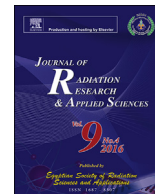


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Ammonium sulfate preparation from phosphogypsum waste

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ABSTRACT

The Egyptian phosphogypsum waste is treated using sulfuric acid prior the ammonium sulfate production. The relevant factors that would affect the removal efficiencies of some impurities are studied. The optimum conditions of the treatment are 8 M sulfuric acid solution and 1/4 solid/liquid ratio for 30 min contact time at 80 °C. Moreover, the optimum conditions of the ammonium sulfate preparation are 10 g of the suspended impure or purified phosphogypsum in 40 ml of 3% ammonium sulfate solution (as initiator), 1/4 solid/liquid ratio at pH7 at an addition of an excess ammonium carbonate, and 150 rpm stirring speed for 4.0 h contact time at 55 °C as well as the 5 mg of barium chloride is added to remove the radium in the ammonium sulfate product. Finally, the ammonium sulfate is crystallized and the chemical analysis of the product shows 20% nitrogen and 23.6% sulfur. Therefore, the purity of the obtained ammonium sulfate is 95% from the purified phosphogypsum.

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1. Introduction

Ammonium sulfate is found in the most everyday applications because of their unique chemical and physical properties. Ammonium sulfate is used in the agricultural fertilizer as a source of nitrogen and sulfur for plant nutrients (Schwenke & McMullen, 2009). Ammonium sulfate is a mixed fertilizer which contains nitrogen and sulfur nutrients (FAO, 2003; Tisdale, 2002). The ammonium sulfate provides the ideal solution that has a quick sulfate release tied in with the season-long availability of degradable sulfur (Hoef, Sawyer, Heuvel, Schmitt, & Brinkman, 1985; Dudal, 2002).

A large amount of waste product phosphogypsum (PG) is produced during the production of wet process phosphoric acid by decomposition of phosphate rock with sulfuric acid. Phosphoric acid plants are really phosphogypsum plants because they make much more gypsum than phosphoric acid (Abu-Eishah & Abu-Jabal, 2001; Klaus, Hurth, & Hoechst, 2002; Parreira, Kobayashi, & Silvestre, 2003; UNSCEAR, 2000; Yang, Liu, Zhang, & Xiao, 2009). The

phosphogypsum is mostly disposed without any treatment, usually by dumping in large stockpiles. These are generally located in coastal areas close to phosphoric acid plants, where they occupy large land areas and cause serious environmental damage. The PG is a powdery material that is composed mainly of calcium sulfate dihydrate (>90% gypsum) and sodium fluorosilicate (Na₂SiF₆) (Berish, 1990; Kacimi, Simon-Masseron, Ghomari, & Derriche, 2006). Due to the residual phosphoric, sulfuric and hydrofluoric acids contained within the porous PG, it is considered an acidic by-product (pH < 3). The PG obtained from the filter cake usually has a free moisture content of 25–30%. The chemical composition of PG from different sources is determined where the main components (%) are CaO, sulphate, SiO₂, Al₂O₃, Fe₂O₃, P₂O₅, and F while the trace metal contents (mg/kg) of the PG are arsenic, silver, barium, cadmium, chromium, lead, mercury and selenium that elements are on the Environmental Protection Agency (EPA) list of potentially toxic elements. It can also be seen that the PG generally has high total contents of Ag, Au, Cd, Se, Sr, some lanthanides and Y (Tayibi, Choura, López, Alguacil, & López-Delgado, 2009).

With regard to the radioactivity, it is mentioned that the PG contains relatively high levels of U-series radionuclides naturally present in the phosphate rock. The most important source of PG radioactivity is reported to be radium (²²⁶Ra) (EPA, 2004; El-Didamony, Gado, Awwad, Fawzy, & Attallah, 2013; Langmuir &

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Riese, 1985; Rutherford, Dudas, & Samek, 1994; USEPA, 2002). Phosphogypsum was treated using different techniques for industrial uses and applications (Khadilkar et al., 2009; Klaus, 1974; Krempff, 1976; Manjit, 2002; Manjit, Garg, & Rehsi, 1993). Ammonium sulfate was produced from many sources whereas it was the oldest synthetic fertilizer (Blouin, Livingston, & Getsinger, 1970; Meline, Faucett, Davis, & Shirley, 1971; Reddy et al., 2011; Sharma, 1991, p. 455; Yong, Mulligan, & Fukue, 2007, p. 211).

The aim of the present study is the production of ammonium sulfate from its economic constitutions, due to the huge amounts of phosphogypsum waste currently produced annually by the phosphate industry in Egypt. The long-term storage and maintenance of the hazard material present economic as well as potential environmental concerns. This study has been found necessary to face that problem. To realize the objectives of this work, the various parameters of the impurities removal upon the phosphogypsum waste have experimentally been optimized by the sulfuric acid. Moreover, the parameters for the ammonium sulfate preparation are studied.

