Improvement in Dispersion Properties of Graphite Powder by $Al(OH)_3$ Coating through Turbidimetry Measurements

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Surface treatment is one of the most efficient ways to improve the water wettability and dispersion properties of graphite powders used in refractory castables. In this study, the above mentioned properties of graphite powders coated with aluminium hydroxide ($Al(OH)_3$) via mechanical milling were investigated. The results showed that dispersion properties of the graphite powders were greatly improved when the amount of coated aluminium hydroxide was more than 5 % by weight. The results revealed further that dispersion properties of graphite powders increased with increasing mechanical milling time.

1 Introduction

Graphite is widely used as refractory material due to many excellent properties such as thermal shock and high corrosion resistance. However, because of poor water wettability and dispersion, graphite could not be used satisfactorily in refractory castables, in which it easily segregates thereby decreasing its advantageous effects [1 – 3].

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The poor aqueous wettability and dispersion behaviour of graphite is associated with the presence of few hydrophilic functional groups, such as -OH or -COOH on its surface. To improve the wettability and dispersion of graphite in water, various methods have been investigated, including the use of surfactants, coating the graphite surface with other materials, and making graphite micropellets or briquettes [4 – 6].

Coating the surface of graphite with materials, which have better wettability and dispersion properties, is considered one of the most efficient ways to improve the behaviour of graphite. Pitch, silicon carbide, CVD (chemical vapour deposition) silicon carbide and high speed impact treatment coatings have also been used because of their improved wettability and dispersion properties. Unfortunately, however, these methods and materials are too expensive to be commercially viable. *S. Zhang* and *W.E. Lee* [7] have reviewed the advantages and disadvantages of the various methods. The methods of surface treatment, hydrophilic treatment by surface-active agents and surface coating by hydrophilic agents are commonly known. If it is possible to coat hydrophilic agents uniformly on the surface of flake graphite by a cheaper coating method, the property of graphite's surface can approach that of hydrophilic agents, and the method could be commercially viable. Therefore, in this study, aluminium hydroxide or $Al(OH)_3$ has been chosen as hydrophilic agent, which is not expensive, to coat it on the surface of flake graphite by mechanical milling (using a ball mill). The advantages of the turbidimetry method for determining the stability of suspensions are that it is a simple and easy way, which is not costly and one can measure it as soon as possible without any special instructions. So we decided to introduce turbidimetry as one of the easy ways for measuring the stability of graphite suspensions.

Tab. 1 **Specifications of graphite powder used as starting material**

Density	2,21 g/cm^3
Water content	0,33 %
Ash content	5,36 %
Particle size (d_{50})	8,28 µm
Specific surface area	1,56 m^2/g

Tab. 2 **Graphite specimens code**

Specimen code	Milling time [h]	$Al(OH)_3$ [%]
0-0	0	0
0-5	5	0
0-10	10	0
0-15	15	0
5-0	0	5
5-5	5	5
5-10	10	5
5-15	15	5
10-0	0	10
10-5	5	10
10-10	10	10
10-15	15	10

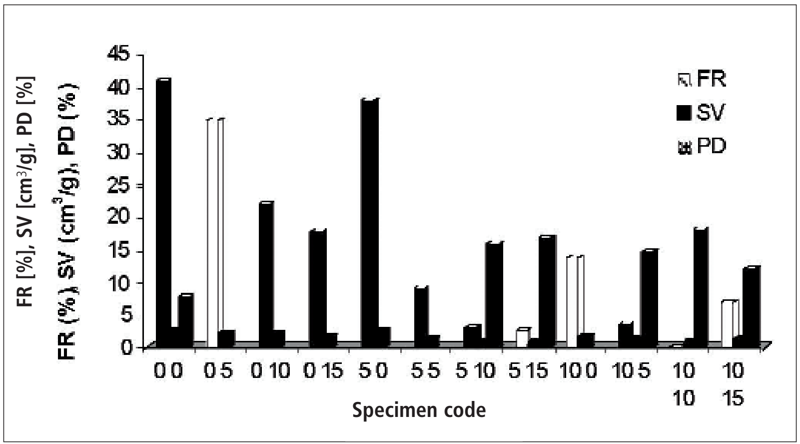

Fig. 1 **Floating ratio (FR), sedimentation volume (SV) and packing density (PD) of as-received and coated graphite powder suspensions**

