

Kinetics and Adsorption Isotherm Study of Removal of Zn^{+2} ions from Aqueous Solution Using Thermal Power Plant Fly Ash

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Abstract—In the present investigation, fly ash received from Koradi Thermal Power Plant, India, is used as an adsorbent for the removal of Zn^{+2} ions from aqueous solution. The adsorption study were systematically carried out in a batch process using *Atomic Absorption Spectroscopy (AAS) Technique*, covering different process parameters such as amount of adsorbent, effect of contact time under constant stirring condition and steady state condition, and initial Zn^{+2} ions concentration. Removal tendency of Zn^{+2} ions from aqueous solution were also investigated using lime as an admixture along with the fly ash. It was found that use of lime as an admixture enhances the removal tendency of Zn^{+2} ions. The equilibrium data were processed using both Langmuir and Freundlich isotherm models. Finally, the kinetics of adsorption of Zn^{+2} ions on fly ash particles were tested for different models. The results obtained in the present research work suggest that fly ash along with lime may be a promising adsorbent for Zn^{+2} ions.

Index Terms—Adsorption isotherm, fly ash, pseudo first order reaction, zinc (II) ions.

I. INTRODUCTION

Industrial used water is one of the major sources of aquatic pollution. Among the aquatic pollutants, heavy metals have gained relatively more significance in view of their persistence, bio-magnification and toxicity. The main sources of heavy metals are the mining and metal industries. The removal of heavy metals from waste water because of their toxic effects on living species is extremely important [1]. As metal Zinc is highly reactive and has found widespread use in electroplating and metal finishing processes [2].

Removal of heavy metals from wastewater by chemical precipitation, flotation, ion exchange, adsorption and biological treatment, are often used to remove the heavy metals from waste water. Among numbers of removing

techniques, adsorption is known more useful and economical. The application of low cost and easily available materials in wastewater treatment has recently attracted great interest. Fly ash, a by-product of coal burning power plants, has been used as a potential adsorbent for removal of Zinc and other heavy metals for water pollution control or wastewater treatment [3].

Indian coals have very high ash content. The ash content of coal used by thermal power plants in India varies between 25% and 45%. [4]. However, coal with an ash content of around 40% is predominantly used in India for thermal power generation, causing several disposal problems. A lot of investigations have been reported in the literature on the utilization of fly ash for adsorption of individual pollutants in an aqueous solution. The results are encouraging for removal of heavy metals from industrial wastewater using fly ash [5-8].

The present study was undertaken to evaluate the effectiveness of fly ash in the removal of Zn^{+2} by adsorption. Laboratory batch kinetic and isotherm studies were conducted to determine the adsorption capacity of fly ash. The pH of the solution was maintained between 5.5 – 9.0 (pH limits for effluents as recommended by Central Pollution control Board, India).

II. MATERIALS AND METHODS

A. Fly Ash

Sample of the raw coal fly ash was collected from Unit 7 of Electrostatic Precipitator (ESP) hopper number 5 of a coal-burning thermal power plant situated at Koradi, Nagpur District, Maharashtra, India; it was used without pretreatment. The fly ash powder was dried in the electric oven at 120 °C for at least 24 hours and kept over fused calcium chloride in the desiccators, just prior to use. The chemical composition of fly ash was found to be: SiO_2 (62.6%), Al_2O_3 (24.41%), Fe_2O_3 (4.04%), TiO_2 (0.69%), CaO (0.35%), MgO (0.54%), Na_2O (0.27%), K_2O (0.21%), SO_3 (0.84%), P_2O_5 (0.037%), MnO (0.28%) and Loss on Ignition (1.27%). The particle size distribution was done using standard method by passing the fly ash over the standard size molecular sieves. It was found that 91.12 % fly ash particles have size below 75 μm .

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B. Equipments

Atomic absorption spectra were recorded on an atomic absorption spectrophotometer (Model – GBC 932 AA). Different batch experiments were carried out by measuring atomic absorption spectra at wavelength of 341.5 nm (slit width = 0.2 nm) using Air-acetylene (oxidizing) flame type. For the weighing purpose, digital balance (Make: Metler Toledo, Switzerland) were used having uncertainty to weigh up to ± 0.1 mg. Solution pH was measured by using digital pH meter (Model EQ 621, Make - Equip-tronics, India).

