The Efficiency of Open Lime-Zeolite Channel System in Treatment of Acid Mine Drainage (AMD) Released from Sungun Copper Mine

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Abstract

Sungun copper mine is located inside Arasbaran forest protected area of Iran and AMD drained from tailing dump of the mine is a big challenge between the mine authorities and the residents, NGOs and Iranian Environmental Protection Organization. As the first AMD treatment system in Iran, a passive open lime-zeolite channel was implemented in Sungun mine and the efficiency was evaluated.

Increasing of pH cannot decrease the concentration of heavy metal down to permissible level and need to be combined with another mechanism to increase the efficiency. A system with combination of zeolite and lime would be an effective treatment method for Cu removal of AMD. The zeolite omits Cu from aqueous phase by means of adsorption (2.86 mg/g based on Langmuir isotherm model) whereas increasing of pH with limestone is recognized as effective mechanism for precipitation of Cu from AMD. However, based on experimental data and site specifications, zeolite- limestone open channel stablished at site to use two mechanisms of buffering and adsorption for treatment of AMD. Combining open zeolite channel with routine open lime channel (OLC) increased the efficiency of AMD treatment system.

A few months after opening of AMD treatment system of Sungun copper mine, remediation efficiency of the system was evaluated using two rounds of sampling and comparing with the pre-treatment sampling results. The results showed that Cu concentration has been decreased from about 45 mg/L down to less than 4 mg/L implying a decrease of more than 91% percent in Cu concentration by lime part of the system. The second sampling which was done after complete installation of the system showed a complete restoration of the AMD related to the copper with decrease of concentration down to detection limit of the instrument (0.03 mg/L). The system also decreased Al concentration 69%, Fe concentration 55% and Zn below detection limit. Furthermore, the treatment system has increased pH value to neutral condition which has the main rule in immobilization of the metals.

Keywords: AMD, Treatment, Lime, Zeolite, Sungun mine

Introduction

Chemical precipitation, ion exchange, adsorption and ultra-filtration are common treatment processes to eliminate heavy metals from waste waters (Coulson et al. 1994). One of the most popular method for the removal of heavy metals from the waste waters is adsorption (Fala et al. 2006, Cui et al. 2006), especially when large quantities or certain wastes are released from mining, industrial or agricultural activities. Sorbents comprise a wide range of organic, inorganic and synthetic products. Among them, CaCO3, dead biomass, blast\ furnace slag, fly ash, clay, tree bark, tea leaves and natural zeolite have potential to remove heavy metals from waste water (Bhattacharyya and Gupta 2006, Günay et al. 2007, Bailey et al. 1999). Treatment systems provide a controlled environment in which natural chemical and biological reactions which help in the treatment of acid mine drainage can occur (DEP 1999). Open limestone channels (OLCs) are passive treatment systems that achieve remediation mostly through chemical means (Kalin et al. 2006). Other passive systems are anoxic limestone drains, aerobic and anaerobic wetlands, and biological and abiotic permeable, reactive barriers (Johnson and Hallberg 2005). Very often, an OLC is preferred due to its low building and maintenance costs. Generally, OLCs are used to treat AMD under conditions of very high flow, because continually-moving water may erode any armoring from the limestone (Pavlick et al. 2005).

In this research Acid Mine Drainage (AMD) samples taken from Sungun copper mine subjected to several treatments through lab scale experiments to find the best scenario of remediation. To optimize final scenarios, another series of treatments have been conducted in batch and semi continuous condition of water flow. The final goal of the study is finding a method or combination of methods to immobilize the heavy metals and decrease their concentration in AMD down to the standard values.

Methods

Water samples have been taken from AMD drained from the tails of porphyry copper mine located at the northwest of Iran (Fig. 1). Sungun deposit hosts 1200 Mt of ore at low grade 0.76% Cu and ~100 ppm Mo (Siahcheshm et al. 2014) so the mining processes produce a huge amount of waste materials and store in tailing dumps. Pore fluids of tailings can become acidic through reactions of surface waters with exposed sulfide minerals. Consequently, AMD can be formed which has low pH to solve heavy metals of the tailings and release them into a river beneath of the dump. The mine tailings are being dumped in a valley named Pakhir valley with an average flow rate of 25 L/s. In order to find the best method for remediation of the AMD a series of experiment were designed.

