

# Two-Phase Stefan Problem for the Modeling of Urea Prilling Tower <sup>†</sup>

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**Abstract:** In the manufacturing of granular urea and ammonium nitrate, the technique of prilling is utilized rather frequently. The liquid droplets that are produced fall along the tower to the ground and become solid due to the removal of heat by the cooling air, which flows in the opposite direction of the stream. Generally, three sequential thermal intervals for the solidification of urea droplets are considered: the cooling of liquid drops, solidification at the freezing temperature of the liquid phase, and cooling of complete solid particles. In this study, the solidification of the urea droplets was considered as a two-phase Stefan problem with a 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.

**Keywords:** prilling; urea; spray crystallization; mathematical modeling; simulation; Stefan problem; free boundary problem



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## 1. Introduction

The technique of prilling is frequently employed in the granular urea and ammonium nitrate industry. The fundamental procedure involves spraying hot liquid from the top of a tower. Simultaneously, a stream of ambient air is collected and supplied from the bottom of the tower. The generated particles fall against the flow of air and solidify due to the heat transfer to the air. The advantages of the process are the nearly uniform shape and size of the produced particles.

In practice, incomplete solidification can readily cause operational issues for prilling towers. As a consequence of inefficient solidification, a low-quality structure is produced, resulting in decreased productivity and profits. Despite the significance of the procedure, there have been few studies on the modeling of a prilling tower. In the work 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 with 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: the cooling of liquid drops, solidification at the 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 particle should change gradually with time. Therefore, the solidification of urea particles is examined in this report as a two-phase Stefan problem in which heat fluxes occur in both liquid and solid phases. The cooling and solidification of the particles are regarded as a single process, from liquid droplets to solid particles, rather than as three distinct stages. Concerning the hydrodynamics of the process, it is supposed that particles attain terminal velocity rapidly. This velocity is required for the estimation of the convective heat transfer coefficient. The boundary condition is the heat flux from the particle to the air by convection.

## 2. 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.

## 3. Terminal Velocity of the Urea Particles

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

$$F_G = F_B + F_D \quad (1)$$

$$\frac{1}{6}\pi d_p^3 \rho_p g = \frac{1}{6}\pi d_p^3 \rho_a g + \frac{1}{2}\rho_a C_D \pi \frac{d_p^2}{4} (v_t + v_a)^2 \quad (2)$$

Therefore, the terminal velocity can be obtained as

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

in which  $v_t$  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,  $\rho_p$  is the particle density ( $\text{kg/m}^3$ ),  $\rho_a$  is the density of the air ( $\text{kg/m}^3$ ),  $d_p$  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 Reynold number. The correlation proposed in Brown and Lawler [5], which fits the range of a Reynold number up to  $2 \times 10^5$ , is used in this study:

$$C_D = \frac{24}{\text{Re}_p} \left(1 + 0.150 \text{Re}_p^{0.681}\right) + \frac{0.407}{1 + \frac{8710}{\text{Re}_p}} \quad (4)$$

where  $\text{Re}_p$  is the particle Reynold number as

$$\text{Re}_p = \frac{d_p (v_t + v_a) \rho_a}{\mu_a} \quad (5)$$

in which  $\mu_a$  is the viscosity of air in  $\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$ .

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 were calculated. After this, the new value of terminal velocity was estimated from Equation (3) and compared to the current terminal velocity. If the difference is small (less than  $1 \times 10^{-8}$ ), the procedure stops and terminal velocity is obtained. If the difference is still high, the procedure is repeated.

#### 4. Heat Transfer as a Two-Phase Stefan Problem

Consider a spherical liquid urea droplet, as shown in Figure 1.

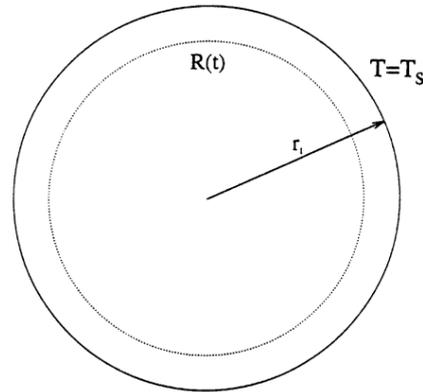


Figure 1. Configuration of the droplet.

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.

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

$$\frac{\partial T}{\partial t} = \frac{\alpha_s}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) , \quad R(t) \leq r < r_p \tag{7}$$

where  $\alpha_j = \frac{k_j}{\rho_j c_j}$ ,  $k_i$ ,  $\rho_i$ , and  $c_i$  ( $j = s, l$ ) 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 is

$$k_s \left( \frac{\partial T}{\partial r} \right)_{R(t)} - k_l \left( \frac{\partial T}{\partial r} \right)_{R(t)} = L \rho_s \frac{dR(t)}{dt} \tag{8}$$

where  $L$  is the latent heat of freezing, respectively.

The initial and boundary conditions are given by

$$T(r, 0) = T_i, \quad 0 \leq r \leq r_1 \tag{9}$$

$$-k \frac{dT}{dr} \Big|_{r=r_p} = h (T_{r=r_p} - T_a), \quad t > 0 \tag{10}$$

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

$$Nu = 2 + 0.6 Re_p^{0.5} Pr^{0.33} \tag{11}$$

where  $Nu = \frac{h d_p}{k_g}$  is the Nusselt number;  $Pr = \frac{c_{p,g} \mu_g}{k_g}$  is the Prandtl number; and  $k_g$ ,  $c_{p,g}$ , and  $\mu_g$  are the thermal conductivity, specific heat capacity, and viscosity of air, respectively.

