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Citation: Review of Scientific Instruments **82**, 086108 (2011);
View online: https://doi.org/10.1063/1.3625267
View Table of Contents: http://aip.scitation.org/toc/rsi/82/8
Published by the American Institute of Physics

Note: Measurement method for sound velocity of melts in large volume press and its application to liquid sodium up to 2.0 GPa

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(Received 17 March 2011; accepted 25 July 2011; published online 19 August 2011)

Based on large volume press and conventional pulse-echo ultrasonic technique, we have overcome the difficulty in determining the length of liquid specimen under high pressure, and the sound velocity in liquid Na has been measured up to 2 GPa. The P-V data deduced by our sound velocity results through equation of state is in an excellent agreement with previous data directly determined by piezometer method. This new experimental technique is convenient and ready for use, being expected to advance investigation on thermodynamic properties of liquid metals and other melts under high pressure. [doi:10.1063/1.3625267]

As a fundamental and physical property of liquids, velocity of sound wave in liquids is very important and useful. Many thermophysical parameters may be derived from sound velocity, such as adiabatic bulk modulus, ratio of isobaric to isochoric heat capacities, Grüneisen gamma and so on.[1] However, to date, there is little direct experimental information on macroscopic thermodynamic properties of liquids or melts under high pressure.

At present, diamond anvils cell (DAC) (Refs. 2 and 3) and large volume press (LVP) (Ref. 4) are two kinds of mainstream apparatus used to investigate elastic property of materials under high temperature and high pressure (HTHP), but these research activities focus prevalently on solid specimens for their application to mineralogy of the Earth's interior.[4] The main reason for few studies on liquids is the enormous difficulty exists in measuring the thickness of liquids under high pressure. Normal container for liquids will deform seriously when suffering deviatoric stress, which make it impossible to get the right length of liquids and high quality ultrasonic signals. Most recently, Decremps*et al*.[5] proposed an innovative technique combining picosecond acoustics in DAC to measure the sound velocity of liquid mercury under HTHP, but the urgent need to explore measurement method for liquid sample under LVP still exists.

In this note, we shall propose a concise and straightforward method to investigate sound velocity of liquid sample in a LVP apparatus based on pulse-echo ultrasonic technique. As the first sample to apply this new method, liquid sodium is chosen because direct experimental P–V data[6] at pressure up to 2 GPa is available for comparison with this study.

The experiment is conducted with a LVP (YJ-3000 ton), which can generate pressure from 0.5 GPa up to 6 GPa. The apparatus is a DIA-type press that is consisted of six anvils[7]. The setup for ultrasonic measurements on this LVP mainly consists of ultrasonic pulser/receiver unit (5077PR, Panametric-NDT), digital oscilloscope (TDS784A, Tektronix) and one ultrasonic transducer with 10 MHz center frequency.[8] A newly developed broadband spectroscopy method on this system[9] is also used for data processing.

Figure 1 shows the cross section of the cell assembly in a pyrophyllite hexahedron for ultrasonic measurements under HTHP. The Tungsten Carbide (WC) anvil and Al_2O_3 buffer (cylinder of 99.7% alumina ceramics) form the acoustic channel for ultrasonic wave transmission between the transducer and liquid specimen. The crucial idea to restrict the thickness of liquid sample is the use of cylindrical WC with a groove as an acoustic reflector. This groove is ultra-precisely machined by slow wire cutting method with a precision less than 1 μm, and full filled with metal sodium as specimen under test. According to deformation theory, hydrostatic stress will not contribute to yielding. As seen in Fig. 1, the WC reflector is surrounded by Teflon tube, thereby providing a pseudo-hydrostatic pressure environment and protecting it from plastic deforming at high pressure. The Ni foil filled the gap between WC reflector and Teflon tube is used to avoid possible chemical reaction and improve sealing effect on melts under high pressure.

