JOURNAL OF CHEMICAL & ENGINEERING DATA

REVIEW

pubs.acs.org/jced

Solubility of Solids in Sub- and Supercritical Fluids: a Review

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ABSTRACT: A review of solubility data for solid compounds in sub- and supercritical fluids published in the literature between 2005 and 2010 is given in tabular form along with the temperature and pressure ranges and the correlation methods applied by the authors for modeling the experimentally determined data. Compounds are classified into groups on the basis of their chemical nature (inorganic, organometallic, aromatic, nonaromatic organic, polymer), except for biological and pharmaceutical compounds, which are considered as a separate group based on their functionality. For each group of compounds, new findings are reviewed and discussed. Supercritical CO_2 was the preferred solvent for most of the applications, although an increasing interest in other solvents at elevated pressure has been observed, especially for fluorinated hydrocarbons, water, and propane.

1. INTRODUCTION

Because of environmental concerns and increasingly strict legislation aiming to lower detrimental environmental impacts as well as growing demand by consumers for more advanced and safe products, new modes of production in the chemical and related industries have been intensively studied in the past years. The studies have been based on progress that has been made in process design and control using the scientific methods of chemical engineering. Among the promising technologies that have been seriously considered for practical applications are also high-pressure technologies and the use of sub- and supercritical fluids, which represent an attractive green alternative to organic solvents conventionally used in various unit operations such as separation processes (extraction, sorption processes, chromatography, drying, membrane separations, etc.), mechanical processes (extrusion, homogenization, emulsification, micronization, crystallization, impregnation, encapsulation, etc.), and chemical and biochemical reactions. Processes with sub- and supercritical fluids have been intensively researched for use in different industrial branches, including agriculture and food, cosmetics, pharmaceuticals, medicine, coatings, textiles, electronics and semiconductors, and waste treatment. For all of these processes, the crucial data are physicochemical data of pure compounds, phase equilibrium data, and mass- and heat-transport data.

Several reviews of high-pressure phase equilibrium data published before 2005 are available in the literature,[1−14] and a compilation of data on solubility in supercritical (SC) CO_2 published from the 1960s to 2004 in both tabular and graphical form and covering 783 different compounds was recently published by Gupta and Shim.[15]

It is important to note that today, 60% of all products sold by chemical companies are crystalline, polymeric, or amorphous solids. These complex materials must have a clearly defined physical shape to meet the designed and desired quality standards. This also applies to pastelike and emulsified products.[16]

The present paper reviews the phase equilibrium data for compounds that are solid at room temperature (and also in some cases their analogous liquid derivatives of low volatility) in systems with supercritical fluids (SCFs) published in the period from 2005 to 2010. On the basis of a review of the latest research on phase equilibria for different systems, the use of SCFs for different applications is discussed.

Compounds are classified into different groups: inorganic compounds, organometallic compounds, biological and pharmaceutical compounds, other aromatic and other nonaromatic organic compounds, and polymers; only data on the solubility of polymers in pressurized fluids have been considered here, while articles dealing with the solubility and diffusivity of gases in polymers have not been included in this review. Similarly, systems involving ionic liquids (ILs) have not been considered because it is generally the case that CO_2 has excellent solubility in many ILs while ILs have little to no solubility in CO_2.[17]

The search strategy was as follows: for the *Journal of Supercritical Fluids* and *Fluid Phase Equilibria*, the tables of contents of all volumes published between 2005 to 2010 were systematically searched. For the *Journal of Chemical & Engineering Data*, *Industrial & Engineering Chemistry Research*, and the *Journal of Chemical Thermodynamics*, online searches for particular keywords (such as solubility and supercritical or phase equilibria and high-pressure) were performed. The above journals were chosen on the basis of the work done by Dohrn et al.,[4] who identified the major journals with the largest numbers of articles dealing with high-pressure fluid-phase equilibria in the years from 2000 to 2004. Finally, an online search (ISI Web of Knowledge) for the same keywords was performed. Considered were the papers for which at least one data point was measured at a pressure of 1 MPa or higher.

2. SYSTEMS INVESTIGATED

The literature review showed that SC CO_2 is the preferred solvent for most of the applications because of its useful properties. It is a clean and versatile solvent and a promising alternative to toxic volatile organic compounds (VOCs) and chlorofluorocarbons (CFCs). SC CO_2 is a good solvent for nonpolar compounds, while the solubility of some polar and ionic

Special Issue: John M. Prausnitz Festschrift

Received: October 29, 2010
Accepted: January 18, 2011
Published: February 22, 2011

compounds having high molecular weight in SC CO_2 is very low, limiting its application in industry. Therefore, there is also an increasing interest in other solvents for use at elevated pressures, especially for fluorinated hydrocarbons (HFCs), water, and propane. Fortunately, polar substances dissolve in HFCs such as trifluoromethane (R23), difluoromethane (R32), pentafluoroethane (R125), 1,1,1,2-tetrafluoroethane (R134a), 1,1,1-trifluoroethane (R143a), 1,1-difluoroethane (R152a), 1,1,1,2,3,3,3-heptafluoropropane (R227ea), and dimethyl ether (DME).[18] The fluids that were investigated as solvents and their properties are summarized in Table 1.

2.1. Inorganic Compounds. The review of systems involving inorganic compounds is given in Table 2. Only the following binary systems involving inorganic compounds were investigated: sulfur in CO_2; phosphate and sulfate salts, monovalent alkali nitrates, and alkali chlorides in H_2O; and alkali-metal iodides in NH_3. Most of the solubility data were measured using subcritical water (SBCW) or supercritical water (SCW). The investigated pressure range was between 8 and 30 MPa, while the temperatures were high (up to 690 K).

The sulfur−CO_2 system was investigated because of problems reported in recent years that elemental sulfur forms deposits in natural gas transmission line systems.[27] Available experimental data on the solubility of sulfur in gases are for much higher ranges of temperature, pressure, and hydrogen sulfide amount than those under transport conditions for natural gas. Therefore, solubility data for sulfur in the simple gases that are components of natural gas, such as CH_4 and CO_2, are needed.[27]

SCW has been considered as a medium of choice for reactions, polymerization, destruction of waste components, gasification of biomass, and particle formation.[21] Applications using SCW often encounter the presence of inorganic compounds in feed streams, most often with a minor concentration. Since the polarity of water in its SC state is reduced, the ability of water to dissolve inorganic compounds is reduced, and salts precipitate and form a solid phase. This can lead to damage of the equipment via erosion, scaling, and corrosion or influence the main reaction

Table 1. Physicochemical Properties[a] of Fluids[19]

fluid	T_c/K	P_c/MPa	ω	$10^{30}\cdot\mu$/C·m
methane (CH_4)	190.6	4.6	0.011	0
ethane (C_2H_6)	305.3	4.87	0.099	0
ethene (C_2H_4)	282.4	5.04	0.087	0
propane (C_3H_8)	369.8	4.25	0.152	0.280
propene (C_3H_6)	365.6	4.665	0.141	1.221
n-butane (C_4H_{10})	425.1	3.8	0.201	0.167
isobutane (R600a)	407.8	3.64	0.185	0.440
1-butene (C_4H_8)[20]	419.5	4.02	0.194	1.001
pentane (C_5H_{12})	469.7	3.37	0.251	1.234
hexane (C_6H_{14})	507.8	3.03	0.299	0.167
dimethyl ether (C_2H_6O)[20]	400.1	5.40	0.276	4.336
difluoromethane (R32)	351.3	5.78	0.277	6.598
trifluoromethane (R23)	299.3	4.83	0.263	5.500
chlorodifluoromethane (R22)	369.3	4.99	0.221	4.863
trichlorofluoromethane (R11)	471.1	4.41	0.189	1.501
1,1-difluoroethane (R152a)	386.4	4.52	0.275	7.545
1,1,1,2-tetrafluoroethane (R134a)	374.2	4.06	0.327	6.865
1,1,1,3,3,3-hexafluoropropane (R236fa)	398.1	3.20	0.377	6.611
carbon dioxide (CO_2)	304.1	7.38	0.224	0
sulfur hexafluoride (SF_6)	318.7	3.75	0.210	0
ammonia (NH_3)	405.4	11.33	0.256	4.903
dinitrogen monoxide (N_2O)	309.5	7.25	0.162	0.537
water (H_2O)	647.1	22.06	0.344	6.188

[a] T_c, critical temperature; P_c, critical pressure; ω, accentric factor; μ, dipole moment.

Table 2. Solubilities of Inorganic Compounds (X) in Pressurized Fluids (S)

X	S	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
$CaCl_2$	H_2O	1045	18.5 to 23.5	660 to 690	enthalpy approach	21
$CaCl_2$	H_2O	1045	11 to 14	623 to 673	ionization and hydration approach	22
$CaHPO_4$	H_2O	643	20.5 to 24.2	665 to 690	enthalpy approach	23
$CaSO_4$	H_2O	1733	18.8 to 23.2	655 to 675	enthalpy approach	23
CsI	NH_3	899	10.97 to 12.83	418.4	empirical approach	24
KCl	H_2O	1043	18 to 23.5	653 to 693	enthalpy approach	25
KI	NH_3	954	9.7 to 14.4	419.5 to 426.0	empirical approach	24
KNO_3	H_2O	607	18 to 23.5	653 to 693	enthalpy approach	25
LiCl	H_2O	878	18 to 23.5	653 to 693	enthalpy approach	25
LiCl	H_2O	878	7 to 11	623 to 673	ionization and hydration approach	22
$LiNO_3$	H_2O	537	18 to 23.5	653 to 693	enthalpy approach	25
$MgCl_2$	H_2O	987	18.5 to 23.5	660 to 690	enthalpy approach	21
$MgSO_4$	H_2O	1397	18.8 to 23.2	655 to 675	enthalpy approach	23
NaCl	H_2O	1074	17 to 24	543 to 683	enthalpy approach, C_p approach, empirical approach, Flory−Huggins approach	26
NaCl	H_2O	1074	18 to 23.5	653 to 693	enthalpy approach	25
NaH_2PO_4	H_2O	>443 (dec.[b])	20.5 to 24.2	665 to 690	enthalpy approach	23
Na_2HPO_4	H_2O	>523	20.5 to 24.2	665 to 690	enthalpy approach	23
NaI	NH_3	934	8.18 to 11.63	418.4	empirical approach	24
$NaNO_3$	H_2O	579	18 to 23.5	653 to 693	enthalpy approach	25
sulfur	CO_2	390 to 393	10 to 30	333 to 363	−	27

[a] T_{fus}: melting temperature of compound X. [b] dec.: decomposes.

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

and processes inside the systems. In order to avoid these problems and predict the influence of these compounds, it is vital to possess knowledge of the properties of the most common inorganic compounds in SCW.[21]

The solubilities of alkali metal iodides in supercritical ammonia (SCA) were studied by Sciaini et al.[24] A very interesting procedure for the destruction of hazardous materials (e.g., chemical weapons) is the use of solvated electrons. Solvated electrons are usually produced by dissolution of alkali metals in ammonia. However, another possibility for obtaining solvated electrons is to generate them by irradiation of iodides dissolved in different solvents, especially SCA, thus allowing the reaction capacity of the medium to be coupled with fine control of the solvent density.[24]

2.2. Organometallic Compounds. SC CO_2 is of interest as an alternative to organic solvents for the extraction of metal ions from waste soils and water and from solid and liquid wastes.[28] However, because of the very poor solubility of metal ions in SC CO_2, direct extraction of metal ions is difficult. Recent studies have demonstrated that noncharged metal complexes with chelating ligands lead to an increase in solubility of metal ions in SC CO_2.[28] For optimizing the extraction process, it is necessary to examine the solubility of metal−chelate complexes in SC CO_2.

Furthermore, SC CO_2 has recently been used to make polymer−metal nanocomposites and metal thin films on high-aspect-ratio surfaces of semiconductors. In these processes, the solubilities of metal complexes in SC CO_2 are the essential data.[29]

Currently, the semiconductor industry requires the development of novel deposition processes. The SCF deposition technique has received much attention for deposition of thin films on high-aspect-ratio surfaces of wafers or microchips. This technique is expected to markedly improve the coatability, embedded quality, and deposition rate and to increase the number of feasible precursors in comparison with existing methods such as physical vapor deposition, chemical vapor deposition, and atomic layer deposition.[30]

Furthermore, noble-metal complexes that are soluble in SC CO_2 have been used for the fabrication of catalysts and nanofunctional materials. In these efforts, an important issue is controlling the size and size distribution of the nanoparticles. These characteristics depend on the nucleation conditions, which are closely related to a solubility change in the SC CO_2 process. Thus, the solubility of these complexes in SC CO_2 is important not only for the success of the process but also for microstructure design.[31]

A large number of metal complexes with various ligands have been investigated experimentally to date.[32] Reviews of these data can be found in the literature.[33−36] It has been found that in general, complexes containing cyclopentadienyl and carbonyl ligands exhibit high solubilities in SC CO_2.[33] Complexes with ligands such as β-diketonate, dithiocarbamate, phosphine, and amine can also be used for applications in CO_2.[34,35]

The latest studies to investigate the influence of the ligands and metal atoms of metal complexes on the solubility in CO_2 are reviewed in Table 3. The studied systems include various β-diketonate [e.g., 2,2,6,6-tetramethyl-3,5-heptanedionate (thd), acetylacetonate (acac), trifluoroacetylacetonate (tfacac), and hexafluoroacetylacetonate (hfacac)], cyclopentadienyl (cp), cyclooctadiene (cod), and dithiocarbamate (dtc) complexes with metals, including silver, copper, nickel, palladium, rhodium, ruthenium, cobalt, chromium, iron, manganese, osmium, vanadium, platinum, potassium, rubidium, titanium, zinc, zirconium, lithium, and uranium (as uranyl ions).

2.3. Biological and Pharmaceutical Compounds. Investigated systems with biological compounds (Table 4) include phenolic compounds, chalcones, carotenoid pigments, fatty acids, amino acids, derivatized amino acids, alkaloids, antioxidants, pyrazines, biocides, and carbohydrates (with a view toward applying the appropriate extraction conditions to fractionate prebiotic sugars from complex carbohydrate mixtures, etc.[98]). Systems with pharmaceutical compounds (Table 4) include antilipemic agents, nonsteroidal anti-inflammatory drugs (NSAIDs), glucocorticoid drugs, anticancer and anti-HIV drugs, calcium channel blockers, antiandrogens, statins, etc.

