



Proceeding Paper **Two-phase Stefan problem for the modeling of urea prilling** tower

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Abstract. The prilling technique is frequently used to make granular urea and ammonium nitrate. The generated droplets fall and become solid due to the heat removal by the cooling air, which flows in a counter-current direction. Generally, three sequential thermal intervals for the solidification of urea droplets are considered: cooling of liquid drops, solidification at freezing temperature of the liquid phase, and cooling of complete solid particles. In this study, the solidification of the urea droplets has been considered as a two-phase Stefan problem with convective flux boundary condition rather than dividing the whole process into three sequential steps. The heat transfer problem was solved numerically using the enthalpy method. The particles were assumed to attain the terminal velocity immediately. The convective heat transfer was determined from the terminal velocity. The temperature distribution of the droplets, and the minimum height for complete solidification at different particle diameters were investigated.

INTRODUCTION

The prilling technique is frequently used to make granular urea and ammonium nitrate. This basic procedure involves spraying a liquid flow from the top of a tower. At the same time, a stream of cooling air collected from the surrounding is fed from the bottom. The generated droplets fall counter-currently and become solid due to the heat removal by the cooling air. The process produces spherical particles with a nearly uniform size.

In practice, prilling towers can easily suffer operating issues due to incomplete solidification. Because of the poor efficiency of the solidification, a low-quality structure is generated, resulting in productivity and profit losses. Despite the importance of the process, only a few studies have been conducted on the modeling of a prilling tower. In the study of Wu, et al. [1], a simple shrinking core model was used to design a new prilling tower. The model is based on a lumped technique in which the temperature is uniform over the entire particle. Alamdari, et al. [2] developed a distributed model. The temperature distribution within the particle was described by a heat transfer equation. Rahmanian, et al. [3] also applied this model to a local industrial tower with a rectangular cross-sectional area. Mehrez, et al. [4] also employed simultaneous mass, heat, and momentum transfers between the two phases to simulate the process. However, in these models, the same three sequential thermal intervals for the solidification of urea droplets are considered: cooling of liquid drops, solidification at freezing temperature of the liquid phase, and cooling of complete solid particles. In this approach, the solidification interval is classified as a Stefan one-phase problem, in which the temperature of the liquid phase is assumed to be constant. This assumption is not natural because the temperature distribution within the

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Copyright: © 2023 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/). particle should change gradually with time. Therefore, in this report, the solidification of urea particles is considered as a two-phase Stefan problem, in which the heat fluxes occur in both two phases, liquid and solid. The cooling and solidification are treated as one process from liquid droplets to complete solid particles instead of dividing into three intervals. About the hydrodynamic of the process, the particles are assumed to be quickly attain the terminal velocity. This velocity is used to estimate the convective heat transfer coefficient. Boundary condition is the convection cooling with air.

Problem formulation

In the model, for simplification, the urea droplets are assumed to fall vertically and quickly attain their terminal velocity. The heat transfer process with the air is described as a two-phase Stefan problem from the top to the bottom of the prilling tower.

Terminal Velocity of the Urea Particles

The urea particles falling inside the tower are subject to three forces: gravitational force (F_G), which is the same direction to the velocity, and buoyancy force (F_B) and drag force (F_D) act in the opposite direction. At stationary, the force balance acting on the particle gives:

$$F_{G} = F_{B} + F_{D} \qquad (1)$$

$$\frac{1}{6}\pi d_{p}^{3}\rho_{p}g = \frac{1}{6}\pi d_{p}^{3}\rho_{a}g + \frac{1}{2}\rho_{a}C_{D}\pi \frac{d_{p}^{2}}{4}(v_{t} + v_{a})^{2} \qquad (2)$$

Therefore, the terminal velocity can be obtained as

$$v_t = \sqrt{\frac{4}{3} \frac{\left(\rho_p - \rho_a\right) g d_p}{\rho_a C_D}} - v_a \qquad (3)$$

In which, v_i is the terminal velocity (m/s) of the particle related to the tower, v_a is the velocity of the air, g is the acceleration of gravity given by g=9.80665 m/s, Q_P is the particle density (kg/m³), Q_a is the density of the air (kg/m³), d_P is the diameter of the spherical particle in (m), and C_D is a dimensionless drag coefficient.

