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Semi-empirical correlation of solid solute solubility in supercritical carbon dioxide: Comparative study and proposition of a novel density-based model

Corrélation semi-empirique de solubilité des solutés solides dans le dioxyde de carbone supercritique: étude comparative et proposition d'un nouveau modèle basé sur la densité

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ABSTRACT

A comprehensive data set on experimental solubility of 210 solid solutes in supercritical CO_2 counting 5550 data points has been used for comparison of the correlation performance of 21 empirical models. On the basis of the comparison results a new eight-parameter density-based model has been proposed. The comparison shows that the three-parameter models are the least accurate. The results also show that models that relate the logarithm of the solubility to the logarithm of solvent density and temperature are more accurate than models that include the pressure. When comparing the overall correlating performance in terms of average absolute relative deviation the proposed model is by far the best with an average absolute relative deviation lying in the range 0.17–81.99% and an average value of 8.88%.

1. Introduction

Supercritical fluid (SCF) technologies have been gaining an increasing attention in process engineering during the past decades due to its various advantages. Carbon dioxide is the most commonly used SCF because it is environmentally benign, nontoxic, nonflammable, cheap, and readily available. As a green solvent, supercritical carbon dioxide ($scCO_2$) finds applications in many conventional and novel emerging technologies such as dry dyeing processing techniques, which operate without wastewater emissions, efficient environmentally friendly and benign new green biotechnological processes, and so forth [1,2]. The accurate knowledge of the phase behavior of the systems involved and, in particular, of the solubility of the solid solutes in high-pressure $scCO_2$ is crucial to the design, optimization, and operation of such processes. The solid solutes of interest, such as dyes or pharmaceuticals, are generally high molecular weight complex molecules and in their majority low volatile solids. This implies that very often their experimental physical properties, such as critical properties and sublimation pressure, are not readily available in the literature. The experimental measurement of the solubility of such compounds in $scCO_2$ is laborious and costly. To avoid expensive and tedious experiments, flexible and robust predictive models are the ideal targets. In the absence of such an ideal predictive model, however,

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simple and accurate enough correlative models are very useful in engineering applications such as used in commercial process simulators. This is the reason why both types of modeling have been and still remain a goal for many research works.

Three major modeling approaches have been used to model the solubility of a solid solute in an SCF for predictive or correlative purposes: equations of state (EoSs) and activity coefficient models, computational intelligence techniques, and density-based semiempirical equations. It should be mentioned that the three approaches rely more or less on experimental data and, therefore, their use outside the range of experimental data from which the models have been obtained is not safe.

EoSs are the straightforward and obvious methods as the solubility is a phase equilibrium problem. Simple cubic EoS with various mixing rules has been extensively used for calculation of solid solute solubility in $scCO_2$ (to cite just few, see, for example, Refs. [3–12]). Detailed review of the use of cubic EoSs in this context has been given by Yazdizadeh et al. [10]. The use of cubic EoSs requires critical properties, acentric factor to compute fugacity coefficient, which is needed along with the sublimation vapor pressure, and the molar volume of the solid solute to compute the solubility. In the absence of experimental values of these solid solute parameters, they are estimated using group contribution (GC) methods. Sublimation vapor pressure, which is very low, and experimental value, unavailable for many solids, may also be considered as an adjusted parameter to be fitted to solubility data as used in Refs. [8,13,14]. The accuracy of the correlation or prediction using the cubic EoS depends greatly on the method used to estimate these solid solute properties [3,4,11]. The effect of mixing rules has been investigated by Yazdizadeh et al. [9,10] in two comparative studies. The first one [9] concerned 52 solid solutes and compared two cubic EoSs combined with four mixing rules and showed a global superiority of Esmaeilzadeh–Roshanfekr EoS [15] combined with Wong–Sandler mixing rule [16] and a global average absolute deviation of 9%. In the second one [10] 23 compounds were considered, five cubic EoSs combined with four mixing rules and the results showed a global best performance of a modified Esmaeilzadeh–Roshanfekr EoS [17] with Wong–Sandler mixing rule [16] and a global average absolute deviation of 7%. Sang et al. [8] considered three cubic EoSs combined with five mixing rules where they used a modification of the calculation procedure by introducing some approximations. They proposed two- and three-adjustable-parameter models for the calculation of the fugacity coefficient. Accordingly, the only solid solute parameter required was the molar volume. They showed that the three-adjustable-parameter model gave a global average absolute deviation of 5.5% for the same data set used in Ref. [10].

Despite their success for fluid mixtures, theoretical –sound state of the art statistical thermodynamic models have not been used for modeling complex molecules such as pharmaceuticals. Statistical associating fluid theory (SAFT)-type models use fluid-specific parameters that usually are fitted to pure fluid experimental data. In common fluids, experimental vapor pressures and liquid densities are typically used. However, for the majority of pharmaceutical complex chemicals such extended experimental data do not exist [18]. Most SAFT-type models require five parameters for each pure associating compound (three for nonassociation ones). Three parameters for nonassociating fluids: the segment number, the interaction energy, and the hard core segment diameter. Two other parameters for associating fluids are the association energy and the affective association volume. These parameters are calculated using GC methods, such as proposed by Tihic et al. [19,20] or estimated from vapor pressure and liquid-density data over extended temperature ranges [21,22]. In the absence of such data for specific compounds, such as polymers and pharmaceuticals, GC methods, such as GC-SAFT [23,24], GC-SAFT variable range (SAFT-VR) [25–32], and GC-SAFT-γ [33–35], are used for the estimation of required properties. As regards modeling the solubility of complex solid molecules in $scCO_2$ using noncubic EoS only few works are reported. This is due to the lack of sufficient experimental data as this family of EoSs uses fluid-specific parameters that usually are fitted to pure fluid experimental data [18]. Tsivintzelis et al. [18] have used nonrandom hydrogen bonding theory to predict the solubility of pharmaceuticals in SCFs. SAFT models have also been used to predict or correlate the solubility of solids in SCFs [36–38].

Activity coefficient models, such as universal functional activity coefficient, the regular solution theory-based models, and more sophisticated models, such as conductor-like screening model (COSMO), cannot be used for high-pressure phase equilibrium calculations without being coupled with another model, namely, an EoS [18]. COSMO-based approach has been used to predict solubility of solids in $scCO_2$ [39–42]. The only experimental data required for solutes are the melting temperature and the melting enthalpy of the solid solute.

Computational intelligence techniques such as artificial neural networks, adaptive neurofuzzy inference system, and least square support vector machine are considered as powerful modeling tools that can map complex highly nonlinear input/output relationships of any systems. This ability has made them suited for a wide range of engineering applications. Applications of such techniques to correlate solubility of various solid solutes in $scCO_2$ have been reported: Refs. [43–51] for artificial neural networks, Refs. [52,53] for least square support vector machine, and Ref. [54] for adaptive neurofuzzy inference system.

The review of the EoS and computational intelligence approaches in modeling the solubility of solid solutes in supercritical solvents reveals that both approaches, whether used for predictive or correlative purposes, with the exception of the COSMO model, rely on experimental data of phase equilibrium and the physical properties of pure solid solute. As regards the EoS approach, phase equilibrium data are required for fitting at least one binary interaction parameter used by mixing rules, fitting other required pure solute properties, or for the validation of models. Regarding computational intelligence methods, experimental data are necessary for training and validation of the model. Pure component properties for complex multifunctional solid solutes, such as pharmaceuticals, are not always available and need to be estimated by GC methods.

Semiempirical modeling is used as a simpler alternative insofar as it does not require any solute property, although there are few models that include some solute parameters such as temperature and enthalpy of fusion and infinite dilute activity coefficient [55], solubility parameter, and molar volume of the solute [56]. In a very recent article [57], we have presented an extensive review of semiempirical models. Comparative studies [58–64] on the performance of semiempirical models show there is no model that presents the best correlation for all the experimental data of all solutes. In the present work, it is intended to carry a larger scale comparative study that includes all semi-empirical models on a more comprehensive data set. It also aims for the improvement of the quality of density-based correlation of the solubility of solid solutes in $scCO_2$.

2. Modeling solid–SCF phase equilibrium

The fugacities of a solid solute in the solid and fluid phases are related by the following fundamental iso-fugacity relationship:

$$f_2^{S} = f_2^{SCF} \quad (1)$$

in which f designates the fugacity and superscripts S and SCF designate solid and SCF phases, respectively.

The fugacity of solid solute in an SCF phase can be written as the product of the solute mole fraction or solubility (y_2), the fugacity coefficient (ϕ_2), and the equilibrium pressure P as follows:

$$f_2^{SCF} = y_2 \phi_2 P \quad (2)$$

The fugacity of the solid phase is usually considered as a pure phase and can expressed as

$$f_2^{s} = P_2^{sub} \phi_2^{sat} \exp\left[\frac{V_2^{s}}{RT}\left(P - P_2^{sub}\right)\right] \quad (3)$$

where P_2^{sub}, ϕ_2^{sat}, and V_2^{s} are the sublimation pressure, the saturated fugacity coefficient, and the molar volume of the solid, respectively. T is the temperature, P is the pressure, and R is the universal gas constant (8.314 J mol^{-1} K^{-1}).

Because of sublimation pressures, ϕ_2^{sat} is equal to 1 for most solutes that do not have a strong tendency of association. Furthermore, because of the incompressibility of solids, values of V_2^{s} can be assumed to be pressure independent.

Hence, y_2 in SCF reduces to

$$y_2 = \frac{P_2^{sub}}{P} \frac{1}{\phi_2^{SCF}} \exp\left[\frac{V_2^{s}}{RT}\left(P - P_2^{sub}\right)\right] \quad (4)$$

Note that ϕ_2^{SCF} is a function of y_2, T, P, and Z^{SCF} (the compressibility coefficient of the supercritical phase), itself a function of y_2, T, and P:

$$\phi_2^{SCF} = \phi\left(T, P, Z^{SCF}, y_2\right) \quad (5)$$

Table 1
Density-based models for the correlation of solid solutes in SCFs.

Model	Equation	Equation no.	Reference
Chrastil	$\ln y_2 = a_0 + a_1 \ln \rho_1 + \frac{a_2}{T}$	(10)	[65]
Adachi and Lu	$\ln y_2 = a_0 + (a_1 + a_2\rho_1 + a_3\rho_1^2) \ln \rho_1 + \frac{a_4}{T}$	(11)	[66]
del Valle and Aguilera	$\ln y_2 = a_0 + a_1 \ln \rho_1 + \frac{a_2}{T} + \frac{a_3}{T^2}$	(12)	[67]
Kumar and Johnston	$\ln y_2 = a_0 + a_1\rho_1 + \frac{a_2}{T}$	(13)	[68]
Bartle et al.	$\ln \frac{y_2 P}{P^{ref}} = a_0 + a_1(\rho_1 - \rho_1^{ref}) + \frac{a_2}{T}$	(14)	[69]
Gordillo et al.	$\ln y_2 = a_0 + a_1 P + a_2 P^2 + a_3 PT + a_4 T + a_5 T^2$	(15)	[70]
Mendez-Santiago and Teja	$T \ln y_2 P = a_0 + a_1\rho_1 + a_2 T$	(16)	[71]
Sung and Shim	$\ln y_2 = \left(a_0 + \frac{a_1}{T}\right) \ln \rho_1 + \frac{a_2}{T} + a_3$	(17)	[72]
Jouyban et al.	$\ln y_2 = a_0 + a_1 P + a_2 P^2 + a_3 PT + a_4\frac{T}{P} + a_5 \ln \rho_1$	(18)	[58]
Sparks et al.	$c_2^* = \rho_{r,1}^{(a_0 + a_1\rho_{r,1} + a_2\rho_{r,1}^2)} \exp\left(b_0 + \frac{b_1}{T_r} + \frac{b_2}{T_r^2}\right)$ with $c_2^* = \frac{c_2}{\rho_{c,1}}$; $\rho_{r,1} = \frac{\rho_1}{\rho_{c,1}}$; $T_r = \frac{T}{T_{c,1}}$	(19)	[73]
Garlapati et Madras	$\ln y_2 = a_0 \ln \rho_1 + \frac{a_1}{T} + a_2$	(20)	[74]
Garlapati et Madras	$\ln y_2 = a_0 + (a_1 + a_2\rho_1) \ln \rho_1 + \frac{a_3}{T} + a_4 \ln(\rho_1 T)$	(21)	[75]
Jafari Nedjad et al.	$\ln y_2 = a_0 + a_1 P^2 + a_2 T^2 + a_3 \ln \rho_1$	(22)	[76]
Ch and Madras	$y_2 = \left(\frac{P}{P^*}\right)^{(k-1)} \exp\left(\frac{a_0}{T} + a_1\rho_1 + a_2\right)$	(23)	[77]
Bian et al.	$c_2 = \rho_1^{(a_0 + a_1\rho_1 + a_2/\ln T)} \exp\left(\frac{a_3 + a_4\rho_1}{T} + a_5\right) c_2 = \frac{\rho_1 Mw_2 y_2}{Mw_1(1 - y_2)}$	(24) (24a)	[59]
Keshmiri et al.	$\ln y_2 = a_0 + \frac{a_1}{T} + a_2 P^2 + \left(a3 + \frac{a_4}{T}\right) \ln \rho_1$	(25)	[78]
Ali Akbar Amooey	$\ln y_2 = \frac{a_0 + a_1/\rho_1 + a_2/\rho_1^2 + a_3 \ln T + a_4(\ln T)^2}{1 + a_5/\rho_1 + a_6 \ln T + a_7(\ln T)^2 + a_8(\ln T)^3}$	(26)	[60]
Hozhabr et al.	$\ln y_2 = a_0 + \frac{a_1}{T} + \frac{a_2\rho_1}{T} - a_3 \ln P$	(27)	[79]
Khansary et al.	$\ln y_2 = \frac{a_0}{T} + a_1 P + \frac{a_2 P^2}{T} + (a_3 + a_4 P) \ln \rho_1$	(28)	[61]
Bian et al.	$\ln y_2 = a_0 + \frac{a_1}{T} + \frac{a_2\rho_1}{T} + (a_3 + a_4\rho_1) \ln \rho_1$	(29)	[64]
Si-Moussa et al.	$\ln y_2 = a_0 + a_1\rho_1 + a_2\rho_1^2 + a_3\rho_1 T + a_4\frac{T}{\rho_1} + a_5 \ln \rho_1$	(30)	[57]

Table 2
Sources and ranges of solubility data of solid solutes in $scCO_2$.

