

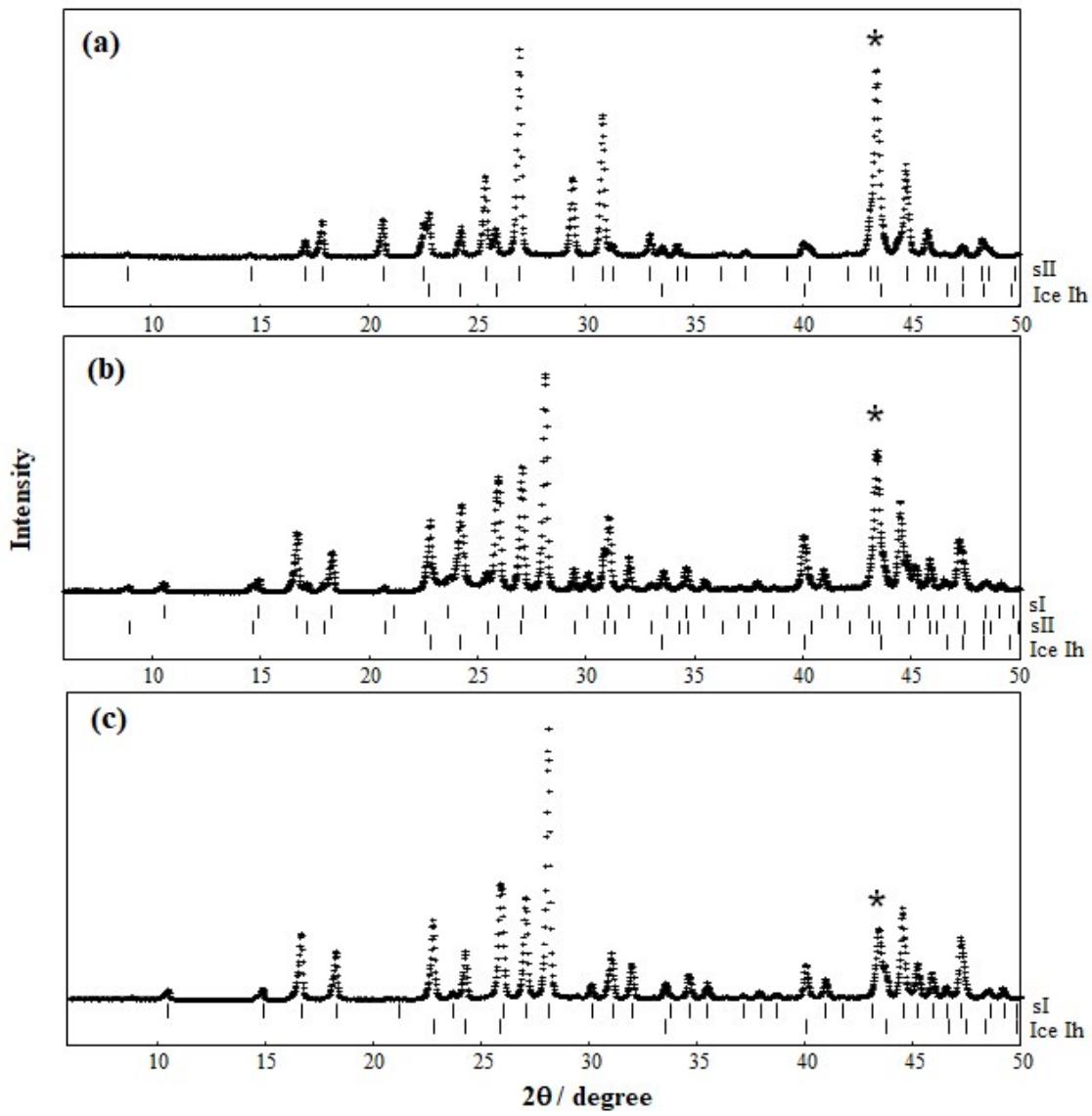
## Supporting Information

### Dissociation kinetics of Propane-Methane and Butane-Methane hydrates below the melting point of ice

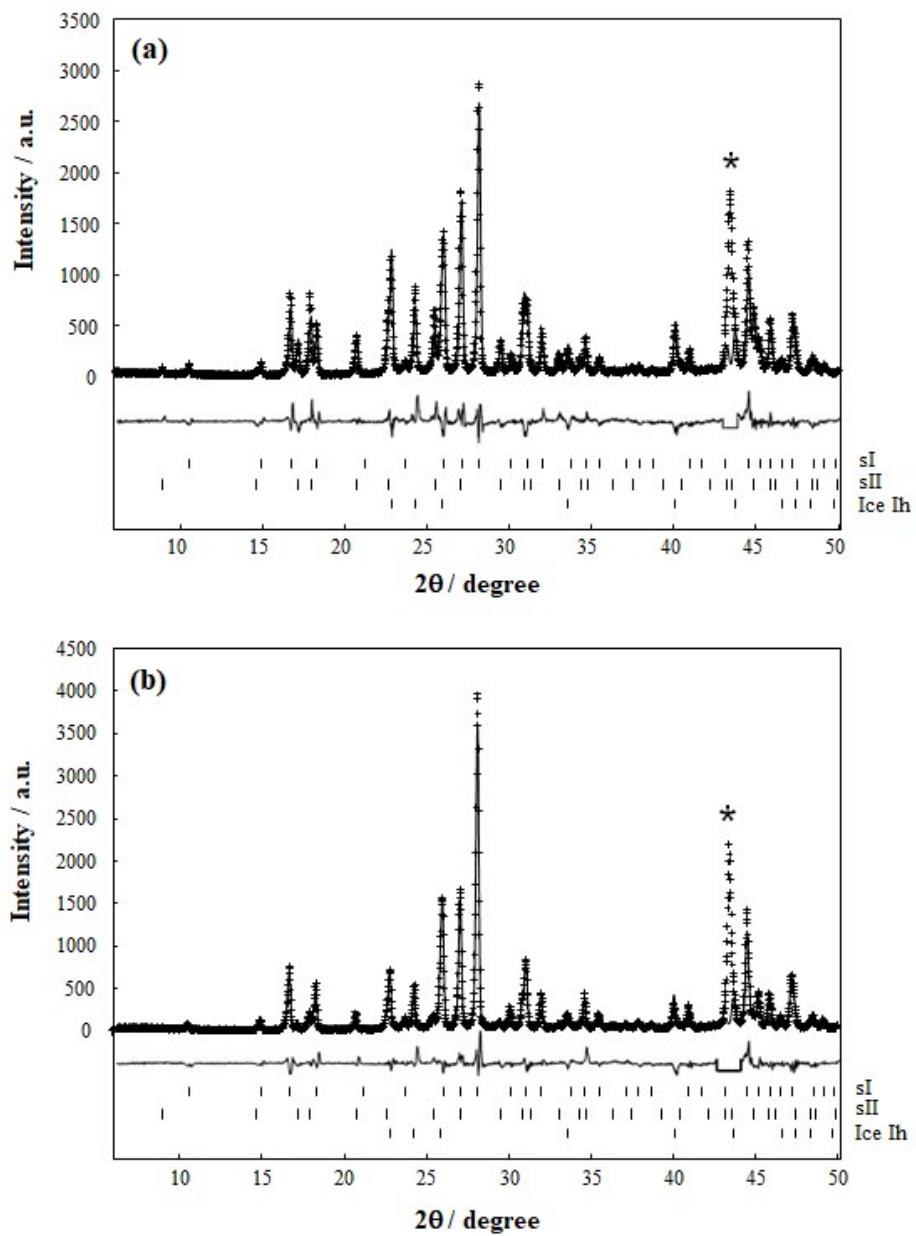
Satoshi Takeya,<sup>\*a</sup> Akihiro Hachikubo<sup>b</sup>

<sup>a</sup> National Institute of Advanced Industrial Science and Technology (AIST), Central 5, 1-1-1 Higashi, Tsukuba 305-8565, Japan.

<sup>b</sup> Kitami Institute of Technology, 165 Koen-cho, Kitami 090-8507, Japan



**Figure S1** Powder X-ray diffraction patterns of the binary gas hydrate of  $\text{C}_3\text{H}_8 + \text{CH}_4$  at 123 K. Here, the bottom dashes represent the calculated peak positions for the sI and sII hydrate and the hexagonal ice. (a)  $\text{C}_3\text{H}_8 + \text{CH}_4$  hydrate ( $\text{C}_3\text{H}_8/\text{CH}_4 = 0.393/0.607$ ) of sII, (b)  $\text{C}_3\text{H}_8 + \text{CH}_4$  hydrate ( $\text{C}_3\text{H}_8/\text{CH}_4 = 0.028/0.972$ ) of sI and sII, and (c)  $\text{C}_3\text{H}_8 + \text{CH}_4$  hydrate ( $\text{C}_3\text{H}_8/\text{CH}_4 = 0.002/0.998$ ) of sI. The asterisk (\*) represents the diffraction peak of the Cu sample holder.



**Figure S2** PXRD pattern of mixture of sI pure  $\text{CH}_4$  hydrate, sII pure  $\text{C}_3\text{H}_8$  hydrate, and ice Ih at 123 K. The plus signs (+) denote the observed intensities; the black solid line was calculated from the crystal structure model. The bottom curve shows the deviation between the observed and calculated intensities, where blank regions are excluded from the analysis because of the presence of Bragg reflections (\*) from the Cu sample holder. The bottom dashes represent the calculated peak positions for sI hydrate, sII hydrate, and hexagonal ice Ih. Volume ratio of the mixtures analyzed by the Rietveld method are (a) sI/sII/Ih = 0.55/0.26/0.19 and (b) sI/sII/Ih = 0.80/0.09/0.11.