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Article

Synthesis and application of aluminum-acetylacetonate-based material for the adsorption of fluoride from water

Síntesis y aplicación de material a base de acetilacetonato de aluminio para la adsorción de fluoruro del agua

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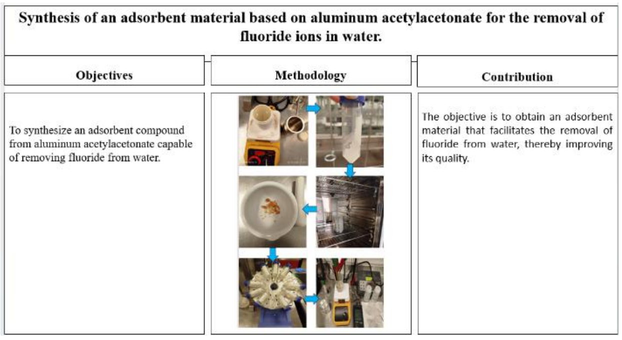
Abstract

Fluorine is a highly electronegative element, wich allows it to from toxic compound thet pose significant health hazards when inhaled or ingested in large quiantities. In thisd study, we report the synthesis of adsorbent materials derived from aluminium acetylacetonate, designed for the removal of fluoride from aqueos solutions. The synthesized materials were characterized using poder X-ray diffraction and FT-IR. Furthermore, adsorption isoterm models were applied to evaluate the efficiency and adsorption behavior of the material toward fluoride ions.



Synthesis of an adsorbent material based on aluminum acetylacetonate for the removal of fluoride ions in water.

Objectives	Methodology	Contribution
To synthesize an adsorbent compound from aluminum acetylacetonate capable of removing fluoride from water.		The objective is to obtain an adsorbent material that facilitates the removal of fluoride from water, thereby improving its quality.

Adsorbent; Aluminum Acetylacetonate; Fluoride Removal; Water Treatment; Isotherm Models

Resumen

El flúor es un elemento altamente electronegativo lo que permite formar compuestos tóxicos que representan un riesgo significativo para la salud cuando se inhalan o ingieren en grandes cantidades. En este estudio se presenta la síntesis de materiales adsorbentes derivados del acetilacetonato de aluminio, diseñados para la remoción de fluoruro en soluciones acuosas. Los materiales sintetizados fueron caracterizados mediante difracción de rayos X en polvo y FT-IR. Además, se aplicaron modelos de isotermas de adsorción para evaluar la eficiencia y el comportamiento del material frente a los iones fluoruro.

Síntesis de un material adsorbente basado en Acetilacetonato de Aluminio para la remoción de ion Fluoruro en agua.

Objetivos	Metodología	Contribución
Sintetizar un compuesto adsorbente a partir de acetilacetonato de aluminio , el cual pueda remover el fluoruro del agua.		Obtención de un material adsorbente que permite remover el fluoruro del agua y mejorar su calidad.

Adsorbente; Acetilacetonato De Aluminio; Eliminación De Fluoruro; Tratamiento De Agua; Modelos Isotermicos

Area: Strengthening the scientific community

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Introduction

Water quality remains a critical public health concern in Lagos de Moreno, Jalisco, where the hydrothermal origin of local water supplies leads to the presence of elevated fluoride concentrations. Excessive fluoride exposure has been associated with severe health issues, including dental and skeletal fluorosis, renal impairment, and central nervous system disorders. According to the Mexican Official Standards, the maximum permissible concentration of fluoride in drinking water is 1.5 mg/L; however, in some regions, reported levels exceed this limit by up to threefold.

To address this issue, several technologies have been explored for fluoride removal, such as ion exchange, filtration, precipitation, redox processes, electrochemical treatments, membrane technologies, and evaporation recovery. While these methods have demonstrated efficiency, their widespread implementation is often limited by high costs and continuous maintenance requirements [1].

This situation highlights the urgent need for the development of alternative, sustainable, and cost-effective materials capable of efficiently removing fluoride from aqueous systems. In this study, we report the synthesis and characterization of novel adsorbent material based on aluminum acetylacetonate.

