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Investigation of diffusion bonding quality by ultrasonic technique

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Abstract

For high-temperature ultrasonic transducers piezoelement-to-metal thermosonic bonding technology was developed based on gold-to-gold diffusion. Bond quality strongly depends on bonding parameters – pressure, temperature, ultrasonic power and time. The first step for achieving good bonds was ultrasonic monitoring of the process. In ultrasonic transducers good diffusion bond means good acoustic coupling of piezoelement to the protector and this coupling must withstand high temperatures. Bond quality tests comprise piezoelement electrodes adhesion issues as well. Bismuth titanate and PZT piezoelements were bonded to thick stainless steel protectors and membranes. For bond quality determination normal-incidence immersion scanning method from metal side was chosen. It is based on measurement of reflection/transmission signals in the bond interface and determination of their ratio which defines acoustic coupling. The whole piezoelement bonding interface was scanned automatically in *x* and *y* directions with respect to 10 MHz wideband ultrasonic transducer, having lateral beam width 1.4 mm at – 6 dB level. With optimal bonding parameters good bond quality was found. Weak or non-bonded areas are absent. Finally thus bonded transducers were used in successful temperature experiments – pulse response measurements up to 400 °C and tests in a liquid Pb/Bi at 290 °C.

Keywords: diffusion bonding, piezoelement, bond quality.

Introduction

For high-temperature ultrasonic transducers we have developed piezoelement-to-metal protector thermosonic bonding technology. This technology is based on a gold-to-gold diffusion, so contacting surfaces were gold electroplated. Bonding parameters – pressure, temperature, ultrasonic power and time – had to be delicately balanced in order to get quality bonds.

Application of ultrasound in bonding process has a decisive role. Due to the tangential relative motion at the bond interface, metal/metal contact is cleaned, and the diffusion of atoms across the bond takes place. The thickness of the diffusion layer is determined by Fick's law [1] and depends on activation energy required to initiate the diffusion. It consists of kinetic (ultrasonic scrub) and thermal (heat) energies.

Thermosonic bonding procedures were optimized using ultrasonic monitoring of the process. Essential, that for the monitoring the same piezoelement is exploited, which is bonded. Ultrasonic monitoring is possible if the protector thickness equals to some wavelengts, what is necessary for observing the back reflection. So during the bonding process the pulse response of the transducer is observed with its transformations, depending on bonding parameters. Wide-band signal of maximal amplitude is seeked. This is the first step necessary for achieving good bonds. Poorly bonded piezoelement demonstrates ringing, well bonded–short pulse. With optimized pressure, temperature, ultrasonic power and time some piezoelements were bonded to the thick protector. Thus determined optimal parameters can be applied for piezoelement bonding to a thin membrane, as ultrasonic monitoring of the process is complicated in this case.

Earlier studies of diffusion bonds have used destructive techniques to estimate the degree of bonding. This technique was found to be inaccurate because a bond can display the tensile strength of the parent material when as 80 % bonding occurs [2].

For quality control of bonds NDT techniques must be used. Ultrasonic inspection is one of the most efficient NDT methods available for a bond evaluation. Longitudinal, shear and Lamb waves can be used. Ultrasonic detection of total disbonds, where an air gap exists between two layers, is always successful. But it is very difficult to quantify the degree of bond weakness [3]. Bond evaluation method usually requires single normal-incidence ultrasonic immersion measurement. When a diffusion bond is not perfect some ultrasonic energy is reflected from the interface separating the two substrates. Reflected amplitude correlates qualitatively with the bond strength, but quantitive correlation is difficult to establish. Normal-incidence ultrasonics can detect and size only relatively large individual disbonds. Often the size of an individual defect is much smaller than insonification area, and effects of many defects are averaged [4]. In the case of a focused transducer more precise mapping can be provided. Imperfect bond can be interpreted in terms of an interfacial spring model [5]. Reflected signal is dependent not only on impedances of contacting materials Z_1 and Z_2, but on angular frequency ω and on distributed spring constants. By measuring the frequency response of a reflected signal one can determine the bond quality. This new promising method has limitations as requires additional information about the bond [4, 6].