C. Preparation of Zn^{+2} Ions Standard Solution

The analytical reagent grade $ZnSO_4 \cdot H_2O$ powder (E-Merck, India, mass fraction purity – 99.9 %) were used for the batch adsorption study. The stock solution of Zn^{+2} (500 mg/l) was prepared by dissolving 4.3966 g $ZnSO_4 \cdot H_2O$ in 2 liters fresh doubly distilled water. Standard stock solution of Zn^{+2} of 1000 mg/L were procured from Merck, India (mass fraction purity - 99.98 %) and were used for the calibration of spectrophotometer.

D. Batch Adsorption and Kinetics Study

The effect of amount of fly ash on the percentage efficiency of removal of Zn^{+2} from the aqueous solution has been studied. For this purpose, aqueous solutions containing of Zn^{+2} (concentration = 20 mg/l to 100 mg/l) were prepared from the stock solution of 500 mg/l. 100 ml aqueous solution were taken in the different conical flask, and fly ash ranging in the amount of 1g to 48 g were added. The mixtures were stirred using magnetic stirrer for 3 hours and then keeping this solution unstirred for 21 hours. The adsorbent were separated from the solution by centrifuge at a speed of 3000 rpm for 5 minutes and the Zn^{+2} solutions were used to undertake measurements of final pH of the solution and atomic adsorption spectra.

To investigate the effect of contact time under the mixing condition and unstirred condition, two set of experiment were carried out using aqueous solution of Zn^{+2} (concentration = 20 mg/l). Amount of fly ash were maintained constant equal to 10 g in 100 ml solution in each of the batch experiments. Eleven samples in different conical flasks were kept unstirred while remaining eight samples in different conical flasks were mixed continuously at 150 rpm speed over the magnetic stirrer for different time intervals. In both the conditions, the adsorbent were separated from the solution by centrifuge at a speed of 3000 rpm for 5 minutes and the supernant Zn^{+2} solution was used to measure atomic adsorption spectra.

Effects of addition of lime as admixture under constant mixing condition and unstirred condition have also been investigated in the present study. For this purpose, batch experiments were carried with 100 ml aqueous solution of Zn^{+2} (20 mg/L) were taken in the conical flask. In each sample, 1.0 g fly ash as adsorbent was added, whereas, the amount of lime were varied as 20 mg/l (pH = 7.5), 30 mg/l (pH = 7.6) and 35 mg/l (pH = 7.9). The studies of addition of higher amount of lime were not undertaken as pH of the solution with higher amount of lime was rising above pH = 9.0. As per the norms of Central Pollution Control Board of

India (CPCB, India) for the industrial waste effluent, the pH should be within 5.5 to 9.0.

The aqueous solutions containing 5 mg/l, 10 mg/l, 20 mg/l, 40 mg/l, 60 mg/l, 100 mg/l and 200 mg/l Zn^{+2} , were prepared from the stock solution (500 gm/L). In the 100 ml of these samples, 10 gm dried fly ash was added. These samples were stirred constantly at 150 rpm for 3 hours to attain the equilibrium time. The adsorbent were separated from the solution by centrifuge at a speed of 3000 rpm for 5 minutes and the supernant Zn^{+2} solution were used to measure atomic adsorption spectra. The Zn^{+2} adsorption isotherm in aqueous solutions were obtained from the study.

III. KINETICS AND ADSORPTION ISOTHERM MODELS

A. Reaction Kinetics

A number of experimental parameters are usually considered in kinetics studies for one adsorption process. Two well-known kinetic models, pseudo-first-order and pseudo-second-order model, were employed to describe the adsorption process. The pseudo-first order, proposed by Lagergren [9] for adsorption analysis, is expressed in the form

$$dq_t / dt = k_1(q_e - q_t) \quad (1)$$

where q_e and q_t are the sorption capacity (mg/g) of Zn^{+2} ions at equilibrium and at a time t , respectively, and k_1 is the rate constant for pseudo first-order sorption (l/min). After integration and applying boundary conditions, viz. that the initial conditions are $q_e - q_t = 0$ at $t = 0$, Eq. (1) becomes:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (2)$$

