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Mathematical modeling of static layer crystallization for propellant grade hydrogen peroxide

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ABSTRACT

Hydrogen peroxide (H_2O_2) is an important raw material widely used in many fields. In this work a mathematical model of heat conduction with a moving boundary was proposed to study the melt crystallization process of hydrogen peroxide which was carried out outside a cylindrical crystallizer. Considering the effects of the temperature of the cooling fluid on the thermal conductivity of crude crystal, the model is an improvement of Guardani's research and can be solved by analytic iteration method. An experiment was designed to measure the thickness of crystal layer with time under different conditions. A series of analysis, including the effects of different refrigerant temperature on crystal growth rate, the effects of different cooling rates on crystal layer growth rate, the effects of crystallization temperature on heat transfer and the model's application scope were conducted based on the comparison between experimental results and simulation results of the model.

\1. Introduction

Since Randolph and Larson have established population balance theory, many work have been done in establishing the crystallization process analysis and solution models, which try to use different theory of nucleation or crystal growth dynamics in crystallization theory research [1–3]. Because of the unique characteristics of melt crystallization, the study of heat transfer is more important than that of mass transfer for melt crystallization process. General heat transfer model is key to the study of melt crystallization kinetics, as well as is of instructive significance to small pilot processes, analysis and optimization of amplification.

To study the transfer characteristics of crystal layer interface in layer melt crystallization, impurities contribution on the interface, transport of impurities concluded in the crystal layer and growth process of crystal layer could deepen the exploration of crystal layer mechanism and theories, raise the purity of crystal layer and separation efficiency, and find the suitable operation ways.

The mathematical models of melt crystallization process are complex. In order to obtain the analytical solution, it is required to solve a series of simultaneous partial differential equations: heat transfer differential equations, heat conduction differential equations, differential equations of motion, mass transfer differential equations and so on. It is not easy to solve these equations, and the complexity is due to that the crystallization process is a non-steady-state process. Temperature, concentration, mass, thermal conductivity and flow rate of the fluid system is variable of time, which is difficult to analyze. Therefore, the study of melt crystallization dynamics can't catch up with that of solution's crystallization. In 1970s, a particular melt crystallization process model is discussed [4]. In 1980s, Myasinkov from former Soviet Union established gradual condensation crystallization model, and Miyake Denisov from Japan improved the suspension crystallization packed bed model. In 1990s, Matsuoka and Garside [5] combined the gradual congealing layer crystallizer with suspended bed tower crystallization modes, and take crystals of the dual system as example to promote a comprehensive kinetic model equations.

Since the layer growth process involves transferring and phase change. The crystal layer position changes with time. The equation to describe the layer growth (such as heat conduction of crystal layer, heat transfer and mass transfer of Melt, interface movement equation) has no analytical solution under normal circumstances. Numerical methods(such as finite difference, finite element and boundary element method) has been the main way to solve such problems. For example, Fukui et al. [6] calculate the growth process of laminar melt crystal between inter-dimensional flat through solving N-S equation, heat transfer and mass transfer equations; Radhakrishnan et al. [7,8] use the finite difference of the grain tube falling film to obtain the numerical solution of layer growth. However, in the numerical calculations, it is essential to discrete the differential equation of transfer, which often introduces errors. Besides, for different boundary conditions, the stability and algorithm needs further study. In addition, numerical calculation methods are complicate, which consumes lots of

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energy to complete, and the results are effected by the algorithm, meshing, calculation steps and other factors.

For specific crystallization operation, the way to obtain semi-analytical solution or approximate solution by simplifying the problem of describing the crystal layer growth is concerned. Feltham et al. [9] finds the semi-analytical solution of board, inner sphere surface and the inner cylindrical surface of the melt solidification process for phase-change issues of the two-component system. Chianese et al. [10] use integral method to conduct the semi-analytical calculation of crystal grown outside the tube, which is consistent with the experiment results of caprolactam crystal layer and naphthalene crystal layer.

The tubular outer tube crystals is adopted in our experiment. Shih and Tsay [11] conducted the experiment to analyze the problem that solid separates out from the cooling cylindrical surface in the saturated solution. Tao and Longwell solved the issue with numerical methods and geometric law [12,13]. Shi introduced the analytic iterative method proposed by Siegel [14,15], and compared the results with the former two methods, finding that second-order analytic iterative method could lead to satisfactory results. Guardani et al. [16,17] adopted the method of Shih to study the layer crystallization process under the assumption that the heat rate between cold wall and cooling medium is steady, in which all the factors' effect were considered and the non-isothermal crystallization process was calculated.

2. Mathematical model

It is illustrated the schematic view of the Melt crystallization process on the cylindrical outer surface of the crystallizer within cool fluid in Fig. 1.