2. Experimental

2.1. Material

2.1.1. Phosphogypsum characteristics

The working phosphogypsum waste is a byproduct obtained from the production of wet process phosphoric acid from rock phosphate at the El Nasr Co. for Intermediate Chemicals on Fayoum City, Egypt. It is a gray colored, damp, fine-grained powder, silt or silty-sand materials. It is mainly $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ with small amounts of phosphate, sand, and clay. The studied phosphogypsum sample is completely analyzed after the complete dissolution by alkali fusion using the suitable analytical techniques.

2.1.2. Samples preparation

The phosphogypsum representative sample is prepared by grinding using a blending mill and quartering method. The separated sample by quartering is milled to -200 mesh size. The pulverized sample is then subjected to the analysis, the treatment and the preparation of ammonium sulfate using the suitable techniques.

2.2. Experimental procedures

2.2.1. Treatment procedures

The treatment process of the phosphogypsum waste is the most important method for the removal of the associated impurities which are contaminating the ammonium sulfate product. Sulfuric acid is used for the treatment of a definite amount to the studied phosphogypsum waste in 250 ml teflon beakers. In this regard, the relevant factors in the treatment are studied to minimize the most associated and harmful impurities. The studied factors will indeed upgrade the concentration of phosphogypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) to a suitable level for the required final preparation of high purity ammonium sulfate. Several treatment experiments are performed to optimize the mentioned relevant factors. In these experiments, the definite amount of impure phosphogypsum sample is progressively treated with different concentrations of sulfuric acid and the solid to aqueous phase ratio ranging from 1/2 to 1/6 for different contact times at different temperatures. In each experiment, the obtained phosphogypsum precipitate is left to settle down after which the slurry is subjected to filtration and the precipitate is thoroughly washed and dried before being directed for the preparation of ammonium sulfate. While the filtrate is analyzed using suitable methods to determine the removal of the most

impurities but fluoride ions is determined in the purified phosphogypsum for the accurate measurement.

2.2.2. Preparation procedures

Ammonium sulfate is prepared by stirring a definite amount of impure or purified phosphogypsum with a suitable amount of ammonium carbonate powder which is suspended in distilled water at different temperatures for different contact times in 250 ml teflon beakers. The reaction operating conditions such as pH of phosphogypsum slurry solution, the amount of ammonium carbonate, solid/liquid phase ratio (S/L), the amount of ammonium sulfate dissolution of phosphogypsum and contact time as well as temperature are studied. The mixture solution is then treated with a few milligrams of barium chloride saturated solution which is used as a carrier to remove the radium ions as barium-radium sulfate precipitate (Habashi, Awadalla, & Zailaf, 1986). After filtration, the ammonium sulfate solution product is obtained as filtrate while the residue is calcium carbonate and most impurities. Subsequently, the filtrate is also dried at 60°C to obtain the purified ammonium sulfate powder.

2.3. Analytical procedures

The quantitative analysis of major oxides SiO_2 , Al_2O_3 , TiO_2 and P_2O_5 in the phosphogypsum sample have been analyzed using a double beam UV–VIS recording Shimadzu UV160A spectrophotometer according to standard methods of analysis while the oxides of Na and K are determined by the flame photometric technique. The Fe_2O_3 , MgO and CaO are chemically determined by titration (Shapiro & Brannock, 1962) whereas the concentrations of the other elements have been analyzed through the inductively coupled plasma optical emission spectrometry (ICP-OES) technique. Whereas the fluoride ion is spectrophotometrically analyzed using the alizarin complexone method after distillation and it is also measured by fluoride electrode (Kandil, Gado, Cheira, Soliman, & Akl, 2016) as well as the activity of radium is obtained by gamma ray photometer. Besides that, the sulfate ions in different samples can be directly measured by the turbidimetric or gravimetric methods (Budesinsky, 1975). Finally, the nitrogen content of ammonium sulfate product is determined by using the Kjeldahl method (Goers, 1980, pp. 37–44). The thermogravimetric analysis of calcium carbonate and ammonium sulfate are obtained using the Perkin-Elmer model TGA 7 Thermogravimetric analyzer which is used to measure the weight loss of the samples.

3. Results and discussion

3.1. Characterization of phosphogypsum

The phosphogypsum basically consists of calcium sulfate dihydrate with small amounts of silica, P_2O_5 , iron, aluminum and fluoride as well as traces of radioactive materials comprises radium and uranium, also heavy metals and lanthanides as presented in Table 1. The harmful impurities in phosphogypsum sample have been affected the production of ammonium sulfate. To obtain the purified ammonium sulfate, the used phosphogypsum must be treated or purified from its impurities prior to the preparation of ammonium sulfate. The chemical analysis of phosphogypsum waste was done using suitable techniques.

3.2. Phosphogypsum treatment

The treatment is used to purify the phosphogypsum sample using sulfuric acid to increase the degree of purity of resultant product. Several series of experiments have been carried out to

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