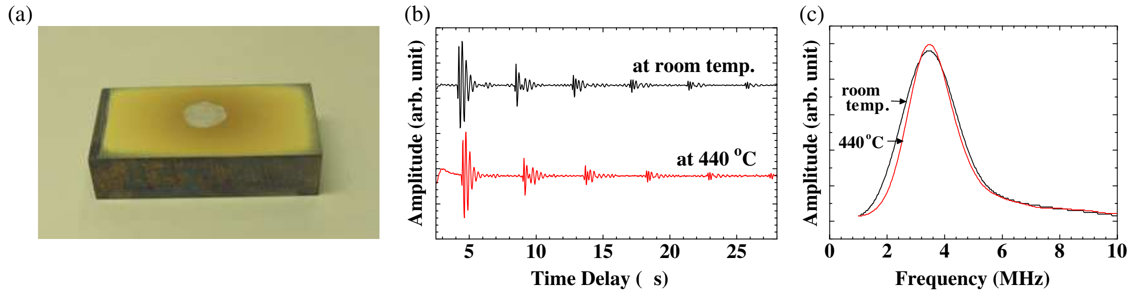

Figure 5. (a) A 200 μm thick film deposited onto a planar steel substrate and its ultrasonic performance (b) in the time and (c) in the frequency domain at room temperature and 440 °C.

served as the ground electrode. The distance and voltage were optimized for different film thicknesses and geometries. The poling time was about 10 min. The corona poling method was chosen because it could pole the thick piezoelectric ceramic film of a large area and on curved surfaces with ease. At room temperature we used 30 μm thick silver paste to form the top electrode to replace the vacuum sputtering reported in [13, 14]. This convenient approach makes the selection of electrode size, which is the sensor size, simple. The silver paste has been tested and its operating temperature was above 440 °C.

3. Ultrasonic performance

3.1. Planar substrates

The fabricated thick BIT piezoelectric films can be used as ultrasonic transducers (UTs). They can operate in pulse–echo, pitch–catch and transmission configurations. Here only the measurements obtained from the pulse–echo geometry are presented. Figures 5(a)–(c) show a 200 μm thick film deposited onto a 6.37 mm thick, 25.4 mm wide and 50.8 mm long steel substrate and its ultrasonic performance at room and 440 °C in the time and frequency domains (of the first echo), respectively. As we can see, the ultrasonic signal shows a signal-to-noise ratio (SNR) of about 16 dB. The SNR is defined as the ratio of the amplitude of the first echo having traveled one round trip through the thickness direction over that of the signals, which are undesired, between the echoes traversing back and forth in the sample. This UT showed a center frequency of 3.6 MHz and had a 3 dB bandwidth of 1.6 MHz. The signal amplitude was reduced by 2 dB from room temperature to 440 °C. At elevated temperatures the electric dipoles lost their alignment slightly from the applied electric field.

In order to determine the optimum area of the top electrode two samples (A and B), which were fabricated at the same time with the same conditions except with slightly different poling conditions, were used. The electrode size varied from 2 to 18 mm. The results measured at room temperature indicated that to obtain the highest strength the optimum diameter of the top electrode for these two film UTs was 11 mm as shown in figure 6. It is noted that in certain situations one needs to use small top electrode to achieve small sensing area and in such a case the signal strength will be sacrificed if the film material is not changed. For example, if PZT/PZT rather than BIT/PZT is used, because of the higher relative dielectric constant of PZT

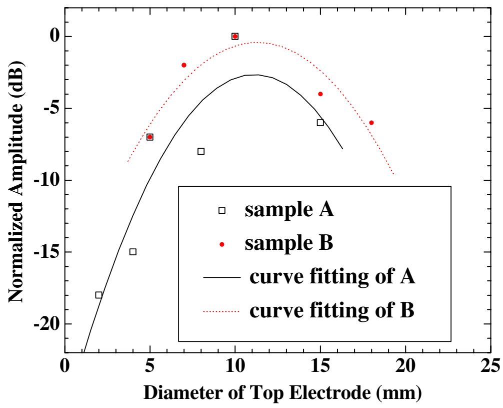

Figure 6. Signal amplitude versus the diameter of the top electrode for two samples.

($\varepsilon_r > 1500$) over BIT ($\varepsilon_r \sim 120$), with the same film thickness the optimum diameter of the top electrode is smaller than that for BIT/PZT films.

