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## CHARACTERIZATION OF COAL FLY ASH AND ITS ABILITY TO REMOVE Ni<sup>2+</sup> IONS FROM AQUEOUS SOLUTION

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The objective of this study was to characterize the coal fly ash produced from an Indian thermal power station. The fly ash was characterized using particle size distribution, chemical composition, scanning electron microscopy (SEM), X-ray diffraction (XRD) and zeta potential. Investigations were also carried out to study the ability of this fly ash to remove the nickel (Ni<sup>2+</sup>) ions from aqueous solution. The effects of various experimental conditions such as amount of fly ash, contact time both in stirring condition and steady state condition, initial metal concentration and pH of the solution on the removal efficiency of Ni<sup>2+</sup> ions were studied at room temperature. In this study, it was found that fly ash can effectively adsorb Ni<sup>2+</sup> ions from aqueous solution under favourable conditions.

**Key words:** Ni<sup>2+</sup> ions fly ash, adsorption, efficiency.

**Karakterizacija letećeg pepela ugljena i njegova sposobnost uklanjanja Ni<sup>2+</sup> iona iz vodene otopine.** Cilj ovog istraživanja bio je karakterizirati leteći pepeo ugljena koji je nastao u indijskoj termo-elektrani. Pepeo je karakteriziran pomoću raspodjele veličine čestica, kemijskog sastava, skenirajuće elektronske mikroskopije (SEM), difrakcije X zraka (XRD) i zeta potencijalom. Istraživači su također proveli studiju mogućnosti uklanjanja nikal (Ni<sup>2+</sup>) iona iz vodene otopine pomoću analiziranog letećeg pepela. Proučavani su učinci različitih eksperimentalnih uvjeta, kao što su količina letećeg pepela, vrijeme kontakta tijekom miješanja i u ravnotežnom stanju, zatim početne koncentracije metala i pH otopine na učinkovitost uklanjanja Ni<sup>2+</sup> iona na sobnoj temperaturi. U ovom istraživanju, utvrđeno je da leteći pepeo može učinkovito adsorbirati Ni<sup>2+</sup> ione iz vodene otopine pod povoljnim uvjetima.

**Ključne riječi:** Ni<sup>2+</sup> ioni, leteći pepeo, adsorpcija, učinkovitost.

### INTRODUCTION

Pollution has been very damaging to aquatic ecosystems, and may consist of agricultural, urban, and industrial wastes containing contaminants such as sewage, fertilizer, and heavy metals that have proven to be very damaging to aquatic habitats and species. Continuous discharge of industrial, domestic and agricultural wastes in rivers and lakes causes deposit of pollutants in

sediments. Such pollutants include heavy metals, which endanger public health after being incorporated in food chain.

Heavy metals cannot be destroyed through biological degradation [1]. Ni<sup>2+</sup> is present in the effluents of silver refineries, electroplating, zinc base casting and storage battery industries. Higher concentrations of nickel cause cancer of lungs, nose and bone.

Dermatitis (Ni itch) is the most frequent effect of exposure to Ni, such as coins and jewellery [2]. Acute poisoning of Ni<sup>2+</sup> causes headache, dizziness, nausea and vomiting, chest pain, tightness of the chest, dry cough and shortness of breath, rapid respiration, cyanosis and extreme weakness [3].

Various methods have been reported for the removal of heavy metals from industrial effluents, such as ion exchange, evaporation, chemical reduction and precipitation, electrolysis, electroplating, ion flotation, adsorption etc. [4] but the selection

of the wastewater treatment methods is based on the concentration of waste and the cost of treatment [5]. Most of these methods suffer from some drawbacks such as high capital and operational cost or the treatment and disposal of the residual metal sludge [6].

Among these, adsorption is by far the most versatile and widely used method for the removal of pollutants due its high removal capacity and ease of operation at large scale. Therefore, the present study was undertaken to evaluate the effectiveness of fly ash in the removal of Ni<sup>2+</sup> by adsorption.

