Interfacial tension of (brines + CO₂). II. CaCl₂(aq), MgCl₂(aq) and Na₂SO₄(aq) at temperatures between (343 and 423) K, pressures between (2 and 50) MPa, and molalities of (0.5 to 5) mol·kg⁻¹

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Abstract

We report the interfacial tensions between carbon dioxide and CaCl₂(aq), MgCl₂(aq) and Na₂SO₄(aq) with molality from (0.49 to 5.0) mol·kg⁻¹. The measurement were made at temperatures between (323 and 423) K at various pressures up to 50 MPa. The pendant drop method was implemented in a high-pressure view cell filled with water-saturated CO₂ into which single drops of brine were injected through a suitable capillary. The expanded uncertainties at 95 % confidence are 0.05 K in temperature and 70 kPa in pressure. For the interfacial tension, the expanded relative uncertainty at 95 % confidence was 1.6%. The results of this study show that interfacial tension increases linearly with molality. Further, at constant temperature and pressure, the interfacial tension is the same function of the positive charge molality for all salts investigated in this work.

Keywords: Brine; Carbon Dioxide; Calcium Chloride; Magnesium Chloride; Sodium Sulfate; Interfacial Tension, High Pressure.

Introduction

The study of interfacial properties between carbon dioxide and the fluids and minerals present in oil reservoirs and deep saline aquifers is fundamental to a scientific understanding of both CO₂-enhanced oil recovery and geological carbon storage. For example, the capillary threshold pressure for caprock breakthrough increases linearly with increasing interfacial tension. However, examination of the literature shows that there have been only very limited experimental studies of the interfacial tension (IFT) between [supercritical](#) CO₂ and brines at reservoir conditions. The available data are scattered and in some cases contradictory. For example, one study showed that the addition of salt (20 g·L⁻¹ NaCl) to the water phase has a negligible effect on the IFT,¹ while other work has shown a distinct increase under salt addition.²⁻⁴ Thus further measurements of high accuracy would be useful in order to clarify the situation.

The object of this research is to investigate the dependence of IFT on temperature, pressure and salinity (including both salt type and molality) over the range of conditions applicable to CO₂ storage in saline aquifers. One objective is to arrive at a universal model, based on such experimental data, for calculating the IFT of any (CO₂ + brine) system.

We recently reported the IFT between [supercritical](#) CO₂ and a mixed brine containing NaCl and KCl at temperatures between (298 and 448) K, pressures between (2 and 50) MPa, and molalities of (1 to 5) mol·kg⁻¹.² To further investigate the dependence of CO₂-brine IFT on the concentration and type of ions we report, in this paper, further results pertaining to aqueous solutions of CaCl₂, MgCl₂ and Na₂SO₄ at temperatures from (343 to 423) K, pressures from (2 to 50) MPa, and molalities of (0.49 to 5) mol·kg⁻¹.

Experiment

Apparatus. Various techniques can be used to determine the interfacial tension between fluid phases. In this study, we used the pendant drop method with axisymmetric drop-shape analysis (ADSA). The ADSA technique is based on matching the profile of the drop with the theoretical profile obtained by numerical integration of the Young-Laplace equation. In this research, commercial software (Advanced DROPimage, Ramé-Hart Instrument Co.) was used to perform this procedure. The apparatus has been described in detail elsewhere.² In brief, it comprised a high-pressure vessel fabricated from Hastelloy C-276, fitted with diametrically opposed sapphire windows, and encased in an insulated aluminium heater jacket. Fluids were injected from high-pressure syringe pumps. The capillary through which brine was delivered entered through the top of the cell so that pendant drops could be formed. Temperature was measured by means of a platinum resistance thermometer, inserted into the wall of the high-pressure apparatus, with an expanded uncertainty of 0.05 K (coverage factor $k = 2$). The pressure was measured by means of a pressure transducer, located in the tubing external to the view cell, with an expanded uncertainty of 70 kPa ($k = 2$).

