NRC Publications Archive Archives des publications du CNRC

Flexible Ultrasonic Array Sensors for Health Monitoring

Kobayasi, M.; Wu, K. -T.; Song, L.; Liu, Q.; Jen, C. -K.

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. /
La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version
acceptée du manuscrit ou la version de l'éditeur.

Publisher's version / Version de l'éditeur:

Proceedings of the SPIE Smart Structures and Materials, 2007., 2007-03-19

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=26ac2830-9733-48c9-a4e6-712ec367dfb8>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=26ac2830-9733-48c9-a4e6-712ec367dfb8>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

National Research
Council Canada

Conseil national de
recherches Canada

Canada

Flexible ultrasonic array sensors for health monitoring

M. Kobayashi^a, K.-T. Wu^b, L. Song^b, Q. Liu^b and C.-K. Jen^a

^aIndustrial Materials Institute, National Research Council of Canada, 75 Blvd. de Mortagne,
Boucherville, Quebec J4B 6Y4, Canada;

^bDepartment of Electrical and Computer Engineering, McGill University, 3480 University Street,
Montreal, Quebec H3A 2A7, Canada

ABSTRACT

Flexible ultrasonic array transducers which can be attached to the desired structures or materials for nondestructive testing and structural health monitoring applications at room and elevated temperatures are developed. These flexible ultrasonic transducers (UTs) arrays consist of a thin polyimide membrane with a bottom electrode or stainless steel foil, a piezoelectric lead-zirconate-titanate (PZT) composite film and top electrodes. The flexibility is realized owing to the porosity of piezoelectric film and the thinness of substrate and electrodes. Top and bottom electrode materials are silver paste, silver paint or electroless plated nickel alloys. The UT array is configured by the several top electrodes. The flexible UT has been successfully tested at 150°C and also immersed into water as immersion ultrasonic probe operated in the pulse-echo mode with good signal to noise ratio.

Keywords: Flexible piezoelectric ultrasonic transducer, high temperature ultrasonic measurement, nondestructive testing and array transducer

1. INTRODUCTION

In-situ structural health monitoring and nondestructive testing (NDT) are necessary to diagnose, identify and characterize defects and/or damages and qualify repairs in many industrial applications, such as aerospace, marine, nuclear structures, etc¹⁻⁴. In addition, non-invasive medical diagnostics^{5,6} are also crucial to improve the human health and living quality. Ultrasonic techniques are frequently used for these purposes because of their subsurface inspection capabilities, fast inspection speed, simplicity and ease of operation. In these applications, ultrasonic transducers (UTs) may need to contact directly with the object to be diagnosed. However, material parts and human body have curved surfaces or complex geometries conventional UTs show poor inspection performance^{1,3}. Flexible UTs are suitable for these applications because they insure the self-alignment to the object surface even with curved or complex geometry so that the transmitted ultrasonic energy can be maximized to improve diagnoses³.

The required characteristics of flexible UTs are capability to transfer ultrasonic energy into various materials e.g. industrial steels, graphite/epoxy composites, human flesh, etc with efficiency, reliability and cost effectiveness. Often industrial applications require high temperature (HT) operation of transducers to perform in-situ characterization of materials, real-time process monitoring, and NDT^{7,8}. This leads to the research interest in developing flexible transducers, which may be operated at HT.

In commercially available flexible transducers, piezoelectric polymers such as polyvinylidene fluoride⁹ and piezoelectric ceramic/polymer composites^{7,10} are mainly used as piezoelectric materials. Both materials include polymer which prevents the use of such flexible UTs at elevated temperature. Other flexible HTUTs have been also reported^{11,12}. Because of single crystal films used, in order to provide the flexibility, the thickness of the piezoelectric film is thin, from 0.2 to 10µm. Thus operating frequency was normally higher than 30MHz and it might not be suitable for diagnostics of thick and highly attenuating materials such as graphite/epoxy composites and human flesh.

This study proposes a new concept for flexible transducers to meet these demands. They consist of a thin polyimide membrane or stainless steel (SS) foil, a bottom electrode if the substrate is polyimide membrane, a piezoelectric ceramic film and a top electrode. The fabrication process of the piezoelectric film is based on the sol-gel spray technique^{13,14}. The piezoelectric lead-zirconate-titanate (PZT) particles are dispersed in PZT sol-gel solution for producing thick PZT/PZT composite film. The top electrode can be made of silver paste¹⁴ or paint. The flexibility is achieved because of the porosity in the composite PZT/PZT films and thinness of the polyimide membrane or SS foil, and top and bottom

electrodes. PZT/PZT composite was selected because of its high piezoelectric strength and Curie temperature of 350°C. Ultrasonic performance in pulse-echo mode of these flexible UTs will be investigated.

