Numerical Simulations of Solidification Characteristics of Molten Slag Droplets in Radiant Syngas Coolers for Entrained-Flow Coal Gasification

Bo Wang, Jianyong Qiu, Qinghua Guo, Yan Gong, Jianliang Xu, and Guangsuo Yu

ABSTRACT: The high-temperature syngas and molten slag droplets discharged from entrained-flow coal gasifiers contain a large amount of heat energy, which can be efficiently recovered by radiant syngas coolers (RSCs). However, it is hard to know the solidification degree of molten slag droplets at the outlet of an RSC during industrial operations. In this work, the industrial-scale RSC and molten slag droplet models are established to predict the solidification degree of slag droplets at the outlet of the RSC. Then, the effects of slag diameter, syngas flow field, slag initial temperature, slag porosity, and slag pore structure are investigated by numerical simulations, and residence time as well as complete solidification time are calculated by coupling of a discrete-phase model and a solidification model. The results indicate that as the slag droplet diameter increases, the residence time of the slag droplet shortens, but the complete solidification time increases. When the slag droplet diameter is greater than or equal to 3.0 mm, the complete solidification time is larger than the residence time, and the slag droplet cannot solidify completely at the outlet of the RSC. The solidification degree in the windward zone is greater than that in the leeward zone. Although the slag initial temperature has little effect on the solidification, a lower slag initial temperature is still conducive to a greater solidification degree. Additionally, the pore structure facilitates solidification, and the promoting effect of penetrated pores is more remarkable than that of closed pores. A larger porosity is also beneficial to accelerate the solidification of molten slag droplets and increase the solidification degree.

1. INTRODUCTION

Coal gasification technology plays an important role in the high efficiency and clean utilization of coal resources.1 Nowadays, entrained-flow coal gasification has become the mainstream technology due to its wide adaptability of coal types, high gasification efficiency, high effective syngas (CO + H2, syngas) yield, and low pollution emission.2−4 The syngas produced by coal gasification contains plenty of sensible heat due to the high-temperature operating conditions of an entrained-flow gasifier.5 Thus, it is necessary to recover this kind of heat to improve the gasification efficiency.

A radiant syngas cooler (RSC) is regarded as a crucial heat-recovery device in entrained-flow coal gasification units.6−8 The syngas produced from the gasifier enters the RSC, carrying a great number of fly ash particles and molten slag droplets, and all of them transfer heat to the membrane wall.5,10 However, the molten slag droplets may collide with the membrane wall and the bottom cone of the RSC, and they are more likely to deposit on the wall when they are in the molten state.11,12 The slag deposition and accumulation will strongly affect the heat-recovery efficiency and the safety operation of the RSC.13,14 Hence, it is of great importance to gain a clear understanding of the heat transfer and solidification characteristics of molten slag droplets in the industrial RSC.

It is hard to carry out experimental investigations on the industrial RSC due to its harsh operating conditions and large scales. Therefore, in previous studies, the numerical simulation method was used to explore the solidification characteristics of molten slag droplets. Xing et al.15 established a two-dimensional model to investigate the heat-transfer process of steel slag granules cooled in nitrogen, and Sun et al.16 simulated the solidification process of slag droplets cooled in air by a three-dimensional model. However, the variable thermal physical properties and the temperature range during the phase change process were not considered in both of their studies. However, Liu et al.17 studied the solidification behaviors of molten slag droplets with the temperature...
model and concluded that variable thermal physical properties affect the cooling process of blast furnace (BF) slag droplets significantly. The subsequent studies took the effects of these factors into account. Zhu et al.18 and Gao et al.19 developed one-dimensional and two-dimensional models, respectively, to investigate the crystallization characteristics of BF slag droplets under air-cooling conditions. Peng et al.20 and Hu et al.21 explored the solidification process of the single and multiple slag droplets by using the combined evaporation and solidification models, respectively, and the same conclusion was given that the solidification time of slag droplets reduced as humidity ratio and air velocity increased. However, among most of these research studies, molten slag droplets transfer heat with the ambient air, which deviates far from the operating conditions of an industrial RSC. The heat transfer process in an industrial RSC is more complex, and the syngas consists of various kinds of gases. Molten slag droplets exchange heat not only with the high-temperature syngas but also with the membrane wall. In addition, it is noted that the thermal physical properties of syngas are quite different from those of air. What is more, none of the above studies have considered the effect of slag porosity on solidification characteristics. However, the devolatilization and combustion gasification reactions as well as slag droplet collision and adhesion could cause pores in molten slag droplets.22

In the present work, a two-dimensional industrial-scale RSC model is established to obtain the flow and temperature fields in an RSC, and the residence time of molten slag droplets in the RSC is obtained by a discrete-phase model. Additionally, three-dimensional molten slag droplet models are developed to investigate the solidification characteristics of molten slag droplets in the RSC. Furthermore, effects of different operating conditions on solidification characteristics are also discussed in this work, and the slag porosity is taken into consideration. The residence time, liquid fraction distribution, and complete solidification time are presented in simulation results, and the solidification degree of molten slag droplets at the outlet of the RSC can be predicted by comparing the residence time and complete solidification time. The simulation results are beneficial to achieve a better understanding of the solidification process of the molten slag droplet in RSCs as well as provide guidance for the safe slag discharge of RSCs.