The quality of the diffusion bond can be determined by measuring the anharmonic content of a transmitted through the bond ultrasonic wave. The anharmonicity is caused by weak bonds. The source of nonlinearity is located in the rim of delaminations in the interface [7]. Dissimilar material bonding joint quality can be evaluated from echo signal intensity and its phase by an automatic C-scan test method [8]. For example, anodic bonding interface [9] was successfully examined with 20 MHz focused ultrasonic transducer. Well-bonded area/area of interface ratio was determined only from echo-signal variation. Despite successful applications of diffusion bonding, there is a lack of good quality assessment methods. Defects found in diffusion bonds often are small, and acoustic microscopy is considered as a highly encouraging new line of investigations due to the very high frequency of ultrasound used [2]. An attempt is known to apply air-coupled ultrasonics for testing of diffusion bonds, where immersion or contact is not wanted or even dangerous. A frequency method was proposed based on the study of resonances of the whole plate-plate structure [10].

There is known a specific bond type–so called kissing bond. It represents disbonds where surfaces are in intimate contact but with a weak bonding. These kissing bonds cannot be detected under classical NDT inspection techniques because there is no noticeable separation between the adherent and adhesive surfaces. These bonds are examined using ultrasonic resonance spectroscopy, frequencies $f > 25$ MHz are necessary [11].

In ultrasonic transducers good diffusion bond means good acoustic coupling of piezoelement to the protector, and this coupling must withstand high temperatures. More, bond quality tests comprise piezoelement electrodes adhesion issues as well. In principle, the bond strength is not critical in this case. Piezoelement-to-metal bonding and the product application are specific, so quality testing has its specific features. The chosen normal-incidence immersion scanning method is based on measurement of reflection/transmission signals and determination of their ratio, which defines the bond quality and acoustic coupling.

Objective of this paper was ultrasonic investigation of thermosonic diffusion bonding of piezoelectric ceramic elements to a steel substrate.

Experimental investigations

Test principles

Simple ultrasonic tests of bonding quality may be provided with commercial delay line transducers of smaller dimensions and higher frequency in order to get the necessary lateral resolution. Measurements are performed from the protector side. Bonding interface in these tests must be protected from water which in used as a coupling medium between this transducer and protector. Protector-to-piezoelement bonding quality evaluation is based on the fundamental transmission-reflection laws:

- In a non bonded area the ultrasonic incident wave is completely reflected from this interface, if the ultrasonic beam cross-section is smaller than this area. In this case no piezoelement back reflection 2 exists;
- In perfectly bonded case both transmited and reflected waves are observed. The amplitudes of these signals can be calculated from the acoustic impedances of these two media. In this case maximal piezoelement back reflection 2 can be seen. It must be significantly larger than reflection from the bonding interface;
- In the case of a poor bonding the interface reflection 1 can be larger than the back reflection 2.

These simple tests give fast overall evaluation of a larger bonded area. For “point-by-point” detailed tests scanning in a water tank was provided using 10 MHz ultrasonic transducer of smaller dimensions, developed by authors. It has Ø3 mm PZT piezoelement and soldered composite metallic backing. The transducer is wideband, its $\Delta f/f$=106 % and the ultrasonic beam width is 1.4 mm at –6 dB level. The principle of the bond quality evaluation is presented in Fig. 1.

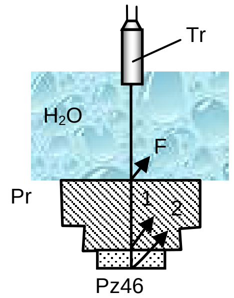

Fig. 1. **Bonding quality determination principle: Tr – ultrasonic transducer; F – front reflection, not used; Pr – protector; 1 – bonding interface reflection; 2 – piezoelement back reflection**

The measurement set-up for the first investigation of the bonding quality is shown in Fig. 2. Semi-manual scanning is provided with two *x-y* micrometers. The amplitudes of reflected signals are measured by digital oscilloscope.