3.2. Curved surfaces

In order to demonstrate the ability of the sol–gel spray technology, samples with curved surfaces were also used. Figures 7(a)–(c) show a 40 μm thick BIT/PZT film deposited onto a cylindrical steel shell with an outer diameter of 25.4 mm and its ultrasonic performance at 440 °C in the time and frequency domains (of the first echo), respectively. The center frequency, 3 dB bandwidth and SNR were 30 MHz, 14 MHz and 26 dB, respectively. Similarly, figures 8(a)–(c) show a 90 μm thick BIT/PZT film deposited onto a spherical steel ball with a diameter of 19 mm and its ultrasonic performance at 440 °C in the time and frequency domains (of the first echo), respectively. The center frequency, 3 dB bandwidth and SNR were 9.5 MHz, 4 MHz and 40 dB, respectively.

In addition to the cylindrical and spherical convex surfaces shown in figures 7(a) and 8(a) respectively, we also used the sol–gel spray technology to coat the thick (>80 μm) BIT/PZT film onto cylindrical and spherical concave surfaces of steel substrates, which are shown in figures 9 and 10, respectively. The ultrasonic performance of these UTs as high temperature ultrasonic focusing lenses will be presented elsewhere.

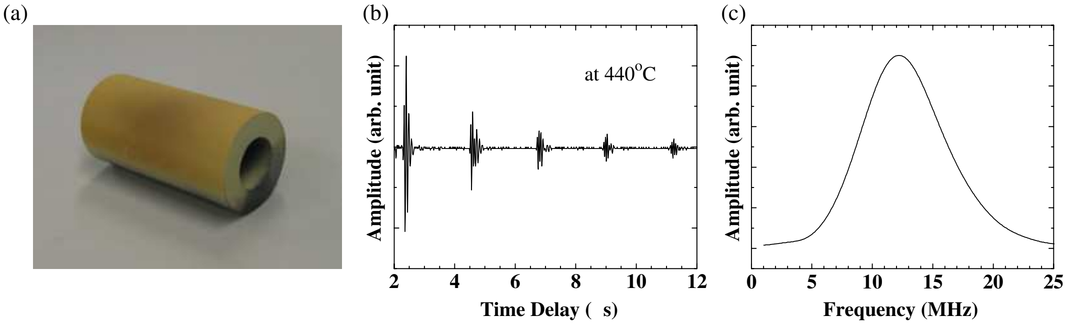

Figure 7. (a) A 40 μm thick BIT/PZT film deposited onto a cylindrical steel shell of 25.4 mm outer diameter and its ultrasonic performance (b) in the time and (c) in the frequency domain at 440 °C.

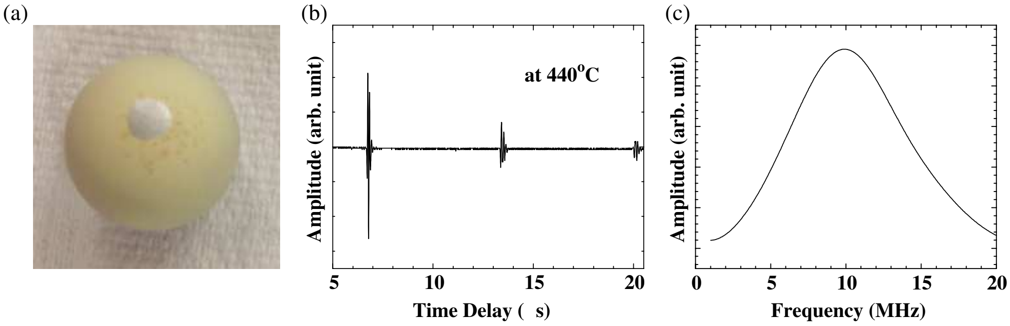

Figure 8. (a) A 90 μm thick BIT/PZT film deposited onto a spherical ball of 19 mm outer diameter and its ultrasonic performance (b) in the time and (c) in the frequency domain at 440 °C.

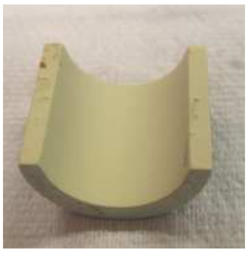

Figure 9. Thick BIT/PZT film deposited on a cylindrical concave surface.

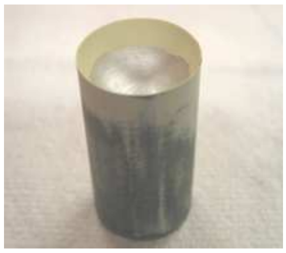

Figure 10. Thick BIT/PZT film deposited on a spherical concave surface.

