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Experimental study of extracting alumina from coal fly ash using fluidized beds at high temperature

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ABSTRACT

Experiments are conducted to investigate the process of alumina extraction from coal fly ash generated by coal-fired power plants located in the northern parts of China. Coal ash and ammonium sulfate are mixed and granulated under the effect of binder material. The high temperature fluidized bed reactor is used for the first time to recover the alumina in solid state. The whole process is simplified and operated continuously to make sure its feasibility for the industry. The effect of operating parameters, temperature, reaction time and reactants ratio on efficiency is discussed. The experimental results show that this method has the potential to recover alumina efficiently. And results also show that 60% reaction time is saved when compared with the acid process. The maximum of extracting efficiency reaches nearly 90% at high temperature under laboratory conditions.

1. Introduction

High-alumina coal ash is an industreferenrial byproduct which is generated in coal-fired power plants in most of the western and central parts of China and also in the smelting and chemical industries [1]. With the increasing consumption of coal, the output of fly ash from coal-fired power plants has become the largest industrial solid waste in China [2]. Annual generation is continuously increasing from 155 million tones in 2002 to 620 million tones in 2015 [3]. Common coal ash disposal ultimately lead to the accretion of coal fly ash on wide open ground. Inappropriate management and accumulation of coal ash will be danger to both environment and human health [4]. To utilize coal ash and decrease its damage, we need to change coal ash into high value-added products. Usually, these materials are rich in Al_2O_3 such that the range is nearly 50% and are equivalent to mid-grade bauxite ores [5]. Considering its high alumina content, high-alumina coal fly ash can be utilized as substitute for bauxite, which otherwise needs to be imported into China in large quantities. Recovering alumina from coal fly ash provides a significant opportunity for converting waste materials to a new aluminum source [6]. And it will be a good alternative and could achieve significant economic and environmental benefits as well. In recent years, there have been widespread concerns about the threat of coal fly ash and the continual increase of the bauxite trade price around the world. The increasing awareness of environmental protection and ecological balance has prompted recovering alumina from coal ash to be a research hotpot [7]. Hence, the study of alumina extraction from coal fly ash has attracted extensive attention in recent times.

A number of water or alkaline leaching processes for recovering alumina have been reported and can be broadly classified into sintering processes, hydro-chemical process, acid processes and some other special processes. The sintering processes usually couple a reaction of coal ash with sintering agent powder to form soluble alumina compounds under high temperature [8]. The sinter is then treated with water or Na_2CO_3 solution to draw the alumina compounds from reactants, and the pregnant solution is then subsequently reacted to precipitate out [9]. One of the greatest advantages of this technology is solid industrial foundations. Fundamental knowledge and experiences of sintering process could be drawn on to improve the process. It also has the advantage of being a simple process and mature equipment system [10]. However, the disadvantages include a high energy consumption, an instability of sinter process resulting from narrow sinter temperature and low extraction rate. Sulfur trioxide or ammonia was produced in the sintering process with the addition of auxiliary materials and poses a risk to the surrounding area [11]. Serious dust pollution can easily lead to a very bad working condition. And the dissolution of small amount of silica during leaching is unavoidable which will results in the decrease of the crystallization rate.

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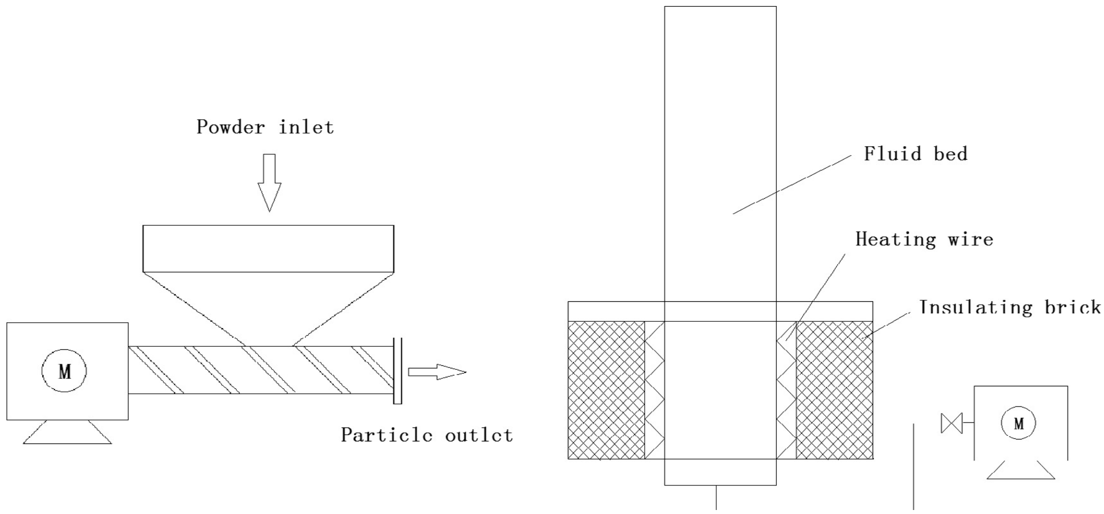