## MATERIALS AND METHODS

### Preparation of Ni<sup>2+</sup> ions standard solution

The analytical reagent grade nickel sulphate hexahydrate (Merck, India, mass fraction purity – 99.9 %) was used for the batch adsorption study. The stock solution of Ni<sup>2+</sup> (500 mg/l) was prepared by dissolving 4.4783 g NiSO<sub>4</sub> x 6H<sub>2</sub>O in 2 liters freshly prepared double distilled water. To carry out

various studies, this stock solution of 500 mg/l was diluted by adding the double distilled water, as per the requirement of the study to prepare solutions having different concentration of Ni<sup>2+</sup> ions (10, 20, 40, 60, 100 and 200 mg/l).

### Equipments

Study of particle size distribution of fly ash was done by passing the fly ash over the standard size molecular sieves. To determine the major mineral components of fly ash, X-ray diffraction spectra (XRD) were recorded over the 2θ range of 0 to 65° using Philips X-Pert-ProXRD diffractometer equipment with a CuKα radiation source. Scanning electron microscopy (SEM) analysis was carried out using scanning electron microscope (Model Philips SEM 515) between 25 kV to 30 kV to study the surface texture and morphology of received

fly ash sample. The zeta potential of fly ash sample at different pH was determined by a Zetameter System 4.0 (Zetameter Inc. Staunton, VA 24402, USA).

Atomic absorption spectra were recorded on an atomic absorption spectrophotometer (Model – GBC 932 AA). For the weighing purpose, digital balance (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, Equiptronics, India).

## 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.

## Experimental

The fly ash was used without any pretreatment. The fly ash was characterized for its particle size distribution, X-ray diffraction spectra (XRD), Scanning electron microscopy (SEM) analysis and zeta potential.

To investigate the effect of fly ash dose, contact time, initial pH of the solution and initial concentration of Ni<sup>2+</sup> ions under the stirring condition and steady state condition, two set of experiment were carried out using aqueous solution of Ni<sup>2+</sup> ions of various concentrations as required. For the study of fly ash dose, contact time, and initial pH of the solution, the Ni<sup>2+</sup> ions concentration in the aqueous solution was maintained at 20 mg/l. The required solution was stirred if required at 150 rpm over the magnetic stirrer. After studying the effect of fly ash dose, the quantity of fly ash was maintained at 10 g per 100 ml of the solution having concentration of 20 mg/l.

The effect of initial ion concentration (10, 20, 40, 60, 100 and 200 mg/l) on the removal efficiency of fly ash is also studied. The pH range of the reaction mixture was maintained within 5.5 to 9.0 which were strictly as per the limits of pH for industrial discharge water, as given by the environment (protection) rules of India. After the experiment, the supernatant clear solutions were separated from the solution by centrifuge at a speed of 3000 rpm for 5 minutes. The resultant Ni<sup>2+</sup> ions solution were used to undertake measurements of final pH of the solution and atomic adsorption spectra to determine the amount of Ni<sup>2+</sup> ions adsorbed by the fly ash. The removal efficiency (%) of fly ash dose can be calculated by using the following formula.

$$\text{Efficiency (\%)} = \frac{\text{Amount of Ni}^{2+} \text{ ions adsorbed (mg/l)}}{\text{Total conc. of Ni}^{2+} \text{ ions (mg/l)}} * 100$$

## RESULTS AND DISCUSSION

### Characterization of fly ash

The chemical composition of fly ash is presented in Table 1.

Table 1. Chemical composition of the fly ash sample, expressed in wt. %  
 Tablica 1. Kemijski sastav uzorka letećeg pepela, izražen u %

Constituent	wt. %
SiO <sub>2</sub>	65.01
Al <sub>2</sub> O <sub>3</sub>	24.41
Fe <sub>2</sub> O <sub>3</sub>	4.04
TiO <sub>2</sub>	0.69
CaO	0.35
MgO	0.55
Na <sub>2</sub> O	0.22
K <sub>2</sub> O	0.21
SO <sub>3</sub>	0.15
P <sub>2</sub> O <sub>5</sub>	0.037
Mn	0.28
Loss on Ignition	2.04

According to the ASTM C-618 this fly ash can be classified as class F for having a less than 5% SO<sub>3</sub> content and loss on ignition less than 6% with a greater than 70% content of three components-SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> [7]. The pH of the fly ash was found to be in between 6.9 to 7.9.

