

Supplementary material

Measurements and predictions of densities and viscosities in CO₂ + hydrocarbon mixtures at high pressures and temperatures: CO₂ + *n*-pentane and CO₂ + *n*-hexane blends.

Alejandro Moreau^a, Ilya Polishuk^b, José J. Segovia^a, Dirk Tuma^c, David Vega-Maza^a, M. Carmen Martín^{a,*}

^a TermoCal Research Group, Research Institute on Bioeconomy, University of Valladolid, Paseo del Cauce 59, 47011 Valladolid, Spain.

^b Department of Chemical Engineering, Ariel University, Ariel, 40700, Israel

^c BAM Federal Institute for Materials Research and Testing, D-12200 Berlin, Germany

* corresponding author, e-mail: mcarmen.martin@eii.uva.es

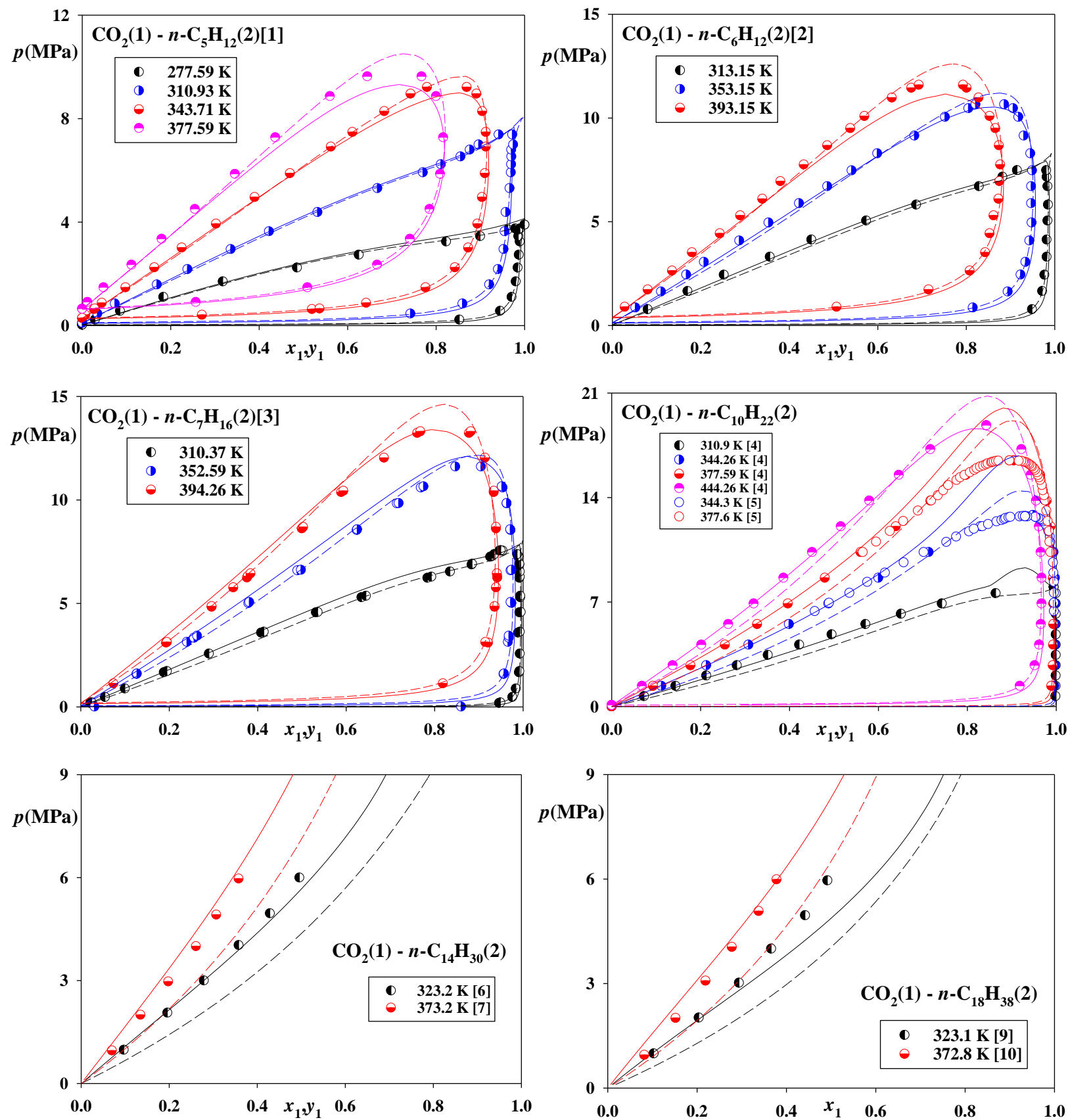


Figure S1. Phase equilibria in the systems $\text{CO}_2(1) - n\text{-alkanes}(2)$. Points – experimental data [1-10].

Solid lines – predictions of CP-PC-SAFT with $k_{12} = 0.09$. Dotted lines – predictions of PC-SAFT with $k_{12} = 0.12$.