The purpose of this work is to evaluate their structural properties and adsorption performance toward fluoride ions, aiming to provide a promising alternative for water treatment technologies in affected regions.

1. Materials and methods

1.1. Materials

Aluminum acetylacetonate, trans-1,2-diaminocyclohexane-N,N,N',N'-tetraacetic acid monohydrate [DCTA], glacial acetic acid, and sodium chloride were purchased from Aldrich and used as received without further purification.

Sodium fluoride [NaF], solvents, salts, and other reagents were of analytical grade and used as received without additional purification. All solutions were prepared using deionized water.

1.1.1. Synthesis of aluminum-acetylacetonate-based material

The synthesis was carried out via a solvothermal method [2]. A total of 4.15 g of aluminum acetylacetonate [Al[AcAc]$_3$] was dissolved in 60 mL of methanol [MeOH], followed by the dropwise addition of 0.512 g of sodium hydroxide [NaOH] previously dissolved in 5.00 mL of distilled water.

The resulting solution was transferred into a Teflon-lined stainless steel autoclave and subjected to thermal treatment at 150 °C for 6 hours in a convection oven.

The resulting gel was washed with methanol and centrifuged at 6000 rpm for 15 minutes to remove residual impurities. This washing and centrifugation step was repeated three times.

The purified product was then dried in an oven at 70 °C for 24 hours and subsequently ground to a fine powder using a mortar and pestle.

1.1.2. Preparation of TISAB II [Total Ionic Strength Adjustment Buffer]

TISAB was used as a buffering solution to adjust the total ionic strength according to the Mexican Official Standard NOM-201-SSA1-2002. It was prepared by placing 500 mL of deionized water in a 1.0 L beaker, followed by the addition of 57 mL of glacial acetic acid, 58 g of NaCl, and 4.0 g of DCTA.

The mixture was stirred until complete dissolution. The pH was then adjusted to between 5.0 and 5.5 by the slow addition of 6.0 N NaOH under continuous stirring. Finally, the volume was made up to 1.0 L in a volumetric flask.

1.2. Characterization

Infrared spectroscopic measurements were performed on a Frontier FT-IR/FIR spectrometer [PerkinElmer; DTGS detector; KBr beamsplitter for mid-IR and aluminum grid/polypropylene beamsplitter for far-IR], using attenuated total reflection [ATR] or KBr disc technique.

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The XRD patterns were recorded from 10° to 80° on a PANalytical X-ray diffractometer [Model Empyrean] with monochromatized Cu Ka radiation [l = 1.5406 Å]. Crystallite sizes [D_C] were calculated from the line broadening of the X-ray diffraction peaks, applying the Debey–Scherrer equation [1] [3],

$$D_C = \frac{k\alpha}{\beta \cos\theta} \quad [1]$$

where b is the breadth of the observed diffraction line at its half-intensity maximum [FWHM], k is the so-called shape factor, which usually takes a value of about 0.9, l is the wavelength of X-ray source used in XRD and q is the angle of reflection.

1.3. Adsorption Studies

Batch adsorption experiments were conducted to evaluate the equilibrium and kinetic behavior of fluoride adsorption onto aluminum-acetylacetonate-based material. A stock fluoride solution [1000 mg/L] was prepared by dissolving 2.21 g of NaF [previously dried at 105 °C for one hour and cooled in a desiccator] in deionized water.

The solution was diluted to one liter and stored in polyethylene bottles. Working solutions with concentrations ranging from 10 to 200 mg/L were prepared by dilution prior to use. All experiments were performed in triplicate. Adsorption tests were carried out in polypropylene Erlenmeyer flasks using a reaction volume of 50 mL.

Experimental parameters such as initial fluoride concentration and contact time were varied or held constant depending on the study objective. After the agitation period, samples were centrifuged at 6000 rpm, and the residual fluoride concentration was measured using an ion-selective electrode [Hanna Instruments HI98402], mixing the supernatant with TISAB II in a 1:1 volume ratio.

