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Communication

Studies on a Mg-Al-Zn Alloy as an Anode for the Removal of Fluoride from Drinking Water in an Electrocoagulation Process

The present study provides an electrochemical coagulation process for the removal of fluoride from drinking water using a Mg-Al-Zn alloy, magnesium, aluminum and mild steel as anodes and stainless steel as cathode. Various parameters which may affect the removal efficiency of fluoride from water, such as pH, concentration of fluoride, current density, temperature and co-existing ions were studied. Electrochemical coagulation was carried out for 30 min at an initial fluoride concentration of 5.0 mg/L and this was reduced to 0.2 mg/L. A maximum removal efficiency of 96% was achieved with a magnesium alloy as anode and stainless steel as cathode at a current density of 0.2 A/dm² and a pH of 7.0.

Keywords: Drinking water; Electrocoagulation; Fluoride

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1 Introduction

Fluorine is an essential element for human health. However, an excess intake of fluoride can lead to various diseases such as osteoporosis, arthritis, brittle bones, cancer, infertility in women, brain damage, Alzheimer's syndrome, and thyroid disorder [1]. Fluoride concentrations less than 1 mg/L are beneficial to prevent skeletal and dental problems. The 1984 World Health Organization (WHO) guidelines suggest optimum levels of fluoride concentration to be 1 to 1.2 mg/L [2]. Fluoride ions can be found in wastewaters derived from semiconductor, metal processing, fertilizer and glass-manufacturing industries [3–5]. The discharge of such wastewaters into surface water can lead to the contamination of groundwater. Many people in the world are affected by the fluorosis, especially in China, India, Pakistan and Thailand [6].

In order to meet water quality standards, further treatment of water is required. Fluoride removal techniques may be based on the principles of adsorption [7], ion exchange [8], precipitation-coagulation [9, 10], membrane separation process [11, 12] and electro dialysis [13–15]. Several adsorbent materials have been tried in the past to find an efficient and economical defluorinating agent. Activated alumina, activated carbon, activated alumina coated silica gel, calcite, activated saw dust, activated coconut shell carbon, activated fly ash, groundnut shell, coffee husk, rice husk, magnesite, serpentine, tricalcium phosphate, bone charcoal, activated soil sorbent, carbon, defluoron-1, defluoron-2, etc., are some of the different adsorbent materials reported in the literature [16–24]. The most commonly used adsorbents are activated alumina and activated carbon. The adsorption process is highly pH selective, has a low adsorption capacity, poor physical integrity, it requires acidification and

pretreatment and its effectiveness for fluoride removal reduces after each regeneration. In the case of ion-exchange processes, fluoride can be removed from water with a strongly basic anion-exchange resin containing quaternary ammonium functional groups. But the limitations like the cost of the resin and regeneration and waste disposal make the process uneconomical. In the coagulation-precipitation method lime and alum are the most commonly used coagulants. The addition of lime leads to precipitation of fluoride as insoluble metal fluoride and raises the pH value of the water up to 11–12. The limitations are a very high maintenance cost, the amount of space needed and high residual aluminum in treated water. In the recent years, membrane processes has emerged as a preferred alternative to provide safe drinking water. Due to disadvantages like the high cost of the membrane, brine disposal and post treatment of water the process is prevented from being economical.

Recent research has demonstrated that electrochemistry offers an attractive alternative to the above-mentioned traditional methods for treating wastewaters [25–31]. Electrocoagulation, which is one of these techniques, is the electrochemical production of destabilization agents that brings about charge neutralization for pollutant removal and it has been used for water or wastewater treatment. The advantages of electrocoagulation include high particulate removal efficiency, compact treatment facility, and possibility of complete automation.