Materials and methods. Pure deionised and degassed water (electrical resistivity $> 18 \text{ M}\Omega\cdot\text{cm}$) was used. Carbon dioxide was supplied by BOC with a specific minimum mole-fraction purity of 0.99995 in a cylinder fitted with a dip tube to permit withdrawal of liquid. Magnesium chloride hexahydrate, calcium chloride hexahydrate and anhydrous sodium sulphate were purchased from Sigma-Aldrich, with mass fraction purities of ≥ 0.99 . Solutions were prepared gravimetrically with the relative uncertainties in mass being below 0.01 %. Thus the relative uncertainty of molality was most probably limited only by the purity of the salts and was taken to be approximately 0.5 %. The solutes were fully dissolved in the water, first, by shaking and then by

sonicating them in closed containers in an ultrasonic bath. The brines were degassed by sonicating for 15 min immediately prior to use.

To obtain the IFT by the pendant drop method, the difference between the densities of the two fluids needs to be known. In this work, the coexisting phase densities were not measured and were instead calculated as in our previous work.² Based on the findings of Hebach et al.,⁵ the density of the CO₂-rich phase was taken to be that of pure CO₂ at the same temperature and pressure and was evaluated from the equation of state of Span and Wagner⁶ as implemented in NIST standard reference database 23 for thermodynamic and transport properties of fluids (REFPROP).⁷ The density ρ_{brine} of the CO₂-free brine was obtained from measurements made in our laboratory,⁸ and the small change in density upon saturation with CO₂ was estimated from the relation

$$1/\rho = (1-w)/\rho_{\text{brine}} + w/\rho_{\text{CO}_2}^* . \quad (1)$$

Here, ρ is the density of the solution, w is the mass fraction of dissolved CO₂, which we evaluated from the model of Duan *et al.*,⁹ and $\rho_{\text{CO}_2}^*$ is the inverse of the partial specific volume of CO₂ in solution, which was taken to be 1260 kg·m⁻³ independent of temperature and pressure.¹⁰ In the present case, the calculated density difference [between the two phases](#) was never less than 230 kg·m⁻³ and never deviated by more than 4 % from the density difference between pure CO₂ and CO₂-free brine. For purposes of estimating the uncertainty of [the interfacial tension](#), we take the standard uncertainty $\delta(\Delta\rho)$ in the density difference to be one tenth of the difference between the calculated density of the CO₂-saturated brine and the density of the CO₂-free brine. We note that recalculation of γ is straightforward if, for example, more

reliable experimental values of the density differences were to become available. The experimental procedure followed was identical to that described previously.²

Results and discussion

The new IFT measurements extend our earlier work to a wider range of brines varying not only in concentration but also in composition (both anions and cations). Table 1 detailed the systems studied and the ranges of molality m , temperature T and pressure p . $\text{CaCl}_2(\text{aq})$ and $\text{MgCl}_2(\text{aq})$ were studied at molalities of (2.5 and 5.0) $\text{mol}\cdot\text{kg}^{-1}$; for $\text{Na}_2\text{SO}_4(\text{aq})$ the molalities were (0.49 and 0.98) $\text{mol}\cdot\text{kg}^{-1}$ (restricted by the saturation limit at ambient temperature). Each system was studied at various temperatures between (343 and 423) K and various pressures between (2 and 50) MPa. The experimental data, 232 in number, are given in tables 2 to 7. The relative standard deviation $\sigma(\gamma)/\gamma$ of the IFT data gathered at each state point was computed from a number of repeated measurements; on average it was 0.7 % and in all cases it was < 2 %. The overall relative standard uncertainty u_r of γ was calculated from the relation

$$u_r^2 = \left[\frac{1}{\gamma} \left(\frac{\partial \gamma}{\partial T} \right)_{p,m} \delta T \right]^2 + \left[\frac{1}{\gamma} \left(\frac{\partial \gamma}{\partial p} \right)_{T,m} \delta p \right]^2 + \left[\frac{1}{\gamma} \left[\frac{\partial \gamma}{\partial m} \right]_{p,T} \delta m \right]^2 + \left[\frac{\delta \Delta \rho}{\Delta \rho} \right]^2 + \left[\frac{\sigma(\gamma)}{\gamma} \right]^2. \quad (2)$$

Combining the standard uncertainties δT , δp , δm and $\delta \Delta \rho$ in the temperature, pressure, molality and density difference respectively with the largest values of the corresponding partial derivatives found in the ranges investigated, the average u_r for all state points was found to be 0.8%. Thus the expanded relative uncertainty of γ at 95% confidence was 1.6%.