No.	Solid solute	Mw (g/mol)	*T* (K)	*P* (MPa)	$y_2 \times 10^4$	N_i	Reference
1	(*S*)-Boc-piperazine	285.3900	308.20–328.20	90.10–205.00	13.70–65.20	21	[81]
2	1,8-Dihydroxy-9-anthrone	226.2274	308.00–348.00	101.00–355.00	1.33–4.87	54	[82]
3	10-Undecenoic acid	184.2800	308.00–333.00	100.00–180.00	4.00–174.00	18	[83]
4	1-Chloro-2,4-dinitrobenzene	202.5520	308.00–313.00	95.00–145.00	17.80–61.90	12	[84]
5	1-Hydroxy-2,4-dimethyl-9-anthrone	238.2812	308.00–348.00	101.00–355.00	0.24–1.06	54	[82]
6	1-Hydroxy-2-ethyl-9-anthrone	238.2812	308.00–348.00	101.00–355.00	1.08–9.78	54	[82]
7	1-Hydroxy-2-methyl-9-anthrone	224.2546	308.00–348.00	101.00–355.00	0.87–7.05	54	[82]
8	1-Hydroxythioxanthone	228.2660	308.00–348.00	121.00–354.00	0.99–4.24	45	[85]
9	2,2′-Oxybis(*N*,*N*-dibutylacetamide)	356.5432	313.00–333.00	88.00–131.00	15.20–86.00	15	[86]
10	2,2′-Oxybis(*N*,*N*-diethylacetamide)	240.2988	313.00–333.00	87.00–133.00	22.30–1171.00	15	[86]
11	2,2′-Oxybis(*N*,*N*-dihexylacetamide)	468.7558	313.00–333.00	90.00–164.00	10.60–53.60	15	[86]
12	2,3-Dimethylaniline	121.1800	313.00–333.00	80.00–200.00	3.30–95.20	18	[87]
13	2,4-Dinitrophenol	184.1100	308.00–348.00	121.60–483.70	12.40–140.00	41	[88]
14	2,5-Dinitrophenol	184.1100	308.00–348.00	121.60–486.40	19.10–94.40	45	[88]
15	2-Methoxybenzoic acid	152.1500	308.20–328.20	99.80–246.90	90.00–1110.00	23	[89]
16	2-Naphthol	144.1700	308.10–328.10	100.50–300.00	2.49–12.30	18	[90]
17	2-Phenyl-4*H*-1,3-benzoxazin-one	223.2270	308.15–328.18	651.46–951.27	0.80–4.50	26	[91]
18	2-Propenamide	71.0800	308.20–323.20	90.00–400.00	0.12–1.63	28	[92]
19	2-Trifluoromethylbenzoic acid	190.1190	308.20–323.20	93.40–226.00	13.70–85.50	21	[93]
20	3-Acetylpyridine	121.1400	313.15–343.15	100.00–260.00	69.00–1619.00	36	[94]
21	3-Aminobenzoic acid	137.1360	308.00–328.00	100.00–210.00	0.02–0.07	15	[95]
22	3-Nitrotoluene	137.1360	313.00–333.00	80.00–200.00	1.50–59.20	18	[87]
23	3-Trifluoromethylbenzoic acid	190.1190	308.20–323.20	94.10–225.40	55.10–469.00	21	[93]
24	4-Aminoantipyrine	203.2400	308.20–328.20	100.50–220.40	1.16–3.67	21	[96]
25	4-Aminosalicylic acid	153.1350	308.00–328.00	110.00–210.00	0.04–0.36	15	[97]
26	4-Dimethylaminoantipyrine	231.2940	308.20–328.20	100.30–220.30	5.25–25.50	21	[96]
27	4-Hydroxybenzaldehyde	122.1200	308.00–328.00	110.00–210.00	0.07–1.38	15	[98]
28	4-Trifluoromethylbenzoic acid	190.1190	308.20–323.20	96.80–224.40	1.06–4.56	21	[93]
29	5-Fluorouracil	130.0770	308.15–328.15	125.00–250.00	0.04–0.15	18	[91]
30	5-Hydroxymethylfurfural	126.1100	314.10–343.20	97.40–195.80	5.06–23.84	22	[99]
31	Acenaphthene	154.2080	308.00–338.00	121.60–354.60	23.95–47.37	36	[100]
32	Acetylferrocene	228.0680	308.00–348.00	77.00–221.00	2.50–79.20	18	[101]
33	All-*trans* retinoic acid	300.4351	308.20–318.20	102.00–303.00	0.01–0.18	15	[102]
34	Amical-48 (diiodomethyl *p*-tolyl sulfone)	422.0200	318.00–338.00	100.00–300.00	0.06–1.15	18	[103]
35	Ammonium benzoate	139.1520	308.00–328.00	110.00–210.00	0.02–0.05	15	[104]
36	Amoxicillin	365.4040	308.15–338.15	160.00–400.00	0.10–72.30	28	[105]
37	Anastrozole	293.3700	308.00–348.00	122.00–355.00	0.04–3.80	45	[106]
38	Anthracene	178.2290	308.10–328.10	100.00–300.00	0.42–1.13	15	[90]
39	Antipyrine	188.2260	308.20–328.20	100.40–220.40	2.09–7.17	21	[96]
40	Aspirin	180.1570	308.15–328.15	120.00–250.00	0.63–3.47	24	[107]
41	Atorvastatin	558.6400	308.00–348.00	121.60–354.60	0.17–14.46	45	[108]
42	Atropine	289.3690	308.00–348.00	122.00–355.00	0.60–16.70	45	[109]
43	Azadirachtin	720.7140	308.15–333.15	100.00–260.00	0.01–0.15	54	[110]
44	Azelaic acid	188.2210	313.15–333.15	100.00–300.00	0.01–0.08	14	[111]
45	Azobenzene	182.2212	308.20–318.20	91.00–253.00	36.40–98.40	10	[112]
46	Azodicarbonamide	116.0788	308.15–328.15	100.00–300.00	0.09–0.20	26	[91]
47	Beclomethasone dipropionate	521.0420	338.00–358.00	213.00–385.00	0.07–0.34	21	[113]
48	Benzamide	121.1366	308.00–328.00	110.00–210.00	0.02–0.16	15	[104]
49	Benzenesulfonamide	172.2050	308.15–328.15	110.00–210.00	0.65–2.90	15	[114]
50	Benzocaine	165.1891	308.00–348.00	122.00–355.00	10.30–121.20	40	[115]
51	Benzoin	212.2400	308.15–328.15	111.30–244.30	0.41–4.10	19	[116]
52	Bisacodyl	361.4000	308.00–348.00	122.00–355.00	0.36–5.83	39	[117]
53	Budesonide	430.5340	338.00–358.00	213.00–385.00	0.06–0.29	21	[113]
54	Caffeine	194.1900	313.00–353.00	199.00–349.00	2.83–11.30	24	[118]
55	Cannabinol	310.4299	314.00–334.00	130.00–202.00	1.26–2.17	34	[119]
56	Capecitabine	359.3501	308.00–348.00	15.20–35.40	0.08–1.59	40	[120]
57	Carbamazepine	236.2686	308.00–348.00	122.00–355.00	0.11–0.94	39	[109]
58	Carvedilol	406.4000	308.00–338.00	160.00–400.00	0.11–50.10	28	[121]
59	Cefixime trihydrate	507.5000	308.00–328.00	183.00–335.00	0.002–0.003	18	[122]
60	Celecoxib	381.3700	323.20–343.20	150.00–300.00	0.02–0.15	18	[123]
61	Cetirizine	388.8880	308.15–338.15	160.00–400.00	0.11–49.20	28	[124]
62	Chlormezanone	273.7000	308.20–328.20	119.30–239.90	0.05–5.45	21	[125]
63	Chlorothalonil	265.9100	318.00–338.00	100.00–300.00	0.13–2.68	23	[103]
64	Chlorpheniramine maleate	390.8600	308.00–338.00	160.00–400.00	0.15–4.26	24	[126]
65	Cholesterol	386.6500	313.15–333.15	100.00–250.00	0.02–1.45	24	[127]
66	Cholesteryl acetate	428.6900	308.15–328.15	90.00–240.00	0.044–10.40	24	[127]
67	Cholesteryl benzoate	490.7600	308.15–328.15	120.00–270.00	0.05–0.53	20	[127]
68	Cholesteryl butyrate	456.7400	308.15–328.15	100.00–240.00	0.22–8.93	20	[127]

(*continued on next page*)

Table 2 (*continued*)

No.	Solid solute	Mw (g/mol)	T (K)	P (MPa)	$y_2 \times 10^4$	N_i	Reference
69	Cinnarizine	368.5000	308.15–328.15	139.70–240.00	0.03–2.05	21	[128]
70	Climbazole	292.7600	313.20–333.20	105.50–398.90	6.20–48.80	24	[129]
71	Clobetasol propionate	466.9700	308.00–348.00	122.00–355.00	0.004–0.035	44	[130]
72	Clofenamic acid	247.6800	313.00–333.00	120.00–360.00	0.04–0.35	24	[131]
73	Clofibric acid	214.6000	308.20–328.20	100.10–220.20	2.57–8.56	21	[132]
74	Clotrimazole	344.8370	308.00–348.00	122.00–355.00	0.04–1.06	45	[133]
75	Clozapine	326.8230	318.00–348.00	121.60–354.60	0.04–0.42	27	[134]
76	Codeine	299.3642	308.00–348.00	122.00–355.00	1.00–12.30	45	[109]
77	Corosolic acid	472.7000	308.15–333.15	80.00–300.00	0.01–743.00	40	[135]
78	Cortisone acetate	402.4800	308.15–373.15	82.40–226.50	0.005–0.23	30	[136]
79	Cyproheptadine	278.4000	308.00–338.00	160.00–400.00	0.81–30.90	28	[137]
80	Cyproterone acetate	416.9380	308–348	122.00–355.00	0.13–2.61	40	[138]
81	Desoxycorticosterone acetate	372.4970	308–348	122.00–355.00	0.01–1.39	45	[130]
82	Dexamethasone	392.4610	308.15–328.15	151.10–348.30	0.013–0.028	30	[139]
83	Diazepam	284.7400	308.00–348.00	122.00–355.00	1.80–11.10	45	[109]
84	Dichlone	227.0400	313.00–333.00	70.70–325.80	0.07–2.51	23	[140]
85	Diclofenac acid	331.3400	308.15–338.15	120.00–400.00	0.40–19.80	32	[141]
86	Diflunisal	250.1980	308.20–328.20	93.00–246.00	0.02–0.08	21	[142]
87	Diheptyl-2,2′-oxidiacetate	330.4596	313.00–333.00	87.00–141.00	21.99–81.66	15	[143]
88	Dinonyl-2,2′-oxidiacetate	386.5659	313.00–333.00	91.00–142.00	14.86–67.10	15	[143]
89	Dipentadecyl-2,2′-oxidiacetate	554.8848	313.00–333.00	110.00–193.00	5.39–14.03	15	[143]
90	Dipentyl-2,2′-oxidiacetate	274.3532	313.00–333.00	85.00–125.00	22.31–125.05	15	[143]
91	Ditridecyl-2,2′-oxydiacetate	498.7790	313.00–333.00	99.00–156.00	5.57–18.22	15	[143]
92	Diundecyl-2,2′-oxidiacetate	442.6722	313.00–333.00	92.00–145.00	7.44–24.34	15	[143]
93	Diuron	233.0900	308.20–328.20	100.00–200.00	0.009–3.18	19	[144]
94	Docetaxel	807.8792	308.00–338.00	152.00–354.00	0.37–5.03	32	[120]
95	Dutasteride	528.5000	308.00–348.00	122.00–355.00	0.001–1.60	45	[145]
96	Epicatechin	290.2680	313.15–343.15	120.00–260.00	0.007–0.18	60	[146]
97	Ergosterol	396.6500	318.15–333.15	120.00–240.00	0.03–0.30	15	[147]
98	Ethanamide	59.0700	308.20–323.20	90.00–400.00	2.38–31.62	30	[92]
99	Ethylene glycol	62.0680	313.15–353.15	70.00–190.00	83.00–429.00	35	[148]
100	Exemestane	296.4000	308.00–348.00	122.00–355.00	0.13–18.76	45	[106]
101	Fenofibrate	360.8310	308.20–328.20	100.10–220.20	18.30–66.60	21	[132]
102	Ferrocene	186.0310	308.00–348.00	80.00–244.00	8.90–31.20	22	[101]
103	Ferulic acid	194.1840	301.50–333.50	120.00–280.00	0.02–0.12	18	[149]
104	Finasteride	372.5000	308.00–348.00	122.00–355.00	0.83–3.33	45	[145]
105	Fluoranthene	202.2506	308.15–328.15	86.00–249.00	1.13–9.30	28	[150]
106	Fluoxetine hydrochloride	345.7900	308.15–338.15	160.00–400.00	0.35–8.12	28	[151]
107	Flurbiprofen	244.2609	303.15–323.15	89.00–245.00	0.22–1.97	27	[152]
108	Flutamide	276.1000	308.00–348.00	122.00–355.00	0.05–5.09	45	[145]
109	Fluvastatin	411.4700	308.00–348.00	121.60–354.60	0.22–6.01	45	[108]
110	Gabapentin	171.2400	308.00–338.00	160.00–400.00	2.31–73.60	28	[153]
111	Gemfibrozil	250.3400	308.20–328.20	100.10–220.20	0.29–41.90	21	[132]
112	Geraniol	154.2500	308.00–333.00	100.00–180.00	27.00–252.00	18	[83]
113	Geranyl butyrate	224.3400	308.15–333.15	100.00–180.00	22.00–250.00	18	[83]
114	Hinokitiol	164.2000	313.20–333.20	101.40–358.40	5.91–24.90	32	[154]
115	Hydrocortisone	362.4600	308.15–373.15	99.00–226.50	0.0005–0.0073	26	[136]
116	Ibuprofen	206.2808	308.15–318.15	80.00–220.00	0.53–58.40	29	[155]
117	Imipramine HCl	316.8700	313.50–323.50	300.00–500.00	0.05–0.10	10	[156]
118	Iodopropynyl butylcarbamate	281.0900	308.15–333.15	87.60–341.50	12.00–47.00	27	[157]
119	Isoniazid	137.1393	308.00–313.00	130.00–185.00	0.005–0.06	18	[158]
120	Juglone	174.1528	308.20–328.20	92.00–244.00	0.20–15.90	18	[159]
121	Ketoconazole	531.4310	308.00–348.00	122.00–355.00	0.005–1.75	45	[133]
122	Ketoprofen	254.2806	313.15–328.15	90.00–250.00	0.03–1.88	15	[160]
123	Lamotrigine	256.0910	318.00–338.00	121.60–354.60	0.004–0.046	27	[134]
124	Letrozole	285.3100	308.00–348.00	122.00–355.00	0.01–0.83	45	[106]
125	Levonorgestrel	384.5000	308.00–338.00	12.20–35.50	0.004–0.03	36	[161]
126	Levulinic acid	116.1152	313.00–342.40	94.10–182.70	4.91–90.00	34	[162]
127	Lovastatin	404.5500	308.00–348.00	121.60–354.60	0.11–1.14	45	[108]
128	Mandelic acid	152.1500	308.15–328.15	101.00–230.60	0.27–29.04	21	[116]
129	*m*-Dinitrobenzene	168.1100	308.00–328.00	95.00–145.00	1.90–55.40	18	[84]
130	Medroxyprogesterone acetate	386.2460	308.00–348.00	122.00–355.00	0.40–4.13	40	[138]
131	Mefenamic acid	241.2900	308.15–338.15	160.00–400.00	0.83–59.40	28	[163]
132	Megestrol acetate	312.5000	308.00–338.00	12.20–35.50	0.03–8.70	36	[161]
133	Meloxicam sodium salt	373.3800	303.00–323.00	149.00–255.00	0.04–0.13	15	[164]
134	Menadione	172.1800	313.00–333.00	97.20–306.70	7.50–10.06	18	[140]
135	Metaxalone	221.3000	308.20–328.20	119.50–240.00	0.04–1.48	21	[125]
136	Methimazole	114.1700	308.00–348.00	122.00–355.00	0.54–18.99	40	[165]
137	Methocarbamol	241.2000	308.20–328.20	119.90–220.50	0.61–4.87	21	[125]
138	Methyl gallate	184.1500	313.00–333.00	100.00–500.00	0.001–0.042	27	[165]
139	Methyl salicylate	152.1473	343.15–423.15	90.00–310.00	5.10–753.30	44	[167]

Table 2 (*continued*)

