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Influence of Al_2O_3 content on crystallization behaviors of blast furnace slags in directional solidification process

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ABSTRACT

Focusing on the heat recovery of BF slag by dry granulation technology, an improved directional solidification method combines with X-ray diffraction (XRD) is promoted to explore the influence of Al_2O_3 content on crystallization behaviors of BF slags in a phase-change cooling process. In the current research, the crystallization process is on real-time reflected by the temperature distribution inside the slag. Moreover, the type and content evolutions of crystal phase were obtained by analyzing the XRD patterns. Furthermore, Scanning Electron Microscope (SEM) was adopted to investigate the morphology evolution of the crystal phase. The results indicated that with an increase in Al_2O_3 content, the primary crystal structure transformed from Akermanite to Gehlenite. Moreover, the transformation of the crystal structure led to a decrease of critical cooling rate and an increase in critical supercooling degree. Furthermore, the crystal phase precipitated in a varying temperature interval, which was governed by the average cooling rate and primary crystal structure. In addition, the growth mode of the crystal phase transformed from columnar pattern to equiaxed pattern with a decrease in average cooling rate. Furthermore, the transformational cooling rate of growth mode decreased with increasing the Al_2O_3 content.

1. Introduction

Blast furnace slag (BF slag) is one of the main by-products in iron-making process, which carries tremendous thermal energy. In China, the output of crude steel was more than 804 million tons in 2015, meanwhile the production of BF slag was nearly 241 million tons [1]. That means, the total waste heat contained in the BF slag equaled the calorific value of 14 million tons standard coal, corresponding to 13% of energy consumption in the iron-making process [2]. Moreover, BF slag will show different phase types (glassy phase and crystal phase), once it experiences various cooling processes. The glassy slag will be produced in a fast cooling, which is increasingly utilized as cementitious materials due to the better hydraulic activity and high content of calcium silicates [3]. By contrast, the economic value of crystal slag formed in a slow cooling is quite limited. In order to obtain glassy slag for the subsequent utilization, water quench with a high cooling rate is the most popular treatment. Unfortunately, obvious deficiencies are presented during the course of water quenching, such as water consumption and pollution, sulfide (SO_2 and H_2S) emissions and a thorough waste of sensible and latent heat [4,5].

Aiming at water saving, environment protection and heat recovery, dry granulation heat recovery technology has been energetically proposed and then, various techniques such as mechanical crushing method [6–8], air blast method [9,10] and centrifugal granulated method [11,12] have been developed. Among them, centrifugal granulation method is the most feasible one, through which the liquid slag is granulated into small droplets [13,14] and cooled by air at the same time. However, the heat transfer capability of air is quite lower than water. In order to obtain glassy slag the cooling air with a high velocity is essential which debases the quality of the recovered waste heat seriously. On the other hand, cooling air with low velocity is beneficial to recover more waste heat, but the cooling rate may be too slow to get glassy slag. In consequence, a major challenge of the dry granulation heat recovery is to simultaneously recover the utmost residual heat and obtain the glassy slag. At present, the majority of the iron ores used in China are imported from abroad, leading the content of the main components in different steel and iron plants is various. For instance, the content of Al_2O_3 was less than 10% in general slag, while the content can exceed 20% in the steel and iron works used the ore imported from Brazil or Australia [15]. Furthermore, the Al_2O_3 content shows a great influence on slag

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physical properties and crystallization behaviors, and subsequently affects the waste heat recovery from BF slag [16,17]. Accordingly, it is essential to investigate the effect of Al_2O_3 content on the crystallization behaviors of BF slag.

Actually, crystallization behaviors are of great importance in lots of engineering applications, such as metals casting, crystal growth and coal gasification. In these processes, matter is subject to the crystallization behaviors change with temperature which is changing nonlinearly in a phase change cooling process. Moreover, the nonlinear variation of temperature is further enhanced by the plentiful latent heat released in the crystallization process. Furthermore, the quantity of latent heat is governed by the crystal content. Therefore, the crystallization behaviors in a phase change cooling process is always a challenging task because of the complicated heat and mass transfer. The crystallization behaviors under simplified cooling conditions (constant cooling rate or quenching temperature) have been carried out by many researchers [18,19]. For instance, the crystallization behaviors of a Brazilian BF slag under a constant cooling rate were investigated by Fredericci et al. [20]. In this paper, the crystallization onset and ending temperature was obtained through the differential scanning calorimetry (DSC) curve. In addition, XRD was adopted by Wang et al. [21] to explore the effect of quenching temperature on the crystallization behaviors of coal slag. The results indicated that the crystal phase content appeared a maximum value with an increase in quenching temperature. By contrast, single/double hot thermocouple technique (S/DHTT) made it possible to explore the crystallization process in a visualisation way [22–24]. For instance, SHTT was used by Sun et al. [15] to investigate the influence of Al_2O_3 content on the crystallization behaviors of synthetic slags. The results showed that the critical cooling rate experienced a minimum value with the Al_2O_3 content increased to 16.6 wt%. However, crystallization behaviors in a phase change cooling process is quite different from those simplified conditions. Additionally, the microstructure evolution inside alloys after solidification was widely investigated using directional solidification technique [25–27]. Moreover, the phase change cooling process was traced accurately by the thermocouples. Therefore, the combination between XRD, SEM and directional solidification technique make it possible to explore the crystallization behaviors of BF slag in phase change cooling process.