At the first step, the AMD samples were subjected to five remediation materials including granular activated carbon, natural zeolite, iron filing, lime, and poly aluminum chloride (PAC). All of these materials except PAC were prepared in the same of size between 1.4 to 2.3 mm and 1 g of each material was added to the 100 mL of drain and stirred for 2 hours in 20 OC in an incubator shaking with a rate of 200 rpm. Concentration of residual Cu in the treated drainage was measured by atomic absorption spectrometry (AAS) using a Novaa 400, Analytik-Jena machine.

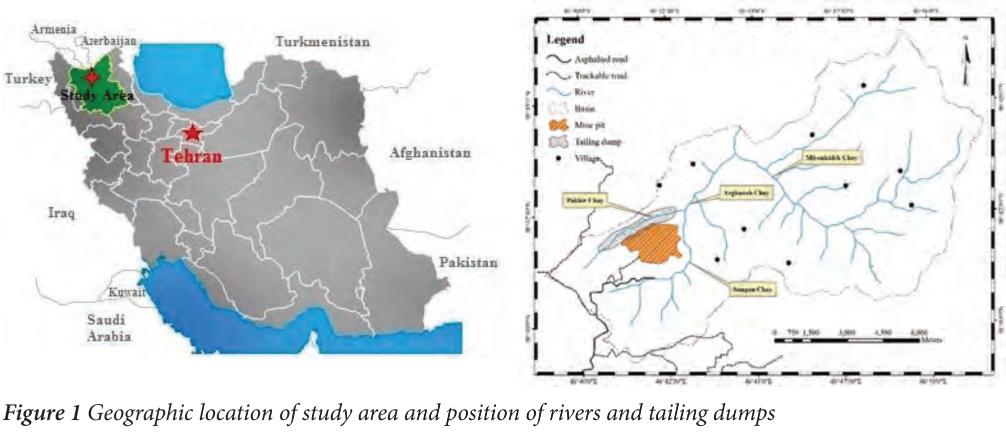

Figure 1 *Geographic location of study area and position of rivers and tailing dumps*

***Table 1** Physical properties and chemical composition parameters of AMD collected from "Sungun" mine*

Parameters	Unit	Value
pH	-	5.5
Total Dissolved Solid(TDS)	mg/L	1020
Cu2+	mg/L	34
Mn2+	mg/L	5.78
Ni2+	mg/L	0.51
Cd2+	mg/L	0.1
Al3+	mg/L	0.97
Fe3+	Mg/L	0.05

To evaluate the ability of PAC through coagulation-flocculation process on AMD treatment, 1.0 g of PAC and 100 mL of drainage sample were poured into an Erlenmeyer and the solution was stirred for 2 min in 20 oC in an incubator at a stirring speed of 200 rpm. Then it was kept for 30 min for efficient deposition of developed clots. Finally, Cu concentration in the treated drainage was determined. Two best options were selected according to the removal efficiency, cost and availability of materials and then the following batch and continuous tests were conducted on them.

To simulate the AMD treatment process, a physical model with dimension of 120 × 50 × 10 cm3 was made and placed on the bench with a slope of 2% to provide the necessary head for flow. To evaluate the performance of zeolite as one of the selected materials in AMD treatment under a semi continuous condition, the above mentioned reactor was filled up to 5 cm with zeolite grains of 2 to 5 mm. The drainage was entered to reactor with a flow rate of 7 L/h which caused a travel time about 21 min for the interred drainage through the model. Samples was taken with intervals of 30 min and assessment of Cu, Mn, Al and Ni concentration in the untreated and treated AMD samples were performed using AAS.