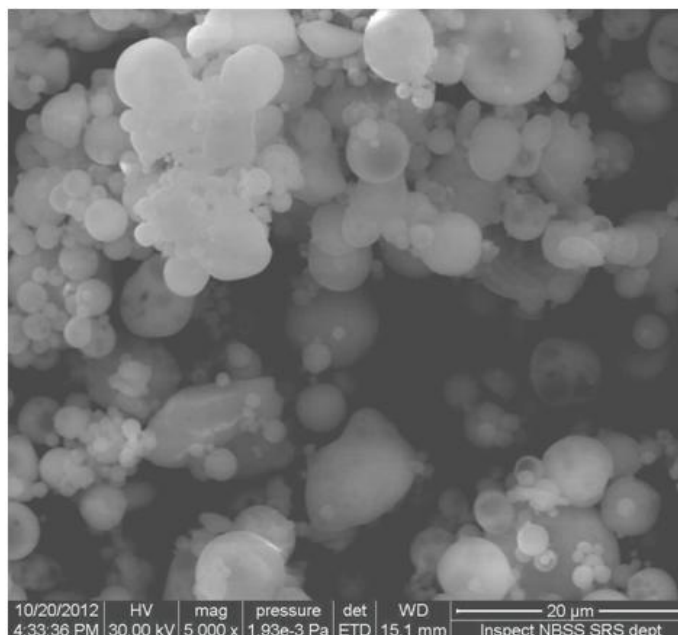
The particle size distribution was done using standard method by passing the fly ash over the standard size molecular sieves. The particle size distribution of the fly ash sample is shown in Table 2. It was found that 91.12 % fly ash particles have size below 75 μm.

Table 2. Particle size distribution of the fly ash sample  
 Tablica 2. Raspodjela veličine čestica u uzorku letećeg pepela

Particle size in μm	Left over fly ash in g (Total quantity = 500 g)	wt. %
> 1000	0.14	0.028
1000 - 600	0.24	0.048
600 - 425	0.26	0.052
425 - 300	0.42	0.084
300 - 212	0.88	0.176
212 - 150	5.62	1.124
150 - 75	36.82	7.364
< 75	455.62	91.124

Figure 1 shows the SEM micrograph of a coal fly ash sample at 5000x magnification. It is seen that, fly ash particles are

mostly spherical in shape, whereas small amount of irregular shaped particles are also present.

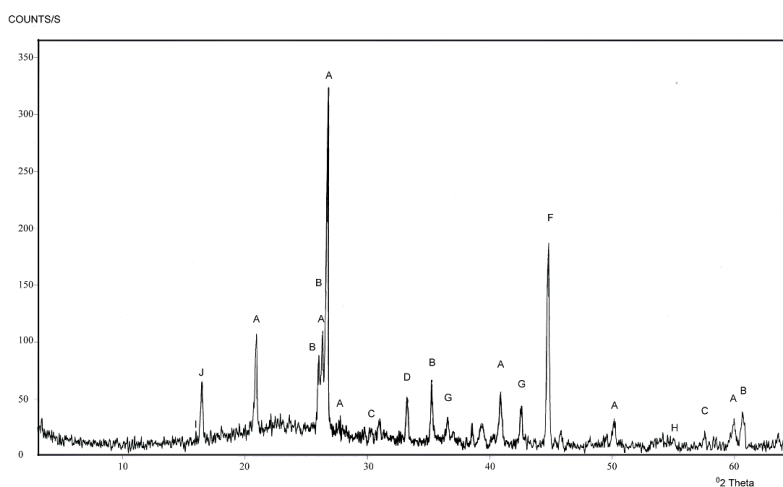


**Figure 1.** SEM image of fly ash sample (magnification: 5000x)

**Slika 1.** SEM snimka uzorka letećeg pepela (povećanje 5000x)

The Figure 2 shows the X-ray diffraction pattern for the fly ash sample. The observed peaks were assigned to their respective components. It can be seen from

the diffractogram of fly ash sample that components are mainly present in their oxide form.



