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Whitening of bauxite tailings☆

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A B S T R A C T

The main waste product generated by bauxite flotation in China is aluminosilicate tailings. The dark color caused by high iron content limits the application of bauxite tailings in polymers. We developed a new method for whitening bauxite tailings by adding phosphoric acid. The whiteness of bauxite tailings was considerably improved from 19% to 73% with the addition of 17 g of 85% phosphoric acid and calcination at temperature 550 °C. XRD, EDS and FTIR analyses showed that the particles of the whitened bauxite tailings are coated with a mixture of phosphates and polyphosphates. The amount of the both increased with increasing phosphorus content. N2 adsorption analysis indicated that the surface area of the whitened bauxite tailings gradually decreased, whereas pore size expanded with increasing phosphorus content.

1. Introduction

At present, natural minerals as fillers are widely used in coatings, plastics, paper, ink and daily chemical industry [1–3]. The whiteness is one of the most important properties of natural inorganic fillers. Typically, carbon and iron depress the whiteness of kaolin, talc, pyrophyllite, calcium carbonate and other natural minerals. Calcination at high temperature is the best way to eliminate carbon. The iron is usually removed from natural minerals by magnetic separation, acid pickling, reduction, oxidation, flotation and other methods [4,5].

Bauxite tailings are aluminosilicate wastes generated during the improvement of the aluminum to silicon ratio in bauxite by beneficiation methods. Reports show that 0.2 t of aluminosilicate tailings is generated for every 1 t bauxite ore processed by flotation. In China, a large quantity of bauxite tailings is stored in the tailing dam after flotation [6]. The main minerals found in this waste product are diaspore, kaolinite, illite, and pyrophyllite. Its primary chemical components are silica, alumina, and iron oxide. Investigations on the utilization of bauxite tailings in building materials [7], fillers [8], and refractory materials [9] have been carried out in recent years. However, because of high iron content of the waste, its large-scale application has been rendered infeasible. The limitation of mass use of bauxite tailings as fillers is the darkening of the fillers resulted from high iron content of bauxite tailings. In order to expand the application volume of bauxite tailings, people attempted to improve the whiteness of bauxite tailings by removing iron from bauxite tailings, but failed, because the iron of bauxite tailings distributed in numerous minerals which mutually embed with complicated relationship and fine particles.

So, it is very urgent to find new and economical method to whiten bauxite tailings. In our experiments, phosphoric acid was added to bauxite tailings, and expected to react with the iron of bauxite tailings under certain conditions, to achieve the purpose of whitening bauxite tailings.

2. Materials and methods

2.1. Materials

Bauxite tailings were obtained from the Zhongzhou bauxite flotation plant in China. The tailings are composed of 57.2% Al_2O_3, 19.8% SiO_2, 4.1% TiO_2, 0.5% MgO, 0.6% CaO, 5.1% TFe, 0.69% K_2O, and 0.82% Na_2O. By X-ray powder diffraction, diaspore, kaolinite, illite, anatase, hematite, and quartz were determined as the main minerals in bauxite tailings. The bauxite tailings were milled for 5 h using a planet-ball mill to reduce particle size. The particle size of the milled bauxite tailings ranged from 0.02 to 50 μm. Of these, 85% exhibited a particle size of −10 μm, as determined by a laser granulometer (CILAS-1064). The phosphoric acid used (AR grade) was of 85% concentration.

2.2. Whitening of bauxite tailings

In the experiment, two process routes were employed to whiten bauxite tailings, which are pre- and post-reaction calcinations. The former is that the milled bauxite tailings firstly is calcined at 600 °C for 2 h in flowing air to eliminate the organic matter, then react

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with the phosphoric acid to get whitened bauxite tailings; the latter is that the milled tailings firstly react with the phosphoric acid, then calcined in flowing air to eliminate the organic matter, finally bauxite tailings was whitened.

2.3. Characterization

The whiteness of the samples was measured using an automatic albedo meter and following the GB/T 5950–2008 criterion of the National Standards of the People's Republic of China. The crystalline structure of the samples is determined by a D/max-γ A diffractometer (Cu Kα radiation, $\lambda = 0.154056$ nm) studies. Surface morphology and surface chemical compositions of samples were analyzed by scanning electron microscopy (SEM, JOEL-JSM-6700 F) and energy dispersive spectroscopy (EDS) analysis. The fourier transform infrared spectra (FTIR) of the samples were recorded on a Nicolet Nexus 670 spectrometer using a KBr disk dispersed. The N_2 adsorption and desorption isotherms were measured using a Quantachrome Autosorb-1 M surface area and porosity analyzer after the samples were degassed under vacuum at 300 °C for 3 h. BET surface areas were calculated from the BET plot, and pore size distributions were calculated using the BJH model.

