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PZT-based high-temperature ultrasonic dual mode transducer applicable to cubic-anvil apparatus for sound velocity measurement

WEI SONG[1], CHANG SU[2], SHUAI WANG[1,3] AND YONGGANG LIU[1]*

[1]*Key Laboratory of High-Temperature and High-Pressure Study of the Earth's Interior,*
Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
[2]*School of Earth Sciences, Institute of Disaster Prevention, Sanhe 065201, China*
[3]*University of Chinese Academy of Sciences, Beijing 100049, China*

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The ultrasonic transducer is an indispensably critical component for sound velocity measurement. With the increasing demand of high-temperature ultrasonic dual mode transducer applicable to cubic-anvil apparatus, where experiments of high-pressure and high-temperature sound velocity are routinely conducted, a PZT-based ultrasonic dual mode transducer was presented in this study. It was made of a sandwich of two PZT piezoceramic wafers, the upper one generating P-waves and the lower one generating S-waves. The transducer was directly bonded onto a WC anvil of cubic-anvil apparatus using high-temperature adhesive, and then heated in an oven while measuring the bottom echoes from the WC anvil. The results showed that the high-temperature transducer could work up to 140 °C. The transducer features low cost, easy fabricating, and high-quality signals, and so we believe it is useful for sound velocity measurement at high temperatures in cubic-anvil apparatus.

Keywords: Ultrasonic transducer, High temperature, PZT, Dual mode, Sound velocity, Cubic-anvil apparatus.

1 INTRODUCTION

Cubic-anvil apparatus in conjunction with ultrasonic technique is one of the most commonly used method for measuring the sound velocities of rocks,

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*Corresponding author: liuyonggang@vip.gyig.ac.cn minerals, and material at high pressures and high temperatures. Ultrasonic transducer is the core element of ultrasonic system and it is normally positioned on the rear surface of tungsten carbide (WC) anvil of cubic-anvil apparatus. In the actual measurement process, when the sample is heated, the heat is inevitably transmitted from the sample to the WC anvil and transducer. For example, it was found that the temperature of the rear surface of the WC anvil could gradually reach up to approximately 100 °C as the sample was heated to 1100 °C using a typical sample assembly in the so-called YJ-3000t cubic-anvil apparatus (Figure 1) installed in Institute of Geochemistry, Chinese Academy of Sciences, China, even a water-cooling circulation in the steel sleeve surrounded the anvil was provided. In that case, a high-temperature ultrasonic transducer is much desired.