No.	Solid solute	Mw (g/mol)	*T* (K)	*P* (MPa)	$y_2 \times 10^4$	N_i	Reference
140	Methylparaben	152.1473	308.00–348.00	122.00–355.00	1.08–12.13	40	[117]
141	Metronidazole benzoate	275.7600	308.00–348.00	122.00–355.00	0.70–45.50	40	[115]
142	*m*-Nitrophenol	139.1100	308.00–348.00	121.60–486.40	3.94–44.50	49	[88]
143	*N*-(4-Ethoxyphenyl)ethanamide	179.2160	308.00–328.00	90.00–190.00	0.09–0.43	16	[77]
144	Nabumetone	228.3000	308.20–328.20	100.00–220.00	0.39–26.80	21	[168]
145	Naproxene	230.2592	313.10–333.10	89.60–193.10	0.02–0.32	18	[169]
146	Nicotinamide	122.1246	313.15–373.15	55.00–305.00	0.10–31.40	24	[170]
147	Nicotinic acid	123.1100	313.15–373.15	45.00–302.00	0.003–0.104	22	[168]
148	Nifedipine	346.3350	333.15–373.15	126.00–296.00	0.05–0.71	29	[171]
149	Niflumic acid	282.2200	313.20–353.20	190.00–310.00	0.07–0.21	21	[123]
150	Nimodipine	418.4403	313.00–333.00	100.00–250.00	0.006–0.423	21	[87]
151	Nitrendipine	360.3600	308.00–328.00	80.00–200.00	0.01–0.63	15	[172]
152	Norfloxacin	319.3308	313.15–323.15	99.40–303.30	0.01–0.24	15	[173]
153	Ofloxacin	361.3675	318.15–323.15	101.50–300.60	0.004–0.013	10	[173]
154	*O*-Nitrobenzoic acid	167.1200	308.00–328.00	100.00–210.00	0.06–0.36	15	[174]
155	*O*-Tolidine	212.2900	308.00–328.00	110.00–210.00	0.05–0.48	15	[175]
156	Oxymatrine	264.3700	308.15–328.15	110.40–210.70	0.007–0.025	18	[176]
157	Oxymetholone	332.4770	308.00–328.00	121.00–305.00	0.16–1.49	20	[122]
158	Paclitaxel	853.9061	308.15–328.15	100.00–275.00	0.012–0.062	21	[91]
159	*p*-Dimethylaminoazobenzene	225.2890	308.20–318.20	91.00–253.00	4.37–12.50	10	[112]
160	Penicillin G	334.3901	313.15–333.15	100.00–350.00	0.04–0.63	18	[70]
161	Penicillin V	350.3800	314.85–334.85	79.90–280.50	3.24–4.26	24	[177]
162	Pentoxifylline	278.3000	308.15–328.15	119.80–2400	0.30–13.70	21	[128]
163	Phenazopyridine	213.2385	308.00–348.00	122.00–355.00	0.44–20.21	45	[165]
164	Phenol	94.1112	333.15–363.15	100.00–350.00	11.40–906.40	33	[178]
165	Phenylbutazone	317.4458	308.20–328.20	100.00–220.00	0.20–26.50	21	[168]
166	Phenylephrine hydrochloride	203.6660	308.15–338.15	160.00–400.00	1.01–28.90	28	[179]
167	*p*-Hydroxyazobenzene	198.2206	308.20–318.20	91.00–253.00	25.70–63.60	10	[112]
168	Picric acid	229.1100	308.00–348.00	121.60–486.40	0.22–26.50	50	[88]
169	Pindolol	248.3210	298.00–318.00	80.00–275.00	0.66–1.92	30	[180]
170	Piracetam	142.2000	308.15–328.15	120.00–240.00	0.08–0.37	21	[128]
171	Piroxicam	331.3460	308.15–338.15	160.00–400.00	0.12–5.12	28	[181]
172	*p*-Methylbenzenesulfonic acid	172.2020	318.00–328.00	80.00–210.00	0.20–0.74	15	[182]
173	*p*-Nitroaniline	183.1300	308.00–328.00	110.00–210.00	0.06–0.43	15	[183]
174	*p*-Nitrophenol	139.1100	308.00–348.00	121.60–486.40	2.23–19.70	49	[88]
175	Prednisolone	360.4400	308.15–373.15	82.40–226.50	0.00004–0.00099	28	[136]
176	Prednisone	358.4300	308.15–373.15	82.40–226.50	0.0001–0.0058	28	[136]
177	Propranolol	259.3434	308.00–348.00	122.00–355.00	3.58–239.61	45	[165]
178	Propyl 4-hydroxybenzoate	180.2010	308.15–328.15	94.10–220.20	0.19–6.12	21	[116]
179	Propyl *p*-hydroxybenzoate	180.2000	308.15–328.15	80.00–230.00	0.038–6.175	18	[184]
180	Protocatechualdehyde	138.1200	313.00–333.00	100.00–500.00	0.004–0.046	24	[166]
181	Protocatechuic acid	154.1200	313.00–333.00	100.00–500.00	0.0004–0.0259	24	[166]
182	*p*-Toluenesulfonamide	171.2200	308.15–328.15	80.00–210.00	0.05–0.51	15	[185]
183	Pyrocatechol	110.1106	333.15–348.15	100.00–350.00	1.22–38.47	22	[178]
184	*rac*-Boc-piperazine	285.3900	308.20–328.20	102.00–204.00	1.24–5.90	19	[81]
185	Racemic paroxetine	329.3650	308.20–328.20	88.30–242.00	0.73–11.84	24	[186]
186	Resorcinol	110.1100	308.15–338.15	120.00–400.00	1.10–9.73	32	[187]
187	Rosuvastatin	481.5380	308.00–348.00	121.60–354.60	0.23–2.44	45	[108]
188	Salicylamide	137.1360	308.20–328.20	100.00–220.00	0.09–2.10	21	[167]
189	Salicylic acid	138.1200	308.15–318.15	92.60–157.90	0.97–3.98	20	[188]
190	Simvastatine	418.5662	308.00–348.00	121.60–354.60	0.02–5.35	45	[108]
191	Spironolactone	416.5730	308.00–338.00	160.00–400.00	0.63–50.10	28	[189]
192	Sulindac	356.4100	308.15–338.15	160.00–400.00	0.37–86.90	28	[190]
193	Syringic acid	198.1700	313.00–333.00	100.00–500.00	0.003–0.127	27	[191]
194	TCMTB [2-(thiocyanomethylthio)benzothiazole]	238.3600	323.00–338.00	100.00–300.00	0.02–13.90	12	[103]
195	Tebuconazole	307.8200	323.00–338.00	100.00–300.00	0.04–18.57	12	[103]
196	Testosterone	288.4200	308.15–373.15	82.40–226.50	0.07–0.90	30	[136]
197	Tetramethylpyrazine	136.2000	318.00–338.00	100.00–300.00	100.00–1310.00	15	[192]
198	Theobromine	180.1700	313.00–353.00	193.00–344.00	0.09–0.47	23	[118]
199	Theophylline	180.1700	313.00–353.00	199–349.00	0.10–0.33	24	[118]
200	Thymidine	242.2286	308.15–328.15	100–300.00	0.01–0.08	25	[91]
201	Thymol	150.2176	308.15–323.15	78.00–250.00	8.37–147.00	17	[193]
202	Tolfenamic acid	461.7100	313–333	120.00–360.00	0.01–0.18	24	[131]
203	Triclocarban	315.5800	313.20–333.20	109.30–389.60	0.90–8.70	24	[129]
204	Triphenylene	228.2879	308.15–328.15	85.00–252.00	0.01–0.42	28	[150]
205	Triphenylmethyl chloride	278.7754	308.15–338.15	150.00–400.00	2.93–27	18	[194]
206	Triphenyltin chloride	385.4500	308.15–328.15	150.00–300.00	2.71–10.96	12	[192]
207	Uracil	112.0868	308.15–338.15	150.00–400.00	0.000769–0.0034	23	[195]
208	Vanillic acid	168.1400	313–333	85.00–500.00	0.0143–0.607	28	[191]
209	Xanthohumol	354.3960	328.15–358.15	350.00–950.00	0.012–0.17	15	[196]
210	Zopiclone	388.8080	313–333	100.00–250.00	0.015–0.219	21	[87]

Table 3
Estimated model parameters (a_i) of the model proposed in this work (Eq. 32) for the 210 solid solutes considered.

No.	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7
1	−105.07	0.083	-4.48×10^{-5}	6.59×10^{-5}	1.32	-2.08×10^{-3}	−24.58	−1.85
2	−1531.82	−0.021	-3.45×10^{-7}	9.44×10^{-5}	4.50	-4.41×10^{-3}	−2.92	175,549.35
3	−4084.36	−0.060	1.46×10^{-5}	1.38×10^{-4}	12.65	-1.32×10^{-2}	2.92	437,823.79
4	−0.97	0.073	-6.70×10^{-5}	4.56×10^{-4}	2.89	-5.14×10^{-3}	−80.05	−0.44
5	8205.31	−0.016	-5.75×10^{-6}	1.10×10^{-4}	−25.39	2.61×10^{-2}	−4.19	−879,263.63
6	−1982.72	−0.013	1.33×10^{-7}	5.77×10^{-5}	5.95	-5.97×10^{-3}	−0.40	217,476.69
7	573.76	0.010	-7.18×10^{-6}	4.24×10^{-5}	−1.91	2.14×10^{-3}	−5.20	−54,779.81
8	−546.60	−0.012	2.81×10^{-6}	3.79×10^{-5}	1.59	-1.58×10^{-3}	0.82	58,878.77
9	374.66	0.219	-1.10×10^{-4}	-2.37×10^{-5}	−1.19	1.91×10^{-3}	−43.89	1.06
10	−525.74	−0.357	2.08×10^{-4}	-1.38×10^{-4}	0.15	-9.85×10^{-5}	101.71	−5.43
11	−382.66	−0.163	9.82×10^{-5}	-1.12×10^{-4}	0.64	-8.50×10^{-4}	53.66	−3.04
12	−1.66	−0.006	1.44×10^{-6}	9.60×10^{-6}	−0.06	2.26×10^{-5}	2.16	2.23
13	11,328.33	0.018	-2.33×10^{-6}	2.17×10^{-7}	−34.56	3.52×10^{-2}	−5.74	−1,234,124.05
14	−392.25	0.001	-6.50×10^{-6}	7.90×10^{-5}	1.32	-1.41×10^{-3}	−8.26	46,727.41
15	−85.82	−0.061	1.76×10^{-5}	5.13×10^{-5}	−0.07	1.37×10^{-4}	17.55	−7.53
16	−83.09	0.050	-1.15×10^{-5}	-6.50×10^{-5}	0.53	-6.75×10^{-4}	−5.92	−10.79
17	−885.78	−0.689	2.25×10^{-4}	7.63×10^{-5}	−2.30	3.56×10^{-3}	244.70	−4.01
18	−262.35	0.001	-2.34×10^{-6}	2.89×10^{-5}	1.63	-2.56×10^{-3}	−1.91	−1.69
19	1.84	−0.044	1.76×10^{-5}	-1.18×10^{-5}	−0.78	1.35×10^{-3}	19.53	2.52
20	−251.12	−0.064	1.38×10^{-5}	1.28×10^{-4}	0.53	-4.15×10^{-4}	5.66	30,058.65
21	−95.39	0.002	9.40×10^{-7}	-3.53×10^{-6}	0.43	-5.89×10^{-4}	0.48	0.15
22	−11.28	−0.007	1.23×10^{-6}	1.35×10^{-5}	−0.02	-2.87×10^{-5}	2.62	2.48
23	133.12	0.021	3.05×10^{-6}	-1.31×10^{-4}	−1.66	2.97×10^{-3}	15.94	−15.54
24	−74.34	0.058	-1.49×10^{-5}	-8.87×10^{-5}	0.31	-3.33×10^{-4}	−2.11	0.85
25	−25.71	−0.019	5.32×10^{-6}	2.56×10^{-5}	−0.18	3.73×10^{-4}	6.14	1.78
26	−360.01	0.190	-1.32×10^{-5}	-5.80×10^{-4}	1.24	-1.19×10^{-3}	12.32	0.63
27	−141.29	−0.023	1.67×10^{-5}	-6.03×10^{-5}	0.09	8.10×10^{-5}	17.82	−0.78
28	598.29	−0.080	2.73×10^{-5}	4.44×10^{-5}	−4.58	7.28×10^{-3}	22.11	6.32
29	−2460.28	−1.203	3.79×10^{-4}	-6.65×10^{-5}	−0.84	1.47×10^{-3}	494.35	−24.73
30	−692.41	−0.051	2.41×10^{-5}	1.05×10^{-5}	1.96	-2.04×10^{-3}	14.30	63,063.17
31	−9800.68	−0.0002	3.53×10^{-6}	-3.46×10^{-5}	30.61	-3.19×10^{-2}	6.55	1,033,864.84
32	−13,205.45	0.023	-1.31×10^{-5}	5.01×10^{-5}	40.60	-4.14×10^{-2}	−9.42	1,437,033.73
33	−236.85	−0.004	9.77×10^{-6}	-5.92×10^{-5}	0.87	-1.17×10^{-3}	11.85	0.60
34	−156.11	−0.017	1.04×10^{-5}	-1.48×10^{-5}	0.58	-7.95×10^{-4}	7.76	0.72
35	−190.06	0.113	-1.17×10^{-5}	-2.68×10^{-4}	0.91	-1.04×10^{-3}	−3.41	0.83
36	1954.65	0.088	-2.45×10^{-5}	-5.31×10^{-5}	−5.82	6.02×10^{-3}	−9.43	−223,043.77
37	−2792.52	0.025	-9.65×10^{-6}	1.15×10^{-5}	8.51	-8.53×10^{-3}	−4.84	303,725.89
38	43.35	0.011	-4.59×10^{-6}	7.94×10^{-6}	−0.31	5.27×10^{-4}	−2.20	2.05
39	−115.46	0.057	-9.23×10^{-6}	-1.33×10^{-4}	0.39	-4.02×10^{-4}	2.56	21.70
40	145.93	0.185	-5.94×10^{-5}	-5.96×10^{-5}	0.55	-6.99×10^{-4}	−52.95	−1.75
41	7678.04	−0.075	1.73×10^{-5}	1.27×10^{-4}	−23.47	2.37×10^{-2}	9.28	−847,146.30
42	852.15	0.030	-1.00×10^{-5}	1.22×10^{-5}	−2.55	2.63×10^{-3}	−4.10	−98,485.91
43	−1645.58	−0.004	4.24×10^{-6}	-1.65×10^{-5}	5.13	-5.37×10^{-3}	4.70	165,008.13
44	−1.95	−0.138	3.39×10^{-5}	2.11×10^{-4}	−0.50	6.00×10^{-4}	18.21	3.11
45	0.93	0.012	1.17×10^{-6}	-4.92×10^{-5}	−0.23	5.31×10^{-4}	2.41	−0.27
46	−716.84	−0.427	1.50×10^{-4}	-8.76×10^{-5}	−1.17	1.95×10^{-3}	171.93	−5.92
47	11,214.79	5.746	-1.90×10^{-3}	7.50×10^{-5}	−0.32	4.38×10^{-4}	−2179.38	93.76
48	−227.10	0.121	-1.59×10^{-5}	-2.37×10^{-4}	1.37	-1.72×10^{-3}	−10.71	−1.82
49	107.14	−0.006	-1.81×10^{-6}	7.24×10^{-5}	−0.62	9.96×10^{-4}	−4.78	4.22
50	−2958.54	−0.061	7.14×10^{-6}	1.47×10^{-4}	8.72	-8.72×10^{-3}	4.93	328,517.55
51	−26.69	0.098	-2.40×10^{-5}	-1.34×10^{-4}	0.25	-1.56×10^{-4}	−10.95	6.24
52	−164.84	−0.068	1.10×10^{-5}	1.71×10^{-4}	0.22	-8.52×10^{-5}	3.13	24,939.84
53	1060.66	0.514	-1.82×10^{-4}	8.90×10^{-5}	−0.35	4.70×10^{-4}	−198.27	8.60
54	34.68	0.004	-3.01×10^{-6}	4.46×10^{-5}	−0.11	1.64×10^{-4}	−5.71	5.29
55	−1339.30	−0.054	5.01×10^{-5}	-2.49×10^{-4}	6.55	-9.78×10^{-3}	46.62	−18.29
56	1164.46	0.181	-5.51×10^{-5}	-4.82×10^{-5}	−2.69	2.71×10^{-3}	−51.34	−110,426.70
57	3303.72	0.214	-7.27×10^{-5}	2.94×10^{-5}	−9.04	9.37×10^{-3}	−79.39	−314,535.29
58	−19,001.50	−0.308	1.25×10^{-4}	-1.47×10^{-4}	57.79	-6.04×10^{-2}	130.40	1,924,912.82
59	−53.00	0.038	-9.72×10^{-6}	-2.77×10^{-5}	0.52	-7.32×10^{-4}	−10.48	−0.54
60	−51.01	0.025	-5.36×10^{-6}	-3.85×10^{-5}	0.04	7.42×10^{-5}	2.27	−1.51
61	−1138.32	0.108	-3.25×10^{-5}	1.40×10^{-5}	3.90	-3.89×10^{-3}	−30.78	132,743.12
62	50.19	0.107	-3.55×10^{-5}	-2.14×10^{-5}	0.19	-1.32×10^{-4}	−24.43	−16.81
63	12.49	0.012	-1.32×10^{-5}	7.90×10^{-5}	0.11	-1.88×10^{-4}	−8.57	−1.40
64	1562.77	−0.141	4.12×10^{-5}	1.04×10^{-4}	−5.29	5.21×10^{-3}	41.52	−202,400.70
65	9228.66	−0.009	-2.13×10^{-6}	5.99×10^{-5}	−28.64	2.96×10^{-2}	0.64	−995,613.08
66	119.60	−0.070	9.87×10^{-6}	1.75×10^{-4}	−1.03	1.53×10^{-3}	7.48	3.78
67	−237.86	0.042	-6.03×10^{-6}	-1.26×10^{-4}	0.69	-8.09×10^{-4}	13.81	−1.24
68	−40.73	0.124	-3.66×10^{-5}	-7.46×10^{-5}	0.88	-1.20×10^{-3}	−27.01	−2.77
69	671.21	0.654	-1.88×10^{-4}	-3.30×10^{-4}	0.89	-7.66×10^{-4}	−180.49	2.74

Table 3 (*continued*)