2. MODEL DESCRIPTION

2.1. Industrial-Scale RSC Model. The high-temperature syngas-carrying molten slag droplets flow into the RSC from the connection of the gasification chamber and the RSC. The syngas releases sensible heat and the molten slag droplets release both sensible heat and latent heat, and they flow out of the RSC after the heat transfer process. The main body of the industrial RSC is a cylinder, which has a rotating body structure. Therefore, the axisymmetric model can be used for the numerical simulation of the industrial-scale RSC in Fluent software. Accordingly, the physical model of the industrial-scale RSC is simplified as a two-dimensional model in this study, as shown in Figure 1, where \( R_\text{in} \) is the radius of the inlet, \( R_\text{out} \) is the radius of the outlet, and \( D \) is the diameter of the straight part of main body. \( H_\text{total} \) represents the total height of the RSC, and \( H_\text{body} \) represents the height of the membrane wall of the RSC. The angles of the upper cone and the bottom cone are 30 and 60°, respectively.

![Figure 1. Two-dimensional model of an industrial-scale RSC.](https://doi.org/10.1021/acsomega.1c02406)

2.2. Molten Slag Droplet Models. Based on previous research studies,23,24 the numerical simulations are carried out under the following assumptions:

1. Molten slag droplets are regarded as spherical during the whole process.
2. The phase change temperature is not a constant, and the liquid zone, mushy zone, and solid zone occur successively during the solidification process.
3. Radiant heat transfer of the molten slag droplet is considered.
4. Natural convection inside the molten slag droplet is neglected.

2.2.1. Nonporous Molten Slag Droplet Model. In this work, the solidification characteristics of nonporous molten slag droplets are discussed first, and a three-dimensional spherical molten slag droplet model is established. The internal structure and model schematic of the nonporous molten slag droplet cooled inside the industrial-scale RSC are shown in Figure 2. The establishment of this model is mainly based on the simplification method of a fixed coordinate system proposed in Section 4.1, which simplifies the calculation domain to a single molten slag droplet and the surrounding fluid. After simplification, part of the simulation results of the RSC model in Figure 1 can be used as the boundary conditions of this model.

2.2.2. Porous Molten Slag Droplet Models. Entrained-flow coal gasification takes place at a very high temperature and pressure in a reducing atmosphere. Under such working conditions, the compositions of syngas such as \( \text{H}_2 \) and \( \text{CO} \) show a high solubility in the molten slag,27 and these dissolved gases will be released to form small bubbles in the molten slag during the cooling process in the RSC. These bubbles pull up the surrounding viscous molten slag and form closed pores in the molten slag. The released gases move in all directions and form irregular pores. However, in order to simplify the simulations, it is assumed that the released gases move regularly in six directions (up, down, front, rear, left, and right), and the closed pore is reasonably simplified as a three-dimensional cross-shaped pore, as shown in Figure 3. When
the temperature of the molten slag decreases, the surface tension of these bubbles in the molten slag increases, the radius of curvature decreases, and the additional pressure increases. These bubbles would burst when the additional pressure is too large to keep a single bubble in balance. With more and more small bubbles bursting, larger bubbles will be formed, which is prone to cause a stress concentration. Finally, the molten slag droplet breaks when the stress exceeds the yield limit of the molten slag droplet. The breakup of the molten slag droplet leads to the formation of slit-shaped pores.28,29 The slit-shaped pore is simplified as the penetrated pore model as shown in Figure 3.

The closed pore structure and penetrated pore structure are considered to improve molten slag droplet models. The pores of a closed pore structure are inside the molten slag droplet and the external syngas cannot contact the pores directly. The pores of the penetrated pore structure directly contact the external syngas and the syngas can flow through the penetrated pores. In addition, the molten slag droplet porosity ($\phi$) can be estimated as

$$\phi = \frac{v_p}{v_p + v_m} \times 100\%$$

where $v_p$ and $v_m$ represent the pore volume and the matrix volume, respectively.

2.3. Mathematical Models. 2.3.1. Turbulence Model. Ni et al.31,32 have validated that the simulation results obtained by a realizable $k$–$\varepsilon$ model are consistent with the experimental results. Therefore, the realizable $k$–$\varepsilon$ model is adopted to simulate the gas phase in this work, and the corresponding equations are shown below

$$\frac{\partial (\rho k)}{\partial t} + \frac{\partial (\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \mu + \frac{\mu_t}{\sigma_k} \frac{\partial k}{\partial x_i} \right] + G_k + G_b + Y_M + S_k$$

$$\frac{\partial (\rho \varepsilon)}{\partial t} + \frac{\partial (\rho \varepsilon u_i)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \frac{(\mu + \frac{\mu_t}{\sigma_\varepsilon}) \frac{\partial \varepsilon}{\partial x_i}}{\sigma_\varepsilon} \right] + \rho C_1 S_\varepsilon - \rho C_2 \frac{\varepsilon}{k} C_{\varepsilon} G_b + S_\varepsilon$$

where $G_k$ is the generation of turbulence kinetic energy due to the mean velocity gradients, $G_b$ represents the generation of turbulence kinetic energy due to buoyancy, $Y_M$ is the contribution of the fluctuating dilatation in compressible turbulence to the overall dissipation rate, $C_2$ and $C_1$ are constants, and $\sigma_k$ and $\sigma_\varepsilon$ are the turbulent Prandtl numbers for $k$ and $\varepsilon$, respectively. $S_k$ and $S_\varepsilon$ are user-defined source terms.