Although, there are numerous reports related to removal of fluoride by electrocoagulation using aluminum as the anode [32–38]. However, reports of using magnesium and magnesium alloy as the anode material for the removal of fluoride by electrocoagulation are scarce. Apart from the above, the main disadvantage of aluminum electrodes is the residual aluminum level (The USEPA guidelines suggest maximum contamination is 0.05–0.2 mg/L) present in the treated water due to cathodic dissolution [39]. This will create health problems like cancer. In the case of magnesium electrodes,

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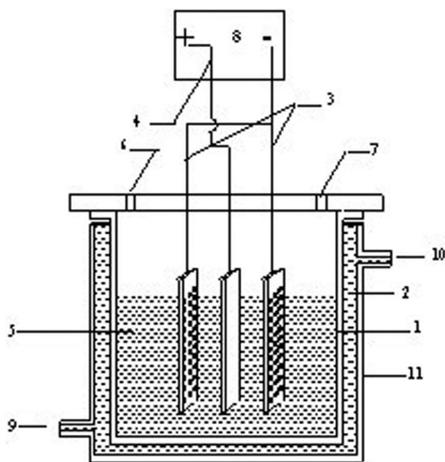


Figure 1. Laboratory scale cell assembly, (1) cell; (2) thermosstatic water; (3) stainless steel cathode; (4) anode; (5) electrolyte; (6 and 7) holes to introduce pH sensor and thermometer; (8) dc source; (9) inlet of thermosstatic water; (10) outlet of thermosstatic water and (11) thermostat.

there is no such disadvantage, as the USEPA guidelines suggest maximum values of magnesium in water to be 30 mg/L.

This article presents the results of the studies undertaken on the electrochemical removal of fluoride using a magnesium alloy, magnesium, aluminum and mild steel as anodes and stainless steel as the cathode. To optimize the maximum removal efficiency of fluoride, different parameters like the effect of anode materials, initial fluoride concentration, temperature, pH, current density and co-existing ions like carbonate, phosphate, silicate and arsenic were studied.

2 Experimental

2.1 Cell Construction and Electrolysis

The electrolytic cell (see Fig. 1) consisted of a 1.0 L Plexiglas vessel that was fitted with a polyvinyl chloride (PVC) cell cover with slots to introduce the electrodes, pH sensor, a thermometer and the electrolytes. Magnesium alloy (Magnesium Elektron Ltd., AZ31 consisting of 3.0 wt% Al and 1.0 wt% Zn), magnesium (commercial grade, India) aluminum (commercial grade, India) and mild steel (commercial grade, India) of surface area 0.02 m² acted as the anode. The cathodes were stainless steel (SS 304; SAIL, India) sheets the same size as the anode, placed at an interelectrode distance of 0.005 m. The temperature of the electrolyte was controlled to the desired value with a variation of ± 2 K by adjusting the rate of flow of thermosstatically controlled water through an external glass cooling spiral. A regulated direct current (DC) was supplied from a rectifier (10 A, 0–25 V, Aplab model).

The fluoride (NaF, Analar Reagent) was dissolved in tap (drinking) water for the required concentration (5–50 mg/L). 0.90 L of solution was used for each experiment, which was used as the electrolyte. Table 1 shows the composition of the water. The pH of the electrolyte was adjusted, if required, with 1 M HCl or 1 M NaOH solutions before adsorption experiments.

2.2 Analysis

The analysis of fluoride was carried out using a Fluoride Ion Selective Electrode (TOA-DKK, Japan). Magnesium, aluminum, zinc and

Table 1. Composition of the water.

S. No.	Parameters	Simulated Water
1	pH	7.01
2	TDS, mg/L	350
3	Calcium, mg/L as Ca	60
4	Magnesium, mg/L as Mg	10
5	Chloride, mg/L as Cl	95
6	Fluoride mg/L as F	5–50 ^{a)}
7	Sulphate, mg/L as SO ₄	11
8	Silicate, mg/L as SiO ₂	5–15 ^{a)}
9	Arsenic, mg/L as As	0.2–5 ^{a)}
10	Phosphate, mg/L as P	5–50 ^{a)}
11	Carbonate, mg/L	5–250 ^{a)}

^{a)} The addition of ions added dependent on the concentration required.

iron were characterized by Ion Chromatography (Metrohm Ltd.). The SEM and EDAX analysis of magnesium hydroxide was carried out with a Scanning Electron Microscope (SEM) made by Hitachi (model s-3000h). The Fourier transform infrared spectrum of magnesium hydroxide was obtained using Nexus 670 FTIR spectrometer made by Thermo Electron Corporation, USA.

