

Correction to the 'Interfacial Tension of (Brines + CO₂): (0.864 NaCl + 0.136 KCl) at Temperatures between (298 and 448) K, Pressures between (2 and 50) MPa, and Total Molalities of (1 to 5) mol·kg⁻¹ by Xuesong Li, Edo Boek, Geoffrey C. Maitland, and J. P. Martin Trusler published in the Journal of Chemical Engineering Data 2012, 57, 1078–1088

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Introduction

Li *et al.* [1] reported interfacial tension measurements between carbon dioxide and the mixed brine (0.864 NaCl + 0.136 KCl) over wide ranges of temperature, pressure and total salt molality. We have determined that their data on the isotherm at 298.15 K for the salt molaity of 0.98 mol·kg⁻¹ are erroneous; results at other temperatures and salt molalities reported in [1] are not affected by the error. We report herein new data, measured at $T = 298.15$ K and at pressures between (2 and 51) MPa, to replace the corresponding isotherm reported in Table 2 of the original reference.

Experimental Section

Apparatus.

As in the original work [1], we made use of the pendant-drop method for carrying out interfacial tension (IFT) measurements. The apparatus, which is a new version of the equipment described in [1], is shown schematically in Figure 1. Standard uncertainties were 0.025 K for temperature and 35 kPa for pressure.

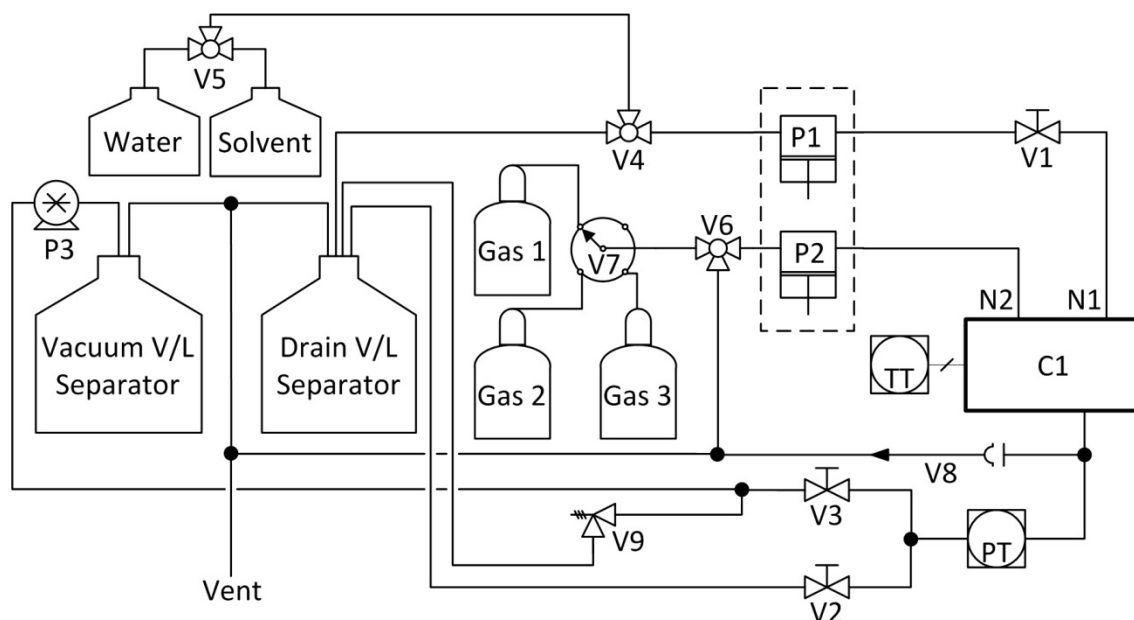


Figure 1. Interfacial tension apparatus, where the CO₂ liquid dip-tube cylinder provides pressurised pure CO₂ to the view cell. C1: optical cell with stirrer; P1, P2: high-pressure Quizix pumps with cooling jacket; P3: vacuum pump; TT: platinum resistance thermometer (Pt100); PT: flow-through pressure transducer; N1, N2: injection ports; V1, V2, V3: high-pressure valves; V4, V5, V6: three-way valves; V7: four-way switch; V8: rupture-disc safety head; V9: relief valve.