**Figure 2.** XRD spectra of fly ash sample

**Slika 2.** XRD spektar uzorka letećeg pepela

Where: A – quartz , silicon dioxide (SiO<sub>2</sub>)  
 B – mulite, aluminum silicon oxide ( 3Al<sub>2</sub>O<sub>3</sub>x2SiO<sub>2</sub>)  
 C – magnetite (Fe<sub>3</sub>O<sub>4</sub>)  
 D – hematite (Fe<sub>2</sub>O<sub>3</sub>)  
 E – leucite, potassium aluminumsilicate K(AlSi<sub>2</sub>O<sub>6</sub>)  
 F – magnesium silicate (Mg<sub>2</sub>SiO<sub>4</sub>)  
 G – silicon oxide (SiO<sub>2</sub>)  
 H – rutile, (TiO<sub>2</sub>)  
 I – calcium aluminum sulphite hydrate (Ca<sub>6</sub>Al<sub>2</sub>O<sub>6</sub>(SO<sub>3</sub>)x32H<sub>2</sub>O)

The values of Zeta potential of fly ash at different pH are shown in Table 3.

**Table 3.** Zeta potential of fly ash at different pH values

**Tablica 3.** Zeta potencijal letećeg pepela kod različitih pH vrijednosti

Sr. No	Initial pH	Final pH	Zeta potential, mV
1	2.0	2.3	-13.2
2	3.0	3.7	-27.9
3	4.0	4.8	-31.8
4	5.0	6.1	-32.5
5	6.0	6.3	-34.8
6	7.0	7.1	-35.2
7	8.0	7.9	-35.7
8	9.0	8.8	-37.6

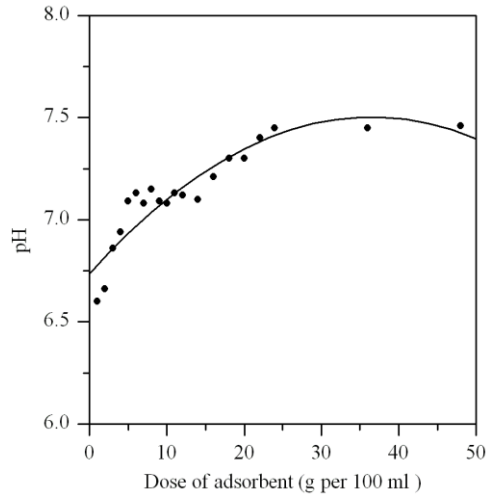
It is seen from the Table 3 that the values of zeta potential are negative within pH range of 2.0 up to 9.0. It was also found that zeta potential increases continuously with pH of the solution. From the close scrutiny of this table, it can be seen that the charge on the surface of the fly ash is negative over a varied range of the pH of aqueous medium which results in the forma-

tion of the attractive forces among the fly ash and positively charged metal ions in the solution. The result in the present research work indicates that the adsorption of heavy metal ions (positively charged) on fly ash is favorable process as the adsorbent particles had shown negative surface charge even in the highly acidic medium.

### Effect of fly ash dose on pH of the solution

To study the effect of dose on the pH of Ni<sup>2+</sup> solution, the aqueous solutions containing 20 mg/l of Ni<sup>2+</sup> were prepared. Fly ash dose from 1 gm per 100 ml to 48 g par 100 ml of solution was added and it was

stirred for three hours and kept in steady state for twenty one hours. The effect of fly ash dose on the pH of the solution have been studied and plotted in the Figure 3.



