# Table 1. Basic properties of the used materials.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Compound | Formula | MW (g/mol) | Tm (K)  | λmax(nm) |  CAS number | Minimum purityBy supplier |
| Clemastine Fumarate | C21H26ClNO.C4H4O4 | 459.96 | 451.15 | 270 | 14976-57-9 | 99% |
| Carbon dioxide | CO2 | 44.01 |  |  | 124-38-9 | 99.99% |
| Methanol | CH3OH | 32.04 |  |  | 67-56-1 | 99.9% |

# Table 2. Solubility of *Clemastine Fumarate* inScCO2 at various temperatures and pressures

#  (The experimentalstandard deviation was obtained by . Expanded uncertainty (U)=*k\*ucombined* and the relative combined standard uncertainty *ucombined/y*=

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Temperature (K)a | Pressure (MPa)a | Density of SC-CO2 (kg/m3) [2] | y2 × 104(Mole fraction) | Experimental standard deviation , S(ȳ)× (104) | S (Equilibrium solubility) (g/L) | Expanded uncertainty of Mole fraction (104U) |
| 308 | 12 | 769 | 0.0161 | 0.0005 | 0.0130 | 0.0012 |
| 15 | 817 | 0.0202 | 0.0010 | 0.0173 | 0.0022 |
| 18 | 849 | 0.0247 | 0.0010 | 0.0219 | 0.0023 |
| 21 | 875 | 0.0284 | 0.0008 | 0.0260 | 0.0020 |
| 24 | 896 | 0.0384 | 0.0002 | 0.0360 | 0.0017 |
| 27 | 914 | 0.051 | 0.0010 | 0.0488 | 0.0030 |
| 318 | 12 | 661 | 0.0248 | 0.0006 | 0.0171 | 0.0017 |
| 15 | 744 | 0.0395 | 0.0005 | 0.0307 | 0.0021 |
| 18 | 791 | 0.0431 | 0.0020 | 0.0357 | 0.0044 |
| 21 | 824 | 0.0513 | 0.0020 | 0.0442 | 0.0046 |
| 24 | 851 | 0.0599 | 0.0009 | 0.0532 | 0.0032 |
| 27 | 872 | 0.0697 | 0.0020 | 0.0636 | 0.0050 |
| 328 | 12 | 509 | 0.0282 | 0.0010 | 0.0150 | 0.0024 |
| 15 | 656 | 0.0414 | 0.0008 | 0.0284 | 0.0025 |
| 18 | 725 | 0.0471 | 0.0020 | 0.0357 | 0.0045 |
| 21 | 769 | 0.0558 | 0.0010 | 0.0449 | 0.0032 |
| 24 | 802 | 0.0778 | 0.0030 | 0.0652 | 0.0069 |
| 27 | 829 | 0.0886 | 0.0040 | 0.0767 | 0.0089 |
| 338 | 12 | 388 | 0.0359 | 0.0010 | 0.0145 | 0.0026 |
| 15 | 557 | 0.046 | 0.0020 | 0.0268 | 0.0045 |
| 18 | 652 | 0.0515 | 0.0007 | 0.0351 | 0.0027 |
| 21 | 710 | 0.0593 | 0.0010 | 0.0440 | 0.0033 |
| 24 | 751 | 0.086 | 0.0040 | 0.0676 | 0.0087 |
| 27 | 783 | 0.0941 | 0.0030 | 0.0771 | 0.0073 |

aStandard uncertainty u are u(T) = ±0.1 K; u(p) = ± 0.1 MPa. The value of the coverage factor k=2 was chosen on the basis of the level of confidence of approximately 95 percent.

**Table 3.** Properties of *Clemastine fumarate* and CO2a.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Substance** | **Tc(K)** | **Pc(MPa)** |  | **Vs**× **10-4 (m3/mol)** | **T(K)** |
|  |  |  |  |  | **Psub (Pa)** f |
|  |  |  |  |  | 308 | 318 | 328 | 338 |
| Clemastine Fumarate | 901.25b | 1.409c | 0.337d | 364.764e | 0.0114 | 0.02699 | 0.0603 | 0.1277 |
| CO2 | 304.18 | 7.38 | 0.225 |  |  |  |  |  |

a Critical temperature: Tc; Critical pressure: Pc; Acentric factor : ; Solid molar volume: Vs; Temperature:T.

bEstimated by Fedors method51,52.

c Estimated by the Joback modification of Lydersen’s method52.

d Estimated by Lee-Keslervapour pressure relations. (Note: The required normal boiling temperature (at 1.0 atm), Tb is estimated with Klincewicz relation, Tc =50.2-0.16M + 1.41Tb were M is molecular weight)52.

e Estimated by Immirzi, A.; Perini, B method52,53.

f Estimated by Lee-Keslervapour method52.

