Introduction

In the perspective of the Carbon Capture and Storage (CCS) in geological reservoir aquifer, knowledge of the thermo-physical properties such as viscosity and density of the Carbon Dioxide (CO$_2$) aqueous solutions are essential (Wang et al., 2011). Viscosity, for example is one of the key important parameter, not only in the transport pipeline system but also for the subsurface storage and process systems (IEAGHG, 2011). Another property of particular importance to CO$_2$ storage is the density of the brine aqueous solution. If the dissolution of gas into brine causes an increase of brine density and this density difference may also be large enough to trigger Rayleigh instability, which can strongly enhance dissolution processes due to mixing. These two properties play a significant role in understanding the behaviour of injected CO$_2$ stream throughout the CCS scheme and consequently in oil and gas exploitation (Ozah et al., 2005).

Viscosity and density of CO$_2$ aqueous solutions are extensively studied over a wide range of pressure and temperature and available in the open literature. However, due to the vast variety of the possible CO$_2$ mole fractions with different type of brine compositions (different type of salts present), as well as with the different operating ranges of pressure and temperature in CCS processes, further viscosity and density experimental data or results have to be generated. Therefore, in this communication, new viscosity and density data of CO$_2$ aqueous solutions (in pure water, single salt NaCl and mixed salts brine) are generated at 353.15 K, 373.15 K, 393.15 K and 423.15 K, and in a pressure ranges up to 70 MPa. PVT laboratory from Heriot Watt University was utilized for this thermo-physical properties measurements. Apart from filling the literature gaps, this new thermo-physical data generation are used to validate the current predictive tools for this CCS purposes.

Material and Apparatus

In this study, pure CO$_2$ from BON N4.5 (99.995 mol%) is used to measure both viscosity and density of the CO$_2$ aqueous solution measurements. NaCl salt with a concentration of 0.35 M and mixed salt brine aqueous solution was prepared gravimetrically by adding the salts which the information was taken from the real field aquifer brine information.

![Schematic diagram of the High Pressure – High Temperature (HPHT) viscosity and density experimental setup.](image-url)
Viscosity and density measurements were performed using High Pressure and High Temperature (HPHT) setup (Chapoy et al., 2013). This rig facility was previously designed and manufactured at Institute of Petroleum Engineering, Heriot Watt University (HWU), with capacity of maximum working pressure up to 200 MPa (29,000 psia) and working temperature of 250 °C (523.15 K). The set-up is located inside the oven, manufactured by BINDER GmbH, where it was capable of being used at temperatures from -70 °C to 200 °C (203.15 K to 473.15 K) with uncertainties of ±0.05 °C. The schematic diagram of this setup is shown in Figure 1.

The setup is consisted of two small cylinders, with volume of 25 cm³, connected to each other through a capillary tube with measured length of 14.78 metres and a temperature-dependent calibrated internal diameter. An oscillating U shape vibrating tube densitometer Anton Paar DMA-HPM was used and connected to the set-up. Two three-way valves, located one on top of the cylinders connected to capillary tube and another one on top of the densitometer, are installed to introduce fluids or samples into the cylinders, tube system and densitometer. The base side of the two cylinders are connected to opposite sides of a push-pull, motor driven mercury pump. This pump can move the sample fluid forwards and backwards between the two cylinders. Apart from this, there is also a hand pump connected to the system, manufactured by Sitec Engineering of Switzerland to control the pressure of the entire fluid system by injection and withdrawal of mercury. Two Quartzdyn pressure transducer (model: QS 30K-B) with the design pressure up to 207 MPa or up to 30,000 psia with a standard uncertainty of ±0.02 MPa were connected to record the system pressure.