Equation (2) can be transformed to nonlinear form as

$$q_t = q_e(1 - e^{-k_1 t}) \quad (3)$$

The pseudo second-order equation [10] can be written as:

$$dq_t / dt = k_2(q_e - q_t)^2 \quad (4)$$

where q_e and q_t are the sorption capacity (mg/g) of Zn^{+2} ions at equilibrium and at a time t , respectively, and k_2 is the rate constant for pseudo second-order sorption (g/(mg.min)). For the boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$, the integrated form of Eq. (4) becomes:

$$1/(q_e - q_t) = 1/q_e + k_2 t \quad (5)$$

which is the integrated rate law for a pseudo second-order reaction [9]. This has a linear form:

$$(t/q_t) = 1/(k_2 q_e^2) + (1/q_e).t \quad (6)$$

B. Adsorption Isotherm Study

The isotherm plays a practical role in predictive modeling procedure for analysis and to design the sorption systems. The data obtained from batch sorption studies were analyzed according to Langmuir and Freundlich adsorption isotherms equations. To optimize the design of a sorption system for the

sorption of Zn^{+2} ions from the aqueous solution, it is important to establish the most appropriate correlation for the equilibrium curves.

According to, Langmuir [9] theory, the sorption takes place at specific homogeneous sites within the sorbent. Freundlich [11] isotherm is derived by assuming a heterogeneous surface with a non-uniform distribution of heat of sorption over the surface. The standard models of Langmuir (Equation (7)) and Freundlich (Equation (8)) used are represented below:

$$(C_e / q_e) = (C_e / q_m) + 1 / (K_a \cdot q_m) \quad (7)$$

where, C_e is equilibrium concentration of Zn^{+2} ions ($mg \cdot L^{-1}$), q_e is solid phase concentration of Zn^{+2} ions ($mg \cdot g^{-1}$), q_m ($mg \cdot g^{-1}$), and K_a ($L \cdot mg^{-1}$) are empirical constants denoting mono layer capacity or limiting sorption, signifies the solid phase concentration, corresponding to the complete coverage of available sorption sites and energy of sorption respectively, can be evaluated from the slope and intercept of the linear plot of C_e / q_e against C_e .

$$\ln q_e = \ln K_f + (1/n) \cdot \ln C_e \quad (8)$$

where, K_f is the Freundlich characteristic constant [$(mg \cdot g^{-1})(L \cdot g^{-1})^{1/n}$] and $1/n$ is the heterogeneity factor of sorption, obtained from intercept and slope of $\ln q_e$ versus $\ln C_e$ linear plot respectively.

IV. RESULTS AND DISCUSSION

A. Effect of Amount of Fly Ash on pH of Solution and Removal of Zn^{+2} Ions

Effect of amount of fly ash added on the pH of the Zn^{+2} ions from the aqueous solution have been depicted in Fig. 1. It can be seen from the figure that pH of the solution increases continuously with addition of fly ash. Sudden increase in pH of solution occurs with addition of 7 gm fly ash per 100 ml sample. Effect of amount of fly ash adsorbent on the removal of Zn^{+2} ions from the aqueous solution have been depicted in Fig. 2. It can be seen from the figure that amount of Zn^{+2} ions removed increases with increase in amount of fly ash added. It can be easily seen from the above figure that 18 gm fly ash per 100 ml sample adsorbed 20 ppm Zn^{+2} ions completely.

B. Effect of Contact Time on the Removal of Zn^{+2} Ions

Effect of contact time on the removal of Zn^{+2} ions from the aqueous solution under constant stirring condition have been depicted in Fig. 3. It can be observed from the this figure that amount of Zn^{+2} ions adsorbed on the fly ash increases almost linearly up to 2 hours constant stirring. But rate of uptake of Zn^{+2} ions lowered with further increase in the contact time.

The equilibrium time of contact required to remove Zn^{+2} ions from the solution was observed to be 3 hrs in the present work.