The phase equilibrium data of these compounds in SCFs are necessary for designing separation or formulation processes such as micronization, impregnation, and encapsulation of drugs in biopolymer nano- or microparticles for controlled-release formulations. For these applications, CO_2−drug−polymer systems have been studied.[54]

For this group of compounds, the most investigated solvent is CO_2, but the potential use of other solvents such as SBCW, HFCs, propane, and ethane have also been investigated. For example, Liu et al.[111] have reported that the solubility of the polar solute L-proline in the subcritical HFC R134a is much higher than in the most commonly used supercritical solvent, CO_2.

Water is a solvent with tunable polarity. As the temperature of water increases, its polarity decreases. The reduction in polarity enables water to dissolve a variety of hydrophobic organic compounds. Recently, SBCW [defined as water between (373 and 573 K)] was used to dissolve a number of pharmaceutical ingredients, which were then precipitated by injecting the hot SBCW/pharmaceutical solutions into room-temperature water. The sizes of the resulting particles varied from (0.5 to 100) μm.[66]

SC CO_2 is generally a poor solvent for a wide range of hydrophilic and polar substances. One possibility for enhancing the solubility is to use cosolvents. Another very promising approach for enhancing the solubility is to use water-in-SC CO_2 (W/C) microemulsions.[123] A W/C microemulsion is formed when small-diameter droplets of polar chemicals are dispersed in CO_2. The aqueous phase especially disperses as nanosized droplets are surrounded by a monolayer of surfactant molecules in the continuous CO_2-rich phase of the W/C microemulsion. Using SC CO_2 to create a W/C microemulsion may be advantageous in some chemical processes, such as extraction, cleaning, nanoparticle synthesis, and chemical reactions. Knowledge of the solubilization amount is therefore essential for the application of these processes based on W/C microemulsions.[123]

Another way of enhancing the solubility of polar compounds is to add CO_2-philic groups as side chains to molecules of the compounds. Recently, hydrocarbons substituted with carbonyl groups have been shown to be highly soluble in supercritical carbon dioxide.[99] Dilek et al.[99] and Hong et al.[112] investigated sugar acetates, which are a class of materials with the potential for high solubility in carbon dioxide. The high solubility of acetylated molecules in CO_2 is attributed to the Lewis acid−Lewis base interaction between CO_2 and the carbonyl group. Weak but cooperative hydrogen bonding between the hydrogen of the carbonyl group and the oxygen of CO_2 is the second reason for the enhanced solubility of these sugar acetates.[99]

Table 3. Solubilities of Organometallic Compounds (X) in Pressurized Fluids (S) + Cosolvents (C)

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
Ag(acac)[b]	CO_2		373 (dec.[n])	10 to 30	333	–	32
Ag(acac)[b]	CO_2		373 (dec.[n])	10 to 30	313	Chrastil	31
Ag(thd)[c]	CO_2		n.a.[o]	10 to 30	333	–	32
$Co(acac)_3$[b]	CO_2		483 to 486	10 to 28.1	313 to 343	Chrastil	30
$Co(cp)_2$[d]	CO_2		445 to 447	10 to 17.5	333	–	32
$Co(thd)_3$[c]	CO_2		523 (dec.[n])	10 to 19.8	313 to 343	Chrastil	29
$Co(thd)_3$[c]	CO_2		523 (dec.[n])	10 to 17	333	–	32
$Cr(acac)_3$[b]	CO_2		483	12.3 to 30.3	313 to 343	Chrastil	30
$Cr(acac)_3$[b]	CO_2	methanol	483	10.1 to 20.3	318	–	37
$Cr(acac)_3$[b]	CO_2	ethanol	483	10.1 to 20.3	318	–	37
$Cr(acac)_3$[b]	CO_2	TFE[k]	483	10.1 to 20.3	318	–	37
$Cr(acac)_3$[b]	CO_2	HFP[l]	483	10.1 to 20.3	318	–	37
$Cr(acac)_3$[b]	CO_2	BTMP[m]	483	10.1 to 20.3	318	–	37
$Cr(cp)_2$[d]	CO_2		445 to 447	10 to 17.5	333	–	32
$Cr(thd)_3$[c]	CO_2		501 to 506	10 to 18	313 to 343	Chrastil	29
$Cr(thd)_3$[c]	CO_2		501 to 506	10 to 15	333	–	32
$Cu(acac)_2$[b]	CO_2		557 to 561 (dec.[n])	10 to 30	333	–	32
$Cu(acac)_2$[b]	CO_2		557 to 561	10 to 30	313	Chrastil	31
$Cu(thd)_2$[c]	CO_2		471	10 to 17.5	333	–	32
$Fe(cp)_2$[d]	CO_2		445 to 446	10 to 17.5	333	–	32
$Fe(cp)_2$[d]	H_2O		445 to 447	5	313 to 433	–	38
$Fe(thd)_3$[c]	CO_2		452 to 458	10 to 15	333	–	32
K(thd)[c]	CO_2		n.a.[o]	10 to 20	333	–	32
Li bis(trifluoroethyl)(dtc)[e]	CO_2		n.a.[o]	9.9 to 24.9	298 to 318	Chrastil	39
Li dibutyl(dtc)[e]	CO_2		n.a.[o]	8 to 24.9	298 to 318	Chrastil	39
Li diethyl(dtc)[e]	CO_2		n.a.[o]	8.5 to 25	298 to 318	Chrastil	39
Li dipropyl(dtc)[e]	CO_2		n.a.[o]	10 to 25	298 to 318	Chrastil	39
$Mn(cp)_2$[d]	CO_2		445 to 447	10 to 30	333	–	32
$Mn(thd)_3$[c]	CO_2		433 to 443	10 to 15	333	–	32
$Ni(acac)_2$[b]	CO_2		503 (dec.[n])	10 to 30	333	–	32
$Ni(cp)_2$[d]	CO_2		445 to 447	10 to 17.5	333	–	32
$Ni(thd)_2$[c]	CO_2		498	10 to 25	333	–	32
$Os(cp)_2$[d]	CO_2		499 to 501	10 to 25	333	–	32
$Pd(acac)_2$[b]	CO_2		473 to 524 (dec.[n])	10 to 30	333	–	32
$Pd(acac)_2$[b]	CO_2		473 to 524	10 to 30	313	Chrastil	31
$Pt(acac)_2$[b]	CO_2		522 to 525	10 to 30	313	Chrastil	31
$Pt(cod)me_2$[f,g]	CO_2		n.a.[o]	10 to 20	333	–	32
Rb(thd)[c]	CO_2		n.a.[o]	10 to 20	333	–	32
$Rh(acac)_3$[b]	CO_2		536 to 537	10 to 30	313	Chrastil	31
Rh(acac)(cod)[b,f]	CO_2		411 to 413	10 to 20	333	–	32
$Ru(acac)_3$[b]	CO_2		533	10 to 30	313	Chrastil	31
$Ru(cp)_2$[d]	CO_2		467 to 471	10 to 17	333	–	32
$Ru(thd)_3$[c]	CO_2		n.a.[o]	10 to 15	333	–	32
$Ru(thd)_2(cod)$[c,f]	CO_2		n.a.[o]	10 to 15	333	–	32
$Ti(thd)_3$[c]	CO_2		n.a.[o]	10	333	–	32
$UO_2(acac)_2dmso$[b,h]	CO_2		n.a.[o]	10 to 25	313	Chrastil	28
$UO_2(hfacac)_2dmso$[i,h]	CO_2		n.a.[o]	10 to 25	313	Chrastil	28
$UO_2(tfacac)_2dmso$[j,h]	CO_2		n.a.[o]	10 to 25	313	Chrastil	28
$V(cp)_2$[d]	CO_2		438 to 440	10	333	–	32
$Zn(thd)_2$[c]	CO_2		417	10 to 15	333	–	32
$Zr(thd)_4$[c]	CO_2		605 to 612	10 to 17.5	333	–	32

[a] T_{fus}: melting temperature of compound X. [b] acac: acetylacetonate. [c] thd: 2,2,6,6-tetramethyl-3,5-heptanedionate. [d] cp: cyclopentadienyl. [e] dtc: dithiocarbamate. [f] cod: cyclooctadiene. [g] me: methyl. [h] dmso: dimethyl sulfoxide. [i] hfacac: hexafluoroacetylacetonate. [j] tfacac: trifluoroacetylacetonate. [k] TFE: 2,2,2-trifluoroethanol. [l] HFP: 1,1,1,3,3,3-hexafluoro-2-propanol. [m] BTMP: 3,5-bis(trifluoromethyl)phenol. [n] dec.: decomposes. [o] n.a.: data not available.

Furthermore, solubility enhancements by fluorination have been reported. Higashi et al.[140] measured the solubilities of 2-, 3-, and 4-trifluoromethylbenzoic acids in supercritical carbon dioxide and compared the data with those for

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 4. Solubilities of Biological and Pharmaceutical Compounds (X) in Solvents (S) + Cosolvents (C)

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
acebutolol	CO_2		solid, n.a.[g]	8 to 27.5	298 to 318	−	40
acetaminophen	CO_2		441 to 445	20	323	−	41
acetaminophen	CO_2		441.2	11.14 to 25	313.3	M-S-T, Chrastil-G-M	42
acetazolamide	CO_2	ethanol	531 to 532	15 to 20.1	313 to 323	Chrastil, M-S-T−Sauceau	43
2-acetoxybenzoic acid	CO_2		408 to 409	10.1 to 28.0	308 to 328	−	45
2-acetoxybenzoic acid	CO_2	methanol	408.2 to 411.2	10 to 20	318 to 328	PR EOS	53
2-acetoxybenzoic acid	CO_2	ethanol	408.2 to 411.2	10 to 20	318 to 328	PR EOS	53
2-acetoxybenzoic acid + benzoic acid	CO_2		−	10.1 to 28.0	308 to 328	−	45
2-acetoxybenzoic acid + 2-hydroxybenzoic acid	CO_2		−	10.1 to 28.0	308 to 328	−	45
2-acetoxybenzoic acid + 2-hydroxybenzoic acid + benzoic acid	CO_2		−	10.1 to 28.0	308 to 328	−	45
2-acetoxybenzoic acid + D,L-PLA[b]	CO_2		−	7.5 to 10.0	313.15	−	54
2-acetoxybenzoic acid + L-PLA[b]	CO_2		−	7.5 to 10.0	313.15	−	54
N-acetyl-L-phenylalanine	CO_2		438.2 to 440.2	7.8 to 23.5	308.2	K-J	116
N-acetyl-L-phenylalanine ethyl ester	CO_2		solid, n.a.[g]	7.8 to 23.5	308.2 to 328.2	K-J	116
N-acetyl-L-tyrosine	CO_2		422.2 to 425.2	7.8 to 23.5	308.2	K-J	116
N-acetyl-L-tyrosine ethyl ester	CO_2		351.2 to 354.2	7.8 to 23.5	308.2 to 328.2	K-J	116
2-acetylpyrazine	CO_2		351.2	up to 20	298 to 373	PR EOS	44
4-aminoantipyrine	CO_2		382.15	10 to 22	308.2 to 328.2	M-S-T, Chrastil, solution model	46
4-aminobenzene sulfonamide	CO_2		437 to 439	11 to 21	308.2 to 328.2	Chrastil	47
4-aminobenzene sulfonamide	CO_2	acetone	437 to 439	11 to 21	308.2 to 328.2	Chrastil-G	47
4-aminobenzoic acid	CO_2		460 to 462	8 to 21	308.2 to 328.2	M-S-T	48
2-aminobenzoic acid	CO_2		420.35	9.6 to 23.7	313 to 323	M-S-T	49
2-aminobenzoic acid	CO_2	menthol	420.35	9.6 to 23.7	313 to 323	M-S-T−Thakur and Gupta	49
anastrozole	CO_2		354.2	12.2 to 35.5	308 to 348	Chrastil, dV-A, K-J, Bartle, Yu, Gordillo	50
antipyrine	CO_2		385.65	10 to 22	308.2 to 328.2	M-S-T, Chrastil, solution model	46
artemisinin	CO_2		429.2 to 430.2	11 to 31	313 to 333	Chrastil, M-S-T	51
artemisinin	CO_2		429.2 to 430.2	10 to 25	308.2 to 328.2	Chrastil, Bartle, M-S-T, PR EOS, SRK EOS	52
astaxanthin	CO_2		solid, n.a.[g]	10 to 42	313 to 333	M-S-T	55
atenolol	CO_2		421 to 425	8 to 27.5	298 to 318	−	40
atorvastatin	CO_2		432.2 to 463.7	12.16 to 35.46	308 to 348	Chrastil, Bartle, K-J, M-S-T	56
azodicarbonamide	CO_2		224 to 225	10 to 30	308 to 328	−	41
beclomethasone dipropionate	CO_2		390 to 393	21.3 to 38.5	338 to 358	Bartle	58
benzoic acid	CO_2		395.6	12.12 to 29.8	338	−	59
benzoic acid	CO_2		395.6	up to 20.7	403.3 to 458.4		60
benzoic acid	H_2O		395.6	5	298 to 473	empirical model	61
benzoic acid	CO_2		395	10.1 to 28.0	308 to 328	−	45
bis(cyclohexyl)DH-PDC [c]	CO_2		546 to 548	12.2 to 35.5	328 to 348	Bartle	62
bis(4-cyclohexylbutyl)DH-PDC [c]	CO_2		437 to 438	12.2 to 35.5	338 to 358	Bartle	62
bis(2-cyclohexylethyl)DH-PDC [c]	CO_2		488 to 489	12.2 to 35.5	338 to 358	Bartle	62
bis(cyclohexylmethyl)DH-PDC [c]	CO_2		535 to 536	12.2 to 35.5	328 to 348	Bartle	62
bis(3-cyclohexylpropyl)DH-PDC [c]	CO_2		474 to 475	12.2 to 35.5	338 to 358	Bartle	62
bis(phenylbutyl)DH-PDC [c]	CO_2		409 to 410	12.2 to 35.5	338 to 358	Bartle	63
bis(phenylethyl)DH-PDC [c]	CO_2		477 to 478	12.2 to 35.5	338 to 358	Bartle	63
bis(phenylmethyl)DH-PDC [c]	CO_2		479 to 481	12.2 to 35.5	338 to 358	Bartle	63
bis(phenylpentyl)DH-PDC [c]	CO_2		401 to 402	12.2 to 35.5	338 to 358	Bartle	63
bis(phenylpropyl)DH-PDC [c]	CO_2		413 to 414	12.2 to 35.5	338 to 358	Bartle	63
bis(phenyltolyl)DH-PDC [c]	CO_2		511 to 512	12.2 to 35.5	338 to 358	Bartle	63
bixin + β-carotene	CO_2		−	35	313 to 333	PR EOS	64
boldine	CO_2		435.2 to 437.2	8 to 40	298 to 333	M-S-T	65