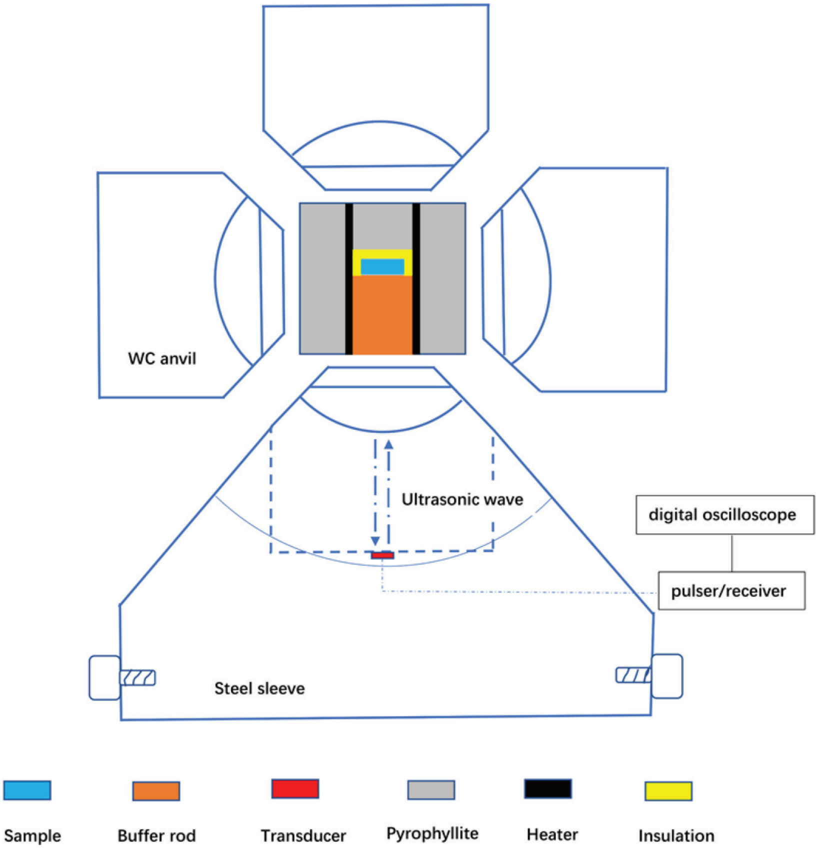


FIGURE 1
Schematic configuration of a typical assembly of ultrasonic measurement in the YJ-3000t cubic-anvil apparatus. Only the bottom WC anvil surrounded by steel sleeve is presented for clearness.

It is well known that high temperature is harmful to the ultrasonic measurements mainly because (1) high temperature weakens the piezoelectric effect, making the transducer capability of generating and receiving ultrasonic wave decrease or even lose; and (2) high temperature degenerates the coupling effect of the ultrasonic couplant; and (3) high temperature increases the attenuation of ultrasonic wave propagation in medium.

Currently, most of our experiments of sound velocity measurement in cubic-anvil apparatus would be conducted at temperatures below 1200 °C, whereas the temperatures of WC anvil would not exceed 120 °C. Thus, the objective of this paper was to develop a high-temperature ultrasonic transducer capable of operating continuously at temperature of at least 120 °C.

Although lots of high-temperature transducers are commercially available, they are mainly fabricated for non-destructive testing and/or structural health monitoring, and might not suitable to cubic-anvil apparatus, owing to the rigorous duty cycle. For example, the Olympus Company is selling ultrasonic transducers with cylindrical delay lines suitable for high-temperature operation up to 480 °C, but it only can maintain 10 seconds, followed by cooling in air at least one minute in order to avoid permanent damage to the transducer caused by overheating [1]. In addition, longer delay line certainly helps the reduction of the temperature of transducer, unfortunately, it seems unrealistic because of the narrow space limited by the inherent structural characteristics of the cubic-anvil apparatus. For the same reason, mode conversion type transducer [2] is also not applicable. Although ultrasonic transducers based on spray coated PZT film for in situ operation for temperatures up to 200 °C was introduced recently [3], it was only applied to P-wave measurements and yielded low amplitudes of signals. Undoubtedly, the ultrasonic transducer capable of simultaneously measuring P-wave and S-wave is more advanced and functional [4–11].

The most promising and simple solution is probably bonding the high-temperature piezoelectric crystals, which can simultaneously generate and receive P- and S- waves, onto the surface of WC anvil using effective bonding method. Among the candidate piezoelectric materials for geophysical high pressure research, such as lithium niobate ($LiNbO_3$) [12], lead zirconate titanate ($Pb(Zr,Ti)O_3$, known as PZT), barium titanate, and quartz, PZT piezoceramic was chosen in this study since it possess good piezoelectric properties and high electromechanical coupling factors and, more importantly, it is low cost [13].

Ultrasonic transducers with PZT type piezoelectric elements are already used in sound velocity measurements for years in high-pressure apparatus. In 1967, Van Steveninck [14] developed the concept of stacking PZT ceramic transducers to produce both P- and S-waves in solids, but the obtained ultrasonic signals were not easy to recognized. Hemsing made some modifications in the stacked system such as the building of damping material on the top of the ceramic transducers [15]. However, none of them tested the temperature effect on transducers since their sound velocity measurements were performed at room temperature. In order to measure the different waves at the same time, Kern et al. [16–18] and Scheu et al. [19] placed small P- and S-wave PZT single mode transducers side by side on the low temperature side of the piston of a cubic-anvil apparatus. Their rock samples were normally heated to only 600–700 °C, thus, no special requirements for the transducers are needed except for a cooling system. Manufacturing technology of dual mode PZT transducers with different cut angles and thickness was developed by Voleišis et al [20]. Moreover, impressed operation of transducers soldered to the steel rods and measurements were provided in a temperature range 20–160 °C. Their experiments showed good performance of developed dual mode PZT transducers. However, operation of soldering PZT transducers to the surface of WC anvil would be too highly technical to easily implement.