2. FABRICATION AND CHARACTERIZATION OF PIEZOELECTRIC FILMS

The detailed fabrication process of sol-gel spray technique can be found from previous publications^{13,14}. Piezoelectric PZT powders in this study, were purchased. Fine and submicron size powders were dispersed into PZT sol-gel solution by the ball milling method to achieve the paint for spray. When non-conductive substrates i.e. polyimide membranes were used, it was required to fabricate the bottom electrode layer before spray coating of piezoelectric PZT/PZT composite film. In this investigation, two methods, spray coating and electroless plating, were chosen to build up bottom electrode layer. For spray coating, silver colloid was sprayed directly onto polyimide membrane with using an airbrush, and cured around 120°C. The thickness of the sprayed silver layer was 5-10µm. For electroless plating, polyimide membranes were immersed in electroless nickel bath containing nickel salt, reducing agent such as sodium hypophosphate, and complexing agent for nickel, after pretreatment process. The electroless bath was heated up to 90°C and the immersion time was about 10minutes. The thickness of electroless plated nickel alloy layer was less than 1µm. When SS foils were used as substrates, it served as bottom electrode. An airbrush was then used to spray the PZT/PZT sol-gel composite directly onto bottom electrodes of the polyimide membranes or SS foils. Paper masks were used to serve as the shadow mask during the spray coating. After each coating, thermal treatments were carried out. Multiple layers were made in order to reach desired film thickness.

Each layer had a thickness of 5-15µm and eight to twelve layers were made in order to reach the desired thickness. The film thickness was measured by a micrometer. The films were then electrically poled using the corona discharging technique. For corona poling, the temperature of the polyimide membrane and that of SS foil were around 120°C and a high positive voltage supplied from a 28kV DC power supply was fed into a thin and sharp needle located several centimeters above the PZT/PZT film coated on the substrates. The distance and voltage were optimized for different film thicknesses and geometries. The poling time was about 10 minutes. The corona poling method was chosen because it could pole the piezoelectric film over a large area with different curvatures and with ease¹⁴. Finally, the top electrode was fabricated at room temperature by Silver Kwik-Stik from SPI Supplies, PA. When the tip of the pen was pressed against the film surface, it retracted into the pen barrel, allowing silver colloid to flow through the opening. The opening and trace width of the pen is approximately 1.0mm and 2.2mm, respectively. The thickness of the silver top electrode was about 20µm with fairly good uniformity. This convenient approach makes the selection of electrode size, i.e. the UT size, simple. The silver paste had been tested and its operating temperature could be up to at least 220°C.

In this investigation, the thickness range of the PZT/PZT films fabricated by sol-gel spray technique was from 50 to 120µm, and it provided an operation frequency range from 2.4 to 13.8MHz. This frequency range is commonly preferred for NDT and health monitoring applications because it provides sufficient time delay resolution and allows acceptable ultrasonic attenuation in metals, composites and human flesh. It is noted that PZT sol-gel solution contributed as bonding material between the PZT powder and the bottom electrode on polyimide membrane or SS foil. The relative dielectric constant of the PZT/PZT films was around 140 measured by a Hewlett Packard 4192A LF Impedance Analyzer at 1 kHz. This value was lower than that of PZT/PZT on a bulk substrate that was around 320¹⁵. It was probably due to the different temperature profiles during the thermal treatments. A porosity of more than 20 volume % was estimated from the relatively low dielectric constant of the PZT/PZT films¹⁵. The electromechanical coupling coefficient k_t of the composite film was measured to be near 0.2. The d_{33} measured by an optical interferometer was 30×10^{-12} m/V.

3. FLEXIBLULTRASONIC TRANSDUCER WITH DIFFERENT SUBSTRATES

3.1 Flexible U T using polyimide membrane as substrates

Firstly, 60µm thick polyimide membranes are used for the investigation because polyimide can sustain 350°C and the sol-gel PZT/PZT multilayer fabrication process. Its acoustic impedance is close to that of liquid such as water, polymer composite and human flesh. Because the polyimide film is an insulator, both colloidal silver spray and nickel alloy electroless plating methods were developed to coat a conductive layer onto the piezoelectric composite film side of the film as the bottom electrode before the coating of the piezoelectric PZT/PZT composite films.

