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A new prediction method for the viscosity of the molten coal slag. Part 2: The viscosity model of crystalline slag

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ABSTRACT

The complex chemical composition of coal slag separated out different types and shapes of crystals during the cooling process. The morphology of the crystal in the molten slag influenced the viscosity of the coal slag. The aim of this paper was to obtain the viscosity model of crystalline slag using a new calculation method. The model of the suspension viscosity obtained by the previous study was used to predict the viscosity of molten slag. The correction factor (β) was introduced in the model of the suspension viscosity. The solid phase volume fraction (φ) in the molten slag was calculated by FactSage. The liquid phase viscosity (η_O) was fitted by the viscosities of molten slag in high temperature section. Ten kinds of crystalline slags were applied in this study to prove the accuracy of the modified viscosity model. The viscosity of molten slag predicted by the model (CSM) agreed with experiment data.

1. Introduction

Entrained flow gasification technology is a type of clean coal conversion technology, which plays an important role in reducing carbon dioxide emissions and improving energy efficiency. It allows for various combinations of electricity, liquid fuels, hydrogen, chemicals and heat with the characters of high efficiency and fuel flexibility [1]. Entrained flow gasifier usually operates at high temperature (above the ash flow temperature) to ensure a suitable slagging condition for the stable operation [2,3]. The viscosity of molten slag is the key factor in determining whether the slagging condition is smooth and stable. The viscosity of molten slag exhibited a rapid increase because of the increase of crystals in the molten slag when temperature was lowered below a certain temperature which was referred as the temperature of critical viscosity (T_{cv}) [4,5].

Many factors affect the viscosity of molten slag, including composition, cooling rate, residence time and so on. And these factors influence the viscosity of molten slag by changing the volume fraction of crystal phase in the molten slag. A number of scholars have studied the factors that affect the crystallization of slag. Fredericci et al. [6] considered the crystallization mechanism of an unaltered blast-furnace slag composition. They suggested that most crystallization was on surface and this suggestion was confirmed by the study of nucleation kinetics. Xuan et al. [7–10] studied the influences of CaO, Fe_2O_3, SiO_2/Al_2O_3 on crystallization characteristics of synthetic coal slags. With the increase of CaO, crystallization of the slag became significant, especially in those with a calcium range between 15% and 35%. The crystallization temperature increased but only slightly. With a higher ratio of Fe_2O_3, more crystallization heat was released and the crystallization shifted to a higher temperature, potentially leading to a higher T_{cv} in viscosity. The kinetics under isothermal 1100 °C showed that the growth rate of crystals increased with the addition of iron oxide. As the S/A ratio increased in the range from 1.5 to 3.5, energy barrier was significantly lowered which increased the crystallization ratio. Shen et al. [11] studied the effect of cooling process on the generation and growth of crystals in coal slag. The variation of the cooling process obviously affected the crystallization behavior of molten slag. Low cooling rate benefited the generation of the crystals and long residence time below the initial crystallization temperature promoted the generation and growth of crystal. Louhich et al. [12] reproduced the experimental T dependence of the crystallization temperature with numerical calculations based on standard models for the nucleation and growth of hard-sphere crystals, classical nucleation theory and the Johnson-Mehl-Avrami-Kolmogorov theory. These results suggested that deep analogies existed between hard-sphere colloidal crystals and pluronics micellar crystals, in spite of the difference in particle softness. Studies about

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glassy slag are relatively rich in the glass industry. Karamanov et al. [13] summarized results of the crystallization of iron-rich glasses. The results indicated that magnetite and pyroxene were the main crystal phases and that the kinetics of pyroxene formation could be explained as growth on a fixed number of magnetite nuclei. Pacurariu et al. [14] studied non-isothermal crystallization kinetics of some glass-ceramics with pyroxene structure. Many other scholars [15–19] have also studied the effect of other factors on the slag crystallization process including residual carbon, trace elements and so on.

About the viscosity of molten slag, Kondratiev et al. [20], Ilyushechkin et al. [21] and Zhang et al. [22] studied the characters of flowability and T_{cv} of coal ash slag. Kong et al. [23–25] proposed the internal and external factors influencing the slag viscosity at high temperature, including CaO content, residual carbon and cooling rate. It was found that the viscosities of both the glassy and the crystalline slag were declining when the cooling rate increased. However, when the temperature was lower than the flow temperature, the impact of the cooling rate on crystal slag was more obvious. At the same time, T_{cv} also decreased with increasing cooling rate. Residual carbon in the slag was the internal factor affecting the slag viscosity. With the content of residual carbon increasing, the slag viscosity also increased. When the content of residual carbon was more than 5%, a significant impact on the slag viscosity appeared. They also studied the effect of CaO content on the viscosity curve. When the CaO content increased, the slag viscosity increased, which exhibited the same properties as the viscosity of crystal slag versus S/A ratio. Other documents also mentioned the influence of silicon, aluminum and other trace elements on the slag viscosity. Zhang et al. [26] found that the viscosity of CaO-SiO_2-Al_2O_3-CaF_2 system increased monotonously with gradually increasing substitution content of Al_2O_3 for SiO_2. Feng et al. [27] produced that the viscosity of slag decreased with the increase of TiO_2 content. Wang et al. [28] found that the viscosity of liquid slag decreased with the addition of V_2O_5 increasing at high temperature. Ilyushechkin et al. [29] studied the viscosity of high-iron slags from Australian coal. Folkedahl et al. [30] considered the effect of atmosphere on the viscosity of selected bituminous and low-rank coal ash slags. Under the air atmosphere, the slag viscosity was related to the mole ratio of basic to acidic oxides. However, the changes in viscosity observed in using hydrogen instead of air atmosphere appeared to be related to the amount of iron which was originally in the slag.