In this study, we fabricated an ultrasonic dual mode transducer, which was made of a sandwich of two PZT ceramic wafers, the upper one generating P-waves and the lower one generating S-waves. Then, the transducer bonded onto the WC anvil substrate using high-temperature adhesive was heated in oven while pulse echo measurements were conducted to test its high-temperature performance.

2 EXPERIMENTAL PROCEDURE

2.1 Fabrication of dual mode transducer

For the measurements of sound velocities of rocks and minerals, the transducers with 1–5 MHz central frequency are often used. According to the concept of staking PZT ceramic [14, 15] to fabricate a dual mode transducer, a P- wave and a S-wave PZT ceramic are necessary. However, while P-wave PZT ceramic wafers with 3.5 MHz frequency and silver electrode at both sides were directly commercially available (PZT-4, Daobo Ultrasonic Electronics Co., Ltd, Changzhou, China), the S-wave PZT ceramic wafer was surprisingly not conveniently available. Alternatively, we decided to manufacture the S-wave PZT ceramic wafer from a large thick P-wave PZT ceramic column (PZT-4, Anbo Machine Factory, Shaoxing, China).

According to the theoretical and experimental conclusions [20, 21], as shown in Figure 2, when the cut angle $\Theta = 90$ °, that is perpendicular to the large thick P-wave PZT ceramic column, we can obtain a S-wave PZT ceramic wafer if the cut planes were fitted with electrodes. A diamond wire cutting machine was used for this cutting procedure. Initially, the wafer thickness was cut to approximately 0.4 mm. Later, it was lapped and polished to get approximately 3.5 MHz frequency, using 9, 3, 1 μm diamond lapping film gradually. The final dimensions were $9 \times 9 \times 0.31$ mm for S-wave ceramic wafer and $6 \times 6 \times 0.64$ mm for P-wave ceramic wafer, respectively. What needs to be emphasized is that achieving uniform thickness of the ceramic wafer in the whole its area is crucial to obtain “perfect” echo signals [20]. In this study, the thickness difference of the ceramic wafer in the whole its area was less than approximately 10 μm.

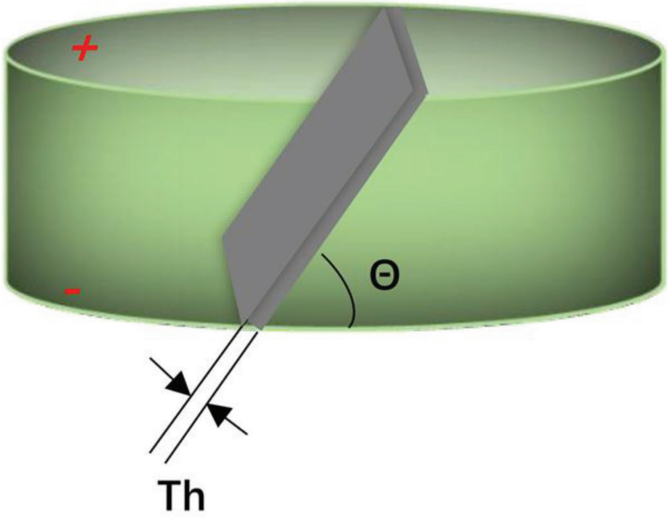


FIGURE 2
The schematic diagram of manufacture the S-wave PZT piezoceramic wafer from a large thick P-wave PZT piezoceramic column as the cut angle Θ = 90 ° and the cut planes were fitted with electrodes. Th: thickness of piezoceramic wafer.