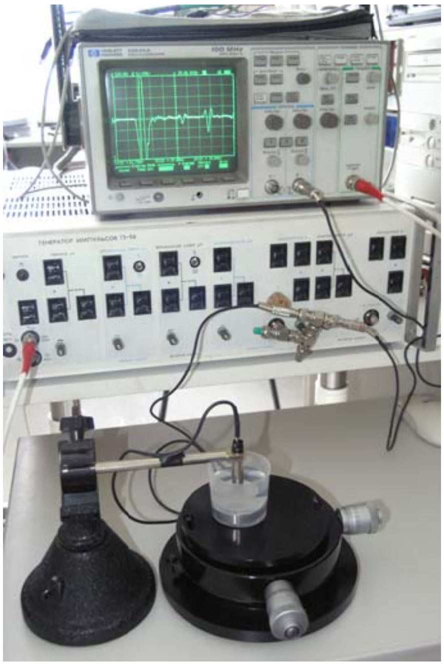

Fig. 2. **Measurement set-up for the first investigations**

Test objects

Bonding quality of some structures was investigated. First of all, experiments were provided with bismuth titanate piezoelement bonded to a thick protector. Due to the ultrasonic monitoring of the pulse response during this bonding process, good bonding results were expected. Electrodes were made new on bare piezoelement especially for the bonding purposes using Ni electroless and Au electroplating processes. Second, PZT and bismuth titanate piezoelements were bonded to the membranes, λ/2 and λ thicknesses. Ultrasonic monitoring was problematic here, thus earlier determined bonding parameters were applied expecting that they were optimal. Later we managed to observe the pulse response and adjust the pressure, etc. In one case of the experiments manufacturer's Ag electrodes were left on the piezoelement, but additionally Au electroplated. So, different quality was anticipated in various bondings to membranes.

Pulse and frequency responses

Before the diffusion bonding experiments it was interesting to know what pulse response of the transducer can be expected in a thick protector if bonding would be ideal. For that purpose 5 MHz Ø12.6 mm bismuth titanate piezoelement was accurately soldered with Rose solder (T_{melt} ~ 100 °C) to the 15 mm (13 λ) thickness protector, made from stainless steel AISI-316L (Fig. 3). For better soldering stainless steel was Ni and Ag electroplated.

Fig. 3. **Stainless steel thick protector with bismuth titanate piezoelement, soldered or thermosonically bonded**

Pulse and frequency responses of the soldered piezoelement are shown in Fig. 4. Piezoelement is highly damped and therefore possesses wide frequency band $\Delta f/f$=110% at –6 dB level.

In the case of a thick protector some piezoelements were bonded with optimized and identical thermosonic bonding parameters. We observed surprisingly identical pulse responses as well (Fig. 5). It means that bond quality is the same. More, pulse and frequency responses of soldered and thermosonically bonded piezoelements are very similar. Signal amplitude for bonded piezoelement is only 4% lower. In our numerous experiments it was impossible to increase the amplitude. May be, we attained the limit, which is conditioned by a lot of factors and especially by quality of gold coated contacting surfaces of piezoelement and protector. Surfaces were flat (tested for Newton rings) and polished, but some roughness obviously remained.

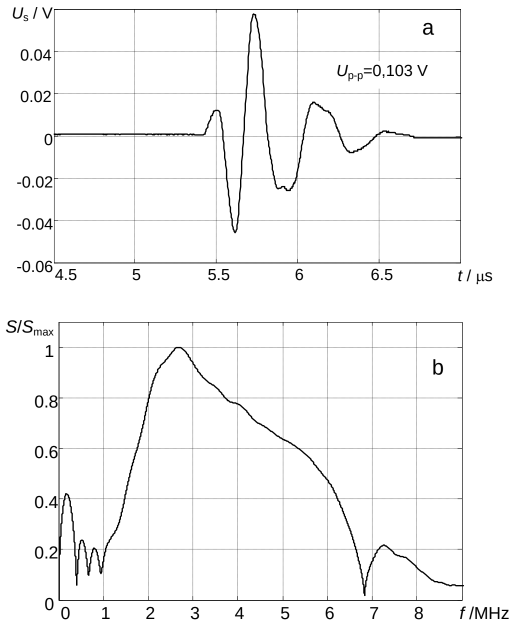

Fig. 4. **Pulse (a) and frequency (b) responses of the soldered piezoelement to the thick protector**