3 Results and Discussion

3.1 Effect of Anode Material

It is well known that water treatment with coagulants such as aluminum (alum, Al(SO₄)₃ · 18 H₂O), ferric chloride (FeCl₃) and ferric sulfate (Fe(SO₄)₃ · 7 H₂O) are effective in removing fluoride from drinking water. It has also been reported that calcium salts have been tested for the removal of fluoride from ground water. Fluoride present in the water will be removed by adsorption with metal hydroxides produced from the respective coagulants. The main disadvantage for the above process is the presence of anions like chloride and sulfate that will reduce the removal efficiency and will increase the Total Dissolved Solids (TDS) in the treated water. So, to overcome the above difficulties, in the present investigation magnesium alloy, magnesium, aluminum, and mild steel were used as anode materials (in situ generation of the coagulants) and stainless steel was used as the cathode material. The electrochemical ion generation has several distinct advantages. Coagulants introduced without corresponding sulfate or chloride ions are more efficient at removing contaminants from water. By eliminating competing anions and using a highly pure coagulant source, lower residual metal concentrations are obtained and less sludge is produced than when metal salts are utilized. A contaminant free ion source allows maximum adsorptive removal of the various dissolved forms of metals that could be present and require treatment. Contaminants present in industrial grade ferrous sulfate and aluminum salts end up in either the treated effluent or sludge cake. If flow rates or contaminant loads fluctuate, chemical treatment systems are difficult to operate but this is not in the case with electrochemical processes.

During the electrolysis of magnesium alloy, magnesium, aluminum or iron, hydroxides of micro-flocs are formed rapidly by anodic dissolution. After the electrolysis process, the water was gently stirred for few minutes for agglomeration of micro-flocs into larger flocs that settle more easily. During this flocculation process all kinds of micro particles and negatively charged ions are attached to

Table 2. Effect of different anode materials on the removal of fluoride from water.

Anode material	Voltage (V)	Concentration of Fluoride (mg/L)		Removal Efficiency (%)
		Initial	Final	
Magnesium alloy	1.5	5.0	0.2	96
Magnesium	2.1	5.0	2.2	56
Aluminum	2.2	5.0	0.35	93
Mild steel	2.1	5.0	1.7	66

Conditions: pH of the electrolyte, 7.0; temperature, 305 K; cathode, stainless steel; current density, 0.2 A/dm²; duration, 30 min.

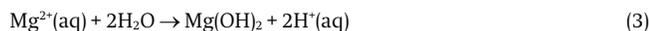
the flocs by electrostatic attraction. Fluoride is also adsorbed onto coagulated flocs. The possible reactions of magnesium alloy, aluminum and mild steel for the formation of hydroxides are as follows:

At the cathode:

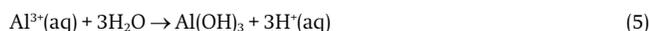


At the anode:

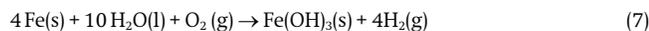
(when magnesium alloy/magnesium is used as the anode material),



(when aluminum is used as the anode material),



(when iron is used as the anode material),



Fluoride adsorbed on hydroxide flocs can be removed by filtration. From Tab. 2 it was concluded that the magnesium alloy electrode was more effective in removing fluoride than mild steel and the removal efficiency was very close to the aluminum electrode. The removal efficiencies for magnesium alloy, magnesium, aluminum and mild steel are 96, 56, 93 and 66% respectively. To evaluate the alternative anode material for fluoride removal from water, further studies were carried out with magnesium alloy as the anode material.

3.2 Effect of Initial Fluoride Concentration

Figure 2 shows the effect of fluoride concentration on its removal from the water in the concentration range of 5 to 50 mg/L. From the table it can be seen that as fluoride concentration increased, the removal efficiency decreased from 96 to 20%. This is because with increasing fluoride concentration, the ratio of aqueous fluoride to available complexation sites increased, leading to competition for complexation sites. With increasing competition the rate of fluoride removal became limited by the generation rate of new sites for