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 4. Continued

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
budesonide	CO_2		494 to 505	21.3 to 38.5	338 to 358	Bartle	58
budesonide	H_2O		499.65	7	298 to 473	M-UNIFAC	66
budesonide	H_2O	methanol	499.65	7	298 to 473	M-UNIFAC	66
budesonide	H_2O	ethanol	499.65	7	298 to 473	M-UNIFAC	66
caffeine	CO_2		500 to 501	15	313	−	67
caffeine	CO_2	water	500 to 501	15	313	−	67
caffeine	CO_2	ethanol	500 to 501	15	313	−	67
caffeine	CO_2	water + ethanol	500 to 501	15	313	−	67
caffeine	CO_2	ethanol	500 to 501	15.2 to 30.0	323.2 to 343.2	−	68
caffeine	CO_2	isopropanol	500 to 501	15.2 to 30.0	323.2 to 343.2		68
camphor	CO_2		448 to 450	3.42 to 14.94	304 to 354	PR EOS	69
camphor	C_3H_8		448 to 450	1.0 to 4.89	304 to 384	PR EOS	69
camphor	$CO_2 + C_3H_8$		448 to 450	2.4 to 15.02	304 to 354	PR EOS	69
cannabinol	CO_2		349.2 to 350.2	13.0 to 20.2	314 to 334	PR EOS	70
capsaicin	CO_2		335 to 338	7.81 to 25.62	298.2 to 328.3	M-S-T, Chrastil-G-M	42
capsaicin	CO_2		335 to 338	6 to 40	298 to 318	M-S-T, PR EOS, GC EOS	71
capsaicin	CO_2		335 to 338	10.4 to 22.9	313 to 333	M-S-T	72
carnosic acid	CO_2	ethanol	solid, n.a.[g]	27.5 to 40	313.2 to 333.2	GCA EOS	73
β-carotene	CO_2		451	12. to 20	313 to 323	−	74
β-carotene	CO_2	ethyl acetate	451	2.52 to 10.88	303 to 343	PR EOS	75
β-carotene	CO_2	ethanol	451	3.12 to 11.73	303 to 343	PR EOS	75
β-carotene	CO_2		453 to 455	8.5 to 25.3	298 to 313	M-S-T, Chrastil-G-M	42
β-carotene	CO_2		449 to 457	15 to 28	313.2 to 333.2	PR EOS	76
β-carotene	CO_2	ethanol	449 to 457	15 to 28	313.2 - 333.2	Chrastil-modified	77
β-carotene	CO_2	acetone	449 to 457	15 to 28	313.2 - 333.2	Chrastil-modified	77
β-carotene	CO_2	*n*-hexane	449 to 457	15 to 28	313.2 - 333.2	Chrastil-modified	77
catechin	CO_2	ethanol + water	448 to 450	15 to 19	313	−	78
cholesteryl benzoate	CO_2		423	16.0 to 27.0	318.2	−	79
cholesteryl benzoate + cholesteryl butyrate	CO_2		−	16 to 24	318.2	PR EOS	79
cholesteryl benzoate + cholesteryl butyrate	CO_2		−	12.0 to 24.0	308.2 to 328.2	PR EOS, Chrastil, K-J, Bartle, M-S-T	80
cholesteryl butyrate	CO_2		372	14.0 to 24.0	318.2	−	79
cinnamic acid	CO_2		406.15	12.3 to 23.61	308.2 to 328.2	Chrastil, M-S-T, PR EOS, SRK EOS	81
trans-cinnamic acid	CO_2		406.2	10 to 40	313.2 to 353.2	SRK EOS, GCA EOS	82
trans-cinnamic acid	CO_2	ethanol	406.2	10 to 40	313.2 to 353.2	SRK EOS, GCA EOS	82
climbazole	CO_2		368 to 371	10 to 40	313.2 to 333.2	PR EOS, QLF EOS	83
clofibric acid	CO_2		394.15	10 to 22	308 to 328	M-S-T, Chrastil, solution model	84
clozapine	CO_2		456	12.16 to 35.46	318 to 348	Chrastil, Bartle, K-J, M-S-T	85
p-coumaric acid	CO_2	ethanol + water	487 (dec.[h])	15 to 19	313	−	78
coumarin	CO_2		344	11 to 24	308 to 328	PR EOS	86
diflunisal	CO_2		492 to 493	9.1 to 24.6	308.2 to 328.2	PR EOS	87
4-dimethylaminoantipyrine	CO_2		381.15	10 to 22	308.2 to 328.2	M-S-T, Chrastil, solution model	46
2,3-dimethylpyrazine	CO_2		285	up to 20	298 to 373	PR EOS	44
3,5-dinitrobenzoic acid	CO_2		477.2 to 479.2	10 to 21	308 to 328	−	88
3,5-dinitrobenzoic acid	CO_2	ethanol	477.2 to 479.2	10 to 21	318	Sovova, modified Sovova	88
3,5-dinitrobenzoic acid	CO_2	ethylacetate	477.2 to 479.2	10 to 21	308 to 328	Sovova, modified Sovova	88
3,5-dinitrobenzoic acid	CO_2	*n*-propanol	477.2 to 479.2	10 to 21	318	Sovova, modified Sovova	88

dx.doi.org/10.1021/je1011373 |*J. Chem. Eng. Data* 2011, 56, 694–719

Table 4. Continued

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
3,5-dinitrobenzoic acid	CO_2	ethylene glycol	477.2 to 479.2	10 to 21	318	Sovova, modified Sovova	88
1,3-diphenyl-2-propenone	CO_2		328 to 332	9 to 22.17	308 to 338	Chrastil, PR EOS	89
dodecanoic acid	CO_2		316.4	9.9 to 22.7	308	PR EOS	109
dodecanoic acid	CO_2	ethanol	316.4	9.9 to 22.7	308	PR EOS	109
dodecanoic acid	CO_2	isoamyl alcohol	316.4	9.9 to 22.7	308	PR EOS	109
dutasteride	CO_2		515	12.1 to 35.5	308 to 348	Chrastil, Bartle	90
epigallocatechin gallate	CO_2		495.2 to 497.2	15 to 35	313	−	91
epigallocatechin gallate	CO_2	acetone	495.2 to 497.2	15 to 35	313	−	91
epigallocatechin gallate	CO_2	ethanol	495.2 to 497.2	15 to 35	313 to 333	Chrastil-G, M-S-T, PR EOS	91
ethyl *p*-aminobenzoate	CO_2		365	8 to 21	308 to 328	Chrastil	92
ethyl *p*-hydroxybenzoate	CO_2		388 to 391	8 to 21	308 to 328	Chrastil	92
ethyl *p*-hydroxybenzoate + ethyl *p*-aminobenzoate	CO_2		−	8 to 21	308 to 328	Chrastil	92
ethylvanillin	CO_2		349.8	8.1 to 30.15	313.2 to 353.2	PR EOS	93
ethylvanillin	R23		349.8	3.2 to 24.75	313.2 to 333.2	Chrastil	94
ethylvanillin	R134a		349.8	6.05 to 29.0	313.2 to 333.2	Chrastil	94
ethylvanillin	R236fa		349.8	5.75 to 26.6	313.2 to 333.2	Chrastil	94
o-ethylvanillin	CO_2		337.2	10.3 to 30.2	313.2 to 353.2	PR EOS	93
o-ethylvanillin	R23		337.2	4.33 to 20.13	313.2 to 333.2	Chrastil	94
o-ethylvanillin	R134a		337.2	4.75 to 26.05	313.2 to 333.2	Chrastil	94
o-ethylvanillin	R236fa		337.2	4.25 to 27.0	313.2 to 333.2	Chrastil	94
exemestane	CO_2		428.3	12.2 to 35.5	308 to 348	Chrastil, dV-A, K-J, Bartle, Yu, Gordillo	50
felodipine	CO_2		415.2 to 417.2	8 to 25	298 to 318	Chrastil	95
fenofibrate	CO_2		358.65	10 to 22	308 to 328	M-S-T, Chrastil, solution model	84
ferulic acid	CO_2		441.2 to 445.2	10 to 35	308 to 338	Chrastil	96
finasteride	CO_2		525	12.1 to 35.5	308 to 348	Chrastil, Bartle	90
fluoranthene	CO_2		380 to 383	10.1 to 28.0	308 to 328	−	45
fluoranthene + benzoic acid	CO_2		−	12 to 28	318	−	45
fluoranthene + 2-hydroxybenzoic acid + benzoic acid	CO_2		−	12 to 28	318	−	45
(±)-*trans*-4-(4′-fluoro-phenyl)-3-hydroxymethyl-1-methyl-piperidine	CO_2		395 to 397	9 to 24	308.2 to 328.2	Chrastil	139
5-fluorouracil	CO_2		282 to 286	12.5 to 25	308 to 328	−	41
flurbiprofen	CO_2		383 to 385	10.76 to 22.12	303 to 323	PR EoS	97
flurbiprofen	CO_2	methanol	383 to 385	10.03 to 24.07	303 to 313	PR EoS	97
flutamide	CO_2		384	12.1 to 35.5	308 to 348	Chrastil, Bartle	90
fluticasone propionate	CO_2		545 to 546	38.5	358	−	58
fluvastatin	CO_2		467.0 to 470.0	12.16 to 35.46	308 to 348	Chrastil, Bartle, K-J, M-S-T	56
galactose	CO_2	ethanol + water	436.2	10 to 30	333 to 373	SRK EOS	98
β-D-galactose pentaacetate	CO_2		412 to 415	9 to 16	308 to 323	−	99
gemfibrozil	CO_2		335.15	10 to 22	308 to 328	M-S-T, Chrastil, solution model	84
griseofulvin	H_2O		492	7	413 to 443	M-UNIFAC	101
griseofulvin	CO_2		493	9.51 to 23.52	308 to 323	M-S-T	102
griseofulvin	CO_2	menthol	493	9.6 to 23.9	313 to 323	M-S-T−Thakur and Gupta	102

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 4. Continued

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
heptanoic acid	CO_2		262.7	8.5 to 20	313.2 to 333.2	Chrastil, dV-A, Chrastil-A-L	103
hexadecanoic acid	CO_2		336.1	10 to 26.1	313 to 318	M-S-T, Chrastil-G-M	42
hexadecanoic acid	CO_2	ethanol	336.1	up to 20	303 to 343	PR EOS	124
hexadecanoic acid	CO_2		336.1	12.8 to 22.6	308 to 318	M-S-T	125
hexadecanoic acid	CO_2	ethanol	336.1	12.8 to 22.6	308 to 318	M-S-T	125
hexadecanoic acid	CO_2	3-methyl-1-butanol	336.1	12.8 to 22.6	308 to 318	M-S-T	125
hexadecanoic acid	CO_2		336.1	10.1 to 23.4	313 to 318	M-S-T	126
hexadecanoic acid	CO_2	ethanol	336.1	8.2 to 24.6	313	M-S-T	126
hexadecanoic acid	CO_2	2-propanol	336.1	10.9 to 20.7	313	M-S-T	126
hexadecanoic acid	CO_2		336.1	12.8 to 22.6	328 to 338	PR EOS, M-S-T, Chrastil	127
hydrogenated castor oil	CO_2		n.a.[g]	4.23 to 32.28	353 to 403	−	104
2-hydroxybenzamide	CO_2		413.58	10 to 22	308.2 to 328.2	Chrastil, M-S-T, solution model	115
2-hydroxybenzoic acid	H_2O		432	5	298 to 473	empirical model	61
2-hydroxybenzoic acid	CO_2		432	10.1 to 28.0	308 to 328	−	45
2-hydroxybenzoic acid + benzoic acid	CO_2		−	10.1 to 28.0	308 to 328	−	45
2-hydroxybenzoic acid + D,L-PLA [b]	CO_2		−	7.5 to 10.0	313.2	−	54
2-hydroxybenzoic acid + L-PLA [b]	CO_2		−	7.5 to 10.0	313.2	−	54
2-hydroxy-5-sulfobenzoic acid	CO_2		378 to 383	8 to 21	308 to 328	Chrastil	135
2-hydroxy-5-sulfobenzoic acid + *p*-aminobenzoic acid	CO_2		−	8 to 21	308 to 328	Chrastil	135
ibuprofen	CO_2	acetone	346 to 349	10	298	Wubbolts	105
ibuprofen	CO_2	ethanol	346 to 349	10	298	Wubbolts	105
ibuprofen	CO_2		346 to 349	12 to 23	313	−	41
ibuprofen + D,L-PLA [b]	CO_2		−	7.5 to 10.0	313.15	−	54
ibuprofen + L-PLA [b]	CO_2		−	7.5 to 10.0	313.15	−	54
iodopropynyl butylcarbamate	CO_2		337 to 341	8.76 to 34.15	313.2 to 333.2	PR EOS, QLF EOS	106
isoniazid	CO_2		444.2 to 446.2	13.0 to 18.5	308 to 313	Chrastil, M-S-T	107
ketoprofen	CO_2		366 to 367	6.5 to 30	298 to 328	−	108
ketoprofen + PLGA[d]	CO_2		−	6.5 to 30	298 to 328	−	108
lactose	CO_2	ethanol + water	496.2	10 to 30	333 to 373	SRK EOS	98
lactulose	CO_2	ethanol + water	442.2	10 to 30	333 to 373	SRK EOS	98
lamotrigine	CO_2		491	12.16 to 35.46	318 to 348	Chrastil, Bartle, K-J, M-S-T	85
letrozole	CO_2		454.2	12.2 to 35.5	308 to 348	Chrastil, dV-A, K-J, Bartle, Yu, Gordillo	50
lovastatin	CO_2		447.5	12.16 to 35.46	308 to 348	Chrastil, Bartle, K-J, M-S-T	56
lovastatin	CO_2	DCM[f]	447.5	6.4 to 45	303.3 to 333.3	−	110
lycopene	CO_2		450	10 to 42	313 to 333	M-S-T	55
lycopene	CO_2		445 to 446	20 to 40	323 to 353	modified PR EOS	76
β-D-maltose octaacetate	CO_2		432	up to 22	253 to 323	−	112
menthol	CO_2		305 to 307	7.29 to 23.56	308 to 323	M-S-T	102
4-methoxyphenylacetic acid	CO_2		360.15	11.61 to 23.61	308.2 to 328.2	Chrastil, M-S-T, PRE OS, SRK EOS	81
2-methoxypyrazine	CO_2		liquid	up to 20	298 to 373	PR EOS	44
methyl 2-hydroxybenzoate	CO_2		265 to 266	9 to 31	343.2 to 423.2	Chrastil, PR EOS	113
2-methylpyrazine	CO_2		244	up to 20	298 to 373	PR EOS	44
mitotane	CO_2		349 to 351	up to 22	298.2 to 333.1	PR EOS	114
nabumetone	CO_2		353.15	10 to 22	308.2 to 328.2	Chrastil, M-S-T, solution model	115
nadolol	CO_2		solid, n.a.[g]	8 to 27.5	298 to 318	−	40
naproxen	CO_2	acetone	426 to 427	10	298	Wubbolts	105
naproxen	CO_2	ethanol	426 to 427	10	298	Wubbolts	105
naproxen	CO_2		430 to 431	12 to 28	313	−	41
naproxen	CO_2		427.7	15 to 40	313 to 353	K-J, M-S-T, EOS and fugacity approach	117