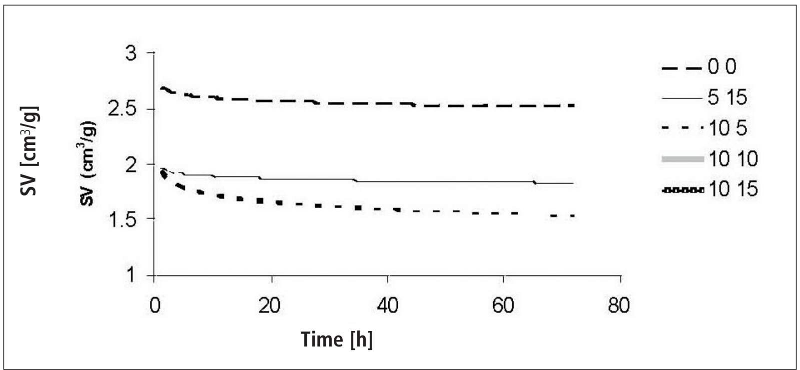

Fig. 2 **Sedimentation volume (SV) versus time of as-received and coated graphite powder suspensions**

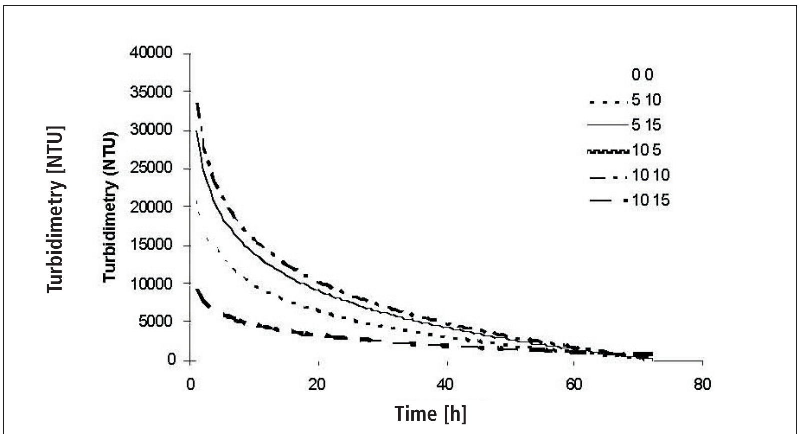

Fig. 3 **Turbidity of as-received and coated graphite powder suspensions**

2 Experimental

German flake graphite, $Al(OH)_3$ (Merck, purity > 99,5 %, particle size < 45 m), carboxymethyl cellulose (CMC) and distilled water were used as the main raw materials. Tab. 1 lists the properties of graphite used in this treatment. Particle size distribution of graphite was studied by laser diffraction micro-analyser Master Sizer 2000 (*Malvern*). Scanning electron microscopy images were made using a LEO1455VP (*LEO*) microscope. The turbidity was determined using the 2100N turbidimeter (*Hach*). Flake graphite and $Al(OH)_3$ were mixed with various amounts of $Al(OH)_3$ for 0, 5, 10, and 15 h in a ball mill (Tab. 2).

CMC was solubilized in water of 60 °C temperature. The surface wettability of untreated and treated graphite was compared with graphite prepared by the following process: 20 g of coated (or uncoated) graphite and 100 cm³ of 0,1 mass-% CMC solution were mixed with a heater stirrer for 1 h and then in a mechanical mixer (*IKA*, Germany) for 1 h (1200 rpm). After this procedure the suspensions were put in a cylinder (15 mm in diameter and 150 mm in height). The height of the graphite sediment layer and the amount of floating graphite were measured. The sediment volume (SV), packing density of the sediment layer (PD) and floating ratio (FR) were calculated according to:

SV [cm³/g] =
volume of sedimentation layer/weight of sedimentation layer,

PD [%] =
1/(SV · graphite true density) · 100,

FR =
weight of floating graphite/total weight of graphite.

SV and FR should ideally be low and PD high for a good wettability.

3 Results and discussion

Fig. 1 shows the floating ratio (FR) of as-received and coated graphite powder suspensions. FR of as-received graphite was about 40 %, whereas those of samples 5-10, 5-15, and 10-10 decreased to 2 %, 1,8%, and 0 %, respectively. The results showed that $Al(OH)_3$ coatings improved water wettability of graphite, but specimens 5 – 15 and 10 – 10 showed the greatest effect. This is confirmed by the SV data shown in Fig. 1, in which specimens 5 – 15 and 10-10 showed the lowest SV value indicating good packing and water wettability.