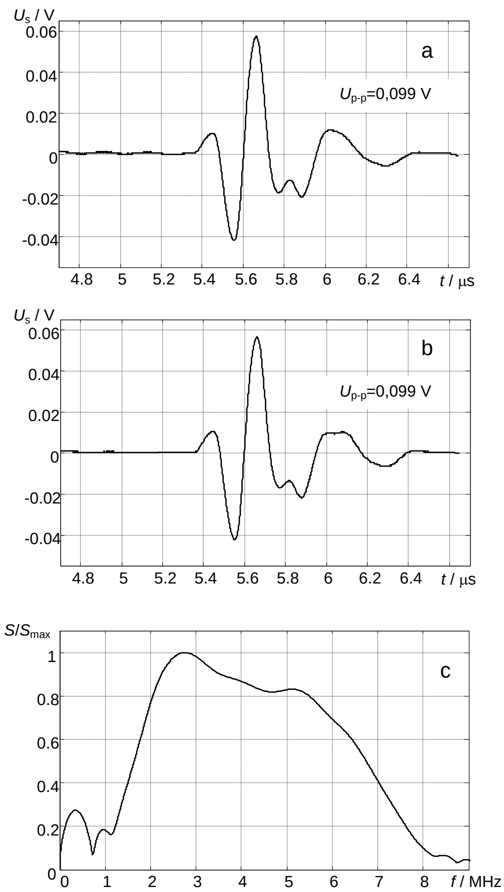

Fig. 5. **Pulse responses (a, b) of the two identical thermosonically bonded piezoelements and their frequency response (c).**

Transmission/reflection calculations

Before the experiments it is necessary to calculate what ratio of reflections amplitudes we can expect in the case of ideal bonding. Transmission T and reflection R coefficients of plane waves at the boundary of two media 1 and 2 can be calculated according to the known formulas:

$$T_{12} = 2Z_2/(Z_1 + Z_2), \quad (1)$$

$$T_{21} = 2Z_1/(Z_1 + Z_2), \quad (2)$$

$$R_{12} = (Z_2 - Z_1)/(Z_1 + Z_2), \quad (3)$$

here: Z_1 = 45 MRayl is the acoustic impedance of stainless steel, Z_2 = 26.2 MRayl is the acoustic impedance of bismuth titanate.

As it was mentioned in the introduction, testing of diffusion bonded materials has its pecularities. Ultrasonic wave normal incidence transmission and reflection coefficients in the case of imperfect interfaces modeled by springs are given by [4]:

$$T_{12} = \frac{2Z_2}{Z_2 + Z_1 - i(\omega / K_n)Z_1 Z_2}, \quad (4)$$

$$R_{12} = \frac{Z_2 - Z_1 + i(\omega / K_n)Z_1 Z_2}{Z_2 + Z_1 - i(\omega / K_n)Z_1 Z_2}. \quad (5)$$

An important feature of these transmission and reflection coefficients is the existence of the frequency-dependent term; K_n is distributed spring constant per unit area (N/m3). There are known attempts to provide spectroscopic measurements of T_{12} and R_{12} in the frequency range 5 – 15 MHz and from these results to determine diffusion bond strength, characterized in MPa. For this purpose broadband 5 MHz and 10 MHz transducers were used. The method must be combined with other information about the bond, thus has a limited application.

Usually the coefficient K_n is not known in advance, therefore for interpretation of our point-by-point measurements and scanning results, we decided to apply more simple Eq. 1-3. At the same time we keep in mind the fact that the results may a little depend on frequency, as well as on bonding interlayers. Piezoelement electrodes, gold coatings of electrodes and steel, as well as gold foil may have total thickness more than 30 μm. Silver's acoustic impedance is 39 MRayl, gold's – 63 MRayl.

With mentioned above acoustical impedances of stainless steel and bismuth titanate the transmission coefficient is T_{12} = 0.73. For the first reflection from bonding interface steel-piezoelement, the reflection coefficient is

$$R_{\mathrm{I}} = R_{12} = -0.26$$

The transmission coefficient from piezoelement to steel T_{21} = 1.25, the reflection coefficient piezoelement-air R_{2air} = – 1. The second reflection from the piezoelement back surface is

$$R_{\mathrm{II}} = T_{12} \times R_{2air} \times T_{21} = -0.91.$$

Thus both reflections have the same reversed phase and their ratio is – 0.91/ – 0.26 = 3.5. It means that according to Eq.1-3 for ideal bonding the ratio of the experimental reflections amplitudes is 3.5.

Scanning of a thick protector

First "by-hand" scanning of the interface was made along the piezoelement diameter for thermosonically bonded and soldered (for comparison) versions of transducers. Typical signals are shown in Fig. 6. In principle, there is no difference in these signals. This means, that bonded and soldered piezoelements demonstrate the same quality of coupling. The wideband 10 MHz ultrasonic transducer guarantees the necessary spatial resolution in the case of 400 μm piezoelement thickness. "Point-by-point" tests were provided manually with micrometer screws scanning the bonded area with a respect to the immersion transducer.

