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Characteristics and morphology of CH$_4$/CO$_2$/N$_2$ mixed hydrates using multistep depressurization after hydrate swapping

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Natural gas hydrates are a potentially very large source of energy in the near future. CH$_4$-CO$_2$ swapping is a kind of win-win exploitation method to produce CH$_4$ from hydrate reservoirs while simultaneously storing CO$_2$. To mitigate the problem of low efficiencies of CH$_4$ production and CO$_2$ storage, CH$_4$-CO$_2$ swapping combined with depressurization has been investigated intensively. However, few studies focus on multistep depressurization for mixed CH$_4$/CO$_2$/N$_2$ hydrates after swapping. This work presents characteristics and morphology of well-controlled multistep depressurization after CO$_2$/N$_2$ or CO$_2$/air gas injection into CH$_4$ hydrates, with a view to exploiting this large resource. A high-pressure cell was used to form CH$_4$ hydrate within unconsolidated sand. Mixed gas of CO$_2$/N$_2$ or CO$_2$/air was then injected into the cell to perform the swapping step. Subsequently, pressure release was conducted in steps to produce gas mixtures. Temperatures at the top and bottom as well as pressure in the cell were recorded. The gas compositions collected from produced gas at each multistep depressurization were analyzed via gas chromatography. The morphology of mixed hydrate dissociation was studied separately in a sapphire cell using same procedures, with temperature, pressure, gas compositions and real-time photos recorded correspondingly. The effects of CO$_2$ concentration, gas release position, air composition and L-methionine on CH$_4$ recovery, CO$_2$ storage and mixed hydrate morphology were comprehensively investigated in the high-pressure cell and the sapphire cell. Three evaluation parameters, i.e. CH$_4$ or CO$_2$ mole gas fraction (X$_{CH_4}$ / X$_{CO_2}$), CH$_4$ recovery percent (RCH$_4$) and CO$_2$ storage ratio (SCO$_2$) were calculated to quantitatively analyze the exploitation efficiency.

The results from the high-pressure cell showed that 30mol%CO$_2$ in CO$_2$/N$_2$ injection gas was the most beneficial to improve CH$_4$ concentration. The highest CH$_4$ concentration was 70.2mol% compared with 13mol% and 20mol% for CO$_2$. The highest SCO$_2$ of 18.8% was obtained for gas release from top rather than bottom, while CH$_4$ recovery efficiencies were almost same regardless of gas release position. The results of SCO$_2$ and morphology from the sapphire cell indicated that air could increase CO$_2$ storage by inducing more CO$_2$-containing hydrate reformation during swapping and multistep depressurization. A similar effect of enhanced CO$_2$ storage for L-methionine addition was observed in morphology study. The highest SCO$_2$ of 42.4% was obtained with L-methionine compared to 18.8% without it. In addition, water production was noticed in the morphology experiments when the stepdown pressure was reduced just below mixed hydrate stability pressure. The exploitation results and morphologies in this work confirmed the optimized CO$_2$ concentration in diluent CO$_2$-containing gas for multistep depressurization after swapping exploitation. CO$_2$ storage could be efficiently enhanced by injecting CO$_2$-air gas or adding L-methionine. The stop point for multistep depressurization should be controlled at pressures just above CO$_2$ hydrate stability pressure. This investigation shows that high efficiencies of CO$_2$ storage and CH$_4$ recovery can be achieved through well-controlled multistep depressurization.

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