Samples and standards were placed in 100 mL polyethylene beakers and continuously stirred with a magnetic stirrer during analysis. The equilibrium fluoride adsorption capacity q_e [mg/g] was calculated using the following equation:

$$q_e = \left(\frac{C_0 - C_e}{m}\right) V \quad [2]$$

where C_0 and C_e are the initial and equilibrium fluoride concentrations [mg/L], m is the mass of the adsorbent [g] and V is the solution volume [L].

1.3.1 Adsorption Kinetics

To evaluate the fluoride adsorption rate onto MgO-1 and MgO-2 nanoparticles and to determine the minimum contact time required to reach equilibrium at neutral pH and 25 °C, batch kinetic experiments were performed using various contact times.

For each test, 50.0 mg of adsorbent was added to 30 mL of fluoride solution with a concentration of 10.0 mg/L. The suspensions were agitated for 0.5, 1, 2, 3, 4, 5, 6, 7 and 8 hours. The amount of fluoride adsorbed at a given time t, denoted q_t [mg/g], was calculated as follows:

$$q_t = \left(\frac{C_0 - C_t}{m}\right) V \quad [3]$$

where C_t [mg/L] represents the fluoride concentration at time t.

Capítulo I - 1.3.2. Adsorption Isotherms

Equilibrium adsorption studies were conducted to obtain the adsorption isotherms. A mass of 50 mg of adsorbent was mixed with 30 mL of fluoride solutions with varying concentrations [10, 25, 50, 75, 100, 125 and 150 mg/L]. These suspensions were agitated at a pH of approximately 6.5–7.0 and at 25 °C for 1 hour, as defined by the adsorption kinetics study.

Afterward, the samples were centrifuged, and the fluoride concentration was measured using the ion-selective electrode method.

Box 1

Figure 1

Product obtained after solvothermal synthesis and crystals obtained after sample drying

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2. Results and discussion

2.1. Material characterization

Infrared spectroscopy

The products were characterized by infrared spectroscopy in the 4000-400 cm^{-1} range. Aluminium compound [see Fig. 2] shown a wide and intense band at 3313 cm^{-1} related to asymmetric and symmetric vibrations n[OH] [4]. The presence of this vibration suggest the presence of water molecules in a hydrogen bonds arrangements in the Al-AcAc system. The IR spectra of this compound confirmed the bidentate binding mode of the AcAc ligand to the Al atom.

The bands observed at 1558 and 1344 cm^{-1}, assigned to the n[CO] and n[C-C] of AcAc vibrations indicates chelation of acetylacetonate to the Al center due to the shifting to lower frequencies of these two bonds: in free acetylacetone two C–O bonds give a band pattern at 1600 and 1500 cm^{-1} whereas two C–C bonds give a band pattern at 1450 and 1260 cm^{-1}.

Acetylacetone is known to show tautomerism, exhibiting the keto n[C=O] and enol n[C=C-OH] form with peaks at 1709 and a broad band of 1640 to 1530 cm^{-1}, respectively [4-5].

The bands at 1427-1445 and 1373-1414 cm^{-1} are attributed to n[CC] vibrations present in the AcAc ligand, whereas the bands at 1036 and 846 cm^{-1} are due to bending d[CCH], combined with n[CC] stretch vibrations.

The n[Al-OAcAc] vibrations appear at 452 cm^{-1} with an average band intensity, whereas 608 cm^{-1} presents bands of similar intensity due to characteristic bending vibrations [d[Al-O-H]$_{br}$] of a hydroxo bridge between the metal nuclei of the Al-AcAc compound, as in the 886-733 cm^{-1} region there are no vibrations related to the AcAc ligand [6].

The Al-O[H]br group exhibits bending and stretching vibrations outside the n[Al-O] plane below 600 cm^{-1}, which combines with Al-OH vibrations and superposition, so these bands are not informative [7].

Box 2

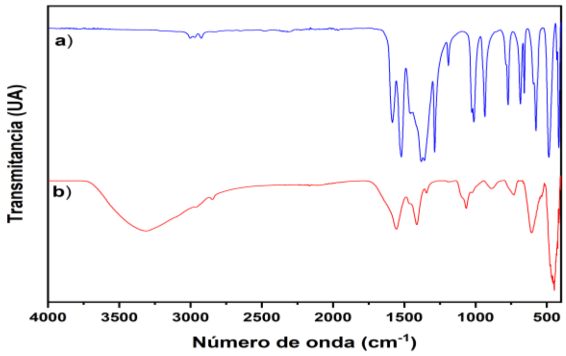

Figure 2

FTIR spectra of a) the acetylacetone ligand and b) aluminum-acetylacetonate-based material.

