

CO₂ velocity measurement and models for temperatures up to 200 °C and pressures up to 100 MPa

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ABSTRACT

Studies on how the velocity of CO₂ is affected by temperature and pressure are important for understanding seismic properties of fluid and rock systems with a CO₂ component. We carried out laboratory experiments to investigate velocity of CO₂ in temperatures ranging from –10 °C to 200 °C and pressures ranging from 7 MPa to 100 MPa, in which CO₂ is in a liquid phase. The results show that under the above conditions, in general, the velocity of CO₂ increases as pressure increases and temperature decreases. Near the critical point (31.1 °C and 7.38 MPa), the velocity of CO₂ reaches a minimum and has a complicated behavior with temperature and pressure conditions due to the CO₂ transition between gas and liquid phases. We also developed preliminary empirical models to calculate the velocity of CO₂ based on newly measured data.

INTRODUCTION

Recently, CO₂ has been considered a major factor in global warming as a result of the greenhouse effect. The increasing concentration of CO₂ in the atmosphere has been associated with human activity. The oil industry is a key contributor to controlling CO₂ emissions and with the environmental industry and the government, they work together to protect the Earth's environment. Extensive studies on the properties of CO₂ have been undertaken experimentally and theoretically to find ways to minimize its impact on the environment (Bachu, 2000; Benson et al., 2005). Consequently, CO₂ sequestration, injection, and storage are increasingly important strategies used to fight the global warming trend (Skov et al., 2002). However, strategic technology on such enormous scales of time, space, and economics is far from maturity. Many challenges remain unresolved (Bachu, 2000; Onishi et al., 2007; Sijacic et al., 2008), for example, where we

try to store CO₂, how we control injected CO₂, how we ensure CO₂ will stay where it is, and so on.

The bottom line is that we need the capability to monitor CO₂ movement and distribution. Technologies developed in the energy industry to enhance oil recovery in different reservoirs, such as seismic monitoring of fluid movement and distribution (Wang et al., 1998; Xue et al., 2005), the influences of changes in in-situ pressure and temperature conditions, and monitoring of stress and fracture distribution and development, can play a crucial role in CO₂ sequestration operations (Lazaratos and Marion, 1997; Majer et al., 2006). Because fluids are the main target for monitoring, we have invested much effort to investigate seismic properties of hydrocarbon fluids (gas, oil, and brine) (Batzle and Wang, 1992; Han and Batzle, 2000a, 2000b). Carbon dioxide as a basic chemical compound has unique properties: It is a nontoxic, colorless, odorless, and nonflammable gas that is economically produced and purified. In the energy industry, CO₂ has been used as a major injection fluid (gas) because of its superior low viscosity, low surface tension, and high miscibility with oil.

Carbon dioxide also shows promise in displacing methane from coal and natural gas hydrates. It has not been a target of study as a globally harmful material subject to sequestration until recently. We need to investigate the seismic properties of CO₂ as a target fluid in the earth's system over a wide range of temperature and pressure conditions. Carbon dioxide has been injected into the ocean bottom at a depth of several thousands of meters with temperatures as low as 4 °C and pressures as high as several tens of megapascals (Zhang, 2005). Carbon dioxide has been injected into subsurface reservoirs in which conditions could be above 200 °C and 100 MPa. Near the surface, under pipeline conditions, the temperature of CO₂ is usually in the range of –20 °C to 50 °C and pressure is in the range of 5 MPa to 25 MPa (Folas et al., 2007). Carbon dioxide is usually in a liquid or supercritical phase under the above temperature and pressure conditions.

Many theoretical equations of state (EOS) have been developed for gases (e.g., Span and Wagner, 1996). Some of these relationships have been used to develop publicly available property calculation

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software (Huber, 2007). However, CO₂ is such a unique gas with complex behavior that those equations are unable to accurately predict its thermodynamic properties (such as seismic velocity) (Batzle and Han, 1998), especially over the entire range of temperatures and pressures required for CO₂ operations. In addition, there are limited laboratory measurements of acoustic properties of CO₂ beyond 40 MPa.

In this paper, we present newly measured velocity data of CO₂ in a wide range of temperatures from -10°C to 200°C and pressures from 7 MPa up to 100 MPa. We also develop preliminary empirical models to calculate the velocity of CO₂ based on measured data.

PHASE DIAGRAM OF CO₂

The pressure and temperature phase diagram of CO₂ shows three phases separated by phase boundaries between solid, liquid, gas, and supercritical fluid in conjunction with the triple point (0.518 MPa and -56.6°C) and the critical point (7.38 MPa and 31.1°C), as shown in Figure 1 (after Shakhshiri, 2008). For CO₂ as an injected (storage) fluid under our measurement conditions, we mainly cover part of liquid- and supercritical liquid-phase regions as shown in Figure 1.