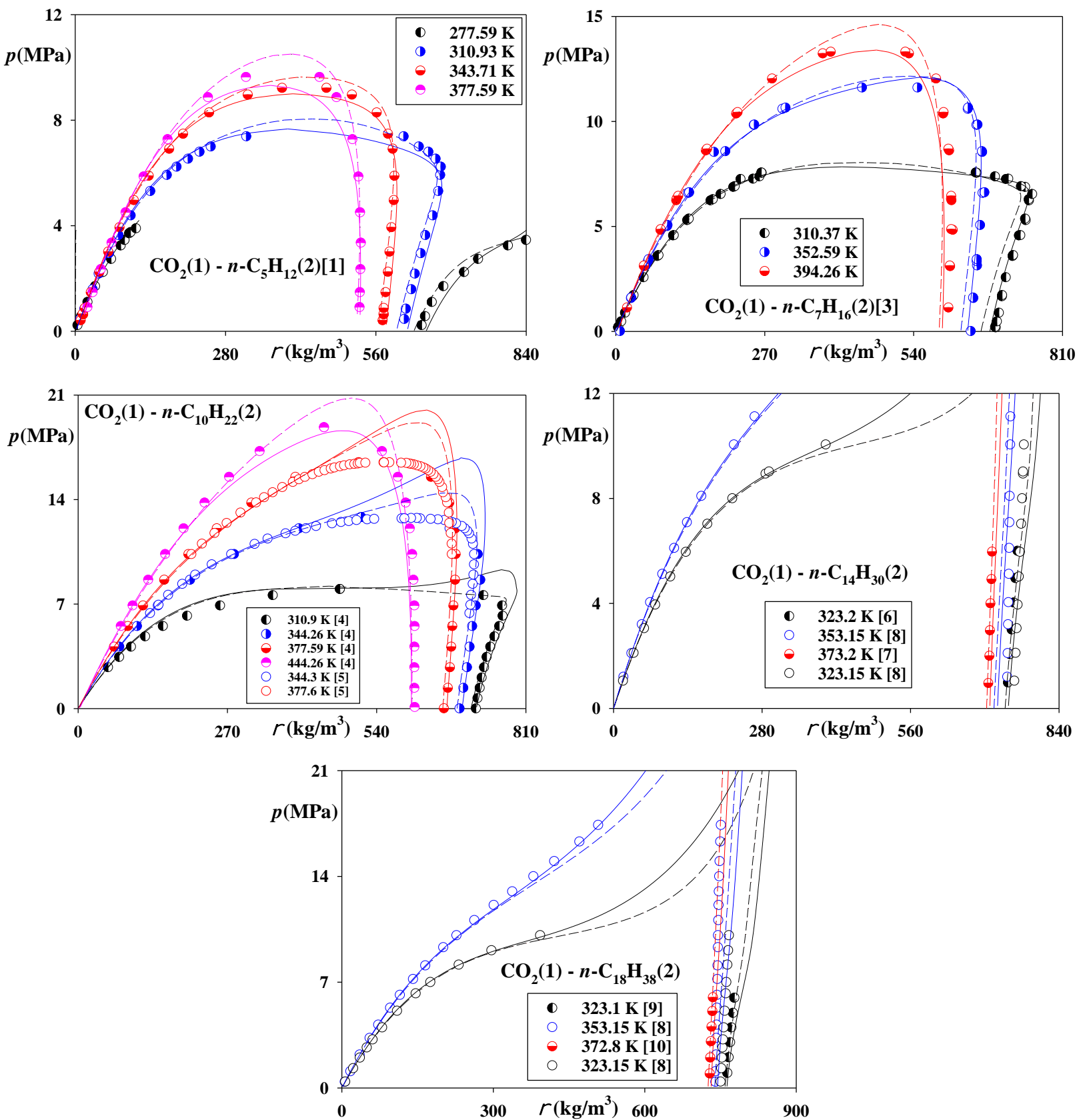


Figure S2. Saturated phase densities of the systems $\text{CO}_2(1) - n\text{-alkanes}(2)$. Points – experimental data [1,3-10].

Solid lines – predictions of CP-PC-SAFT with $k_{12} = 0.09$. Dotted lines – predictions of PC-SAFT with $k_{12} = 0.12$.