No.	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7
70	101.91	0.110	-3.27×10^{-5}	-6.30×10^{-5}	−0.05	1.91×10^{-4}	−24.39	1.91
71	−2234.78	−0.045	1.50×10^{-6}	1.93×10^{-4}	6.75	-6.79×10^{-3}	−9.01	257,254.20
72	−81.15	0.044	-1.07×10^{-5}	-4.84×10^{-5}	0.41	-4.81×10^{-4}	−4.05	−2.43
73	−584.03	0.199	-2.44×10^{-5}	-5.37×10^{-4}	2.84	-3.80×10^{-3}	7.58	−3.85
74	246.33	0.033	-1.02×10^{-5}	-7.38×10^{-6}	−0.72	7.64×10^{-4}	−3.25	−32,096.23
75	50.19	−0.185	3.64×10^{-5}	3.55×10^{-4}	−0.56	5.21×10^{-4}	14.70	2.15
76	−2563.28	−0.078	1.94×10^{-5}	1.34×10^{-4}	7.57	-7.63×10^{-3}	8.38	279,924.96
77	51,417.64	−0.049	-2.31×10^{-5}	2.93×10^{-4}	−160.07	1.66×10^{-1}	3.75	−5,507,449.07
78	−11.31	−0.032	1.59×10^{-5}	2.13×10^{-5}	−0.21	3.21×10^{-4}	6.31	1.26
79	−717.56	0.144	-4.74×10^{-5}	7.99×10^{-5}	3.17	-3.27×10^{-3}	−61.25	106,770.33
80	452.85	−0.085	2.19×10^{-5}	1.64×10^{-4}	−1.55	1.55×10^{-3}	4.99	−46,538.55
81	−350.16	−0.026	3.00×10^{-6}	8.53×10^{-5}	0.83	-6.91×10^{-4}	1.57	42,383.68
82	2576.44	−0.069	1.52×10^{-5}	1.10×10^{-4}	−8.49	9.05×10^{-3}	9.10	−269,462.56
83	145.56	−0.040	7.81×10^{-6}	1.04×10^{-4}	−0.52	5.54×10^{-4}	−0.73	−12,431.30
84	232.96	−0.072	1.18×10^{-5}	1.50×10^{-4}	−1.60	2.34×10^{-3}	6.50	8.38
85	3580.56	−0.036	7.25×10^{-6}	7.43×10^{-5}	−11.12	1.14×10^{-2}	6.73	−395,352.20
86	240.50	−0.055	-2.89×10^{-7}	2.07×10^{-4}	−1.39	2.02×10^{-3}	−3.65	5.82
87	−243.80	−0.179	8.83×10^{-5}	1.77×10^{-5}	0.11	-1.94×10^{-4}	46.11	−1.30
88	−659.43	−0.171	8.83×10^{-5}	-2.60×10^{-4}	0.89	-1.12×10^{-3}	94.34	−8.06
89	−2624.87	−1.201	4.12×10^{-4}	-2.41×10^{-4}	0.10	1.46×10^{-4}	498.22	−22.53
90	−312.90	−0.067	3.99×10^{-5}	-2.59×10^{-5}	1.22	-1.83×10^{-3}	21.33	−3.88
91	1293.79	0.663	-2.86×10^{-4}	2.29×10^{-4}	−1.11	1.57×10^{-3}	−226.17	30.25
92	−7198.49	−5.097	2.27×10^{-3}	-1.55×10^{-4}	0.60	-7.51×10^{-4}	1461.03	−74.64
93	476.02	−0.295	3.43×10^{-5}	6.00×10^{-4}	−3.44	4.37×10^{-3}	34.00	−132.83
94	3749.41	0.069	-3.24×10^{-5}	1.39×10^{-4}	−11.02	1.14×10^{-2}	−45.12	−374,414.07
95	860.66	0.065	-1.99×10^{-5}	-2.68×10^{-5}	−2.46	2.48×10^{-3}	−9.44	−98,727.09
96	387.57	0.000	-3.17×10^{-7}	2.78×10^{-6}	−1.19	1.21×10^{-3}	3.01	−52,764.44
97	−40.90	−0.062	2.84×10^{-6}	2.00×10^{-4}	0.27	-5.48×10^{-4}	−0.86	3.73
98	−65.30	0.002	-4.13×10^{-6}	3.36×10^{-5}	0.40	-6.11×10^{-4}	−1.96	5.24
99	−902.14	−0.031	4.74×10^{-6}	8.00×10^{-5}	2.59	-2.52×10^{-3}	0.81	103,437.48
100	2766.57	0.054	-8.54×10^{-6}	-8.46×10^{-5}	−7.92	7.68×10^{-3}	−3.01	−327,864.36
101	−551.60	0.281	-4.77×10^{-5}	-6.35×10^{-4}	2.91	-3.81×10^{-3}	−3.79	−12.88
102	334.39	−0.044	1.49×10^{-5}	4.95×10^{-5}	−1.18	1.25×10^{-3}	6.75	−39,319.98
103	8621.92	−0.019	2.17×10^{-5}	-2.28×10^{-4}	−28.04	2.97×10^{-2}	48.90	−949,369.22
104	−425.91	−0.056	1.89×10^{-6}	2.17×10^{-4}	1.26	-1.22×10^{-3}	−9.59	61,284.65
105	−76.07	0.038	-1.10×10^{-5}	-5.30×10^{-5}	0.26	-2.77×10^{-4}	0.43	36.77
106	0.16	−0.201	6.48×10^{-5}	2.67×10^{-5}	−1.20	1.18×10^{-3}	78.68	−51,364.38
107	−5.17	0.046	-1.06×10^{-5}	-1.26×10^{-5}	0.31	-3.39×10^{-4}	−14.06	0.60
108	−677.14	−0.104	1.55×10^{-5}	2.35×10^{-4}	1.65	-1.54×10^{-3}	7.87	82,605.10
109	915.94	−0.016	-3.56×10^{-6}	1.17×10^{-4}	−2.83	2.94×10^{-3}	−5.88	−94,980.76
110	−1663.48	0.211	-5.86×10^{-5}	-1.22×10^{-4}	5.99	-6.14×10^{-3}	−48.99	190,361.87
111	−288.61	−0.040	2.79×10^{-5}	-1.50×10^{-4}	0.31	-2.32×10^{-4}	38.92	−14.73
112	13,211.97	−0.034	1.98×10^{-6}	1.31×10^{-4}	−41.69	4.38×10^{-2}	−2.97	−1,390,552.83
113	−3716.29	−0.134	4.82×10^{-5}	1.37×10^{-4}	11.07	-1.13×10^{-2}	20.92	394,533.29
114	−194.12	0.006	1.38×10^{-7}	-6.99×10^{-5}	0.52	-6.85×10^{-4}	15.69	0.17
115	−434.95	0.004	3.54×10^{-5}	-2.46×10^{-4}	1.53	-1.94×10^{-3}	25.13	2.49
116	−160.03	0.048	-2.34×10^{-5}	2.49×10^{-5}	1.15	-1.73×10^{-3}	−9.81	−48.21
117	195.60	7.771	-2.08×10^{-3}	-1.36×10^{-3}	104.33	-1.62×10^{-1}	−3225.35	4.15
118	127.43	−0.021	-9.84×10^{-7}	8.65×10^{-5}	−0.77	1.13×10^{-3}	−1.23	3.22
119	−980.73	−0.137	1.67×10^{-4}	-1.48×10^{-3}	−4.86	1.04×10^{-2}	276.44	−2.50
120	41.32	0.018	-1.92×10^{-6}	-4.23×10^{-5}	−0.50	8.82×10^{-4}	3.08	−1.55
121	−1936.35	0.101	-2.90×10^{-5}	-5.30×10^{-5}	6.44	-6.69×10^{-3}	−20.54	204,158.22
122	0.97	0.071	-3.74×10^{-5}	8.39×10^{-5}	0.65	-1.02×10^{-3}	−24.95	1.66
123	−48.07	0.033	-1.03×10^{-5}	-1.79×10^{-5}	0.28	-3.53×10^{-4}	−5.04	0.94
124	−2999.33	0.053	-2.60×10^{-5}	5.89×10^{-5}	9.47	-9.67×10^{-3}	−18.88	330,889.01
125	−407.18	0.008	2.20×10^{-6}	-1.37×10^{-5}	1.18	-1.12×10^{-3}	−0.70	41,782.09
126	1452.11	0.004	-1.72×10^{-6}	4.59×10^{-6}	−4.46	4.53×10^{-3}	1.03	−161,529.34
127	1609.02	0.021	-8.32×10^{-6}	-5.68×10^{-6}	−4.87	4.93×10^{-3}	−2.69	−178,903.05
128	10.68	0.013	-2.88×10^{-6}	2.54×10^{-5}	−0.14	3.60×10^{-4}	−3.43	−1.12
129	−249.41	−0.019	1.79×10^{-5}	-4.63×10^{-5}	1.12	-1.67×10^{-3}	11.04	−1.74
130	−5601.28	−0.029	6.06×10^{-6}	9.34×10^{-5}	16.72	-1.66×10^{-2}	−2.06	625,762.42
131	1943.21	−0.015	5.00×10^{-6}	4.98×10^{-5}	−6.14	6.38×10^{-3}	4.35	−215,444.10
132	1973.71	−0.072	1.95×10^{-5}	1.05×10^{-4}	−6.08	5.97×10^{-3}	12.18	−228,276.88
133	−2087.93	−1.249	3.60×10^{-4}	4.06×10^{-4}	−0.93	1.02×10^{-3}	439.09	−21.03
134	−25.36	−0.006	1.14×10^{-6}	1.30×10^{-5}	0.10	-1.51×10^{-4}	0.58	1.44
135	433.52	0.069	-4.48×10^{-5}	2.43×10^{-4}	−1.36	2.03×10^{-3}	−45.50	3.74
136	−1476.05	−0.108	2.31×10^{-5}	2.02×10^{-4}	4.18	-4.20×10^{-3}	10.03	164,342.67
137	−49.11	0.187	-4.62×10^{-5}	-1.93×10^{-4}	0.99	-1.16×10^{-3}	−33.99	3.39
138	−124.36	0.017	4.46×10^{-6}	-8.00×10^{-5}	0.27	-2.10×10^{-4}	7.25	7.88
139	−161.27	0.013	7.31×10^{-6}	-3.18×10^{-5}	0.37	-2.63×10^{-4}	0.71	18,861.41

(*continued on next page*)

Table 3 (*continued*)

No.	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7
140	439.34	−0.013	1.26×10^{-6}	5.72×10^{-5}	−1.43	1.55×10^{-3}	−1.92	−45,085.94
141	1837.59	0.007	-5.92×10^{-6}	5.63×10^{-5}	−5.85	6.26×10^{-3}	−6.70	−187,579.63
142	−964.63	−0.050	9.53×10^{-6}	1.01×10^{-4}	2.86	-2.97×10^{-3}	5.23	101,856.34
143	154.02	0.059	-2.34×10^{-5}	2.10×10^{-5}	−0.49	8.21×10^{-4}	−19.17	5.30
144	−61.04	−0.060	1.33×10^{-5}	7.55×10^{-5}	−0.19	2.95×10^{-4}	15.81	−0.34
145	−91.13	0.010	1.53×10^{-6}	-3.05×10^{-5}	0.30	-3.54×10^{-4}	2.70	1.25
146	−2044.48	−0.067	1.48×10^{-5}	1.41×10^{-4}	5.85	-5.69×10^{-3}	3.98	233,327.78
147	384.04	−0.019	5.06×10^{-6}	4.45×10^{-5}	−1.22	1.24×10^{-3}	0.80	−44,257.62
148	10.31	−0.079	1.93×10^{-5}	1.40×10^{-4}	−0.30	3.69×10^{-4}	7.80	1.62
149	−79.29	−0.052	1.24×10^{-5}	6.04×10^{-5}	0.01	-3.90×10^{-5}	12.95	1.50
150	−13.36	−0.069	1.49×10^{-5}	1.09×10^{-4}	−0.38	$5.65 \times 10^{-4} \times 10^{-4}$	12.48	−3.63
151	−77.13	−0.024	8.64×10^{-6}	2.73×10^{-5}	0.16	-1.76×10^{-4}	6.26	−0.45
152	186.97	−0.257	-2.02×10^{-5}	1.06×10^{-3}	0.45	-1.71×10^{-3}	−33.22	3.37
153	0.34	0.003	5.40×10^{-5}	-4.32×10^{-4}	−1.84	3.62×10^{-3}	41.42	1.20
154	−32.81	0.016	-3.91×10^{-6}	-1.36×10^{-5}	−0.01	1.89×10^{-4}	−0.04	−2.69
155	−3.39	0.017	-5.48×10^{-6}	-8.53×10^{-6}	−0.13	3.20×10^{-4}	−0.94	1.02
156	−249.26	0.078	-9.95×10^{-6}	-1.58×10^{-4}	1.46	-2.01×10^{-3}	−5.99	−1.82
157	−9.69	−0.037	3.71×10^{-6}	9.18×10^{-5}	−0.17	2.25×10^{-4}	5.57	2.35
158	−1835.62	−0.845	2.97×10^{-4}	-1.29×10^{-4}	0.71	-8.42×10^{-4}	329.42	−11.27
159	0.31	0.049	-3.71×10^{-6}	-1.24×10^{-4}	−0.18	5.30×10^{-4}	−1.42	−1.50
160	44.26	−0.081	1.29×10^{-5}	2.07×10^{-4}	−0.28	2.97×10^{-4}	1.10	2.75
161	−94.41	−0.024	1.21×10^{-5}	4.80×10^{-6}	0.31	-4.18×10^{-4}	6.19	−0.93
162	−12.30	0.079	-2.34×10^{-5}	-5.22×10^{-5}	0.18	-9.26×10^{-5}	−11.59	16.92
163	1429.40	−0.016	5.10×10^{-7}	8.54×10^{-5}	−4.41	4.54×10^{-3}	−3.38	−152,612.74
164	−36.56	0.021	-4.22×10^{-6}	-1.76×10^{-5}	0.16	-1.78×10^{-4}	−1.57	−2.21
165	−69.00	−0.056	1.37×10^{-5}	5.96×10^{-5}	−0.18	2.93×10^{-4}	16.86	−8.38
166	5830.12	−0.206	5.82×10^{-5}	1.24×10^{-4}	−19.23	1.99×10^{-2}	63.96	−658,885.28
167	−0.99	−0.013	4.42×10^{-6}	5.06×10^{-6}	−0.24	4.69×10^{-4}	4.59	−1.05
168	−2923.03	−0.098	3.07×10^{-5}	7.40×10^{-5}	8.53	-8.70×10^{-3}	24.20	305,416.57
169	112.44	−0.040	1.13×10^{-5}	8.74×10^{-5}	−0.77	1.17×10^{-3}	1.00	3.11
170	106.56	0.105	-2.97×10^{-5}	-7.50×10^{-5}	−0.15	4.01×10^{-4}	−23.36	3.67
171	−13,659.77	0.031	-2.01×10^{-5}	1.97×10^{-4}	43.02	-4.46×10^{-2}	−39.27	1,487,919.98
172	−18.24	−0.011	1.87×10^{-6}	2.54×10^{-5}	0.00	2.29×10^{-5}	1.31	4.05
173	−42.82	0.044	-1.03×10^{-5}	-5.78×10^{-5}	0.20	-1.71×10^{-4}	−4.44	3.12
174	−32.24	−0.016	4.67×10^{-6}	2.82×10^{-5}	0.00	3.89×10^{-5}	3.71	−1.83
175	−100.38	−0.058	3.57×10^{-5}	7.10×10^{-7}	0.05	-1.74×10^{-5}	14.02	0.55
176	2.14	−0.086	3.24×10^{-5}	1.38×10^{-4}	−0.38	5.56×10^{-4}	8.87	−1.92
177	1346.38	−0.098	2.06×10^{-5}	2.00×10^{-4}	−4.41	4.60×10^{-3}	6.60	−141,805.22
178	23.81	0.022	-6.93×10^{-6}	9.39×10^{-6}	−0.15	3.36×10^{-4}	−5.20	−0.23
179	−160.90	−0.147	3.83×10^{-5}	2.34×10^{-4}	0.50	-8.99×10^{-4}	17.55	−2.76
180	−11.19	−0.008	9.67×10^{-6}	-4.01×10^{-5}	−0.49	9.44×10^{-4}	10.36	0.69
181	−184.93	−0.095	1.65×10^{-5}	1.87×10^{-4}	0.68	-1.16×10^{-3}	13.21	44.17
182	−78.04	−0.005	5.66×10^{-6}	-2.25×10^{-5}	0.23	-2.75×10^{-4}	4.42	−1.27
183	0.94	0.034	-8.40×10^{-6}	-3.84×10^{-5}	−0.03	1.21×10^{-4}	−3.12	−0.20
184	−16.28	0.065	-3.08×10^{-5}	3.83×10^{-5}	0.57	-8.45×10^{-4}	−19.31	11.93
185	50.56	−0.037	5.90×10^{-6}	8.97×10^{-5}	−0.49	7.47×10^{-4}	3.63	0.36
186	−1572.06	0.028	-8.59×10^{-6}	1.77×10^{-5}	5.10	-5.30×10^{-3}	−12.28	170,879.91
187	472.29	0.045	-1.67×10^{-5}	6.58×10^{-6}	−1.35	1.42×10^{-3}	−9.97	−49,967.76
188	−35.35	−0.044	9.82×10^{-6}	5.59×10^{-5}	−0.23	3.75×10^{-4}	11.37	0.70
189	−0.04	0.045	-1.20×10^{-5}	-3.13×10^{-5}	0.13	-7.83×10^{-5}	−9.23	0.14
190	−1016.15	0.041	-1.71×10^{-5}	3.20×10^{-5}	3.20	-3.22×10^{-3}	−11.13	113,622.34
191	−9888.68	0.148	-5.24×10^{-5}	8.59×10^{-5}	32.16	-3.36×10^{-2}	−68.81	1,082,525.80
192	−1468.07	0.055	-2.61×10^{-5}	1.18×10^{-4}	4.82	-4.76×10^{-3}	−30.13	175,156.62
193	−415.39	−0.122	4.34×10^{-5}	1.38×10^{-5}	1.04	-1.53×10^{-3}	43.61	20.11
194	2.14	0.071	-4.37×10^{-5}	9.66×10^{-5}	0.55	-8.69×10^{-4}	−22.63	2.17
195	0.06	0.022	-2.85×10^{-5}	1.67×10^{-4}	0.45	-7.23×10^{-4}	−18.02	−0.04
196	11.47	−0.070	1.73×10^{-5}	1.34×10^{-4}	−0.32	4.28×10^{-4}	6.92	0.70
197	−18.14	−0.011	3.92×10^{-6}	9.96×10^{-6}	−0.11	3.03×10^{-4}	3.19	6.16
198	−303.35	−0.184	6.24×10^{-5}	6.66×10^{-5}	0.16	-2.57×10^{-4}	53.32	−4.75
199	−141.35	−0.248	6.43×10^{-5}	2.52×10^{-4}	−0.53	5.40×10^{-4}	50.60	−0.51
200	−1602.81	−0.520	1.86×10^{-4}	-9.83×10^{-5}	3.36	-5.11×10^{-3}	203.24	−11.97
201	718.88	−0.018	8.52×10^{-6}	-1.12×10^{-6}	−4.84	7.68×10^{-3}	7.37	6.19
202	−38.32	−0.006	-2.87×10^{-6}	5.73×10^{-5}	0.17	-2.20×10^{-4}	−2.08	1.94
203	−44.52	0.123	-3.11×10^{-5}	-1.02×10^{-4}	0.88	-1.17×10^{-3}	−26.52	3.52
204	−127.08	0.009	1.63×10^{-6}	-6.44×10^{-5}	0.23	-2.16×10^{-4}	10.89	0.53
205	−1.50	0.105	-2.81×10^{-5}	-9.46×10^{-5}	0.34	-3.21×10^{-4}	−18.43	−0.20
206	44.33	−0.134	2.24×10^{-5}	2.29×10^{-4}	−0.97	1.28×10^{-3}	24.08	−5.17
207	−1972.88	0.128	-2.30×10^{-5}	-1.91×10^{-4}	8.10	-1.00×10^{-2}	−15.63	145,287.58
208	−22.21	−0.031	3.99×10^{-6}	8.16×10^{-5}	−0.03	2.56×10^{-5}	2.85	2.61
209	605.76	0.371	-1.06×10^{-4}	-6.44×10^{-6}	0.95	-1.28×10^{-3}	−152.58	6.90
210	−170.01	0.005	9.12×10^{-6}	-8.17×10^{-5}	0.57	-7.11×10^{-4}	8.98	−0.19

Table 4
AARD (%) for 210 compounds calculated for each of the 22 studied correlations.