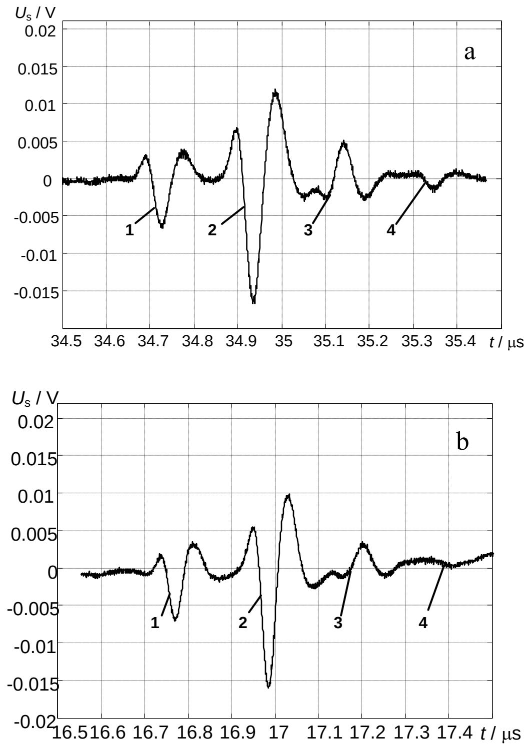

Fig. 6. **Observed signals in the case of a thick protector for thermosonically bonded (a) and soldered (b) piezoelement: 1 – interface reflection; 2 – Pz46 back reflection; 3, 4 – multiple reflections, not used**

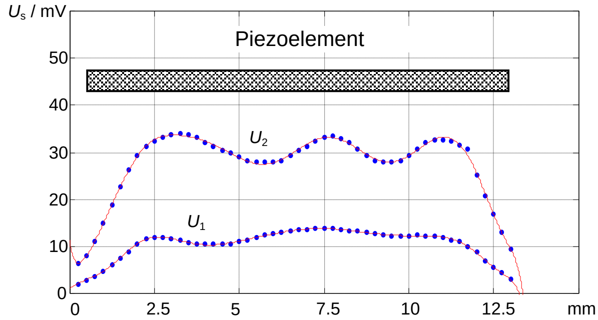

Fig. 7. **Scanning results along the piezoelement diameter: U_1 – interface reflection; U_2 – Pz46 back reflection; on the top the actual piezoelement size is shown**.

The scanning step was 0.25 mm, amplitudes of the interface and back reflections as well as their ratio were determined for each measurement point. The pulse response of this transducer was shown in Fig. 5. Scanning results for the bonded piezoelement are presented in Fig. 7. Variations of U_1 and U_2 are rather smooth, what allows to suggest that there are no significant defects in the bonding. When only the part of ultrasonic beam is reflected near the both edges of the piezoelement, U_1 and U_2 amplitudes decrease. Thus if the bond quality is evaluated only from U_1 and U_2 signals, false evaluation of the bond quality can be made near the edges of a piezoelement.

The same scanning was provided for the soldered version, pulse response of which has a slightly larger amplitude (Fig. 4), than the bonded. U_1 dependence was more uniform, U_2 – very similar (not shown here).

The back/interface reflections ratio U_2/U_1 characterizes the bond quality completely, not depending on the beam position towards piezoelement edges. The results in Fig. 8 show that the thermosonic bonding quality along the diameter is rather uniform. The mean ratio value is 2.7 (52 results). The maximal value is 3.2, the minimal – 2.2.

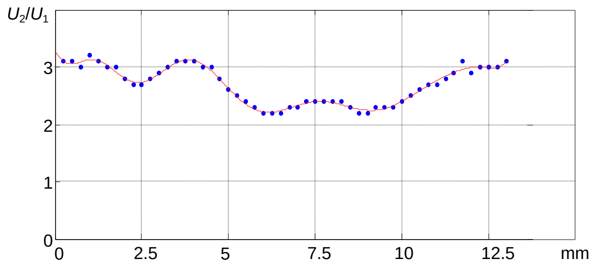

Fig. 8. **Variation of back/interface reflections ratio U_2/U_1.**

It was expected, that for the soldered version this ratio must be larger, close to the theoretical value 3.5. Unfortunately, the mean value is only 2.3 with variations between 1.9 (one result) and 3.0 (one result as well). The conclusion must be made, that simple transmission-reflection Eq. 1-3 for absolute measurement are not correct. But the application of exact formulas Eq. 4, 5 is problematic due to the unknown constant K_n. More, apparently this constant is different for thermosonic bonding and soldering, for thick protector and membranes.