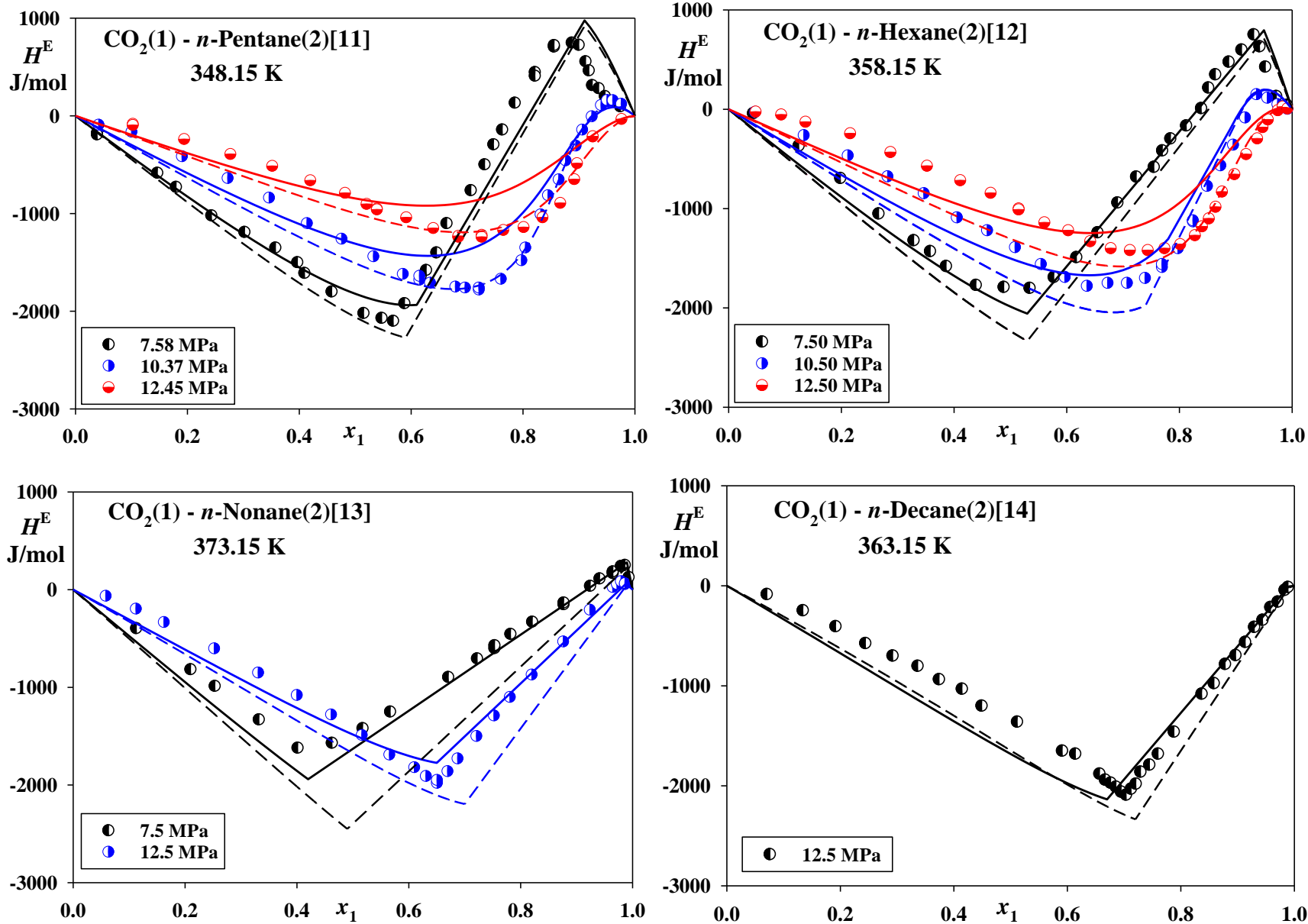


Figure S3. Excess enthalpies in the systems CO₂(1) – *n*-alkanes(2). Points – experimental data [11-14].

Solid lines – predictions of CP-PC-SAFT with $k_{12} = 0.09$. Dotted lines – predictions of PC-SAFT with $k_{12} = 0.12$.

Table S1. Absolute Average Deviations% in predicting densities and speeds of sound by CP-PC-SAFT ($k_{12} = 0.09$) and PC-SAFT ($k_{12} = 0.12$).

System	Property	AAD%	AAD%	x_1 range	T range (K)	p range (MPa)	Number of points	Ref.
		CP-PC-SAFT	PC-SAFT					
CO ₂ (1) – <i>n</i> -pentane(2)	density	1.9	0.6	0 – 0.5999	273.15 – 373.15	0.1 – 100	461	This work
CO ₂ (1) – <i>n</i> -hexane(2)	density	1.3	0.7	0 – 0.5999	273.15 – 373.15	0.1 – 100	462	This work
CO ₂ (1) – <i>n</i> -heptane(2)	density	1.4	0.4	0 – 0.8849	303.35 – 313.25	10.11 – 70.62	84	[15]
CO ₂ (1) – <i>n</i> -decane(2)	density	1.4	1.0	0.207 – 0.73	313.15 – 410.15	3.4 – 76.4	49	[16]
CO ₂ (1) – <i>n</i> -decane(2)	density	1.4	1.2	0.1182 – 0.9539	318.15 – 358.15	10 – 130	294	[17]