No.	(10)	(11)	(12)	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)	(26)	(27)	(28)	(29)	(30)	(32)
1	21.08	9.99	21.08	28.65	21.05	17.35	23.18	13.73	12.96	**6.20**	21.14	14.74	18.42	12.89	9.85	13.77	10.08	13.880	15.31	11.69	8.62	6.34
2	19.34	10.70	19.34	11.80	19.09	10.47	18.59	18.47	8.40	10.63	19.31	10.64	12.17	11.41	6.70	12.55	7.00	12.007	11.83	9.56	8.67	**6.64**
3	24.35	9.48	24.35	13.96	20.21	9.41	19.53	21.08	12.55	9.02	24.36	9.46	12.24	9.59	8.49	12.17	9.18	9.76	9.45	8.91	**7.45**	7.58
4	5.38	2.37	5.38	4.07	7.00	**0.78**	6.83	4.94	3.60	2.37	5.38	3.70	3.91	3.90	1.57	3.40	6.95	3.973	4.02	1.66	1.16	1.17
5	46.65	43.30	46.65	44.95	43.95	45.80	43.09	46.42	43.37	40.90	46.53	43.46	45.40	43.38	41.59	45.89	**38.06**	43.202	46.76	44.81	42.94	39.98
6	13.30	7.74	13.30	8.79	10.28	14.13	9.92	12.43	7.37	7.28	13.26	7.83	10.70	7.67	5.98	11.16	6.33	8.229	9.45	8.40	7.67	**5.84**
7	17.12	12.78	17.12	13.50	14.95	17.37	13.76	16.23	10.77	**9.52**	17.03	12.00	15.00	12.96	11.21	16.31	9.73	12.797	14.98	13.57	11.41	9.56
8	9.44	3.61	9.44	3.62	10.74	7.42	9.47	7.46	3.27	3.46	9.40	3.43	5.31	3.59	2.91	5.20	**2.86**	4.137	5.53	3.66	3.39	2.99
9	9.16	9.09	9.16	9.02	8.53	9.09	9.05	8.42	3.21	3.14	9.02	3.29	8.48	8.56	3.26	4.08	6.57	9.078	3.51	8.09	7.56	**3.06**
10	6.23	6.00	6.23	7.14	7.00	5.98	7.38	5.96	4.56	5.78	6.21	5.99	5.75	7.09	5.08	6.04	4.70	7.070	6.16	5.22	**4.13**	4.30
11	12.34	8.80	12.34	10.59	10.62	9.87	9.96	9.97	**7.69**	8.89	12.38	9.32	12.25	10.48	9.01	9.71	9.07	9.946	10.70	8.87	8.60	7.99
12	3.63	3.39	3.63	10.81	10.42	11.89	11.72	3.19	5.68	3.18	3.64	3.38	3.38	9.74	3.21	**3.08**	3.38	11.115	10.82	3.20	4.78	3.11
13	33.35	24.00	33.36	26.70	31.12	27.31	29.58	29.76	26.67	23.99	33.27	24.19	26.97	27.31	24.03	26.37	24.35	27.200	27.99	23.98	23.66	**18.10**
14	18.40	13.51	18.40	14.35	18.03	11.59	17.54	18.54	12.46	13.22	18.46	14.10	16.85	14.36	11.61	17.06	12.18	15.412	15.28	11.78	**10.86**	11.62
15	3.54	2.87	3.54	3.78	6.63	12.40	6.60	3.54	4.37	2.99	3.51	2.88	2.79	3.57	2.60	3.03	3.06	3.618	2.95	3.14	2.75	**2.59**
16	6.46	4.29	6.46	6.21	8.06	13.41	6.69	5.34	4.87	4.22	6.47	5.32	6.40	4.80	4.20	5.38	9.94	4.438	5.85	4.96	3.90	**3.41**
17	31.91	23.55	31.91	30.98	32.82	**13.29**	33.22	28.66	21.32	15.01	31.89	17.11	22.95	25.64	17.55	22.68	26.03	24.541	23.81	23.87	21.66	14.21
18	13.15	9.39	13.15	9.67	19.06	15.30	18.39	11.97	8.45	**7.18**	13.16	7.96	12.00	9.34	7.28	11.21	9.09	9.800	10.16	9.28	8.66	7.37
19	7.59	5.13	7.59	14.31	10.38	17.59	10.44	7.22	8.41	4.40	7.58	5.77	7.00	10.49	5.93	7.31	4.78	10.628	8.51	6.15	5.10	**4.27**
20	14.68	9.67	14.68	9.75	13.99	8.46	14.61	14.89	5.56	10.44	14.67	10.21	7.65	9.06	5.32	5.93	5.48	10.179	7.65	7.74	4.94	**4.55**
21	5.95	2.65	5.95	2.61	10.03	6.58	9.67	4.33	4.60	2.22	5.95	2.22	3.84	2.66	**1.97**	3.30	6.91	2.928	3.02	2.69	2.74	2.16
22	3.57	3.10	3.57	13.79	11.49	14.18	13.10	**2.32**	6.04	2.91	3.60	3.05	3.20	12.01	2.44	2.37	3.11	13.865	11.00	2.53	3.14	2.38
23	19.92	12.88	19.92	32.29	25.32	31.29	26.39	19.48	17.78	6.57	19.90	6.66	14.94	21.43	**6.13**	15.83	10.44	20.850	16.39	8.03	7.34	6.26
24	9.60	**6.33**	9.60	17.43	11.42	18.02	12.33	8.84	7.13	6.38	9.59	7.44	7.49	9.86	7.62	7.59	7.95	10.196	8.84	7.82	6.73	6.65
25	3.56	2.42	3.56	4.93	4.79	11.21	3.94	2.39	4.15	**1.56**	3.50	1.66	1.88	2.78	1.73	2.26	6.66	2.702	3.53	2.29	1.92	1.58
26	21.92	23.69	21.92	26.60	24.19	31.63	23.60	21.97	23.17	24.04	21.89	23.79	22.51	25.77	21.47	24.01	**17.43**	25.408	24.15	21.94	22.05	21.59
27	6.50	3.52	6.50	6.82	8.07	16.53	7.47	4.17	7.70	2.53	6.46	4.91	4.32	6.56	3.40	4.18	7.40	6.431	5.54	3.39	**1.89**	1.99
28	16.62	15.18	16.62	20.01	23.99	11.48	23.96	16.59	8.53	**7.20**	16.58	10.25	16.15	18.87	10.96	16.08	10.61	19.112	15.73	15.68	14.90	7.23
29	8.72	8.22	8.72	8.52	9.02	5.20	8.76	8.20	6.74	5.27	8.70	6.04	8.01	8.80	5.76	7.69	6.90	8.767	8.17	7.83	6.72	**4.53**
30	8.62	2.66	8.62	4.97	8.63	3.88	8.13	7.02	2.52	2.37	8.61	4.19	4.65	2.99	4.31	3.19	2.70	2.924	4.03	4.28	2.56	**2.25**
31	12.96	12.79	12.96	14.01	16.13	14.39	16.23	12.46	15.34	11.50	13.02	11.72	13.97	13.67	12.33	12.46	11.83	14.157	13.12	12.47	12.43	**8.95**
32	24.87	24.48	24.87	26.19	25.29	19.68	26.95	24.29	20.28	24.56	24.76	23.97	23.77	25.98	22.99	24.59	23.39	28.374	23.55	23.86	24.05	**17.03**
33	7.58	5.81	7.58	12.65	9.90	27.24	9.85	7.11	11.42	5.34	7.65	6.08	7.23	9.71	6.04	6.80	11.18	9.742	5.03	6.85	6.53	**4.89**
34	9.17	7.10	9.17	10.69	11.01	14.08	11.00	8.14	9.78	6.63	9.19	8.44	8.95	10.55	8.26	8.12	9.29	10.612	8.58	8.30	7.81	**6.20**
35	10.07	5.43	10.07	6.39	12.74	9.15	11.47	3.60	5.60	5.31	10.06	6.15	7.11	5.51	**2.81**	3.53	5.52	5.240	5.74	3.83	4.02	2.83
36	36.64	36.42	36.64	39.79	49.29	17.36	50.08	36.54	**15.69**	36.42	36.59	36.61	25.89	32.06	36.41	20.92	44.94	34.397	31.29	36.42	37.04	35.88
37	11.01	7.22	11.02	9.51	7.25	17.26	6.73	9.81	10.15	6.25	10.83	7.38	8.85	6.62	7.41	9.26	7.05	6.721	10.06	7.65	6.97	**4.49**
38	5.74	3.93	5.74	5.57	9.10	12.38	7.99	4.58	4.45	3.45	5.73	3.87	4.98	3.87	4.08	4.49	9.00	3.603	5.25	4.30	**3.43**	3.49
39	8.12	6.15	8.12	16.68	10.65	17.63	11.22	8.04	6.91	6.02	8.09	6.01	6.11	9.73	5.50	6.03	9.44	9.591	8.76	5.55	5.77	**5.31**
40	5.15	3.00	5.14	5.66	5.57	5.82	5.16	5.35	5.65	2.63	5.19	4.71	5.98	4.92	4.45	5.39	6.21	5.151	5.44	4.65	3.31	**2.28**
41	18.88	17.66	18.87	17.56	18.28	16.45	19.90	18.04	19.45	16.95	19.02	16.98	20.41	17.55	15.38	16.68	14.44	18.975	16.80	15.18	16.24	**12.56**
42	12.97	4.04	12.97	10.58	3.71	30.56	8.64	10.70	11.94	3.98	12.86	5.90	10.13	**3.64**	5.44	9.09	4.62	6.655	10.64	5.51	3.84	3.89
43	4.59	2.67	4.59	5.05	8.53	10.27	8.03	3.88	5.25	2.68	4.60	3.36	3.26	4.39	3.43	**2.60**	7.72	4.441	3.70	3.43	3.05	2.88
44	24.28	21.51	24.28	22.44	24.73	27.82	24.75	24.57	17.69	21.54	24.28	21.86	18.35	22.43	19.82	16.81	27.25	22.989	**15.80**	20.22	17.04	16.82
45	2.17	1.16	2.17	3.30	11.92	9.69	11.33	0.69	5.77	1.16	2.17	1.95	2.00	2.53	0.32	0.74	0.39	2.347	1.51	0.59	0.70	**0.17**
46	15.88	10.61	15.88	15.10	17.15	7.67	17.17	15.89	9.85	8.43	15.87	11.12	11.87	12.98	9.93	10.98	13.86	12.945	10.53	12.34	10.16	**7.01**
47	100.92	35.72	100.92	95.23	96.50	111.05	98.32	118.98	92.93	36.51	100.88	93.44	98.71	85.47	81.82	93.80	111.90	84.770	95.83	96.76	35.20	**35.02**
48	13.82	5.48	13.82	8.09	15.35	6.31	14.00	6.24	4.32	5.57	13.82	5.45	9.33	5.84	2.90	5.93	10.42	5.510	3.65	3.98	4.61	**2.36**

(*continued on next page*)

Table 4 (*continued*)