**Table 4.** Correlation constants for the exiting empirical models.

|  |  |  |  |
| --- | --- | --- | --- |
| Model | Correlation Parameters | AARD% | R2 |
| Alwi-Garlapati model | =1.8759; =-18.105; =2.0667 | 14.00 | 0.809 |
| Bartel et al., model | =14.719; =-7179.9; =6.6739×10-3 | 20.2 | 0.765 |
| Bian et al., model | = -4.9614; =5.5723×10-3; =2030.5; =-9.9873 ; =9.6915  | 11.2 | 0.927 |
| Chrastil model | =3.0938; = -11.003; = -4907.2 | 16.7 | 0.785 |
| Ref. Chrastil model | =3.0813; = -21.619; = -4216.6 | 16.7 | 0.784 |
| Garlapati-Madras model | =-755.12; =859.01; =0.9875; =-8597.4; =-10.722 | 14.6 | 0.818 |
| Mendez-Teja model | =-8479.4; =1.9629; =14.617 | 21.69 | 0.706 |
| Sodeifian et al., model | =-42.487; =-6.9315×10-4; =2.4265; =-4.2127×10-4; =1.929×10-2; =62.052 | 8.78 | 0.929 |
|  |  |  |  |
| Tippana-Garlapati model | =8.334×10-7; =1.3157×10-5; =-3.3583×10-7; =5.6805×10-7; =-1.3913×10-5; =7.7736×10-7 | 7.57 | 0.951 |
| Mahesh-Garlapati model | =-14.614; =-2.4145; =3.3127 | 17.9 | 0.797 |

**Table 5**. Correlation constants of PREoS+VdW2 combination.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Model | Correlation parameters | T=308K | T=318K | T=328K | T=338K |
|  |  | Temperature Dependent Parameters |
| PREoS-VdW2Temperature Dependent Parameters |  | 0.58814 | 0.55098 | 0.55315 | 0.53741 |
|  | 0.5856 | 0.52813 | 0.52218 | 0.48034 |
|  | AARD% | 3.99 | 2.5572 | 7.5542 | 13.067 |
|  |  | Temperature Independent Parameters |
| PREoS-VdW2 Temperature Independent Parameters |  | 0.79788 |  |  |  |
|  | 0.74029 |  |  |  |
|  | 0.27409 |  |  |  |
|  | -221.54 |  |  |  |
|  | 11.305 |  |  |  |
| AARD% | 8.2458 |  |  |  |

**Table 6.** Correlation constants of cluster models.

|  |  |  |  |
| --- | --- | --- | --- |
| Model | Correlation Parameters | AARD% | R2 |
| New model | =0.10756; = 443590; = 1357.1;= -9115.7 | 10.3 | 0.936 |
| K.-W. Chen et al., model | = 0.10794; = 6093.7; = -70.319 | 12.1 | 0.913 |

**Table 7**. Summary of thermodynamic properties.

|  |  |
| --- | --- |
| Model | Property |
|  | Total Enthalpy, ΔHtotal(kJ/mol) | Enthalpy of SublimationΔHsub(kJ/mol) | Enthalpy of Solvation,(kJ/mol) |
| Chrastil model | 40.798a |  | -18.896e; -12.282f |
| Reformulated Chrastil Model | 35.056b |  | -24.638g-6.54h |
| Bartle et al., model |  | 59.694c(approximate value) |  |
| PREoS +vdW2 modelAs temperature independent  |  | 28.516d(average value) |  |

eObtained as a result of difference between the ΔHsubc and ΔHtotala

fObtained as a result between the ΔHsubd and ΔHtotala

gObtained as a result of difference between the ΔHsubc and ΔHtotalb

hObtained as a result between the ΔHsubd and ΔHtotalb

**Table 8.** Computed AIC and AICc values.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Model | SSE.1011 | Np | N | AIC | AICc |
| **Existing density models** |  |  |  |  |  |
| Alwi-Garlapati model | 2.41888 | 3 | 24 | -636.5 | -635.30 |
| Bartel et al., model | 3.29465 | 3 | 24 | -649.54 | -648.34 |
| Bian et al., model | 94.47 | 5 | 24 | -675.52 | -672.19 |
| Chrastil model | 2.69392 | 3 | 24 | -654.37 | -653.17 |
| Reformulated Chrastil model | 2.70787 | 3 | 24 | -654.25 | -653.05 |
| Garlapati-Madras model | 2.36986 | 5 | 24 | -653.45 | -650.11 |
| Mendez-Teja model | 3.2662 | 3 | 24 | -649.75 | -648.55 |
| Sodefian et al., model | 1.03868 | 6 | 24 | -671.24 | -666.30 |
| Tippana-Garlapati model | 61.5158 | 6 | 24 | -683.82 | -678.88 |
| Mahesh\_Garlapati model | 2.64529 | 3 | 24 | -654.81 | -653.61 |
| **Cluster models** |  |  |  |  |  |
| New cluster model | 82.7593 | 4 | 24 | -680.70 | -678.59 |
| K.-W. Chen et al., model | 1.10632 | 3 | 24 | -675.73 | -674.53 |
| **EoS Model** |  |  |  |  |  |
| PREoS model +vdW2 Mixing Rule | 84.4568 | 5 | 24 | -678.21 | -674.88 |