Implications of the CH_4 – Flue Gas Replacement in the Structure H Hydrate System as Revealed by ^{13}C NMR and a High Pressure Micro-Differential Scanning Calorimeter

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The CH_4-flue gas replacement in naturally occurring gas hydrates has attracted significant attention due to its potential as a method of exploitation of clean energy and sequestration of CO_2. In this study, 2,2-dimethylbutane (Neohexane, NH) was used as a sH hydrate former, and the compositions of flue gas were CO_2 (10%) + N_2 (90%) and CO_2 (20%) + N_2 (80%). The shifts of the quaternary phase ($H-L_W-L_H-V$) equilibrium of initial and replaced hydrates verified that flue gas was successfully replaced with the CH_4 in the sH hydrates. The ^{13}C NMR confirmed that the CH_4-flue gas replacement reaction proceeded without structural transition and the CO_2 molecules also occupied cages of structure H hydrates. A high pressure micro-differential scanning calorimeter (HP μ-DSC) provided reliable hydrate stability conditions and heat of dissociation values in the porous silica gels after the replacement, which confirmed that CH_4 in the hydrates was successfully replaced with flue gas. A heat flow change associated with the dissociation and formation of hydrates was not noticeable during the CH_4 – flue gas replacement in sH hydrates.