SUPPORTING INFORMATION

A production method under surveillance: Laboratory pilot-scale simulation of CH₄-CO₂ exchange in a natural gas hydrate reservoir

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Fig. S1 Phase and component distributions during depressurization in E1; a) CH₄ and CO₂ distribution at t₁ (before depressurization) between a gas phase (partition coefficient = 0) and a gas hydrate phase (partition coefficient = 1); CH₄ and CO₂ distribution at given constraints (blue: water availability; grey: phase saturation; yellow: effluent gas composition); grey: possible CH₄:CO₂ partition states based on phase saturation constraints assuming an error of ±2.5% (Sₘ + Sₖ + Sₕḥ = 0.975 to 1.025) (dark grey) or an error of ±10% (light grey). The overlay of all given constraints results in overall possible CH₄ and CO₂ distributions as shown by red areas (Light red: Error ±10%, bright red: Error ±2.5%). b) Phase and component inventories based on possible partition states prior to depressurization at t₂ of experiment E1; Gas hydrate (GH) and gas phase CH₄:CO₂ ratios refer to the average of the bright red region for partitioning coefficients in the upper diagram.
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(upper bar chart); the lower bar chart shows the theoretical CH₄:CO₂ gas hydrate composition that would be in equilibrium with the detected gas phase; numbers in red show the CH₄:CO₂ molar ratios in the corresponding phase; c,d) show the corresponding results at time t₂.

Fig. S2  Phase and component distributions during depressurization in E2; a) CH₄ and CO₂ distribution at t₁ (before degassing) between gas phase (partition coefficient = 0) and gas hydrate phase (partition coefficient = 1); b) Phase and component inventories based on overall possible partition states prior to depressurization at t₁ of experiment E2; For further details see Supplementary Fig. S1.
Fig. S 3 Temperature evolution during the five injection intervals and equilibration periods in experiment E1 (supplement to the top panel of Figure 5 in the main manuscript). While on the left the complete temperature ranges are plotted, the figures on the right show more detailed changes in the temperature range of 7 – 12 °C. In Fig. S3 d (injection interval 4) the negative excursion of temperatures in T0, T1 is due to a strong temporary pressure decrease (p= 3.5 MPa) at the outlet with subsequent degassing and gas hydrate decomposition.
Fig. S 4 Temperature evolution during the first injection interval (0 – 4 h) and equilibration period (up to 14 h) of experiment E1 visualized as 2D heat maps. Temperatures between 13.5 – 33° C are summarized in one color.