



Entropy generation minimization for isothermal crystallization processes with a generalized mass diffusion law



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ABSTRACT

Entropy generation minimization for a class of isothermal crystallization processes with a generalized mass diffusion law is investigated in this paper. For the given total mass of crystals, the optimality condition corresponding to the MEG (Minimum Entropy Generation) of the process is obtained firstly, and special cases for both the linear [$g \propto \Delta(\mu)$] and the diffusive [$g \propto \Delta(c)$] mass diffusion laws are further derived. The optimization results are also compared with the two different mass diffusion strategies of CKCC (Constant Key Component Concentration) and CMDF (Constant Mass Diffusion Flux) operations. The results indicate that for the case with the linear law, the entropy generation rate for the MEG of the process keeps constant, and the total entropy generations for the strategies of CKCC and CMDF operations are equal to each other; while for the case with the diffusive law, the entropy generation rate for the MEG of the process decreases with the time, and the strategy of CMDF is superior to that of CKCC. The results can provide important guidelines for optimal design and operation of the crystallization processes in engineering.

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1. Introduction

The theory of EGM (Entropy Generation Minimization) or FTT (Finite Time Thermodynamics) has achieved great progress since the mid 1970s [1–23]. Bejan [24] first optimized the geometry of heat transfer tubes for the MEG (Minimum Entropy Generation) caused by both heat transfer and fluid flow viscosity. Some authors investigated optimal temperature configurations for the MEG of heat exchange processes with Newtonian heat transfer law [$q \propto \Delta T$] [25–30], and further discussed effects of different thermal-resistance models [31–37]. Berry et al. [3] and Tsirlin et al. [38,39] derived optimal configurations for the MEG of a series of thermodynamic and chemical processes, which include heat transfer, throttling, isothermal mass transfer, isothermal crystallization, mechanical friction and chemical conversion, and further extended the related results to heat-driven and mechanical separation [40–42], optimal separation sequence for multi-component mixtures [43] and binary distillation separation process optimization [44]. Xia et al. [45] derived the optimal concentration configuration for the MEG of one-direction isothermal mass diffusion

process. For a class of tubular chemical reactors coexisted with the irreversible losses of finite temperature-difference heat transfer, finite pressure-drop fluid flow and finite potential-difference chemical reaction, Nummedal [46] and Johannessen [47] considered that the external furnace gas temperature was controllable completely, and further obtained the optimal temperature configurations of the furnace gas for the MEG of the chemical reactors. Ahmadi and Mehrpooya [48], and Ahmadi et al. [49–53] investigated the optimal thermo-economic performances of solar driven heat engine [48,49] and four-temperature-level absorption refrigeration [50] by applying the theory of FTT, and further performed multi-objective optimizations for the Rankine [51], the nano-scale Braysson [52], the nanoscale Stirling refrigeration [53] cycles. Xu et al. [54] discussed the difference between the irreversibility and available energy loss of heat transfer process in heat exchanger. Qin et al. [55] investigated the ecological performance of four-temperature-level absorption heat transformer with heat resistance, heat leakage and internal irreversibility. Sieniutycz et al. [56] derived the minimal dissipation conditions of systems for the MEG with the given heat load.

Gordon et al. [57] and Xia et al. [58] obtained the optimal temperature configuration of external heat reservoir for the MEG [57] and the MED (Minimum Entropy Dissipation) [58] of the liquid-solid phase change process, respectively. Santoro et al. [59]

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Nomenclature

a	constant to be determined	t	time, s
c	concentration	λ	Lagrangian constant
F	surface area, m ²	μ	chemical potential, J/mol
g	mass-diffusion flux, mol/s	τ	process duration, s
h	mass diffusion coefficient, mol ² /(J · s) or mol/s	$()^*$	optimal
K	proportion factor	$()_c$	related to the linear law
L	Lagrangian	$()_{eq}$	related to the equilibrium
M	mass, kg	$()_{min}$	minimum value
p	pressure, Pa	$()_\mu$	related to the diffusive law
R	universal gas constant, J/(mol · K)		
S	entropy, J/K		
T	temperature, K		