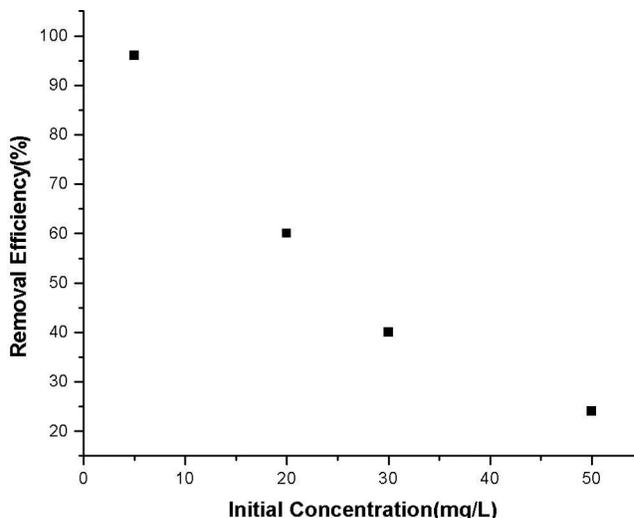


Figure 2. Effect of initial concentration of fluoride on fluoride removal from drinking water. Conditions: solution pH, 7.0; solution temperature, 305 K; anode, magnesium alloy; cathode, stainless steel; current density, 0.2 A/dm²; duration, 30 min.

fluoride adsorption. Under this scenario, the removal kinetics should become zero order with respect to fluoride at high concentrations. The ratio of dissolved fluoride to available magnesium hydroxide was sufficiently low at lower fluoride concentration and there was no competition between fluoride species for complexation sites. This situation resulted in removal kinetics that were first order in fluoride concentration removal from the drinking water.

3.3 Effect of pH

pH is one of the most important factors affecting the performance of electrochemical processes. To examine this effect, a series of experiments was carried out using 5.0 mg/L fluoride containing solutions, with an initial pH varying in the range 3 to 10. The results are presented in Fig. 3. The removal efficiency of fluoride increased with increasing pH and the maximum removal efficiency was obtained at pH 7.0. It was found that the maximum removal efficiency for fluoride was 96% at pH 7.0 and the minimum efficiency was 20% at pH 10. At pH ≤ 6.5, the oxide surfaces exhibited a net positive charge or the positive charge density was higher and adsorption of anionic fluoride was enhanced by coulombic attraction. At alkaline pHs, the oxide surface has a net negative charge and would tend to repulse the anionic fluoride in solution.

3.4 Effect of Current Density

The amount of fluoride removal and the removal rate was increased by increasing the current density [39]. The removal efficiencies were 34, 42, 58, 70 and 96% for current densities of 0.02, 0.04, 0.06, 0.08 and 0.2 A/dm², respectively. The results are presented in Fig. 4. Furthermore, the amount of fluoride removal depended upon the quantity of adsorbent generated, which is related to the time and current density [40]. As expected, the amount of fluoride adsorption increased with an increase in current density, which indicated that

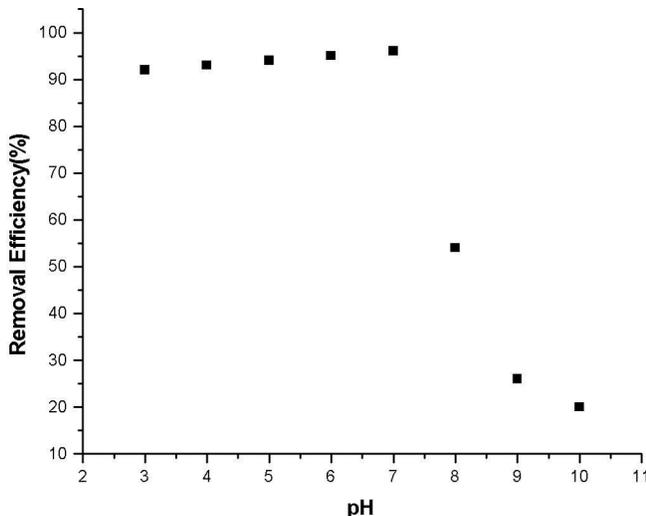


Figure 3. Effect of electrolyte pH on the removal of fluoride from drinking water. Conditions: fluoride concentration, 5.0 mg/L; solution temperature, 305 K; anode, magnesium alloy; cathode, stainless steel; current density, 0.2 A/dm²; duration, 30 min.