CO ₂ (1) – <i>n</i> -decane(2)	density	0.9	0.8	0 – 0.951	283.15 – 398.15	0.1 – 130	274	[18]
CO ₂ (1) – <i>n</i> -decane(2)	density	0.9	0.4	0.3011 – 0.9517	283.15 – 393.15	10 – 100	299	[19]
CO ₂ (1) – <i>n</i> -dodecane(2)	density	1.7	1.0	0.1014 – 0.6022	283.15 – 393.15	10 – 100	299	[19]
CO ₂ (1) – <i>n</i> -dodecane(2)	density	1.3	0.5	0 – 0.9877	303.15 – 313.15	10 – 70	126	[20]
CO ₂ (1) – <i>n</i> -pentadecane(2)	density	1.3	0.8	0.376 – 0.757	313.15 – 410.15	6.36 – 76.37	47	[21]
CO ₂ (1) – <i>n</i> -hexadecane(2)	density	1.1	1.2	0 – 0.727	298.14 – 473.85	0.61 – 120.86	267	[22]
CO ₂ (1) – <i>n</i> -heptadecane(2)	density	0.9	0.4	0 – 0.98	303.15 – 333.15	10 – 70	194	[23]
CO ₂ (1) – <i>n</i> -heptane(2)	speed of sound	1.6	8.2	0 – 0.885	303.35 – 313.25	10.11 – 70.62	79	[15]
CO ₂ (1) – <i>n</i> -dodecane(2)	speed of sound	2.6	11.2	0 – 0.85278	303.15 – 313.15	10 – 70	84	[20]
CO ₂ (1) – <i>n</i> -heptadecane(2)	speed of sound	3.1	13.8	0 – 0.85	303.15 – 333.15	10 – 70	149	[23]