EXPERIMENTS

For our velocity measurements, we are using wave propagation in the megahertz range. Under our experimental conditions, CO₂ is in a fluid phase. Velocity depends on bulk modulus \mathbf{K} and density ρ as

$$V_p = \sqrt{\frac{K}{\rho}}. \quad (1)$$

The bulk modulus here is the dynamic or adiabatic modulus. Compressibility values (inverse of bulk modulus) can be derived from the density behavior with pressure but these would be the static or isothermal values. The adiabatic and isothermal values can differ by around 50% in most regions but this difference increases substantially as the critical point is approached.

The measurement system consists of a test vessel, a transfer (storage) vessel, temperature control, and pressure control, as shown in Figure 2. The test vessel is composed of titanium and connected with

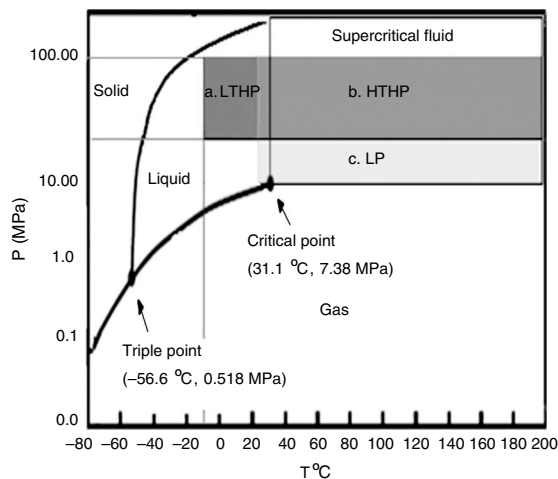


Figure 1. Phase diagram of CO₂ and experimental investigated areas: (a) LT and HP range; (b) HT and HP range; (c) LP range.

the transfer vessel. We use an ISCO digital pump (65D) to control the pressure of the transfer vessel and use a pressure transducer (Precise Sensors 6550) with 0.1% accuracy to measure the pressure of the CO₂ fluid in the test vessel. A silicon hot pad wrapped around the surface of the test vessel is used to heat the vessel. Temperature is controlled by a controller (Digi-Sense 2186-00) with a dual thermal couple system: one is placed on the outside surface of the test vessel to control the temperature of the hot pad and another is placed inside the sample chamber to monitor the temperature of CO₂ fluid to within 0.1°C accuracy. There are two acoustic transducers placed on both sides of the fluid-sample chamber. One is a transmitter and the other is a receiver. Velocity measurement depends on distance L (i.e., sample length) between the two transducers and traveltime t of acoustic wave between the transducers as

$$V = \frac{L}{t}. \quad (2)$$

Distance between the transducers varies slightly with variation of pressure and temperature. We use distilled water as a standard sample to calibrate the distance between the transducers as a function of pressure and temperature conditions. We apply the International Association for the Properties of Water and Steam — Industrial Formulation 1997 for the Thermodynamic Properties of Water and Steam (IAPWS-IF97; Wagner and Kruse, 1998) and the Fluid Application Geophysics program (FLAG program — a computer program developed by the Fluids/DHI consortium) as standard for the velocity of distilled water. In this way, the calibrated distance between the transducers is known at a given condition and velocity depends only on measured traveltime. This method is suitable for a stable system and high pressure and temperature measurement. However, we need to regularly check the calibration.

In an alternative technique, we use an acoustic probe to measure velocity directly. We apply a micrometer to move one transducer (with a limit of the sample pressure < 2000 psi) and measure distance change (ΔL) and traveltime change (Δt). In this way, velocity can be calculated directly as

$$V' = \frac{\Delta L}{\Delta t}. \quad (3)$$

We compare velocity V' with velocity V to calibrate the distance between the transducers. In the region near the critical point, we must use a different set of transducers and a resonant method to measure the extremely attenuated signal and low velocity. Considering controlled pressure and temperature conditions, the accuracy of ve-

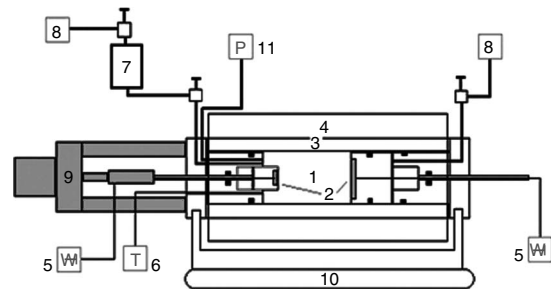


Figure 2. Schematic experimental setup: (1) fluid sample; (2) transducers; (3) sample vessel; (4) temperature controller; (5) scope; (6) temperature indicator; (7) CO₂ storage vessel; (8) digital pump; (9) micrometer; (10) system holder; (11) pressure indicator.