Fig. 1 also shows the packing densities (PD) of as-received and coated graphite suspensions. The coated graphite achieves higher PD values than as-received graphite, and specimen 10-10 had the highest PD value. The sedimentation volume (SV) versus time from as-received graphite suspensions is much higher than that of coated graphite suspension (Fig. 2). For example, after 3 h the SV of as-received graphite

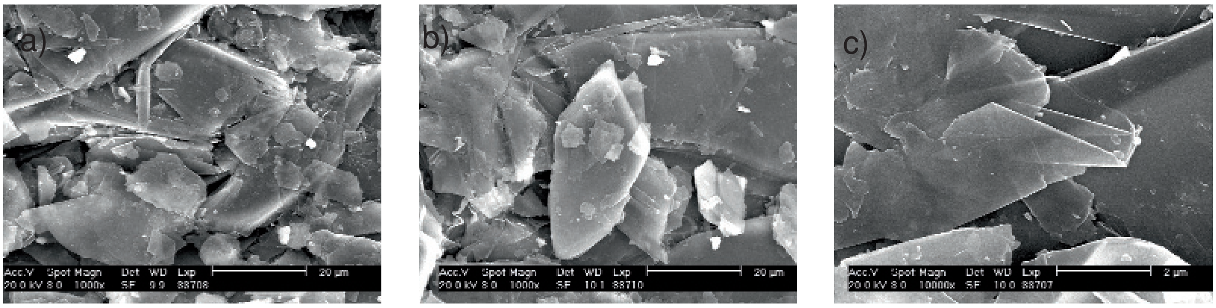

Fig. 4 **SEM photograph of raw materials and treated graphite, a) 0-0, b) 5-5, c) 10-10 specimens**

was 2,6 cm^3/g, whereas that of 10-10 was 1,8 cm^3/g.

As-received graphite (specimen 0-0) surfaces have poor water wettability, attributable to few water-affinity-functional groups such as -OH on its surface. By mechanical milling $Al(OH)_3$ can be stuck on the surface of flake graphite through repeated mechanical work such as impact force, compressive force, friction force and shearing force [8].

The spinning of graphite and $Al(OH)_3$ particles caused them to collide with each other and with the cylinder wall, activating the graphite surface and causing $Al(OH)_3$ to be absorbed on to the graphite surface. Two observations are believed to indicate improved water wettability and dispersion were that the graphite flakes became more rounded. Also much more -OH functional groups are present in the 10-10 and 5-15 specimens. Specimens 5-0 or 10-0 showed worse water wettability due to a lower coverage of graphite, whereas 10-10 had the greatest water wettability indicating the highest coverage of graphite.

Fig. 3 shows the turbidity of the specimens during 72 h. In turbidimetry the amount of light passing through a solution is measured. The higher the turbidity, the smaller the quantity of light transmitted. Turbidity is the reduction in transparency of a continuous phase due to light scattering and absorption caused by a dispersed phase. Thus it is a function of the size and concentration of the dispersed species [9–11].

Compared with as-received graphite, all coated graphite suspensions showed much more turbidity. Due to the coatings, turbidity had increased from 800 NTU (as-received graphite) to a very high level (close to 99999 NTU). These facts indicated that coatings on graphite surfaces improved the turbidity and dispersion properties of graphite. For example, specimen 5-15 showed slightly lower turbidity than specimen 10-10, indicating that 10-10 had the best turbidity. It was -OH functional groups in the coatings, which led to the improvement in graphite's turbidity. Fig. 4 shows SEM images of the graphite powders coated with various amounts of $Al(OH)_3$. With increasing mechanical milling time, the size of graphite powders became uniform [12, 13]. The aspect ratio of flake graphite is high, and it has the state of a plate shape. Treated graphite coated by $Al(OH)_3$, which can be attained by mechanical milling, has a smaller aspect ratio than raw flake graphite and become more spherical after treatment. As shown in figures, $Al(OH)_3$ was evenly distributed on the surfaces of graphite powders and the thickness of the $Al(OH)_3$ layer increased with increasing amount of $Al(OH)_3$ or mechanical milling time.

5 Conclusion

The wettability and dispersion properties of graphite powder suspensions coated with $Al(OH)_3$ via mechanical milling were investigated by sedimentation volume and turbidimetry measurements. The conclusions are as follows:

The wettability and dispersion properties of the graphite powders were significantly improved when the amount of $Al(OH)_3$ was > 5 mass-%. The turbidimetry of the graphite suspensions increased with increasing amount of $Al(OH)_3$ and mechanical milling time. The specimens prepared with 5 mass % of $Al(OH)_3$ and 10 h mechanical milling time achieved the best results.

Acknowledgements

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