

Thermodynamic optimization for dissociation process of gas hydrates



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ABSTRACT

The dissociation process of gas hydrates is also the discharging process of the gas hydrate cool storage system. In order to reduce the entropy generation rate of the gas hydrate dissociation process, this paper takes the entropy generation minimization as the optimization objective to perform thermodynamic optimization for the related process. By establishing thermodynamic optimization model of the gas hydrate dissolution process based on entropy generation analysis, both the optimal control strategy and the optimal heating rate of the gas hydrate dissolution process are determined. The entropy generation rate related to the optimal heating rate decreases by 7.5% compared with normal situation. The research results can provide important guidelines for optimal design and operation of the dissolution process of gas hydrates related to the gas hydrate cool storage system.

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1. Introduction

Gas hydrates are a class of crystalline inclusion compounds with nonstoichiometric composition, consisting of guest molecules trapped in a lattice of polyhedral water cages [1,2]. Gas hydrates have received increasing attention, mainly due to the natural gas hydrate as a clean carbon-based energy source [3,4], hydrate-based CO₂ separation [4–7], potential exploitation of hydrates for gas storage [4,8], transportation and gas separation [4,9], produce gas from gas hydrates [4,9–12], etc. Chatti et al. [13] reviewed the benefits and drawbacks of clathrate hydrates in their interest areas in details. Most of the refrigerants formed with water as refrigerant gas hydrates are particularly attractive for offpeak building cooling, because they freeze at temperatures higher than ice and their phase change latent heat is similar to that of ice [14–30]. Discharging process is the refrigerant gas hydrate dissolution process using the heating medium for heating. The applications of gas hydrates involve complex thermodynamic and kinetic problems which need to establish the relationship between the dissolution rate and thermodynamic variables. The heat transfer in the dissolution process of gas hydrates is somewhat similar to nucleate boiling of liquids. The researchers of Holder's lab firstly obtained measurements on the rate of heat transfer to a solid hydrate phase which decomposed into gas and liquid phases simultaneously [31].

Kamath et al. [32] put the dissolution process of gas hydrate as nucleate boiling of liquids and established the model of the hydrate dissociation process. Selim et al. [33] developed a mathematical model for hydrate dissociation and the results from the solution of the governing differential equations describing the rate of dissociation can be used to estimate the amount of hydrate dissociated as a function of time. Kim et al. [34] developed an intrinsic model for the kinetics of hydrate decomposition which indicated that the decomposition rate was proportional to the particle surface area and to the difference in the fugacity of methane at the equilibrium pressure and the decomposition pressure. Jamaluddin et al. [35] established the model of decomposition of a synthetic core of methane hydrate by coupling intrinsic kinetics with heat transfer rates. Simulation results indicated that the global rate of decomposition can be affected significantly by moving from a heat transfer controlled regime to a regime where both heat transfer and intrinsic kinetics by changing the system pressure. Clarke et al. [36] developed a new mathematical model which accounted for the distribution of particle sizes in the hydrate phase to determine the rate constant of decomposition. Recently, the research on the decomposition of gas hydrate is still in development. The dissociation kinetics at different heating rates was studied in Refs. [37], where it was shown that even very slow heating can significantly affect the intensity of dissociation. The experiments of dissociation of methane hydrate under external pressure revealed that not the curvature, but the heat flux value to regulate the dissociation rate and the change in diffusion [38]. By applying the CDM “(Consecutive Desorption and Melting Model)”, the process of gas hydrate

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Nomenclature

A_2	coefficient related to the characteristics of the dissolution process of the hydrate medium [$J \cdot s^{1/2} / (K \cdot m^3)$]	P_m	pressure at critical decomposition point (Pa)
B_2	coefficient related to the characteristics of the heat transfer process of the hydrate medium ($K/s^{1/2}$)	P^*	optimal pressure (Pa)
c	the specific heat [$J/(kg \cdot K)$]	$\dot{Q}$	rate of heat transfer (W)
F_H	sum of the gas hydrate crystals surface area (m^2)	R	universal gas constant [$8.314 J/(mol \cdot K)$]
F	area of the heat exchanger (m^2)	T	temperature (K)
f	fugacity of the hydration medium (Pa)	T_m	temperature at critical decomposition point (K)
H_r	heat of the hydrate reaction, i.e., the latent heat of liquid–solid phase change (J/mol)	T^*	optimal temperature (K)
K^*	dissolution rate constant of gas hydrates [$mol/(m^2 \cdot s \cdot Pa)$]	t	time (s)
K'_f	heat transfer coefficient [$W/(m^2 \cdot K)$]	U_3	gas hydrate dissolution rate (mol/s)
L	Lagrange function	U_3^*	optimal dependence of the gas hydrate dissolution rate (mol/s)
$\dot{m}$	mass flow rate (kg/s)	Greek symbols	
n_2	total mole of generated gas hydrates at different time (mol)	λ	Lagrange multiplier
$\bar{n}$	total mole of gas hydrates (mol)	σ_2	entropy generation rate of the dissolution process of gas hydrates (W/K)
n^*	optimal dependence of the total gas hydrate mole on time (mol)	μ	chemical potential (J/mol)
p'	pressure (Pa)	Π	phenomenological coefficient [$((mol)^{4/3} K)/(J \cdot s)$]
		Subscripts	
		d	cool discharging process
		eq	equilibrium
		f	heat transfer medium