locity measurement of the CO₂ sample is better than 0.5%. Velocity data on the CO₂ sample are measured using an acoustic system consisting of a Pulser-receiver (Panametric 5055PR) and a LeCroy digital oscilloscope (WaveSurfer 42Xs). Although waveforms change with temperature, the first arrival is always distinct, as seen in Figure 3. All waveforms were recorded in the scope. We can pick the first arrival accurately and systematically with cross references of different waveforms in the same time base.

Considering different behaviors of CO₂ velocity in the wide range of temperature and pressure and different experiment set-ups, we need to separate the region of interest into three parts, as shown in Figure 1:

- 1) Low temperature (LT) and high-pressure (HP) range within the CO₂ liquid phase: $-10^{\circ}\text{C} \leq T \leq 20^{\circ}\text{C}$ and $20 \text{ MPa} \leq P \leq 100 \text{ MPa}$. Here we use a cooling bath to control the temperature of the vessel containing the CO₂ sample.
- 2) High temperature (HT) and HP range within the CO₂ supercritical fluid: $25^{\circ}\text{C} < T \leq 200^{\circ}\text{C}$ and $20 \text{ MPa} \leq P \leq 100 \text{ MPa}$. We use a simple heating pad to control the temperature of the CO₂ sample.
- 3) Low pressure (LP) range near the critical point of CO₂: $7 \text{ MPa} < P < 20 \text{ MPa}$.

Near the critical point (31.1°C and 7.38 MPa), CO₂ is in a transition phase between liquid and gas. Its properties change drastically. The acoustic velocity decreases and nears zero as the critical point is approached (see, for example, Walas, 1985). We also apply much denser sampling in pressure (1 MPa interval) and temperature (5°C interval) to examine in detail the velocity of CO₂ at the phase transition between liquid and gas.

In our measurements, we used industry-grade CO₂ (provided by Matheson Tri-Gas[®]) of 99.5% purity for measurement. In the earth, CO₂ will occupy the pore space with other fluids such as water or hydrocarbons. These compounds, when mixed with CO₂, can alter its properties substantially. The behavior of such mixtures is important but beyond the scope of this paper.

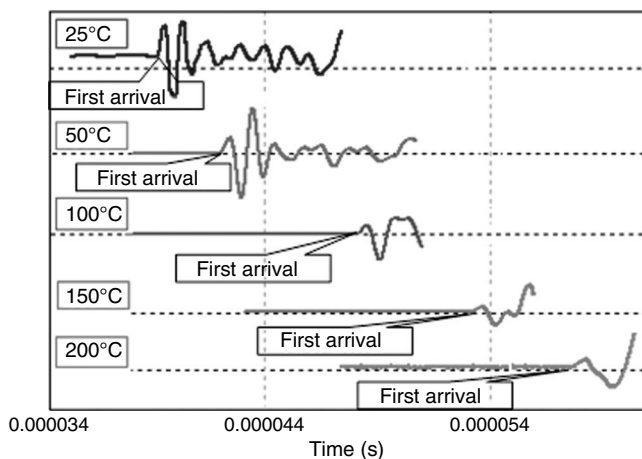


Figure 3. Waveforms of transmission P-wave received at pressure $P = 100 \text{ MPa}$ for different temperatures from 25°C to 200°C .

MEASURED RESULTS AND DISCUSSION

We present measured data in the following three parts.

Velocity of CO₂ in the LT and HP range

Low temperature (LT) means below room temperature (-10 to 20°C) and HP ranges from 20 MPa to 100 MPa , as shown in Figure 1. Here, CO₂ is in a liquid form. Velocity data measured in the LT and HP range are shown in Figure 4. The velocity increases as pressure increases at a given constant temperature and decreases as temperature increases at a given constant pressure. We also observed that the gradient of velocity with respect to pressure gradually reduces with increasing pressure and decreasing temperature (larger pressure effect at a higher temperature and a larger temperature effect at a lower pressure), as indicated in Figure 4. However, the influence of the temperature effect to reduce velocity decreases with increasing temperature. According to equation 1, because the bulk modulus and density are functions of temperature and pressure, the resulting velocity change will depend on the competing relative change between bulk modulus and density. When the temperature remains constant, the increase of the pressure requires that more molecules be packed in a unit volume. This decreases the distance between molecules and causes an increase in density. The repulsive force between molecules in a liquid phase follows a nonlinear relationship with distance rather than a simple linear relationship, as in Hooke's law. When the liquid is compressed, the relative increase of the bulk modulus is greater than the relative increase of density, resulting in a velocity increase with a pressure increase.

