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Estimation of Gas Composition and Cage Occupancies in $\text{CH}_4\text{-C}_2\text{H}_6$ Hydrates by CP-MAS ^{13}C NMR Technique

[Masato Kida](#)¹⁾, [Hirotohi Sakagami](#)¹⁾, [Nobuo Takahashi](#)¹⁾, [Akihiro Hachikubo](#)²⁾, [Hitoshi Shoji](#)²⁾, [Yasushi Kamata](#)³⁾⁵⁾, [Takao Ebinuma](#)³⁾, [Hideo Narita](#)³⁾ and [Satoshi Takeya](#)⁴⁾

1) Dept. of Materials Science, Kitami Institute of Technology

2) New Energy Resources Research Center, Kitami Institute of Technology

3) Methane Hydrate Research Laboratory, National Institute of Advanced Industrial Science and Technology (AIST)

4) Research Institute of Instrumentation Frontier, National Institute of Advanced Industrial Science and Technology (AIST)

5) (Present) Disaster Prevention Technology Div., Railway Technical Research Institute

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CP-MAS ^{13}C NMR measurements were carried out on mixed gas hydrates containing $\text{CH}_4\text{-C}_2\text{H}_6$. The changes in NMR chemical shift values for CH_4 and C_2H_6 clearly corresponded to the structural changes in the hydrate structure. The encaged gas compositions estimated by the integrated ^{13}C NMR signal intensities agreed well with the dissociated gas compositions measured by gas chromatography. Therefore, the gas composition in mixed gas hydrates can be directly estimated from the ^{13}C NMR spectra. The cage occupancies of the small and large cages of the hydrates were estimated from the ^{13}C NMR spectra on the basis of a statistical thermodynamic model. The large cages were almost fully occupied with guest molecules, whereas small cage occupancy decreased with increasing C_2H_6 concentration. Therefore, large cages are highly preferentially occupied by C_2H_6 molecules rather than CH_4 molecules.

Keywords: [Gas hydrate](#), [Natural gas](#), [Gas composition](#), [Cage occupancy](#), [¹³C NMR](#)



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