**Figure 3.** The plot of variation in pH against dose of adsorbent (fly ash)

**Slika 3.** Dijagram promjene pH u odnosu na količinu adsorbensa (leteći pepeo)

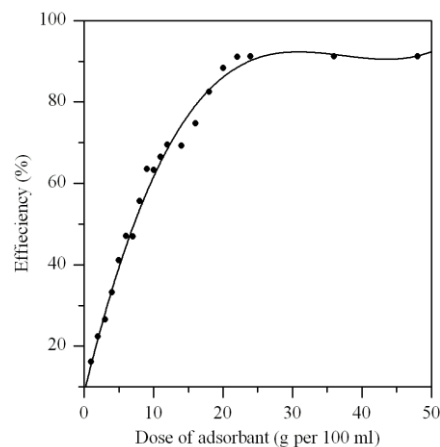
Figure 3 indicates that as the dose of fly ash increases from 1 gm per 100 ml solution to 22 gm per 100 ml of the solution, the pH of the solution varies from 6.6 to 7.4.

Further increase in the dose of fly ash, does not increase the pH because the pH of fly ash is in the range of 6.9 to 7.9 which keeps the pH of the solution near to neutral.

### $\text{Ni}^{2+}$ removal capacity of fly ash

To study the effect of dose on the percentage removal efficiency of  $\text{Ni}^{2+}$  from the aqueous solution, the aqueous solutions containing 20 mg/l of  $\text{Ni}^{2+}$  were prepared. Fly ash dose from 1 gm per 100 ml to 48 g per 100 ml of solution was added and it was

stirred for three hours and kept in steady state for next twenty one hours. The supernatant solution was separated from the solution and all the results generated from AAS has been shown in the Figure 4.



**Figure 4.** The plot of variation in removal efficiency (%) against dose of adsorbent (fly ash)

**Slika 4.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na količinu adsorbensa (leteći pepeo)

Figure 4 indicates that percentage removal efficiency of fly ash increases continuously from 16.2 % for 1 g adsorbent up to 91.1 % for 22 g of adsorbent. It was also observed that with further increase in quantity of adsorbent, the % of Ni<sup>2+</sup> ions removed from the aqueous solution does not varies to much extent and remains almost constant.

The reason for such type of trend can be explained as follows: Initially when the amount of fly ash is lower, the number of available adsorption sites increased by increasing adsorbent dosage. Thus increase in adsorption with increasing adsorbent dosage is mainly due to the unsaturated adsorption sites through the adsorption reaction [8]. While, at very high doses of fly ash in the batch process, the inter-particle

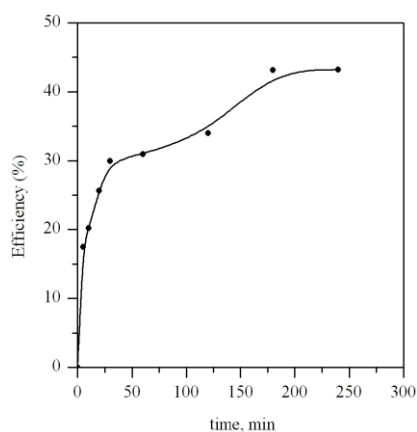
interactions among the fly ash molecules, such as aggregation, caused by high adsorbent concentration takes place. Such aggregation would lead to a decrease in the total surface area of the adsorbent [9] and the percentage efficiency of removal of Ni<sup>2+</sup> ions does not varied as in the case of lower dose of adsorbent. It can be concluded that when mass of adsorbent in the solution is less, the magnitude of percentage efficiency varies very sharply.

It was noticed that when 10 g fly ash adsorbent is added in the reaction mixture, almost 50 percent removal efficiency of Ni<sup>2+</sup> ions was achieved. Therefore, it was decided to restrict the dose of fly ash to a maximum of 10 g per 100 ml of solution in the further study.

### Effect of contact time

To study the effect of contact time of fly ash on the percentage removal efficiency of Ni<sup>2+</sup> from the aqueous solution, the aqueous solutions containing 20 mg/l of Ni<sup>2+</sup> and fly ash (10 g per 100 ml) were prepared. These solution were kept in constant stirring condition and the steady state condition.

After the specified contact time, the supernent solution was separated and all the results generated from AAS are used to determine the amount of Ni<sup>2+</sup> ions adsorbed. The results obtained have been plotted in Figure 5 and under steady state condition in the Figure 6.