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 4. Continued

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
naproxen	H_2O		427.7	7	403 to 443	M-UNIFAC	118
naringenin	CO_2		520.2 to 523.2	10.3 to 44.5	313 to 333	M-S-T	119
N-carbobenzoxy-L-phenylalanine	CO_2		358.2 to 360.2	7.8 to 23.5	308.2	K-J	116
N-carbobenzoxy-L-tyrosine	CO_2		368.2	7.8 to 23.5	308.2	K-J	116
nitrendipine	CO_2		430.2 to 431.2	8 to 25	298 to 318	Chrastil	95
nonanedioic acid	CO_2		382.2 to 384.2	10 to 30	313.2 to 333.2	Chrastil, M-S-T, PR EOS	57
nonanoic acid	CO_2		282.2	10 to 30	313.2 to 333.2	Chrastil-A-L	120
nonanoic acid	CO_2		282.2	8.5 to 30	313.2 to 333.2	Chrastil, dV-A, Chrastil-A-L	103
octadecanoic acid	CO_2		343	12.8 to 22.6	328 to 338	PR EOS, M-S-T, Chrastil	127
octadecanoic acid	CO_2		343	12.8 to 22.6	308 to 318	M-S-T	125
octadecanoic acid	CO_2	ethanol	343	12.8 to 22.6	308 to 318	M-S-T	125
octadecanoic acid	CO_2	3-methyl-1-butanol	343	12.8 to 22.6	308 to 318	M-S-T	125
octanioc acid	CO_2		288.2 to 290.2	8.5 to 30	313.2 to 333.2	Chrastil, dV-A, Chrastil-A-L	103
γ-oryzanol	C_3H_8	chloroform	408 to 410	up to 17	303 to 353	−	121
oxymatrine	CO_2		480 to 481	11 to 21	308.2 to 328.2	Bartle	122
oxymatrine	CO_2		480 to 481	30	328.2	−	123
oxymatrine	$H_2O + CO_2$		480 to 481	30	328.2	−	123
oxymatrine + PFPE-NH_4 [e]	CO_2		−	30	328.2	−	123
oxymatrine + PFPE-NH_4 [e]	$H_2O + CO_2$		−	15 to 23	308.2 to 328.2	−	123
paclitaxel	CO_2		213 to 217	10 to 30	308 to 328	−	41
patchoulol	CO_2		328 to 333	10 to 25	313 to 323	−	128
phenacetin	CO_2		407	9 to 19	308 to 328	PR EOS, M-S-T	129
2-(phenoxy)acetic acid	CO_2		371.65	11.75 to 22.43	308.2 to 328.2	Chrastil, M-S-T, PR EOS, SRK EOS	81
2-phenyl-4*H*-1,3-benzoxazin-4-one	CO_2		123 to 125	10 to 30	308 to 328	−	51
phenylbutazone	CO_2		378.58	10 to 22	308.2 to 328.2	Chrastil, M-S-T, solution model	115
phytosterol	CO_2		411.5	14.33 to 31.19	323 to 343	−	59
pindolol	CO_2		440 to 444	8 to 27.5	298 to 318	−	40
D-pinitol	CO_2		452 to 458	10 to 40	313 to 333	SRK EOS, GC EOS	130
piperine	CO_2		404 to 408	10 to 20	293 to 333	Chrastil, dilute solution model	131
progesterone	CO_2		406.11	up to 10	283 to 368	PR EOS	132
progesterone	C_3H_8		406.11	up to 10	283 to 368	PR EOS	132
progesterone	C_4H_{10}		406.11	up to 10	283 to 368	PR EOS	132
L-proline	R134a		501 (dec.) [h]	5 to 25	288 to 328	−	111
L-proline	CO_2		501 (dec.[h])	10 to 36	308 to 332	−	111
propranolol	CO_2		solid, n.a.[g]	8 to 27.5	298 to 318	−	40
propyl *p*-hydroxybenzoate	CO_2		370.2	8.0 to 23.0	308.2 to 328.2	Chrastil	133
propyl *p*-hydroxybenzoate	CO_2	cyclohexane	370.2	8.0 to 23.0	308.2	Chrastil-cosolvent	133
propyl *p*-hydroxybenzoate	CO_2	cyclohexane + acetone	370.2	8.0 to 23.0	308.2	Chrastil-cosolvent	133
propyl *p*-hydroxybenzoate	CO_2	acetone	370.2	8.0 to 23.0	308.2	Chrastil-cosolvent	133
puerarin	CO_2	ethanol	460.2 to 462.2	2 to 20	308 to 328	−	134
pyrazine	CO_2		325.2	up to 20	298 to 373	PR EOS	44
quercetin-3-glucoside	CO_2	ethanol + water	solid, n.a.[g]	15, 19	313	−	78
resveratrol	CO_2	ethanol + water	526 to 528	15, 19	313	−	78
rosuvastatin	CO_2		435.0	12.16 to 35.46	308 to 348	Chrastil, Bartle, K-J, M-S-T	56
rp70 (hardened rapeseed oil)	CO_2		n.a.[g]	3.15 to 30.78	333 to 367	−	104
rp70 (hardened rapeseed oil)	C_3H_8		n.a.[g]	0.99 to 7.18	328 to 363		104

dx.doi.org/10.1021/je1011373 |*J. Chem. Eng. Data* 2011, 56, 694–719

Table 4. Continued

X	S	C	t_{fus}/k [a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref.
simvastatin	CO_2		408.0 to 411.0	12.16 to 35.46	308 to 348 K	Chrastil, Bartle, K-J, M-S-T	56
simvastatin	CO_2	DCM [f]	408.0 to 411.0	6.4 to 45	303.3 to 333.3	−	110
tagatose	CO_2	ethanol + water	407.2	10 to 30	333 to 373	SRK EOS	98
tetradecanoic acid	CO_2		327.6	9.9 to 22.7	308 to 318	PR EOS	109
tetradecanoic acid	CO_2	ethanol	327.6	9.9 to 22.7	308 to 318	PR EOS	109
tetradecanoic acid	CO_2	isoamyl alcohol	327.6	9.9 to 22.7	308 to 318	PR EOS	109
Δ^9-tetrahydrocannabinol	CO_2		solid, n.a.[g]	13.2 to 25.1	315 to 345	PR EOS	136
tetramethylpyrazine	CO_2		343.2 to 353.2	10 to 30	318 to 338	Chrastil	96
theophylline	CO_2	ethanol	545	10	309.2	−	48
theophylline	CO_2	ethanol + DCM [f]	545	10	309.2	−	48
theophylline	CO_2		544.2 to 546.2	10 to 44	313 to 333	M-S-T	137
thymidine	CO_2		186 to 188	10 to 30	308 to 328	−	41
α-tocopheryl succinate	CO_2		349 to 350	15.0 to 30.0	313 to 323	M-S-T	138
triclocarban	CO_2		527 to 529	10 to 40	313.2 to 333.2	PR EOS, QLF EOS	83
2-trifluoromethylbenzoic acid	CO_2		380.7	9.34 to 22.6	308.2 to 323.2	PR EOS	140
3-trifluoromethylbenzoic acid	CO_2		376.8	9.41 to 22.54	308.2 to 323.2	PR EOS	140
4-trifluoromethylbenzoic acid	CO_2		497.3	9.68 to 22.44	308.2 to 323.2	PR EOS	140
3,4,5-trihydroxybenzoic acid	CO_2	ethanol	524 to 528	10 to 40	313.2 to 333.2	SRK EOS, GCA EOS	100
tripalmitin	CO_2		337 to 339	3.24 to 24.75	336 to 363	PR EOS	104
uracil	CO_2		>573	15 to 40	308 to 338	Chrastil, Chrastil-W	141
urea	CO_2		406 to 408	10 to 30	313 to 353	PR EOS	142
urea	CO_2	ethanol	406 to 408	15, 30	313 to 333	PR EOS	142
vanillin	CO2		355.5	8 to 27.65	313.2 to 353.2	PR EOS	93
vanillin	R23		355.5	3.13 to 20.25	313.2 to 333.2	Chrastil	94
vanillin	R134a		355.5	1.9 to 20.58	313.2 to 333.2	Chrastil	94
vanillin	R236fa		355.5	1.18 to 20.5	313.2 to 333.2	Chrastil	94
o-vanillin	CO_2		313.6	7.83 to 30.37	313.2 to 353.2	PR EOS	93
o-vanillin	R23		313.6	2.4 to 25.2	313.2 to 333.2	Chrastil	94
o-vanillin	R134a		313.6	5.2 to 24.6	313.2 to 333.2	Chrastil	94
o-vanillin	R236fa		313.6	7.9 to 26.3	313.2 to 333.2	Chrastil	94

[a] T_{fus}: melting temperature of compound X. [b] PLA: polylactic acid. [c] DH-PDC: 1,4-dihydro-2,6-dimethyl-4-(1-methyl-5-nitroimidazol-2-yl)-3,5-pyridinedicarboxylate. [d] PLGA: poly(lactide-*co*-glycolide). [e] PFPE-NH_4: ammonium carboxylate perfluoropolyether. [f] DCM: dichloromethane. [g] n.a.: data not available. [h] dec.: decomposes.

nonfluorinated compounds. The results obtained can represent the tendency of solubility enhancement by fluorination.[140]

2.4. Aromatic Compounds. Aromatic compounds that are not included in the previous group are summarized in Table 5. These include bipyridine derivatives and hydroxyquinoline derivatives used as chelating agents, anthraquinone-type disperse dyes, photochromic dyes, disperze azo dyes, polar acid dyes, pesticides, polycyclic aromatic hydrocarbons (PAHs), substituted phenols, etc.

The most commonly used SC solvent for these compounds was CO_2, but the solvent power of propane, water, and HFCs was also investigated. Investigations of the solubility of PAHs in propane indicated excellent solvent properties of propane for the extraction of polyaromatics in comparison with SC CO_2.[149] The values of the mole fraction of anthracene in propane are 2 orders of magnitude higher than those reported in the literature for anthracene in CO_2.[149]

The solubilities of two different bisphenol compounds [2,2-bis(4-hydroxyphenyl)propane (bisphenol A) and 2,2-bis(4-hydroxyphenyl)hexafluoropropane (bisphenol AF)] in three SCFs (CO_2, R152a, and R134a) were investigated by Liu et al.,[156] and the influence of the fluorine content of the solute or solvent on the solubility was studied. The results demonstrated that even a modest amount of fluorine substitution for hydrogen has a significant impact on the solubility levels of compounds. CO_2 is a poor solvent for bisphenol A and a modest solvent for bisphenol AF. This is in agreement with previous studies showing that fluorinated compounds, relative to their hydrocarbon analogues, have higher solubilities in CO_2.[156] The comparison of the two HFC solvents used in this study showed once again that fluorine substitution has a significant impact on the strength of solvents that have the same polarizability and dipole moment. R152a showed a greater solvent power than R134a because of the differences in molar volumes and the distances of fluorine and hydrogen separation in these two solvents.[156]

The solubilities of a variety of unsaturated carboxylic acids in SC R134a were determined by Abbott et al.,[144] who demonstrated that polar solutes have much higher solubilities in R134a than in CO_2 and that the energy requirements needed to obtain similar solubilities in pure CO_2 are over an order of magnitude higher than those for the R134a system.