To evaluate the effect of solution pH on the ability of zeolite on copper removal from the drainage, 0.5 L drainage poured into a series of Erlenmeyer flasks and initial pHs were adjusted from 7 to 10 using NaOH. The treatments incubated for two days at room temperature to reach complete precipitation.

Results and Discussion

According to the chemical analysis of AMD sample collected from the toe of tailings of Sungun mine (Table 1), it is acidic with pH of 5.5 and present a high conductivity and high concentration of some heavy metal ions (Cu, Mn, Ni). These concentrations is higher than the permissible value recommended by WHO standard (1987-1999). The specific cyanic color of AMD and river bed is due to the high concentrations of copper in the solution. It is clear that this water drainage introduces sulphuric acid and toxic heavy metals (Cu, Mn and Ni) into the environment.

The initial concentration of Cu cations in the untreated AMD and its concentration in the treated samples have been shown in Fig. 2. All the treatment processes except coagulation and flocculation using PAC, can reduce

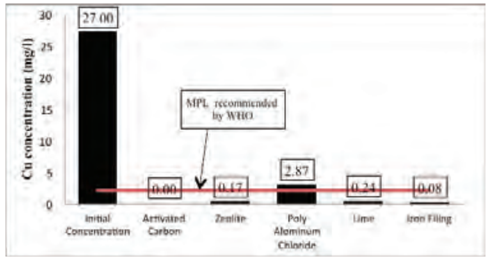

***Figure 2** Ability of various treatment processes on the decrease in copper concentration in batch scale model. (MPL: Maximum Permissible Level)*

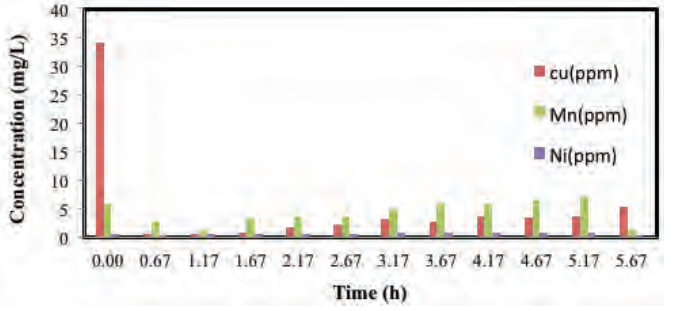

Figure 3 *Concentration of Cu, Mn and Ni heavy metals during semi continuous test using zeolite*

Cu concentration to reach permissible level. This indicates that the coagulation and flocculation process has lower efficiency than the sorption and pH reduction methods for Cu removal from AMD. Based on the results of this experiment and considering the local availability and low cost of zeolite and lime, these materials were selected for further investigations.

The results of treatment tests using zeolite (Fig. 3) revealed that concentrations of Cu, Mn and Ni ions in the initial and treated drainage sample indicate a good performance, especially in the removal of copper, but over time its performance decreased due of saturated of the surface active sites. In order to measure zeolite adsorption ability, a series of experiments were performed.

Through another test, AMD treatment using lime was investigated and the results demonstrated (Fig. 4) that the efficiency of lime for the removal of copper from solution is less than zeolite, and its ability decreases gradually during the test.

Treatment System Designing

According to the results of the experiments an open channel system was designed for treatment of Pakhir valley AMD in Sungun copper mine complex. To design this treatment system Pakhir valley discharge was measured for one year which show an average annually discharge of $0.025 m^3/s$. Also maximum instantaneous discharge with return periods of 25 and 50 years were calculated 20.5 and $6 m^3/s$, respectively.