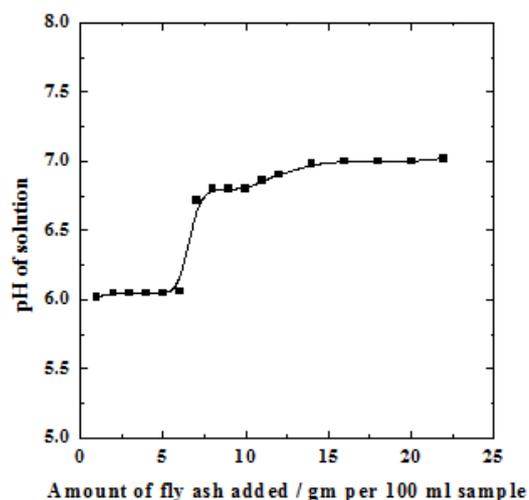


Fig. 1. Variation in final pH of the solution against amount of fly ash added per 100 ml sample.

(Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; contact time = 3 hrs stirring and 21 hrs without stirring)

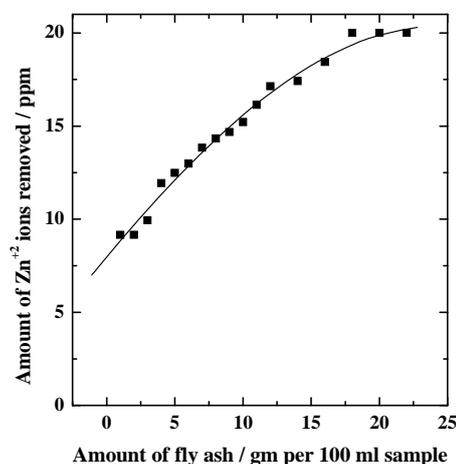


Fig. 2. Variation in amount of Zn^{+2} ions removed from solution against amount of fly ash added per 100 ml sample.

(Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; contact time = 3 hrs stirring and 21 hrs without stirring)

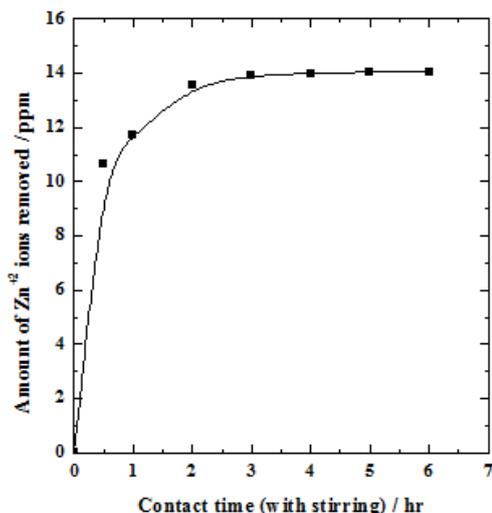


Fig. 3. Variation in amount of Zn^{+2} ions removed from solution against contact time under constant stirring condition.

(Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; Amount of Fly ash = 10 gm / 100 ml sample)

Effect of contact time on the removal of Zn^{+2} ions from the aqueous solution under steady state condition have been depicted in Fig. 4. It can be seen from the fig. 4 that amount of Zn^{+2} ions adsorbed on the fly ash increases continuously with rise in contact time.

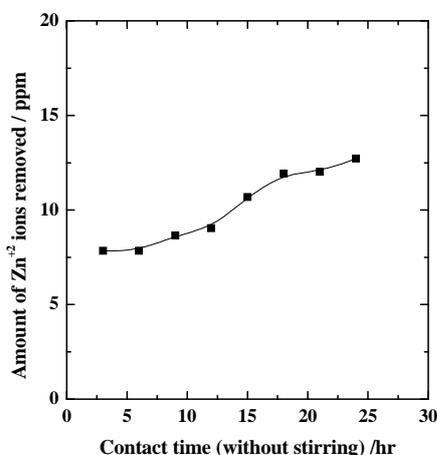


Fig. 4. Variation in amount of Zn^{+2} ions removed from solution against contact time under unstirred condition. (Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; Amount of Fly ash = 10 gm / 100 ml sample)

The maximum percentage of removal efficiency of Zn^{+2} ions was found to be 69.7% under 3 hrs of constant stirring whereas only 63.6% ions were adsorbed on the fly ash after 24 hours when the reaction mixture were kept under without stirring condition. Thus, from the results obtained in the present work, it can be concluded that Zn^{+2} ions from the aqueous solution removed effectively when through mixing were done.