A promising application of SCFs that has been developed in recent years because of environmental problems and economic considerations is a new attractive dyeing method with SCFs. The

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 5. Solubilities of Other Aromatic Compounds (X) in Solvents (S) + Cosolvents (C)

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
acenaphthene	H_2O		365 to 368	5 to 10	323 to 573	−	143
α-acetamidocinnamic acid	R134a		467	5 to 20	378	PR EOS	144
Acid Red 57 + hexadecyltri-methylammonium bromide	CO_2	methanol	solid, n.a.[d]	25 to 32.5	308 to 348	Bartle	145
Acid Red 57 + dodecyl-trimethylammonium bromide	CO_2	methanol	solid, n.a.[d]	25 to 32.5	318 to 373	Bartle	146
acridine	H_2O		383.2	5	313.2 to 363.2	−	147
anthracene	CO_2		489.7	11.7 to 20.1	318.2	−	148
anthracene	C_3H_8		489.7	4.7 to 11.3	298 to 405	PR EOS	149
anthracene	H_2O		487 to 489	5 to 10	323 to 573	−	143
anthracene	H_2O		483 to 488	4 to 8	313 to 483	solubility-vs-*T* model	150
anthracene + phenanthrene + carbazole	CO_2		−	11.1 to 20.1	308.2 to 318.2	−	148
anthracene oil	CO_2		n.a.[d]	15.1 to 20.1	308.2 to 318.2	−	148
9,10-anthraquinone	H_2O		558.0	5	313.2 to 433.2	solubility-vs-*T* model	151
anthrone	H_2O		428.2	5	313.2 to 423.2	solubility-vs-*T* model	151
1,2-benzanthracene	H_2O		430 to 432	4 to 8	313 to 423	solubility-vs-*T* model	150
biphenyl	CO_2		342 to 345	10 to 30	353 to 383	−	152
bis(2-(2-butoxyethoxy)ethyl)-2,2-bipyridine-4,4-dicarboxylate	CO_2		312.2 to 313.2	10.4 to 18.8	313 to 333	Bartle	153
bis(2-butoxyethyl)-2,2-bipyridine-4,4-dicarboxylate	CO_2		314.2 to 315.2	10.4 to 18.8	313 to 333	Bartle	153
1,4-bis(butylamino)-9,10-anthraquinone	CO_2		392	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(dodecylamino)-9,10-anthraquinone	CO_2		352	10 to 30	310 to 340	Carnahan−Starling−van der Waals EOS	155
1,4-bis(dodecylamino)-9,10-anthraquinone	N_2O		352	7.6 to 12.7	310 to 330	Carnahan−Starling−van der Waals EOS	155
bis(2-(2-ethoxyethoxy)ethyl)-2,2-bipyridine-4,4-dicarboxylate	CO_2		340.2 to 341.2	10.4 to 18.8	313 to 333	Bartle	153
1,4-bis(ethylamino)-9,10-anthraquinone	CO_2		468.5	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(hexadecylamino)-9,10-anthraquinone	CO_2		366.5	15 to 180	320 to 340	Carnahan−Starling−van der Waals EOS	155
1,4-bis(hexadecylamino)-9,10-anthraquinone	N_2O		366.5	10 to 160	310 to 330	Carnahan−Starling−van der Waals EOS	155
2,2-bis(4-hydroxyphenyl) hexafluoropropane	R152a		435.2	up to 15	343 to 453	−	156
2,2-bis(4-hydroxyphenyl) hexafluoropropane	R134a		435.2	10 to 20	403 to 453	−	156
2,2-bis(4-hydroxyphenyl) hexafluoropropane	CO_2		435.2	up to 200	303 to 413	−	156
2,2-bis(4-hydroxyphenyl) propane	R152a		431.2	up to 65	378 to 453	−	156
2,2-bis(4-hydroxyphenyl) propane	R134a		431.2	65	473	−	156
2,2-bis(4-hydroxyphenyl) propane	CO_2		431.2	200	473	−	156
1,4-bis(methylamino)-9,10-anthraquinone	CO_2		493	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(1-methyl ethylamino)-9,10-anthraquinone	CO_2		443.5	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(octylamino)-9,10-anthraquinone	CO_2		349	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(pentylamino)-9,10-anthraquinone	CO_2		389	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
1,4-bis(propylamino)-9,10-anthraquinone	CO_2		406	4.6 to 20	299 to 346	Chrastil, Chrastil-S-S, Bartle, dV-A, M-S-T	154
2,2-bis(*p*-tolyl) hexafluoropropane	CO_2		357.3	5.62 to 25.1	293.9 to 382.4	−	157
2,2-bis(*p*-tolyl) hexafluoropropane	R134a		357.3	0.58 to 8.71	295.2 to 447.7	−	157
2,2-bis(*p*-tolyl) hexafluoropropane	R152a		357.3	0.58 to 8.71	294.9 to 447.9	−	157
2,2-bis(*p*-tolyl)propane	CO_2		351.8	14.41 to 71.61	304.3 to 373.7	−	157
2,2-bis(*p*-tolyl)propane	R134a		351.8	1.93 to 12.19	336.4 to 448.7	−	157
2,2-bis(*p*-tolyl)propane	R152a		351.8	0.43 to 9.38	294.8 to 448.6	−	157
bitumen	Toluene		n.a.[d]	4.5 to 14.5	603.2 to 633.2	Chrastil	158

dx.doi.org/10.1021/je1011373 |*J. Chem. Eng. Data* 2011, 56, 694–719

Table 5. Continued

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
bitumen	Methanol		n.a.[d]	6.5 to 22.5	553.2 to 623.2	Chrastil	158
p-bromobenzaldehyde	CO_2		334.2	9 to 13.5	303 to 333	PR EOS	159
carbazole	CO_2		519.5	10.1 to 20.1	318.2	−	148
carbazole	H_2O		519.5	5	313.2 to 433.2	−	147
carbonylhydridotris[tris-(*p*-trifluoromethylphenyl) phosphine]rhodium	CO_2		solid, n.a.[d]	9 to 11.9	320 to 333	Chrastil	160
carbonylhydridotris-(triphenylphosphine) rhodium	CO_2		solid, n.a.[d]	9.9 to 16.9	333	Chrastil	160
4-chlorophenol	CO_2		313 to 318	8.8 to 15.6	308 to 313	Chrastil, M-S-T	162
4-chlorophenol	CO_2	methanol	313 to 318	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
4-chlorophenol	CO_2	acetone	313 to 318	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
4,4-diaminodiphenylmethane	CO_2		367.7	11 to 20	313.2 to 333.2	PR EOS, Chrastil, M-S-T	161
dibenzofuran	CO_2		355	6.5 to 27.5	301.3 to 338.2	Chrastil, PR EOS	163
dibenzofuran	CO_2	methanol	355	9.3 to 20.6	318.2 to 338.2	Pérez	164
dibenzofuran	CO_2	acetic acid	355	10.3 to 20.5	318.2 to 338.2	Pérez	164
dibenzofuran	H_2O		355.3	5	313.2 to 353.2	−	147
dibenzothiophene	H_2O		371.8	5	313.2 to 363.2	−	147
2,4-dichlorophenol	CO_2		315 to 316	8.8 to 15.6	308 to 313	Chrastil, M-S-T	162
2,4-dichlorophenol	CO_2	methanol	315 to 316	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
2,4-dichlorophenol	CO_2	acetone	315 to 316	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
dicofol	CO_2		344.4	10 to 25	293.2 to 313.2	K-J, Chrastil	165
1,3-dihydro-3,3-dimethyl-1-isopropyl-6-(2,3-dihydroindole-1-yl)spiro[2*H*-indole-2, 3-3*H*-naphtho[2,1-*b*] [1,4] oxazine	CO_2		450.2 to 452.2	10 to 26	308 to 328	Chrastil, Bartle, M-S-T, Z-E, PR EOS, SRK EOS	166
1,4-dihydroxy-9,10-anthraquinone	CO_2		471.2 to 472.2	15.2 to 35.5	308 to 348	PR EOS	167
1,4-dihydroxy-2-(prop-2-enyl)-9,10-anthraquinone	CO_2		412.2 to 413.2	15.2 to 35.5	308 to 348	PR EOS	167
9,10-dimethylanthracene	H_2O		455.2 to 457.2	5	313.2 to 448.2	solubility-vs-*T* model	168
4,6-dimethyldibenzothiophene	H_2O		426 to 430	5	313.2 to 423.2	−	147
2,5-dimethyl phenol	CO_2		348 to 350	10.1 to 28	308	−	169
2,3-dimethyl phenol	CO_2		346 to 348	10.1 to 28	308	−	169
1,4-dinethoxy-9,10-anthraquinone	CO_2		493.2 to 496.2	15.2 to 35.5	308 to 348	PR EOS	167
Disperse Blue 56	CO_2		453.4	15 to 30	353.2 to 393.2	Chrastil, M-S-T	171
Disperse Blue 56	CO_2	ethanol	453.4	15 to 30	353.2 to 393.2	Chrastil, M-S-T	171
Disperse Blue 56	CO_2	DMSO[c]	453.4	20 to 30	353.2	Chrastil, M-S-T	171
Disperse Blue 79	CO_2	ethanol	413	16 to 24.1	353 to 393	PR EOS, Chrastil-G, Ferri density-based	172
Disperse Blue 183	CO_2		solid, n.a.[d]	12 to 28	343.2 to 383.2	Chrastil, M-S-T	170
Disperse Orange 3	CO_2	methanol	468	16 to 24	353 to 393	Chrastil, K-J, Bartle, M-S-T	173
Disperse Orange 3	CO_2	ethanol	468	16 to 28	353 to 393	PR EOS, Chrastil-G, Ferri density-based	172
Disperse Red 60	CO_2		460	11 to 23	353 to 393	Bartle	174
Disperse Red 60	CO_2		460	10 to 30	333.2 to 393.2	MF-NLF EOS	175
Disperse Red 73	CO_2		solid, n.a.[d]	12 to 28	343.2 to 383.2	Chrastil, M-S-T	170
Disperse Red 73	CO_2		solid, n.a.[d]	12 to 28	343 to 383	Chrastil, M-S-T	176
Disperse Red 73 + Disperse Blue 183	CO_2		−	12 to 28	343.2 to 383.2	Chrastil, M-S-T	170
Disperse Red 73 + Disperse Yellow 119	CO_2		−	12 to 28	343 to 383	Chrastil, M-S-T	176
Disperse Violet 1	CO_2		516.3	15 to 30	353.2 to 393.2	Chrastil, M-S-T	177
Disperse Violet 1	CO_2	ethanol	516.3	15 to 30	353.2 to 393.2	Chrastil, M-S-T	177
Disperse Violet 1	CO_2	DMSO[c]	516.3	20 to 30	353.2	Chrastil, M-S-T	177
Disperse Yellow 54	CO_2		533	15 to 30	353.2 to 393.2	Chrastil, M-S-T	179
Disperse Yellow 54	CO_2	ethanol	533	15 to 30	353.2 to 393.2	Chrastil, M-S-T	179
Disperse Yellow 54	CO_2	DMSO[c]	533	20 to 30	353.2	Chrastil, M-S-T	179
Disperse Yellow 54	CO_2		543	10 to 30	333.2 to 393.2	MF-NLF EOS	175
Disperse Yellow 119	CO_2		solid, n.a.[d]	12 to 28	343 to 383	Chrastil, M-S-T	176
Disperse Yellow 184	CO_2		481 to 483	12.1 to 35.5	308 to 348	Chrastil, K-J, Bartle, M-S-T	178
Disperse Yellow 184-modified	CO_2		492 to 494	12.1 to 35.5	308 to 348	Chrastil, K-J, Bartle, M-S-T	178

dx.doi.org/10.1021/je1011373 |*J. Chem. Eng. Data* 2011, 56, 694–719

Table 5. Continued

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
Disperse Yellow 232	CO_2		467 to 469	12.1 to 35.5	308 to 348	Chrastil, K-J, Bartle, M-S-T	178
Disperse Yellow 232-modified	CO_2		498 to 500	12.1 to 35.5	308 to 348	Chrastil, K-J, Bartle, M-S-T	178
disperse dye: 6-{(*E*)-2-[4-(diethylamino)phenyl]-1-diazenyl}-2-propyl-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione	CO_2		555 to 556	12.2 to 35.5	308 to 348	Chrastil	180
disperse dye: ethyl 2-[6-{(*E*)-2-[4-(diethylamino)-2-methylphenyl]-1-diazenyl}-1,3-dioxo-1*H*-benzo[*de*]isoquinolin-2(3*H*)-yl] acetate	CO_2		568 to 569	12.2 to 35.5	308 to 348	Chrastil	180
disperse dye: ethyl 2-[6-{(*E*)-2-[3-hydroxy-2-naphthyl]-1-diazenyl}-1,3-dioxo-1*H*-benzo[*de*]isoquinolin-2(3*H*)-yl] acetate	CO_2		650 to 651	12.2 to 35.5	308 to 348	Chrastil	180
1-hydroxy-9,10-anthraquinone	CO_2		457.2 to 459.2	15.2 to 35.5	308 to 348	PR EOS	167
2-hydroxy-1-[4-(2-hydroxyethoxy)phenyl]-2-methyl-1-propanone (Irgacure 2959)	CO_2		362.2 to 393.2	10 to 26	308.2 to 328.2	Chrastil, Bartle, M-S-T, PR EOS	181
1-hydroxy-4-(prop-2-enyloxy)-9,10-anthraquinone	CO_2		400.2 to 401.2	15.2 to 35.5	308 to 348	PR EOS	167
6-methoxy-1-tetralone	R134a		351	5 to 20	378	PR EOS	144
2-methylanthracene	H_2O		477.2 to 479.2	5	313.2 to 453.2	solubility-vs-*T* model	168
p-methylbenzene sulfonic acid	CO_2		solid, n.a.[d]	8 to 21	308 to 328	Chrastil	182
p-methylbenzene sulfonic acid	CO_2	ethyl acetate	solid, n.a.[d]	8 to 21	308	Chrastil-cosolvent	182
p-methylbenzene sulfonic acid	CO_2	ethyl acetate + ethanol	solid, n.a.[d]	8 to 21	308	Chrastil-cosolvent	182
p-methylbenzene sulfonic acid	CO_2	ethanol	solid, n.a.[d]	8 to 21	308	Chrastil-cosolvent	182
p-methylbenzene sulfonic acid	CO_2	acetone	solid, n.a.[d]	8 to 21	308	Chrastil-cosolvent	182
2-methyl-*N*-phenylacetamide	CO_2		383.2	10 to 22	308.2 to 328.2	PR EOS, SRK EOS, Chrastil, M-S-T	183
4-methyl-*N*-phenylacetamide	CO_2		421.7	10 to 22	308.2 to 328.2	PR EOS, SRK EOS, Chrastil, M-S-T	183
1,5-naphthalenediamine	CO_2		463.1	11 to 20	313.2 to 333.2	PR EOS, Chrastil, M-S-T	161
naphthalene	CO_2		353	10 to 20.5	313.1 to 328.1	M-S-T	184
naphthalene	CO_2		353	up to 20.6	372.5 to 430.7	SRK EOS	60
naphthalene	CO_2		353	12 to 28	308	—	41
naphthalene	H_2O		350 to 355	4 to 8	313 to348	solubility-vs-*T* model	150
naphthalene + D,L-PLA[b]	CO_2		—	7.5 to 10	313.15	—	54
naphthalene + L-PLA[b]	CO_2		—	7.5 to 10	313.15	—	54
p-nitrobenzoic acid	CO_2		510 to 513	8 to 21	308 to 328	M-S-T	185
p-nitrobenzoic acid	CO_2	cyclohexane	510 to 513	8 to 21	308 to 328	M-S-T-cosolvent	185
p-nitrobenzoic acid	CO_2	ethanol	510 to 513	8 to 21	308 to 328	M-S-T-cosolvent	185
p-nitrobenzoic acid	CO_2	cyclohexane + acetone	510 to 513	8 to 21	308 to 328	M-S-T-cosolvent	185
p-nitrobenzoic acid	CO_2	acetone	510 to 513	8 to 21	308 to 328	M-S-T-cosolvent	185
permethrin	CO_2		319.5	10 to 25	293.2 to 313.2	K-J, Chrastil	165
phenanthrene	CO_2	acetone	371 to 373	1 to 12.5	295 to 350	—	186
phenanthrene	CO_2		371 to 373	11.3 to 33.2	308.2	—	106
phenanthrene	H_2O		372.4	5	313.2 to 363.2	—	147
phenanthrene	CO_2		372	10.1 to 20.1	318.2	—	148
phenanthrene	CO_2		372	16 to 28	318	—	41
9,10-phenanthrenequinone	H_2O		482 to 485	5	313.2 to 473.2	solubility-vs-*T* model	151
phenanthridine	H_2O		379.9	5	313.2 to 363.2	—	147
phenazine	H_2O		444.0	5	313.2 to 433.2	—	147
phenothiazine	H_2O		458.4	5	313.2 to 453.2	—	147
phenoxathiin	H_2O		328.8	5	313.2 to 328.2	—	147
phenoxazine	H_2O		429.9	5	313.2 to 413.2	—	147