A similar analysis tells us that increasing temperature at constant pressure will cause a relatively larger decrease in the bulk modulus than in density, thus, resulting in a decrease in velocity (Figure 5). The temperature effect on the decrease of velocity shows a trend to increase with decreasing pressure, especially at a pressure of 20 MPa , as shown in Figure 5. Carbon dioxide as an LP liquid is less confined and temperature shows a greater effect in reducing CO₂ velocity. Overall, we can summarize that the velocity data of the CO₂ liquid in LT and HP conditions is typical of a liquid.

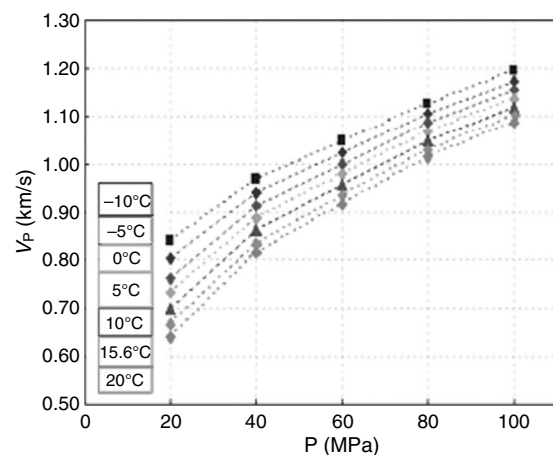


Figure 4. Measured velocities of CO₂ as a function of pressure P for different temperatures from 20°C to -10°C .

Velocity properties in the HT and HP range

Here HT means higher than room temperature (25–200 °C). The examined pressure range is high (HP), from 20 MPa to 100 MPa. According to the phase diagram in Figure 1, CO₂ in this range is in a supercritical fluid phase.

Measured velocity data are shown in Figure 6. Similar to velocity data measured at an LT and HP range, the velocity of CO₂ in an HT and HP range increases as pressure increases at constant temperature and decreases as temperature increases at constant pressure. However, the temperature effect becomes complicated, as shown in Figure 7. At HP (>40 MPa), velocity of CO₂ decreases with increasing temperature in a monotonic manner but as pressure is lowered the dependence on temperature decreases. Eventually, at lower pressure and elevated temperature (the 20 MPa line in Figure 7), the velocity increases with increasing temperature. This type of temperature dependence is more typical of light gases. Thus, in this supercritical region, we begin to see the transition from liquid-like to gas-like behavior.

Velocity of CO₂ around the critical point

We also measured the CO₂ velocity near the critical point. Properties of CO₂ as a supercritical fluid are very sensitive to temperature and pressure as we approach the critical point (Walas, 1985). A very small drop in pressure can significantly change the phase state of CO₂ from liquid-like to gas-like. Remember that a passing acoustic wave is a small perturbation in pressure. Near the critical point, the molecules transition between liquid- and gas-like phases, resulting in rapid density change, decrease of velocity, and increase of wave attenuation. Therefore, we must apply a different measurement set-up and much finer measurement intervals around the critical point. For temperatures from 25 °C to 45 °C, we use a 5 °C interval and for pressures from 7 MPa to 13 MPa we use a 1 MPa interval.

As shown in Figure 8, the top two curves with temperatures of 25 °C and 30 °C (below the critical point temperature of 31.1 °C) exhibit the typical liquid velocity-pressure relationship. However, when the temperature approaches and then exceeds the critical temperature (velocity data measured at 35 °C, 40 °C, and 45 °C), the velocity-pressure relationship can be clearly separated into a gas-like regime and a liquid-like regime with minimum velocity between

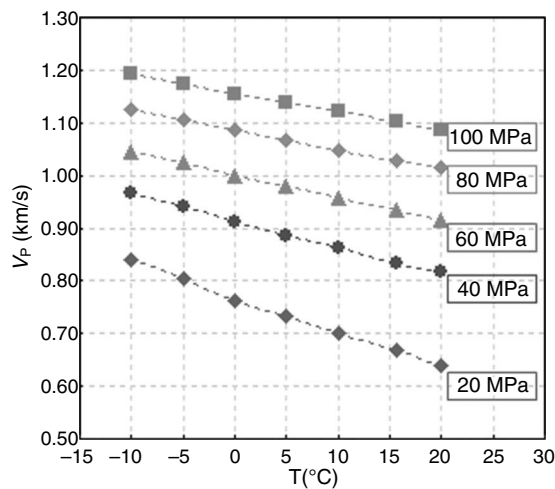


Figure 5. Measured velocities of CO₂ as a function of temperature T for different pressures from 20 MPa to 100 MPa.

