Clathrate hydrate formation in ternary system of methane, water and ethylene carbonate
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Introduction
Gas hydrates are ice-like crystalline molecular complexes that are composed of water and suitably sized guest molecules like methane. Gas hydrates have drawn much attention in various fields. Formation of gas hydrate plugs may cause serious problems for oil and gas transportation in pipelines. On the other hand, gas hydrates are also used for other applications such as natural gas storage and transportation. Extensive researches on the screening of appropriate additives with inhibition and promotion effects in gas hydrate systems have been presented in literature. It was demonstrated [1] that cyclopentanone C5H9O (CP) additive has a promotion effect on the formation of methane hydrate and shifts hydrate phase boundaries to lower pressure and higher temperature. Ethylene carbonate C2H4O3 (EC) is the structure analogue of more hydrophobic CP.

Aim
Aim of this study is to investigate hydrate formation in ternary system of methane, water and ethylene carbonate and to measure phase equilibrium conditions for this system.

Materials & Methods
The following materials were used:
1. distilled water;
2. methane (Moscow’s Gas Refinery Plant, >99.99 % pure);
3. Ethylene Carbonate (Sigma Aldrich, >99 % pure).

We used the Gas Hydrate Autoclave GHA350 to prepare the gas hydrates and to measure their dissociation conditions. Three-phase equilibrium conditions L–V–H (liquid water solution-vapor-hydrate) were determined by the isochoric method. New experimental results are reported for pressures ranging from 4 to 22 MPa and for mass fraction of ethylene carbonate ranging from 0.05 to 0.25 (0.011 to 0.064 for mole fraction).

Results & Discussion
The experimental results show that the addition of ethylene carbonate in the system shifts hydrate phase boundaries to higher pressure and lower temperature in contrast to more hydrophobic cyclopentanone.

Conclusions
We measured the three phase equilibrium conditions in the methane, water and ethylene carbonate system. Ethylene carbonate shifts hydrate phase boundaries to higher pressure and lower temperature. Despite the similarity of cycle dimensions and chemical structure, these compounds behave in the opposite way. CP is a thermodynamic hydrate promoter whereas EC acts as thermodynamic hydrate inhibitor.

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References

Figure 1 – Schematic illustration of experimental procedure
Figure 2 – Comparison of equilibrium conditions for gas hydrates in CH4+H2O+EC and CH4+H2O+CP systems