dx.doi.org/10.1021/je1011373 |*J. Chem. Eng. Data* 2011, 56, 694–719

Table 5. Continued

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
N-phenylacetamide	CO_2		387.5	10 to 22	308.2 to 328.2	PR EOS, SRK EOS, Chrastil, M-S-T	183
9-phenylanthracene	H_2O		426.2 to 428.2	5	313.2 to 423.2	solubility-vs-*T* model	168
phenylboric acid	CO_2		490 to 493	10 to 30	353 to 383	−	152
4-phenylphenol	CO_2		439 to 440	10.1 to 28	308	−	169
4-phenylphenol + 2,3,5-trimethylphenol	CO_2		−	10.1 to 28	308	−	169
4-phenylphenol + 2,4,6-trimethylphenol	CO_2		−	10.1 to 24	308	−	169
4-phenyltoluene	CO_2		317 to 320	10 to 30	353 to 383	−	152
o-phthalic acid	CO_2		483.2 to 484.2	8 to 21	308.2 to 328.2	M-S-T	187
o-phthalic acid + *p*-aminobenzoic acid	CO_2		−	8 to 21	308.2 to 328.2	M-S-T	187
1-(prop-2-enyloxy)-9,10-anthraquinone	CO_2		413.2	15.2 to 35.5	308 to 348	PR EOS	167
pyrene	H_2O		429	5 to 10	323 to 573	−	143
quinacridone	H_2O		663	24	453.4 to 592.9	Anderson	188
Solvent Brown 1	CO_2	methanol	427	16 to 24	353 to 393	Chrastil, K-J, Bartle, M-S-T	173
Solvent Brown 1	CO_2	ethanol	427	18 to 30.1	353 to 393	PR EOS, Chrastil-G, Ferri density-based	172
sulfanilamide	CO_2		438.2	11 to 21	308 to 328	−	189
p-terphenyl	H_2O		485 to 486	4 to 8	313 to 483	solubility-vs-*T* model	150
4-*tert*-butyl phenol	CO_2		371 to 374	10.1 to 28	308	−	169
thianthrene	H_2O		429.6	5	313.2 to 423.2	−	147
thioxanthone	H_2O		487.9	5	313.2 to 473.2	solubility-vs-*T* model	151
p-toluenesulfonamide	CO_2		411.2	11 to 21	308 to 328	−	189
p-toluenesulfonamide	CO_2	ethanol	411.2	11 to 21	308 to 328	−	189
p-toluenesulfonamide	CO_2		411.2	8.0 to 21.0	308 to 328	Chrastil	190
p-toluenesulfonamide	CO_2	ethanol	411.2	8.0 to 21.0	308 to 328	M-S-T−Sauceau	190
p-toluenesulfonamide	CO_2	glycol	411.2	8.0 to 21.0	308	M-S-T−Sauceau	190
p-toluenesulfonamide + sulfanilamide	CO_2		−	11 to 21	308 to 328	Chrastil, M-S-T	189
p-toluenesulfonamide + sulfanilamide	CO_2	ethanol	−	11 to 21	308 to 328	Chrastil-G, M-S-T−Sauceau	189
2,4,6-trichlorophenol	CO_2	acetone	337 to 339	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
2,4,6-trichlorophenol	CO_2		337 to 339	8.8 to 15.6	308 to 318	Chrastil, M-S-T	162
2,4,6-trichlorophenol	CO_2	methanol	337 to 339	8.8 to 15.6	313	Chrastil-G, M-S-T-cosolvent	162
2,3,5-trimethylphenol	CO_2		365 to 368	10.1 to 28	308	−	169
2,4,6-trimethylphenol	CO_2		341 to 344	10.1 to 28	308	−	169
triphenylene	H_2O		468 to 471	4 to 8	313 to 468	solubility-vs-*T* model	150
triphenylmethyl chloride	CO_2		382 to 386	15 to 40	308.2 to 338.2	Gordillo, Chrastil	191
triphenylphosphine	CO_2		352 to 354	9 to 17.8	320 to 333	Chrastil	160
triphenyltin chloride	CO_2		376 to 379	15 to 30	308.2 to 338.2	Gordillo, Chrastil	191
triptycene	H_2O		525.2 to 527.2	5	313.2 to 513.2	solubility-vs-*T* model	168
tris(*p*-trifluoromethylphenyl)phosphine	CO_2		343 to 348	9 to 13.9	320 to 333	Chrastil	160
1,3,5-tri-*tert*-butylbenzene	CO_2		343.2	up to 20	298 to 328	−	192
2,4,6-tri-*tert*-butylphenol	CO_2		398 to 403	6.7 to 66.9	301 to 343	−	193
xanthene	CO_2		373.7	8 to 24	308.2 to 328.2	PR EOS, Chrastil	194, 195
xanthene	H_2O		373.7	5	313.2 to 368.2	solubility-vs-*T* model	151
xanthone	CO_2		447.2	12 to 30	308.2 to 328.2	PR EOS, Chrastil	194, 195
xanthone	H_2O		434.1	5	313.2 to 433.2	solubility-vs-*T* model	151

[a] T_{fus}: melting temperature of compound X. [b] PLA: polylactic acid. [c] DMSO: dimethyl sulfoxide. [d] n.a.: data not available.

SCF dyeing method has some advantages in comparison with conventional water-based dyeing: (1) the solubility of the dye can be controlled by pressure, allowing possible control of the dyeing strength and color; (2) contaminated wastewater streams are not produced; (3) washing and drying of the dyed fabric is not necessary; and (4) the SCF and the remaining dye in the cell of the solid textile can be reused after dyeing. Thus, the dyeing procedure is shorter than that for conventional methods.[145]

Dyeing of textile fibers in SC CO_2 has been limited to synthetic fibers using nonpolar disperse dyes, which are easily soluble in the SC CO_2, but the dyeing of natural fibers in this medium is still under development because of the low solubility of water-soluble polar dyes. In order to dye natural fibers, it is necessary to make such dyes soluble in SCFs. For this reason, the polar dyes are reacted with a hydrophobic ion-pairing reagent, after which they can be solubilized in SC CO_2.[145] Özcan and Özcan[145] measured the solubility of Acid Red 57 by ion-pairing with hexadecyltrimethylammonium (HDTMA) bromide and dodecyltrimethylammonium (DTMA) bromide[146] in SC CO_2. The results showed that Acid Red 57 is insoluble in SC CO_2 even with methanol as a cosolvent and that even the ion-pair forms of the acid dye (Acid Red 57−DTMA and −HDTMA) are essentially insoluble in SC CO_2 and require the addition of methanol as a cosolvent for measurable solubility to be achieved.

2.5. Nonaromatic Organic Compounds. Nonaromatic organic compounds that are not included in the group of biological and pharmaceutical compounds are summarized in Table 6; these include acyclic and cyclic compounds, such as keto acids, carboxylic acids, series of diglycolic acid esters, surfactants for microemulsions, diamondoid hydrocarbons, high-molecular-mass *n*-alkanes, *n*-alcohols, and alcohol ethoxylates, etc. The

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

solvents used for these compounds were propane and HFCs in addition to CO_2.

Liu et al.[18] measured the solubilities of a series of sodium bis(2-ethylhexyl) sulfosuccinate analogue surfactants in SC CO_2 and SC R134a. The solubilities of these surfactants were much higher in R134a than in SC CO_2. The solubilities in SC CO_2 increased with increasing number of carbon atoms in the surfactant, whereas those in R134a decreased with increasing number of carbon atoms in the surfactant.[18]

Schwarz et al.[197] studied the phase equilibria of high-molecular-mass 1-alcohols in supercritical propane. It was observed that the hydroxyl group has the effect of increasing the phase-equilibrium pressure because of the difference in polarity, thus decreasing the solubility of the alcohols in propane relative to that of *n*-alkanes.[197]

2.6. Polymers. The solubilities of polymers in CO_2, HFCs, DME, and hydrocarbons such as ethane and propane were measured at very high pressures up to 300 MPa (Table 7). The thermodynamic data for polymers in SCFs are important for the design of polymerization processes, fractionation, processing technologies, product development, and industrial applications.

SC CO_2 is a good solvent for many low-molecular-weight compounds and a few polymers, but it is generally a very poor solvent for high-molecular-weight polymers. However, its solubility in many polymers is substantial, being influenced by temperature, pressure, and weak interactions with the groups of the polymer. Dissolved CO_2 causes a reduction in the viscosity of the polymers by increasing their free volume. Thus, they are plasticized, allowing processing at lower temperatures. SC CO_2 has been used successfully as a polymerization medium and as a solvent, antisolvent, plasticizer, or blowing agent in polymer processing, including polymer modification, formation of polymer composites, polymer blending, microcellular foaming, and particle production.

However, it has been previously demonstrated that polymers possessing electron-donating functional groups display Lewis acid−base interactions in CO_2 with carbonyls, acetates, and ethers, which significantly enhance the solubilities of the polymers in CO_2. Small sugar acetates are known to be extraordinarily CO_2-philic, and poly(vinyl acetate) (PVAc) is one of the most CO_2-soluble nonfluorous polymers identified to date.[224]

The solubility of an amorphous high-molecular-weight polymer with pendant sugar acetates [poly(1-*O*-(vinyloxy)ethyl-2,3,4,6-tetra-*O*-acetyl-β-D-glucopyranoside), P(AcGIcVE)] in SC CO_2 was studied by Tapriyal et al.[224] The solubility of this polymer in CO_2 was compared to the solubilities of PVAc and polylactic acid (PLA) in CO_2. The results showed that PVAc remains the most CO_2-soluble high-molecular-weight oxygenated hydrocarbon polymer identified to date, followed by P(AcGIcVE) and amorphous PLA.

In the work of Milanesio et al.,[225] cloud points of binary mixtures of propane, diethyl ether (DEE), or DME with polybutadiene (PB) and mixtures of DEE with high-density polyethylene (PE) were experimentally determined. The temperature range studied was from (323 to 433) K for PB + DME and PB + propane and from (373 to 463) K for PB + DEE and PE + DEE. The results showed that in PB-containing binary systems, over the ranges of conditions of the experiments, the minimum pressure required to guarantee homogeneity at any temperature is below 20 MPa for DEE, below 30 MPa for DME, and on the order of 50 MPa for propane as the solvent.[225] For the PE + DEE system, a minimum pressure of about 24 MPa is needed in order to keep the system within a single phase.[225]

The *P*, *T* cloud-point curves of high-molecular-weight PE in hydrocarbons, halogenated hydrocarbons, oxygen-containing hydrocarbons, and their mixtures at high temperatures and high pressures were measured by Kojima et al.[221] In addition to the solvents included in Table 7, that study also examined the hydrocarbons butane, 2-methylbutane, 2,2-dimethylbutane, 2-methylpentane, 2,3-dimethylbutane, 3-methylpentane, and heptane and the halogenated hydrocarbons chloroethane, 2-chloropropane, 2-chlorobutane, and dichloromethane. It was found that the cloud-point pressures of the solutions showed a good correlation with the critical temperatures of the solvents.[221]

In the work of Galy et al.,[226] the phase behavior of poly(ethylene oxide)-*b*-poly(propylene oxide)-*b*-poly(ethylene oxide) triblock copolymers in liquid and SC CO_2 was studied by cloud-point measurements. The results showed that such trade hydrocarbonated surfactants are fairly soluble (0.001 mass fraction) in CO_2 under relatively mild conditions of temperature and pressure (T < 338 K, P < 30 MPa).

3. MODELING

There are two major approaches to modeling solubilities in SCFs: equation of state (EOS) modeling and empirical density-based correlations.[154] EOS modeling usually requires the selection of the most appropriate equation and mixing rules and also knowledge of pure-component parameters of the solute. Thus, solubility modeling by EOS is a challenging task. On the other hand, the use of empirical models does not require knowledge of additional solute properties and is therefore quite popular in chemical engineering, where such models have often been found to be very successful in correlating existing solubility data.

3.1. Empirical Models. The empirical models most often applied for modeling of the solubilities of solids in SCFs are summarized in Table 8.

