



A comparative study of chemical precipitation and electrocoagulation for treatment of coal acid drainage wastewater

M.S. Oncel^a, A. Muhcu^a, E. Demirbas^b, M. Kobyas^{a,*}

^a Gebze Institute of Technology, Department of Environmental Engineering, 41400 Gebze, Turkey

^b Gebze Institute of Technology, Department of Chemistry, 41400 Gebze, Turkey

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ABSTRACT

The present study provided a quantitative comparison between chemical precipitation and electrocoagulation (EC) for removal of heavy metals such as Fe, Al, Ca, Mg, Mn, Zn, Si, Sr, B, Pb, Cr and As from coal mine drainage wastewater (CMDW) at a laboratory scale. The optimum pH for removal of most of heavy metals from CMDW by the chemical precipitation using sodium hydroxide was 8 except for Ca, Sr and B (pH 10 or higher). The removal efficiencies at the optimum pH were varied from 28.4% to 99.96%. Influence of current density and operating time in the EC process was explored on the removal efficiency and operating cost. Results from the EC process showed that the removal of metals present in CMDW increased with increasing current density and operating time. The EC process was able to achieve higher removal efficiencies (>99.9%) at an electrocoagulation time of 40 min, a current density of 500 A/m² and pH of 2.5 as compared to the results obtained with the chemical precipitation at pH 8. The operating costs at the optimum operating conditions were also determined to be 1.98 €/m³ for the EC and 4.53 €/m³ for the chemical precipitation. The EC process was more effective than the chemical precipitation with respect to the removal efficiency, amount of sludge generated and operating cost. Electrocoagulation has the potential to extensively eliminate disadvantages of the classical treatment techniques to achieve a sustainable and economic treatment of polluted wastewater.

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Introduction

Acid mine drainage (AMD) generated from active and abandoned mining is a serious environmental problem with the potential of severe contamination of surface and groundwater [1–3]. AMD most commonly initiates from coal mining as materials exposed to water and atmospheric oxygen during the mining process and contains high concentration of FeS₂ [4]. In general, drainage from coal mines is not only of low pH but also includes high levels of sulphate and heavy metals such as Fe, Al, Mn, Ca and Zn [5,6]. High content of toxic metals and high acidity in AMD adversely affects surface water, groundwater and soil. These properties of mine drainage disrupt stream ecosystems and further aggravate the problem by creating yellow or white sediments [7]. Moreover, AMD is a highly acidic aqueous solution and formed through the chemical reaction of surface and shallow subsurface water with rocks containing sulphur-bearing minerals to give sulphuric acid. Heavy metals are not biodegradable and tend to accumulate in living organisms and many heavy metal ions are known to be toxic or carcinogenic. Toxic heavy metals for the treatment of industrial wastewaters include zinc, copper, nickel,

mercury, cadmium, lead and chromium [8]. Therefore, these wastewaters must be treated before being discharged into the environment.

Passive and active treatment methods have been applied for the treatment of AMD [5,9]. Passive systems include using anoxic limestone drains, aerobic wetlands, compost reactors, permeable reactive barriers and packed bed iron-oxidation bioreactors [10]. Bioreactors represent a passive-treatment option for removal of sulphate and metals from AMD. However, their treatment performance can be quite variable depending on a number of factors including organic substrate sources and their degradation, mine-water chemistry, microbiological diversity and activities, reactor configuration and hydraulics [11]. Wetlands also are ineffective in areas with rocky soils and steep slopes [12]. Close proximity to floods and large land requirements negatively impact wetland use. The most widely used active treatment process for AMD is based on chemical neutralization and hydroxide precipitation of metals [13,14]. Most active treatment involves pH adjustment and removal by precipitation as a result of the formation of oxy/hydroxides. pH adjustment is needed for treatment of large quantities of AMD. Active treatment enhances the treatment efficiency with using of chemicals but causes a large economic burden owing to the high cost of maintenance and chemicals, and this process requires continuous operation [5]. The disadvantages of the traditional chemical treatment are high cost

* Corresponding author. Tel.: +90 262 6053214; fax: +90 262 6538490.
E-mail address: kobyas@gyte.edu.tr (M. Kobyas).

of the chemical reagents, inefficient removal of sulphate and production of a bulky sludge needed to be disposed of. Passive systems are economical as compared with the active treatment but require longer retention times and greater space. Therefore, they are not appropriate for treatment of large-scale mine drainage. Although the passive treatment has been implemented on full-scale sites in several countries, treatment efficiency can be uncertain because of seasonal changes in flow rate and temperature, and the systems are apt to fail during long-term operation.

Current AMD treatment technologies are either inadequate or too expensive. Thus high cost of conventional treatment technologies has produced economic pressure and has caused engineers to search for cost effective and environmental friendly technologies to treat AMD. In the past decades, research efforts have been directed towards advanced techniques for removal of heavy metals from AMD as well as industrial wastewater. Several techniques such as chemical precipitation, oxidation, reduction, coagulation, solvent extraction and adsorption have been commonly employed for the removal of heavy metal ions [8,15].