these two regimes. With decreasing pressure, CO₂ velocities in the liquid regime tend to decrease to a minimum due to a drastic decrease in modulus near the critical point and then to increase because density decreases drastically as CO₂ transitions from a liquid phase to a gas phase. The transition between the two regimes approximate-

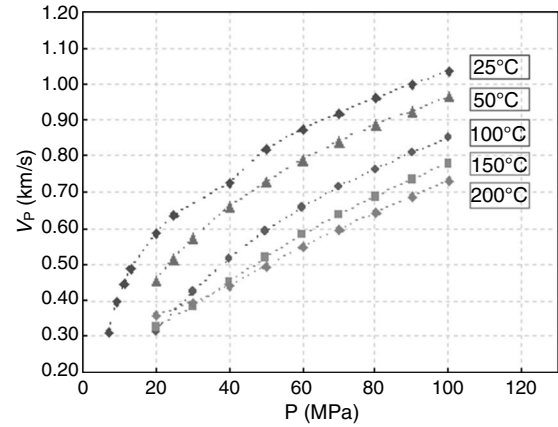


Figure 6. Measured velocities of CO₂ as a function of pressure P for different temperatures from 25 °C to 200 °C.

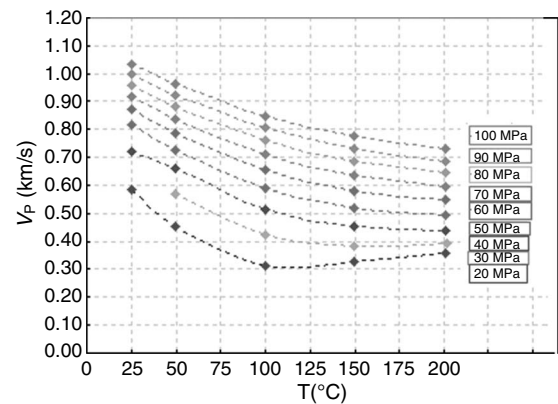


Figure 7. Measured velocities of CO₂ as a function of temperature T for different pressures from 20 MPa to 100 MPa.

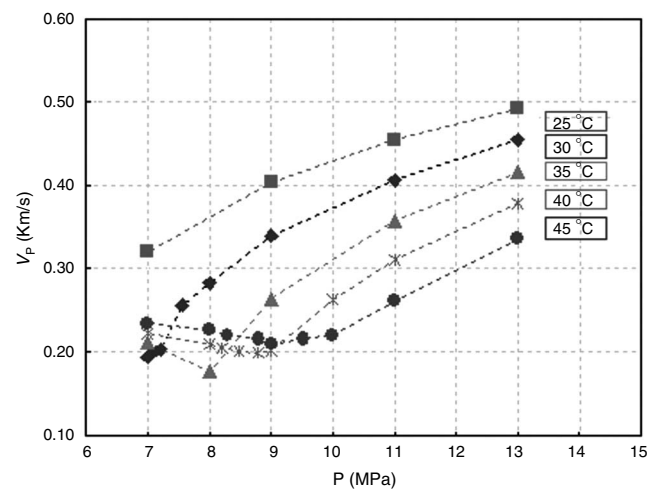


Figure 8. Measured velocities of CO₂ as a function of pressure P for the range near the CO₂ critical point.

ly corresponds to the CO₂ critical point pressure of 7.38 MPa. We also observe similar complicated temperature effects on velocities of CO₂ near the critical point (Figure 9). Thus, although we can reside in a single-phase supercritical state, behavior can be either liquid-like (HP) or gas-like (LP). For CO₂ in a liquid-like state, velocities will decrease with increasing temperature but the opposite is true for CO₂ in a gas-like state. High pressure tends to drive CO₂ to be a liquid whereas HT tends to drive CO₂ to be a gas. Velocities of CO₂ are determined by balanced temperature and pressure effects, especially near the critical point.

EMPIRICAL MODELS

Based on our three measured groups of data that cover wide ranges of pressures (7–100 MPa) and temperatures (–10–200 °C), we took several approaches to describe the velocity of CO₂. Initially, this included examining published models and then comparing them to the EOS for CO₂ (Duan et al., 1992; Span and Wagner, 1996; Batzle and Han, 1998).