There are few previous data with which to compare the new results. However, in Figure 1 we compare our results for the IFT of $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at $T = 373$ K and $m = 2.5$ $\text{mol}\cdot\text{kg}^{-1}$ with results for the same system from Aggelopoulos *et al.*³ at $m = (1.8 \text{ and } 2.7)$ $\text{mol}\cdot\text{kg}^{-1}$. [To permit a](#)

more quantitative comparison, the data of Aggelopoulos *et al.*³ were interpolated linearly to $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$, fitted by a cubic function of pressure and then compared with the present results; we find agreement within $1 \text{ mN}\cdot\text{m}^{-1}$ everywhere within the overlapping pressure range.

Variation with pressure and temperature. For all brine types and concentrations, γ is observed to decrease with increasing pressure when the salinity and the temperature are kept constant. The results for $\text{CaCl}_2(\text{aq})$ for $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$ are taken as an example and plotted in figure 2. It is clear that γ decreases continuously with increasing pressure, rapidly at first and then more gradually at high pressures. Figure 3 shows the same data plotted along isobars as a function of temperature and it can be seen that the trends, while smooth, are more complicated. This has also been noted in the literature.^{4, 11-13}

Variation with salinity. The effect of salinity (both ion type and molality) on water surface tension under ambient conditions is documented in the literature.^{14, 15} Our results indicate that, for a given salt at constant temperature and pressure, the interfacial tension **increases** linearly with molality. However, again at constant temperature and pressure, different salts can exhibit the same or different effects on the IFT. For example, CaCl_2 , MgCl_2 and Na_2SO_4 all behave essentially the same but have a larger effect than brines containing NaCl and KCl . Figure 4 compares the present results for $\text{CaCl}_2(\text{aq})$ at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$ with our earlier data² for $(0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at $m = 5.0 \text{ mol}\cdot\text{kg}^{-1}$ as a function of time after creation of the drop at $T = 423.15 \text{ K}$ and $p = 2 \text{ MPa}$. It is observed that the IFT of CO_2 against both brines are essentially identical at $\gamma = (54.2 \pm 0.2) \text{ mN}\cdot\text{m}^{-1}$. The present results also show that $\text{MgCl}_2(\text{aq})$ and $\text{CaCl}_2(\text{aq})$ behave almost identically at the same temperature, pressure and molality. Figure 5 compares the IFT data for CO_2 with $\text{CaCl}_2(\text{aq})$ and $\text{MgCl}_2(\text{aq})$, both at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$, and $(0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at $m = 5 \text{ mol}\cdot\text{kg}^{-1}$, as functions of pressure along isotherms at $T =$

(343, 373 and 423) K. The results for each brine system appear to be the same to within the experimental uncertainty.

In view of the behaviour observed experimentally, we propose that the IFT between CO₂ and brines containing the chlorides or sulphates of Na, K, Mg and Ca may be represented as a linear function of the positive-charge molality, m^+ , which may be defined for a mixed brine by

$$m^+ = \sum_{i=1}^n z_i m_i . \quad (3)$$

Here, m_i is the molality and z_i is the charge number of the i th cation, and n is the number of cationic species present in the brine. Thus, for a single salt $M_v^{z+}X_{(z/y)v}^{y-}$, m^+ is the product of the stoichiometric number v and the charge number z of the cation M. On grounds of electrical neutrality, the negative-charge molality is identical. Figure 6 illustrates the proposed relation. Here we plot as a function of m^+ at $T = 373.15$ K and various pressures the IFT of pure water (from Georgiadis¹⁶), CaCl₂(aq) and (0.864 NaCl + 0.136 KCl)(aq) (reported previously²). A good linear correlation is observed at every pressure. The results for both MgCl₂(aq) and Na₂SO₄(aq) also obey the same relation.