obtained the optimal pressure configuration of the external gas phase for the minimum wasted work of the gas-liquid phase change process. Mironova et al. [60] and Bi et al. [61] performed thermodynamic optimization of the crystallization process for the MEG. Refs. [3,39,60,61] investigated the crystallization processes mainly based on the linear phenomenological law [$g \propto \Delta(\mu)$] in the theory of LIT (Linear Irreversible Thermodynamics) [62–65], while the real mass diffusion processes in engineering always obey Fick's diffusive law [$g \propto \Delta(c)$], and optimization for the MEG of the crystallization process with [$g \propto \Delta(c)$] does not have been performed until now. Different heat transfer laws resulted in different optimal reservoir-temperature configurations for the MEG of heat exchange processes with the same heating load [31–37]. Similarly, different mass transfer laws may result in different optimal key-component-concentration configurations for the MEG of crystallization processes with the same amount of crystals grown. Besides, one of aims of FTT is to pursue generalized optimization rules and results. Based on Refs. [3,39,60,61], this paper will further investigate the MEG of the crystallization process with a generalized mass diffusion law. Different from Refs. [3,39,60,61], in which only the optimal strategy of the MEG for the linear law [$g \propto \Delta(\mu)$] was investigated, the strategies of the MEG for the cases with both the linear [$g \propto \Delta(\mu)$] and the diffusive [$g \propto \Delta(c)$] laws will be investigated, and the optimization results will be further compared with those of CKCC (Constant Key Component Concentration) and CMDF (Constant Mass Diffusion Flux) operations. The MEG performance bounds of the crystallization processes and the corresponding pathway conditions can be used to design the thermodynamically most efficient crystallization processes, to estimate how close is a conventional mass diffusion operation to the thermodynamic limit, to construct areas in a process state space that are thermodynamically feasible, etc. [39].

2. Physical model

Since the concentration of the key component in the solution is higher than its equilibrium concentration, the key component crystallizes out of solution on to the surfaces of the crystals already present, i.e. the crystallization process occurs. The original dimensions of the crystals are unknown, and some distribution can be postulated to characterize them. The pressure and the temperature of the solution are known, and are denoted as p and T , respectively. The concentration of the key component in the solution is denoted as c_1 , while its equilibrium concentration is denoted as c_{eq} , and the corresponding chemical potentials are, respectively, given by

$$\mu_1 = \mu_0(T, p) + RT \ln c_1 \quad (1)$$

$$\mu_2 = \mu_0(T, p) + RT \ln c_{eq} \quad (2)$$

where $\mu_0(T, p)$ is the chemical potential of the crystallizing substance in the reference state, and R is the universal gas constant. The mass diffusion flux is denoted as $g(c_1, c_{eq})$, the surface area of the crystals is denoted as F , and their mass is denoted as M . Since $g(c_1, c_{eq})$ and F depend on F and M , respectively, $g(c_1, c_{eq})$ is also the function of M . The initial mass of a crystal is given by $M_i(0)$. For simplicity, all of the crystals are assumed to be sphere such as salicylic acid [66], cefotaxime sodium [67] and clopidogrel sulfate [68] crystals. Their surface area F is proportional to $M_i^{2/3}$, i.e. F is a convex function of M_i as shown in Fig. 1. The net surface area F_Σ for the case of averaging over all of the crystals with the total mass M_i is smaller than that calculated by assuming that all of the crystals are the same and have the same mass. The entropy generation is proportional to the mass-diffusion coefficient, which increases with the increase in the surface area of the crystals. As a result, the application of the dependence $\bar{F}_\Sigma = KM_\Sigma^{3/2}$ provides a low limit value for the entropy generation. This fact will be used in the derivations below.

In terms of the law of mass conservation, the mass M of the crystals is given by

$$\frac{dM}{dt} = g(M^{2/3}, c_1, c_{eq}), \quad M(0) = M_0, \quad M(\tau) = \bar{M}. \quad (3)$$

where τ is the process duration, and $g(M^{2/3}, c_1, c_{eq})$ is the mass diffusion flux, which is a function of mass diffusion area $\bar{F}_\Sigma(M^{2/3})$, c_1 and c_{eq} . $g(M^{2/3}, c_1, c_{eq})$ follows the relations $\partial g / \partial c_1 > 0$, $\partial g / \partial c_{eq} < 0$ and $g = 0$ when $c_1 = c_{eq}$. For the crystallization process

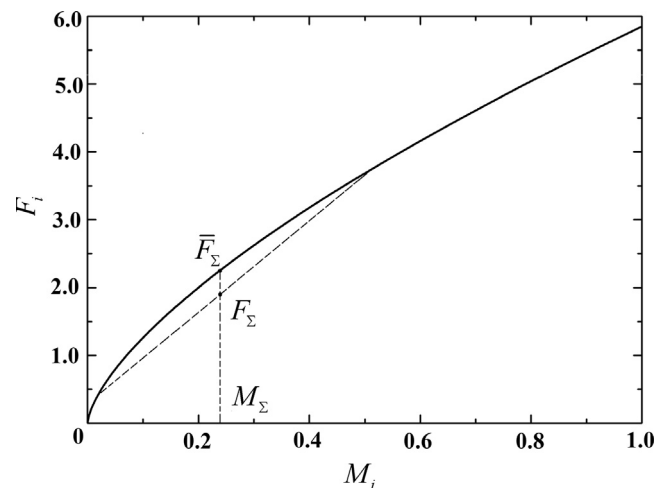


Fig. 1. The characteristic dependence of a crystal's surface on its mass.

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