The whole piezoelement bonding interface as well was scanned automatically in x and y directions with the step 0.25 mm. Results for reflection from interface and the reflections ratio are presented in Fig. 9.

Distribution of U_2 shows smaller signals near the piezoelement edges, what is evident from Fig.9a results. The distribution of U_2/U_1 (Fig.9b) is rather smooth in the whole area, only some results demonstrate a larger ratio. We can conclude that thermosonic bonding quality is good; there are no unbonded areas.

The scanning results of the bonding area were grouped according to the signals ratio:

1.9÷2.2, 14%;
2.3÷2.6, 32%;
2.7÷3.0, 36%;
3.1÷3.4, 13%;
3.5÷3.8, 5%.

These results correlate well with point-by-point measurements along the diameter (Fig. 8).

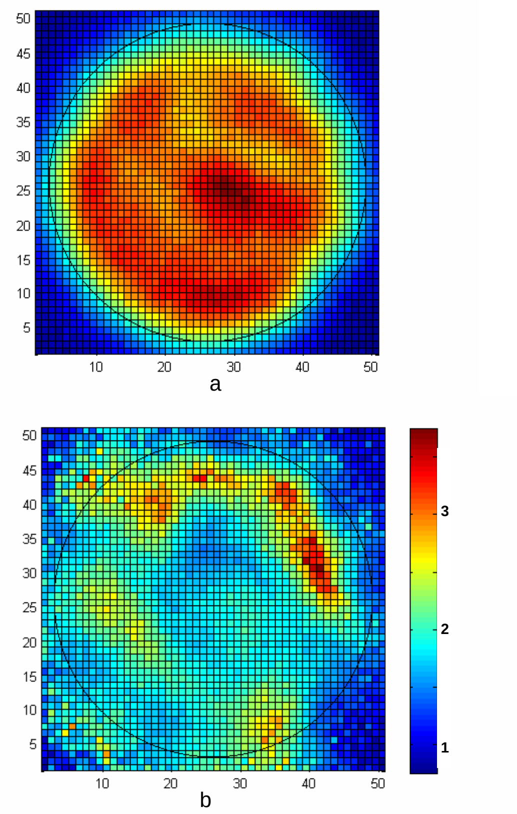

Fig. 9. **Thermosonic bonding quality detailed investigation in the whole area of the piezoelement: distribution of U_2 (a) and U_2/U_1 (b).**

High temperature experiments

Detailed NDT investigation of diffusion bonding is very important, but the final bond quality evaluation is made in real high-temperature experiments with thus bonded ultrasonic transducer (Fig. 10). Experiments were of two types: first of all a pulse response of the bonded piezoelement with a thick protector was observed at higher temperatures (up to 400 °C), and after that this ultrasonic transducer was used for ultrasound propagation experiments in a liquid Pb/Bi. Both experiments revealed excellent piezoceramics-to-metal thermosonic bonding quality and good operation at high temperatures.

Bonding to membrane results

As it was mentioned above, these thermosonic bonding experiments were made with different success, due to the original manufacture's electrodes adhesion and thermosonic monitoring problems. As an example Fig. 11 presents these results.

The bonding quality in different areas of piezoelement is quite different – from acceptable to no bonding at all. It is supposed, that a poor electrode adhesion is partially responsible for these results. The unreliable PZT-Ag electrode adhesion was noticed before the bonding.

When original piezoceramics electrodes were removed and b y authors were made completely new ones,