3.3. Thin metal sheets

Thin metal sheets together with thick piezoelectric coatings may also serve as smart structures in a two-layer or multilayer configuration. In this section we deposited 90 μm thick BIT/PZT film on a 254 μm thick stainless steel, a 127 μm thick titanium, a 25 μm thick nickel and a 127 μm thick copper sheet as shown in figures 11(a)–(d), respectively. Due to the small thickness and 650 °C annealing temperature of BIT/PZT film the titanium, nickel and copper films distorted somewhat. Figures 12(a) and (b) show the ultrasonic performance in the time and frequency domains (of the first echo) respectively, at 440 °C for the configuration shown in figure 11(a). The films shown in figures 11(b)–(d) have been also tested but the results are not shown here.

4. Self-support BIT/PZT film

Thin piezoelectric sheet and shells may be of interest for surface bonding or interior embedding for the smart material and structure applications. During our fabrication procedures described above, if the substrate surface is well polished, the adhesion between this surface and the BIT/PZT film is weak; this film with its substrate curvature may be detached from the substrate. Figures 13 and 14 show the self-support BIT/PZT films with a substrate shape of figures 5(a) and 8(a) respectively, and a thickness of more than 40 μm. Alternatively we could use rapid and many thermal cycles to weaken the adhesion between the substrate surface and the BIT/PZT film and to detach the film.

5. NDE applications

It is known that the broadband MHz frequency UT is suitable for defect or void detection. Here we would like to demonstrate that these thick film UTs at 440 °C could perform this defect detection. A 200 μm thick BIT/PZT film was deposited onto a 12.7 mm thick steel plate with a side-drilled hole of 1.5 mm diameter. The four traces from top to bottom indicate that the

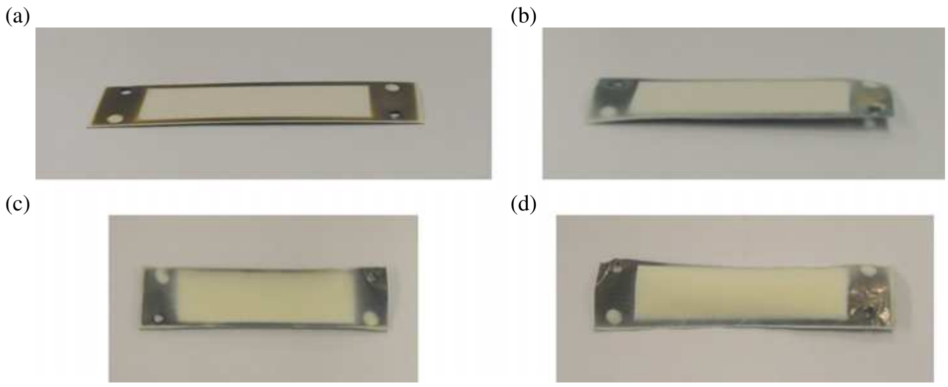

Figure 11. Thick BIT/PZT film deposited on a (a) 254 μm thick stainless steel, (b) 127 μm thick titanium, (c) 25 μm thick nickel and (d) 127 μm thick copper sheet.

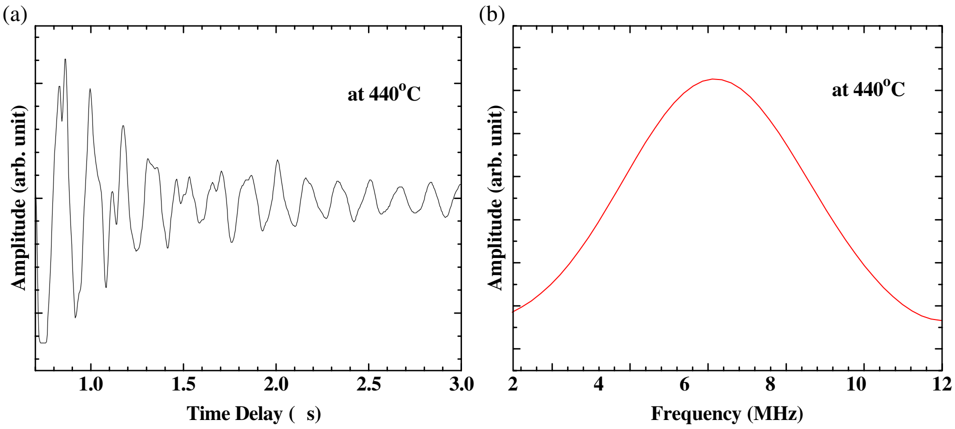

Figure 12. Ultrasonic performance of a 90 μm thick BIT/PZT film deposited on a 254 μm thick stainless steel sheet in (a) the time and (b) the frequency domain at 440 °C.