C. Effect of Initial Concentration Zn^{+2} Ions

Effect of initial Zn^{+2} ions concentration on the removal tendency of fly ash, from the aqueous solution have been depicted in Fig. 5. It was observed that amount of Zn^{+2} ions concentration adsorbed on the fly ash increases with the increase in the initial Zn^{+2} ions concentration in the solution. On the other hand, percentage efficiency of Zn^{+2} ions removal were found to be decreased with rise in the initial Zn^{+2} ions concentration in the solution (Fig. 6).

D. Effect of Lime as an Admixture on the Removal of Zn^{+2} Ions

To understand the effect of lime added as admixture along with the fly ash, the graphs have been plotted showing variation in amount of Zn^{+2} ions removed against contact time, under constant mixing condition in Fig. 7. It was observed that 20 ppm Zn^{+2} ions were removed completely in all the condition. Thus, conclusion can be drawn that even addition of small amount of lime i. e. as low as 20 ppm, as an admixture along with 10 gm fly ash as an adsorbent for 1 hour effectively remove the Zn^{+2} ions from the solution. This might be due to the fact that addition of lime increases the pH of the solution which enhances the precipitate formation of $Zn(OH)_2$.

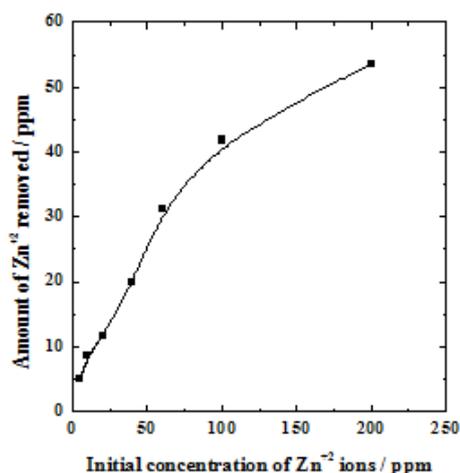


Fig. 5. Variation in amount of Zn^{+2} ions removed from solution against initial concentration of Zn^{+2} ions in solution. (Experimental Condition: Amount of Fly ash = 10 gm / 100 ml sample, contact time: 1 hr stirring)

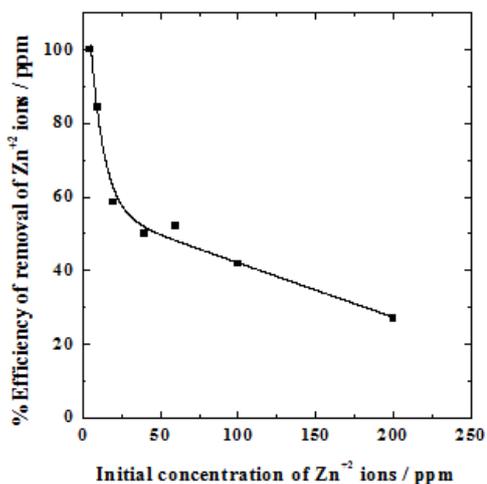


Fig. 6. Variation in percentage efficiency of Zn^{+2} ions removal from solution against initial concentration of Zn^{+2} ions in solution. (Experimental Condition: Amount of Fly ash = 10 gm / 100 ml sample, contact time: 1 hr stirring)

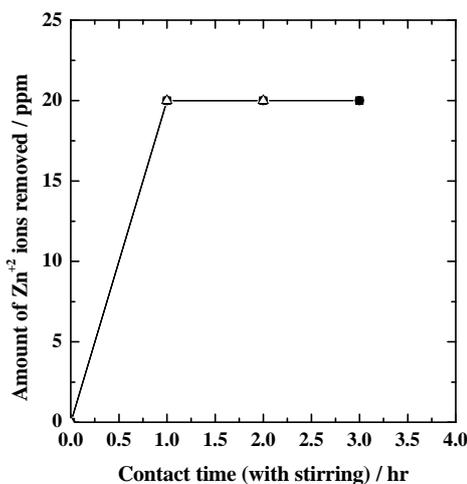


Fig. 7. Variation in amount of Zn^{+2} ions removed from solution against contact time under constant stirring condition in presence of lime as an admixture. (□-□, 20 ppm lime; ●-●, 30 ppm lime; Δ-Δ, 35 ppm lime) (Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; Amount of Fly ash = 10 gm / 100 ml sample)