At a molecular level, the explanation for the observed behaviour may be that the ions, being excluded from the CO₂-rich phase, have a negative affinity for the interface and are restricted to the bulk of the aqueous phase. The resulting gradient in ionic strength near the interface leads to an enhanced attraction of the water molecules towards the bulk of the aqueous phase, thereby increasing the work required to expand the interfacial area and amplifying γ . This effect is expected to increase with both ion concentration and ion charge. The special ion effect on surface tension has been studied for brine systems under ambient condition by several authors.^{14,}

Empirical equation. In our previous work,² we developed an empirical correlation for interfacial tension based on the results for (0.864 NaCl + 0.136 KCl)(aq) in which γ was represented as a linear function of total salt molality with coefficients that dependent upon T and p . We now apply the same function, with the same parameters, but generalize it by replacing total salt molality m by the positive-charge molality m^+ . The correlation is therefore written

$$\gamma/(\text{mN} \cdot \text{m}^{-1}) = A[m^+ / (\text{mol} \cdot \text{kg}^{-1})] + B \quad (4)$$

where A is a simple linear function of T and p ,

$$A = a_0 + a_1(p/\text{MPa}) + a_2(T/\text{K}), \quad (5)$$

and B , representing $\gamma/(\text{mN} \cdot \text{m}^{-1})$ for the ($\text{CO}_2 + \text{H}_2\text{O}$) system, is given by

$$B = [b_{00} + b_{01}(T/\text{K}) + b_{02}(T/\text{K})^2] + [b_{10} + b_{11}(T/\text{K})](p/\text{MPa})^{-1} + [b_{20} + b_{21}(T/\text{K})](p/\text{MPa})^{-2} + [b_{30} + b_{31}(T/\text{K})](p/\text{MPa})^{-3}. \quad (6)$$

The parameters appearing in equation (4) and (5) are given in table 8, and the ranges of validity for the correlation are $323.15 \leq T/\text{K} \leq 448.15$ and $p \leq 50 \text{ MPa}$.

Eq. (4) was used, without further adjustment, to predict the IFT of the brines investigated in this work and the relative deviations are shown in figure 7. The average absolute relative deviation for the entire data set is 2% with the majority of the data predicted within $\pm 3\%$. Thus we deduce that Eq. (4) may be useful in predicting the IFT of other ($\text{CO}_2 + \text{brine}$) systems at positive-charge molalities up to about $10 \text{ mol} \cdot \text{kg}^{-1}$, subject to the restrictions on temperature and pressure noted above.

Conclusions

We have measured the IFT for $\text{CO}_2 + \text{CaCl}_2(\text{aq})$, $\text{CO}_2 + \text{MgCl}_2(\text{aq})$ and $\text{CO}_2 + \text{Na}_2\text{SO}_4(\text{aq})$ over wide ranges of temperature, pressure and molality. The results clearly demonstrate a linear dependence of γ upon the positive-charge molality m^+ in the brine over the whole range investigated. The results, together with previous data for the mixed salt system (0.864 NaCl + 0.136 KCl)(aq), all conform to a simple empirical correlation which may be useful as a predictive tool for other brine systems.

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Figure 1. Interfacial tension of the $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at $T = 373$ K as a function of pressure: ■, this work at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$; ○, Aggelopoulos *et al.*³ at $m = 2.7 \text{ mol}\cdot\text{kg}^{-1}$; △, Aggelopoulos *et al.*³ at $m = 1.8 \text{ mol}\cdot\text{kg}^{-1}$.

Figure 2. Interfacial tension of $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at $m = 2.5 \text{ mol} \cdot \text{kg}^{-1}$ as a function of pressure on different isotherms: ▲, $T = 343.15 \text{ K}$; ✕, $T = 373.15 \text{ K}$; □, $T = 393.15 \text{ K}$; ○, $T = 423.15 \text{ K}$.

Figure 3. Interfacial tension of $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at $m = 2.5 \text{ mol} \cdot \text{kg}^{-1}$, as a function of temperature at different pressures: ◆, $p = 2 \text{ MPa}$; ■, $p = 4 \text{ MPa}$; ▲, $p = 6 \text{ MPa}$; ×, $p = 8 \text{ MPa}$; *, $p = 10 \text{ MPa}$; ●, $p = 12 \text{ MPa}$; +, $p = 14 \text{ MPa}$; ◇, $p = 18 \text{ MPa}$; □, $p = 20 \text{ MPa}$; △, $p = 30 \text{ MPa}$; ○, $p = 50 \text{ MPa}$.