No.	(10)	(11)	(12)	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)	(26)	(27)	(28)	(29)	(30)	(32)
49	12.57	5.54	12.57	6.99	11.68	7.30	10.61	8.38	5.28	5.03	12.55	5.25	8.16	6.33	**4.69**	7.25	14.31	6.174	7.92	5.74	5.32	4.75
50	11.82	11.99	11.82	15.31	12.22	15.15	13.10	12.00	8.33	11.17	11.69	11.13	10.68	12.22	6.26	11.05	6.35	13.225	12.55	11.16	9.67	**4.87**
51	6.52	5.55	6.52	10.55	6.44	10.26	6.70	6.53	4.36	4.79	6.49	5.15	4.66	5.50	4.77	3.87	4.78	4.412	6.13	4.76	**2.80**	3.10
52	15.97	10.85	15.97	12.52	14.31	10.68	13.82	15.94	7.17	9.99	15.87	12.23	12.82	12.44	**6.10**	12.96	6.58	13.404	15.06	11.02	9.16	6.60
53	12.57	10.11	12.57	12.35	14.16	**8.05**	14.07	12.50	8.55	9.80	12.55	12.07	9.42	10.33	11.86	8.69	13.00	10.45	10.04	12.29	9.83	9.39
54	4.95	3.43	4.96	3.45	5.66	3.39	3.55	3.20	2.88	2.92	4.71	2.95	3.07	3.44	2.86	3.20	2.90	3.05	5.42	3.24	2.90	**2.85**
55	31.41	26.99	31.40	31.71	32.63	6.06	32.72	23.29	24.87	6.02	31.42	8.78	32.46	32.11	11.83	21.95	5.94	31.92	31.62	23.33	19.87	**4.96**
56	10.49	5.87	10.49	9.66	8.59	11.53	10.38	11.66	11.71	5.68	10.64	9.09	12.66	8.88	8.56	11.07	19.58	10.45	11.44	8.57	6.95	**5.44**
57	19.56	13.08	19.56	16.21	19.34	16.06	18.91	19.39	15.04	12.22	19.42	15.83	16.95	17.17	15.43	17.63	22.47	17.04	18.02	15.78	**11.85**	12.10
58	58.53	42.33	58.52	54.43	60.78	35.86	59.69	50.96	40.29	38.15	58.69	41.07	49.80	39.18	40.35	46.92	38.70	38.55	43.57	45.19	42.90	**33.70**
59	2.39	2.33	2.39	2.37	3.66	0.64	3.46	2.38	3.01	0.70	2.40	0.70	2.88	2.36	0.61	2.11	0.76	2.39	1.81	1.95	2.22	**0.61**
60	2.26	2.25	2.26	5.08	2.29	6.21	2.17	2.18	2.87	1.80	2.19	1.80	1.76	2.29	1.44	1.75	**1.33**	1.95	2.93	1.74	1.42	1.35
61	7.42	5.36	7.42	7.64	5.09	14.90	6.41	6.21	8.12	4.17	7.06	5.00	5.57	5.07	4.62	5.84	29.17	4.79	11.18	5.43	**4.11**	4.21
62	5.79	4.12	5.79	7.75	**3.93**	16.12	5.22	5.61	4.76	4.10	5.75	5.14	5.47	3.94	4.79	5.61	17.12	4.54	7.34	4.97	4.13	4.15
63	23.98	15.71	23.98	18.58	19.11	20.99	19.69	23.46	19.45	16.63	23.97	19.19	21.87	17.93	17.76	21.77	21.70	18.89	19.06	17.61	**14.48**	14.89
64	9.19	8.83	9.18	8.68	9.76	7.55	10.46	9.08	12.85	6.71	9.52	6.62	11.71	8.83	**6.08**	6.91	7.27	9.67	6.51	6.54	7.35	6.29
65	7.10	4.97	7.10	7.91	6.14	19.74	6.50	6.78	4.91	5.12	7.09	5.28	5.38	5.43	**4.22**	4.74	5.63	6.04	5.23	4.41	4.45	4.34
66	12.13	7.19	12.13	10.78	9.09	32.55	9.96	11.91	4.72	7.09	12.16	8.99	10.65	8.16	5.38	9.09	4.89	9.54	9.44	6.87	6.31	**4.54**
67	6.80	6.13	6.80	8.55	6.93	10.44	6.88	6.74	8.46	6.19	6.84	6.49	6.39	6.97	5.79	6.22	**5.72**	6.71	6.66	5.83	5.94	5.81
68	6.61	5.86	6.61	8.66	6.85	18.69	7.62	6.62	**4.73**	5.17	6.70	4.76	7.07	6.44	5.06	6.57	7.24	7.45	6.07	5.84	6.53	5.50
69	6.80	5.35	6.81	7.54	5.84	8.21	5.97	5.93	4.80	4.75	6.65	5.89	6.22	5.85	5.97	5.63	13.69	5.29	8.35	6.04	4.12	**3.88**
70	5.25	4.14	5.25	8.20	4.06	11.64	3.54	5.27	3.72	3.31	5.22	3.83	3.18	4.07	3.37	2.83	5.96	2.75	5.51	3.37	2.39	**2.30**
71	21.26	14.38	21.26	15.69	20.57	10.75	20.05	21.43	12.02	14.61	21.23	15.82	15.35	15.92	9.59	14.80	10.89	17.58	16.71	12.48	10.95	**8.92**
72	5.31	4.99	5.31	8.69	5.49	16.45	**4.78**	5.08	6.76	5.01	5.30	5.32	5.17	5.41	5.18	4.97	9.32	5.18	5.99	5.04	4.80	4.92
73	47.16	35.81	47.16	54.07	83.43	20.59	82.08	47.18	24.08	35.73	47.21	36.68	37.84	38.63	21.36	34.77	28.08	38.95	33.40	23.66	26.87	**19.63**
74	8.54	2.83	8.54	11.47	2.81	26.76	7.01	7.38	8.63	2.71	8.47	4.76	7.55	2.90	4.58	7.03	25.41	5.50	8.34	4.54	3.71	**2.65**
75	28.28	24.09	28.28	24.29	25.64	18.14	26.23	30.72	17.77	24.02	28.30	24.82	25.43	25.79	17.13	22.72	38.34	27.77	25.86	18.75	**15.02**	18.06
76	17.91	12.67	17.91	12.15	17.59	11.61	16.55	15.85	**10.69**	12.72	17.87	12.26	13.69	12.63	10.72	14.28	24.64	13.03	13.31	12.43	12.68	12.26
77	92.28	60.85	92.28	714.91	630.01	454.94	855.59	113.30	**56.47**	56.82	92.49	76.48	66.92	356.62	102.86	89.85	85.24	669.08	69.05	116.94	71.36	81.99
78	29.60	14.46	29.60	24.80	29.58	13.35	29.60	24.66	23.84	12.14	29.33	17.32	26.71	21.87	12.71	21.66	12.70	21.23	37.53	21.31	15.40	**10.73**
79	10.30	7.23	10.30	8.56	9.00	9.01	9.92	10.45	**6.93**	7.19	10.28	8.46	10.19	8.44	7.60	8.90	24.79	9.91	9.93	7.51	7.10	7.11
80	27.71	11.04	27.71	18.48	27.52	4.53	26.32	24.26	10.25	10.24	27.73	10.69	10.24	11.88	7.67	10.32	18.19	12.31	9.70	8.62	5.55	**4.49**
81	13.84	8.22	13.85	9.91	8.13	16.12	8.19	11.45	5.82	6.98	13.68	7.64	9.44	7.39	5.79	9.88	5.68	8.28	12.39	8.72	7.90	**5.62**
82	8.85	7.72	8.85	8.44	16.91	5.73	16.77	8.85	**4.39**	7.73	8.85	8.29	6.72	8.21	7.50	6.48	8.46	8.21	5.89	7.53	7.42	7.00
83	16.66	7.47	16.66	10.85	19.32	3.85	18.30	14.79	5.00	7.40	16.66	7.54	8.00	9.14	3.42	7.93	19.14	9.44	8.11	4.58	3.53	**2.96**
84	21.90	20.29	21.90	27.97	30.70	23.74	31.65	14.74	16.51	21.24	21.89	21.13	21.99	28.17	**9.32**	15.12	14.57	29.85	24.19	17.93	10.56	12.75
85	9.54	6.60	9.54	7.46	9.68	20.26	10.17	9.56	13.19	5.40	9.61	5.31	8.11	6.70	4.22	5.81	4.29	8.61	5.84	4.24	5.01	**4.06**
86	19.74	12.47	19.74	14.09	17.02	11.60	17.09	19.48	10.05	13.02	19.73	13.29	15.49	14.25	10.60	15.54	25.14	14.50	14.52	11.20	11.05	**9.87**
87	3.70	3.57	3.70	4.61	4.63	8.75	5.38	3.69	3.77	3.69	3.69	3.80	3.70	4.62	3.64	3.72	3.69	4.72	3.72	3.62	**3.54**	3.68
88	6.68	6.07	6.68	7.17	7.05	9.38	7.36	6.70	6.27	6.31	6.69	6.45	6.41	7.40	4.46	6.18	**4.21**	7.53	6.29	5.51	5.78	4.52
89	6.57	3.93	6.57	7.26	6.77	3.48	7.43	5.48	5.59	3.33	6.56	3.51	6.61	6.24	3.46	5.47	3.56	7.54	4.24	3.67	2.68	**2.55**
90	7.56	7.55	7.56	7.75	8.03	6.52	8.15	7.53	1.38	1.47	7.64	1.45	7.99	7.27	1.45	2.37	2.21	7.12	3.21	7.57	7.95	**1.22**
91	4.83	4.73	4.83	5.57	5.11	3.57	5.54	4.92	**1.60**	2.40	4.73	2.80	3.42	4.46	2.69	3.39	3.43	5.56	3.29	4.17	3.89	2.11
92	8.13	4.45	8.13	8.49	8.50	5.11	8.68	8.16	8.23	3.87	8.14	6.70	8.21	8.61	6.33	7.78	6.08	8.71	8.07	6.95	4.63	**3.54**
93	112.31	86.65	112.31	122.17	114.93	119.25	114.57	99.13	106.46	87.74	112.31	87.10	108.04	114.74	71.76	97.92	**40.72**	115.27	119.79	87.11	87.27	79.83
94	8.45	6.14	8.46	8.47	8.64	6.10	7.90	8.47	6.34	5.91	8.39	8.34	8.61	8.09	6.40	8.33	6.70	8.18	9.09	7.63	5.83	**5.48**
95	11.23	3.98	11.23	14.03	5.78	34.24	11.52	10.70	10.80	4.08	11.28	7.23	11.15	5.45	6.72	10.01	6.14	8.58	10.26	6.94	5.05	**3.92**
96	0.93	0.93	0.93	6.89	3.43	6.24	5.09	0.95	7.02	0.92	0.92	0.94	4.62	3.07	0.93	0.95	1.79	4.38	0.88	0.95	2.00	**0.85**
97	7.19	6.94	7.19	7.99	7.61	5.56	8.10	6.86	3.49	6.13	7.20	6.03	6.35	7.34	**2.42**	3.82	2.80	8.16	4.97	2.45	2.48	2.43
98	6.26	5.28	6.26	8.70	12.05	17.46	11.36	6.39	5.89	5.05	6.26	6.06	6.43	4.94	5.38	6.43	4.97	5.50	6.43	5.53	**4.55**	4.62
99	14.16	13.44	14.16	13.53	17.91	**4.87**	19.82	12.56	6.41	13.26	14.17	13.02	8.32	10.68	6.15	6.41	5.83	10.77	10.89	6.86	6.75	4.96

100	22.53	18.16	22.52	18.71	20.44	22.87	21.89	22.28	26.38	10.75	22.84	10.85	26.11	18.80	10.16	19.91	27.67	21.13	17.71	17.05	20.60	**8.63**
101	69.54	45.50	69.54	81.04	122.67	26.85	120.77	69.53	30.19	45.44	69.61	49.06	52.66	54.45	25.66	47.18	38.49	55.05	47.63	28.27	32.74	**24.10**
102	10.45	4.93	10.45	11.32	13.94	7.76	14.62	10.79	6.34	4.59	10.43	9.48	6.29	11.60	6.96	6.39	7.90	13.04	7.89	9.61	2.85	**2.72**
103	31.20	31.38	31.21	31.64	62.67	**6.83**	61.67	29.35	7.36	28.20	31.12	28.89	16.48	12.27	27.72	16.69	28.02	12.19	11.84	27.72	27.73	26.54
104	24.37	16.43	24.37	21.02	25.97	14.83	25.63	24.56	15.28	16.35	24.37	18.85	20.90	20.85	12.01	19.00	23.94	21.36	21.22	13.14	**11.57**	12.03
105	10.96	8.60	10.96	23.98	16.13	36.60	17.06	10.55	9.67	8.18	10.93	7.89	9.16	13.43	7.88	9.16	7.91	13.36	10.86	7.89	**7.81**	8.37
106	10.58	9.67	10.57	10.89	11.34	13.29	11.24	10.56	13.63	9.15	10.61	10.24	11.31	10.79	10.13	10.45	10.50	11.10	9.48	10.45	9.82	**9.14**
107	10.25	**5.15**	10.26	6.76	12.94	7.61	11.89	7.45	5.85	5.16	10.24	5.39	6.89	5.77	5.36	6.19	11.32	5.81	5.97	5.40	5.23	5.16
108	19.86	19.77	19.86	21.76	20.09	21.04	21.23	20.63	13.39	19.77	19.83	19.65	19.33	19.95	13.11	15.90	15.32	21.41	18.84	17.54	15.63	**12.57**
109	15.78	8.28	15.78	9.89	10.81	13.97	11.65	15.32	6.87	8.13	15.73	10.09	12.04	9.45	4.40	11.42	4.00	11.32	12.19	6.28	3.71	**3.02**
110	10.80	10.31	10.80	13.20	10.66	15.76	11.13	10.54	11.77	10.21	10.79	10.82	10.65	10.73	10.49	10.19	16.30	10.18	12.58	10.45	9.61	**9.27**
111	17.53	10.67	17.53	27.73	21.57	30.23	22.84	16.31	15.42	11.57	17.50	11.79	13.85	18.38	11.11	13.98	**9.31**	18.38	16.48	10.86	11.35	11.39
112	16.34	8.06	16.34	9.50	14.27	11.62	13.55	13.50	12.06	8.19	16.32	8.59	12.97	9.39	8.02	11.63	8.28	9.20	12.16	8.60	8.13	**7.73**
113	25.92	8.73	25.92	20.24	27.14	8.08	25.93	20.49	7.77	8.67	25.90	16.31	9.04	8.85	15.37	8.99	9.74	8.84	9.35	16.50	7.36	**6.49**
114	8.97	4.32	8.97	11.26	5.97	12.23	6.04	8.85	5.97	4.29	8.97	4.30	4.48	5.54	3.21	4.28	3.83	5.00	3.54	3.59	3.82	**3.00**
115	53.13	40.69	53.13	66.36	74.04	46.84	86.95	51.94	40.31	34.35	53.22	52.96	52.86	55.87	48.58	49.34	28.23	59.17	55.67	52.74	44.26	**20.88**
116	10.81	8.40	10.81	22.37	11.84	52.86	13.36	11.21	11.35	8.53	10.81	11.90	10.43	9.67	11.69	10.63	10.47	10.64	9.73	11.86	**6.22**	8.09
117	17.61	12.95	17.61	17.63	17.37	7.25	17.39	17.54	**7.20**	12.95	17.61	12.99	9.41	16.82	11.36	7.37	17.51	16.56	7.64	8.15	7.67	7.68
118	7.11	6.30	7.11	6.60	10.06	10.31	10.29	6.94	5.85	5.10	7.09	6.30	6.68	6.53	**2.69**	6.39	5.42	6.86	6.41	6.01	5.51	3.46
119	8.65	8.33	8.65	8.35	8.80	5.73	8.59	6.71	6.51	6.91	8.63	6.89	8.24	8.30	**5.21**	5.70	6.88	8.21	6.73	5.53	5.37	5.29
120	6.89	6.69	6.89	15.60	6.85	35.96	7.30	4.93	7.18	4.45	6.81	4.57	6.45	6.93	4.80	4.51	4.22	7.40	7.98	4.61	4.08	**3.96**
121	20.95	9.32	20.94	13.26	13.24	22.36	14.38	19.79	18.40	7.61	21.13	11.91	21.99	12.59	11.53	18.89	43.60	14.10	14.67	12.68	12.20	**6.54**
122	10.95	8.49	10.95	15.87	11.53	23.71	13.82	8.97	6.20	8.35	10.95	10.84	10.92	10.38	8.38	9.31	7.99	12.51	9.86	8.38	**6.18**	6.25
123	58.09	**2.97**	58.09	37.83	68.61	10.90	68.50	47.77	7.32	3.42	58.09	11.68	30.73	20.30	6.60	24.60	42.77	20.37	23.47	10.79	4.31	2.98
124	19.38	14.86	19.38	21.50	16.44	20.91	19.11	19.25	18.75	15.23	19.51	19.87	22.06	16.34	18.27	19.84	17.07	18.49	19.50	18.33	13.96	**13.53**
125	15.00	9.64	15.00	11.26	17.96	12.72	16.60	11.62	11.02	10.03	14.98	9.99	10.68	9.60	10.08	**9.51**	14.95	9.58	10.52	9.65	9.54	9.98
126	114.23	19.48	114.23	98.66	131.35	16.82	131.24	55.55	**16.75**	19.82	114.22	24.37	40.16	25.30	22.69	36.90	67.19	25.26	19.92	22.54	18.75	19.27
127	5.33	4.08	5.33	9.66	4.46	12.18	5.50	5.05	7.90	4.02	5.40	4.95	6.32	4.54	4.82	4.88	4.81	5.50	4.25	4.95	4.29	**3.22**
128	18.77	3.90	18.77	4.73	8.30	9.75	7.19	13.22	4.49	3.41	18.76	3.58	10.18	4.62	3.36	7.57	36.50	4.80	7.75	4.11	3.56	**3.18**
129	8.43	4.78	8.43	7.94	10.21	8.29	10.54	7.94	4.04	2.37	8.44	6.18	4.70	7.48	6.29	4.82	9.90	8.13	4.97	7.12	3.97	**2.29**
130	35.00	24.13	35.00	27.37	33.87	24.05	32.08	33.06	26.21	25.74	34.95	25.69	25.05	24.58	22.49	27.77	44.13	24.87	26.21	24.97	23.64	**21.68**
131	6.91	3.61	6.91	3.89	5.33	10.63	4.43	5.42	5.96	3.74	6.74	3.82	3.85	3.55	3.49	3.89	18.98	4.27	6.98	3.62	3.42	**3.37**
132	24.60	22.38	24.60	22.23	26.02	24.61	25.68	24.51	23.88	19.62	24.73	20.04	21.81	22.40	20.80	19.99	19.07	23.06	**18.96**	21.31	21.10	19.39
133	5.73	4.29	5.73	5.98	6.45	5.84	6.22	5.72	4.95	4.20	5.72	5.71	5.35	5.98	5.73	4.89	5.39	6.06	5.74	5.78	3.20	**2.93**
134	1.51	1.32	1.51	1.29	11.81	1.52	11.65	1.53	1.15	0.75	1.51	0.77	1.39	1.29	0.83	1.00	0.67	1.41	1.35	22.59	0.60	**0.56**
135	27.10	22.96	27.10	22.26	27.89	13.81	27.14	25.60	**11.68**	23.69	27.05	23.93	20.98	20.05	25.57	22.18	45.20	21.39	19.93	12.24	22.89	25.49
136	18.98	14.05	18.98	13.74	18.62	11.33	18.03	18.88	9.08	14.28	18.96	14.12	13.89	13.82	10.10	12.43	10.34	15.11	13.49	**4.21**	9.46	8.99
137	5.58	4.01	5.58	4.79	5.56	9.60	4.47	4.30	5.96	4.02	5.58	4.36	4.96	4.09	4.12	4.28	10.54	3.79	5.13	6.42	3.82	**2.96**
138	11.24	8.17	11.24	12.20	9.98	42.34	9.02	6.70	16.23	8.21	11.25	8.15	9.73	9.11	6.38	6.00	13.77	8.80	7.63	12.27	**4.92**	6.19
139	28.77	16.45	28.77	18.75	15.79	15.75	12.01	23.59	27.08	14.11	28.61	14.42	26.35	13.94	11.74	10.96	18.31	12.55	17.52	**5.84**	11.69	10.17
140	11.74	5.85	11.75	6.79	13.63	6.14	12.35	8.93	5.02	5.10	11.67	5.15	7.14	6.33	**3.72**	7.24	13.25	6.38	7.47	14.85	5.21	3.82
141	20.88	15.55	20.89	16.13	17.73	14.07	16.05	15.17	14.43	9.30	20.63	9.96	15.56	16.13	9.62	14.80	10.40	15.28	19.48	**7.18**	12.86	8.07
142	12.01	11.06	12.01	11.43	14.14	12.16	14.45	12.12	9.22	10.30	12.06	10.23	11.43	10.91	7.18	8.03	7.04	12.74	11.25	**5.35**	6.49	6.54
143	8.37	4.57	8.37	5.66	8.86	4.03	7.91	6.88	5.06	**3.09**	8.34	4.51	6.56	5.69	4.14	6.37	9.90	5.47	7.44	3.82	3.85	3.19
144	10.72	2.89	10.72	23.79	17.61	21.18	19.54	6.20	9.02	2.94	10.71	3.72	7.11	10.81	3.47	5.59	10.08	10.85	5.68	3.74	1.75	**1.39**
145	6.53	3.83	6.53	4.21	5.43	10.72	5.31	5.45	4.94	3.81	6.54	4.08	5.45	4.25	3.67	3.84	**3.46**	4.14	4.34	34.97	3.77	3.52
146	67.44	67.50	67.44	87.83	114.14	29.99	135.37	36.18	44.09	66.29	67.57	65.40	56.40	53.50	24.31	27.56	45.49	48.56	77.13	**19.37**	25.23	27.36
147	27.60	21.41	27.60	21.50	31.60	15.00	35.25	25.39	16.02	21.16	27.60	21.28	17.57	19.01	**14.11**	18.83	19.62	19.61	26.28	14.49	17.68	15.24
148	19.07	16.11	19.07	17.08	18.34	11.54	21.47	19.14	10.00	16.36	19.07	16.36	13.97	17.70	11.15	13.15	11.77	19.97	17.14	**1.05**	12.16	10.69
149	2.60	1.91	2.60	4.45	2.87	2.52	4.46	1.17	3.21	1.18	2.83	1.17	4.67	2.75	1.04	0.93	0.87	4.34	1.87	7.68	0.86	**0.86**
150	9.30	7.38	9.30	18.77	14.67	18.23	16.55	7.32	8.45	7.38	9.28	8.08	8.92	14.11	6.48	6.65	7.29	15.69	10.14	8.70	**4.80**	5.00
151	9.18	7.79	9.18	19.47	18.88	24.73	20.80	9.03	9.11	7.67	9.17	9.38	8.27	18.17	8.44	7.55	9.21	20.02	9.68	13.59	8.25	**6.67**
152	33.28	29.32	33.28	33.06	37.41	13.02	37.79	29.97	13.42	29.66	33.29	31.33	29.22	32.50	12.87	19.66	34.82	32.84	27.33	**9.00**	14.93	11.62