The next step was the bonding, which is the critical challenge of most ultrasonic transducer, because ultrasonic transducer is not just of the piezoelectric material but the whole transducer consisting of piezoelectric material, electrodes, adhesive, wire, and connections. All those elements must be reliably joined together in order to get a good acoustic coupling in the high-temperature environment. Meanwhile, the ultrasonic transducer also must be reliably coupled to WC anvil.

We made the following strategies. Firstly, we decided to bond the S-wave PZT ceramic wafer onto the surface of WC anvil substrate using a high-temperature adhesive (Wellmid 2120, Wellmid Electronics Co., Ltd, Shenzhen, China). Secondly, we bonded a piece of Cu foil (9 × 9 × 0.02 mm) on the top of the S-wave ceramic wafer as electrode using silver conductive epoxy (ZB2562, Jonby New Material Technology Co., Ltd, Nanjing, China). Thirdly, we bonded the P-wave PZT ceramic wafer on the Cu foil using Wellmid 2120 again. The maximum rated temperature is 150 °C for Wellmid 2120, and is 200 °C for ZB2562, respectively. The Wellmid 2120 and ZB2562 were cured at 110 °C for one hour with post cure heating of the bond for additional two hours at the same temperature in the oven. To obtain a thin, uniform and strong bond layer, which is also crucial to obtain high-quality echo signals, a pressure of 1–3 MPa was exerted at every step of the bonding process. Finally, three 0.1 mm diameter enameled copper wires, named wire 1, 2, 3, were connected to the top electrode of the P-wave wafer, to the edge of the Cu foil electrode, and to the WC anvil using ZB2562 silver conductive epoxy, respectively.

Figure 3 gives the schematic diagram of the fabricated ultrasonic dual mode transducer attached on WC anvil. The thickness of the WC anvil substrate was 53 mm and the diameter were 70 mm. The two yellow plastic sheets were used to prevent possible electrical short. For P-wave measurements, wire 1 and wire 2 were connected to the positive electrode and negative electrode of ultrasonic pulse generator/receiver unit, respectively. For S-wave measurements, wire 2 and wire 3 were connected to the positive electrode and negative electrode of the ultrasonic pulse generator/receiver unit, respectively.

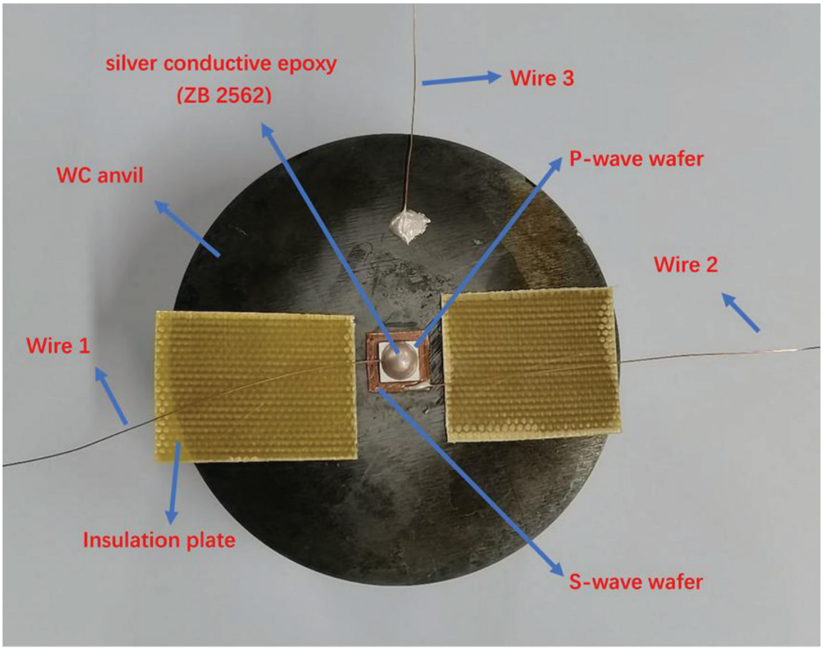


FIGURE 3
The fabricated stacked PZT ultrasonic transducer attached on WC anvil.

Allowable temperature limits of transducer components were shown in Table I.