Obviously, the thickness of liquid sodium under HTHP can be determined by the depth of the groove (1.5 mm depth, 3.0 mm width), and the dimension change of WC reflector under pressure can be estimated by well known Stress-Strain relationship in its elastic behavior.[10] According to definition of bulk modulus, if pressure is small relative to its bulk modulus, there is a simple formula to estimate the volume change with the pressure,

$$\frac{\Delta V}{V_0} \approx \Delta P/3K_0.$$

As the bulk modulus K_0 of WC is approximately greater than 400, when the pressure ΔP reaches 2 GPa, the rela-

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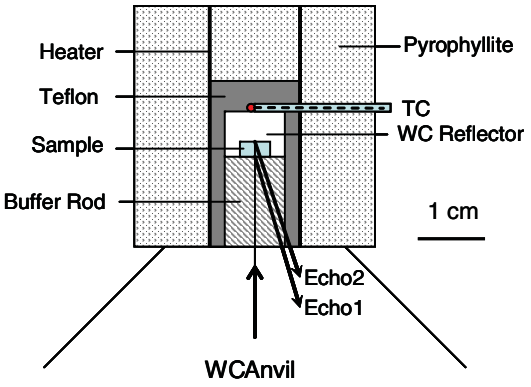

FIG. 1. (Color online) Cross section of sample assembly.

tive volume change equals to 0.16%. Thermal expansion coefficient α of WC is $\sim 5\times 10^{-6}$, so when temperature reach 500 K, the volume change becomes

$$\frac{\Delta V}{V_0} = \alpha \times \Delta T = 5 \times 10^{-6} \times (500 - 293) = 0.1\%.$$

These two volume changes will offset each other, so finally the volume change is 0.06%. If we change the volume into linear thickness change, we will find

$$\frac{\Delta L}{L_0} \approx \frac{\Delta V}{V_0} \times \frac{1}{3} = 0.06\% \times \frac{1}{3} = 0.02\%.$$

It is a very small correction in our experimental conditions, with no sense for consideration if comparing to other error sources such as ultrasonic system and so on, so we will not consider the negligible correction of pressure effect on the WC.

Upon recovery from each high pressure experiment, careful examination of the WC reflector and Al_2O_3 buffer were performed to make sure their surfaces have no visible cracks, and no measurable changes in dimensions have been observed by digital micrometer with 1 μm resolution.

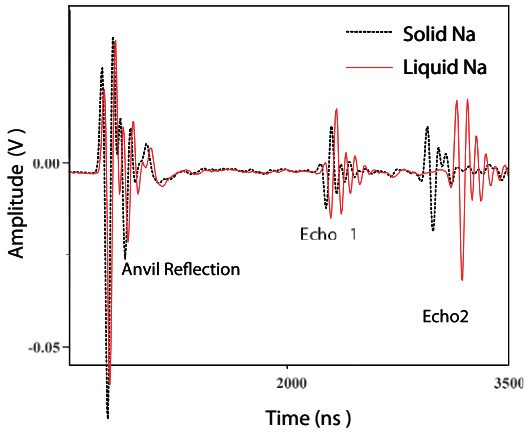

FIG. 2. (Color online) Received signals before and after Na melting at 0.7 GPa and 425 K. Dashed black waveforms for echoes before melting; solid red waveforms for echoes after melting.

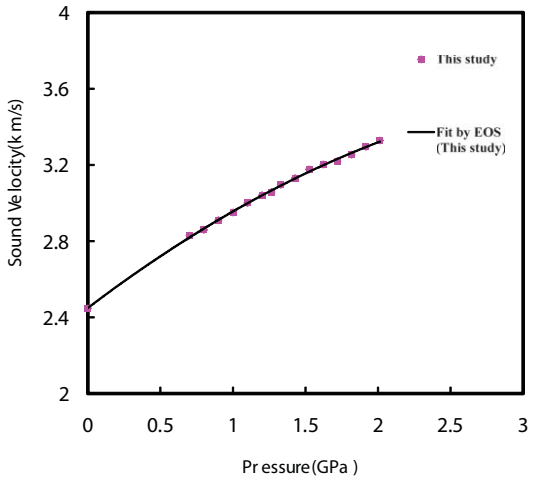

FIG. 3. (Color online) Sound velocities in liquid Na as a function of pressure at 493 K. Pink squares for this study up to 2 GPa.

