

# A laboratory procedure of measuring ultrasonic properties of CO2 saturated fluids

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#### Summary

CO<sub>2</sub> storage opportunities into geological structures aiming to reduce their emissions into atmosphere have led to many different studies related to seismic monitoring by qualitatively tracking the movement of CO<sub>2</sub> plume in reservoir and quantitatively measure the amount of CO<sub>2</sub> in place with other fluids (oil and/or brine). Recent ultrasonic measurements for CO<sub>2</sub> saturated samples shows the effects of CO<sub>2</sub>'s vapor-liquid-supercritical phase transitions on the overall rock seismic response. We have also recently observed influence of the rate of cooling and heating on CO<sub>2</sub> condensation and evaporation in a saturated synthetic rock sample. Acoustic properties of endmember CO<sub>2</sub> and CO<sub>2</sub> saturated fluids at various temperatures and pressures, therefore, would be useful studies in order to develop theoretical modeling of these observed responses using theories that will include the CO<sub>2</sub> properties more fully. An adapted version of the traditional double reflectors pulse echo method is built to make the measurements of speed of sound in fluids affected by temperature and pressure in order to aid in the interpretation of field seismic observations. As of calibration purposes the speeds of sound of pure water are measured in a temperature range of 10° C to 70° C and at pressures up to 60 MPa. The uncertainty of these measurements is around 0.1% comparing with the formulation of International Association for the Properties of Water and Steam (IAPWS-95). Further measurements in water with 10<sup>5</sup> ppm salinity show the speeds of sound lie between 0.02% and 0.04% within Wilson's empirical formulations of sound speeds in sea water. However the experimental set-up is now in the process of being modified to take measurements in CO<sub>2</sub> saturated fluids at temperatures and pressures up to 100° C and 80 MPa, respectively, using the pulse-echo double reflectors method and later the results will be compared with observations from CO<sub>2</sub> saturated rock samples.

#### Introduction

The excess of greenhouse gases (e.g., carbon dioxide  $(CO_2)$ ) in atmosphere is the most addressed cause of anthropogenic warming on the earth. The possibility of capturing and storing  $CO_2$  into subsurface geologic structures provides hope that  $CO_2$  emissions into atmosphere can be reduced.

Recent ultrasonic measurements using a pulse-transmission method for fully CO2 saturated core samples from the Weyburn-Midale carbonates over a variety of temperatures and pressures representative of subsurface conditions show the change in seismic velocities that indicates CO2's vapor-liquid-supercritical phase transitions (e.g., Njiekak et al., 2013). In one of our previous studies we observed the influence of the rate of cooling and heating on CO<sub>2</sub> condensation and evaporation in a

saturated synthetic rock sample (Kofman et al., 2013). In order to aid the interpretation of this observation we have built an adapted version of the traditional double reflectors pulse echo method to measure the ultrasonic properties of endmember  $CO_2$  and  $CO_2$  saturated fluids at various temperatures and pressures. Here we discuss the advantages of the method and some results with pure water and brine that also reflect the accuracy of the technique.

# **Theory and Method**

Figure 1 shows the experimental technique that includes single transmitting-receiving 1 MHz piezoelectric transducers (PZT) placed between two stainless steel reflectors separated using spacers of unequal length. This reflectors set-up is placed inside a pressure vessel filled with the fluid which properties would be measured. The newly built experimental set-up is incorporated with a chiller system and has the ability to control the pressure of the fluid (confining pressure) up to 80 MPa. Temperature of the fluid can be regulated in the range from ~ 0° C to ~ 110° C. One of the advantages of this method includes measurement of the flight time of the signal by just considering the time difference between the onsets of first echoes from the two reflectors (Muringer et al., 1985; Ball and Trusler, 2001; and Meiera and Kabelac, 2006). However, another advantage is the ability to tune the time interval and the amplitude of the second burst such that the first echo from the second signals exactly cancels the second echo from the first signal. This time interval then equals the time difference between the arrival of the first and second echoes of the first signal received at the transducer. The cancellation of the signals is highly sensitive to the time interval adjustment and therefore can provide a resolution of less than 5 ppm (Meiera and Kabelac, 2006). Flight times of the pulse are observed at a high sampling rate using a digital oscilloscope. Linear thermal expansivity and isothermal compressibility of stainless steel are also used to calculate the transit time difference corrections with pressure and temperature variations. Plane waves deviate during their propagation in fluid so the diffraction corrections are also considered in speed calculation.

The speed of sound can be expressed as,

$$u = 2\Delta L / \Delta t$$

where  $\Delta L$  = Path length difference,  $(L_2 - L_1)$ ;  $\Delta t$  = Transit time difference,  $(t_2 - t_1)$ ;

The variation of the path length difference with temperature (T) and pressure (P) can be obtained from the following formula,

$$\Delta L(T,P) = \Delta L(T_0, P_0) \left\{ 1 + \langle \alpha \rangle (T - T_0) - \frac{1}{3} \langle \beta \rangle (P - P_0) \right\}$$

where  $\langle \alpha \rangle$  = Mean value of the linear thermal expansivity ( $\alpha$ ) of the stainless steel at pressure  $P_0$  over the temperature interval [ $T_0$ , T], and

 $\langle \beta \rangle$  = Mean value of the isothermal compressibility ( $\beta$ ) of the stainless steel at temperature *T* over the pressure interval [ $P_0$ , P].

# Examples

Speeds of sound of pure water are measured in the temperature range from 10° C to 70° C and at pressures up to 60 MPa. The waveforms (at 50° C) and the speeds with pressure variations (at 30° C and 50° C) are shown in Figures 2 and 3. The uncertainty of these measurements is around 0.1% comparing with the formulation of International Association for the Properties of Water and Steam (IAPWS-95) (Lin and Trusler, 2012). The PZT was later covered with silicone conformal coating to prevent the conductive behavior in the presence of salt water. Similar measurements were then conducted in water with 10<sup>5</sup> ppm salinity. Figure 4 shows the speeds of sound in the brine lie between 0.02% and 0.04% within speeds from Wilson's empirical formulations for sea water.

# Conclusions

In reality  $CO_2$  would stay with brine and/or oil in the geological formation. Therefore we have tried to implement a well accurate double reflectors pulse echo method to measure the ultrasonic properties of endmember  $CO_2$  and  $CO_2$  saturated fluids. The demonstrations with pure water and brine show their high accuracy of the technique. We are in the process for further measurements with  $CO_2$ .

### Acknowledgements

We would like to thank Carbon Management Canada (CMC-NCE) and the Natural Sciences and Engineering Research Council of Canada (NSERC) for their generous support.

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Figure 1: Image of ultrasonic pulse echo cell. 1. Stainless steel reflector, 2. PZT transducer, 3. PZT holdertransducer sandwiched between two stainless steel disc (Delrin used in between for insulating PZT with steel), and 4. Three rods (spacers).



Figure 2: Waveforms of the echoes of the double refelctors method at pressures from 1 to 60 MPa and temperature of 50° C. The differences of onsets of first two echoes provide the transit time.



Figure 3: Speeds of sound in pure water at temperatures of 30° C and 50° C as a function of pressure are plotted along data from Lin and Trusler, 2012. The differences among them are due to the small variation in tempearure and pressure (e.g., they measured at exact 50° C whereas our measurements were done at 50.5° C and also had pressure difference of ~0.1 MPa).



Figure 4: Speeds of sound in water with 10<sup>5</sup> ppm salinity at 30° C as a function of pressure. The data is plotted along with speeds from Wilson's empirical formula for sea water (Wilson, 1960).