For the fabrication of bottom electrode on 60µm thick polyimide membrane colloidal silver spray method was used as shown in Fig.1a at first. Five to ten microns thick silver paint was deposited between the top polyimide membrane and the 60µm thick PZT/PZT composite as shown in Figs.1b and 1c without and with top electrode, respectively. The top

electrode was made by silver paste and its thickness was around $20\mu\text{m}$. Care was taken to strengthen the adhesion between this bottom electrode layer and the polyimide membrane. The PZT/PZT film thickness was obtained using ten layers of coating process. Such fabrication process implies that the polyimide membrane has sustained ten thermal cycles during the drying and firing of the PZT/PZT composite film and also the corona poling heating process. In order to optimize the size of the top electrode, experiments were carried out. The results of signal amplitude versus the diameter of the top electrode shown in Fig.2 indicates that the optimal electrode size for this flexible UT is 10mm in diameter.

Fig. 1. A flexible UT using polyimide membrane as the substrate with sprayed silver film as top electrode; (a) schematic diagram, (b) without and (c) with top electrode.

Fig. 2. Signal amplitude versus the diameter of the top electrode of the flexible UT shown in Fig.1c.

The flexible UT shown in Fig.1c was then was pressed onto a 13.8mm thick SS plate at room temperature and 150°C . HT oil couplant was placed between the probing side of the polyimide membrane and SS plate. Figs.3a and 3b show the transducer response, in time and frequency domains respectively, in pulse-echo mode at room temperature. L_2 and L_4 are the first and second round trip echoes through the thickness of the SS plate. The center frequency, the 6dB band width and SNR of the L_2 echo are determined to be 11.3MHz, 5.1MHz and 24dB, respectively. The ultrasonic response of the transducer at 150°C is shown in Fig.4. The center frequency, the 6dB bandwidth and SNR of the L_2 echo at this operating temperature is 9.3MHz, 3.7MHz and 27dB, respectively. It is observed that the signal strength of the L_2 echo at room temperature was decreased by about 5dB and the frequency bandwidth reduced by 1.4MHz as flexible UT operated at 150°C . At 150°C , the time delay of the L_2 echo traveling in SS substrate had $0.14\mu\text{s}$ more delay than that measured at room temperature.

Fig. 3. Ultrasonic performance of the flexible UT shown in Fig.1c pressed onto a 13.8mm thick SS plate in (a) time and (b) frequency domain at room temperature. Pulse-echo mode was used.

Fig. 4. Ultrasonic performance of the flexible UT shown in Fig.1c pressed onto a 13.8mm thick SS plate in (a) time and (b) frequency domain at 150°C. Pulse-echo mode was used.

The flexible UT shown in Figs.1b and 1c can be made as UT array if many top electrodes can be added onto the surface of poled PZT/PZT composite film. This demonstrates the simplicity of the technique to achieve UT array. The corona poling technique has been used to pole such piezoelectric film up to 100mm by 100mm area in one time. Fig.5 shows a flexible four UTs array using 60μm thick polyimide membrane as the substrate. It has four top electrodes. The electrical connection and protection of the UT array will be introduced in Section 3.2.

Fig. 5. A flexible four UTs array using polyimide membrane as the substrate.

The second approach to fabricate the bottom electrode onto the top surface of the 60μm polyimide membrane was using the electroless plating technique as shown in Fig.6a. The pretreatment including cleaning, chemical roughening, sensibilization, activation and reduction process was operated for polyimide membranes before the immersion into the electroless nickel bath. After the pretreatment, polyimide membranes were immersed into hypophosphite bath heated up to 90°C for 10minutes in order to deposit nickel alloy bottom electrode layer onto polyimide membrane. The thickness of electroless nickel alloy layer was less than 1μm. In order to optimize the size of the top electrode, experiments were carried out. The results of signal amplitude versus the diameter of the top electrode shown in Fig.6 indicates that the optimal electrode size for this flexible UT is 11mm in diameter.

Fig. 6. A flexible UT using polyimide membrane as the substrate with electroless coated nickel alloy film as bottom electrode; (a) schematic diagram and (b) with top electrode.

Fig. 7. Signal amplitude versus the diameter of the top electrode of the flexible UT shown in Fig.6b.