TABLE 1
Allowable temperature limits of transducer components

Component	Allowable temperature limit
PZT wafer	360 °C (Curie temp.)
High-temperature adhesive (Wellmid 2120)	150 °C (maximum temp.)
Silver conductive epoxy (ZB2562)	200 °C (maximum temp.)
Silver sputter layer	962 °C (melting temp.)
Copper foil layer	1083 °C (melting temp.)
Copper wire	1083 °C (melting temp.)

2.2 In-oven pulse echo measurements

The high-temperature pulse echo measurements were performed in an oven. Figure 4 shows the schematic of the experimental set-up for the high-temperature pulse echo measurements. To precise determination of temperature, a K-type thermocouple was placed contacting the WC anvil, very close (~1 mm) to the transducer. Temperature was increased from 20 °C to 140 °C at 20 °C/hour rate. After reaching the target temperature, temperature was hold for 10 minutes before the echo signals were saved. The uncertainty in the temperature measurement was approximately 5 °C, the maximum operating temperature of the Wellmid 2120 adhesive is 150 °C, so the maximum heating temperature was set to 140 °C.

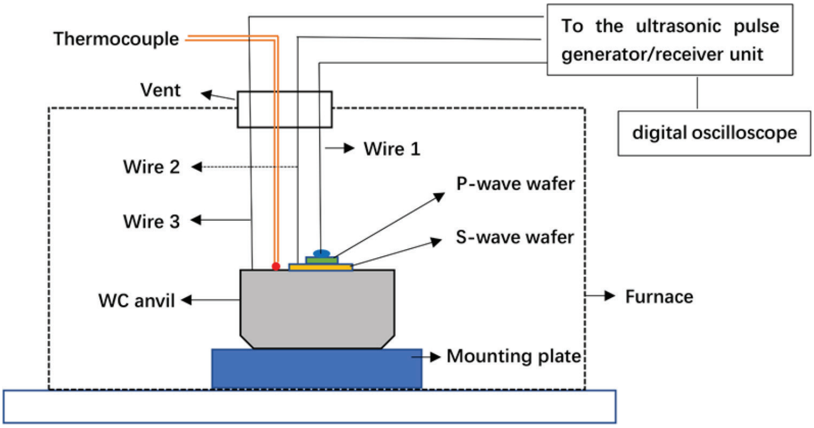
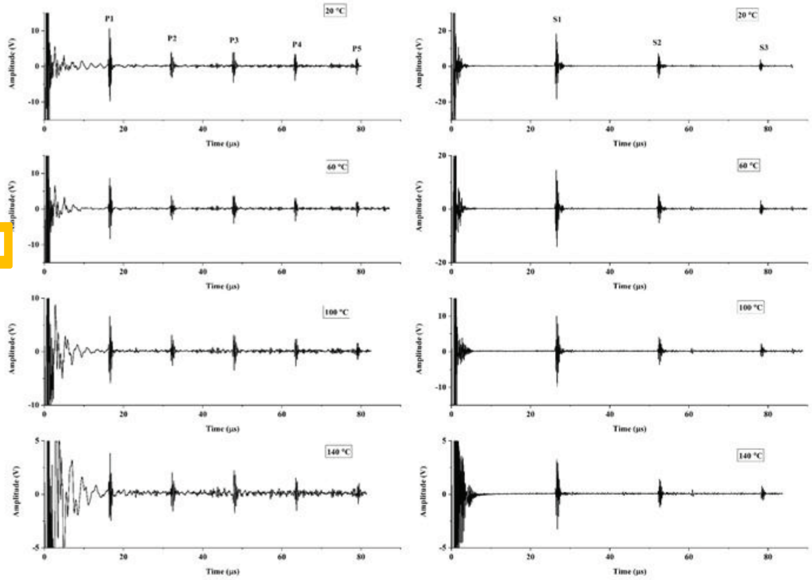


FIGURE 4
The schematic of the experimental set-up for the high-temperature pulse echo measurements.