Figure 4. Interfacial tension as a function of time at $T = 423.15$ K and $p = 2$ MPa: ●, $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$; □, $\text{CO}_2 + (0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at $m = 4.95 \text{ mol}\cdot\text{kg}^{-1}$.²

Figure 5. Interfacial tension between CO_2 and three different brines as a function of pressure on different isotherms: (a), $T = 343.15 \text{ K}$; (b), $T = 423.15 \text{ K}$. Δ , $\text{CaCl}_2(\text{aq})$ at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$; $\circ$, $(0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at $m = 4.95 \text{ mol}\cdot\text{kg}^{-1,2}$; $\square$, $\text{MgCl}_2(\text{aq})$ at $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$.

Figure 6. Interfacial tension of (CO₂ + brine) as a function of the positive charge molality at $T = 373.15$ K: ■, $p = 2$ MPa; ◆, $p = 4$ MPa; ▲, $p = 6$ MPa; +, $p = 10$ MPa; ●, $p = 12$ MPa; □, $p = 14$ MPa; ×, $p = 16$ MPa; △, $p = 18$ MPa; ○, $p = 20$ MPa; *, $p = 30$ MPa; ◇, $p = 40$ MPa; +, $p = 50$ MPa. Black symbols refer to (0.864 NaCl + 0.136 KCl)(aq)² and red symbols refer to CaCl₂(aq).

Figure 7. Deviations $\Delta\gamma$ of the interfacial tension γ of different (CO_2 + brine) systems from the predictions of equation (3) at temperatures between (343 and 423) K: ○, $\text{CaCl}_2(\text{aq})$ and $\text{MgCl}_2(\text{aq})$ at $m^+ = 5.0 \text{ mol}\cdot\text{kg}^{-1}$; ●, $\text{CaCl}_2(\text{aq})$ and $\text{MgCl}_2(\text{aq})$ at $m^+ = 10.0 \text{ mol}\cdot\text{kg}^{-1}$; △, $\text{Na}_2\text{SO}_4(\text{aq})$ at $m^+ = 0.98 \text{ mol}\cdot\text{kg}^{-1}$; ▲, $\text{Na}_2\text{SO}_4(\text{aq})$ at $m^+ = 1.96 \text{ mol}\cdot\text{kg}^{-1}$.

Table 1. Total salt molalities m , temperatures T and pressures p studied in this work.

Salt type	$m/(\text{mol}\cdot\text{kg}^{-1})$	T/K	p/MPa
CaCl_2	2.50	343, 373, 393, 423	2, 4, 6, 8, 10, 12, 14, 16, 18, 20, 25, 30, 35, 40, 45, 50
	5.00	343, 373, 393, 423	
MgCl_2	2.50	343, 373, 393, 423	
	5.00	343	
Na_2SO_4	0.49	373	2, 4, 6, 8, 10, 12, 14, 16
	0.98	343	

Table 2. Interfacial tension γ for $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at temperatures T , pressures p and molality $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
343.15	2	1134.2	65.7	393.15	2	1108.3	60.6
343.15	4	1096.9	59.6	393.15	4	1078.9	56.9
343.15	6	1052.2	53.6	393.15	6	1046.7	53.5
343.15	8	996.2	48.5	393.15	8	1011.5	49.6
343.15	10	922.9	44.8	393.15	10	972.8	46.9
343.15	12	825.6	42.4	393.15	12	930.7	44.8
343.15	14	715.5	41.3	393.15	14	885.4	42.9
343.15	16	625.2	40.7	393.15	16	838.0	41.4
343.15	18	561.4	40.2	393.15	18	790.0	39.8
343.15	20	515.3	39.7	393.15	20	743.2	38.9
343.15	25	439.2	38.3	393.15	25	640.7	36.6
343.15	30	389.9	37.4	393.15	30	562.9	35.4
343.15	35	353.4	36.9	393.15	35	504.5	34.7
343.15	40	324.5	36.5	393.15	40	459.0	33.8
343.15	45	300.5	35.8	393.15	45	422.2	33.1
343.15	50	279.9	35.2	393.15	50	391.7	32.5
373.15	2	1119.7	62.2	423.15	2	1089.4	55.1
373.15	4	1087.7	58.2	423.15	4	1063.2	52.4
373.15	6	1051.8	54.2	423.15	6	1035.3	49.5
373.15	8	1011.0	50.0	423.15	8	1005.5	47.0
373.15	10	964.6	46.8	423.15	10	973.9	45.2
373.15	12	911.9	44.4	423.15	12	940.6	42.9
373.15	14	853.5	42.5	423.15	14	905.8	41.1
373.15	16	791.9	40.6	423.15	16	869.8	39.8
373.15	18	731.5	39.6	423.15	18	833.3	38.8
373.15	20	676.5	38.9	423.15	20	797.0	37.6
373.15	25	570.5	37.2	423.15	25	710.7	35.0
373.15	30	498.9	36.0	423.15	30	636.3	33.5
373.15	35	447.3	35.1	423.15	35	575.1	32.1
373.15	40	407.6	34.5	423.15	40	525.1	31.1
373.15	45	375.6	34.0	423.15	45	483.9	30.6
373.15	50	349.0	33.5	423.15	50	449.2	29.8

