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Removal of zinc and cadmium from an aqueous solution using limestone

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Abstract

This study investigates the removal of Zn(II) and Cd(II) ions from aqueous solutions using local Georgian limestone as a natural adsorbent. Limestone is a cost-effective, readily available, mechanically durable, and environmentally friendly mineral. For the first time, the adsorption capacity of this specific mineral was systematically evaluated. The effects of key operational parameters, including adsorbent dosage, contact time, initial metal ion concentration, and solution pH, on the adsorption efficiency of zinc and cadmium were thoroughly examined. Optimal process parameters were identified, yielding adsorption efficiencies of 82% for zinc and 69% for cadmium. The corresponding maximum adsorption capacities were determined to be 28 mg/g for zinc and 2.30 mg/g for cadmium.

Keywords: Limestone; Heavy Metals; Adsorption; Zinc; Cadmium; Degree of Adsorption.

1. Introduction

Contamination of aquatic ecosystems by heavy metals represents one of the most critical environmental challenges today. Rapid global industrialization, urbanization, and agricultural activities continue to increase toxic metal concentrations in both surface and groundwater resources. Heavy metals such as Cd, Pb, Zn, Cu, and Cr are characterized by high toxicity, persistent chemical stability, and a strong tendency to bioaccumulate in living organisms. Although essential trace elements like copper and zinc are necessary for human physiological processes, they pose severe health hazards when threshold limits are exceeded. The permissible concentration of zinc in drinking water is established at 1-3mg/L; exceeding these levels induces both acute and chronic toxicity, causing neurological symptoms, abdominal pain, diarrhea, dizziness, and depression [1, 2]. Among heavy metals, cadmium poses an exceptionally grave risk due to its high toxicity and propensity to cause severe health conditions, including damage to the kidneys, liver, blood vessels, skeletal system, and cardiovascular network [3, 4]. Consequently, the development of efficient, cost-effective, and environmentally safe technologies for removing heavy metal ions from industrial wastewater is of paramount importance. A wide array of treatment techniques has been developed for contaminated water, including flotation, chemical and electrochemical precipitation, membrane separation, ion exchange, photo-extraction, and adsorption [5, 6]. Among these, adsorption is widely recognized as one of the most promising methodologies owing to its high efficiency, eco-friendly nature, easy availability, and low operational cost. In recent years, substantial research has focused on utilizing natural minerals as inexpensive, environmentally friendly adsorbents [7, 8]. Limestone is a naturally occurring, calcium-bearing sedimentary mineral featuring a highly developed porous structure, making it an attractive candidate for heavy metal remediation. It exhibits a distinct ability to neutralize acidic waters and capture various pollutants, particularly heavy metal ions. Its high operational performance stems from its inherent porosity, which provides ample surface reactivity toward dissolved ionic species. Numerous studies have demonstrated the

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utility of natural limestone as an adsorbent for purifying aqueous media from heavy metals such as (Pb^{2+}, Cd^{2+}, Zn^{2+}, Cu^{2+}) [9, 10]. In Georgia, limestone is predominantly mined for construction purposes in the village of Kvemo Bodbe (Kakheti region) under state mining licenses. The objective of this work is to evaluate local Georgian limestone as an effective adsorbent for removing Zn^{2+} and Cd^{2+} ions from aqueous solutions under systematically varied experimental conditions.

2. Material and methods

2.1. Materials and Reagents

Natural limestone from the village of Kvemo Bodbe (Kakheti, Georgia) was used as an adsorbent in the purification of aqueous solutions from zinc and cadmium ions. The limestone was used without any prior treatment; it was crushed, sieved through a mesh, and the 1-2mm fraction was collected.

Samples were finely powdered using vibration mill RETSCH RS200. Main and microelements were determined by means of X-Ray fluorescence spectrometry method using X-Ray spectrometer SPECTROSCOUT with Cu-Rh X-Ray tube.

Table 1 Elemental composition of the natural Georgian limestone sample**.**

Adsorbents	CaO	SiO_2	Fe_2O_3	Al_2O_3	TiO_2	MgO	Loss on ignition
Limestone	50.61	0.5	0.55	0.13	-	2.85	43.36

The remaining ~2% of the sample mass was below the detection/quantification limit of the XRF analysis and remains unidentified.