The ultrasonic bottom echoes from the WC anvil substrate were measured using a digital oscilloscope (Tektronix DPO2024B, USA), and an ultrasonic pulse generator/receiver unit (CTS-8077PR, Guangdong Goworld Co., Ltd., Shantou, China). The damping resistance was adjusted to 50 Ω, and the data was collected with 512 times averaging to improve signal-to-noise ratio (S/N). An excitation voltage of 200 V and an excitation pulse frequency of 3.33 MHz were provided to the ultrasonic transducer in the experimental trials. All settings were not modified during the measurements.

3 RESULTS AND DISCUSSION

The waveforms of multiple echoes from the WC anvil substrate are shown in Figure 5, where (a) is the waveforms of the P-wave measurements, while (b) is the waveforms of the S-wave measurements from 20 °C to 140 °C. As shown in this figure, multiple bottom echoes were obtained clearly at each temperature.



(a) (b)

FIGURE 5
The waveforms of multiple echoes (labeled as P1-P5 and S1-S3) from the WC anvil substrate by (a) P-wave and (b) S-wave transducer.

As stated above, the P-wave and S-wave measurements were separately conducted by connecting wire 1 and 2, wire 2 and 3, respectively, to the ultrasonic pulse generator/receiver unit. Interestingly, when wire 1 was connected to the positive electrode and, wire 3 was connected to the negative electrode of the ultrasonic pulse generator/receiver unit, P-wave and S-wave echo signals were simultaneously observed in the time domain, as shown in Figure 6. It can be seen that the amplitudes of echo signals in dual mode are much lower than those in single mode.

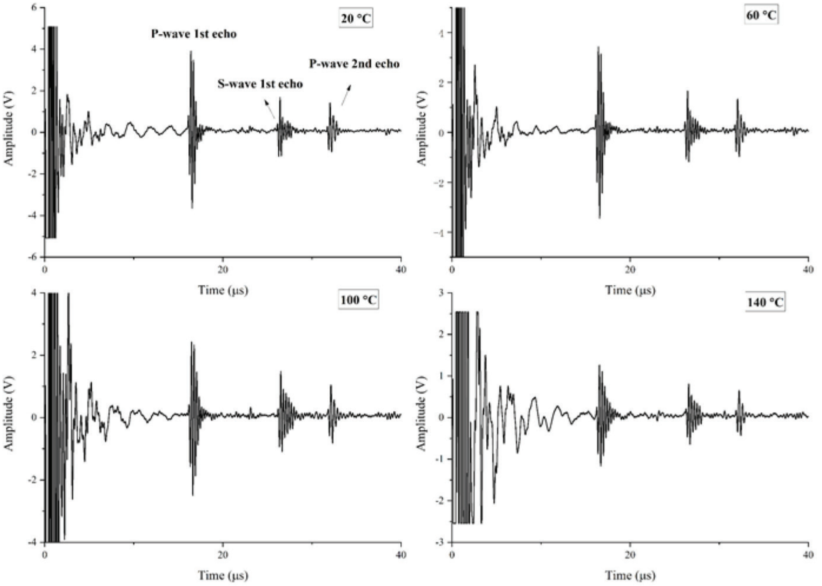


FIGURE 6
The waveforms of echoes from the WC anvil substrate by dual mode transducer.

It also can be seen that in both pure mode (Figure 5) and double mode (Figure 6), as the temperature rises, the reverberation signals grow while the echo signals from the WC anvil decrease. This can be explained by the deterioration of the acoustical contact between the ultrasonic transducer and the WC anvil with the increased temperature.

The waveforms of the first echo are compared in Figure 7, where (a) is for P-wave and (b) is for S-wave measurement. The echo positions in the time range delayed with increased temperature. This is explained by the fact that the velocity and the thermal expansion of the WC substrate changed with temperature. It is worth pointing out that those echo signals have high amplitudes, S/N ratios and narrow pulses, which are very useful for the sound velocity measurements.

