Small-Angle X-ray Scattering Measurement of Ionic Liquid-CO₂ Systems

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1 Introduction

Despite being salts, ionic liquids (ILs) that show liquid phase at room temperature have high CO₂ solubility and selectivity compared with gases included in the exhaust gas such as N₂, O₂, and H₂. Fluctuations, inhomogeneity of molecular distributions of the systems, provide mesoscopic structural information and are estimated by small-angle scattering methods for X-rays and neutrons. Containing heavy atoms such as fluorine, sulfur, some ionic liquids have higher X-ray absorbing power than some molecular liquids. Furthermore, in order to perform the measurement of IL-CO₂ systems at phase equilibrium without agitation, shorter exposure time is needed, which requires much higher intensity of X-ray source to perform scattering experiment.

2 Experiment

A high-pressure sample holder made entirely of titanium and a titanium alloy was designed to vary the path length precisely and perform stable operation at high temperature and pressure, while retaining a constant path length[1]. Imidazolium-based ionic liquid, 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide [C₂mim][NTf₂] was used as a sample. The measurement for IL-CO₂ mixtures was carried out at 314 K with pressures of 0.1, 1, 5, 10, 15, and 20 MPa under the process of pressure application using the apparatus at the BL-6A. Also, using samples which has different alkyl-chain length ([Cₙmim][NTf₂] ( n = 2, 4, 6, 8 )) at ambient condition, operation was performed.

An X-ray beam was monochromatized to λ = 1.50 Å and the observable s-region was 0.02 Å⁻¹ ~ 0.20 Å⁻¹, where the scattering parameter, s, is defined as 4πsinθ/λ (2θ: scattering angle, λ: wavelength). I(0), scattering intensities at s = 0, directly relate to the mesoscale structural fluctuation, are evaluated from the obtained SAXS intensities.

3 Results and Discussion

Fig.2 and 3 shows the intensities of [C₂mim][NTf₂]-CO₂ system and of [Cₙmim][NTf₂], respectively. I(0), shown in Fig.4, was obtained from the extrapolation of the each plot. At the region over 10 MPa, I(0) got two or three times as I(0) under 10 MPa, implying the structural change of IL at higher pressure region. And as alkyl-chain becomes longer, I(0) at ambient condition showed higher value.

References
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