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## Thermodynamic Stability Conditions, Methane Enrichment, and Gas

## Uptake of Ionic Clathrate Hydrates of Mine Ventilation Air

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**Abstract:** Abatement of methane from mine ventilation air (MVA) is a significant challenge faced by coal mining industry. A promising method for methane capture from gas mixture is clathrate hydrate formation. In search of suitable and cost-effective low-dosage promoters for hydrate-based methane capture processes, this paper reports the pressure requirement for the hydrate formation of simulated MVA (0.5 vol% CH<sub>4</sub> + 99.5 vol% air) and its potential for methane extraction, in the presence of tri-n-butyl phosphine oxide (TBPO) or tetra-n-butyl ammonium bromide (TBAB) at three different initial loadings (5 wt%, 15 wt%, and 26 wt%). An isochoric equilibrium step-heating pressure search method was used to measure the hydrate phase equilibrium conditions at the temperature range of (277.61 to 295.54) K and pressure range of (0.23 to 19.11) MPa. It was found that at a given initial loading, TBPO was largely more effective than TBAB in reducing the pressure requirement for hydrate formation of MVA. At a given temperature, the equilibrium pressures of the clathrate hydrates were indifferent to the change in the initial loading of TBPO from 5 wt% to 26 wt%, in contrast to those of TBAB. Gas composition analysis by gas chromatography confirmed that CH<sub>4</sub> could be significantly enriched in the ionic clathrate hydrates, and the highest methane enrichment ratio obtained in the present work was 300%, with TBPO at initial loading of 5 wt%. At this relatively low loading, within a given period of 5 hours, TBPO also led to higher gas uptake compared with TBAB. The advantages of TBPO as a promoter of MVA hydrate were discussed.

**Keywords:** Ventilation air methane; Ionic clathrate hydrate; Tri-n-butylphosphine oxide; Tetra-n-butyl ammonium bromide; Phase equilibrium

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