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Phase Stability Conditions of Carbon Dioxide and Methane Clathrate Hydrates in the Presence of KBr, CaBr<sub>2</sub>, MgCl<sub>2</sub>, HCOONa, and HCOOK Aqueous Solutions: Experimental Measurements and Thermodynamic Modelling

Arash Kamari, Hamed Hashemi, Saeedeh Babaee, Amir H Mohammadi, Deresh Ramjugernath

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## **ACCEPTED MANUSCRIPT**

Phase Stability Conditions of Carbon Dioxide and Methane Clathrate Hydrates in the Presence of KBr, CaBr<sub>2</sub>, MgCl<sub>2</sub>, HCOONa, and HCOOK Aqueous Solutions: Experimental Measurements and Thermodynamic Modelling

Arash Kamari <sup>a</sup>, Hamed Hashemi <sup>a</sup>, Saeedeh Babaee <sup>a</sup>, Amir H Mohammadi <sup>\*a,b</sup>, Deresh Ramjugernath <sup>\*\*a</sup>

Abstract – Experimental data on dissociation conditions of carbon dioxide clathrate hydrates in the presence of KBr, CaBr<sub>2</sub>, MgCl<sub>2</sub>, HCOONa, and HCOOK aqueous solutions, as well as methane clathrate hydrates in the presence of HCOONa and HCOOK aqueous solutions have been measured and are reported in this study. Measurements were conducted in temperature and pressure ranges of (266.9 to 284.5) K and (1.47 to 9.56) MPa, respectively, using an isochoric pressure-search method. Results reveal that at varying concentrations, the aqueous salt solutions have a satisfactory inhibition effects on both carbon dioxide and methane hydrate formation, causing a shift in the P-T (pressure-temperature) equilibrium cruves to higher pressures and lower temperatures. A thermodynamic model based on the solid solution theory of van der Waals and Platteeuw (vdW-P) was used for estimation of hydrate dissociation conditions in the presence of the aqueous salt solutions. The UNIQUAC approach, along with the Debeye-Huckel method, was applied to calculate the activity coefficient of water in the presence of salt. The UNIQUAC interaction parameters between water and anions and cations were optimized using the measured hydrate dissociation data. The model results provide a reasonable agreement with the experimental data, with an average absolute deviation (ARD%) below 0.3%.

**Keywords -** Gas hydrate; Dissociation condition; Phase equilibria; Experimental data; Thermodynamic modelling, UNIQUAC

<sup>&</sup>lt;sup>a</sup> Thermodynamics Research Unit, School of Engineering, University of KwaZulu-Natal, Howard College Campus, King George V Avenue, Durban 4041, South Africa

<sup>&</sup>lt;sup>b</sup> Institut de Recherche en Génie Chimique et Pétrolier (IRGCP), Paris Cedex, France

<sup>\*</sup>Corresponding authors Email: <u>a.h.m@irgcp.fr</u> & <u>amir\_h\_mohammadi@yahoo.com</u>

<sup>\*\*</sup>Corresponding authors Email: ramjuger@ukzn.ac.za

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