The variation in the amount of Zn^{+2} ions adsorbed on the fly ash particles against contact time under steady state condition with varied amount of lime i. e. 20 ppm, 30 ppm and 35 ppm have been plotted in Fig. 8. It was observed that 18.8 ppm Zn^{+2} ions were removed within 6 hrs and maximum 19.4 ppm were removed within 24 hrs unstirred condition when the amount of lime added was 20 ppm in the solution. But with rise in the amount of lime i. e. 30 ppm entire Zn^{+2} ions were removed with 6 hrs unstirred condition.

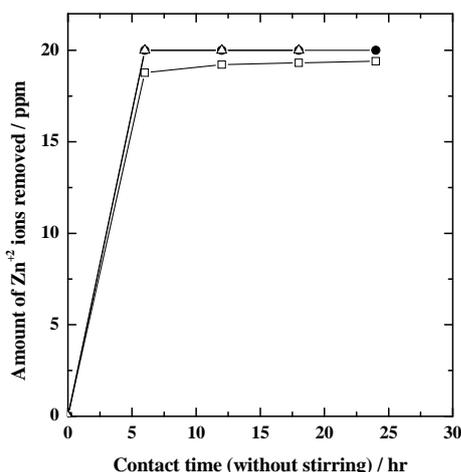


Fig. 8. Variation in amount of Zn^{+2} ions removed from solution against amount of lime as an admixture under unstirred condition. (\square - \square , 20 ppm lime; \bullet - \bullet , 30 ppm lime; Δ - Δ , 35 ppm lime) (Experimental Condition: Initial Concentration of Zn^{+2} ions = 20 ppm; Amount of Fly ash = 10 gm / 100 ml sample)

The variation in the amount of Zn^{+2} ions adsorbed on the fly ash particles against amount of lime as an admixture along with 1 gm fly ash for samples having different initial concentration of Zn^{+2} ions i. e. 20 ppm, 50 ppm and 100 ppm have been plotted in Fig. 9.

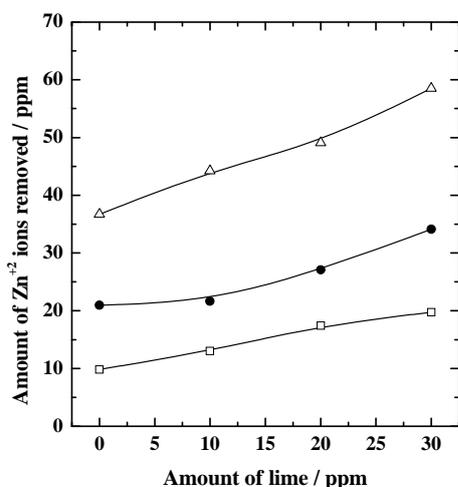


Fig. 9. Variation in amount of Zn^{+2} ions removed from solution against amount of lime as an admixture. (\square - \square , 20 ppm initial concentration of Zn^{+2} ; \bullet - \bullet , 50 ppm initial concentration of Zn^{+2} ; Δ - Δ , 100 ppm initial concentration of Zn^{+2}) (Experimental Condition: Amount of Fly ash = 1.0 gm / 100 ml sample, contact time: 1 hr stirring)

The maximum efficiency of removal of Zn^{+2} ions from the solution was observed when amount of lime added was 30 ppm in all the three conditions.