The fabrication of PZT/PZT composite film and the top electrode and poling technique were the same as those described for the silver sprayed bottom electrode samples. Then the flexible UT shown in Fig.6b was pressed onto a 13.8mm thick SS plate at room temperature and 150°C. HT oil couplant was also placed between the probing side of the polyimide membrane and SS plate. Figs.8a and 8b show the transducer response, in time and frequency domains respectively, in pulse-echo mode at room temperature. L_2 and L_4 are the first and second round trip echoes through the thickness of the SS plate. The center frequency, the 6dB band width and SNR of the L_2 echo are determined to be 13.8MHz, 5.5MHz and 17dB, respectively. The ultrasonic response of the transducer at 150°C is shown in Fig.9. The center frequency, the 6dB bandwidth and SNR of the L_2 echo at this operating temperature is 11.3MHz, 5.1MHz and 22dB, respectively. It is observed that the signal strength of the L_2 echo at room temperature was decreased by about 6dB and the frequency bandwidth reduced by 0.4MHz as the flexible UT operated at 150°C.

Fig. 8. Ultrasonic performance of the flexible UT shown in Fig.6b pressed onto a 13.8mm thick SS plate in (a) time and (b) frequency domain at room temperature. Pulse-echo mode was used.

Fig. 9. Ultrasonic performance of the flexible UT shown in Fig.6b pressed onto a 13.8mm thick SS plate in (a) time and (b) frequency domain at 150°C. Pulse-echo mode was used.

3.2 Flexible U T using SS foil as substrates

Since SS substrate can sustain operation temperature more than several hundred degrees Celsius ($> 700^{\circ}\text{C}$), it may be brazed or welded onto metallic structures for HT operation. It may be also glued onto metallic, polymer or graphite/epoxy composite materials for NDT and structure health monitoring applications. The techniques described in Section 2 were used to fabricate the PZT/PZT composite films. The top electrodes were also made by silver paste. Fig.10 illustrates two views of a PZT/PZT 120 μm thick composite film five UTs array directly fabricated onto a 75 μm thick SS foil with this particular merit. Thinner SS foils may also be used as reported in ref.¹⁶. The entire transducer array structure was sandwiched by 60 μm thick polyimide membranes excluding the probing side of the SS foil (the side opposite to thick piezoelectric film) so that it may be protected from the moisture in the environment and can operate at temperatures up to 150°C ¹⁶. Copper strips were used for electrical connections. Such protection and electrical connection techniques can be also used to fabricate UT array with polyimide membranes as substrates mentioned in Section 3.1.

Fig. 10. A flexible PZT/PZT 120 μm thick composite film five UT array using a 75 μm thick SS foil.

When this flexible UT array was immersed into water for days the ultrasonic performance was remained the same. Fig.11a shows the water immersion measurement result from UT3 which was 46 mm away from the front surface of a 25.4 mm thick flat aluminum plate. L^1 was the reflected echo from the front surface. L_2 and L_4 were the first and second round trip echoes through the thickness of the aluminum plate. The frequency domain of the echo L_2 is shown in Fig.11b. It can be seen that the echoes have good signal to noise ratio even the alignment was not perfect due the flexibility of the UT array and its slight curvature caused by the residue stress induced during the fabrication of PZT/PZT composite. Such type of flexible transducers was also used for NDT of plastics and graphite/epoxy samples at room temperature and metal substrate with curved and flat surfaces at 150°C ¹⁶.

Fig. 11. Ultrasonic performance of the flexible ultrasonic transducer (UT3) in (a) time and (b) frequency domains for water immersion measurement at room temperature.

4. CONCLUSIONS

Flexible UTs consisting of 60 μ m thick polyimide membrane with a bottom electrode or 75 μ m thick SS foil, a piezoelectric PZT/PZT composite films and top around 20 μ m thick silver paste electrodes have been developed. The bottom electrodes on polyimide membranes were made by two methods; silver paint spray and electroless plating of nickel alloy which provide the thickness of 5 to 10 μ m and less than 1 μ m electrode, respectively. The flexibility of the transducer was realized owing to the porosity of piezoelectric film and the thinness of substrate and electrodes. PZT/PZT composite was chosen as piezoelectric film because of its high piezoelectricity and its thickness was from 50 to 120 μ m fabricated by a spray technique. The piezoelectric constant d_{33} of PZT/PZT was measured by the optical interferometer. The UT array was configured by the several top electrodes. For protection purpose polyimide membranes were glued onto the top electrodes of the UT array. Copper thin and narrow strips were used as the flexible wire for electrical connections. Such UT array can be attached to the desired structures or materials for NDT and health structural monitoring applications at room and elevated temperatures. The flexible UT array was successfully tested at 150°C and also immersed into water as immersion ultrasonic probe operated in the pulse-echo mode with good signal to noise ratio.