We found that these model results either do not fit well with measured data or are too complicated for application; therefore, we developed empirical models. Because of the complexity near the critical point, we have not yet obtained a simple, accurate, unified empirical model for the three groups of data. However, here we present three empirical models to describe velocities of CO₂ valid under restricted temperature and pressure ranges.

CO₂ velocity model (LTHP) for the LT and HP range

Based on experimental results, the following polynomial model is suitable to describe CO₂ velocity as a function of temperature and pressure for this range:

$$V_p = a + bT + cT^2 + dP + eP^2 + fTP, \quad (4)$$

where $a = 0.61571$, $b = -0.00709$, $c = 0.00002$, $d = 0.00833$, $e = -0.00003$, $f = 0.00004$, V_p is in kilometers per second, T in degrees Celsius, and P in megapascals.

The comparison of calculated CO₂ velocities with measured data is shown in Figure 10. The model correctly matches the CO₂ velocity in the temperature and pressure range ($-10^\circ\text{C} \leq T \leq 20^\circ\text{C}$ and $20 \text{ MPa} \leq P \leq 100 \text{ MPa}$) to within an error of 0.7%.

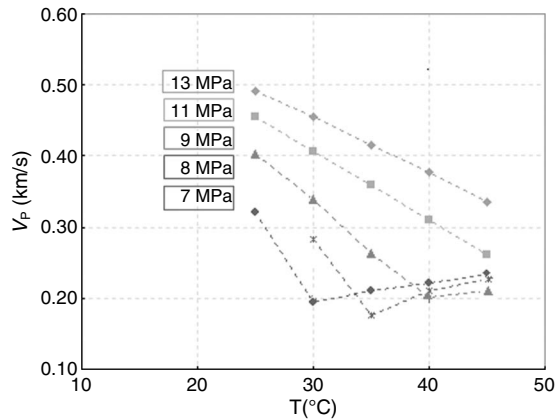


Figure 9. Measured velocities of CO₂ as a function of temperature T for the range near the CO₂ critical point.

CO₂ velocity model (HTHP) for the HT and HP range

We can use the following model for the HT and HP range:

$$V_p = (1 - W_f)V_{lp} + W_fV_{hp}, \quad (5)$$

where

$$W_f = \frac{W_{f1} + |W_{f1}|}{2(0.5 + |W_{f1}|)}, \quad (6)$$

$$V_{lp} = 150 + 120T_{pr} - [9 + 175(1.5 - T_{pr})^2]P_{pr}, \quad (7)$$

$$V_{hp} = 45 + 600(a - T_{pr})^3 + 246|P_{pr} - S_{hp}|^{0.44}, \quad (8)$$

and

$$W_{f1} = (P_{pr} + 7.661 - bT_{pr})^9, \quad (9)$$

$$P_{pr} = \frac{P}{7.386} + 4.20831 \frac{T^{1.43}}{(c - T)(1 + 1.249P)}, \quad (10)$$

$$T_{pr} = \frac{1}{304.21} [T_{abs} - d(304.21 - T_{abs})], \quad (11)$$

$$T_{abs} = T + 273.15. \quad (12)$$

The values of coefficients a , b , c , and d in equations 8–11 are $a = 1.71$, $b = 7.789$, $c = 312$, and $d = 0$.

Figure 11 shows the modeled versus computed CO₂ velocity. This model is valid for the range of temperatures (from 25 °C to 200 °C) and pressures (from 20 MPa to 100 MPa) and is accurate to within an error of 1% except several temperature points from 50 °C to 100 °C and pressures below 40 MPa.

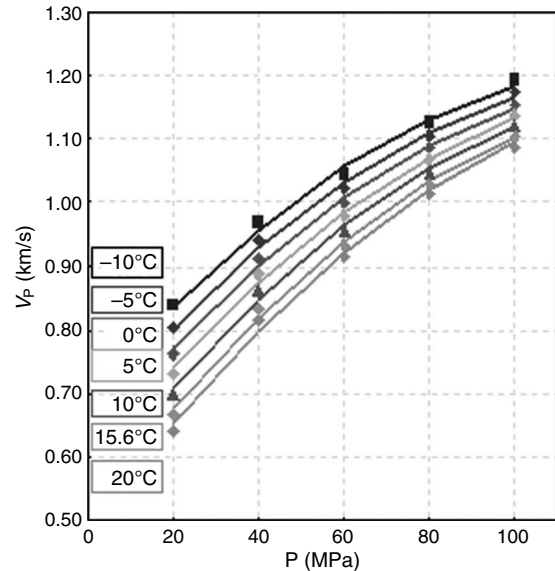


Figure 10. Calculated (solid lines) and measured (symbols) velocities of CO₂ as a function of pressure P for different LT from 20 °C to –10 °C.