(*continued on next page*)

Table 4 (*continued*)

No.	(10)	(11)	(12)	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)	(26)	(27)	(28)	(29)	(30)	(32)
153	15.09	8.85	15.09	13.41	23.31	6.29	23.11	11.94	10.50	8.83	15.09	9.17	11.01	8.76	8.43	7.80	12.63	8.92	8.42	**3.77**	6.64	4.99
154	5.92	3.66	5.92	6.62	5.11	16.06	4.71	4.32	6.06	**2.85**	5.86	3.07	4.07	3.32	3.52	4.10	11.77	3.71	5.23	3.58	3.12	2.89
155	3.99	3.40	3.99	5.97	4.96	11.37	4.03	3.54	5.02	**2.85**	3.95	2.94	3.19	3.38	3.12	3.55	6.40	3.35	4.35	7.24	2.97	2.90
156	9.93	7.31	9.93	7.80	12.68	4.84	12.17	8.33	5.28	6.19	9.93	6.39	8.42	7.23	6.02	8.09	6.77	7.27	4.81	**3.00**	7.59	5.61
157	5.23	3.83	5.23	8.82	4.28	8.25	5.68	3.08	3.87	3.83	5.24	3.74	4.50	3.36	2.54	2.92	**2.52**	3.73	3.79	10.85	2.83	2.59
158	18.71	8.63	18.71	18.50	22.24	9.03	28.83	12.67	9.30	8.65	18.70	12.13	17.16	9.21	7.92	11.04	11.78	9.89	10.08	**2.41**	8.47	8.07
159	7.35	3.68	7.35	4.25	15.08	8.93	14.18	2.97	5.43	3.70	7.35	4.23	5.47	4.02	2.13	3.12	4.86	3.81	2.71	16.32	2.80	**2.05**
160	38.88	23.00	38.88	28.68	41.27	14.40	40.33	36.82	11.04	22.30	38.88	22.63	24.42	22.43	16.49	24.05	38.49	22.76	19.19	**9.11**	15.57	15.95
161	10.61	3.76	10.61	10.73	16.78	13.28	17.60	10.58	5.42	3.69	10.61	9.07	6.98	9.80	9.13	7.10	8.80	10.15	8.30	6.05	4.97	**3.67**
162	6.14	5.69	6.15	10.10	6.19	15.75	6.98	6.09	6.54	5.70	6.11	5.99	5.90	5.99	6.04	5.75	12.96	5.95	6.61	7.55	5.61	**5.54**
163	16.38	7.43	16.38	8.75	14.21	10.25	13.21	14.30	7.20	7.42	16.34	8.31	10.92	8.78	6.59	11.08	25.15	9.32	10.89	**6.14**	6.67	6.56
164	18.98	5.20	18.98	5.81	6.09	15.70	6.76	15.79	11.90	5.23	18.97	5.90	12.73	5.24	5.88	10.34	7.63	5.99	8.17	**4.63**	6.23	5.38
165	11.00	2.99	11.00	27.12	20.55	23.61	22.81	6.83	8.87	3.05	11.00	4.44	7.17	12.22	3.76	6.11	3.37	11.88	6.81	11.67	2.63	**2.39**
166	12.12	10.81	12.13	11.60	13.21	13.16	12.32	11.66	13.14	10.75	12.07	11.35	11.00	11.77	11.20	11.25	11.39	11.74	11.57	**1.95**	10.41	9.51
167	2.03	1.16	2.03	4.78	10.81	10.33	10.21	2.04	5.47	1.17	2.03	1.98	2.00	3.15	1.18	2.03	1.69	3.21	1.85	13.85	1.19	**1.10**
168	15.85	10.71	15.85	13.11	19.36	21.44	18.88	15.76	12.21	10.85	15.84	13.89	10.13	13.04	13.71	11.31	20.50	14.27	11.93	**8.85**	9.19	10.18
169	33.62	8.64	33.62	24.01	44.64	10.93	43.77	24.97	14.08	8.38	33.61	8.21	18.09	12.49	9.19	16.36	31.38	12.45	13.76	**3.97**	7.67	7.32
170	5.27	4.27	5.27	4.58	6.78	5.34	5.71	3.94	3.37	3.05	5.20	3.23	4.59	4.44	3.24	3.66	4.25	4.04	5.28	13.60	2.75	**2.54**
171	17.81	16.03	17.81	16.36	17.30	15.33	18.09	17.80	15.17	14.30	17.95	14.04	16.81	16.50	13.60	14.42	14.14	17.84	14.40	**2.87**	13.86	11.27
172	4.29	4.03	4.29	5.65	11.05	6.61	11.37	3.78	3.32	3.79	4.30	4.05	3.72	5.63	**2.73**	2.91	3.02	6.25	3.76	3.40	3.09	2.84
173	3.95	3.49	3.95	5.48	5.73	8.03	4.51	3.31	3.38	3.66	3.95	3.68	4.03	3.36	3.42	3.27	6.57	3.32	4.10	4.48	3.42	**3.13**
174	8.95	4.59	8.95	4.87	12.15	11.51	10.61	7.54	6.25	4.52	8.94	4.49	5.23	4.71	3.83	5.16	15.15	5.43	5.58	30.38	3.78	**3.73**
175	57.18	19.60	57.18	38.28	44.13	22.26	38.11	42.74	38.04	17.97	56.83	29.75	51.81	36.26	25.26	29.66	**12.30**	31.40	55.77	35.29	19.03	19.18
176	66.45	27.04	66.46	37.02	42.47	29.87	43.70	55.15	36.81	26.64	65.86	33.86	39.76	31.61	24.71	22.91	19.81	28.48	48.31	**13.22**	21.04	17.40
177	24.89	15.83	24.89	17.44	24.38	9.53	23.10	23.16	7.47	15.82	24.84	15.74	14.51	16.31	10.73	14.05	28.99	17.27	15.17	**4.19**	11.29	9.58
178	11.82	3.41	11.82	4.12	7.23	9.59	5.62	7.53	4.59	**2.88**	11.80	3.93	7.23	3.88	4.26	5.41	19.62	3.69	6.95	26.29	3.15	2.99
179	41.24	30.40	41.24	45.08	51.88	30.97	53.76	30.36	30.19	28.59	41.25	39.95	31.23	41.83	23.80	20.32	32.06	44.29	31.38	**13.99**	14.18	17.33
180	17.49	13.54	17.49	17.69	16.20	44.08	15.61	14.39	14.34	12.11	17.44	13.12	13.55	15.54	12.64	13.63	22.67	15.13	14.82	14.36	14.24	**11.81**
181	20.47	19.84	20.46	26.37	22.66	40.32	25.47	20.39	16.85	20.38	20.51	19.07	19.47	22.69	14.48	17.55	14.91	25.80	19.08	**4.08**	11.60	14.44
182	4.09	3.05	4.09	10.98	11.23	15.91	11.89	3.95	4.55	3.10	4.09	4.19	3.95	9.00	3.82	3.80	3.89	9.26	6.04	7.44	4.91	**2.42**
183	12.17	6.58	12.17	9.13	5.85	14.53	5.72	10.44	10.34	6.45	12.17	8.34	11.15	5.86	7.14	9.94	8.70	**5.72**	8.52	11.19	6.57	6.48
184	12.01	9.50	12.01	12.62	10.72	14.08	11.11	12.09	11.43	9.06	12.00	11.26	11.96	10.72	11.09	12.12	11.10	11.08	12.02	**4.05**	9.28	8.87
185	6.80	4.05	6.80	5.52	8.62	12.88	8.00	6.22	3.25	3.91	6.79	3.92	4.31	4.89	3.02	4.63	14.08	5.45	4.76	5.12	3.65	**2.73**
186	23.81	3.59	23.81	17.02	35.83	**2.72**	35.32	15.27	5.69	4.05	23.80	5.55	12.72	5.77	5.11	11.99	20.26	5.82	3.65	7.00	3.42	3.77
187	8.65	3.71	8.65	9.73	3.86	18.77	5.24	9.12	7.69	**3.18**	8.68	7.20	8.89	3.97	6.92	8.84	6.36	5.03	9.83	3.57	3.82	3.44
188	7.72	2.75	7.72	16.63	11.35	16.40	12.71	5.40	7.69	2.69	7.71	3.55	6.05	9.07	3.43	5.01	2.74	9.25	4.31	3.61	**1.78**	1.95
189	5.19	3.61	5.19	3.64	6.72	5.77	6.45	4.31	3.70	3.61	5.19	3.64	3.86	3.64	**3.41**	3.83	6.15	3.69	3.87	9.59	3.63	3.67
190	11.80	6.57	11.80	12.76	6.34	23.06	9.13	11.31	9.38	6.38	11.71	9.73	11.11	6.37	8.98	11.29	7.62	8.27	12.95	8.41	**5.92**	5.98
191	13.38	12.01	13.37	13.50	13.79	7.19	14.80	11.84	20.04	8.39	13.53	9.84	18.79	13.36	7.61	11.41	9.88	14.19	13.75	9.22	9.68	**5.35**
192	9.74	8.44	9.75	10.83	8.54	12.75	9.32	9.70	12.69	7.42	9.52	8.84	9.19	8.55	6.74	9.46	13.11	8.42	10.97	19.76	6.70	**6.40**
193	24.44	12.03	24.44	74.07	48.02	152.92	54.07	19.17	27.74	12.24	24.47	19.67	22.12	31.72	20.54	19.43	18.01	35.35	18.24	78.67	12.24	**10.86**
194	183.64	33.67	183.64	96.44	113.46	32.24	114.62	170.72	22.28	39.08	183.64	79.50	132.34	78.27	41.57	132.97	297.07	78.44	86.67	46.87	**14.28**	28.91
195	159.80	21.42	159.80	80.71	97.17	18.82	96.91	145.81	21.72	24.47	159.80	55.34	103.14	55.58	21.28	103.87	236.23	55.86	54.32	33.44	**13.39**	16.15
196	34.91	31.72	34.91	54.25	62.02	23.77	77.21	33.75	23.15	30.81	34.75	35.01	30.01	49.45	23.50	25.11	28.38	53.18	43.96	**3.37**	24.35	20.97
197	3.60	2.90	3.60	5.52	8.38	8.33	8.81	3.59	10.54	2.30	3.59	2.74	2.03	5.01	2.83	2.99	**1.59**	5.41	2.90	4.61	3.63	2.13
198	6.75	4.48	6.75	6.04	8.98	3.47	8.22	6.18	4.41	3.75	6.75	4.43	4.87	3.50	4.43	3.83	4.08	3.54	3.28	5.26	**3.17**	3.21
199	5.33	5.34	5.33	5.39	5.88	4.55	5.71	5.27	4.19	5.32	5.33	5.33	5.44	5.36	4.24	4.82	4.71	5.53	5.09	23.63	4.61	**3.28**
200	27.21	23.02	27.21	26.99	28.23	**12.17**	28.24	27.23	22.89	13.88	27.24	14.59	23.31	24.52	14.50	22.41	21.17	25.55	22.11	18.35	22.88	12.94
201	18.54	18.36	18.54	20.19	19.67	30.84	19.68	18.55	17.30	12.75	18.51	13.15	18.10	18.90	17.93	18.22	14.33	18.71	18.15	**3.70**	18.23	12.73
202	5.41	4.53	5.41	6.12	6.66	11.23	6.57	5.40	7.91	4.48	5.42	4.71	5.10	4.50	3.70	4.31	3.68	5.14	4.81	5.34	3.60	**3.58**
203	7.18	5.83	7.18	6.48	8.95	12.15	7.06	5.37	**3.56**	5.82	7.19	5.88	6.83	5.78	4.71	5.17	4.79	5.44	6.05	9.44	5.15	3.94

204	11.30	8.95	11.30	18.87	13.91	23.19	15.10	10.73	12.17	8.86	11.30	9.10	10.40	13.83	9.07	10.01	9.47	14.68	11.81	**5.64**	9.22	9.28
205	5.75	5.39	5.75	8.85	6.20	11.36	5.76	5.78	6.71	5.81	5.82	5.86	6.10	5.87	5.35	5.40	5.27	5.74	6.25	**5.03**	5.60	5.27
206	6.80	4.63	6.80	8.28	4.38	5.01	5.12	5.15	**2.56**	4.52	6.81	4.77	5.82	4.06	3.43	5.29	4.37	4.24	6.47	44.53	4.76	3.44
207	50.21	49.86	50.21	51.16	51.58	10.34	53.05	47.57	55.91	10.44	50.63	10.15	53.50	51.16	7.61	44.97	12.40	52.40	44.25	**5.96**	44.55	7.86
208	13.85	9.40	13.85	9.31	18.22	26.14	19.09	13.33	8.26	9.40	13.87	8.73	9.99	9.30	**4.90**	8.40	6.27	10.86	7.94	10.98	5.61	5.96
209	12.81	11.03	12.80	13.38	12.90	**3.40**	13.68	10.93	13.44	3.73	13.04	4.51	14.15	12.52	4.56	10.63	4.52	13.72	7.75	5.48	11.92	3.77
210	6.71	5.96	6.71	9.05	10.62	13.03	10.14	5.45	5.32	5.95	6.71	6.56	6.54	8.12	5.28	5.45	6.27	7.90	7.26	**0.83**	5.77	4.24
Min.	0.93	0.93	0.93	1.29	2.29	0.64	2.17	0.69	1.15	0.70	0.92	0.70	1.39	1.29	0.32	0.74	0.39	1.41	0.88	0.59	0.60	0.17
Max.	183.64	86.65	183.64	714.91	630.01	454.94	855.59	170.72	106.46	87.74	183.64	93.44	132.34	356.62	102.86	132.97	297.07	669.08	119.79	116.94	87.27	81.99
Overall AARD	18.80	11.94	18.80	22.27	23.74	19.94	25.79	16.96	12.63	10.87	18.79	12.69	15.02	16.11	10.67	13.52	15.37	18.90	14.53	12.56	10.39	**8.88**

$$Z^{SCF} = Z(T, P, y_2) \tag{6}$$

Eqs. 5 and 6 are the basis for calculating the solubility of a solid solute in an SCF by an EoS. The quality of predictions or correlations depends on the EoS and mixing rules to be used. Calculations are very sensitive to the accuracy of the required solute properties [3,4,38].

Taking the logarithm of Eq. 4 gives

$$\ln y_2 = \ln P_2^{sub} - \ln P - \ln \phi_2^{SCF} + \frac{V_2^s}{R}\frac{P}{T} - \frac{V_2^s P_2^{sub}}{R}\frac{1}{T} \tag{7}$$

The volumetric properties of the solvent are governed by the equation of state of a real gas:

$$P = ZRT\rho_1 \tag{8}$$

The solubility of the solute at a given T and P can be related to the density and the coefficient of compressibility of the pure solvent by replacing the pressure in Eq. 7:

$$\ln y_2 = \ln P_2^{sub} - \ln(ZRT\rho_1) - \ln \phi_2^{SCF} + V_2^s Z\rho_1 - \frac{V_2^s P_2^{sub}}{R}\frac{1}{T} \tag{9}$$

All of the density-based models that have been proposed can be considered as a simple empirical approximation to the right-hand side of Eq. 9, which generally does not require any solute property. The next section presents a brief review of density-based model for correlation of solid solubility data.