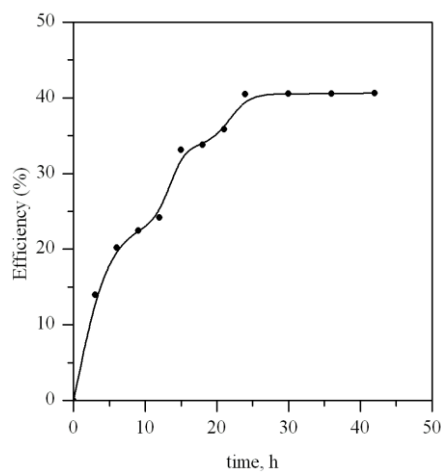
**Figure 5.** The plot of variation in removal efficiency (%) against contact time, under constant stirring

**Slika 5.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na vrijeme kontakta tijekom konstantnog miješanja



It can be seen from the Figure 5 that the removal efficiency of aqueous solution of Ni<sup>2+</sup> ions concentration of 20 mg/l having fly ash as 10 g per 100 ml, increases with increase in contact time when solution was kept in at constant stirring condition. It can be easily seen from the figure that the removal efficiency is very high at the beginning of the adsorption but with increase in contact time it increases but at slow rate.

The removal efficiency which reached 30.96% in just one hour when the solution was stirred at 150 rpm and 10 gm fly ash per 100 of the solution was added to it. This removal efficiency has been increased to about 43 % when the contact time was increased to 3 hour and after that increase in contact time does not increase the removal efficiency, under the similar experimental conditions.



**Figure 6.** The plot of variation in removal efficiency (%) against contact time, under steady state  
**Slika 6.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na vrijeme kontakta u ravnotežnom stanju

It is depicted from the Figure 6 that the removal efficiency of Ni<sup>2+</sup> ions (conc. 20 mg/l and fly ash 10 g per 100 ml) under steady state condition increases at a very slow rate, with increase in contact time. It can be easily seen from the figure that the removal efficiency reached 13.96% in 3 hours and to about 40 % when the contact time was increased to 24 hour under the similar experimental conditions.

### Effect of initial pH of solution

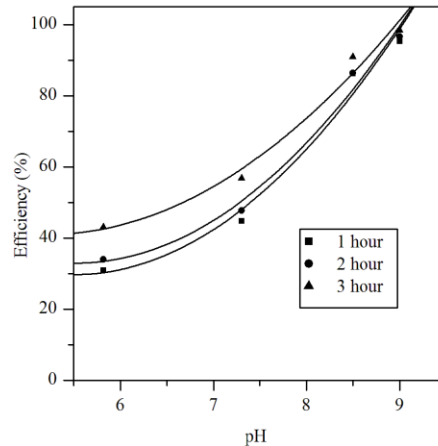
Effects of initial pH of solution, under constant stirring condition and steady state condition have also been investigated in the present study. The aqueous solution of Ni<sup>2+</sup> ions concentration of 20 mg/l having fly ash as 10 g per 100 ml are prepared and pH of

By observing the Figure 5 and Figure 6, it can be concluded that the greater removal efficiency of Ni<sup>2+</sup> ions from the aqueous solution can be achieved in a short duration, by thorough mixing of adsorbent i. e. fly ash in the aqueous solution of metal ion. It is also indicated that the removal of Ni<sup>2+</sup> ions was mainly attributed to the effective adsorption reaction and not a precipitation reaction.

the reaction mixture was maintained within 5.5 to 9.0 which were strictly as per the limits of pH for industrial discharge water, as given by the Environment (Protection) Rules (India).

To understand the effect of pH along with the fly ash, the graphs have been plotted showing the variation in percentage efficiency of removal of  $\text{Ni}^{2+}$  ions

against pH at different contact time, under constant stirring condition in Figure 7 and under steady state condition in Figure 8.