In the present work, four kinds of slag with Al_2O_3 contents ranging from 8.5 to 17% were prepared to analyze the influence of Al_2O_3 on the crystallization behaviors. An improved directional solidification technique was used to obtain the temperature distribution inside the slag. XRD was applied to analyze the evolution of crystal phase structure and content along the vertical direction after the solidification. In addition, the crystal phase morphology was performed, using a Scanning Electron Microscope (SEM). Moreover, DSC was used to detect the glass transition temperature and liquidus temperature respectively. Based on mentioned methods, the evolution of crystal phase structure and content, critical average cooling rate, critical supercooling degree and crystal growth mode under various Al_2O_3 content was experimentally investigated. These investigations may provide theoretical guidance for the dry graduation and waste heat recovery technique in choosing the operating conditions.

2. Experimental procedure

2.1. Experimental system and method

As shown in Fig. 1, the experimental apparatus for the directional solidification of the BF slag mainly consisted of 3 parts: cooling system, test unit and measurement system. Moreover, the preceding studies have given detailed information about the experimental set-up used for the directional solidification trials [28]. Furthermore, the experimental set-up is covered by Morgan brick. And the thickness of the insulation layer is 80 mm. In addition, some improvements have been done to make sure the heat is directionally extracted only through the water-cooled bottom. For example, the diameter of the stainless crucible decreased to 32 mm. Moreover, to better fix the stainless crucible and insulation layer, a cooler made of graphite was added to the cooling system. Furthermore, to avoid the heat exchange between the BF slag and the cooling water in the outer peripheral surface of the stainless crucible, the bottom of the crucible was stacked together with the cooler cover using the heat-resistant adhesive.

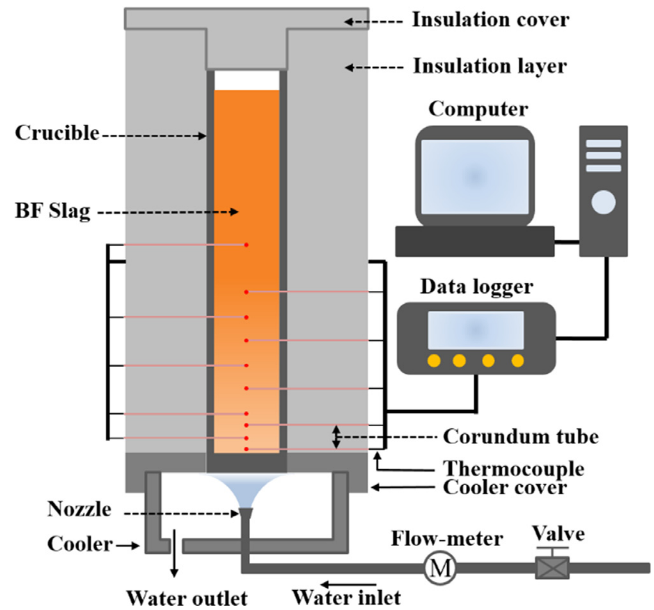

Fig. 1. Schematic representation of the experimental setup.

Prior to the experiment, the stainless crucible was heated to 1100 °C in advance, and the BF slag was heated to 1550 °C in a heating furnace (Model CQ-GG10A, LYCQ, China). Then the melting BF slag contained in a graphite crucible was taken out from the heating furnace using crucible tongs and rapidly poured into the stainless crucible. After that, put the insulation cover over the insulation layer and switched on the cooling water valve simultaneously. In addition, temperatures measured by the thermocouples were recorded by the data acquisition unit and displayed on the computer screen. Eventually, the experiment was finished when the temperature of the slag declined to 250 °C.

After the experiment, the stainless crucible was removed from the test unit, and the slag samples at different positions were cut into slices with a thickness of 1 mm using a diamond wire cutting machine (Model WXD-170, Mike, China). Moreover, to investigate the evolution of crystal growth mode, the slag sample was cut in half along the vertical direction. Then SEM images of these samples (the enlargement rate from 2k to 50k) were obtained by a SEM instrument (Model S-4800, Hitachi, Japan) to analyze the evolution of the crystal phase morphology along the vertical direction. After that, the slag slices were ground into powder using an agate mortar, and the maximum size of the powder was 38 μm. Eventually the XRD patterns of these powdered samples were acquired to determine the evolution of the crystal phase content along the vertical direction, using a XRD system (Model D/max-1200, Rigaku, Japan). The detection was conducted at the conditions of 40 kV and 40 mA using Cu Kα with a scanning speed of $2°\cdot min^{-1}$. In addition, the glass transition temperature (T_g) and the liquidus temper-