References

- [1] G. J. Besserer, D. B. Robinson, Equilibrium-phase properties of *n*-pentane-carbon dioxide system. J Chem. Eng. Data 18 (1973) 416–419.
- [2] Y.-H. Li, K. H. Dillard, R. L. Robinson Jr. Vapor-liquid phase equilibrium for carbon dioxide-*n*-hexane at 40, 80, and 120 °C. J Chem. Eng. Data 26 (1981) 53–55.
- [3] H. Kalra, H. Kubota, D. B. Robinson, H.-Joo Ng. Equilibrium phase properties of the carbon dioxide-*n*-heptane system. J. Chem. Eng. Data 23 (1978) 317–321.
- [4] H. H. Reamer, B. H. Sage. Phase equilibria in hydrocarbon systems. Volumetric and phase behavior of the *n*-decane-CO₂ system. J Chem. Eng. Data 8 (1963) 508–513.
- [5] N. Nagarajan, R. L. Robinson, Jr. Equilibrium phase compositions, phase densities, and interfacial tensions for CO₂ + hydrocarbon systems. 2. CO₂ + *n*-decane. J Chem. Eng. Data 31 (1986) 168–171.
- [6] M. Kariznovi, H. Nourozieh, J. Abedi. Phase composition and saturated liquid properties in binary and ternary systems containing carbon dioxide, *n*-decane, and *n*-tetradecane. J. Chem. Thermodyn. 57 (2013) 189–196.
- [7] H. Nourozieh, M. Kariznovi, J. Abedi. Measurement and correlation of saturated liquid properties and gas solubility for decane, tetradecane and their binary mixtures saturated with carbon dioxide. Fluid Phase Equilib. 337 (2013) 246–254.
- [8] N. Li, C. W. Zhang, Q. L. Ma, L. Y. Jiang, Y. X. Xu, G.-J. Chen, C. Y. Sun, L. Y. Yang. Interfacial tension measurement and calculation of (carbon dioxide + *n*-alkane) binary mixtures. J Chem. Eng. Data 62 (2017) 2861–2871.
- [9] H. Nourozieh, B. Bayestehparvin, M. Kariznovi, J. Abedi. Equilibrium properties of (carbon dioxide + *n*-decane + *n*-octadecane) systems: experiments and thermodynamic modeling. J Chem. Eng. Data 58 (2013) 1236–1243.