Materials.

The source and purity of the chemical samples used are given in Table 1. Prior to preparing the brine solutions, the salts were dried in an oven at $T = 373.15$ K. Solution was prepared gravimetrically, with the relative uncertainties in mass below 0.01 %. The total salt molality of the solution studied was $0.98 \text{ mol}\cdot\text{kg}^{-1}$, with composition $[(1 - y) \text{ NaCl} + y \text{ KCl}](\text{aq})$, where $y = 0.136$ is the mole fraction of KCl in the salt. The brine solutions were degassed under vacuum for 15 min before use.

Table 1. Description of chemical samples, where x is mole fraction, w is mass fraction and ρ_e is electrical resistivity.

Chemical Name	Source	Purity as Supplied	Additional Purification
Carbon dioxide	BOC	$x \geq 0.99995$ ^a	None
Sodium chloride	Sigma Aldrich	$w \geq 0.995$ ^a	Oven dried
Potassium chloride	Sigma Aldrich	$w \geq 0.99$ ^a	Oven dried
Water	Millipore Direct-Q UV3	$\rho_e > 18 \text{ M}\Omega\cdot\text{cm}$ at $T = 298 \text{ K}$	Degassed

^a Purities are as stated by the supplier.

Validation.

The pendant drop method was validated by measurement of the surface tension of water at $T = 298.11$ K. The measured value was $(71.97 \pm 0.1) \text{ mN}\cdot\text{m}^{-1}$, in close agreement with the value $(71.98 \pm 0.05) \text{ mN}\cdot\text{m}^{-1}$ obtained from the IAPWS recommended correlation by means of the REFPROP 9.1 database [2]. (Brine + CO₂) interfacial tensions were also measured at $T = 373.15$ K, and three pressures between (2 and 20) MPa for validation; the results, reported in Table 2, were in good agreement with those reported by Li et al. [1].

Table 2. Interfacial tension γ for CO₂ + (0.864 NaCl + 0.136 KCl)(aq) at temperature $T = 373.15$ K, pressures p , and total salt molality $m = 0.98 \text{ mol}\cdot\text{kg}^{-1}$.^a Also given is the calculated density difference $\Delta\rho$ between the aqueous and non-aqueous phases, the interfacial tension value γ_0 from Li et al. [1] at the same conditions, and the difference $\Delta\gamma = \gamma - \gamma_0$.

p/MPa	T/K	$\Delta\rho/(\text{kg}\cdot\text{m}^{-3})$	$\gamma/(\text{mN}\cdot\text{m}^{-1})$	$\gamma_0/(\text{mN}\cdot\text{m}^{-1})$	$\Delta\gamma/\gamma_0$
2.0	373.19	968.13	55.51	55.40	0.2 %
10.0	373.23	815.04	39.98	39.70	0.7 %
20.0	373.51	528.65	32.65	31.90	2.4 %

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

Data Analysis.

The interfacial tension was evaluated as described in the original work [1] with the exception that the density difference between the coexisting phases was calculated according to the slightly revised method described by Chow et al. [3; 4].

Results

IFT measurements were made for carbon dioxide and the brine of total molality $0.98 \text{ mol}\cdot\text{kg}^{-1}$ at $T = (298.13 \pm 0.06) \text{ K}$ over a range of pressures from (2 to 51) MPa. The results are given in Table 3 and Figure 2. The standard relative uncertainties $u_r(\gamma)$ of the interfacial tensions were estimated from the relation:

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

The relative standard deviation $\sigma(\gamma)/\gamma$ of the IFT data at each state point was evaluated from 3 repeated measurements and the average was found to be 0.3 %; in all cases it was < 1.2 %. The relative uncertainty of molality was most likely limited by the purity of the salts and was estimated to be 0.5 %. The overall combined relative uncertainty of interfacial tension of all state points is 0.9 %, and the expanded relative uncertainty at 95 % confidence is 1.8 %.