2.1. Density-based model for correlation of solid solubility data

In a recent article [57], we have presented an extensive review of density-based empirical models. Chronologies of their development, the experimental findings, and theoretical explanations of some of the model parameters have been presented. Table 1 shows semiempirical models used to correlate the experimental solubility data that have been proposed by different authors in the last four decades with the aim of improving the quality of correlation of new experimental data. Models that require solute properties, which are not readily available for compounds of interest such as pharmaceuticals and dyes, have not been considered in this study. In Table 1, y_2 is the solubility of the solid solute in mole fraction, ρ_1 is the density of the SCF (in kilograms per cubic meter), T is the temperature (in Kelvin), P is the pressure (in bar), a_i represents the characteristic parameters determined by fitting experimental data of the solute/solvent considered. For the Sparks et al. [73] model, solubility is expressed in a dimensionless form, whereas in the Bain et al. [64] model the solute solubility, c_2, is in kilograms per cubic meter. In both models c_2 is related to y_2 by Eq. 24a, in which Mw_1 and Mw_2 are the molecular weights of the solvent and the solute, respectively. It is worth mentioning that some of the models do not relate the solubility explicitly to the density of the SCF solvent. The model proposed in our previous work [57] is based on the following facts: (1) models that relate the solubility of the solid solute to temperature, pressure, and the density of the SCF violate the phase rule, because the volumetric properties (P, T, and

ρ_1) of $scCO_2$ are governed by the phase rule, hence only two variables need to be fixed; (2) just as the density expansion of the virial EoS is more accurate than the pressure expansion at increased pressures [80], relating the solubility to density at isothermal condition instead of pressure would lead, a priori, to better correlation.

2.2. Proposed model

On the basis of the two arguments mentioned by Si-Moussa et al. [57], the proposed model is derived first by a modification of Gordillo et al. [70] model where the pressure is replaced by the density of the solvent, which leads to the following equation:

$$\ln y_2 = a_0 + a_1\rho_1 + a_2\rho_1^2 + a_3\rho_1 T + a_4 T + a_5 T^2 \tag{31}$$

As the linear relationship between $\ln(y_2)$ and $\ln \rho_1$ in isothermal conditions, as well as the inverse of temperature effect on $\ln(y_2)$, has been considered in many models that demonstrated acceptable correlation performance in previous comparative studies [57–64], these two terms ($a_6 \ln \rho_1 + a_7/T$) are added to Eq. 31. The proposed model is therefore

$$\ln y_2 = a_0 + a_1\rho_1 + a_2\rho_1^2 + a_3\rho_1 T + a_4 T + a_5 T^2 + a_6 \ln \rho_1 + a_7/T \tag{32}$$

The eight parameters a_0–a_7 are obtained from regression of experimental data.

3. Results and discussion

In the present study, a data set of 210 solid solutes counting 5550 data points of experimental solubility in $scCO_2$, compiled from the literature published in the last 25 years, is used for comparison of correlation performance of 22 empirical models. Table 2 displays the range of experimental temperature, pressure, solubility, N_i number of experimental points, and source of the 210 selected binary solute–$scCO_2$ systems.

The optimization procedure, for fitting the model parameters, reduces the percentage average absolute relative deviation (AARD %) and is defined as

$$\mathrm{AARD}(\%) = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{y_2^{\mathrm{exp}} - y_2^{\mathrm{cal}}}{y_2^{\mathrm{exp}}}\right| \tag{33}$$

Initial approximations of model parameters are first obtained by genetic algorithm function (ga MATLAB function) through several runs to avoid convergence to local minimum. These approximations are then used as initial guess for least-square fitting with Levenberg–Marquardt algorithm as minimization option (lsqcurvefit MATLAB function).

Table 3 shows the results of parameter estimation (a_i) of the model proposed in this work (Eq. 32) for the 210 systems considered. Parameters of the other models can be accessed in the Supplementary material.

The AARDs (%) of each model equation with each solid compound are shown in Table 4 where the minimal AARD for the correlation of the solubility data of each binary system is shown in bold. It is clear that empirical models, considered in this study, as a whole correlate quite satisfactorily the data set with a minimal AARD (0.17%) obtained by the proposed model (Eq. 32) for component 45 (azobenzene) and a maximum AARD (56.47%) obtained by Jouyban et al. model (Eq. 18) for component 77 (corosolic acid). The average of minimal AARDs is 6.87%. Therefore, 0.17% represents the minimum of minimums and 56.47% represents the maximum of minimums and 6.87% represents the average of the minimums for the data set considered. It is worth mentioning that the value of 56.77% was obtained for corosolic acid for which the experimental data present presumably a misprint, which results in excessive AARD.

The new model was compared with the 21 most commonly used semiempirical models in terms of AARD (%) and the results are presented in Table 5, in which bold values represent the best performance for a group of models with the same number of adjustable parameters. Among the three parameter models, Garlapati and Madras [74] (Eq. 20) and Chrastil [65] (Eq. 10) models were found to correlate better with an average AARD of 18.79%, whereas Jafari Nedjad et al. [76] model (Eq. 22) was found to be better among four parameter models with an average AARD of 15.02%. Comparing five parameter models, Adachi and Lu [66] correlation (Eq. 11) gave the best global AARD of 11.94%. For the set of six parameter models, Si-Moussa et al. [57] model (Eq. 30) shows a slight advantage over Sparks et al. [73] (Eq. 19) and Bian et al. [59] (Eq. 24) models and a

Table 5
Comparison of overall AARD according to the number of model parameters.

Number of parameters	Model equation	Overall AARD %	Number of parameters	Model equation	Overall AARD %
3	(10)	18.80	5	(21)	12.69
3	(13)	22.27	5	(25)	13.52
3	(14)	23.74	5	(28)	14.53
3	(16)	25.79	5	(29)	12.56
3	(20)	**18.79**	6	(15)	19.94
4	(12)	18.80	6	(18)	12.63
4	(17)	16.96	6	(19)	10.87
4	(22)	**15.02**	6	(24)	10.67
4	(23)	16.11	6	(30)	**10.39**
4	(27)	18.90	8	(32)	**8.88**
5	(11)	**11.94**	9	(26)	15.37

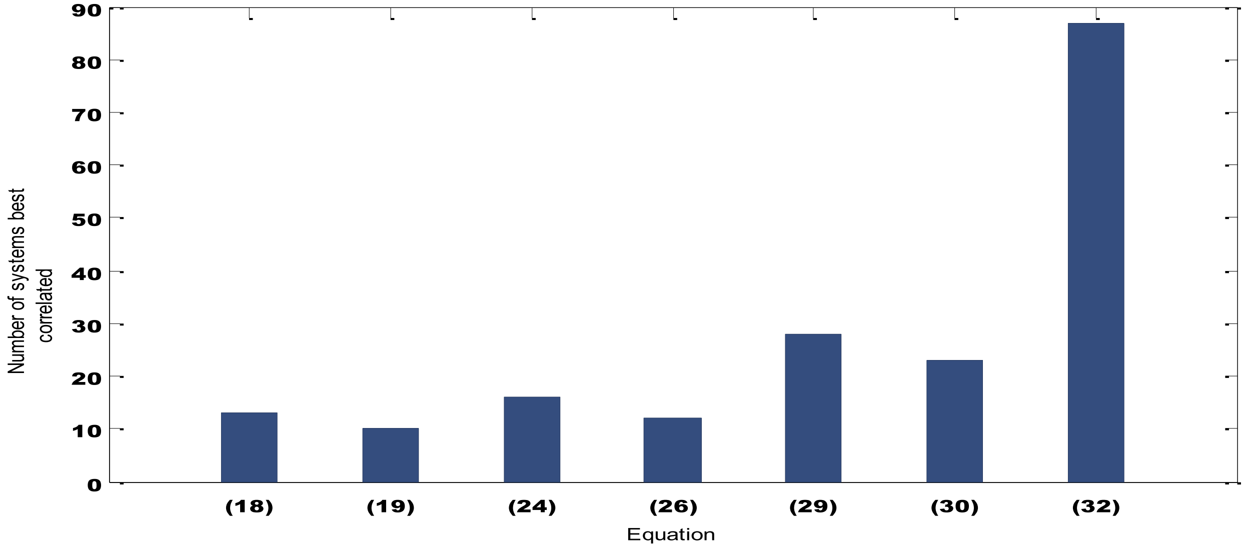

Fig. 1. The number of systems best correlated by each model equation.

clear superiority over Jouyban et al. [58] (Eq. 18) and Gordillo et al. [70] (Eq. 15) models with an average AARD of 10.39%. To summarize, it is clear from Table 5 that the more parameters the model has the more accurate correlation performance are, except the nine-parameter model of Amooey [60] (Eq. 26) with a global AARD of 15.37%. The eight-parameter model proposed in this work shows the smallest AARD (8.88%) among all of the models considered. These findings are further illustrated in Fig. 1, which represent the correlations that best correlated 90% of systems considered. Clearly, the model proposed in this work is superior to the other models as it best correlates 87 (41%) systems of the 210 considered. The second best is the five-parameter model of Bian et al. [64] (Eq. 29) with 28 (13%) systems.

It should be mentioned that models that include the pressure as variable tend to give a relatively poor correlation. This is well demonstrated by the correlation results of the same data set of Jouyban et al. [58] model (Eq. 18) (12.63%) compared to Si-Moussa et al. [57] model (Eq. 30) (AARD = 10.39%). A more illustrative example, which supports this evidence, is the comparison of Gordillo et al. [70] (Eq. 15) and its modification (Eq. 31), not included in the present study, where there is a net difference between the correlation performance of the two equations (AARD of 19.94% and 11.09%, respectively).

Fig. 2 displays the scatter plot of the entire data set of the solid solute solubility calculated using the model proposed in this work (Eq. 32) versus experimental solubility (5550 experimental points). These plots are generated using the *postreg* function of MATLAB, trace the calculated solubility as a function of experimental solubility.

In Fig. 2, the first bisector shows the exact fit between the correlated solubilities and experimental data, whereas the cross points demonstrated the real correlated solubility data by the proposed correlation versus experimental data. In detail, the closer the points to the solid line, the more accurate correlated solubility data means. On the basis of the obtained results, it can be concluded that the proposed correlation was able to correlate the solubility of solids in $scCO_2$ with an acceptable deviation.

Figs. 3 and 4 show comparisons between calculated and experimental solubility for the best (menadione) and worst (corosolic acid) correlation results.

4. Conclusions

The comparative study of semiempirical models on a fairly large database carried out in this work shows that the

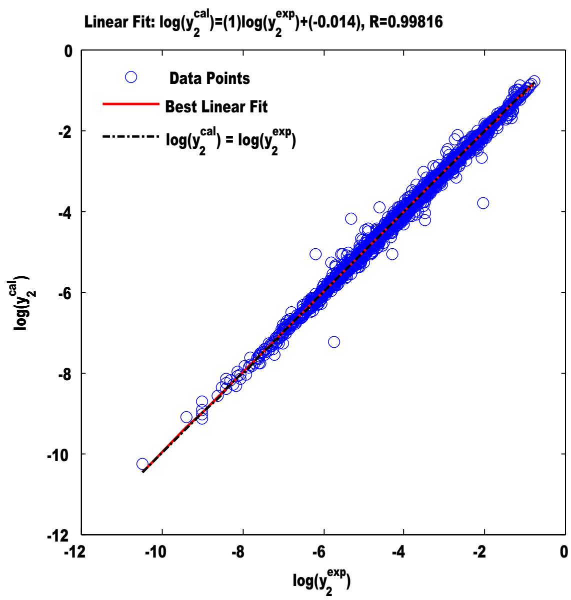

Fig. 2. Scatter plot of the entire dataset of solid solutes solubility calculated using the model proposed in this work (Eq. 32) versus experimental solubility.

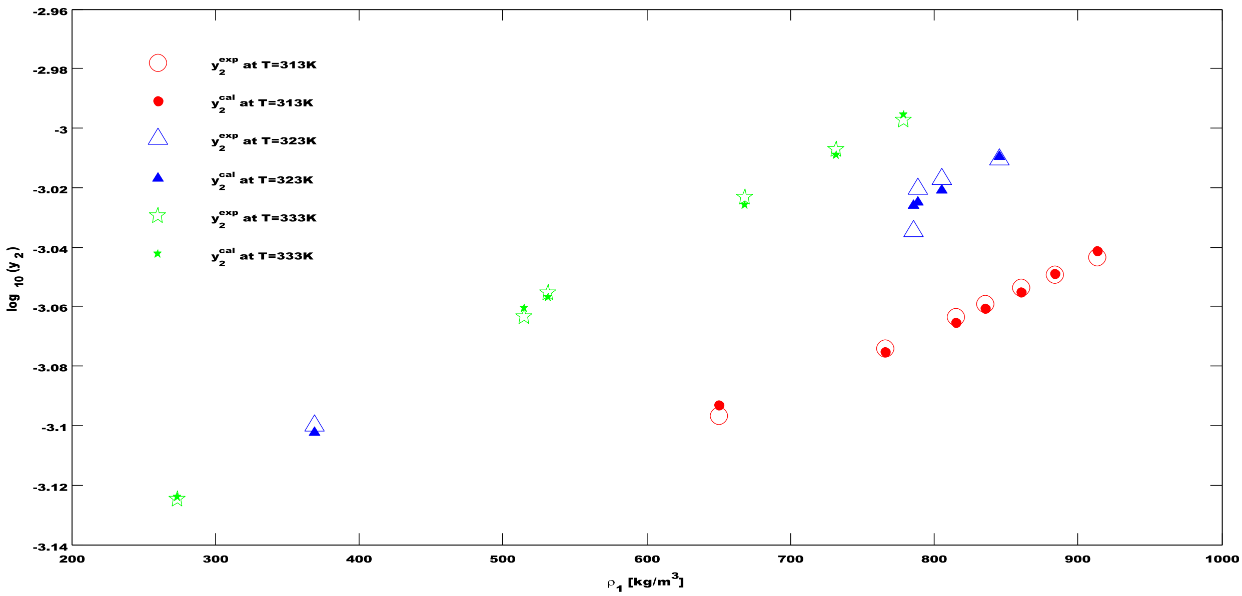

Fig. 3. Mole fraction solubility ($\log_{10}(y_2)$) of menadione in $scCO_2$ as a function of carbon dioxide density and temperature.

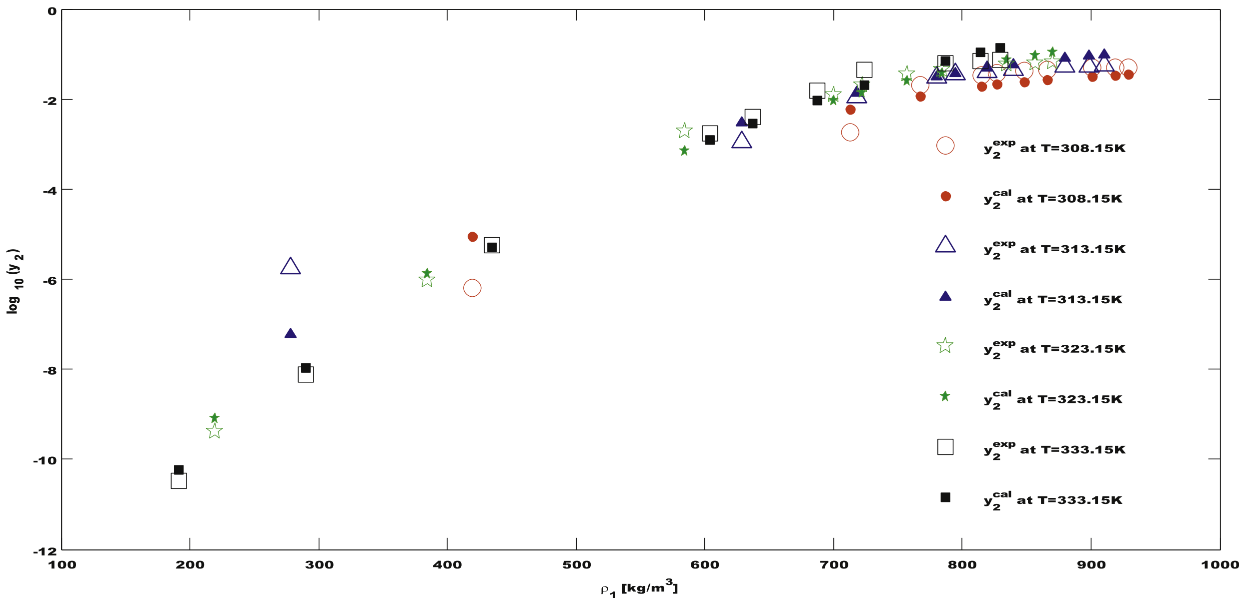

Fig. 4. Mole fraction solubility ($\log_{10}(y_2)$) of corosolic acid in $scCO_2$ as a function of carbon dioxide density and temperature.

solubility data of solid solutes in $scCO_2$ are better correlated by models that relate the logarithm of the solubility in the mole fraction to the density of solvent and temperature with more than five adjustable parameters. The model must include a term of the logarithm of the density, a polynomial of the density, and a term that accounts for the effect of temperature. As to the data set considered, semiempirical models correlate quite satisfactorily the data within the AARD range of 0.17–56.47% and an average value of 6.87%. However, when comparing the global correlating performance of each model, the proposed model has by far the best correlating performance with an AARD lying in the range of 0.17–81.99% and an average value of 8.88%. The second best correlation is (Eq. 30) [30] where the AARD ranges in 0.60–87.27% and an average value of 10.39%.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.crci.2018.02.006.

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