- [10] M. Kariznovi, H. Nourozieh, J. Abedi, Experimental results and thermodynamic investigation of carbon dioxide solubility in heavy liquid hydrocarbons and corresponding phase properties. *Fluid Phase Equilib.* 339 (2013) 105–111.
- [11] J. J. Christensen, P. W. Faux, D. Cordray, R. M. Izatt. The excess enthalpies of (carbon dioxide + pentane) at 348.15, 373.15, 413.15, 470.15, and 573.15 K from 7.58 to 12.45 MPa. *J. Chem. Thermodyn.* 18 (1986) 1053–1064.
- [12] J. J. Christensen, T. A. C. Walker, R. S. Schofield, P. W. Faux, P. R. Harding, R. M. Izatt. The excess enthalpies of (carbon dioxide+hexane) at 308.15, 358.15, and 413.15 K from 7.50 to 12.50 MPa. *J. Chem. Thermodyn.* 16 (1984) 445–451.
- [13] D. R. Cordray, L. D. Gunderson, J. J. Christensen, J. L. Oscarson, R. M. Izatt. The excess molar enthalpies of $[x\text{CO}_2 + (1 - x)\{0.5\text{CH}_3(\text{CH}_2)_4\text{CH}_3 + 0.5\text{CH}_3(\text{CH}_2)_{10}\text{CH}_3\}]$ at the temperatures (308.15, 323.15, 373.15, and 413.15) K and at the pressures 7.50 MPa and 12.50 MPa, and of $\{x\text{CO}_2 + (1 - x)\text{CH}_3(\text{CH}_2)_7\text{CH}_3\}$ at 373.15 K and 7.50 MPa and 12.50 MPa. *J. Chem. Thermodyn.* 23 (1991) 941–949.
- [14] J. J. Christensen, D. Cordray, R. M. Izatt. The excess enthalpies of (carbon dioxide+decane) from 293.15 to 573.15 K at 12.50 MPa. *J. Chem. Thermodyn.* 18 (1986) 53–61.
- [15] J.-P. Bazile, D. Nasri, A. W. S. Hamani, G. Galliero, J.-L. Daridon. Excess volume, isothermal compressibility, isentropic compressibility and speed of sound of carbon dioxide + *n*-heptane binary mixture under pressure up to 70 MPa. I Experimental Measurements. *J. Supercrit. Fl.* 140 (2018) 218–232.
- [16] M. E. Kandil, N. M. Al-Saifi, A. S. Sultan. Simulation and measurements of volumetric and phase behavior of carbon dioxide + higher alkanes at high pressure: $\text{CO}_2 + n$ -decane at temperatures (313–410) K and pressures up to 76 MPa. *Int. J. Greenh. Gas Cont.* 53 (2016) 198–206.

- [17] A. M. Chacon Valero, F. X. Feitosa, H. B. de Sant'Ana. Density and volumetric behavior of binary CO₂ + *n*-decane and ternary CO₂ + *n*-decane + naphthalene systems at high pressure and high temperature. *J. Chem. Eng. Data* 65 (2020) 3499–3509.
- [18] O. Fandiño, L. Lugo, J. J. Segovia, E. R. López, M. J. P. Comuñas, J. Fernández. High pressure densities of carbon dioxide + dipentaerythritol hexaheptanoate: New experimental setup and volumetric behavior. *J. Supercrit. Fl.* 58 (2011) 189–197.
- [19] J. Zambrano, F. V. Gómez-Soto, D. Lozano-Martín, M. Carmen Martín, J. J. Segovia, Volumetric behaviour of (carbon dioxide + hydrocarbon) mixtures at high pressures. *J. Supercrit. Fl.* 110 (2016) 103–109.
- [20] J.-P. Bazile, D. Nasri, A. W. S. Hamani, G. Galliero, J.-L. Daridon. Density, speed of sound, compressibility, and excess properties of carbon dioxide + *n*-dodecane binary mixtures from 10 to 70 MPa. *J. Chem. Eng. Data* 64 (2019) 3187–3204.
- [21] M. E. Kandil. Measurements and modeling of volumetric and phase behavior of carbon dioxide + higher alkanes: CO₂ + *n*-Pentadecane at temperatures 313 to 410 K and pressures up to 77 MPa. *J. Chem. Eng. Data* 63 (2018) 1389–1396.
- [22] M. Mohammed, F. Ciotta, J. P. Martin Trusler. Viscosities and densities of binary mixtures of hexadecane with dissolved methane or carbon dioxide at temperatures from (298 to 473) K and at pressures up to 120 MPa. *J. Chem. Eng. Data* 62 (2017) 422–439.
- [23] J.-P. Bazile, D. Nasri, G. Galliero, J.-L. Daridon. Density, speed of sound, compressibility, and excess properties of the carbon dioxide + *n*-heptadecane binary mixture from 10 to 70 MPa. *J. Chem. Eng. Data* 66 (2021) 3245–3257.