As expected, the peak-to-peak amplitudes of the first echo consistently decreased in the heating process (Figure 8). The rate of decrease of the peak-to-peak amplitudes is -0.12V/°C in P-wave, and -0.26V/°C in S-wave. In the high-temperature study by Voleišis et al. [11], where the PZT ultrasonic dual

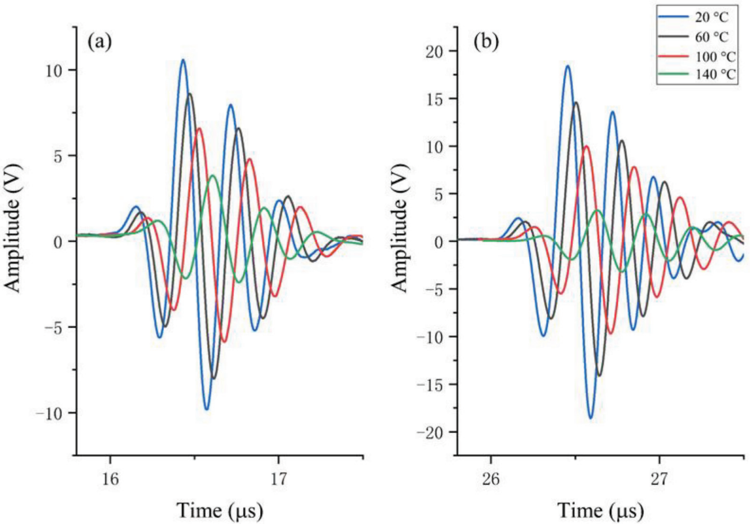


FIGURE 7
The waveforms of the first echo from the WC anvil substrate by (a) P-wave and (b) S-wave transducer.

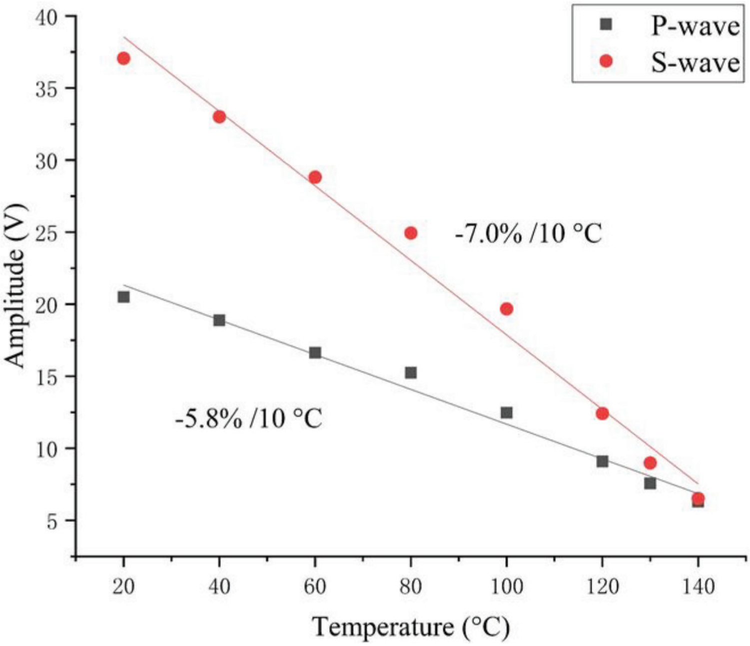


FIGURE 8
The peak-to-peak amplitudes of the first echo decrease as the temperature grows.

mode transducer was soldered to the steel rod, the rate of decrease of the amplitudes of the reflected signals in P-wave and S-wave were almost the same. Therefore, it indicated that the larger reduction of amplitude in S-wave signals is because the transmission of S-wave is more susceptible to the deterioration of the acoustical contact (adhesive) than that of P-wave.

Figure 9 shows the temperature effect on frequency spectrum of the first echo. A slight decrease in the frequency is observed as the temperature grows from 20 to 140 °C, that is 0.3 MHz for P-wave and 0.2 MHz for S-wave signal. Similar phenomena was observed in literature [3, 22]. In addition, no influence of temperature on the general shape of the frequency spectra of the received signals and, the temperature only affects the amplitude of the frequency spectra, which reduces with the increasing temperature.

