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Laboratory measurements of seismic velocities of CO_2/brine mixtures at elevated temperatures up to 70°C and pressures up to 38 MPa.

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SUMMARY

Phase equilibrium of CO_2-brine fluids is important to studies related to CO_2 sequestration in deep saline aquifers and CO_2 enhanced oil recovery. Furthermore, the study of the seismic properties of CO_2 saturated and not saturated brines as pore fluids helps to understand their influence on the 4D seismic properties of rocks.
In this work we reported results of measurements of the acoustic velocities in brines with dissolved CO_2. We investigate the effects of pressure (from 2 MPa to 38 MPa), temperature (from 30 ˚C to 70 ˚C), and salinity (0, 10,000ppm, 20,000ppm, 40,000ppm, 100,000ppm) of KCl-NaCl brines on ultrasonic velocities. We also study the time lapse effect of CO_2 dissolution into the brine.
It has been found that CO_2 dissolution in brine has significantly changed acoustic properties of brine.

Key words: Supercritical CO_2, brine, saturation, P-wave velocities, rock physics.
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INTRODUCTION

Understanding the acoustic velocities of water, brines and CO_2 is essential in order to understand the seismic properties of hydrocarbon reservoirs, optimization of enhanced oil recovery (EOR) and especially for monitoring of the geological storage (CCS). Understanding CO_2 properties has great importance since the climate changes, high demands reducing CO_2 levels and finding alternative options of storing excess CO_2 in geological formations.

Batzle and Wang (1992) summarized all available data in that time on ultrasonic velocities in brines and produced and empirical relations between P-wave velocities, pressures, temperatures and salinities in brines. This empirical relationship is widely used to in modelling. Recently Han et .al (2009) reported experimental result on researches the acoustic properties of pure CO_2 up to 100 MPa and 200°C. Sun and Han (2009) reported first results on acoustic measurements of mixtures of CO_2 in distilled water. However, despite numerous data sets of ultrasonic velocities and densities in water and brines at elevated temperatures and pressures have been collected throughout the years, limited information for mixtures of brines and carbon dioxide is available in the open literature.

To fill this gap in this paper we are going to report the results of measurements of ultrasonic velocities as a function of pressure, temperature and salinity.

EXPERIMENT SETUP

Figure 1 shows the setup of an experiment specifically designed in order to measure P-wave velocities of fluids, saturated and not saturated with CO_2. The setup consists of cylindrical stainless steel pressure cell, which contains the liquid. Two ultrasonic P-wave-type transducers (V103-RM, Panametrics) are connected to both sides of the pressure cell. Central frequency of these transducers is 1 MHz. The transducers are connected to a pulser/receiver (5077PR, OLYMPUS) that produces a square wave to excite P wave and amplified the signal, which is monitored by digital oscilloscope (TDS 3034C, Tektronix). P-wave velocities are calculated by measuring the arrival times of the waves. Flexible heaters with controlled temperature are wrapped around the pressure cell, when two thermometers are used on the upper and the bottom sides of the pressure cell in order to monitor the temperature. The CO_2 is being stored in a big gas cylinder with an external pressure gauge. The CO_2 flows from the cylinder to an injection pump (Teledyne ISCO D-series pump) that controls the pressure of the gas/liquid inside the measurement cell. The water/brine flows form the tube to the pressure cell through another injection pump (.LC-20A, Shimadzu Ltd.) that can also control the pressure in the cell.

Experimental apparatus was calibrated and different temperatures and pressures using brines with different salinities. The results of calibration were compared with ultrasonic velocities calculated using Batzle and Wang (1992) empirical relationship.

