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Acid mine drainage treatment by coagulation process using a synthesized sulphate-based coal fly ash coagulant

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ABSTRACT

The study focused on the synthesis of a composite coagulant containing polymeric sulphates of Fe, Al, Si, Mg, and Ca from coal fly ash (CFA). Thereafter, the study assessed the performance of the produced composite coagulant in the treatment of acid mine drainage (AMD).

The CFA, a by-product of coal combustion rich in Fe, Al, Si, Mg, and Ca oxides was used in this investigation to synthesize a composite coagulant. This was performed using direct pressure H_2SO_4 leaching process. The results of the study found that the recoveries of Fe, Al, Si, Mg, and Ca from CFA were influenced by the concentration of H_2SO_4, temperature, solid to liquid ratio, and time. Subsequently, the extraction process of metals and silicon were optimized using the concentration of H_2SO_4, temperature, solid to liquid ratio, and time process variables.

The raw and processed CFA samples were analysed. The results showed that particle size distribution of the leach residues compared to raw changed during the leaching process. The SEM analysis revealed that new phases were present in the treated CFA sample compared to raw CFA. The morphology of CFA residues revealed that porous and stem-like structures were formed after leaching.

Investigations of kinetic mechanism, based on the shrinking core model, showed that the rate of the dissolution of CFA is controlled by mixed controlled mechanism (diffusion and chemical reaction). Investigation of the Arrhenius expression for Fe, Al, and Si revealed that Fe required more activation energy (59769.35 J/mole) for its acid dissolution, followed by Al (7248.228 J/mole) and silicon (5751.126 J/mole). The kinetic model study showed that zero-order kinetic model fitted mainly the investigated elements.

Before treatment by coagulation process, the quality of the raw AMD sample was determined in terms of elemental composition, total dissolved solids (TDS), and turbidity. The synthesized complex coagulant performed better in the removal of Al, Fe, Mn, Mg, Ca, Si, Zn, Ni, TDS, and turbidity in AMD treatment. The removal of these impurities was affected strongly by coagulant dosage, pH, and time.

The optical microscopy and SEM/EDS investigations of the floc formation revealed that pollutants adsorption onto hydroxide precipitates occurred during the coagulation process.

A comparison of the conventional iron and aluminium sulphate coagulants showed that the produced composite coagulant performed better than conventional coagulants in terms of removal of Mg, Ca, Si, Zn, and TDS. However, the performance of the conventional iron and aluminium sulphate coagulants was quite similar for the removal of Al, Fe, Mn and Ni.

In general, little information is available about the equilibrium capacities and adsorption kinetics of CFA-based coagulant for Al, Fe, Mn, Mg, Ca, Si, Zn, and Ni in aqueous solution. Furthermore, very limited information is available on the thermodynamic and kinetics aspects of adsorption of other element compounds onto other hydroxide precipitate-based adsorbents. Therefore, in order to better understand the adsorption process equilibrium and kinetics, a comprehensive study was conducted. The adsorption isotherm study showed that Temkin and BET equations best fitted the coagulation process. Both, Temkin and BET equations revealed that the adsorption energy was less than zero (i.e., exothermic behaviour). The chemical reaction kinetic model study suggested the applicability of the pseudo-second-order kinetic model. The molecule diffusion kinetic model investigation showed that multi adsorption stage occurred during the process. The intraparticle diffusion coefficients for the different pollutants were calculated from Weber–Morris and Boyd's models; and the results showed that the intraparticle diffusion coefficients, of the specific pollutants are in the range of $1.8\ 10^{-11}$ to $0.57\ 10^{-13}$ cm^2/sec and $1.05\ 10^{-12}$ to $1.87\ 10^{-12}$ cm^2/sec, respectively, (except for Ni and Ca for the Weber–Morris model).

The development of models for both processes, coagulant synthesis by leaching of CFA and AMD treatment by the synthesized coagulant, was performed with a new approach that fits the behaviour of the coagulant production and AMD treatment based on the software called "Eureqa Newtonian". The results showed that the model equations (y is the concentration and *t* the time) to predict the dissolution of Fe, Al, Si, Mg, and Ca from CFA in acidic medium are, respectively: $y = 57.3 - \frac{9.4\ t}{1.2-(0.3\ t)^{52.7}}$ (R^2 value of 0.95); $y = -79.9 + 52.7\ t - 9.1\ t^2 + 0.5\ t^3 + 62.6\ t^{-1}$ (R^2 value of 0.99); $y = 1.01\ t + 5.9$ (R^2 value of 1); $y = 58.7 + 4.1\ t - 0.2\ t^2$ (R^2 value of 0.91); and $y = 5.6\ t - 0.04\ t^3$ (R^2 value of 0.98).

However, for the prediction of AMD coagulation behaviour, results showed a polynomial equation form as $Ax^n + Bx^{n-1} + \cdots + Fx + G = 0$. The model equations can be summarized as follows where the input and output variables are time and concentration, respectively: the equation obtained for Al was $y = 0.01x^2 - 0.59x + 10.61$ with $R^2 = 0.87$, for Fe as $y = 0.0001x^4 - 0.01x^3 + 0.41x^2 - 8.13x + 63.44$ with $R^2 = 0.99$, for Zn as $y = 0.054x^2 - 2.56x + 47.44$ with $R^2 = 0.78$, for Si as $y = -0.0011x^3 + 0.12x^2 - 3.53x + 86.09$ with $R^2 = 0.96$, for Mn as $y = -0.0003x^3 + 0.028x^2 - 0.66x + 57.69$ with $R^2 = 0.98$, for Mg as $y = 0.0055x^2 - 0.22x + 66.66$ with $R^2 = 0.90$, for Ca as $y = 0.017x^2 - 1.14x + 92.44$ with $R^2 = 0.88$, and for Ni as $y = -0.0002x^4 + 0.022x^3 - 0.62x^2 + 3.9x + 96.19$ with $R^2 = 0.98$.

In addition, a mathematical model was developed based on the first principle of material balance to predict the dissolution of CFA with time. The dissolution model of a partial differential equation was applied to describe the mass transfer of element as it moves through the CFA particle towards the solution. The results showed that mathematical model for the CFA dissolution as function of time is as follows:

$$y(z) = 6.98 \text{ x } 10^{-11}\, e^{42.10 \text{ x } 10^{6} z} + \ 3.09 \text{ x } 10^{4} z\, e^{42.10 \text{ x } 10^{6} z}$$