The density-based models are based on the linear relationship between the logarithm of the solubility and the logarithm of the solvent density (or the solvent density itself) within a certain range of pressure and temperature. Generally, these models give good results for a solvent-density region from 10 MPa to approximately 30 MPa, where not only technical SCF processes usually operate but also most solubility data are collected.[154] Such a correlation is bound to fail for more extreme density regimes, such as when the solvent density becomes liquidlike under the experimental conditions.[154]

The Chrastil,[227] Kumar and Johnston (K-J),[230] Bartle,[231] Mendez-Santiago and Teja (M-S-T),[232] and del Valle and Aguilera (dV-A)[235] models and their modifications (see Table 8) relate the solubility of a solute (j) to the temperature and density of the SCF. Yu et al.[236] and Gordillo et al.[237,238] proposed equations that relate the solubility of the solute to the pressure and the temperature. In these models, ρ, T and P are the density of the SCF, temperature, and pressure, respectively, γ_j is the mass concentration of the solute, y_j is the mole fraction of the solute, and the a_i are the model constants. In the Bartle model,[231] P_{ref} is the reference pressure (0.1 MPa) and ρ_{ref} is reference density (700 kg·m^{-3}).

In the Chrastil model,[227] a_1 is an association number that represents the average number of SCF molecules in the solvated complex. The constant a_0 depends on the molecular weights of the solute (M_j) and supercritical fluid (M_{SCF}).

Table 6. Solubilities of Nonaromatic Organic Compounds (X) in Solvents (S) + Cosolvents (C)

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
adamantane	H_2O		482 to 485	5	313 to 493.2	−	196
bis(2,2,3,3,3-pentafluoropropyl) 2-sulfosuccinate sodium salt	CO_2	water	511.9 to 512.9	12 to 25	308 to 338	−	198
bis(2,2,3,3,3-pentafluoropropyl) 2-sulfosuccinate sodium salt	R134a	water	511.9 to 512.9	up to 40	308 to 333	−	198
bis(2,2,3,4,4,4- hexafluorobutyl) 2-sulfosuccinate sodium salt	R134a	water	414.6 to 417.8	up to 40	308 to 343	−	198
crotonic acid	R134a		344	5 to 20	378	PR EOS	144
crotonic acid	CO_2		344	5 to 20	378		144
1-decanol	C_3H_8		278 to 280	3.7 to 6.76	378 to 408	−	197
diamantane	H_2O		solid, n.a.[b]	5	313 to 493.2	−	196
diamantane	C_3H_8		solid, n.a.[b]	0.3 to 14	360 to 510 K	−	199
diamantane	C_2H_6		solid, n.a.[b]	0.3 to 200	360 to 510	−	199
dibutyl 2,2′-oxidiacetate	CO_2		liquid	8.6 to 12.7	313 to 333	Bartle, Chrastil	200
dibutyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	25	318	−	201
dibutyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	10 to 30	308 to 338	−	18
dibutyl-2-sulfosuccinate sodium salt	CO_2		solid, n.a.[b]	10 to 30	308 to 338	−	18
didecyl-2,2′-oxidiacetate	CO_2		312.4	9.3 to 15.2	313 to 333	Bartle, Chrastil	200
didodecyl-2,2′-oxidiacetate	CO_2		314.7	9.2 to 14.5	313 to 333	Bartle, Chrastil	200
diheptyl-2,2′-oxidiacetate	CO_2		liquid	8.7 to 14.1	313 to 333	Bartle	202
diheptyl-2,2′-oxidiacetate	CO_2		liquid	12.6 to 17.7	343 to 363	Bartle	203
dihexadecyl-2,2′-oxidiacetate	CO_2		323.3	13.4 to 20.4	313 to 333	Bartle, Chrastil	200
dihexyl-2,2′-oxidiacetate	CO_2		liquid	8.4 to 12.4	313 to 333	Bartle, Chrastil	200
dihexyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	25	318	−	201
dihexyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	10 to 30	308 to 338	−	18
dihexyl-2-sulfosuccinate sodium salt	CO_2		solid, n.a.[b]	10 to 30	308 to 338	−	18
dinonyl-2,2′-oxidiacetate	CO_2		liquid	9.1 to 14.2	313 to 333	Bartle	202
dinonyl-2,2′-oxidiacetate	CO_2		liquid	13.6 to 19.7	343 to 363	Bartle	203
dioctyl-2,2′-oxidiacetate	CO_2		liquid	8.8 to 13.7	313 to 333	Bartle, Chrastil	200
dioctyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	25	318	−	201
dioctyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	30	318	−	18
dioctyl-2-sulfosuccinate sodium salt	CO_2		solid, n.a.[b]	30	318	−	18
dipentadecyl-2,2′-oxidiacetate	CO_2		solid, n.a.[b]	11 to 19.3	313 to 333	Bartle	202
dipentyl-2,2′-oxidiacetate	CO_2		liquid	8.5 to 12.5	313 to 333	Bartle	202
dipentyl-2,2′-oxidiacetate	CO_2		liquid	11.8 to 16.7	343 to 363	Bartle	203
dipentyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	16.5 to 33	308 to 338	−	201
dipentyl-2-sulfosuccinate sodium salt	R134a		solid, n.a.[b]	30	318	−	18
dipentyl-2-sulfosuccinate sodium salt	CO_2		solid, n.a.[b]	15 to 35	308 to 338	−	201
dipentyl-2-sulfosuccinate sodium salt	CO_2		solid, n.a.[b]	30	318	−	18
ditetradecyl-2,2′-oxidiacetate	CO_2		320	10.3 to 16.9	313 to 333	Bartle, Chrastil	200
ditridecyl-2,2′-oxidiacetate	CO_2		solid, n.a.[b]	9.9 to 15.6	313 to 333	Bartle	202
diundecyl-2,2′-oxidiacetate	CO_2		solid, n.a.[b]	9.2 to 14.5	313 to 333	Bartle	202
diundecyl-2,2′-oxidiacetate	CO_2		solid, n.a.[b]	13.7 to 18.1	343 to 363	Bartle	203
1-docosanol	C_3H_8		338 to 345	4.11 to 8.37	378 to 408	−	197
1-docosanol	C_3H_8		338 to 345	4.11 to 8.37	378 to 408	−	197
1-dodecanol	C_3H_8		295 to 299	3.51 to 6.89	378 to 408	−	197
n-eicosane	C_3H_8		308 to 310	0.84 to 2.19	287.33 to 333.45	PR EOS	204
n-eicosane	CO_2		308 to 310	7.12 to 25.0	299.45 to 333.45	PR EOS	204
ethanamide	CO_2		354.2	9 to 40	308.2 to 333.2	SRK EOS	205
ethyl myristate	C_3H_8		285	3.41 to 6.8	378 to 408	−	206
1-hexadecanol	C_3H_8		322 to 323	4.04 to 7.58	378 to 408	−	197
itaconic acid	R134a		439	3 to 20	378 to 393	PR EOS	144
itaconic acid	CO_2		439	5 to 20	378		144
itaconic acid + methylsuccinic acid	R134a		−	3 to 20	383	PR EOS	144
levulinic acid	CO_2		303 to 306	8.0 to 19.0	313 to 342.4	Chrastil	207
levulinic acid	CO_2	ethanol	303 to 306	8.0 to 19.0	313 to 342.4	Chrastil-G	207

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 6. Continued

X	S	C	T_{fus}/K[a]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
maleic acid	CO_2		410 to 413	7 to 30	318 to 348.2	Chrastil, M-S-T	208
methylsuccinic acid	R134a		388	5 to 20	378 to 383	PR EOS	144
methyl tetradecanoate	C_3H_8		292	3.42 to 6.83	378 to 408	–	206
1-octadecanol	C_3H_8		331 to 334	3.92 to 7.66	378 to 408	–	197
2-propenamide	CO_2		357.7	9 to 40	308.2 to 333.2	SRK EOS	205
tetradecane	C_3H_8		279	2.97 to 6.48	378 to 408	–	206
tetradecanoic acid	C_3H_8		326	3.87 to 8.48	378 to 408	–	206
tridodecylamine	CO_2		289	8 to 40	308.1 to 328.1	Bartle, M-S-T	209
Unilin 550 alcohol	C_3H_8		372	3.46 to 13.88	378 to 408	–	210
Unithox 550 ethoxylate	C_3H_8		372	4.83 to 26.84	378 to 408	–	210

[a] T_{fus}: melting temperature of compound X. [b] n.a.: data not available.

Table 7. Solubilities of Polymers and Monomers (X) in SCFs (S) in Systems with or without Cosolvents (C)

X	S	C	T_{fus}/K[a]	M_w/kg·mol^{-1} [b]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
decyl acrylate	CO_2		n.a.[e]	monomer	3.07 to 22.3	313 to 393	PR EOS	211
decyl methacrylate	CO_2		229	monomer	3.27 to 23.1	313 to 393	PR EOS	211
isobutyl acrylate	CO_2		213	monomer	2.4 to 14.8	313 to 393	PR EOS	212
isobutyl methacrylate	CO_2		236	monomer	3.3 to 15.9	313 to 393	PR EOS	212
L-lactide	CO_2		389 to 392	monomer	10 to 80	275 to 355	PR EOS	213
poloxamer F-127	CO_2	ethanol	293 to 333	12.7	10	298	–	214
poly(ε-caprolactone)	R22		n.a.[e]	14	125	up to 413.2	–	215
poly(ε-caprolactone)	R22 + CO_2		n.a.[e]	14	2.7 to 125.5	323.3 to 415	–	215
poly(ε-caprolactone)	DME[d] + CO_2		n.a.[e]	14	2.7 to 125.5	322.9 to 415	–	215
poly(ε-caprolactone)	CO_2		n.a.[e]	14, 170	200 to 283	393 to 503	–	216
poly(ε-caprolactone)	R22		n.a.[e]	170	4.3 to 45.5	328 to 473	–	216
poly(ε-caprolactone)	C_4H_8		n.a.[e]	170	161 to 284	425 to 485	–	216
poly(ε-caprolactone)	C_3H_6		n.a.[e]	170	182 to 277	419 to 274	–	216
poly(ε-caprolactone)	CO_2	DME[d]	n.a.[e]	170	115 to 283	323 to 473	–	216
poly(ε-caprolactone)	CO_2	R22	n.a.[e]	170	59 to 2 80	323 to 473	–	216
poly(ε-caprolactone)	C_3H_6	DME[d]	n.a.[e]	170	92 to 270	323 to 473	–	216
poly(ε-caprolactone)	C_3H_6	R22	n.a.[e]	170	60 to 260	323 to 473	–	216
poly(ε-caprolactone)	C_4H_8	DME[d]	n.a.[e]	170	32 to 254	313 to 473	–	216
poly(ε-caprolactone)	C_4H_8	R22	n.a.[e]	170	85 to 265	323 to 473	–	216
poly(ε-caprolactone)	DME[d]		n.a.[e]	14, 170	14 to 50	328 to 473	–	216
poly(ε-caprolactone)	CO_2 + dichloromethane		n.a.[e]	n.a.[e]	up to 21	303 to 343	–	217
polycaprolactone	CO_2	ethanol	303 to 313	1	10 to 25	310 to 330	PHSC EOS	218
poly(cylcohexene carbonate)	CO_2		n.a.[e]	12	252 to 335	455 to 480	PC-SAFT	219
poly(cylcohexene carbonate)	CO_2	cyclohexene oxide	n.a.[e]	12, 25, 54	367 to 466	115 to 363	PC-SAFT	219
poly(decyl acrylate)	CO_2		n.a.[e]	130	145 to 194	451 to 479	–	211
poly(decyl acrylate)	CO_2	decyl acrylate	n.a.[e]	130	138 to 210	321 to 473	–	211
poly(decyl acrylate)	C_3H_8		n.a.[e]	130	16.9 to 28.6	332 to 454	–	211
poly(decyl acrylate)	C_3H_6		n.a.[e]	130	9.7 to 27.8	334 to 454	–	211
poly(decyl acrylate)	C_4H_{10}		n.a.[e]	130	1.9 to 14.3	354 to 455	–	211
poly(decyl acrylate)	C_4H_8		n.a.[e]	130	5.3 to 14	395 to 455	–	211
poly(decyl acrylate)	DME[d]		n.a.[e]	130	4.4 to 17.8	374 to 455	–	211
poly(decyl methacrylate)	CO_2	decyl methacrylate	n.a.[e]	100	11.7 to 233.3	314 to 458	–	211
polyethylene	C_3H_8		n.a.[e]	0.8, 7, 23.625, 52, 59.3	5 to 50	360 to 390	–	220
polyethylene	C_2H_6		n.a.[e]	0.8, 7, 23.625, 52, 59.3	30 to 140	370 to 425	–	220