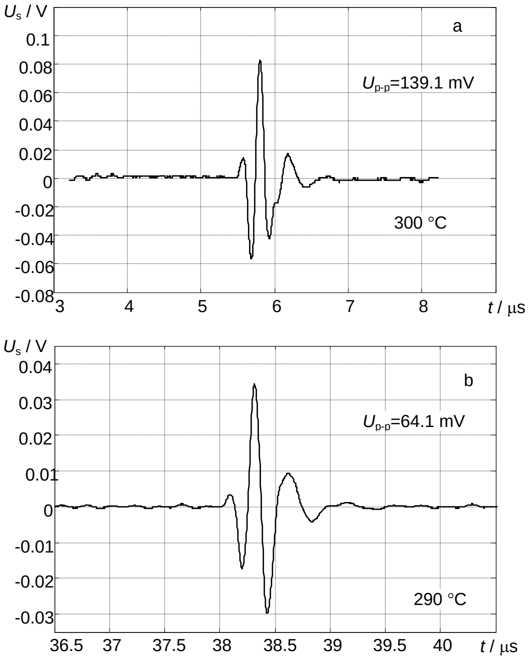

Fig. 10. **Pulse response of the piezoelement bonded to a thick protector at 300 °C (a) and the signal of this transducer observed in a liquid Pb/Bi at 290 °C (b).**

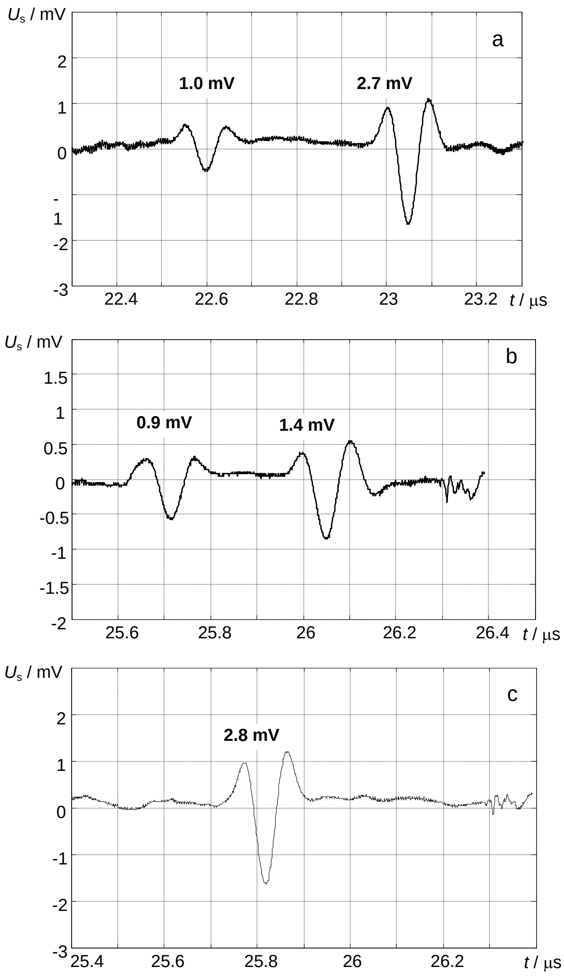

Fig. 11. **Piezoelement-to-membrane thermosonic bonding testing results in different areas of the interface: a – acceptable bonding quality, signal ratio 2.7; b – bonding is poor, ratio 1.5; c – no bonding, back reflection is absent**

much better results were achieved and non-bonded areas were not found (Fig. 12). Optimal parameters were applied during the thermosonic bonding process, as modified ultrasonic monitoring was used.

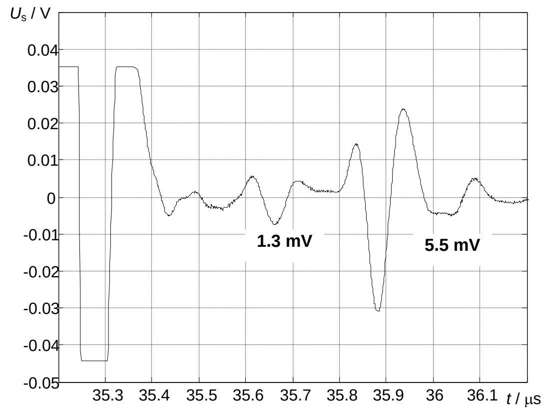

Fig. 12**. Piezoelement-to-membrane perfect bonding results, signal ratio is 4.2.**

Conclusions

Piezoelement-to-metal diffusion bonding is used for high-temperature ultrasonic transducers realization.

Transducers were made applying thermosonic bonding technology. Ultrasonic monitoring of the process is the first step in realization of high quality bonds. The bonded structure was investigated by normal-incidence ultrasonic NDT method. 10 MHz ultrasonic transducer with 1.4 mm lateral beam width was used. More precise mapping can be provided with a focused ultrasonic transducer. The whole bond area was scanned with the step 0.25 mm. Interface and back reflections were determined in each point. Their ratio characterizes the bond quality. Distribution of this signal ratio is rather smooth and uniform in the whole piezoelement area. This ratio is near to the theoretically maximal. Not bonded areas are absent.