In the present experiment, we first raise the pressure to 0.7 GPa, and then at the fixed press load, temperature are slowly heated to 493 K measured by NiCr-NiSi thermal couple (TC), and finally increase the pressure to about 2.0 GPa at constant temperature 493 K. We are able to observe clear echo signals of ultrasonic wave for all P, T conditions. Fig. 2 shows an example of such signals observed before and after melting of Na (425 K, 0.7 GPa). The dashed and solid waveforms reflected by interface of liquid Na and WC reflector are the echoes before and after melting respectively. This transient dynamic process of melting process could be observed clearly in oscilloscope, so it is extremely helpful to decide the melting point of Na at specific pressure. We have recalibrated pressure scale for this new assembly at several different oil pressure of hydraulic system from 65 MPa to 250 MPa. By reading responding melting temperature point of Na at these pressure points, we can get the right pressure in the sample cell based on the melting curve of Na under pressure.[11] It is found the influence of our new assembly on the pressure calibration is less than 10% from previous pressure scale.

Echoes of liquid Na from the interfaces between buffer rod and sample, sample and WC reflector can be evidently recorded with high quality, so the round-trip travel times of ultrasonic wave passing through the liquid Na are precisely determined. The travel time is determined with an error of less than 2 ns corresponding to an uncertainty on the velocity lower than 0.5% even the consideration of length change of WC reflector included.

The pink squares in Fig. 3 show the sound velocity in liquid Na at 493 K with pressure increase up to 2.0 GPa. The

TABLE I. Experimental data[13] of bulk thermal expansion α(0, T) (10^{-4} K^{-1}), densityρ(g cm^{-3}), specific heat capacity Cp (10^{-3} kJ g^{-1} K^{-1}), and molar mass M at temperature of 493 K and zero pressure for calculation. Values of parameters B_T (0, T) (GPa), $B_{T'}$ (0, T) and Z (a pressure- and temperature- independent parameter) are derived by fitting the sound velocity data through the KD EOS[12].

T = 493 K	M	α(0,T)	Cp(0,T)	ρ(0,T)	B_T (0, T)	$B_{T'}$ (0, T)	Z
Liquid Na	22.99	2.52	1.31	0.903	4.73	4.0	0.0715

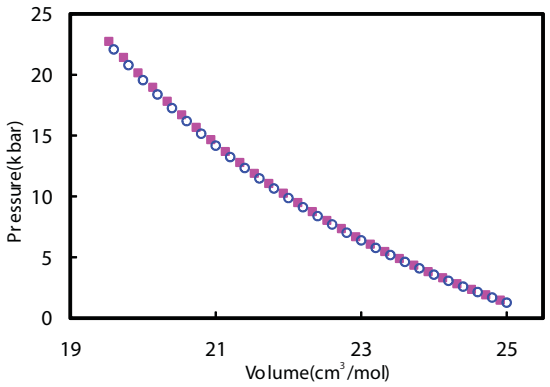

FIG. 4. (Color online) P–V relation of liquid Na at 493 K. Circles for direct experimental P–V data (Ref. 6) and Pink squares for results determined by fitting sound velocity data of this study through the KD EOS (Ref. 12).

method for fitting the sound velocity data with the Kumari-Dass (KD) EOS has been fully described by Kuchhal.[12] Hence, the best-fitted values of isothermal bulk modulus B_T (0, T), and first pressure derivative of the bulk modulus $B'_T(0, T)$ at temperature of 493 K can be obtained and listed in Table I. Once knowing B_T (0, T), $B'_T(0, T)$, the volume data as a function of pressure in liquid Na at 493 K can be calculated out.[12] Fig. 4 shows that the P–V relation determined by this ultrasonic study which is extremely consistent with the direct P–V data measured by Makarenko *et al.*[6] at pressure up to 2 GPa. It demonstrates that our technique is an innovative improvement to existing ultrasonic measurement for liquids or melts under HTHP.

In conclusion, based on LVP and conventional pulse-echoing ultrasonic technique, the sound velocity of liquid Na at a temperature of 493 K has been measured under pressure up to 2 GPa, and the result agrees with the direct P–V data. We successfully overcome the difficulty in measuring the length of specimen in liquid state under high pressure, and extend the pressure limit for liquids into GPas. This experimental method is being expected to advance investigation on thermodynamic properties of liquid metals and other melts in extreme HTHP.

The authors acknowledge support from the National Natural Science Foundation of China (NNSFC) under contract 40974052, the Knowledge Innovation Program of the Chinese Academy of Sciences under contract KZCX2-YW-Q08–3-4, as well as the Mutual Foundation of Huge Scientific Equipment of National Natural Science Foundation of China and Chinese Academy of Science under contract 10979053. We thank Professor Chengming Zhu for technical support.

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