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催化、动力学与反应器

CO_2置换CH_4水合物中CH_4的实验和动力学

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摘要 在自行设计的反应装置中考察了2.8 MPa和3.25 MPa压力下，温度271.2、273.2和276.0 K时CO2气体置换十二烷基硫酸钠（SDS）体系CH4水合物中CH4的置换过程。实验数据表明，在反应的前50 h，CH4水合物的分解速率较快，其后分解速率变慢。冰点以上CH4水合物的分解速率较快。基于动力学数据，建立了SDS体系置换反应过程中CH4水合物的分解动力学模型和CO2水合物的生成动力学模型。计算得到CH4-CO2置换反应过程中CH4水合物的分解活化能为28.81 kJ·mol-1，CO2水合物的生成活化能为68.40 kJ·mol-1。数据表明，CH4水合物的分解可能受置换反应过程中水分子的重排控制，而CO2水合物的生成可能受CO2气体在水合物中的扩散控制。

关键词 CO2 CH4水合物 SDS 置换 动力学模型

分类号

Experimental and kinetic studies on methane replacement from methane hydrate formed in SDS system by using pressurized CO_2

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Abstract

The process and kinetics of CH4 replacement from CH4 hydrate formed in sodium dodecyl sulfate(SDS) system by using pressurized CO2 was observed with a self-designed device at temperatures of 271.2, 273.2 and 276.0 K and at pressures of 2.8, 3.25 MPa.Experimental data showed that the decomposition rate of CH4 hydrate was faster in the first 50 hours than the later time.Above ice point, the decomposition rate of CH4 hydrate was faster than that below ice point.Based on the rate data, kinetic models were developed for CH4 hydrate decomposition and CO2 hydrate formation.In the process of CH4-CO2 replacement in the hydrate, the activation energies were,calculated 28.81 kJ·mol-1 for CH4 hydrate decomposition and 68.40 kJ·mol-1 for CO2 hydrate formation respectively.The analysis of the experimental data and activation energies indicated that CH4 hydrate decomposition was probably dominated by re-arrangement of water molecules in the hydrate and CO2 hydrate formation was probably dominated by diffusion of CO2 in the hydrate.

Key words CO2 CH4 hydrate SDS replacement kinetic model

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