2.3.2. Heat-Transfer Model. The simulations of radiant heat transfer are complicated. Previous studies have indicated that both the P1 model and the discrete ordinates model are suitable for the radiant heat transfer calculation in the industrial RSC,33,34 but the calculation speed of the P1 model is faster than that of the discrete ordinates model.35 Therefore, the P1 model is employed to calculate the radiant heat transfer in the industrial RSC. The equations for the P1 model are as follows

$$-\nabla \cdot q_r = -\nabla \cdot (\Gamma \cdot G) = \alpha G - 4\alpha \sigma T^4$$

$$\Gamma = [3(\sigma + \sigma_k)]^{-1}$$

$$\nabla \cdot (\Gamma \cdot G) = -\alpha G + 4\alpha \sigma T^4 = S_G$$

where $q_r$ is the radiation flux, $G$ is the incident radiation, $\sigma_k$ is the scattering coefficient, $\alpha$ represents the absorption coefficient, $\sigma$ is the Stefan–Boltzmann constant [$\sigma = 5.67 \times 10^{-8} \text{ W/(m}^2\cdot\text{K}^4)$], and $S_G$ is the user-defined radiation source.

2.3.3. Species Transport Model. The species transport model is adopted to calculate the multi-component gas, and the syngas contains various components, including CO, H$_2$, CO$_2$, H$_2$O, and so forth. Therefore, the species transport model is selected to predict the local mass fraction of each component in this work. The general form of the conservation equation is shown below
\[
\frac{\partial}{\partial t}(\rho Y_i) + \nabla \cdot (\rho \bar{v} Y_i) = -\nabla \cdot \mathbf{J}_i + R_i + S_i
\]

(7)

where \( Y_i \) and \( R_i \) are the volume concentration and net rate of production of species \( i \), respectively, \( \mathbf{J}_i \) is the diffusion flux of species \( i \), and \( S_i \) is the rate of creation by addition from a user-defined source.

2.3.4. Discrete-Phase Model. Molten slag droplets in an RSC can be regarded as discrete-phase particles, and their trajectories are predicted by integrating the force balance on the particles in a Lagrangian reference frame. The force balance on the particles can be written as

\[
\frac{d\mathbf{u}_p}{dt} = \frac{\mathbf{u} - \mathbf{u}_p}{\tau_i} + \frac{\bar{g}(\rho_p - \rho)}{\rho_p} + \mathbf{F}_{\text{nu}}
\]

(8)

where \( \mathbf{F}_{\text{nu}} \) is an additional acceleration term, \( \tau_i \) is the molten slag droplet relaxation time, \( \mathbf{u} \) is the syngas velocity, \( \mathbf{u}_p \) is the slag droplet velocity, \( \mu \) is the viscosity of syngas, \( \rho \) is the syngas density, \( d_p \) is the slag droplet diameter, and \( Re \) is the Reynolds number.

2.3.5. Solidification Model. The solidification process of a molten slag droplet is investigated by the enthalpy-porosity technique. The mushy zone is modeled as a "pseudo" porous medium in which the porosity decreases from 1 to 0 as the material solidifies. When the material has fully solidified, the porosity becomes zero. The following equations are applied to calculate the solidification process of a molten slag droplet, and these equations are applicable to all three models proposed in Section 2.2.

The energy equation is given below

\[
\frac{\partial (\rho H)}{\partial t} + \nabla \cdot (\rho \bar{v} H) = \nabla \cdot (\kappa \nabla T) + S_{\text{source}}
\]

(11)

where \( H \) is the enthalpy, \( \rho \) is the density, \( \bar{v} \) is the fluid velocity, and \( S_{\text{source}} \) is the source term.

The enthalpy \( H \) is calculated as the sum of sensible heat and latent heat, and the corresponding equations are shown below

\[
H = h + \Delta H = h_{\text{ref}} + \int_{t_{\text{ref}}}^{t} \varepsilon_p dT + \beta L
\]

(12)

where \( h \) and \( \Delta H \) are the sensible and latent heat, respectively, \( h_{\text{ref}} \) and \( T_{\text{ref}} \) are the reference enthalpy and temperature, respectively, \( \varepsilon_p \) is the specific heat at a constant pressure, and \( \beta \) is the liquid fraction.

The liquid fraction \( \beta \) can be defined as

\[
\beta = \begin{cases} 
0 & \text{if } T < T_{\text{solidus}} \\
\frac{T - T_{\text{solidus}}}{T_{\text{liquidus}} - T_{\text{solidus}}} & \text{if } T_{\text{solidus}} \leq T \leq T_{\text{liquidus}} \\
1 & \text{if } T > T_{\text{liquidus}}
\end{cases}
\]

(13)

where \( T_{\text{solidus}} \) and \( T_{\text{liquidus}} \) are the solidus and liquidus temperatures of the molten slag droplet, respectively.

The momentum sink in the mushy zone is shown below

\[
S = \frac{(1 - \beta)^2}{(\beta + \varepsilon)} A_{\text{mush}}(\bar{v} - \bar{v}_p)
\]

(14)

where \( \varepsilon \) is a small constant (0.001) to prevent the denominator from being equal to zero, \( A_{\text{mush}} \) is the mushy zone constant, and \( \bar{v}_p \) is the pull velocity.

3. BOUNDARY CONDITIONS AND MODEL VALIDATION

3.1. Operating Conditions and Materials. The operating data of an industrial RSC are adopted for numerical simulations. The inlet syngas components and operating conditions are shown in Tables 1 and 2, respectively.