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 3. Interfacial tension γ for $\text{CO}_2 + \text{CaCl}_2(\text{aq})$ at temperatures T , pressures p and molality $m = 5.0 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
343.15	2	1273.9	74.8	393.15	2	1247.3	69.2
343.15	4	1236.3	68.7	393.15	4	1217.6	65.5
343.15	6	1191.2	63.0	393.15	6	1185.1	62.1
343.15	8	1135.0	57.0	393.15	8	1149.5	58.4
343.15	10	1061.4	50.9	393.15	10	1110.6	55.5
343.15	12	964.0	49.9	393.15	12	1068.2	53.0
343.15	14	853.8	49.3	393.15	14	1022.8	50.9
343.15	16	763.2	48.6	393.15	16	975.1	49.5
343.15	18	699.3	48.0	393.15	18	926.9	48.8
343.15	20	653.1	47.2	393.15	20	880.0	48.0
343.15	25	576.7	46.5	393.15	25	777.1	46.2
343.15	30	527.2	45.6	393.15	30	699.0	44.5
343.15	35	490.5	44.5	393.15	35	640.2	43.7
343.15	40	461.4	44.2	393.15	40	594.4	42.9
343.15	45	437.1	43.9	393.15	45	557.4	42.4
343.15	50	416.3	43.6	393.15	50	526.6	42.3
373.15	2	1258.6	72.0	423.15	2	1229.5	64.9
373.15	4	1226.2	67.4	423.15	4	1202.8	62.2
373.15	6	1189.9	63.1	423.15	6	1174.5	58.7
373.15	8	1148.9	58.6	423.15	8	1144.4	54.9
373.15	10	1102.3	55.1	423.15	10	1112.5	52.3
373.15	12	1049.4	52.0	423.15	12	1078.9	50.7
373.15	14	990.7	50.5	423.15	14	1043.8	49.1
373.15	16	929.0	49.4	423.15	16	1007.7	48.2
373.15	18	868.5	48.9	423.15	18	970.9	47.4
373.15	20	813.3	48.2	423.15	20	934.4	46.7
373.15	25	707.0	46.6	423.15	25	847.6	44.9
373.15	30	635.1	45.5	423.15	30	772.8	43.4
373.15	35	583.2	44.8	423.15	35	711.2	42.4
373.15	40	543.2	44.2	423.15	40	660.9	41.6
373.15	45	511.0	43.1	423.15	45	619.3	41.1
373.15	50	484.1	42.4	423.15	50	584.2	40.8