Experimental Procedure

The capillary tube viscosity measurement method has been employed to measure the viscosity of aqueous solutions, with and without CO₂ system (Kashefi, 2012). For each of the test, after applying vacuum to the entire system, the set-up was loaded with the sample mixtures through the injection point at the top of the densitometer. Then after disconnecting the sample cylinder from the system, the sample fluid was pushed through the capillary tube into the other cylinder using the push-pull mercury pump. The temperature of the system was set to the desired condition and the desired pressure was achieved using the hand pump. Once conditions had stabilized, and after isolating the densitometer by closing the related valve, the sample was pumped through the capillary tube at a constant flow rate. To ensure the consistency of the measurements, at each pressure, viscosities were determined at a constant flow rate with six readings were logged, so the reported viscosity data in this study are an average of at least six or nine separate readings. Pumping the sample fluid through the capillary tube by piston pump resulted in a dynamic differential pressure that was monitored and recorded until stable. Then the pump was stopped to record the static differential pressure. The difference between the dynamic and static differential pressure was used as the pressure drop across the tube using Poiseuille equation. During this time, the related valve connected to the densitometer were switched and connected. The sample was injected into the U-tube which is electronically excited. The frequency of the excited U-tube is a function of sample mass in the U-tube. The densities of the sample are determined by the measurement of the oscillation period of the tube.

Modelling

For CO₂ aqueous solution viscosity model prediction, few models were tested and developed. Predictive model based on one reference fluid corresponding states (CS) theory, i.e Pederson and CO₂ Pedersen, predictive models based on two reference fluid corresponding states (CS) theory, i.e., Aasberg-Petersen and CO₂-CS2, predictive models based on extended corresponding states (ECS) theory, i.e, SUPERTRAP and CO₂-SUPERTRAP and Lennard-Jones spheres viscosity model were used to predict the viscosity of the CO₂ aqueous solution. The CO₂–Pederson model predicts the viscosity using the corresponding states theory, while for the CO₂-TRAPP model predicts using the extended corresponding states theory by employing the shape factors. For the density data model prediction, Peng Robinson (Robinson & Peng, 1978) and Soave-Redlich-Kwong (SRK) (Soave, 1972) and GERG-2008 (Kunz & Wagner, 2012) Equation of State (EoS) were used and compared against density experimental data, for validation. Ahmadi et al., (Ahmadi, 2018) also employed the same EoS model predictions to predict the density of the mixture tested in their recent completed study.
Results and Discussions

It is observed that our viscosity results for pure water (for validation) are in excellent agreement with the predictions of the IAPWS viscosity model (Wagner & Kretzschmar, 2008) with calculated AAD=0.25% and well within the uncertainties of the model (1%). The viscosity result for the NaCl solution and mixed salt brine are also in good agreements with the model predictions calculated using a modified Jones and Dole equation. Figure 2 (a) shows the viscosity data obtained for mixed salt brine solution at several isotherms, compared against calculated viscosity models and Figure 2 (b) is the measured viscosity when CO₂ is introduced into the system against calculated viscosity model. Comparison between viscosity data measured for pure water against IAPWS model prediction are also shown in the Figure 2 (b).

Figure 2 (a): Measured and calculated viscosity of mixed salt brine solution at 353.15 K, 373.15 K, 393.15 K and 423.15 K. Figure 2 (b): Viscosity of water and CO₂ aqueous solutions (in mixed salts) at several CO₂ mole fraction at T=423.15 K, against IAPWS prediction (red line).

Density results obtained for each of the aqueous solution with and without CO₂, in pure water, NaCl and mixed salts brine are also compared against model predictions, for validation. Figure 3 (a) shows one of the density result for mixed salt brine, without dissolved CO₂ measured at few isotherm runs, at 353.15 K, 373.15 K, 393.15 K and 423.15 K. And Figure 3 (b) shows the effect of dissolved CO₂ on the densities of the mixed salt brine solution at T=393.15 K (as for an example). Both are in good agreement with model predictions, with less than 0.1% AAD calculated.

Figure 3 (a): Measured and calculated density of mixed salt brine solution at 353.15 K, 373.15 K, 393.15 K and 423.15 K (from top to bottom). Figure 3 (b): Effect of dissolved CO₂ on densities of the mixed salt brine solution at T=423.15 K, (dashed lines no CO₂/red lines xCO₂ = 0.02575)
Conclusion

In this study, viscosity and density of the (CO$_2$) aqueous solutions were measured at few isotherms, at 353.15 K, 373.15 K, 393.15 K and 423.15 K and pressures up to 70 MPa. Excellence agreements against available viscosity and density literature data and model predictions show reliability of the new results obtained from this study. Uncertainties and % AAD calculated for each of the measurements will be demonstrated in the full paper. Apart from filling the literature data gaps, the new viscosity and density thermo-physical data generation will help validating the existing viscosity and density prediction models.

References