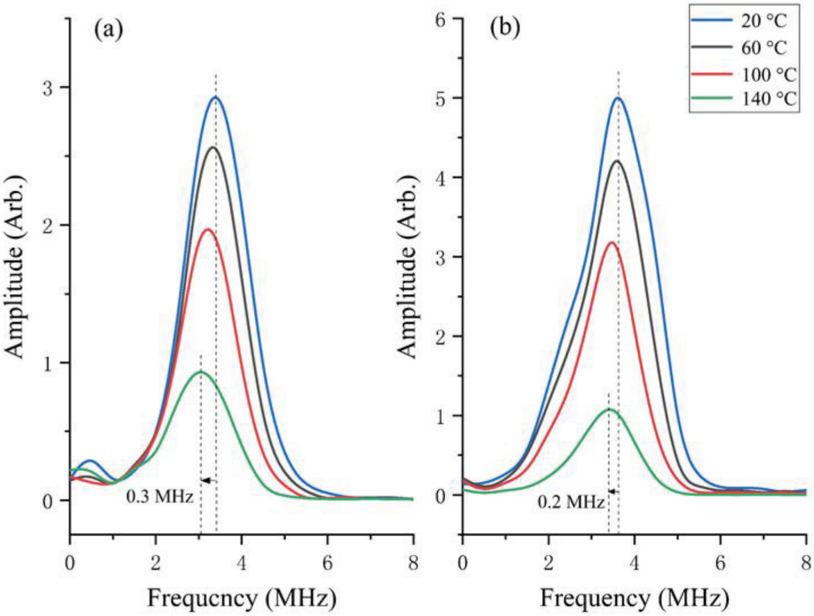


FIGURE 9
The temperature effect on frequency spectrum of the first echo from (a) P-wave and (b) S-wave measurements.

After cooling, the amplitudes of the first echo were found to decrease by approximately 2.5% for P-wave and 9.6% for S-wave, but the S/N ratios were still high. Normally, experienced a temperature of 140 °C would not influence the properties of the PZT type piezoelectric element. For example, Voleišis et al. soldered the PZT piezoelectric element to the steel rods and heated them to 160 °C [20]. They found the signal fully restored after cooling. Thus, the main reason of the decreased amplitudes of the echo seems to be due to the degradation of the adhesive caused by high temperature and/or the slightly debonding of the PZT ceramic wafer caused by thermal stress. This could be a common problem of the ultrasonic transducer coupled on WC anvil using adhesive. Nonetheless, it is still preferred because of its merits, which are low cost, easy fabricating, and yield high-quality signals. On the other hand, other cooling strategies lowering the transducer temperature during heating [23] help to extend the longevity of the transducer. For example, in our previous study, a tunnel excavated in the steel sleeve surrounded the WC anvil was used to blow focused cooling air in a nozzle directly to the transducer to prevent overheating.

With regard to the high-temperature adhesive, we in fact also tested several other adhesives, such as Epotek-353ND, IX28214-2, DP760, KaiSiMi 540, KMD-398, and we found that lots of high-temperature adhesives are intended for rough structurally bonding usage, not acoustically. The adhesives with large particles in their composition easily resulted in cracked piezoelectric material [24] and distorted echo signals when used. Seeking a better candidate high-temperature adhesive, which means better performance at higher temperature with extend duration, is one of the solutions to improve the ultrasonic dual mode transducer.

4 CONCLUSIONS

Using two stacked PZT piezoceramic wafers, the upper one generating P-waves and the lower one generating S-waves, we fabricated a high-temperature ultrasonic dual mode transducer bonded on a WC anvil substrate and investigated its high-temperature performance. The obtained echo signals have high amplitudes, S/N ratios and narrow pulses, which indicated the ultrasonic transducer has a capability to work to 140 °C. Despite the reduction in amplitudes of the echo signals after cooling, the transducer is still preferred owing to its low cost, easy fabricating, and yield high-quality signals. We believe the fabricated dual mode transducer is useful for sound velocity measurements at high temperatures in cubic-anvil apparatus.

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