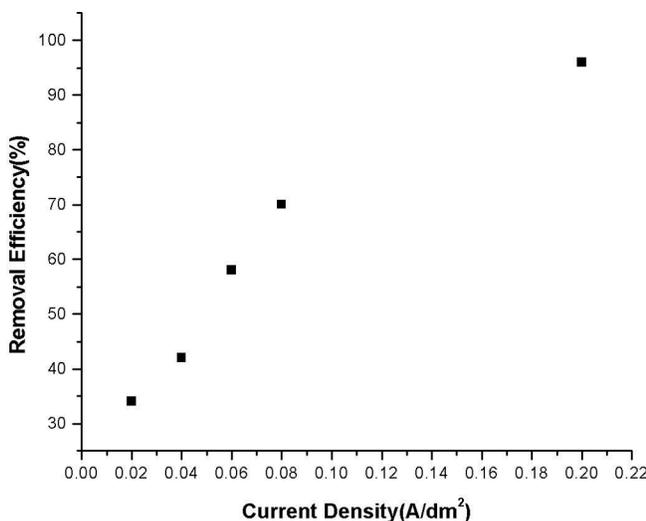


Figure 4. Effect of current density on the removal of fluoride from drinking water. Conditions: fluoride concentration, 5.0 mg/L; solution pH, 7.0; solution temperature, 305 K; anode, magnesium alloy; cathode, stainless steel; duration: 30 min.

adsorption depends up on the availability of binding sites for fluoride.

3.5 Effect of Temperature

The removal efficiencies were 70, 96, 96, 96 and 97% for temperatures of 293, 305, 313, 323, 333 K, respectively. The results are presented in Fig. 5. From the results, it was found that at low temperatures (<305 K) the removal efficiency was 24% lower than that at room temperature. At lower temperatures the dissolution of the anode was less and the amount of formation of hydroxide was very low for complexation with fluoride [41].

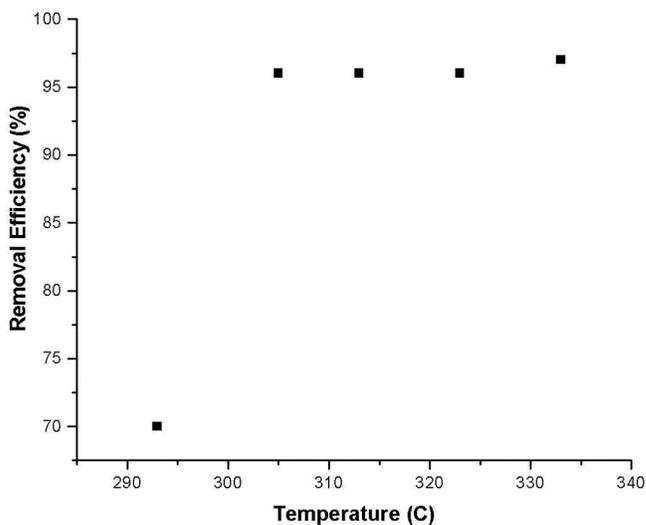


Figure 5. Effect of electrolyte temperature on the removal of fluoride from drinking water. Conditions: fluoride concentration, 5.0 mg/L; solution pH, 7.0; anode, magnesium alloy; cathode, stainless steel; current density, 0.2 A/dm²; duration, 30 min.

3.6 SEM, EDAX and FTIR Analysis

Figure 6 shows the scanning electron microscope (SEM) image of the anode before and after treatment. The SEM image indicated the presence of fine coagulant particles on the surface.

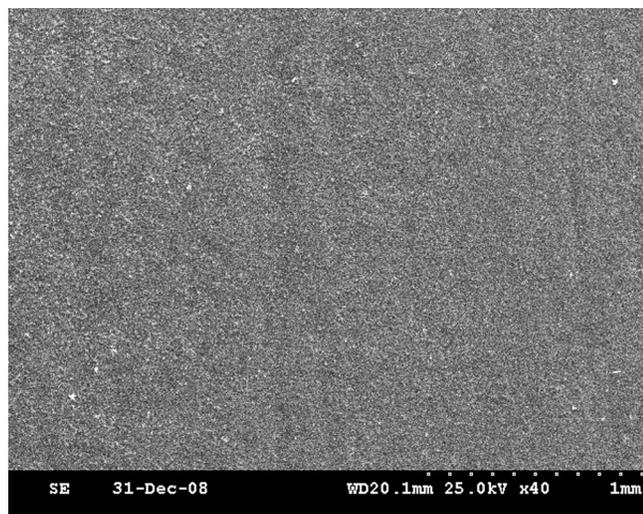
Energy-dispersive analysis of X-rays was used to analyze the elemental constituents of fluoride-adsorbed magnesium hydroxide, as shown in Fig. 7. It showed the presence of fluoride in small amounts appeared in the spectrum, other than the principal elements of Mg and O. EDAX analysis provided direct evidence that fluoride was adsorbed on magnesium hydroxide. Other elements detected in the adsorbed magnesium hydroxide came from adsorption of the conducting electrolyte, chemicals used in the experiments, alloying and the scrap impurities of the anode and cathode.