The designed treatment system is including: initial box covered channel, diversion dam, limestone channel, zeolite channel and diversion channel. The initial box conveys AMD from the toe of the tailing to the diversion dam. Because every day new tailing was dumped into Pakhir valley, to prevent the contact of AMD with new tailings this concrete channel was designed as box and covered form. Limestone channel is a concrete one filling with limestone particles to increase pH which can precipitate heavy metal from equate phase. The particle size is 10 mm to increase contact area. According to the dimension of limestone channel (bed width is 80 cm and the depth is 40 cm) a length of about 120 m is necessary to decrease copper concentration down to 15 mg/L. So, AMD flowing through limestone particle precipitates a part of its copper due to increasing of pH and then inters to zeolite channel. The length of zeolite channel was optimized to be 250 m ac-

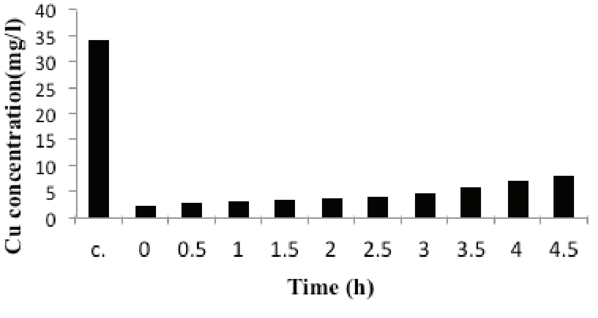

Figure 4 *Concentration of Cu during semi continuous test using lime*

Figure 5 *Open lime-zeolite channel system after construction*

cording to the results of the lab experiments which can provide enough time for absorption of copper. This channel can decrease Cu concentration down to 3 mg/L which is less than its MPL of irrigation water. To increase the resident time of AMD in limestone -zeolite channels several check dam were used in each25 meter.

One year after designing of AMD treatment system for Sungun copper mine, it was constructed and opened in late December 2016 (Fig. 5). In order to evaluate efficiency of system in remediation of AMD two rounds of sampling carried out one month after system opening (when only lime filter works) and three months after (when the both lime and zeolite filters work). The results showed that Cu concentration has been decreased from about 45 mg/L down to less than 4 mg/L implying a decrease of more than 91% percent in Cu concentration by lime part of the system. The second sampling which was done after complete installation of the system showed a complete restoration of the AMD related to the copper with decrease of that concentration down to detection limit of the instrument (0.03 mg/L). Table 2 also showed that this system more than Cu have decreased other metals like Al concentration decrease 69% percent, Fe concentration rate removal is 55% and Zn concentration reduced below detection limit.

Conclusion

The results of laboratory experiments showed that all the investigated compounds except the PAD could decrease the concentration of copper considerably. Due to feasibility of cost efficiency, lime and zeolite were selected to design a treatment system for Sungun copper mind AMD. Based on equilibrium experiments, lime has an ability to reduce the concentration of copper down to 15 mg/L, and changing the amount of lime has a little effect on the reducing of Cu concentration. Zeolite can decrease the concentration much more down to permissible value. Consequently, it seems a system with combination of zeolite and lime would be an effective treatment

Table 2 *System efficiency in terms of pollutant removal at Pakhir River (the values are in mg/L).*

pH	Zn	Pb	Mn	Fe	Cu	Cd	Al	Sampling Site	
6.1	1.92	ND	16.81	NM	44.49	0.31	2.26	Inlet of the system	Winter2017
7.3	1.43	ND	15.08	NM	3.84	0.29	1.25	Outlet of the limestone channels	
-	25.5	-	10.29	NM	91.36	6.45	44.7	Removing Percent	
5.4	3.06	0.38	8.315	1.024	46.35	ND	9.29	Before treatment system	Spring2017
7.1	ND	0.28	0.1450	0.507	ND	ND	2.85	Outlet of the Zeolite Channels	
-	100	26.3	98.2	50.5	100	-	69.3	Removing Percent	

NM: Not Measured ND: Not Detected

method for Cu removal of AMD. Based on experimental data and site specifications, it was recommended to use zeolite- limestone open channel to implement two buffering and adsorption agents for treatment of Sungun mine AMD. Construction of the designed system showed that the efficiency of the system is satisfactory, expectedly. Cu concentration was decreased from 45 mg/L down to less than detection limit (0.3 ppm) which is less than environmental permissible level for surface water. The treatment system also reduced concentration of some other metals like Zn, Al and Mn from 26 to near 100 percent of the initial value.

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