E. Chemical Kinetics

In the present study, pseudo first order and second order reaction kinetics were tested. When $\ln (q_e - q_t)$ were plotted against t , the R^2 value was found to be equal to 0.952. R^2 value for the plot of t / q_t against t (Fig. 10) was found to be equal to 0.999. This implied that adsorption of Zn^{+2} ions from the solution over the fly ash particles followed the second order kinetics. Equilibrium sorption capacity of fly ash (q_e) was calculated to be 15.15 mg/l whereas second order rate constant (k_2) was found to be equal to $0.0092 \text{ l.mg}^{-1}.\text{min}^{-1}$.

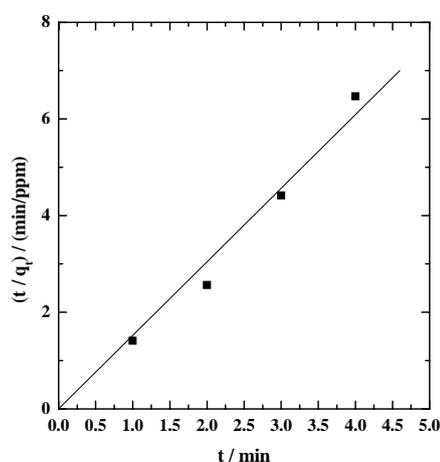


Fig. 10. Second order kinetics for adsorption of Zn^{+2} ions in aqueous solution

F. Adsorption Isotherm Study

The results obtained in batch adsorption experiment were fitted to Langmuir adsorption isotherm using least square fit method as shown in Fig. 11 and the coefficient of regression (R^2) were found to be 0.994.

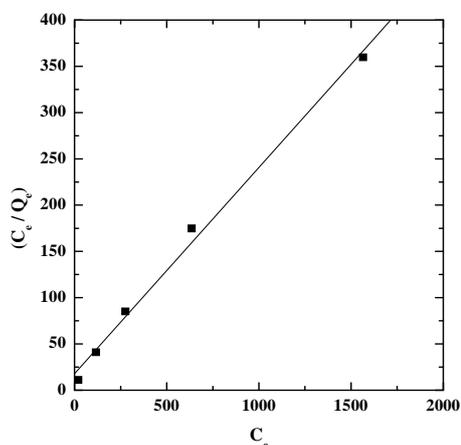


Fig. 11. Langmuir adsorption isotherm of Zn^{+2} ions from aqueous solution using fly ash adsorbent

The values of q_m was estimated to be equal to 4.505 mg.g^{-1} whereas magnitude of K_a was found to be 0.0124 L.mg^{-1} . The results of adsorption were also fitted to the Freundlich Adsorption isotherm using least square fit method which is as shown in Fig. 12. The coefficient of regression (R^2) was found to be 0.982. Therefore, the equilibrium adsorption data of Zn^{+2} ions can be represented more appropriately by the Freundlich models in the studied concentration range. The magnitude of n i. e. reciprocal of heterogeneity factor of sorption was calculated to be equal to 5.076 whereas K_f i. e. the Freundlich characteristic constant was estimated to be equal to 1.0398.

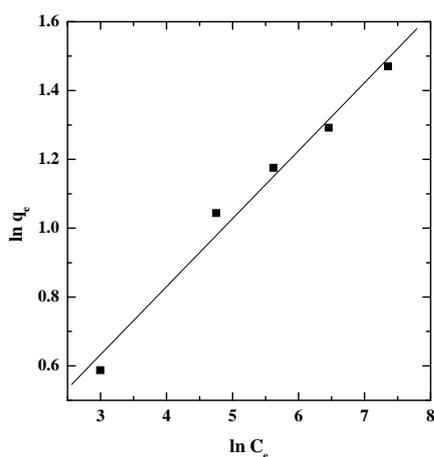


Fig. 12. Freundlich adsorption isotherm of Zn^{+2} ions from aqueous solution using fly ash adsorbent.

V. CONCLUSION

The report predicted from the current paper clearly suggest that use of exhausted coal fly ash from the thermal power plant as an adsorbent for the removal of Zn^{+2} ions from the solution is much economical, effectual and more viable. Lime as an admixture can also be efficiently used to remove Zn^{+2} ions. The different operational parameters observed during the process of investigations reveal that the amount of adsorbent, contact time (stirred and steady state condition), concentrations of the adsorbate, concentration of lime as a admixture govern the overall process of adsorption.

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