3. Results and discussion

3.1. Effect of process route and phosphorous content on whiteness

Two different pathways were used to whiten bauxite tailings, which are pre-reaction calcination and post-reaction calcination. Pre-reaction calcination involved the removal of organics through calcination prior to reaction with phosphoric acid. Post-reaction calcination involved the removal of organics after reaction with phosphoric acid. Fig. 1 illustrates the effect of phosphorous content in both process samples which were calcined at 600 °C for 2 h in flowing air on whiteness. The pre-reaction calcination process resulted in a significant increase in whiteness with increasing phosphorous content, up to 87% whiteness when phosphorous content is higher than 20%. However, at phosphorous content below 12%, its whiteness is far below the post-reaction calcination sample. The post-reaction calcinations process, on the other hand, reached 73% maximum whiteness at 4.5% phosphorous content and then slightly reduced or unchanged with further increasing phosphorus content. The observed decrease in whiteness for samples from the post-reaction calcination was due to the remaining phosphoric acid in product increasing with increasing the addition of phosphoric acid. This remaining phosphoric acid carbonizes the organic matter into fine carbonaceous particles during calcination, which needs a higher calcination temperature for further removal.

The samples from the two processes are different in character. The sample from the pre-reaction calcination behaves very hard lumps, can reach appropriate granularity only through washing with water, breaking and ball milling. While the one from the post-reaction calcination process behaves very loose, and hardly need ball mill. So it is concluded that the post-reaction calcination process is industrially feasible.

3.2. Effect of calcination temperature on whiteness

Organic matter is one of the factors depressing the whiteness of bauxite tailings. Calcinations aim to eliminate the organics. Effect of calcination temperature on whiteness of sample from the post-reaction calcination was also investigated. Fig. 2 shows the relation between the whiteness of samples with 4.5% phosphorous content and calcination temperature. It can be seen that the whiteness depended on calcination temperatures. The whiteness reached the maximum 73% at a calcination temperature of 550 °C, and decreased at calcination temperatures higher than 600 °C. This is because the organic matters of sample have been burned at 550 °C, leading to a considerable increase in whiteness. In further calcination processes at temperatures higher than 600 °C, interactions between a variety of metals and phosphates, as well as yellowish-white iron polyphosphate formation may have occurred [10], resulting in decreasing whiteness.

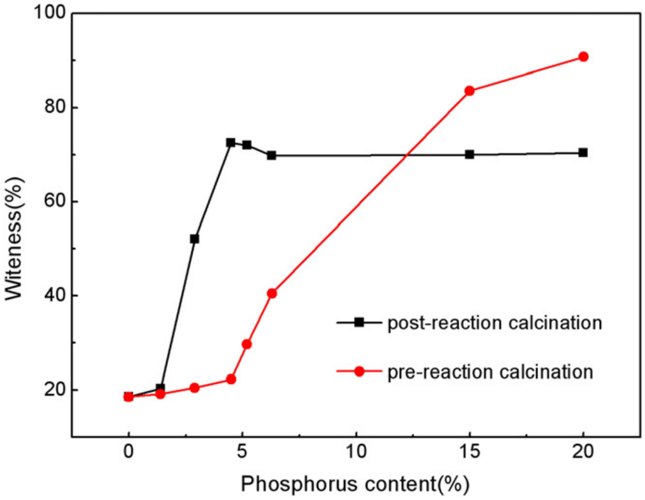

Fig. 1. Effect of process route and phosphorous content on whiteness.

3.3. XRD

The XRD patterns of the samples from the process of the post-reaction calcinations are shown in Fig. 3. Alumina, anatase, quartz and hematite are the main mineral phases of bauxite tailings. The whitening samples contained most of these mineral phases, but the noise of baseline of patterns increased, the diffraction peak intensity of alumina decreased, and a new phase, aluminum phosphate, emerged, indicating that the metal ions on the surface of the particles reacting with phosphoric acid produced more amorphous materials. Hematite was not found in the whitened samples, indicating that hematite reacted completely with phosphoric acid.

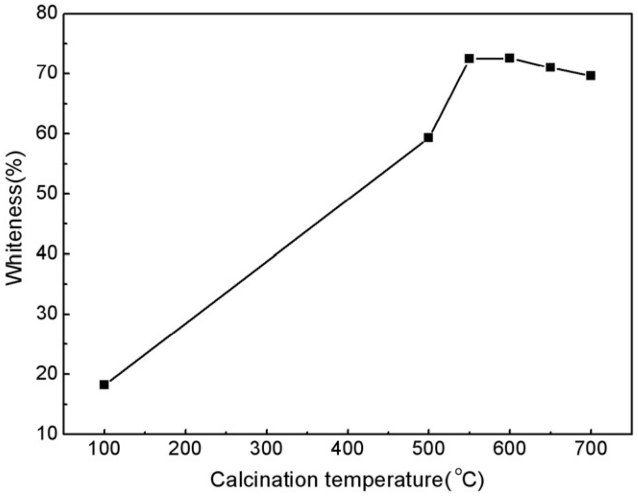

Fig. 2. Effect of calcination temperature on whiteness of sample from the process of the post-reaction calcination.