The final bond evaluation was performed in real high-temperature experiments with thus bonded ultrasonic transducer. The pulse response in a thick protector was observed up to 400 °C, ultrasound propagation experiments in a liquid Pb/Bi were provided.

Both experiments revealed excellent piezoceramics-to-metal thermosonic bonding quality and good operation at high temperatures.

References

1. **Minh-Nhat Ba Le.** Advanced thermosonic wire bonding using high frequency ultrasonic power: optimization, bondability, and reliability. A thesis presented to the Faculty of California Polytechnic State University, San Luis Obispo. 2009.

   http://www.digitalcommons.calpoly.adu/cgi/viewcontent.cgi?article=1181&context

2. **Derby B., Briggs G. A. D., and Wallach E. R. R. E.** Non-destructive testing and acoustic microscopy of diffusion bonds. Journal of Materials Science. ISSN 0022-2461 (Print) 1573-4803 (Online). 2004. Vol. 18, No.8. P. 2345-2353.
3. **Jian X., Guo N., Dixon S. and Grattan K. T. V.** Ultrasonic weak bond evaluation in IC packing. Meas. Sci. Technol. 2006. Vol. 17. P. 2637-2642.

4. **Lavrentyev A. I., Beals J. T**. Ultrasonic measurement of the diffusion bond strength. Ultrasonics. 2000. Vol. 38. P. 513-516.
5. **Fraisse P., Schmit F., Zarembowitch. A.** Ultrasonic inspection of very thin adhesive layers. J. Appl. Phys. 1992. Vol. 72. No. 8. P. 3264-3271.
6. **Ojard G. Hughes M. S., Rehbein D. K., Buck O.** Ultrasonic determination of interface strength in diffusion bonds of dissimilar materials. In: Review of progress in quantitative non-destructive evaluation. Vol. 11B; Proceedings of the 18th Annual Review, Brunswick, ME, July 28-Aug. 2, 1991 (A93-19582 06-38). P. 1347-1354.
7. **Wegner A., Koka A., Janser K., Netzelmanu U., Hirsekorn S., Arnold W.** Assesment of the adhesion quality of fusion-welded silicon wafers with nonlinear ultrasound. Ultrasonics. 2000. Vol. 38. P. 316-321.
8. **Gang Tie, Yasuo Takahashi.** Ultrasonic echo signal fetures of dissimilar material bonding joints. Trans. Nonferrous Met. Soc. China. 2004. Vol. 14. No. 6. http://www.ysxbcn.com/Html_Article/En-3373.html
9. **Zongijie Cao, Huaidong Chen, Jin Xue, Yuwen Wang.** Evaluation of mechanical quality of field-assisted diffusion bonding by ultrasonic non-destructive method. Sensors and Actuators 2005. A 118. P. 44-48.
10. **Windels F., Leroy O.** Air-coupled ultrasonic testing of diffusion bonds. Ultrasonics. 2002. Vol. 340. P. 171-176.
11. **Marty P. N., Dedaï N., Andersson J.** NDT of kissing bond in aeronautical structures. www.ndt.net/article/wcndt2004/pdf/aerospace/193_marty.pdf

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Difuzinio sujungimo kokybės tyrimas ultragarsiniu metodu

Reziumė

Aukštos temperatūros ultragarsiniams keitikliams autoriai sukūrė pjezoelemento ir metalo sujungimo technologiją aukso-aukso dengimų difuzijos pagrindu. Bismuto titanato ir PZT pjezoelementai buvo difuzijos metodu sujungti su ultragarsinio keitiklio nerūdijančio plieno protektoriumi. Sujungimo kokybė tirta ultragarsiniu imersiniu metodu aukštojo dažnio ultragarsiniu zondu skenuojant sujungimo zoną ir matuojant atspindėtus ir perėjusius signalus, taip pat nustatant jų santykį. Taikant optimalius proceso parametrus (slėgį, temperatūrą, ultragarsinio poveikio dozę ir laiką) gauti geros kokybės sujungimai. Šie ultragarsiniai keitikliai buvo tiriami iki 400 °C temperatūroje ir skysto švino/bismuto lydinio aplinkoje.

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