Stock cadmium solutions (1000 mg/L) were prepared from certified standard solutions (CPAchem Ltd.). Zinc stock solutions (1000 mg/L) were synthesized by dissolving high-purity granular zinc metal in nitric acid and diluting to 1 liter with deionized water. Working solutions of required concentrations were prepared by diluting stock solutions with deionized water.

2.2. Experimental part

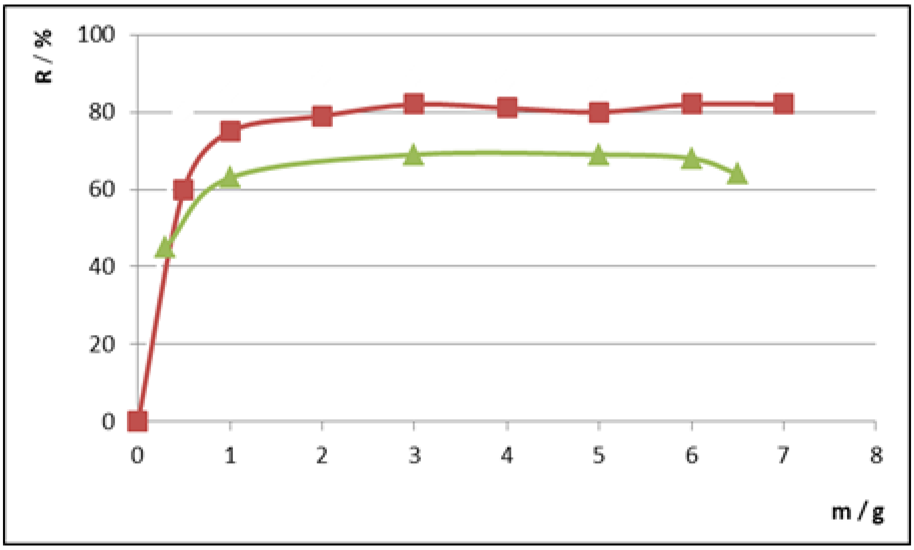

Figure 1 Impact of adsorbent dosage (m, g) on Zn^{2+} and Cd^{2+} adsorption degree (R/%). pH = 5.6, τ = 60 min, t = 20°C, C = 100 mg/L; Green line: Cd^{2+}, Red line: Zn^{2+}.

Batch adsorption experiments for Zn^{2+} and Cd^{2+} ions onto limestone were performed under static conditions at 20^{0}C. The influence of adsorbent dosage, contact time, initial solute concentration, and solution pH was thoroughly evaluated. Experiments were carried out in 250 mL vessels containing 100 mL of metal ion solutions. To determine the optimal sorbent dosage, limestone masses ranging from 0.1g to 6.0 g (0.1, 1.0, 3.0, 5.0, and 6.0 g) were added to 100mL solution volumes. Samples were agitated for 1 hour. As shown in Figure 1, equilibrium removal was reached at 3.0 - 5.0 g, leading to the selection of 3.0 g as the standard dosage for subsequent tests.

The kinetic effect of contact time was evaluated at interval durations of 15, 30, 60, 90, and 120, 150, 190 minutes. Reaction mixtures containing 3.0 g of limestone and 100 mL of solution were shaken, sampled at the specified time intervals, filtered, and analyzed. Adsorption equilibrium was reached within 60 minutes (fig. 2).