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

## 5. 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 the urea particle diameter (1 mm to 2.2 mm), the terminal velocity, which is determined with Equation (3), is used as the steady velocity of the particle. The convective heat transfer coefficient is then obtained with Equation (11). The heat transfer coefficient  $h$  is used as the input and the solidification of the urea droplet is considered as a two-phase Stefan problem with the 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 Table 1.

**Table 1.** Parameters and values of the prilling process.

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

## 6. Results and Discussions

### 6.1. Terminal Velocity and Convective Heat Transfer Coefficient

The terminal velocity and convective heat transfer coefficient of urea particles at different sizes when the air velocity is 0.63 m/s are shown in Figure 2. From the figure, it can be seen that the terminal velocity increases with an increase in particle diameter. The terminal velocity can achieve about 9 m/s when the diameter reaches 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/(m<sup>2</sup>·K) when the particle diameter is 2.4 mm. Therefore, with a higher falling velocity and lower heat transfer efficiency at a larger size of particles, it is more difficult for a coarser droplet to completely solidify in a prilling tower.

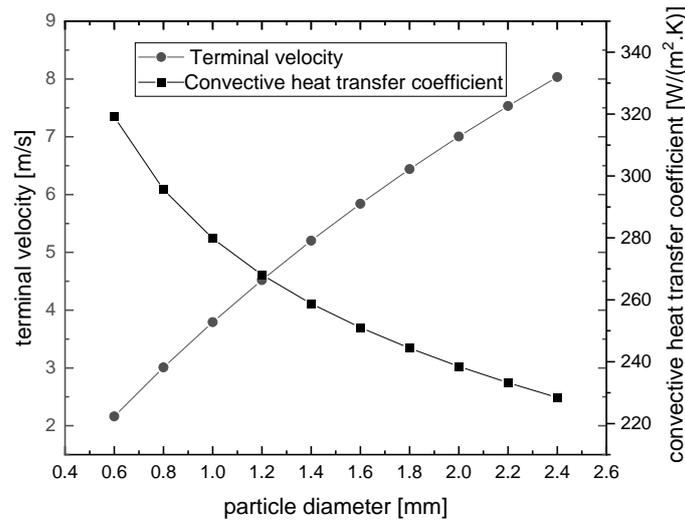


Figure 2. Terminal velocity at different sizes of urea particles.

6.2. Temperature Profiles

The temperature at the center of the droplets for various diameters is shown in Figure 3. 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 a solid and cooled after 50 s.

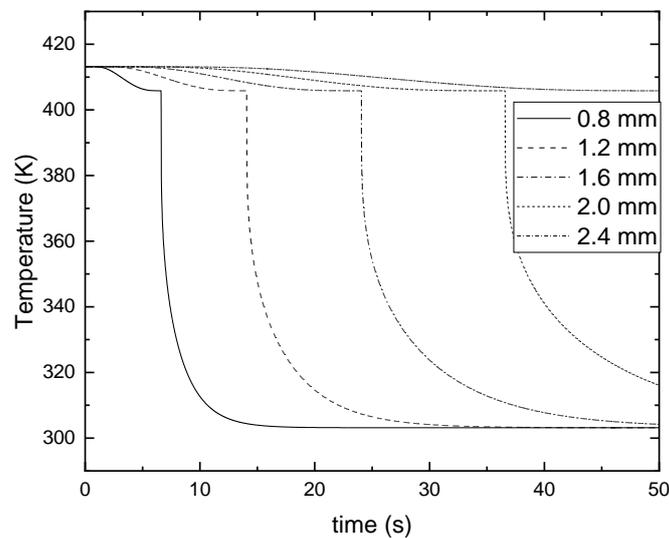
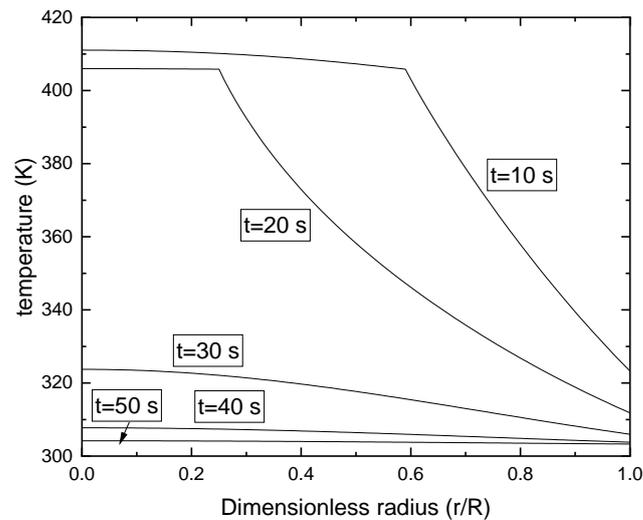


Figure 3. Temperature at the center of the droplets versus time for various particle diameters.

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 Figure 4.

The terminal velocity, the required time, and the minimum height for the complete solidification are summarized in Table 2. The typical height of prilling towers is about 50 m. 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 1.2–2.0 mm, the solidification is partially complete.



**Figure 4.** Temperature distribution versus time of a 1.6 mm particle.

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

Particle Diameter (mm)	Required Time for Complete Solidification (s)	Terminal Velocity (m/s)	Height (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

## 7. Conclusions

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

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**Data Availability Statement:** The data that support the findings of this study are available from the corresponding author, T.-A.N., upon reasonable request.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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