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CRYSTAL STRUCTURE AND STABILITY OF BUTANE CLATHRATE HYDRATE

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ABSTRACT

In this study, powder X-ray diffraction (PXRD) studies of butane (C_4H_{10}) clathrate hydrates were performed. Crystal structure analysis of iso-C_4H_{10} hydrate and n-C_4H_{10} + CH_4 hydrate revealed dynamical disorder and cage occupancies of iso-C_4H_{10} and n-C_4H_{10} molecules in $5^{12}6^4$ large cages. Kinetic stability of iso-C_4H_{10} and n-C_4H_{10} hydrate was examined by temperature ramping method by means of PXRD, and it was suggested that both of them do not show self-preservation phenomena. These results are useful for further understanding of natural gas hydrates.

Keywords: butane, methane, natural gas, hydrate, structure analysis

INTRODUCTION

Natural gas hydrate encages small amount of butane (C_4H_{10}) and pentane (C_5H_{12}) as well as methane (CH_4), ethane (C_2H_6) and propane (C_3H_8). [1]. Although CH_4, C_2H_6, and C_3H_8 hydrate has been investigated from both fundamental and application point of view [2-4], it is still not well known about C_4H_{10} or C_5H_{12} hydrate [5].

Herein, powder X-ray diffraction (PXRD) studies of C_4H_{10} clathrate hydrates were performed, and the effect of structural isomer, n-C_4H_{10} and iso-C_4H_{10}, was examined. Also, the effect of conformation of n-butane, anti and gauche, was examined. *Ab intio* crystal structure determinations of these hydrates were performed. And kinetic stability of both n-C_4H_{10} + CH_4 and iso-C_4H_{10} hydrate structure by temperature ramping were investigated using PXRD method.

EXPERIMENTAL METHODS

Research grade CH_4, n-C_4H_{10} and iso-C_4H_{10} (purity: 99.99%, 99.8% and 99.9%, respectively, supplied by Takachiho Chemical Industry) were used for guest gases of hydrate samples. In the case of n-C_4H_{10} + CH_4 hydrate, finely powdered ice of about 3 g was put into a high-pressure vessel of 20 mL in volume at 255 K. The vessel was evacuated and pressurized by n-C_4H_{10} + CH_4 mixed-gas up to 10 MPa. The temperature was controlled at 274.2 K for more than 1.5 days to melt the ice and to form gas hydrates. After confirmation of no significant pressure decrease (less than 0.01 MPa hour^{-1}), the

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