Table 7. Continued

X	S	C	T_{fus}/K[a]	M_w/kg·mol^{-1} [b]	P_{min}/MPa to P_{max}/MPa	T_{min}/K to T_{max}/K	correlation model	ref
polyethylene	C_2H_4		n.a.[e]	0.8, 7, 23.625, 52, 59.3	30 to 200	370 to 425	−	220
polyethylene	C_5H_{12}		n.a.[e]	102	10 to 23	440 to 510	−	221
polyethylene	R11		n.a.[e]	102	up to 20	440 to 510		221
polyethylene	C_6H_{14}		n.a.[e]	102	up to 15	440 to 510		221
poly(isobutyl acrylate)	CO_2		n.a.[e]	120	91.5 to 182.2	324 to 423	−	212
poly(isobutyl acrylate)	CO_2	isobutyl acrylate	n.a.[e]	120	7 to 91.7	304 to 428	−	212
poly(isobutyl methacrylate)	CO_2		n.a.[e]	200	122 to 204.7	356 to 435	−	212
poly(isobutyl methacrylate)	CO_2	isobutyl methacrylate	n.a.[e]	200	42 to 172.2	316 to 436	−	212
poly(L-lactide)	R22		n.a.[e]	80, 110, 230	3.6 to 26.1	332.3 to 410.8	−	215
poly(L-lactide)	R23		n.a.[e]	80, 110, 230	93.1 to 116	333.1 to 415.3	−	215
poly(L-lactide)	R32		n.a.[e]	80, 110, 230	77.0 to 111.3	334.6 to 416.8	−	215
poly(L-lactide)	R152a		n.a.[e]	80, 110, 230	53.2 to 64.2	352.9 to 415.2	−	215
poly(L-lactide)	DME[d]		n.a.[e]	80, 110, 230	3.1 to 24.9	331.4 to 412.6	−	215
poly(L-lactide)	R22 + CO_2		n.a.[e]	2	3.6 to 44.1	314.2 to 395.7	−	215
poly(L-lactide)-PEG-poly(L-lactide)[c]	CO_2	dichloromethane	n.a.[e]	n.a.[e]	up to 35	313.1 to 338.1	−	222
poly(L-lactide)-PEG-poly(L-lactide)[c]	CO_2	dichloromethane + ethanol	n.a[e]	n.a.[e]	up to 35	313.1 to 338.1	−	222
polystyrene	DME[d]		n.a.[e]	45	25.2 to 44	300.7 to 408.6	−	215
poly(*tert*-butyl acrylate)	CO_2		n.a.[e]	250	121.5 to 206.4	395 to 458	−	212
poly(*tert*-butyl acrylate)	CO_2	*tert*-butyl acrylate	n.a.[e]	250	18.2 to 179.1	319.7 to 425	−	212
poly(*tert*-butyl methacrylate)	CO_2		n.a.[e]	180	191.3 to 257.6	435 to 464	−	212
poly(*tert*-butyl methacrylate)	CO_2	*tert*-butyl methacrylate	n.a.[e]	180	6.24 to 217.3	315 to 436	−	212
poly(tetrafluoroethylene)	CO_2		n.a.[e]	n.a.[e]	10 to 70	318 to 383	Chrastil, Bartle, M-S-T	223
poly(1-*O*-(vinyloxy)ethyl-2,3,4,6-tetra-*O*-acetyl-*β*-D-glucopyranoside	CO_2		n.a.[e]	6, 20, 40	30 to 80	298	−	224
tert-butyl acrylate	CO_2		204	monomer	2.4 to 14.0	313 to 393	PR EOS	212
tert-butyl methacrylate	CO_2		213	monomer	3.3 to 14.7	313 to 393	PR EOS	212

[a] T_{fus}: melting temperature of compound X. [b] M_w: molecular weight of polymer investigated. [c] PEG: poly(ethylene glycol). [d] DME: dimethyl ether. [e] n.a.: data not available.

The constant a_2 in the Chrastil and K-J models is related to total enthalpy change (ΔH) as $a_2 = \Delta H/R$, where R is the gas constant; ΔH is equal to the sum of the enthalpy of vaporization ($\Delta_{vap}H$) and the enthalpy of solvation ($\Delta_{solv}H$) of the solute, i.e., $\Delta H = \Delta_{vap}H + \Delta_{solv}H$.

In the Bartle model, the parameter a_2 can be directly used to estimate $\Delta_{vap}H$ as $\Delta_{vap}H = -a_2R$. On the basis of the values of ΔH and $\Delta_{vap}H$, the value of $\Delta_{sol}H$ can be approximated for each solute−CO_2 system.

In the modification to the Chrastil model proposed by Garlapati and Madras,[126] f^0 is the fugacity of the gas, which was considered to be 1 atm.

Mendez-Santiago and Teja[233] presented a semiempirical density-based model for ternary cosolvent systems by adding a new parameter and the cosolvent concentration (y_c). Because the sublimation pressures of the solid are not often available, a modified M-S-T equation was proposed by Sauceau et al.,[234] in which ρ_f is the density of the fluid (solvent + cosolvent mixture) and P° is the standard pressure.

The M-S-T model developed for ternary cosolvent systems[233] did not yield a satisfactory fit to the data when a solid cosolvent was used. Hence, in the work of Thakur and Gupta,[102] a further-modified form of the M-S-T model was proposed for the solubility of griseofulvin in CO_2 enhanced by menthol as a cosolvent; in this modification, a_2 and a_3 are parameters related to the cosolvent effect and the concentration y_c is the cosolvent mole fraction.

The modification of the Chrastil model proposed by Wang[143] was derived from the solvation concept, the equilibrium constant

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

Table 8. Empirical Models

model (denotation)	correlation	ref
Chrastil	$\ln \gamma_j = a_0 + a_1 \ln \rho + \frac{a_2}{T}$	227
Chrastil modified by Garlapati and Madras (Chrastil-G-M)	$y_j = \left(\frac{RT\rho}{M_{SCF} f^0}\right)^{a_1 - 1} + \exp\left(a_0 + \frac{a_2}{T}\right)$	126
Chrastil modified by Wang (Chrastil-W)	$\ln y_j = a_0 + a_1\rho + \frac{a_2}{T} + a_3 \ln P$	143
Chrastil modified by Sung and Shim (Chrastil-S-S)	$\ln y_j = a_0 + \frac{a_2}{T} + \left(a_1 + \frac{a_4}{T}\right) \ln \rho$	228
Chrastil modified by González (Chrastil-G)	$\ln \gamma_j = a_0 + a_1 \ln \rho + \frac{a_2}{T} + a_3 \ln \gamma_c$	229
Chrastil modified by Adachi and Lu (Chrastil-A-L)	$\ln \gamma_j = a_0 + (e_0 + e_1\rho + e_2\rho^2) \ln \rho + \frac{a_2}{T}$	120
Kumar and Johnston (K-J)	$\ln y_j = a_0 + a_1\rho + \frac{a_2}{T}$	230
Bartle	$\ln\left(\frac{y_j P}{P_{ref}}\right) = a_0 + a_1(\rho - \rho_{ref}) + \frac{a_2}{T}$	231
Mendez-Santiago and Teja (M-S-T)	$T \ln(y_j P) = a_0 + a_1\rho + a_2 T$	232
M-S-T−Thakur and Gupta	$T \ln(y_j P) = a_0 + a_1\rho + (a_2\rho + a_3) y_c$	102
M-S-T-cosolvent	$T \ln\left(\frac{y_j P}{P^{sub}}\right) = a_0 + a_1\rho + a_2 y_c$	233
M-S-T−Sauceau	$T \ln\left(\frac{y_j P}{P^{\circ}}\right) = a_0 + a_1\rho_f + a_2 y_c + a_3 T$	234
del Valle and Aguilera (dV-A)	$\ln \gamma_j = a_0 + a_1 \ln \rho + \frac{a_2}{T} + \frac{a_3}{T^2}$	235
Yu et al.	$y_j = a_0 + a_1 P + a_2 P^2 + a_3 PT(1 - y_2) + a_4 T + a_5 T^2$	236
Gordillo et al.	$\gamma_j = a_0 + a_1 P + a_2 P^2 + a_3 PT + a_4 T + a_5 T^2$	237, 238
Ziger and Eckert (Z-E)	$\ln E = a_1\left[\varepsilon_j \frac{\Delta}{y_{SCF}}\left(2 - \frac{\Delta}{y_{SCF}}\right) - \log\left(1 + \frac{{\delta_{SCF}}^2}{P}\right)\right] + a_0$	239
Anderson et al.	$\ln K = \ln K_r - \frac{\Delta H_r^{\circ}}{R}\left(\frac{1}{T} - \frac{1}{T_r}\right) + \frac{\Delta c_{p,r}}{RT_r(\partial\alpha/\partial T)_{p,r}}\left[\frac{1}{T}\ln\frac{\rho}{\rho_r} - \frac{\alpha_r}{T}(T - T_r)\right]$	240, 188
Pérez et al.	$P = (c_1 + a_1 y_c) {y_j}^2 + (c_2 + a_2 y_c) y_j + (c_3 + a_3 y_c)$	164
Wubbolts et al.	$y_s^{eq} = y_s^0 (1 - x_{CO_2})^{A + Bx_{CO_2}} + y_s^{CO_2} x_{CO_2}$	241
Sovova	$y_2 - y_1 = k {y_3}^m {y_1}^n$	242
modified Sovova	$y_2 - y_1 = k {y_3}^m {y_1}^n e^{b/T}$	88

of the high-pressure reaction, and the rules of phase equilibrium. Therefore, an improved correlation with four adjustable parameters was obtained. In this expression, the parameter a_0 is dependent on the entropy of solvation (ΔS^0) and vapor pressure (P^{sat}) of the solute through the relation $a_0 = \Delta S^0/R + \ln P^{sat}$. The value of a_2 is dependent on the enthalpy of solvation, the molar volume (V_s^s), and the vapor pressure of the solute and is given by $a_2 = -(\Delta H^0 + V_s^s P^{sat})/R$. The constant a_1 is related to the second virial coefficient of the solute (B_{22}), the mixed second virial coefficient (B_{12}), the molar volume, and the association number (k) through the expression $a_1 = -[2(B_{12} - kB_{22}) + (k - 1)B_{11}] + V_s^s$. Finally, a_3 is a function of k given by $a_3 = k - 1$.

Another modification of Chrastil equation, proposed by González,[229] includes the influence of cosolvent on the solute solubility, where γ_c is the mass concentration of the cosolvent and a_1 and a_3 are the association numbers of the SCF and the cosolvent, respectively.

In a modification of the Chrastil equation proposed by Adachi and Lu,[120] the association number a_1 is a quadratic function of the density of the SCF (rather than a constant, as Chrastil assumed).

For each of these models, the coefficients a_i can be obtained by fitting the correlation to the experimental data.

A semiempirical correlation based on regular solution theory and the van der Waals (VdW) EOS was developed by Ziger and Eckert.[239] In this approach, the VdW EOS and mixing rules are used to evaluate the solute's fugacity coefficient in the SCF in terms of solute and solvent solubility parameters. In their expression, $E = y_j P/P_j^{sub}$ is the enhancement factor, defined as the ratio of the observed equilibrium solubility to that predicted by the ideal gas law under the same temperature and pressure conditions; ε_j is a dimensionless energy parameter given by $\varepsilon_j = \delta_j V_j^L/(2.3RT)$; and Δ is the ratio of the solvent's and solute's solubility parameters. Finally, a_1 and a_0 are two empirical constants that are obtained by regression of the experimental data. The parameters δ_j and δ_{SCF} are the solubility parameters of the solute and solvent, respectively, P_j^{sub} is the sublimation pressure of the solid solute, and V_j^L is the solute's molar volume in the liquid state.

Anderson et al.[240] proposed the simplified density model for analyzing dissolution processes under given conditions. In this model, K, ΔH°, and Δc_p denote the equilibrium constant, standard enthalpy, and heat capacity for the dissolution process,

dx.doi.org/10.1021/je1011373 |J. Chem. Eng. Data 2011, 56, 694–719

respectively, R is the gas constant, and T, ρ, and α stand for absolute temperature, density, and thermal expansivity, respectively. The subscript r denotes the reference state of 298.15 K and 0.1 MPa.

Pérez et al.[164] used an empirical correlation in which P is a function of the solute and cosolvent mole fractions and c_1 to c_3 and a_1 to a_3 are fitting parameters.

The empirical equation proposed by Wubbolts et al.[241] describes the dependence of the solubility of a compound on the mole fraction of CO_2 (used as an antisolvent) in the mixture with an organic solvent. It expresses the solubility y_s^{eq} as a function of the concentration of carbon dioxide x_{CO_2} in the solvent mixture (both on a solute-free basis). The form of the equation ensures that the correct solubility is reproduced for the limits of pure solvent (y_s^0) and pure antisolvent ($y_s^{CO_2}$). The equation is fitted to the data with the constants A and B.

A commonly used semiempirical model by Sovova[242] has been widely used to correlate the solubilities of solids in SC CO_2 with cosolvent. In the model, y_2 is the mole-fraction solubility of the solute in SC CO_2 with cosolvent, y_1 is the mole-fraction solubility of solute in pure SC CO_2, and y_3 is the mole fraction of the cosolvent in SC CO_2; k, m, and n are parameters independent of temperature and pressure that are obtained by fitting to experimental data.

The Sovova model contains three adjustable parameters (k, m, and n). It is clear that y_2 is related to y_1 and y_3 but not related to temperature (T). When y_3 remains invariant as T changes, the number of parameters in the equation decreases. Therefore, in the modified Sovova model,[88] a function of temperature (T) is added, and k, m, n, and b are the equation's parameters independent of temperature and pressure; y_1, y_2, and y_3 are the same as those in Sovova model.

For the description of the solubility of a salt/water system, two relevant methods can be taken into consideration. Equations of state can be used as well as empirical and semiempirical approaches. A review of models is given in the literature.[26]

3.2. Thermodynamic Models. EOSs are commonly applied in engineering practice since they can be used to predict the thermodynamic properties of fluids and describe the phase behavior of mixtures over large ranges of temperature and pressure. Detailed reviews of the thermodynamic models applied for predicting phase behavior and modeling aspects in SCF mixtures have been presented elsewhere.[243−245]

The EOSs used for modeling the phase equilibria of the systems in Tables 2to 7 are the Soave−Redlich−Kwong (SRK) EOS, the Peng−Robinson (PR) EOS, the group contribution (GC) EOS, the modified universal functional activity coefficient (M-UNIFAC) model, the perturbed hard-sphere chain (PHSC) EOS, the quasi-chemical nonrandom lattice fluid (QLF) EOS, a regular-solution model with a Flory−Huggins term (a solution model), and the multifluid nonrandom lattice fluid (MF-NLF) EOS.

4. SUMMARY

Most of experimental solubility data for solid compounds in sub- and supercritical fluids reported in the literature between 2005 and 2010 are for binary systems. Data were found for over 380 different pure solid compounds, for which solubilities were measured in different sub- and supercritical fluids without or with cosolvents. Solubility data for binary, ternary, or multicomponent solid mixtures are still limited, as only 29 binary, two ternary, and one multicomponent solid systems have been investigated. In the future, investigations of complex multicomponent solid systems should be performed in order to understand the behavior of real complex systems and the interactions between molecules in such systems. Most of the solubility data are for biological and pharmaceutical compounds. The overview of models used for phase equilibrium calculations showed that the empirical and semiempirical models are preferred for modeling the solubility of solids in SCFs. This is a result of the fact that they do not require the knowledge of additional solute properties and often give satisfying results in correlating experimental solubility data.

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