CO₂ velocity model (LP) near the critical point

Because the LP range from 7 MPa to 20 MPa includes the CO₂ critical point, the velocity is very sensitive to temperatures and pressures. The form of equations 5–12 for the HT and HP range can be used to predict the velocity behavior of this LP range, but the values of coefficients *a*, *b*, *c*, and *d* in equations 8–11 are now *a* = 1.66, *b* = 6.405, *c* = 30,000, and *d* = 40.

The result shown in Figure 12 indicates they are almost matched within temperatures from 25°C to 40°C and the pressure range from 7 MPa to 13 MPa (not accurate near the critical point).

The results of comparing the measured CO₂ velocities to those calculated by these empirical models and by Span and Wagner’s EOS (Span and Wagner, 1996) are shown in Figures 13 and 14. The EOS equation for CO₂ covers the wider temperature and pressure region from the triple-point temperature to 1100 K at pressures up to 800 MPa. The equation is accurate but contains many more terms. Some of the terms are complex exponentials that become difficult to

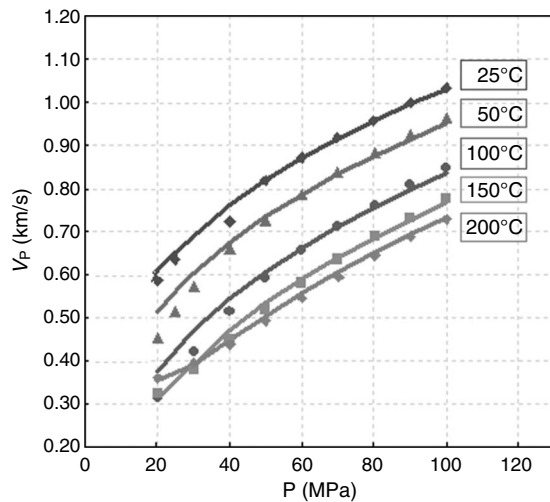


Figure 11. Calculated (solid lines) and measured (symbols) velocities of CO₂ as a function of pressure P for different temperatures from 25°C to 200°C.

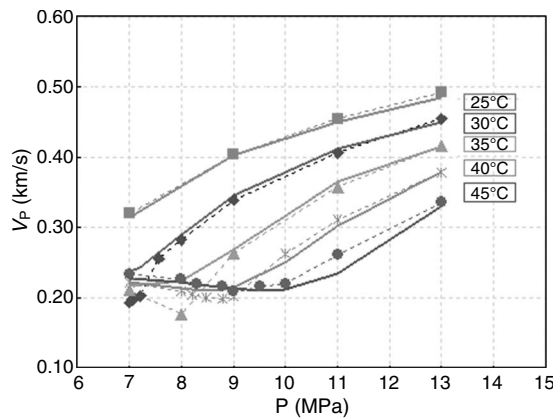


Figure 12. Calculated (solid lines) and measured (dashed lines) velocities of CO₂ as a function of pressure P for the range near the CO₂ critical point.

evaluate accurately. Our new empirical models with fewer terms and a simple exponential are simpler and easier to implement. From Figure 14, we can see that at LT (e.g., < 100°C), the predicted velocity by the EOS model begins to deviate from the measured velocity trend at 50 MPa or lower. The new model is less accurate than Span and Wagner’s in very LP regions but gives a more accurate prediction at elevated pressures.

Xu (2006) describes CO₂ properties by modifying Batzle-Wang’s equations (Batzle and Wang, 1992). Compared to the laboratory-measured data by Wang and Nur (1989), Xu’s calculated velocities produce a better match to the measured data than by using the original Batzle-Wang equations. We also applied our model to that same data set. A comparison of the measured and calculated results is shown in Figure 15, modified from the plot of Xu (2006). In Figure

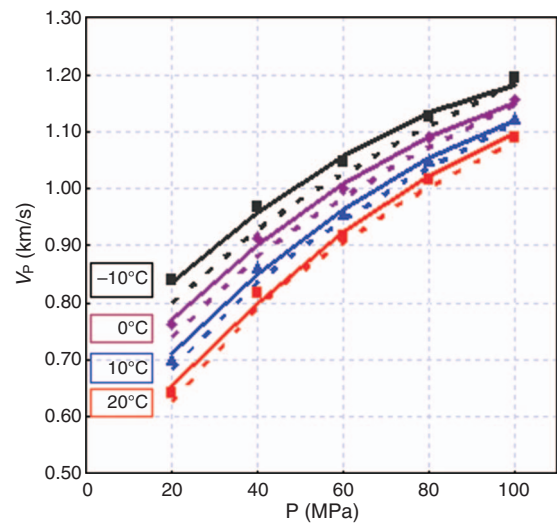


Figure 13. Comparing the calculated (solid lines based on the LTHP model) and measured velocities with the calculated velocities (dashed lines) by Span and Wagner (1996) for the LT and HP range.