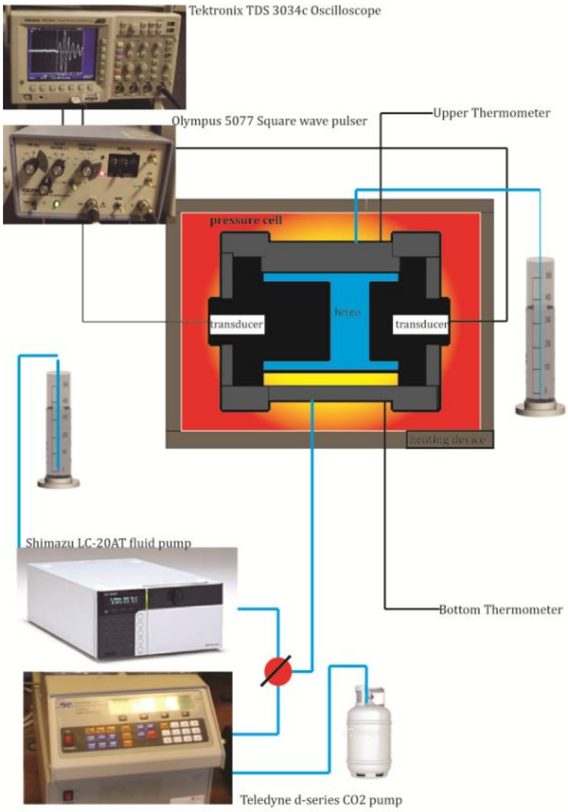

Fig 1. The Experiment Setup.

Fig 2. The pressure cell taken apart.

METHOD

We filled the cylindrical pressure cell with the brines and measured P-wave arrival times when gradually increasing pressures from 2 to 38 MPa for the brines and from 6 to 34 MPa for brines saturated with CO_2. Measurements for CO_2/brine mixtures can be reliable only if pressure in the system is increasing, a sudden drop in pressure may lead to generation of gas bubbles and thus increasing of scattering of ultrasonic waves. The temperature was varied between 30°C to 70°C. We used brines with the folowing composition: 50% of KCl and 50% of NaCl and salinity of 0, 10,000ppm, 25,000ppm, 40,000ppm and 100,000ppm. After measuring the velocities of pure brine 20% of the cell volume was released and filled with CO_2. To check that CO_2 is dissolved into brine and the system came to a thermodynamic equilibrium, we monitored P-wave velocities versus time.

RESULTS

It takes some time for CO_2 to be dissolved in brine. To estimate the time when the system (CO_2-brine) comes to thermodynamic equilibrium we measured time dependence of velocity. Results are shown in the Fig 3. At temperatures less than 50˚C velocity of CO_2/brine mixtures is gradually increasing with dissolution of CO_2, however for higher velocities we can see that velocity is decreasing while CO_2 is mixing.

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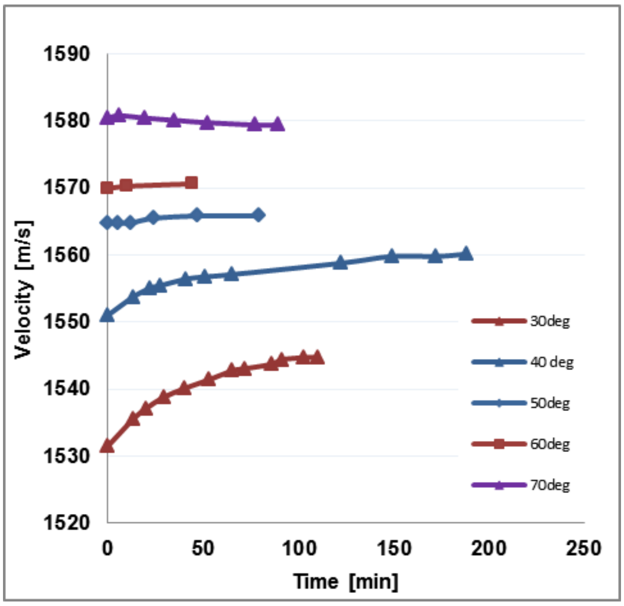

Figure 3. P-wave velocities as a function of time during the CO_2 saturation. The salinity is 10 000ppm, the pressure is 6MPa and each line represents a different temperature.

Figure 4 shows the velocities' time dependence during the saturation process at temperature 40˚C using different brine salinities.