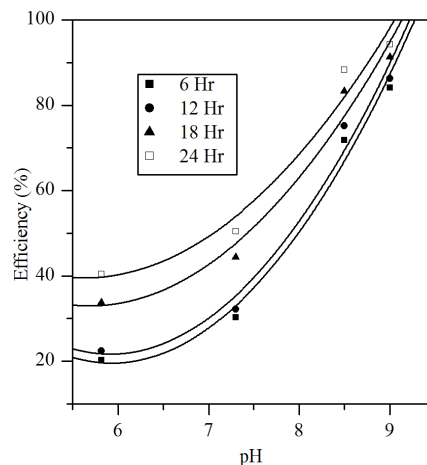


**Figure 7.** The plot of variation in removal efficiency (%) against initial pH at different contact time, under constant stirring

**Slika 7.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na početni pH pri različitom vremenu kontakta, tijekom konstantnog miješanja

It can be seen from the figure 7 that % efficiency of removal of  $\text{Ni}^{2+}$  ions increases with contact time. Also, it can be seen that at lower pH of the solution, the magnitude of percentage removal efficiency varies significantly with the variation in the

contact time but this variation become less significant with the increase in the pH of the solution. Thus, it can be concluded that at higher pH of the solution, the rate of removal of  $\text{Ni}^{2+}$  ions is almost independent of the contact time.



**Figure 8.** The plot of variation in removal efficiency (%) against initial pH at different contact time, under steady state

**Slika 8.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na početni pH pri različitom vremenu kontakta, u ravnotežnom stanju

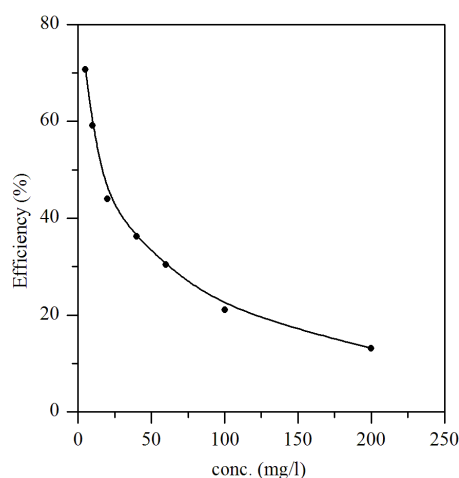
It can be depicted from the Figure 8 that the removal efficiency is proportional to the initial pH of the solution. It was observed that removal efficiency can be achieved

more than 90% as soon as the pH of the solution is 9. On the other hand the removal efficiency is only 20% in six hour contact time when initial pH is 5.82.

### Effect of initial concentration of Ni<sup>2+</sup> ions

To study the effect of initial Ni<sup>2+</sup> ions concentration on removal efficiency of fly ash, batch experiments were carried out in the present research work. For this purpose, Ni<sup>2+</sup> ions having varied concentration such as 5 mg/l, 10 mg/l, 20 mg/l, 40 mg/l, 60

mg/l, 100 mg/l and 200 mg/l were prepared and the removal efficiency of fly ash (10 g per 100 ml) for different initial ion concentration has been depicted in Figure 9.



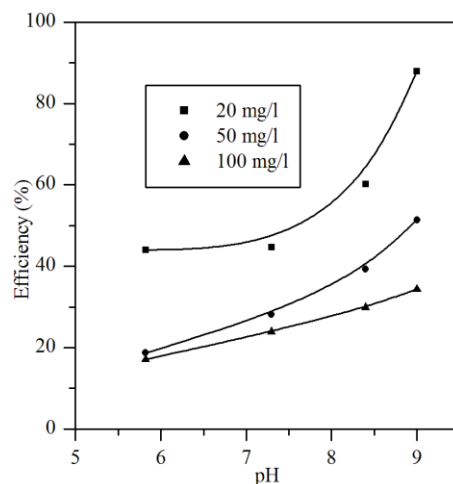
**Figure 9.** The plot of variation in removal efficiency (%) against initial concentration of Ni<sup>2+</sup> ions

**Slika 9.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na početnu koncentraciju Ni<sup>2+</sup> iona

It was observed from the close scrutiny of Figure 9 that percentage removal efficiency of Ni<sup>2+</sup> ions decreased with rise in the initial Ni<sup>2+</sup> ions concentration in the aqueous solution. The percentage removal efficiency of Ni<sup>2+</sup> ions which was 70.74 % when the initial Ni<sup>2+</sup> ions concentration was 5 mg/l and this removal efficiency decreased to 21.05% and 13.17% when the initial Ni<sup>2+</sup>

ions concentration increased to 100 mg/l and 200 mg/l respectively under the similar experimental conditions.