Kondratiev et al. [31], Hosseini et al. [32], and Ramacciotti et al. [33] have proposed models to predict the slag viscosity. These models were based on the method of simplifying the synthesis of oxides, so there were great limitations in the accuracy of prediction. Browing et al. [34] proposed a classical prediction method for the viscosity of molten slag which exhibited Newtonian fluid properties. Fulcher et al. [35] proposed the viscosity prediction model based on temperature correction factor by using Arrhenius equation:

$$\log\eta = A + \frac{B}{T - T_0}. \quad (1)$$

where A is a constant, B is the activation energy, and T_0 is the temperature correction factor.

Urbain et al. [36] established the semi-empirical model to predict the viscosity of Al_2O_3-CaO-FeO-SiO_2 system according to Weymann-Frenkel equation [37], which was the most widely used in recent years.

$$\eta = aTe^{1000b/T}, \quad (2)$$

and

$$\ln(a) = -0.2963b - 11.6725. \quad (3)$$

In the equation above, T is the slag temperature, the unit is K. a and b are the empirical constants, b is related to the slag composition, as a function of the slag system components. Many other researchers, such as Kalmanovitch and Frank [38], Kondratiev and Jak [39] and Hurst et al. [40] had also modified the Urbain model.

Table 1
Chemical composition of coals in the experiment.

Composition/wt%	SiO_2	CaO	Al_2O_3	Fe_2O_3	MgO	Na_2O	TiO_2
SF	37.65	26.16	19.69	11.89	0.96	2.62	1.03
YL	27.20	28.12	25.44	16.65	1.07	0.77	0.48
JJT	32.19	25.91	19.85	16.12	3.12	2.31	0.49
YH	26.66	22.27	20.31	24.98	3.01	2.37	0.41
BD	45.23	36.09	9.51	4.55	1.40	1.10	2.11
NM	48.85	16.22	13.28	15.88	3.42	1.64	0.71
TX	50.26	30.42	1.72	11.82	0.56	2.17	3.05
XJW	50.10	30.67	1.53	11.95	0.56	2.16	3.04
BS	39.31	23.89	9.65	21.20	3.23	1.29	1.45
ZX	55.23	21.97	0.20	19.24	0.55	2.04	0.78

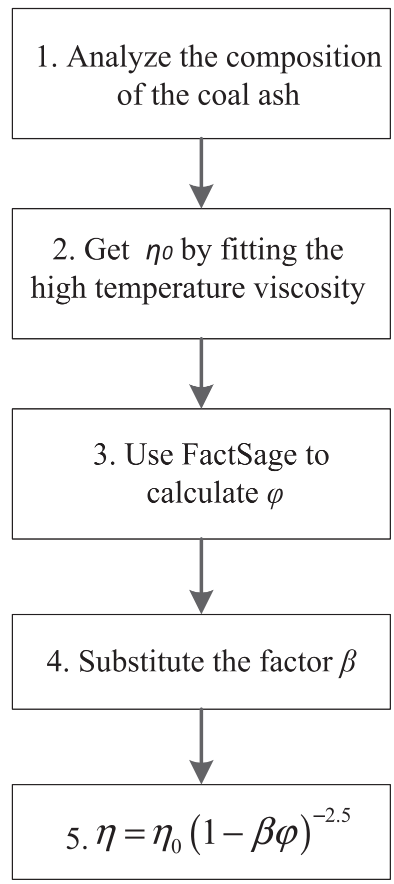

Fig. 1. Schematic diagram of the calculation method.

Reid [41] suggested that the viscosity of the slag at a given temperature can be expressed as an expression of SiO_2 content. Watt et al. [42] studied the viscosity of homogeneous liquid slag in relation to slag composition. The equation was given by

$$\log\eta = \frac{10^7 m}{(T - 150)^2} + c, \quad (4)$$

where η is in poise, and T is in °C. Here

$$m = 0.00835SiO_2 + 0.00601Al_2O_3 - 0.109, \quad (5)$$

and

$$c = 0.0415SiO_2 + 0.0192Al_2O_3 + 0.0276(equiv.\ Fe_2O_3) + 0.0160CaO - 3.92, \quad (6)$$

and slag components are expressed on weight percentages.

It can be seen from the literatures that bulk of irregular crystals generated in the crystalline slag during the crystallization process which improved the difficulty in predicting the viscosity of the