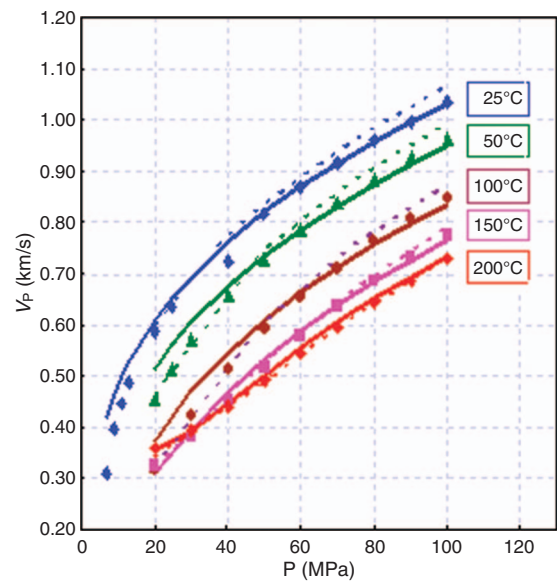


Figure 14. Comparing the calculated (solid lines based on the HTHP model) and measured velocities with the calculated velocities (dashed lines) by Span and Wagner (1996) for the HT and HP range.

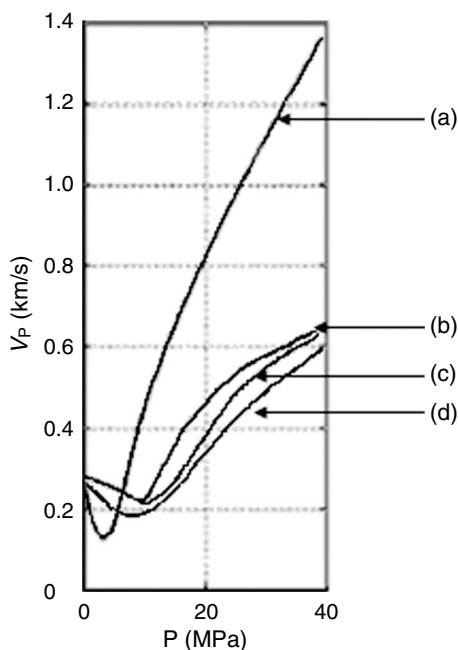


Figure 15. Comparing the calculated and measured CO₂ velocity: (a) Calculated velocity using Batzle-Wang equations; (b) calculated velocity using our model; (c) measured data by Wang and Nur (1989); (d) calculated velocity using Batzle-Wang's equations with modified parameters (Xu, 2006).

15, line a is calculated by using the original Batzle-Wang equations, line b is calculated by using our empirical model, line c represents measured data by Wang and Nur (1989), and line d is calculated by using Batzle-Wang equations with modified parameters by Xu (2006). It can be seen that our model matches the experimental data better when pressure is lower than the critical point (7.38 MPa) and higher than 25 MPa. Xu's results are better between approximately 10 MPa and 20 MPa.

CONCLUSION

We have carried out an experimental study of the acoustic velocity of CO₂ over a wide range of temperatures and pressures ($-10^{\circ}\text{C} \leq T \leq 200^{\circ}\text{C}$ and $7 \text{ MPa} \leq P \leq 100 \text{ MPa}$). In the HP range, $20 \text{ MPa} \leq P \leq 100 \text{ MPa}$, CO₂ is in a liquid-like state and velocities of CO₂ increase as pressure increases and temperature decreases. When pressure falls below 20 MPa with temperature in a range of $25\text{--}45^{\circ}\text{C}$, CO₂ approaches the critical point (31.1°C and 7.38 MPa) and its velocity is highly sensitive to temperature and pressure change. The velocities of CO₂ reach the minimum near the critical point. At pressures above the critical point, liquid-like behavior dominates (V_p decreases with increasing temperature). As pressures drop below the critical point, gas-like behavior dominates (V_p increases with increasing temperature).

We have developed velocity models of CO₂ based on the experimental data and described the relationships of CO₂ velocity with temperature and pressure. Generally, our models fit well with the experimental data. However, more laboratory data are needed to limit the bias of the experimental data and to refine the models, particularly in the critical region.

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