Table 1. Inlet Syngas Components of the Industrial RSC

| Components       | Mole Flow Rate (k mol/h) | Mole Fraction (%) |
|------------------|--------------------------|-------------------|
| H₂               | 2216.03                  | 27.90             |
| CO               | 3164.36                  | 39.85             |
| CO₂              | 1106.63                  | 13.93             |
| H₂O              | 1454.58                  | 18.32             |

Table 2. Operating Conditions of the Industrial RSC

| Variables                     | Value |
|-------------------------------|-------|
| Syngas and particle inlet temperature (K) | 1573  |
| Inlet water temperature of the water wall tube (K) | 592.0 |
| Operating pressure (MPa)       | 6.5   |
| Inlet syngas flow rate (m³/h)  | 1600  |
| Inlet slag mass low rate (m³/h) | 3.16  |

Table 3. Physical Properties of the Molten Slag Droplet

| Parameters | Solid Phase | Liquid Phase |
|------------|-------------|--------------|
| L (J/kg)   | 611,483     | 1423         |
| T_{solidus} (K) | 1283     | -            |
| T_{liquidus} (K)  | 1283     | -            |
| \varepsilon_{slag} | 0.80      | -            |

Thermal physical parameters of the molten slag droplet are listed in Table 3, where \( L \) is the latent heat and \( \varepsilon_{slag} \) is the emissivity of a molten slag droplet. We assume that the emissivity of a molten slag droplet is a constant, and the emissivity of a molten slag droplet is determined by referring to the formula from Gao’s research;19 the calculation formula is written as

\[
\varepsilon_{slag} = 4 \times 10^{-7} (T_{solidus} - 273.15)^2
\]

\[
- 7 \times 10^{-4} (T_{solidus} - 273.15) + 1.087
\]

(15)

In this work, the value of \( \varepsilon_{slag} \) is calculated as 0.8.

The density \( \rho \), specific heat \( c_p \), and thermal conductivity \( \lambda \) are regarded as the functions of slag droplet temperature \( T \), as shown below

\[
\rho(T) = \begin{cases} 
2750 & T > T_{\text{liquidus}} \\
3360 - 0.64T & T_{\text{solidus}} \leq T \leq T_{\text{liquidus}} \\
2840 & T < T_{\text{solidus}}
\end{cases}
\]

(16)
Due to the complex reactions and heat transfer conditions in the entrained-flow gasifier, the diameter distribution of the molten slag droplets discharged from the gasifier tends to be uneven. Therefore, the solidification characteristics of molten slag droplets with different sizes should be considered. Based on the liquid slag thickness distribution at the outlet of the gasification chamber in Zhang’s work,\textsuperscript{40,41} the sizes of molten slag droplets are selected as 1.0, 1.5, 2.0, 3.0, and 5.0 mm in this work. It is worth noting that the effects of size distribution and released latent heat of molten slag droplets are ignored because of the very small volume fraction (about $10^{-5}$) of molten slag droplets.

### 3.2. Grid Independence Validation

Grid independence is necessary to eliminate the effects of grid numbers on the simulation results. In this work, nonporous molten slag droplet models with different grid numbers (38,744, 52,872, 69,632, 83,589, 116,256, 156,512, 184,960, and 236,328) are established.

Figure 4 demonstrates the influence of the grid number on the temperature distribution at $R = 0$ ($R$ is the radius of the molten slag droplet), and it can be seen that the liquid fraction values are independent of the grid numbers when the grid number is over 156,512. Therefore, the grid number of the numerical model is determined to be 184,960, which satisfies the grid independence.

### 3.3. Model Validation

The present model is validated by the experimental results from Ferreira et al.\textsuperscript{42} In their work, solidification experiments were carried out with alloys of two metallic systems, and the experimentally obtained temperatures were also given, which could be used as a validation for the present model. Table 4 shows the thermophysical properties used in Ferreira’s experimental analysis.\textsuperscript{42}

**Table 4. Thermophysical Properties Used in Ferreira’s Experimental Analysis**

| parameters             | solid | liquid |
|------------------------|-------|--------|
| specific heat (J/(kg·K)) | 1092  | 1059   |
| density (kg/m³)       | 2654  | 2488   |
| latent heat (J/kg)    | 381,773 | 921    |
| solidus temperature (K)| 821   |        |
| liquidus temperature (K)|       | 937    |
| initial slag temperature (K)|   |        |
| thermal conductivity (W/(m·K)) | 193 | 89     |

Figure 5 shows the comparison of temperature distribution at a position of $y = 10$ mm between the present simulation value and the experimental value from Ferreira et al.

### 4. RESULTS AND DISCUSSION

#### 4.1. Flow-Field Characteristics in an RSC

Figure 6 shows the velocity distribution and temperature distribution of an RSC. It could be seen that there is a jet core region at the top of the RSC, and a certain range of recirculation areas occurs on both sides of the inlet jet, which is similar to the results predicted by Li et al.\textsuperscript{43} The syngas temperature at the central jet of the industrial RSC is the highest, and it gradually decreases along the flow direction due to the convectional and radiant heat transfer.

Temperature distribution along the axis direction is required for the subsequent calculation of molten slag droplets. Therefore, it (as shown in Figure 7) is obtained by the simulation results of the temperature field, and the corresponding fitting polynomial ($R$-square, 0.9960) can be expressed as

$$T(h) = 1564.74 + 23.6h - 8.86h^2 + 0.55h^3 - 0.0097h^4 \quad (0 \leq h \leq 24.09)$$  \hspace{1cm} (19)

where $h$ is the height along the axis direction and $T(h)$ is the temperature along the axis direction.