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 4. Interfacial tension γ for $\text{CO}_2 + \text{MgCl}_2(\text{aq})$ at temperatures T , pressures p and molality $m = 2.5 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
343.15	2	1112.3	65.5	393.15	2	1090.5	60.6
343.15	4	1074.8	59.6	393.15	4	1060.7	56.9
343.15	6	1030.3	53.9	393.15	6	1028.9	53.3
343.15	8	974.3	49.3	393.15	8	993.6	50.3
343.15	10	901.0	45.5	393.15	10	954.9	47.2
343.15	12	803.6	42.9	393.15	12	912.8	44.5
343.15	14	693.6	41.6	393.15	14	867.5	42.5
343.15	16	603.1	40.7	393.15	16	820.0	40.7
343.15	18	539.3	39.7	393.15	18	771.9	39.4
343.15	20	493.2	39.3	393.15	20	725.1	38.4
343.15	25	417.0	38.2	393.15	25	622.5	36.1
343.15	30	367.6	37.2	393.15	30	544.7	35.4
343.15	35	331.1	36.7	393.15	35	486.2	34.0
343.15	40	302.1	36.1	393.15	40	440.6	33.4
343.15	45	278.0	35.7	393.15	45	403.8	33.1
343.15	50	257.4	35.1	393.15	50	373.2	32.2
373.15	2	1100.3	62.5	423.15	2	1074.1	55.9
373.15	4	1068.0	58.5	423.15	4	1047.5	53.0
373.15	6	1032.3	54.3	423.15	6	1019.8	50.6
373.15	8	991.5	50.4	423.15	8	990.0	47.9
373.15	10	945.1	47.3	423.15	10	958.4	45.0
373.15	12	892.4	43.9	423.15	12	925.1	42.9
373.15	14	833.9	41.7	423.15	14	890.2	41.1
373.15	16	772.3	40.2	423.15	16	854.3	39.3
373.15	18	711.9	38.9	423.15	18	817.8	38.1
373.15	20	656.9	38.1	423.15	20	781.4	37.0
373.15	25	550.7	37.2	423.15	25	695.0	35.3
373.15	30	479.0	35.3	423.15	30	620.5	33.6
373.15	35	427.3	34.4	423.15	35	559.2	32.2
373.15	40	387.6	34.1	423.15	40	509.2	31.5
373.15	45	355.6	33.4	423.15	45	467.8	30.8
373.15	50	328.9	32.9	423.15	50	433.0	30.1

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 5. Interfacial tension γ for $\text{CO}_2 + \text{MgCl}_2(\text{aq})$ at temperatures T , pressures p and molality $m = 5.0 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
343.15	2	1247.2	76.1	343.15	18	671.9	48.4
343.15	4	1209.5	69.9	343.15	20	625.6	47.5
343.15	6	1164.3	60.6	343.15	25	549.0	46.1
343.15	8	1108.0	56.7	343.15	30	499.3	45.2
343.15	10	1034.4	51.3	343.15	35	462.4	44.4
343.15	12	936.8	50.0	343.15	40	433.1	44.2
343.15	14	826.6	49.6	343.15	45	408.6	44.1
343.15	16	735.9	48.6	343.15	50	387.6	43.6

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 6. Interfacial tension γ for $\text{CO}_2 + \text{Na}_2\text{SO}_4(\text{aq})$ at temperatures T , pressures p and molality $m = 0.49 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
373.15	2	993.3	54.6	373.15	18	610.4	33.1
373.15	4	962.3	49.7	373.15	20	555.8	32.2
373.15	6	927.3	45.8	373.15	25	450.5	31.0
373.15	8	887.3	40.1	373.15	30	379.7	28.9
373.15	10	841.5	37.5	373.15	35	328.8	28.9
373.15	12	789.4	37.1	373.15	40	289.8	27.8
373.15	14	731.5	35.6	373.15	45	258.5	27.4
373.15	16	670.3	34.3	373.15	50	232.5	27.0

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 7. Interfacial tension γ for $\text{CO}_2 + \text{Na}_2\text{SO}_4(\text{aq})$ at temperatures T , pressures p and molality $m = 0.98 \text{ mol}\cdot\text{kg}^{-1}$.

T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	T/K	p/MPa	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$
343.15	2	1057.2	60.6	343.15	10	848.1	39.0
343.15	4	1020.7	54.1	343.15	12	751.1	36.7
343.15	6	976.5	47.8	343.15	14	641.4	36.2
343.15	8	921.0	42.2	343.15	16	551.3	35.3

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$ and $u(\gamma) = 0.008\cdot\gamma$.

Table 8. Parameters in equations (3) and (4) for the correlation of the interfacial tension.²

a_0	4.51010×10^{-01}	b_{10}	$-5.3890 \times 10^{+02}$
a_1	6.2018×10^{-03}	b_{11}	$2.0305 \times 10^{+00}$
a_2	3.3654×10^{-03}	b_{20}	$3.8312 \times 10^{+03}$
b_{00}	$-4.1203 \times 10^{+01}$	b_{21}	$-1.1694 \times 10^{+01}$
b_{01}	4.3549×10^{-01}	b_{30}	$-5.1659 \times 10^{+03}$
b_{02}	-7.2502×10^{-04}	b_{31}	$1.5073 \times 10^{+01}$