Figure 8 presents the FT-IR spectrum of fluoride-magnesium hydroxide. The sharp and strong peak at 3698.07 cm⁻¹ was due to the O-H stretching vibration in the Mg(OH)₂ structures. The 1639.72 cm⁻¹ peak indicated the bending vibration of H-O-H. A broad absorption band at 3448.60 cm⁻¹ implied the transformation from free protons into a proton-conductive state in brucite. The strong peak at 475.51 cm⁻¹ was assigned to the Mg-O stretching vibration. The spectrum data was in good agreement with the reported data [42]. Mg-F was observed in -OH stretching region [43].

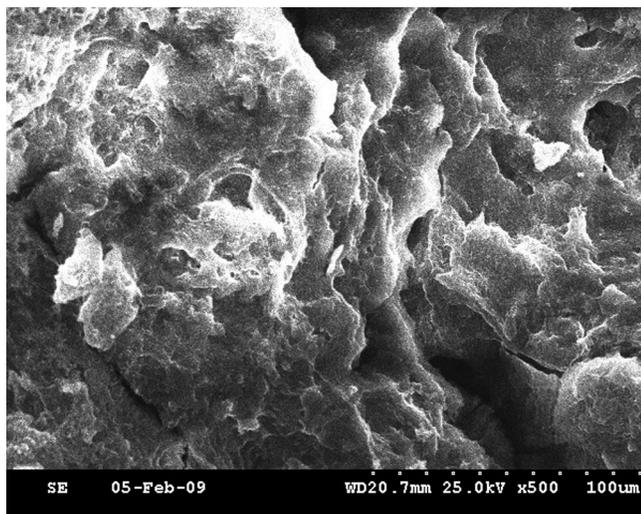
3.7 Effect of Coexisting Anions

3.7.1 Carbonate

The effect of carbonate on fluoride removal was evaluated by increasing the carbonate concentration in the electrolyte from 5 to 250 mg/L. The results are presented in Tab. 3. The removal efficiencies were 96, 96, 66, 54, 28 and 18% for carbonate ion concentrations of 0, 2, 5, 65, 150 and 250 mg/L, respectively. From the results it was found that the removal efficiency of fluoride was not affected by the presence of carbonate below 2 mg/L. A significant reduction in removal efficiency was observed above 5 mg/L carbonate concen-



(a)



(b)

Figure 6. SEM image of the anode (a) before and (b) after treatment.

tration, due to passivation of the anode resulting in a hindrance in the dissolution process of the anode.

3.7.2 Phosphate

The concentration of phosphate ion was increased from 2 to 50 mg/L, which is the contaminant range of phosphate in ground water. The removal efficiency for fluoride was 96, 96, 64, 60 and 54% for 0, 2, 5, 25 and 50 mg/L of phosphate ion, respectively. The results are presented in Tab. 3. There was no change in the removal efficiency of fluoride below 2 mg/L of phosphate in the water. At higher concentrations (and above 5 mg/L) of phosphate, the removal efficiency decreased drastically. This was due to the preferential adsorption of phosphate over fluoride as the concentration of phosphate increased.

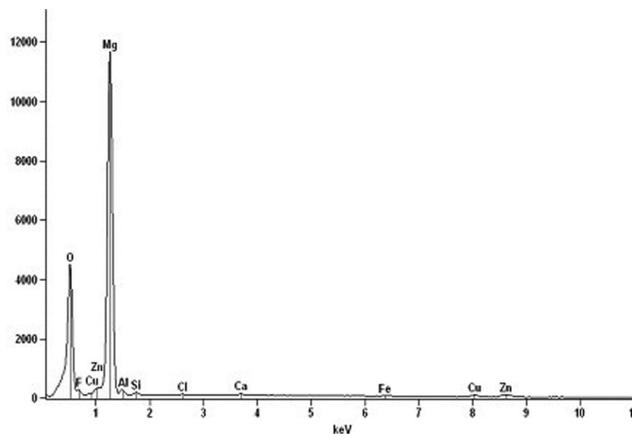


Figure 7. EDAX spectrum of fluoride-adsorbed magnesium hydroxide.