When the change rate of solid-liquid interface temperature t_g is far slower than the growth rate of crystal layer, the heat transfer of melt crystallization kinetics on the cylindrical outer surface could be described with partial differential equations and boundary conditions as follows:

$$\frac{\partial t}{\partial \theta} = \frac{k}{\rho C_p}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial t}{\partial r}\right) \tag{1}$$

$$r_0 \le r \le R(\theta) \tag{2}$$

$$\theta = 0,\ \mathrm{R}(\theta) = \mathrm{r}_0 \tag{3}$$

$$r = R(\theta),\ \frac{dR}{d\theta} = \left.\frac{k}{\rho C_p}\frac{\partial t}{\partial r}\right|_R \tag{4}$$

$$r = R(\theta),\ t = t_{gl} \tag{5}$$

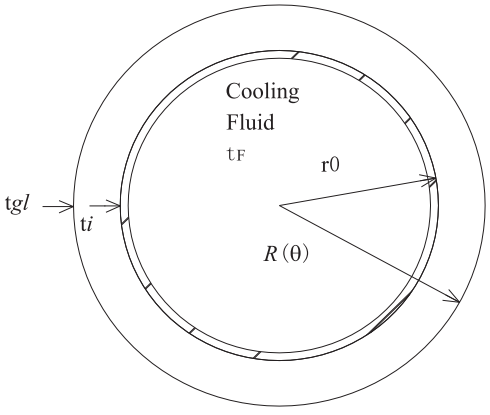

Fig. 1. Cross sectional scheme of the internally cooled tube adopted in the model. r_0–The cylinder outer diameter(crystallization tube). R(θ)–The outer diameter of the crystal which grows with time t_F–the average temperature of the refrigerant inside the tube. t_i–the temperature of the outer surface of cylinder. t_{gl}–the temperature of the solid-liquid interface.

$$r = r_0,\ q = -k\left.\frac{\partial t}{\partial r}\right|_{r_0} = h(t_F - t_i) \tag{6}$$

For the equations above, the analytical iteration method of crystal layer growth inside and outside the tube proposed by Shih and Tsay is applied in this paper. The solution is generated by the algebraic expressions. First, Eqs. (1)–(6) are defined as follows, which are dimensionless.

$$U = \frac{t - t_{gl}}{t_{gl} - t_F} \tag{7}$$

$$\tau = \frac{\lambda\theta}{C_p\rho(r_0)^2} \tag{8}$$

$$x = \ln\left(\frac{r}{r_0}\right) \tag{9}$$

$$X = \ln\left(\frac{R(\theta)}{r_0}\right) \tag{10}$$

$$\beta = \frac{\lambda}{hr_0} \tag{11}$$

$$\alpha = \frac{\Delta H_m}{C_p(t_{gl} - t_F)} \tag{12}$$

$$U_i = \frac{t_i - t_{gl}}{t_{gl} - t_F} \tag{13}$$

The above dimensionless number are taken into the formula (1)–(6), it can be obtained that:

$$e^{2x}\frac{\partial U}{\partial \tau} = \frac{\partial^2 U}{\partial x^2},\quad 0 \le x \le X(\tau) \tag{14}$$

$$\tau = 0,\ X = 0 \tag{15}$$

$$\frac{dX}{d\tau} = \left.\frac{1}{\alpha\cdot e^{2x}}\frac{\partial U}{\partial X}\right|_X \tag{16}$$

$$U(X, \tau) = 0 \tag{17}$$

$$\left.\frac{\partial U}{\partial X}\right|_{x=0} = \frac{1}{\beta}(1 + U_i) \tag{18}$$

Analytic iterative method is used to solve the equations above. Solutions to the equations are:

$$\tau = \frac{1}{4}[1 - 2\alpha\beta - (1 - 2\alpha\beta - 2X)e^{2X}] - G(X) \tag{19}$$

$$U(X, \tau) = -1 + \frac{\alpha(\beta + x)e^{2X} - (\partial I/\partial X)}{\alpha(\beta + X)e^{2X} - (dG/dX)} \tag{20}$$

$$G(X) = \frac{1}{4(\beta + X)}[\beta - 1 + (2\beta - 1)X + (1 - \beta - X)e^{2X}] \tag{21}$$

$$\frac{\partial I}{\partial X} = \frac{1}{4(\beta + X)^2}[1 - 2\beta + 2\beta^2 + (1 - 2\beta - 2X)(\beta + X)e^{2X} + (\beta - 1 + x)e^{2X}] \tag{22}$$

$$\frac{\partial G}{\partial X} = \frac{1}{4(\beta + X)^2}[(1 - 2\beta + 2\beta^2)(1 - e^{2X}) + 2(1 - 2\beta)Xe^{2X} - 2X^2e^{2X}] \tag{23}$$

3. Experiments

Apparatus used in this experiment is shown in Fig. 2. The hydrogen peroxide feed solution is pretreated in the pre-cooling tank with jacket. When the raw material in the pre-cooling tank is cooled to the required