During industrial operations, molten slag droplets would be charged out of an RSC due to different kinds of forces,
including drag force and gravity. If the industrial RSC is taken as the fixed coordinate system, a dynamic mesh should be used to simulate the molten slag droplet, which will lead to an excessive computational cost. Therefore, in this work, the molten slag droplet is chosen as the fixed coordinate system to reduce computational burden. By applying the discrete-phase model in the Fluent software, the relationship between the falling height \( h(t) \) and the falling time of the molten slag droplet is obtained. Then, the relationship between the temperature \( T(t) \) of the syngas around the molten slag droplet and the falling time can also be calculated. Through the change of the fixed coordinate system, the gas–solid two-phase flow is simplified as the relative movement of the syngas phase to the molten slag droplet phase, which means that the molten slag droplet is stationary and the surrounding syngas flows over the surface of the molten slag droplet with the temperature \( T(t) \). Thus, the residence time of slag droplets with different diameters should be discussed.

### 4.2. Residence Time of Molten Slag Droplets

The residence time of molten slag droplets is calculated by a discrete phase model. The numerical results are shown in Figure 8. It is found that the residence time of molten slag droplets decreases as the slag diameter increases, which can be associated with the flow characteristics of different droplets. Large slag droplets tend to flow out of the RSC directly from the central flow channel, but the small slag droplets are more likely to be sucked up by the recirculation regions. Therefore, a longer residence time is caused in the recirculation regions for small droplets.

Furthermore, the fitting equations of falling height along the axis direction and residence time of different molten slag droplets are obtained. The equations are summarized in Table 5, where \( t \) is the residence time and \( h(t) \) is the falling height along the axis direction. Thus, the relationship between flow field temperature and slag droplet residence time can be acquired by combining eq 19 and these equations, which are listed in Table 6. It is noted that these equations in Table 6 will be imported into Fluent software as user-defined functions and be used as boundary conditions for simulation.

### 4.3. Solidification Process of Nonporous Molten Slag Droplets

In this work, the solidification characteristics of nonporous molten slag droplets are investigated first. The liquid fraction and temperature distribution of a molten slag with a diameter of 1.0 mm are shown in Figures 9 and 10, respectively. In Figure 9, it can be observed that the solidification behavior first occurs on the surface of the molten slag droplet and then gradually penetrates toward the central area layer by layer. Furthermore, it can be seen that the solidification degree on the windward side is greater than that on the leeward side. Similarly, temperature distribution has the same distribution trend in Figure 10, and the temperature along the axis direction.

### Table 5. Equations of Falling Height and Residence Time

| Diameter (mm) | Equations | R-square |
|---------------|-----------|----------|
| 1.0           | \( h(t) = 2.96 + 4.05t - 0.177t^2 \) | 0.9900   |
| 1.5           | \( h(t) = 2.48 + 5.34t - 0.30095t^2 \) | 0.9913   |
| 2.0           | \( h(t) = 2.13 + 6.40t - 0.42493t^2 \) | 0.9928   |
| 3.0           | \( h(t) = 1.50 + 8.17t - 0.67874t^2 \) | 0.9946   |
| 5.0           | \( h(t) = 0.64 + 10.36t - 1.03038t^2 \) | 0.9972   |
the windward zone is lower than that in the leeward zone. This can be attributed to the difference in the syngas flow characteristics around the slag droplet, as shown in Figure 11. In the windward zone, the syngas velocity on the surface of the particles is large and there is a large velocity gradient due to the influence of the syngas flow. However, in the leeward zone, the syngas velocity is close to zero due to the appearance of the vortex. Therefore, in the windward zone with a higher velocity gradient, the heat exchange is stronger and the solidification degree is greater.

4.4. Effect of Slag Droplet Diameter. In industrial operations, the diameter of molten slag droplets is often uneven, which is affected by the type of raw coal, gasification conditions, operating loads, and so forth. Therefore, it is sensible to consider the solidification characteristics of molten slag droplets with different diameters.

Figure 12 presents the liquid fraction distribution of different slag droplets at the slag center (R = 0), and it can be seen that the variation of the liquid fraction slows down and the complete solidification time increases as the slag diameter increases from 1.0 to 5.0 mm. This phenomenon can be attributed to the fact that the heat-carrying capacity of the molten slag droplet increases with increasing slag droplet diameter but the specific surface area and the heat transfer coefficient decrease. Consequently, the solidification performance gets worse. It is noted that as the slag diameter increases, the curve of liquid fraction distribution becomes less smooth. This is mainly because the distance from the slag center to the surface increases, which leads to a higher internal thermal resistance. Thus, the latent heat of slag solidification is more difficult to release in time, resulting in less smooth variations of liquid fraction distribution.

A comparison between the residence time and complete solidification time is displayed in Figure 13. The liquid fraction distribution of the Y-section profile and the Z-section profile at the outlet of an RSC are also shown in Figure 13. It can be observed that the complete solidification time of the slag

| diameter (mm) | 1.0 | 1.5 | 2.0 | 3.0 | 5.0 |
|--------------|-----|-----|-----|-----|-----|
| T(1)         | $1568.93 - 66.14t - 74.28t^2 + 33.97t^3 - 6.32t^4 + 0.61t^5 - 3.22 \times 10^{-2}t^6 + 8.73 \times 10^{-4}t^7 - 9.56 \times 10^{-6}t^8$ | $1575.59 - 61.82t - 147.87t^2 + 82.33t^3 - 19.40t^4 + 2.40t^5 - 1.62 \times 10^{-1}t^6 + 5.65 \times 10^{-2}t^7 - 7.96 \times 10^{-3}t^8$ | $1578.95 - 70.56t - 147.42t^2 + 82.72t^3 - 19.61t^4 + 2.40t^5 - 1.62 \times 10^{-1}t^6 + 5.65 \times 10^{-2}t^7 - 7.96 \times 10^{-3}t^8$ | $1575.59 - 61.82t - 147.87t^2 + 82.33t^3 - 19.40t^4 + 2.40t^5 - 1.62 \times 10^{-1}t^6 + 5.65 \times 10^{-2}t^7 - 7.96 \times 10^{-3}t^8$ | $1576.14 - 61.99t - 147.42t^2 + 82.72t^3 - 19.61t^4 + 2.40t^5 - 1.62 \times 10^{-1}t^6 + 5.65 \times 10^{-2}t^7 - 7.96 \times 10^{-3}t^8$ |