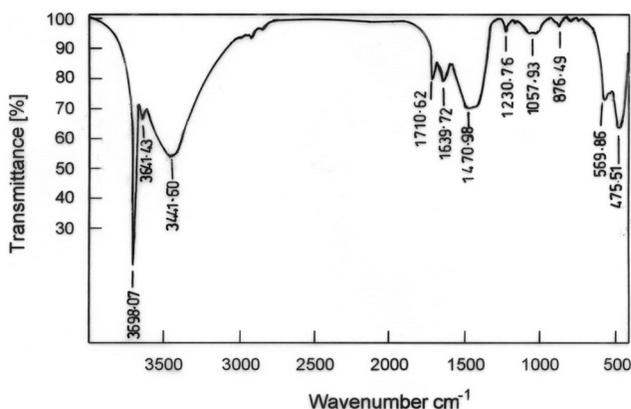


Figure 8. FTIR spectrum of fluoride-adsorbed magnesium hydroxide.

3.7.3 Silicate

The effect of silicate on the removal efficiency of fluoride is presented in Tab. 3. From the results, it was found that no significant change in fluoride removal was observed, when the silicate concentration was increased from 0 to 2 mg/L. The respective efficiencies for 0, 2, 5, 10 and 15 mg/L of silicate are 96, 96, 64, 54 and 30%. The removal of fluoride decreased with increasing silicate concentration from 2 to 15 mg/L. Further increases in silicate concentration decreased the fluoride removal efficiency. In addition to preferential adsorption, silicate can interact with magnesium hydroxide to form soluble and highly dispersed colloids that are not removed by normal filtration.

3.7.4 Arsenic

From the results it was found that the efficiency decreased from 96 to 38% by increasing the concentration of arsenate from 0.2 to 5 mg/L. The effect on removal efficiency of fluoride is presented in Tab. 3. Like phosphate ions, this was due to the preferential adsorption of arsenic over fluoride as the concentration of arsenate increases. When arsenic ions are present in water to be treated they will compete greatly with fluoride ions for binding sites.

Table 3. Effect of the addition of carbonate, phosphate, silicate and arsenic into the electrolyte on the removal of fluoride from water.

Concentration (mg/L)	Voltage (V)	Concentration of Fluoride (mg/L)		Removal Efficiency (%)
		Initial	Final	
Carbonate				
Nil	1.5	5.0	0.2	96
5.0	1.8	5.0	1.7	66
65.0	2.0	5.0	2.3	54
150.0	2.2	5.0	3.6	28
250.0	2.7	5.0	4.1	18
Phosphate				
Nil	1.5	5.0	0.2	96
5.0	1.8	5.0	1.8	64
25.0	2.0	5.0	2.0	60
50.0	2.6	5.0	2.3	54
Silicate				
Nil	1.5	5.0	0.2	96
5.0	1.8	5.0	1.8	64
10.0	2.3	5.0	2.3	54
15.0	2.6	5.0	3.5	30
Arsenic				
Nil	1.5	5.0	0.2	96
0.2	1.5	5.0	0.3	94
0.5	1.5	5.0	1.0	80
2.0	1.9	5.0	2.5	50
5.0	2.0	5.0	3.5	30

4 Conclusions

The results showed that a maximum removal efficiency of 96% was achieved at a current density of 0.2 A/dm² and a pH of 7.0 using magnesium alloy as anode and stainless steel as cathode. The magnesium hydroxide generated in the cell removed the fluoride present in the water and reduced the fluoride concentration to 0.2 mg/L. Further to this, no metal contaminants like Mg, Al, and Zn were present in the treated water. The results indicate that it is possible to use Mg-Al-Zn alloy as an alternative anode material for the removal of fluoride from drinking water. The results also indicate that the process can be scaled up to higher capacity and used to eradicate skeletal and dental problems.

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