Currently, there are a number of studies about hydroxide precipitation process using lime, $\text{Ca}(\text{OH})_2$ and NaOH for removal of heavy metals in the literature. $\text{Cu}(\text{II})$ and $\text{Cr}(\text{VI})$ ions from wastewater were evaluated. Maximum precipitation of $\text{Cr}(\text{III})$ occurred at pH 8.7 with the addition of $\text{Ca}(\text{OH})_2$ and the concentration of chromate was reduced from 30 to 0.01 mg/L. The optimum pH for maximum copper precipitation was 12.0 for both $\text{Ca}(\text{OH})_2$ and NaOH and the concentration of copper was reduced from 48.51 to 0.694 mg/L [16]. Fly ash was used as a seed material to enhance lime precipitation. The fly ash-lime carbonation treatment increased the particle size of the precipitate and significantly improved the efficiency of chromium, copper, lead and zinc removals. The concentrations of chromium, copper, lead and zinc in effluents were reduced from initial concentration of 100.0 mg/L to 0.08, 0.14, 0.03 and 0.45 mg/L [17]. Chemical coagulation and precipitation by lime were employed to treat synthetic wastewater consisting of Zn, Cd, Mn and Mg at the concentration of 450, 150, 1085 and 3154 mg/L. It was found that the optimum pH was greater than 9.5 for treatment of the wastewater to meet the Wastewater Standard of the Ministry of Industry [18]. Hydroxide precipitation may have some limitations: (i) hydroxide precipitation produces secondary wastes such as metal hydroxide sludge and gypsum which are highly regulated and have costly disposal requirements; (ii) some metal hydroxides are amphoteric, and the mixed metals create a problem using hydroxide precipitation since the ideal pH for one metal may put another metal back into solution; (iii) when complexing agents are in the wastewater, they inhibit metal hydroxide precipitation [19].

Removal efficiencies of Cu, Cr and Ni from metal plating wastewater using an Fe–Al electrode pair at a current density of 10 mA/cm², pH 3.0 and an EC time of 20 min with energy and electrode consumptions of 10.07 kWh/m³ and 1.08 kg/m³ were achieved with electrocoagulation (EC) process as 100% [20]. Optimum conditions for removal of $\text{Cr}(\text{VI})$ with a concentration of 1470 mg/L were determined to be 7.4 A, pH 1.84 and 70 min and the removal efficiency by the EC process was 100% [21]. The performance of simultaneous removal of Cu, Ni, Zn and Mn from a model wastewater was investigated with the EC using iron electrodes and removal efficiency of more than 96% was obtained for all metals with a total energy consumption of 49 kWh/m³ at 25 mA/cm² [22]. Treatment of spent final rinse water of zinc phosphate from an automotive assembly plant was performed in an electrochemical cell equipped with aluminium or iron plate electrodes. The highest phosphate and zinc removal efficiencies at optimum conditions were 97.7% and 97.8% for Fe electrode (60.0 A/m², pH 3.0 and operating time of 15.0 min), and 99.8% and 96.7% for Al electrode (60.0 A/m², pH 5.0 and operating time of 25.0 min)

[23]. Removal efficiencies of arsenic in a batch EC reactor using Al and Fe electrodes with monopolar parallel electrode connection mode were 93.5% for Fe electrode at 12.5 min and pH 6.5 and 95.7% for Al electrode at 15 min, pH 7 and at 2.5 A/m² [24]. Ni and Zn removals from Ni and Zn plating processes by the EC using stainless steel electrodes were achieved with 100% at 9 mA/cm² and pH 6 [25]. Separation of some heavy metal ions such as Fe, Ni, Cu, Zn, Pb and Cd with different initial concentrations in the range of 50–600 mg/L and initial pH between 7.5 and 7.8 was studied with electrocoagulation–electroflotation process. The removal rate was 95% at 15 min [26]. The removal efficiency of Mn^{2+} from synthetic wastewater containing 100 mgMn²⁺/L by the EC was obtained for 78.2% at 6.25 mA/cm² and pH 7 [27].

There has been no direct report as yet being published for the treatment of coal mine drainage wastewater (CMDW) by the EC process, despite the considerable success of this process for the treatment of industrial wastewater, groundwater and surface water containing dissolved metal ions in the literature [21–26,28,29,31]. Therefore, the present study was aimed to focus on treatment of CMDW by the EC process using iron plate electrodes. Effects of current density and operating time for the removal of Fe, Al, Mn, Mg, Pb, Zn, Cr, As and Sr from CMDW at a laboratory scale were investigated to determine the optimum operating conditions in the EC process. The chemical precipitation by sodium hydroxide (NaOH) was used to treat CMDW. The operating costs for the EC and chemical precipitation processes at the optimum operating conditions were calculated.

Electrocoagulation and chemical precipitation processes

Electrocoagulation process

EC involves the generation of coagulants in situ by dissolving sacrificial anodes such as aluminium or iron upon application of a direct current. Iron is oxidized in an EC reactor at anodic sites to Fe^{2+} ions which dissolve to Fe^{3+} . The wastewater solution becomes green and bubbles of gas at cathode are observed during the EC process. The effluent becomes clear and then a green and yellow sludge are formed which are attributed to Fe^{2+} and Fe^{3+} hydroxides. The following major reactions take place in the EC process [28–30]:



The metal ions can form wide ranges of coagulated species and metal hydroxides, or precipitate and adsorb dissolved contaminants at an appropriate pH value. When CMDW contains high concentrations of different metals such as Fe, Al, Ca, Mg, Mn, Zn, several competitive reactions occur either at the cathode (metals deposition on the cathode electrodes) or in solution (precipitation and co-precipitation of metals with ferrous hydroxides). An amount of metal(s) is removed by cathodic reduction (metal is formed and deposited on the cathodes electrodes) according to the following reaction (5) [31]:



Several metals can be simultaneously or successively reduced on cathode electrodes. According the electrochemical motive force series the order of metals deposition should be as follows: $\text{Sr} > \text{Ca} > \text{Mg} > \text{Al} > \text{Mn} > \text{Zn} > \text{Cr} > \text{Fe} > \text{Pb}$. Furthermore, the

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