The drag coefficient on a spherical particle depends on the particle Reymold number. The correlation proposed in Brown and Lawler [5], which fits the range of Reynold number up to 2×10^5 is used in this study:

$$C_{D} = \frac{24}{\text{Re}_{p}} \left(1 + 0.150 \,\text{Re}_{p}^{0.681} \right) + \frac{0.407}{1 + \frac{8710}{\text{Re}_{p}}} \tag{4}$$

Where Re_p is the particle Reynold number as

$$\operatorname{Re}_{p} = \frac{d_{p} \left(v_{t} + v_{a} \right) \rho_{a}}{\mu_{a}} \qquad (5)$$

In which μ_a is the viscosity of air in kg.m⁻¹.s⁻¹.

Since the terminal velocity is also included in the drag coefficient calculation, an iteration is required to obtain the result. First, the initial guest for the terminal velocity was assigned. Then the Reynold number and drag coefficient was calculated. After that, the new value of terminal velocity was estimated from Eq. (3) and compared to the current terminal velocity. If the difference is small (less than 1×10^{-8}), the procedure stops and terminal velocity is obtained. If the difference is still high, the procedure is repeated.

Heat transfer as a two-phase Stefan problem

Consider a spherical liquid urea droplet, as shown in **Error! Reference source not** found.



Figure 1. Configuration of the droplet.

At t > 0 the surrounding temperature is given by T_{s} , which is lower than the freezing temperature T_{f} of urea. Then as time proceeds, the droplets will be cooled down by convection and eventually solidify. The system is governed by the system of equations

$$\frac{\partial T}{\partial t} = \frac{\alpha_l}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right) , \quad 0 \le r < R(t) \quad (6)$$
$$\frac{\partial T}{\partial t} = \frac{\alpha_l}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right) , \quad R(t) \le r < r_p \quad (7)$$

where $\alpha_i = \frac{\kappa_i}{\rho_i c_i}$, k_i , ρ_i , c_i (*i*=*s*,*l*) are the thermal diffusivity, thermal conductivity,

density and specific heat capacity of the solid and liquid phase, respectively; R(t) is the position of the solid-liquid interface and $R(0)=r_{P}$.

At the solid-liquid interface, the flux condition

$$k_{s}\left(\frac{\partial T}{\partial r}\right)_{R(t)} - k_{l}\left(\frac{\partial T}{\partial r}\right)_{R(t)} = L\rho_{s}\frac{dR(t)}{dt}$$
(8)

where *L* is the latent heat of freezing, respectively. The initial and boundary conditions are given by

$$T(r,0) = T_{i}, \ 0 \le r \le r_{1}$$
(9)
$$-k \left. \frac{dT}{dr} \right|_{r=r_{p}} = h \left(T_{r=r_{p}} - T_{a} \right), \ t > 0$$
(10)

where h is the convective heat transfer coefficient, which can be obtained from Ranz-Marshall correlation [6]:

$$Nu = 2 + 0.6 \operatorname{Re}_{p}^{0.5} \operatorname{Pr}^{0.33} (11)$$

where $\text{Nu} = \frac{hd_p}{k_g}$ is the Nusselt number, $\text{Pr} = \frac{c_{p,g}\mu_g}{k_g}$ is the Prandtl number, k_g , $c_{p,g}$,

 μ_{g} are thermal conductivity, specific heat capacity, and viscosity of air.

The two-phase Stefan problem describing the solidification of urea particles can be solved numerically using the enthalpy method. The detail of the numerical schema for the inward solidification of a sphere can be found elsewhere such as in [7].

Solution Procedure and Model Parameters

The assumptions and approach used in this study are as follows. When the particles fall down the tower, the terminal velocity is assumed to be attained immediately. Therefore, for each value of urea particle diameter (1 mm to 2.2 mm), the terminal velocity which is determined by Eq. (3) is used as the steady velocity of the particle. The convective heat transfer coefficient is then obtained by Eq. (11). The heat transfer coefficient *h* is used as the input and the solidification of urea droplet is considered as a two-phase Stefan problem with convective flux boundary condition. The system of partial differential equations is solved to describe the solidification of urea droplets. From the result of the simulation of heat transfer, the temperature distribution within the particle versus the time taken can be obtained. The time required for complete solidification is the time at which the temperature at the center of the particle becomes less than the freezing point. The terminal velocity and the time requirement will give the minimum height of the tower, which allows the particle to solidify completely.