ACKNOWLEDGMENTS

The authors are grateful to H. Hébert and J.-F. Moisan for their technical assistance. Financial support from the Natural Sciences and Engineering Research Council of Canada is acknowledged.

REFERENCES

1. O. Roy, S. Mahaut, and O. Casula, "Development of a smart flexible transducer to inspect component of complex geometry: modeling and experiment", *Review of Progress in Quantitative Nondestructive Evaluation*, vol.21, pp.908-914, 2002.
2. D. H. Wang and S. L. Huang, "Health monitoring and diagnosis for flexible structures with PVDF piezoelectric film sensor array", *Journal of Intelligent Material Systems and Structures*, vol.11, pp.482-491, 2000.
3. R. S. Frankle and D. N. Rose, "Flexible ultrasonic array system for inspecting thick composite structures", in *Proc. SPIE*, vol.2459, pp.51-59, 1995.
4. M. V. Gandhi and B. S. Thompson, *Smart materials and structures*. London ; New York : Chapman & Hall, 1992.
5. B. Wilkinson, I. R. Hall, I. S. MacCallum, C. M. McEniery, B. J. Van Der Arend, Y. E. Shu, L. S. MacKay, D. J. Webb, and J. R. Cockcroft, "Pulse-wave analysis: clinical evaluation of a noninvasive, widely applicable method for assessing endothelial function", *Arterioscler Thromb. Vasc. Biol.*, vol. 22, pp. 147-152, 2002.
6. M. Akiyama, N. Ueno, K. Nonaka, and H. Tateyama, "Flexible pulse-wave sensors from oriented aluminum nitride nanocolumns", *Appl. Phys. Lett.*, vol. 82, pp. 1977-1979, 2003.
7. C. Devallencourt, S. Michau, C. Bantignies, and N. Felix, "A 5 MHz piezocomposite ultrasound array for operations in high temperature and harsh environment", in *Proc. IEEE Ultrason. Symp.*, pp.1294-1297, 2004.
8. H. Karasawa, M. Izumi, T. Suzuki, S. Nagai, M. Tamura, and S. Fujimori, "Development of under-sodium three-dimensional visual inspection technique using matrix-arrayed ultrasonic transducer", *Journal of Nuclear Science and Technology*, vol.37, pp.769-779, 2000.
9. L. F. Brown and A. M. Fowler, "High vinylidene-fluoride content P(VDF-TrFE) films for ultrasound transducers", in *Proc. IEEE Ultrason. Symp.*, pp.607-609, 1998.
10. L. J. Bowen, R. L. Gentilman, H. T. Pham, D. F. Fiore, and K. W. French, "Injection molded fine-scale piezoelectric composite transducers", in *Proc. IEEE Ultrason. Symp.*, pp.499-503, 1993.
11. M. Akiyama, N. Ueno, K. Ikeda, K. Nonaka, and H. Tateyama, "High-sensitivity flexible ceramic sensor", *U.S. Patent*, 6 608 427 B2, 2003.
12. M. Akiyama, T. Kamohara, K. Nishikubo, N. Ueno, H. Nagai, and T. Okutani, "Ultrahigh temperature vibration sensors using aluminum nitride thin films and W/Ru multilayer electrodes", *Appl. Phys. Lett.*, vol.86, 022106 (3 pages), 2005.
13. D. A. Barrow, T. E. Petroff, R. P. Tandon, and M. Sayer, "Characterization of thick lead zirconate titanate films fabricated using a new sol gel based process", *J. Appl. Phys.*, vol.81, pp.876-881, 1997.

14. M. Kobayashi and C.-K. Jen, "Piezoelectric thick bismuth titanate/PZT composite film transducers for smart NDE of metals", *Smart Materials and Structures*, vol.13, pp.951-956, 2004.
15. M. Kobayashi, "Development and applications of high temperature piezoelectric ultrasonic transducers", Ph. D. thesis, Department of Electrical and Computer Engineering, McGill University, June 2004.
16. M. Kobayashi, C.-K. Jen, and D. Lévesque, "Flexible ultrasonic transducers," *IEEE Trans. Ultrason. Ferroelect. Freq. Contr.*, vol.53, pp.1478-1486, 2006.