Table 6. Equations of Temperature and Residence Time
be found that the variation of the liquid fraction slows down as the slag droplet initial temperature increases. From the local enlarged drawing (b), it can be observed that the complete solidification times of molten slag droplets with different inlet temperatures are very close to each other. Therefore, it can be concluded that although the effect of slag droplet initial temperature is small, a lower slag droplet initial temperature is still conducive to obtaining a faster solidification rate.

4.6. Effect of Slag Droplet Porosity. The pores in molten slag droplets will decrease the liquid slag content. The thermal physical properties of syngas in those pores are quite different from that of molten slag, which affects the heat-transfer process greatly. Therefore, it is sound to consider the proportion of pores in the molten slag droplet during the heat transfer process, which can be characterized by the slag porosity. In this work, the effect of slag porosity on solidification is discussed based on the closed pore structure model.

Figure 15 sketches the relationship between the complete solidification time and slag porosity. It can be found that the complete solidification time shortens significantly as the slag porosity increases. It is noted that as the slag porosity increases, the decrease of complete solidification time in interval A is greater than that in interval B, indicating that the effect of slag porosity on solidification is not only caused by the slag content. The variation of complete solidification time can also be associated with the fact that the thermal conductivity of syngas is far less than that of molten slag. The porosity is larger and syngas content increases in interval B, resulting in a larger internal heat resistance.

The increasing interval heat resistance hinders the solidification; thus, the variation of complete solidification time is not as obvious as that in interval A.

4.7. Effect of the Slag Pore Structure. Different pore structures cause the slag droplets to have different specific surface areas, which affects the flow characteristics of the syngas and the heat transfer process. Therefore, it is reasonable to discuss the effect of slag pore structures on the solidification characteristics.

Figure 16 reports the liquid fraction distribution of the Y-section profile of slag droplets with different pore structures. It
can be found that the solidification degree of slag droplets with pore structures is greater than that of nonporous slag droplets, which is mainly attributed to the reduction of slag content. Furthermore, the solidification degree of a slag droplet with penetrated pores is greater than that of a slag droplet with closed pores, although the porosity of closed pores (4.8%) is slightly larger than that of penetrated pores (3.8%). The main reason for this phenomenon is that the external syngas directly contacts the internal area of the slag droplet through the penetrated pores, which leads to larger specific surface areas, and enhances the heat exchange between the slag and syngas, resulting in a greater solidification degree.

5. CONCLUSIONS

In the present work, a two-dimensional industrial-scale RSC model and three-dimensional molten slag droplet models are established to explore the solidification characteristics of molten slag droplets in an RSC. In order to predict whether the slag discharge of an industrial RSC is safe, the solidification degree at the outlet of the RSC should be known. Therefore, the solidification degree is analyzed by comparing the residence time and solidification time, which are calculated by numerical simulations of coupling a discrete-phase model and a solidification model. Furthermore, the effects of different operating conditions on solidification characteristics are discussed. The main conclusions are as below:

(1) The residence time of molten slag droplets decreases as the slag diameter increases, which is closely associated with the flow characteristics of slag droplets.

(2) The complete solidification time of molten slag droplets increases as the slag diameter increases. When the slag diameter is greater than or equal to 3.0 mm, the molten slag droplet cannot solidify completely when it reaches the outlet of the RSC.

(3) The solidification degree in the windward zone is greater than that in the leeward zone due to the smaller velocity gradient in the leeward zone.

(4) Although the slag initial temperature has little influence on the solidification process, a lower initial temperature is still beneficial to obtain a greater solidification degree.

(5) The pore structures in slag droplets promote the solidification process and shorten the complete solidification time. Moreover, an increasing slag porosity also shortens the complete solidification time.

■ AUTHOR INFORMATION

Corresponding Authors
Yan Gong – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China; orcid.org/0000-0003-2557-832X; Email: yangong@ecust.edu.cn
Guanguo Yu – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China; State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry and Chemical Engineering, Ningxia University, Yinchuan 750021, PR China; orcid.org/0000-0003-4085-9736; Phone: +86-21-64252974; Email: gsyu@ecust.edu.cn; Fax: +86-21-604251312

Authors
Bo Wang – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China
Jianyong Qiu – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China
Qinghua Guo – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China; orcid.org/0000-0002-9476-7688
Jianliang Xu – Institute of Clean Coal Technology, East China University of Science and Technology, Shanghai 200237, PR China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c02406

Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by the Project of Key Research Plan of Ningxia (2019BCH01001) and the National Natural Science Foundation of China (21878082, 21878094). The Zhejiang Provincial Department of Science and Technology is also acknowledged for this research under its Provincial Key Laboratory Programme (2020E10018).