The model parameters are summarized in Error! Reference source not found..

| Parameters | Values |
|--|------------------------|
| Temperature of urea feed (°C) | 140 |
| Velocity of air (m/s) | 0.63 |
| Density of air (kg/m ³) | 1.166 |
| Viscosity of air (Pa.s) | 1.87×10^{-5} |
| Specific heat capacity of air (kJ/(kg.K)) | 1.005 |
| Thermal conductivity of air (W/(m.K)) | 0.025 |
| Density of solid urea (kg/m ³) | 1335 |
| Freezing temperature of urea (°C) | 132 |
| Thermal conductivity of solid urea (W/(m.K)) | 2.651×10^{-2} |
| Specific heat capacity of solid urea (J/(kg.K)) | 1334 |
| Melting heat (kJ/kg) | 224 |
| Density of liquid urea (kg/m ³) | 1220 |
| Thermal conductivity of liquid urea (W/(m.K)) | 1.3×10^{-2} |
| Specific heat capacity of liquid urea (J/(kg.K)) | 2250 |
| Particle (droplet) diameter range (mm) | 0.6 - 2.4 |

Table 1. Parameters and values of the prilling process.

Results and discussions

Terminal velocity and convective heat transfer coefficient

Terminal velocity, convective heat transfer coefficient of urea particles at different sizes when the air velocity is 0.63 m/s are shown in FIGURE 1. From the figure, it can be seen that the terminal velocity increases with an increase of particle diameter. The terminal velocity can achieve about 9 m/s when the dimameter reach 2.4 mm. On the other hand, when the particle size increases, the convective heat transfer coefficient decreases. The convective heat transfer coefficient becomes as low as 222 W/(m².K) when the particle

diameter is 2.4 mm. Therefore, with higher falling velocity and lower heat transfer efficiency at larger size of particles, it is more difficult for the coarser droplet to completely solidify in a prilling tower.



FIGURE 1 Terminal velocity at different sizes of urea particles

Temperature profiles

The temperature at the center of the droplets for various diameters are shown in **Error! Reference source not found.** From the figure, it can be observed that the solidification takes a longer time for the coarser particle. For the particle with a diameter of 2.4 mm, the center just reaches the freezing point and is still in the liquid phase after 50 s. For the smaller particles, such as the particle with a diameter less than 2.0 mm, the centers are completely transformed to solid and cooled after 50 s.



Figure 3. Temperature at the center of the droplets versus time for variuos particle diameter.

For the particle with a diameter of 1.6 mm (typical size in urea prilling towers), the temperature distribution inside the particle at different times is shown in **Error! Reference source not found.**





The terminal velocity, the required time and the minimum height for the complete solidification are summarized in **Error! Reference source not found.** The typical height of prilling towers is about 50 meters. Therefore, it can be roughly estimated that the particles with diameters less than 1.2 mm can solidify completely. For particles with sizes in the range of 1.2 mm - 2.0 mm, the solidification is partially complete.

Table 2. The terminal velocity, the required time and the minimum height for the complete solidification.

| Particle diameter | Required time for complete | Terminal | Height |
|-------------------|----------------------------|----------------|--------|
| [mm] | solidification [s] | velocity [m/s] | [m] |
| 0.6 | 3.86 | 2.16 | 8.34 |
| 0.8 | 6.62 | 3.01 | 19.92 |
| 1 | 10.03 | 3.79 | 38.03 |
| 1.2 | 14.07 | 4.52 | 63.60 |
| 1.4 | 18.78 | 5.20 | 97.68 |
| 1.6 | 24.08 | 5.84 | 140.64 |
| 1.8 | 30.00 | 6.44 | 193.21 |
| 2 | 36.57 | 7.00 | 256.12 |

CONCLUSIONS

The solidification of the urea droplets has been considered as a two-phase Stefan problem with convective flux boundary conditions. The problem was solved numerically using the enthalpy method. The temperature distribution is smooth for various particle diameters. From the results, the minimum height of the tower for complete solidification of each particle diameter can be approximately estimated. For the typical height of 50 meters, the droplets smaller than 1.2 mm are entirely solidified.

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