Table 3. Interfacial tension γ for $\text{CO}_2 + (0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at temperatures T , pressures p , and total salt molality $m = 0.98 \text{ mol}\cdot\text{kg}^{-1}$.^a Also given is the calculated density difference $\Delta\rho$ between the aqueous and non-aqueous phases.

p/MPa	T/K	$\Delta\rho/\text{kg}\cdot\text{m}^{-3}$	$\gamma/\text{mN}\cdot\text{m}^{-1}$	p/MPa	T/K	$\Delta\rho/\text{kg}\cdot\text{m}^{-3}$	$\gamma/\text{mN}\cdot\text{m}^{-1}$
2.0	298.12	1001.1	60.5	12.5	298.14	199.6	34.9
4.1	298.10	950.0	49.2	16.5	298.15	164.2	34.6
5.0	298.10	915.2	44.1	20.4	298.15	137.9	34.2
6.0	298.10	856.3	38.3	31.2	298.15	88.4	33.4
8.3	298.19	264.3	35.8	41.5	298.16	55.2	32.9
10.3	298.17	227.9	35.3	50.9	298.14	30.9	31.1

^a Standard uncertainties u are $u(T) = 0.025 \text{ K}$, $u(p) = 35 \text{ kPa}$, and $u(\gamma) = 0.009\gamma$.

In Figure 2, we compare the new measurements with the original data [1] and the latter are seen to be substantially too low. The new data, extrapolated to zero pressure, give a brine surface tension of $71.7 \text{ mN}\cdot\text{m}^{-1}$, which compares with the value of $73.7 \text{ mN}\cdot\text{m}^{-1}$ obtained by combining the surface tension of pure water [2] with the salt effect reported by Pegram and Record [5]. This comparison serves as additional validation of the new measurements.

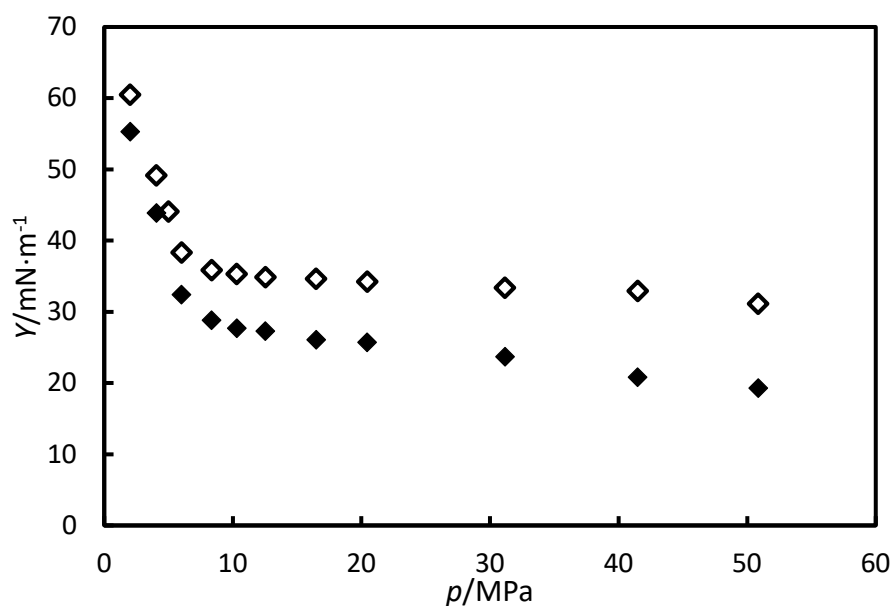


Figure 2. Interfacial tension γ for $\text{CO}_2 + (0.864 \text{ NaCl} + 0.136 \text{ KCl})(\text{aq})$ at total salt molality of $0.98 \text{ mol}\cdot\text{kg}^{-1}$, $T = 298 \text{ K}$ and various pressures p : \diamond , this work; \blacklozenge , Li et al. [1].

Acknowledgements

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