To study the effect of initial pH of the solution of on its removal efficiency for the different concentration of Ni<sup>2+</sup> ions under the simmler experimental conditions, graph is plotted and has been shown in Figure 10.



**Figure 10.** The plot of variation in removal efficiency (%) against initial pH at different initial ion concentration

**Slika 10.** Dijagram promjene učinkovitosti uklanjanja (%) u odnosu na početni pH pri različitim početnim koncentracijama Ni<sup>2+</sup> iona

It is depicted from the Figure 10 that the removal efficiency of the fly ash was maximum, when the initial pH of the aqueous solution was more for any initial ion concentration. When the Ni<sup>2+</sup> ions concentration was 20 mg/l, the maximum removal efficiency of Ni<sup>2+</sup> ions has been increased from 43.96% to

87.92% with the increase in pH of the solution from 5.82 to 9.0. For the same increase in pH of the solution the efficiency has increased from 18.72% to 51.4% for initial Ni<sup>2+</sup> ions of 50 mg/l and it has been increased from 17.14% to 34.32% when the initial Ni<sup>2+</sup> ions concentration was 100 mg/l.

## CONCLUSION

It was observed that SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> contents make up about 87% of the total content of the fly ash used. The coal fly ash sample contains approximately 91% particles of the size lesser than 75 μm and its pH was found in the range of 6.9 to 7.9. The fly ash used in the present study, was classified as class F. The X-ray diffraction pattern for this fly ash sample shows most of the minerals present are in oxide form and are mostly spherical in shape, whereas small amount of irregular shaped particles are also present. It was also found that zeta potential of fly ash increases continuously with pH of the solution.

Experimental results showed there was a high removal rate of Ni<sup>2+</sup> ions by adding fly ash and this removal is probably due to adsorption of Ni<sup>2+</sup> ions on the negatively charged surface of the fly ash. Even, the rate of removal of Ni<sup>2+</sup> ions is enhanced by the increasing the pH of the solution by addition of lime in the reaction mixture. This means the removal of Ni<sup>2+</sup> ions from solution is the combination of adsorption process and the precipitation phenomena. To sum up, fly ash could be used as an effective adsorbent to remove Ni<sup>2+</sup> from aqueous solutions under the optimal conditions.

**REFERENCES**

- [1] G. S. Sayler, J. D. Nelson, and R. R. Colwell, *Appl Microbiol.*; 30(1975)1, 91–96.
- [2] S. Al-Asheh and Z. Duvnjak, *Adv. Environ. Res.* 1(1997),194.
- [3] K. Kadirvelu, , Ph.D. Thesis, Bharathiar University, Coimbatore(1998), India.
- [4] V. K. Gupta, D. Mohan, S. Sharma, and K. T. Park, (1998) *The Environmentalist Journal* 19(1998) 2,129-136.
- [5] A. Rađenović, J. Malina, A. Štrkalj, *The Holistic Approach to Environment* 1(2011)3, 109-120.
- [6] M. Takaoka, T. Kawai, N. Takeda and K. Oshita, *Proceedings of Environmental Engineering Research* 39(2002):403-412.
- [7] A. R. Pourkhorshidi, M. Najimi, T. Parhizkar, F. Jafarpour and B. Hillemeier, *Cement Concrete Comp.*, 32(2010)10, 794–800.
- [8] K. Kadirveli and C. Namasivayam, *Advances in Environmental Research*, 7(2003)2, 471–478.
- [9] A. Ramesh, H. Hasegawa, T. Maki and K. Ueda, *Separation and Purification Technology* 56(2007)1, 90–100.