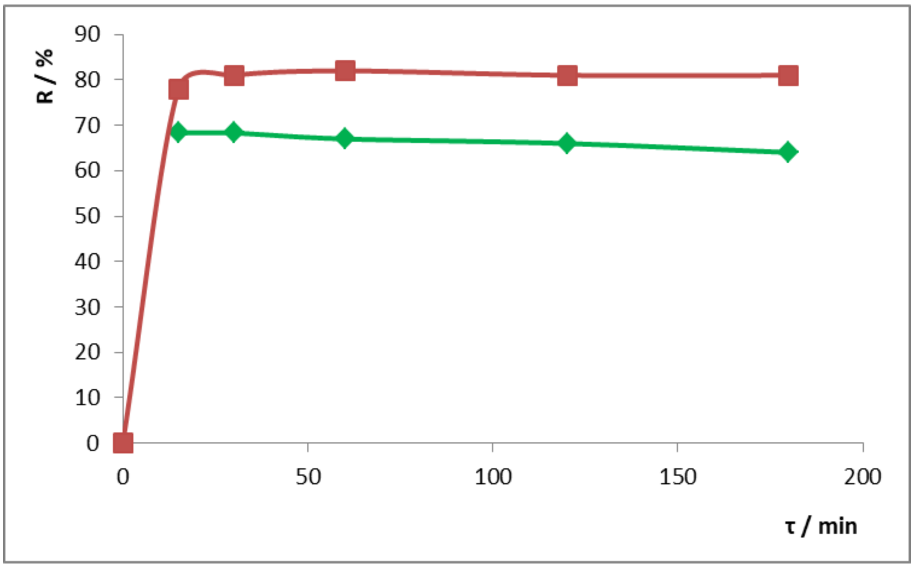

Figure 2 Impact of contact time on Zn^{2+} and Cd^{2+} adsorption degree (R/%). pH = 5.6, t = 20°C, C = 100 mg/L; Green line: Cd^{2+}, Red line: Zn^{2+}.

The effect of initial metal concentration was investigated over the range of 25 - 300 mg/L under optimized conditions 3.0 g sorbent mass, 60 min contact time). Rapid adsorption was observed, reaching saturation equilibrium in the range of 100 -200 mg/L. Consequently, an initial concentration of 100 mg/L was adopted for subsequent tests (fig. 3).

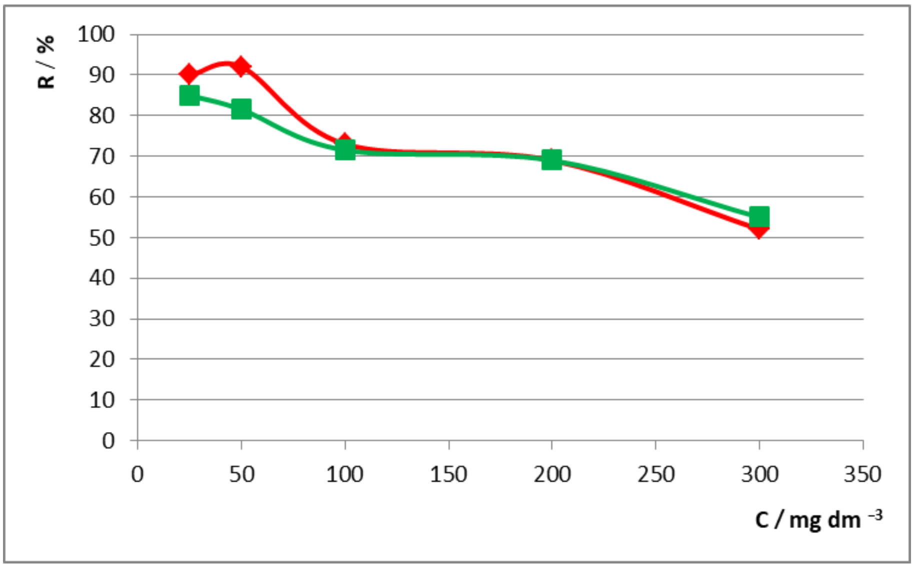

Figure 3 The effect of initial solution concentration on Zn^{2+} and Cd^{2+} adsorption degree (R/%). pH = 5.6, τ = 60 min, t = 20°C; Green line: Cd^{2+}, Red line: Zn^{2+}.

The influence of pH was evaluated across a range of 2.0 to 8.0 at 20⁰C, utilizing a 100 mg/L metal concentration and 3.0 g sorbent dose. Mixtures were agitated for 60 minutes, filtered, and analyzed for zinc and cadmium ion content.

Analyses were taken using atomic-absorption (AAS Perkin-Elmer-200) and chemical methods. Experiments were conducted three times and the mean values with standard deviations were used. The amount of adsorbed zinc and cadmium per unit mass of the adsorbent surface (adsorption capacity) A mg/g and the degree of adsorption R% were calculated using the formulas:

$$A = \frac{C_o - C}{m} \cdot V \text{ mg/g}$$

$$R = \frac{C_o - C}{C_o} \cdot 100\%$$

Where C_0 - initial solution concentration, mg/dm^3;

C- equilibrium (final) concentration;

m - sorbent weight, g;

V - volume of purified solution, dm^3.