decomposition was assumed to comprise two consecutive and repetitive quasi chemical reaction steps [39]. The visual observation indicated that the model of the methane hydrate dissociation could be a consecutive process of solid dissolution, guest diffusion, and vapor phase formation and growth [40]. Molecular dynamics simulation has been widely applied to the analysis of gas hydrate dissociation process. The dissociation mechanisms of gas hydrates (I, II, H) were revealed by analyzing the structural snapshots, radial distribution functions and diffusion coefficients at different temperatures. The diffusion rates of water molecules and guest molecules increased with the rising of temperature [41]. The analytic model of the mass and heat transfers of hydrate dissociation by depressurization and thermal stimulation in porous media was established, theoretical analysis of the effects of the depressurizing rate and the warm water injection temperature on the gas production and heat transfer characteristics of the methane hydrate dissociation were carried out, and the predicted results were in good agree with the experimental results [42,43]. In addition, the decomposition driving force [44], effects of activated carbon particle sizes [45], using microwave and radio frequency in-liquid plasma methods [46], etc. on methane hydrate dissociation and gas production were reported.

In recent years, thermodynamic optimization theory has been applied for optimizing performance of thermodynamic cycles and devices, and has made great progress in the fields of physics and engineering [28,47–57]. The applications of thermodynamic optimization theory include heat engines [47–49], heat pumps [50], refrigerators [51], heat exchangers [52], distillation systems [49], chemical reactions [49], mesoscopic systems [49], quantum systems [49], direct energy conversion devices [52–54], thermal energy storage systems [55–57], and even life processes of animals, superconducting transition phenomena, wind energy systems of the earth [49], etc. However, so far there are a few related references that study on the thermodynamic optimization for solution crystallization process [48,58,59] and crystallization process of refrigerant gas hydrate [57]. There is no related reference that

studies on the thermodynamic optimization for dissolution process. In this paper, the thermodynamic optimization will be performed for the dissolution process of refrigerant gas hydrates.

2. Physical model

From the gas hydrate dissociation kinetics model established in Refs. [31,32,34], it is learned that hydrate dissociation is a heat-transfer-limited process. As gas hydrates (solid) dissociate, they form liquid (water) and gaseous phases. Since all the three phases are present at the dissociating interface, this phenomenon is termed three phase heat transfer. It is reasonable to draw an analogy between nucleate boiling phenomena and heat transfer during hydrate dissociation. Fig. 1 shows schematic diagram of the physical model of gas hydrates dissociation. The overall process of the hydrate decomposition apparently involves two consecutive

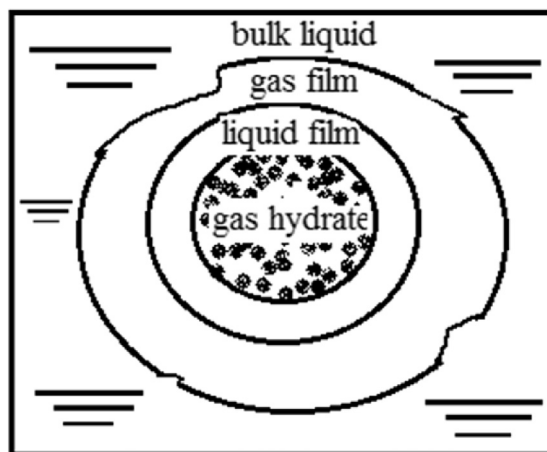


Fig. 1. Schematic diagram of the physical model of gas hydrates dissociation.

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