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Measurements of Binary Diffusion Coefficients in Supercritical Carbon Dioxide

Chang Yi Kong*

Graduate School of Engineering and Research Institute of Green Science and Technology, Shizuoka University, 3-5-1 Johoku, Naka-ku, Hamamatsu, 4328561, Japan

A supercritical fluid (SCF) is an adjustable solvent which is reached at a temperature and pressure higher than its critical point. It can diffuse through solids like a gas, and dissolve materials like a liquid. In addition, close to the critical point, small changes in pressure or temperature result in large changes in density or viscosity, allowing many properties to be tuned. SCFs are suitable as a substitute for organic solvents in a range of industrial processes. Furthermore, there is no surface tension in a SCF, as above the critical temperature there is no phase transition. The applications of SCF is still expanding in the fields of extraction [1,2], food [3], pharmaceutical [4,5], material [6-8] and chemical reaction [9]. The quantitative knowledge of mass transport phenomena such as diffusion coefficients of various compounds in SCF is of considerable important in the design and efficient operation of the newly proposed SCF processes. Experimental methods, compounds and SCFs employed to determine the diffusion coefficients are highlighted in this editorial.

Diffusion is caused by random molecular motion that leads to complete mixing. In chromatography, the diffusion coefficient refers to the diffusion of a solute in a relatively or very dilute solution. So far, a number of experimental methods such as solid dissolution (SD) [10,11], pseudo steady-state solid dissolution (PSTD) [12], photon correlation spectroscopy (PCS) [13], radioactive tracer response (RTR) [14,15], nuclear magnetic resonance (NMR) [16,17], Taylor-Aris dispersion [18,19], modified Taylor-Aris dispersion [20,21] and chromatographic impulse response (CIR) [22,23] have been developed for measuring binary diffusion coefficients in SCFs. Among the above methods, most literature data [24-26] on binary diffusion coefficients in SCFs have been made by the Taylor-Aris dispersion and the CIR methods. The latter involves chromatographic adsorption, and the former does not.

Supercritical (SC) CO_2 is known to be the most stable and an excellent solvent and is normally used in mobile phases for supercritical fluid chromatography (SFC) as well as the applications of SCF over a wide range of fields. The Taylor-Aris dispersion method is adequate for measuring binary diffusion coefficients for lower molecular weight (MW), non and weakly polar compounds in SC CO_2. So far, most of compounds used to measure the binary diffusion coefficients by the Taylor-Aris dispersion method in SC CO_2 have lower MW than 300 and have non or weak polarity.

On the other hand, the modified Taylor-Aris dispersion and the CIR methods can be used to measure polar compounds or higher MW compounds higher than 400 in SC CO_2. In fact, Dahmen et al [21] measured the binary diffusion coefficients of Squalene (MW=410.7) in SC CO_2 by the modified Taylor-Aris dispersion method and Funazukuri et al. [20] also reported those of α-tocopherol (MW=430.7), β-carotene (MW=536.9) and ubiquinone CoQ10 (MW=863.3) in SC CO_2 using by the same method. The CIR method has been employed to measure the binary diffusion coefficients of polar compounds [27] such as benzoic acid, ethanol, methanol, 1-propanol and 2-propanol, and of metal complexes [28-30] such as ferrocene, 1,1'-dimethylferrocene, palladium(II) acetylacetonate, cobalt(III) acetylacetonate and platinum(II) acetylacetonate in SC CO_2. Recently, the binary diffusion coefficients for a number of compounds [31-35] with higher MWs such as β-carotene (MW=536.9), dibenzo-24-crown-8 (MW=448.5), diolein (MW=622.0), α-tocopherol (MW=430.7), triarachidonin (MW=951.5), trierucin (MW=1053.8), triolein (MW=885.4), ubiquinone CoQ10 (MW=863.3) and vitamin K_1 (MW=452.7) have been measured by the CIR method in SC CO_2. Kong group [34] have first measured the binary diffusion coefficients for the highest MW compound of trinervonin with MW of 1137.9 so far. The CIR method is superior to the Taylor dispersion method in diffusion measurements in SC CO_2, especially in the vicinity of the critical point, and for polar or high MW compounds. Although many studies have been paid to the measurements of diffusion coefficients in SC CO_2, there are still few works on the diffusion coefficients for polar compounds, drug compounds, metal complexes, and in the regions near to critical point, at higher temperatures (>100°C) and higher pressures (>40 MPa) region in SC CO_2. In addition, the solvents such as 2,3-dimethylbutane [36], chlorotrifluoromethane, [37], ethane [38-40], ethanol [41], hexane [40-42], propane, [40-43] 2-propanol [44] and water [45] are also used as SCFs, in which the binary diffusion coefficients have been measured for various organic compounds with lower MWs such as 1,3,5-trimethylbenzene, 1-octene, 1-tetradecene, acetone, benzene, m-cresol, naphthalene, n-decane, n-tetradecane, phenanthrene, phenol, p-xylene and toluene.

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***Corresponding author:** Chang Yi Kong, Graduate School of Engineering and Research Institute of Green Science and Technology, Shizuoka University, 3-5-1 Johoku, Naka-ku Hamamatsu 432-8561 Japan; Tel: 053478-1174 E-mail: tcykong@ipc.shizuoka.ac.jp

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