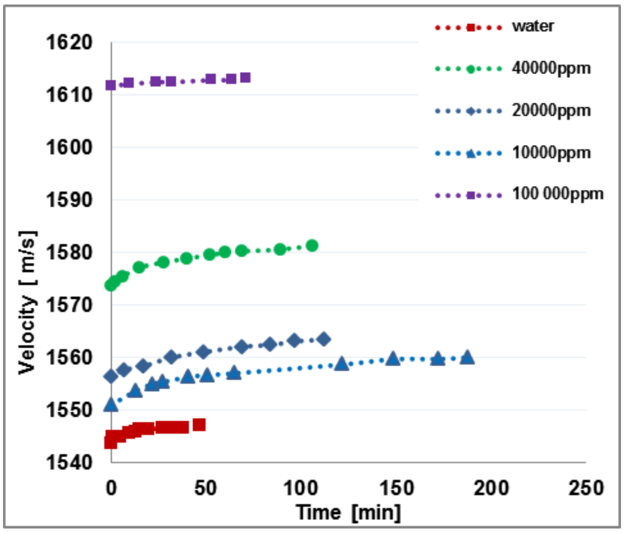

Figure 4. P-wave velocities time dependence during the CO_2 saturation in brine with different salinity. The temperature is 40 ˚C and the pressure is 6MPa.

From the chart above we see that first of all dissolution of CO_2 has significant effects on the velocity of brine.

Figure 5 illustrates how velocities of CO2/brine mixtures depend on pressure in the system.

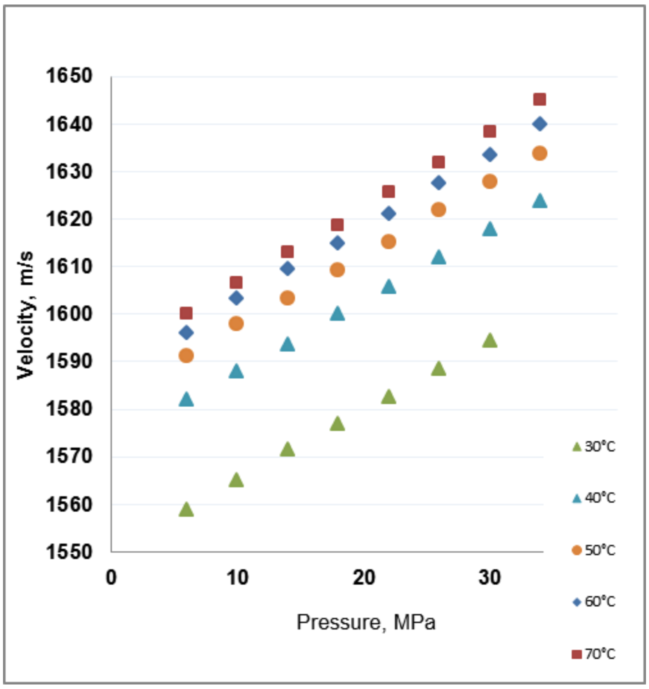

Figure 5. P-wave velocities of CO_2 brine mixtures at different temperatures. Salinity is 40,000ppm.

Dependence of P-wave velocity of salinity is presented in Figure 6. Analogously we see that the dependence of the velocity on pressure is approximately linear.

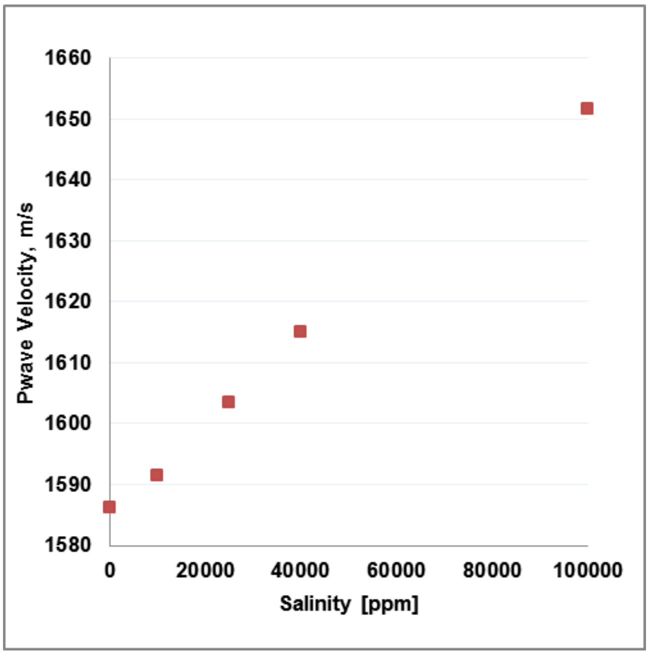

Figure 6. P-wave velocities of CO_2/brine mixtures vs. salinity. The pressure is 22 MPa and the temperature is 50˚C.

