## Sequestering Carbon Dioxide into Complex Structures of Naturally Occurring Gas Hydrates

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Large amounts of  $\mathrm{CH_4}$  in the form of solid hydrates are stored on continental margins and in permafrost regions. We explore here the swapping phenomenon occurring in  $\mathrm{SI}$  and  $\mathrm{SII}$   $\mathrm{CH_4}$  hydrate deposits through spectroscopic analyses and its potential application to  $\mathrm{CO_2}$  sequestration at the preliminary phase. The present outcome of 85%  $\mathrm{CH_4}$  recovery rate in  $\mathrm{SI}$   $\mathrm{CH_4}$  hydrate achieved by the direct use of binary  $\mathrm{N_2}$  +  $\mathrm{CO_2}$  guests is quite surprising when compared with the rate of 64% for a pure  $\mathrm{CO_2}$  guest attained in the previous approach. In addition, the simultaneously-occurring dual mechanism of  $\mathrm{CO_2}$  sequestration and  $\mathrm{CH_4}$  recovery is expected to provide the physicochemical background required for developing a promising large-scale approach with economic feasibility. In the case of  $\mathrm{sII}$   $\mathrm{CH_4}$  hydrates, we observe a spontaneous structure transition of  $\mathrm{sII}$  to  $\mathrm{sI}$  during the replacement and a cage-specific distribution of guest molecules. A significant change of the lattice dimension due to structure transformation induces a relative number of small cage sites to reduce, resulting in the considerable increase of  $\mathrm{CH_4}$  recovery rate.