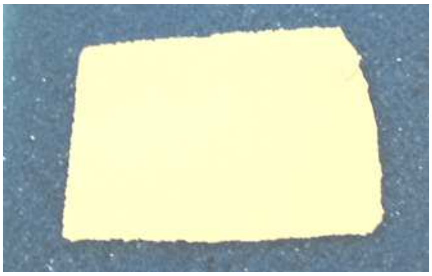

Figure 13. Self-support BIT/PZT sheet.

Figure 14. Self-support BIT/PZT spherical shell.

lengths of this artificial defect under the ultrasonic insonified areas defined by the 11 mm top electrode diameter are 0, 2, 4 and 6 mm, respectively. The large signal at the end of the traces in figure 15 was the reflected echo from the bottom of the steel substrate. This indicates that the growth of the defect can be detected at 440 °C with this high temperature thick BIT/PZT film UT. As the defect became larger, the amplitude of the reflected signal from the defect became larger.

Figure 16 shows that the ultrasonic signals were reflected from a bottom drilled flat hole of 1.0 mm diameter in another steel plate of 12.7 mm coated with a 200 μm thick BIT/PZT film. The four traces from top to bottom show that the lengths of this hole measured from the bottom of the plate are 0, 1, 2 and 3 mm, respectively. The large signal at the end of the traces in figure 16 was the reflected echo from the bottom of the steel substrate. Figure 16 demonstrates that ultrasonic monitoring of the extension of the defect length can be performed at 440 °C, and the time delay of the signal reflected from the defect can be used to find the position of the tip of this bottom drilled flat hole.

6. Conclusions

Piezoelectric composite films of thicknesses ranging from 40 to 200 μm were successfully deposited on steel, stainless steel, titanium, nickel and copper substrates by a sol–gel spray

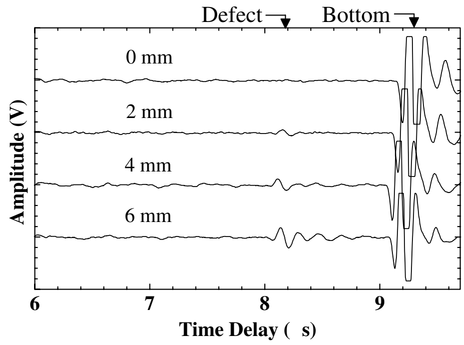

Figure 15. Ultrasonic monitoring of the growth of the artificial horizontal defect at 440 °C.

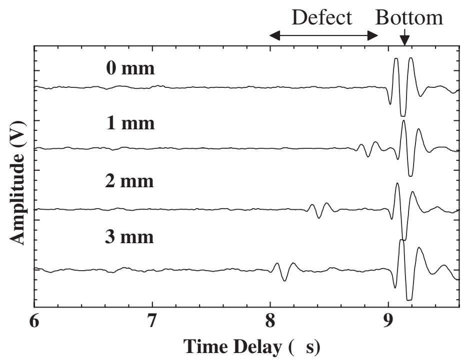

Figure 16. Ultrasonic monitoring of the extension of the artificial vertical defect at 440 °C.

technique. Ball milled bismuth titanate (BIT) fine powders were dispersed into PZT solution to achieve the gel. The gel was then sprayed onto the substrate at room temperature. The films with desired thickness have been obtained through a multilayer coating approach. Piezoelectricity was achieved using the corona discharge poling method, which was convenient for substrates with curved surfaces. The metal substrate served as the bottom electrode and the silver paste forms the top one. For the BIT/PZT film studied the optimal area of the top electrode was found to be 11 mm. The main advantages of sol–gel sprayed thick film sensors, such as that they (1) do not need couplant, (2) can serve as piezoelectric and ultrasonic transducer (UT), (3) can be coated onto curved surfaces and (4) can operate up to 440 °C, have been demonstrated. As UT the center frequencies of the ultrasonic signals generated by these films ranged from 3.6 to 30 MHz and their 3 dB bandwidth was larger than 1.6 MHz.

The thick films have been coated on metallic substrates with planar, spherical concave and convex, and cylindrically concave and convex surfaces. The ultrasonic signals generated and received by these UTs operated in the pulse–echo mode had a signal to noise ratio (SNR) more than 30 dB to evaluate the properties of the substrate onto which they are deposited. These thick films are also effectively coated onto thin sheet metals of thickness down to 25 μm. Self-support films with flat and shell geometries were made. The capability of this thick film UTs for non-destructive evaluation of materials at 440 °C has been verified.

Acknowledgments

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