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Thermal resistances in stirred-tank and tubular reactors for clathrate-hydrate formation: Estimating their reactor-scale dependences

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ARTICLE INFO

Article history:

Received 6 September 2016

Accepted 21 November 2016

Available online 27 November 2016

Keywords:

Clathrate hydrate

Gas hydrate

Heat transfer

Hydrate-forming reactor

Reactor scale-up

ABSTRACT

Efficient formation of clathrate hydrates is the primary condition for establishing the hydrate-based technology for various industrial applications such as the natural gas storage/transport, the capture of CO₂ from industrial flue gases, etc. Because hydrate formation is an exothermic process, the heat discharge from the inside of hydrate-forming reactors can be one of the major rate-controlling processes. The thick stainless-steel walls encasing such reactors configured for high-pressure operations may provide significant resistances to the heat transfer across them. Based on the conventional schemes for calculating the hoop stress in pressure-vessel walls and the convective/conductive heat transfer in cylindrical systems, this paper demonstrates how the thermal resistances to the heat discharge from the inside of each stirred-tank or tubular reactor vary depending on the geometric scale (i.e., the diameter) and the design pressure of the reactor. In general, the thermal resistance to the conductive heat transfer in the reactor wall becomes increasingly significant, relative to the resistance to the convective heat transfer inside the reactor, not only with an increase in the design pressure, but also with an increase in the reactor diameter.

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1. Introduction

Various industrial applications of clathrate hydrates, a class of crystalline solid compounds each consisting of host water molecules and guest molecules of some other (mostly nonaqueous) substance (or substances), have been planned and investigated in the past three-quarters of a century (Sloan and Koh, 2008). Extensive research-and-development efforts have been devoted to several of these applications such as seawater desalination by means of forming and separating hydrate crystals from residual liquids (see, for example, Barduhn, 1967; Kang et al., 2014), the storage and transport of natural gas in the form of hydrates (Mori, 2003; Horiguchi et al., 2011; Rehder et al., 2012; Shin et al., 2016), the pre- or post-combustion separation of CO₂ from fuel-gas mixtures or flue gases by means of hydrate formation (Babu et al., 2015; Dashti et al., 2015; Ma et al., 2016), and the on-board or off-board storage of hydrogen in the form of hydrates (Strobel et al., 2009; Veluswamy et al., 2014). Most of such hydrate-based technologies devised so far have not yet been established for practical use because

of technical difficulties in forming and/or handling hydrates or unclear prospects for the economic success of these new technologies. Efficient hydrate formation is one of the major tasks for establishing these technologies.

In general, the rate of hydrate formation depends on two transport processes: (1) the mass transfer of the hydrate-guest substance to the hydrate-crystal-forming sites inside (or in contact with) a continuous aqueous phase or, on the contrary, the mass transfer of water to the hydrate-crystal-forming sites located in a continuous phase rich with the hydrate-guest substance, and (2) the heat transfer from the hydrate-crystal-forming sites to the outside of the given hydrate-forming system. The latter process is inevitable for continuously removing the heat generated by the exothermic hydrate-forming reaction, thereby maintaining the crystal-forming sites at a desirable temperature. If its operation is planned to be done at a temperature above the water-freezing point, the hydrate-forming system is generally designed such that either the aqueous liquid (liquid water in which the hydrate-guest substance possibly dissolves) or the hydrate-guest substance (typically

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Nomenclature

a	Constant defined in Eq. (10) ($\text{m}^{-1/3}$)
C_1	Empirical factor defined in Eq. (9) (–)
C_2	Constant given by Eq. (12) ($\text{J K}^{-1} \text{m}^{-7/3} \text{s}^{-1/3}$)
C_3	Constant given by Eq. (21) ($\text{J K}^{-1} \text{m}^{-13/5} \text{s}^{-1/5}$)
D	$= 2 R_{\text{in}}$, inside diameter of cylindrical reactor (m)
d	Impeller diameter (m)
\hat{d}	$= d/D$, impeller-to-tank diameter ratio (–)
f	Darcy–Weisbach friction factor (–)
h	Convection coefficient for heat transfer to inside surface of reactor wall ($\text{W m}^{-2} \text{K}^{-1}$)
k	Thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)
L	Axial length relevant to heat transfer (m)
N	Rotational speed (revolutions per second) of stirrer shaft (s^{-1})
Nu_D	$= hD/k_l$, Nusselt number for heat transfer from aqueous phase to reactor wall (–)
p_{in}	Internal pressure (Pa)
p_{out}	External pressure (Pa)
Pr	Prandtl number (–)
r	Radial coordinate (m)
R_{in}	Inside radius of cylindrical reactor (m)
R_{m}	$= (R_{\text{in}} + R_{\text{out}})/2$, mean radius of cylindrical reactor (m)
R_{out}	Outside radius of cylindrical reactor (m)
\hat{R}	$= R_{\text{out}}/R_{\text{in}}$
Re_D	$= u_{\text{av}}D/\nu_l$, Reynolds number for liquid flow through a tube (–)
Re_d	$= Nd^2/\nu_l$, Reynolds number for rotational liquid flow induced by a stirrer (–)
t	$= R_{\text{out}} - R_{\text{in}}$, reactor wall thickness (m)
t_{min}	Allowable minimum value of t (m)
TR_{in}	Thermal resistance to convective heat transfer to internal reactor-wall surface axially extending to L (K W^{-1})
TR_{wall}	Thermal resistance to heat flow crossing reactor wall axially extending to L (K W^{-1})
u_{av}	Average axial velocity of liquid flowing through a tube (m s^{-1})
\dot{V}	Volumetric flow rate ($\text{m}^3 \text{s}^{-1}$)
\dot{W}	Power required for driving a stirrer (W)
\dot{w}	Power required for stirring a unit volume of liquid (W m^{-3})