Figure 7 shows an example of velocity dependence of brine CO_2 mixture on temperature.

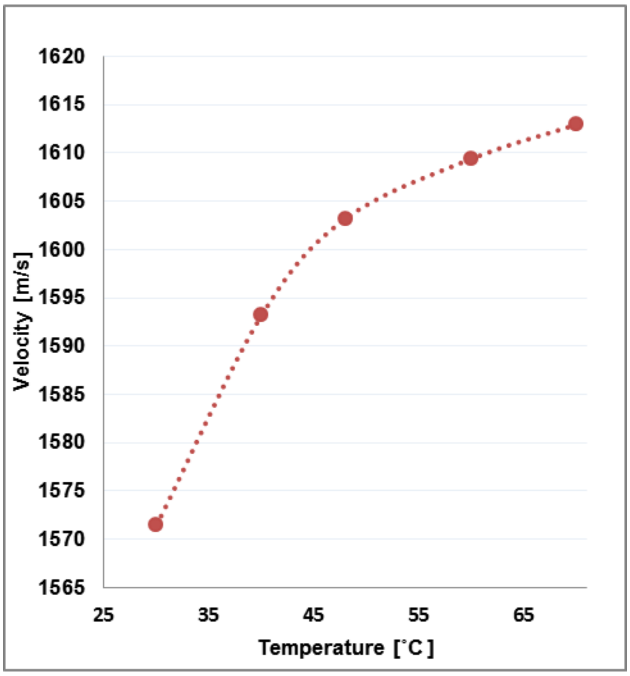

Figure 7. P-wave velocities vs. temperature. The salinity is 40,000ppm. The solution is saturated with CO_2.

The shape of this dependence is similar to results presented by Sun for CO2/water mixture (2009).

Based on results of experiments we have created an empirical formula for dependence of ultrasonic velocity of brines in which CO_2 is dissolved.

We derived a four-dimensional formula by analysing the one-dimensional curves, when modifying one of the variables and holding the rest of them constant. We tried to fit the data to a formula, which structure is:

$$V_{mixture} = a_1 + a_2 \cdot P + a_3 \cdot S + a_4 \cdot P \cdot S + a_5 \cdot T + a_6 \cdot P \cdot T + a_7 \cdot S \cdot T + a_8 \cdot P \cdot S \cdot T + a_9 \cdot T^2 + a_{10} \cdot P \cdot T^2 + a_{11} \cdot S \cdot T^2 + a_{12} \cdot P \cdot S \cdot T^2 + a_{13} \cdot T^3 + a_{14} \cdot S \cdot T^3 + a_{15} \cdot P \cdot T^3 + a_{16} \cdot P \cdot S \cdot T^3$$

Using Matlab we numerically developed preliminary empirical model to calculate the velocities of CO_2/brine mixtures:
The coefficients of the formula are shown in the Table I.

Table 1. Numerically calculated coefficients of the formula.

a_1	*1.512E+03 (m/s)*
a_2	*-1.072E-01*
a_3	*1.477E-04*
a_4	*1.766E-05*
a_5	*1.547E-02*
a_6	*8.369E-02*
a_7	*1.085E-05*
a_8	*-8.949E-07*
a_9	*1.418E-02*
a_{10}	*-1.279E-03*
a_{11}	*1.544E-07*
a_{12}	*1.233E-08*
a_{13}	*-2.562E-04*
a_{14}	*-3.226E-09*
a_{15}	*6.243E-06*
a_{16}	*-4.808E-11*

Where, **V** is in m/s, **P** is in MPa, **S** in ppm and **T** in ˚C.
This empirical formula has been obtained without considering any physical laws. Further research will include a consideration of the first principles and refining the relationship.

Bulk modulus (***K$_f$***) of the CO_2/brine can be obtained from

$$K_f = \rho * V^2$$

where, ρ is density of brine/CO_2 mixture,

Density of CO_2/brine can be estimated using empirical relationships summarized by Inglaer (2011).

CONCLUSIONS

It was experimentally founded that acoustic velocity of brine with CO_2 is different from velocities of pure brine. Empirical formula for brine/CO_2 solution in thermodynamical equilibrium was created.

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