Fig. 1. Schematic diagram of experimental apparatus.

The hydro-chemical process was firstly proposed in 1990s and alumina can be separated from impurities in a wet alkaline process [12]. This process has already been successfully implemented for treatment of red mud and low-grade bauxite [13]. The hydro-chemical process has made great progress in industrial in recent years. The advantages of this process are that it can achieve a high alumina extraction rate and the dealumination slag can be easily decomposed. However, it also has some limitations. The relatively high alkali concentration results in a high viscosity of the mixed slurry [14]. This process was complicated and required higher energy consumption and total cost [15]. High temperature and pressure required led to difficulties to scale-up and improvement of the technology. The purity of product is hard to be controlled with the exist of sodium aluminate hydrate. And the low cycle efficiency of the reaction medium caused by the reaction discipline is another major limitation [16].

In the acid process, the main method now-a-days is that, coal fly ash first reacts with hydrochloric (or sulfuric acid) to generate aluminum chloride (or aluminum sulfate) [17]. The aluminum salts are then crystallized from the acid medium and are subsequently decomposed. This process can thoroughly separate aluminum from silicon; more over, sulfuric, hydrochloric and nitric acids are generally used as leaching agents [18]. Compared with other process, acid process has the advantages of consuming less energy and producing a lower amount of slag. Shemi et al. [19] employed acid process to extract aluminum from coal ash and their results showed that the extraction efficiency is acceptable. Wu et al. [20] leached aluminum from ash by a pressure acid-leaching method. They found that microwave heating has inherent advantages in that it can be selective, controllable and efficient. Xu et al. [21] proposed a new process to extract alumina from ash using NH_4HSO_4 and H_2SO_4 mixed solution. The leaching behavior was investigated and optimized conditions were determined according to their results. Guo et al. [22] performed the research about alumina extraction using hydrochloric acid as the leachant. The addition of NaOH and Na_2CO_3 improved extraction evidently and the mixed additives made the efficiency reached 95% at 700 °C. And the mechanism of improved extraction in the acid solution was analyzed in their work. However, this process still remains pilot-plant scale level and several major drawbacks limit the industrial application of this process, including corrosion, low alumina extraction efficiency, high impurity content and the excessive use of acid and fluoride [23]. The strong acid reaction systems require the need for process equipment to resist corrosion, which will result in a high cost. Also, high recovery cannot be achieved at low acid concentrations and low temperature [24].

Fig. 2. Photo of X-ray diffraction.

Although there are some theoretical research and large-scale experiments, but the extraction efficiency is hard to be controlled in the industry. An industrialized project with an alumina production capacity of 200,000 tones per year was put into production in 2013, but it did not run well and was subsequently stopped because the recovery rate was too low [25]. So such processes are very tough to be fully understood and put into practice. A method that achieves high efficiency and simple process will be essential to the industrial applications for the extraction of alumina from coal ash.

In this paper, we have applied a new method for the recovery of alumina from coal ash by using ammonium sulfate in high temperature fluidized bed to form $NH_4Al(SO_4)_2$ in solid state. Feasibility of the new method has been theoretically proved by researchers [26]. High purity alum is precipitated by the reaction of $NH_4Al(SO_4)_2$, dissolved in water and then followed by crystallization. Owing to the advantages of fluidized bed, such as the good heat and mass transfer performances, high efficiency in gas-solid contact, a classical contact reactor, the fluidized beds have been widely used in the field of combustion, catalytic cracking and synthesis [27]. The process is more simple by using fluidized bed and purity of production is higher than the other process. Here, we just discuss the first stage of recovery of aluminum: the production of ammonium aluminum sulfate using fluidized bed at high temperature. The