■ REFERENCES

(1) Wei, J.; Guo, Q.; Ding, L.; Gong, Y.; Yu, J.; Yu, G. Understanding the Effect of Different Biomass Ash Additions on Pyrolysis Product Distribution, Char Physicochemical Characteristics, and Char Gasification Reactivity of Bituminous Coal. Energy Fuels 2019, 33, 3068–3076.

(2) Kong, X.; Zhong, W.; Du, W.; Qian, F. Three Stage Equilibrium Model for Coal Gasification in Entrained Flow Gasifiers Based on Aspen Plus. Chin. J. Chem. Eng. 2013, 21, 79–84.

(3) Xu, C.; Yuan, Z.; Wang, X. Preparation of TiCl4 with the Titanium Slag Containing Magnesia and Calcia in a Combined Fluidized Bed. Chin. J. Chem. Eng. 2006, 14, 281–288.

(4) Meng, Y.; Jiang, P.; Yan, Y.; Pan, Y.; Wu, X.; Zhao, H.; Sharmin, N.; Lester, E.; Wu, T.; Pang, C. H. An advanced ash fusion study on the melting behaviour of coal, oil shale and blends under gasification conditions using picture analysis and graphing method. Chin. J. Chem. Eng. 2021, 32, 393–407.
(5) Chen, X.; Kong, L.; Bai, J.; Dai, X.; Li, H.; Bai, Z.; Li, W. The key for sodium-rich coal utilization in entrained flow gasifier: The role of sodium on slag viscosity-temperature behavior at high temperatures. Appl. Energy 2017, 206, 1241–1249.

(6) Qiu, J.; Guo, Q.; Wei, J.; Xu, J.; Gong, Y.; Yu, G. Numerical Simulation of Heat Transfer and a Forging Plate Structure in a Radiant Syngas Cooler with Radiation Screens. Ind. Eng. Chem. Res. 2020, 59, 16483–16491.

(7) Zhang, P.; Xu, C.; Kuang, J.; Liu, S.; Xia, Z.; Wu, K.; Huang, Y. Investigation on the ash deposition of a radiant syngas cooler using critical velocity model. Energy Rep. 2020, 6, 112–126.

(8) Qiu, J.; Guo, Q.; Xu, J.; Gong, Y.; Yu, G. Numerical study on heat transfer and thermal stress of the upper cone membrane wall in radiant syngas cooler. Appl. Therm. Eng. 2020, 169, 114845.

(9) Guan, X.; Hewitt, A.; Peng, W.; Vimalchand, P.; Nelson, M.; Pinkston, T.; Madden, D. Particulate control devices in Kemper County IGCC Project. Energy Rep. 2019, 5, 969–978.

(10) He, X.; Qiu, J.; Xu, J.; Liu, H.; Yu, G. Dynamic analysis of heat transfer processes of molten slag in a radiant syngas cooler. J. Chem. Eng. Chin. Univ. 2020, 34, 326–334.

(11) Sefidari, H.; Wiinikka, H.; Lindblom, B.; Nordin, L. O.; Wu, G.; Yazhenskikh, E.; Müller, M.; Ma, C.; Ohman, M. Comparison of high-rank coals with respect to slagging/deposition tendency at the transfer-chute of iron-ore pelletizing grate-klin plants: A pilot-scale experimental study accompanied by thermochemical equilibrium modeling and viscosity estimations. Fuel Process. Technol. 2019, 193, 244–262.

(12) Li, G.; Xu, S.; Zhao, X.; Sun, R.; Wang, C. a.; Liu, K.; Mao, Q.; Che, D. Investigation of chemical composition and morphology of ash deposition in syngas cooler of an industrialized two-stage entrained-flow coal gasifier. Energy 2020, 194, 116901.

(13) Li, X.; Yu, G.; Dai, Z.; Zhou, Z.; Wang, F. Numerical Simulation of Molten Slag Deposition in Radiant Syngas Cooler with a CFD-Based Model. J. Chem. Eng. Jpn. 2016, 49, 69–78.

(14) Zhang, B.; Jin, J.; Liu, H. Modeling Study of the Slag Behaviors and SiC Refractory Wall Corrosion on the Top Cone of a Membrane Wall Entrained-Flow Gasifier. Energy Fuels 2020, 34, 12440–12448.

(15) Xing, H.; Wang, X.; Yue, L.; Zhang, Y. Numerical Simulating for Phase-change Heat Transfer Process of slag Granule. Iron Steel Vanadium Titanium 2010, 31, 79–83.

(16) Sun, Y.; Shen, H.; Wang, H.; Wang, X.; Zhang, Z. Experimental investigation and modeling of cooling processes of high temperature slags. Energy 2014, 76, 761–767.

(17) Liu, X.; Zhou, X.; Liao, Q.; Wang, H. Theoretic analysis on transient solidification behaviors of a molten blast furnace slag particle. CIESC J. 2014, 65, 285–291.

(18) Zhu, X.; Ding, B.; Wang, H.; He, X.-Y.; Tan, Y.; Liao, Q. Numerical study on solidification behaviors of a molten slag droplet in the centrifugal granulation and heat recovery system. Appl. Therm. Eng. 2018, 130, 1033–1043.

(19) Gao, J.; Feng, Y.; Feng, D.; Zhang, Z.; Zhang, X. Solidification with crystallization behavior of molten blast furnace slag particle during the cooling process. Int. J. Heat Mass Tran. 2020, 146, 118888.

(20) Peng, H.; Hu, Z.; Shan, X.; Ling, X.; Liu, L. Study on the solidification characteristics of molten slag droplets cooled by mixed cooling medium. Appl. Therm. Eng. 2019, 149, 939–949.