Greek symbols

ε	Volume fraction (–)
κ	Thermal diffusivity ($\text{m}^2 \text{s}^{-1}$)
μ	Dynamic viscosity (Pa s)
ν	Kinematic viscosity ($\text{m}^2 \text{s}^{-1}$)
σ_{AS}	Allowable stress (Pa)
σ_{UTS}	Ultimate tensile strength (Pa)
σ_{YS}	Yield tensile strength (Pa)
σ_{θ}	Hoop stress (or circumferential stress) (Pa)
$\sigma_{\theta, \text{max}}$	Maximum value of σ_{θ} (Pa)

Subscripts

g	Gas
h	Hydrate
l	Aqueous liquid phase which may be regarded as lw or sl
lw	Pure water in liquid state
sl	Water-based hydrate slurry
w	Reactor-wall material

in the form of a gas possibly containing water vapor) is dispersed into the other in order to extend the phase-separating interfaces, thereby enhancing the mass transfer process (Mori, 2003). The characteristics of the heat transfer process inevitably depends on the state of the above-described fluid-in-fluid dispersion. Because of the high thermal conductivity and high heat capacity of the aqueous liquid, the latter dispersion type in which the aqueous liquid serves as the continuous phase is generally considered to be more favorable for the heat transfer as compared to the former type in which the hydrate-guest substance, mostly in the gaseous state, serves as the continuous phase.

Typically, the hydrate-forming system uses a vessel-type or tubular reactor equipped with an external cooling jacket or a cooling coil through which a liquid coolant is forced to flow (see Fig. 1). While the mass transfer process relevant to the hydrate formation occurs only inside the reactor, the heat transfer process for discharging the heat released by the hydrate formation extends over the three regions aligned in series — the inside of the reactor, the reactor wall, and the inside of the cooling jacket. These regions provide individual thermal resistances against the heat transfer from the inside of the reactor to the coolant flowing in the jacket.¹ This study intends to comparatively evaluate the magnitudes of the two thermal resistances in each hydrate-forming reactor — the resistance to the convective heat transfer inside the reactor and the resistance to the conductive heat transfer inside the reactor wall. In view of the variety of cooling-jacket designs, many alternative materials usable as the coolant, and the arbitrariness in determining the coolant flow rate, we exclude the thermal resistance inside the cooling jacket from the scope of this study. Particular interest is paid in investigating how the two thermal resistances, one inside each reactor and the other in its wall, and their mutual ratio vary with the reactor scale. In quantitatively evaluating the thermal resistances, we specify the following conditions on the geometrical design and the operation of the reactors.

- (i) Each reactor is either (a) a cylindrical stirred-tank reactor into which a hydrate-guest substance in the gas state is continuously supplied (probably via bubbling into a pool of the aqueous liquid) or (b) a cylindrical reactor in which the hydrate-guest substance and liquid water are forced to flow co-currently in the form of a bubbly, plug or slug flow. That is, the aqueous liquid serves as the continuous phase in the major portion of the inside space of the reactor.
- (ii) The reactors are operated in the continuous mode such that both the thermodynamic and fluid-mechanical conditions inside each reactor are practically invariant during each hydrate-forming operation.
- (iii) The hydrate-guest substance is assumed to be pure methane. The pressure inside each reactor is maintained at 4 MPa or 8 MPa, while the bulk temperature inside the reactor is maintained at 275 K. (Note that these conditions are used only at the final stage of calculating the thermal resistances. The procedure of calcula-

¹ The reactor and the cooling jacket may or may not be integrated into a single unit. If the cooling jacket has its own inside wall, this wall provides an additional thermal resistance. If the reactor is wrapped with a helical cooling coil, instead of an annular jacket, the heat flow outside the reactor wall may significantly deviate from the simple radial geometry, thereby complicating the evaluation of the thermal resistances outside the reactor wall.

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