3. Results and discussion

Studies on the adsorption of zinc and cadmium ions on limestone were conducted to establish optimal values for adsorbent dosage, contact time, test solution concentration, and pH value. As shown in Figure 1, an increase in the adsorbent dose leads to a sharp increase in the degree of adsorption, as increasing the quantity of sorbent provides a greater number of available active sites for the adsorption of metal ions. The adsorption percentage increases up to 80% for zinc ions and 63% for cadmium ions, respectively. Beyond this, the rate of adsorption growth decreases, reaching an equilibrium plateau and remaining nearly constant within the range of 3–6 g [11]. These values reflect the dosage-optimization step; the final optimized removal efficiencies (82% Zn, 69% Cd), obtained under the combined optimal conditions, are presented in the Conclusion.

The dependence of the degree of zinc and cadmium adsorption on contact time is presented in Figure 2. This relationship indicates that with an increase in contact time, a rapid increase in the adsorption of zinc and cadmium occurs, with the majority of metal ions being adsorbed within the first 15 minutes, reaching 80% for zinc and 68% for cadmium. Subsequently, the rate of adsorption declines and reaches equilibrium at 69% for cadmium and 82% for zinc, with corresponding adsorption capacities of 2.30 mg/g for cadmium and 28 mg/g for zinc. The initial rate of uptake of metal ions was high because all adsorption sites on the sorbent surface were initially vacant. As the contact time increased, zinc and cadmium ions occupied these binding sites, and after a certain period, nearly all active centers became occupied, which reduced the rate of uptake [12].

The effect of initial concentration on the adsorption capacity for zinc and cadmium ions was evaluated by varying the concentration of the test solution. The initial concentrations of metal ions were varied in the range of 25 mg/L to 300 mg/L. Increasing the initial concentration of ions in the solution leads to an increase in the amount of metal adsorbed per unit mass of sorbent until the binding centers become saturated and reach equilibrium. As shown in Figure 3 within the concentration interval of 100–200 mg/L, the adsorption remains constant, reaching 75% for zinc and 65% for cadmium [13]. These values correspond specifically to the concentration-optimization step; the final removal efficiencies under the overall optimal conditions (82% Zn, 69% Cd) are reported in the Conclusion.

The influence of the pH factor on the adsorption of zinc and cadmium was investigated. The solution pH is a critical factor influencing the adsorption of heavy metals, as it governs the surface charge of the adsorbent, the degree of ionization, and the chemical speciation of metal ions in the solution. It was demonstrated that at low pH values, adsorption is minimal because the adsorbent surface acquires a positive charge (H^+ protons), leading to electrostatic repulsion of metal cations and a decrease in adsorption efficiency. As the pH increases (from 2 to 6), the H^+ concentration decreases, and the surface of the adsorbent becomes more negatively charged with $(OH)^-$ groups, promoting electrostatic attraction of Zn^{2+} and Cd^{2+} ions and increasing the accumulation of metals on the adsorbent. The maximum adsorption is achieved at pH 5.6–6. A number of authors report that high-quality limestone increases the pH above 8 due to the carbonate load in the solution, which stimulates the precipitation of metal hydroxides or carbonates. Thus, precipitation plays an important role in the sorption of heavy metals [14].

However, such carbonate formation is considered unlikely under the conditions of this experiment (pH 5.6), since, according to literature data, the precipitation of metal carbonates requires a pH value above 8 [15].

4. Conclusion

This study was conducted to evaluate the efficacy of local Georgian natural limestone for removing heavy metal ions (Zn^{2+}, Cd^{2+}) from contaminated waters. It was found that adsorption strongly depends on pH, adsorbent dosage, contact time, and initial solution concentration. Optimal process conditions were selected as follows: adsorbent dosage of 3.0g, contact time of 60 min, solution concentration of 100 mg/L, and solution pH of 5.6. Under these conditions, the removal efficiency of zinc on limestone reached 82% and cadmium reached 69%, with corresponding adsorption capacities of 28 mg/g and 2.30 mg/g, respectively. It was demonstrated that limestone possesses significant potential as an effective adsorbent for removing zinc and cadmium ions from contaminated waters. The use of this readily available natural material represents an environmentally friendly and economically feasible approach to reducing heavy metal water pollution.

Compliance with ethical standards

Disclosure of conflict of interest

The authors declare that they have no conflict of interest.

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