(21) Hu, Z.; Peng, H.; Ling, X.; Xu, H.; Li, J. The influence of the interaction between the multiple slag droplets on the solidification characteristics in humid air. Appl. Therm. Eng. 2020, 170, 115012.

(22) Wang, D.; Yang, H.; Wu, Y.; Zhao, C.; Ju, F.; Wang, X.; Zhang, S.; Chen, H. Evolution of pore structure and fractal characteristics of coal char during coal gasification. J. Energy Inst. 2020, 93, 1999–2005.

(23) Liu, Y.; He, R. A comprehensive fractal char combustion model. Chin. J. Chem. Eng. 2016, 24, 1750–1760.

(24) Guo, Q.; Zhang, Z.; Xue, Z.; Gong, Y.; Yu, G.; Wang, F. Coal char particle secondary fragmentation in an entrained-flow coal-water slurry gasifier. J. Energy Inst. 2019, 92, 578–586.

(25) Ding, B.; Liao, Q.; Zhu, X.; Wang, H. Deep insight into phase transition and crystallization of high temperature molten slag during cooling: A review. Appl. Therm. Eng. 2021, 184, 116260.

(26) Ding, B.; Wang, H.; Zhu, X.; He, X.-Y.; Liao, Q.; Tan, Y. Crystalization Behaviors of Blast Furnace (BF) Slag in a Phase-Change Cooling Process. Energy Fuels 2016, 30, 3331–3339.

(27) Beom, M.; Acosta, A.; Rincón, J.; Romero, M. Thermal expansion of slag and fly ash from coal gasification in IGCC power plant. Fuel 2006, 85, 2352–2358.

(28) Pan, C.; Liang, Q.; Guo, X.; Dai, Z.; Liu, H.; Gong, X. Characteristics of Different Sized Slag Particles from Entrained-Flow Coal Gasification. Energy Fuels 2016, 30, 1487–1495.

(29) Sun, L.; Shen, Z. J.; Liang, Q. F.; Xu, J. L.; Liu, H. F. Water-cooled slag physicochemical property in entrained-flow gasifier. Chem. Eng. 2014, 42, 61–65.

(30) Chang, Y.; Yao, Y.; Liu, Y.; Zheng, S. Can cuttings replace cores for porosity and pore size distribution analyses of coal? Int. J. Coal Geol. 2020, 227, 103534.

(31) Ni, J.; Yu, G.; Guo, Q.; Liang, Q.; Zhou, Z. Experimental and Numerical Study of the Flow Field and Temperature Field for a Large-Scale Radiant Syngas Cooler. Ind. Eng. Chem. Res. 2010, 49, 4452–4461.

(32) Ni, J.; Yu, G.; Guo, Q.; Dai, Z.; Wang, F. Modeling and comparison of different syngas cooling types for entrained-flow gasifier. Chem. Eng. Sci. 2011, 66, 448–459.

(33) Wang, K.; Zhang, H.; Chu, S.; Zha, Z. Pyrolysis of single large biomass particle: Simulation and experiments. Chin. J. Chem. Eng. 2021, 29, 375–382.

(34) Yu, G.; Ni, J.; Liang, Q.; Guo, Q.; Zhou, Z. Modeling of Multiphase Flow and Heat Transfer in Radiant Syngas Cooler of an Entrained-Flow Coal Gasification. Ind. Eng. Chem. Res. 2009, 48, 10094–10103.

(35) Krishnamoorthy, G. A computationally efficient P1 radiation model for modern combustion systems utilizing pre-conditioned conjugate gradient methods. Appl. Therm. Eng. 2017, 119, 197–206.

(36) Sundaresan, S.; Ozel, A.; Kolehmainen, J. Toward Constitutive Models for Momentum, Species, and Energy Transport in Gas–Particle Flows. Annu. Rev. Chem. Biomol. Eng. 2018, 9, 61–81.

(37) Ai, W.; Kuhlman, J. M. Simulation of Coal Ash Particle Deposition Experiments. Energy Fuels 2011, 25, 708–718.

(38) Ding, B.; Zhu, X.; Wang, H.; He, X.-Y.; Tan, Y. Numerical investigation on phase change cooling and crystallization of a molten blast furnace slag droplet. Int. J. Heat Mass Tran. 2018, 118, 471–479.

(39) Pan, Y.; Witt, P. J.; Xie, D. CFD simulation of free surface flow and heat transfer of liquid slag on a spinning disc for a novel dry slag granulation process. Progr. Comput. Fluid Dyn. Int. J. 2010, 10, 292–299.

(40) Zhang, B.; Shen, Z.; Liang, Q.; Xu, J.; Liu, H. Modeling the slag flow and heat transfer on the bottom cone of a membrane wall entrained-flow gasifier. Fuel 2018, 226, 1–9.

(41) Zhang, B.; Shen, Z.; Liang, Q.; Xu, J.; Liu, H. Modeling the slag flow and heat transfer with the effect of fluid-solid slag layer interface viscosity in an entrained flow gasifier. Appl. Therm. Eng. 2017, 122, 785–793.

(42) Ferreira, I. L.; Spinelli, J. E.; Pires, J. C.; Garcia, A. The effect of melt temperature profile on the transient metal/mold heat transfer coefficient during solidification. Mater. Sci. Eng. A 2005, 408, 317–325.

(43) Li, X.; Guo, Q.; Qiu, S.; Yu, G. Numerical study on the performance of an adapted radiant syngas cooler with water spray for entrained-flow gasifier. Asia-Pac. J. Chem. Eng. 2016, 11, 346.