X-ray diffraction analysis

The X-ray diffraction pattern [XRD] of aluminium material synthesized under solvothermal method are presented in Fig. 3. In the diffractogram, the crystalline structure present in the sample was identified as boehmite, indexed to [020], [120], [031], [051], [080] and [251] planes of the orthorhombic dipyramidal system of AlO[OH] [JCPDS 21-1307] [8] with the peaks shifted with respect to this pure crystalline phase, which may be due to a distortion generated by the presence of non-stoichiometric water and/or AcAc ligand in the structure [pseudoboehmite] [9]. XRD pattern show a broad peaks typically indicate a material with a small crystallite size, structural disorder, lattice defects, or strain.

Crystallite sizes of the material prepared from aluminum acetylacetonate, calculated using Scherrer's formula, presented reduced values with average crystallite size of the product fell between 1.85 and 4.61 nm.

Box 3

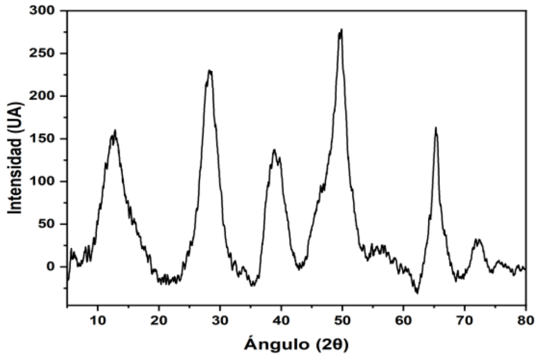

Figure 3

XRD patterns of aluminum-acetylacetonate-based material

Adsorption kinetic studies

As shown in Figure 4, batch experiments indicated that 50 mg of adsorbent material has a good adsorption capacity in 50 mL of fluoride solution at 10 ppm, reaching its equilibrium point in a time between 1 to 2 hours, removing 94% to 95% of fluoride present in the medium, which based on studies described by Pricila et al., [2017] [10], demonstrated that adsorption is favored when stirring ensures a uniform distribution of fluoride ions at the liquid-solid interface, since the liquid film trapped at the adsorbent interface is reduced.

Box 4

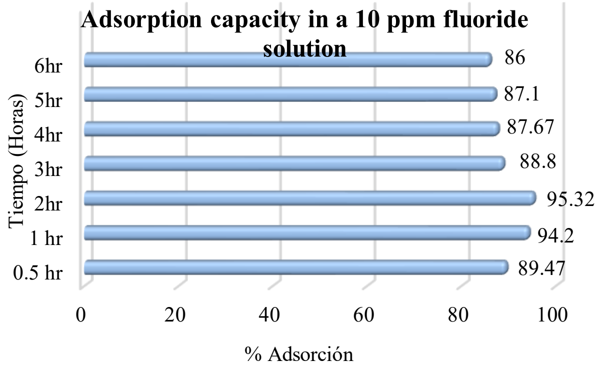

Figure 4

Graphical representation of the adsorption capacity of the compound expressed as a percentage.

Adsorption isotherm studies

To optimize the fluoride removal process, it is essential to understand and describe the distribution of fluoride ions between the two phases [liquid–solid interface].

Adsorption equilibrium occurs when the amount of solute adsorbed onto the surface of the adsorbent material equals the amount desorbed. At this point, the concentration of the equilibrium solution remains constant. By plotting the concentration of the solid phase [q_e] against that of the liquid phase [C_e], the adsorption equilibrium isotherm can be represented.

In this study, the equilibrium data obtained from initial fluoride concentrations ranging from 10 to 150 mg/L at neutral pH and 25 °C [Table 1] were fitted to the two-parameter Langmuir adsorption isotherm model, whose linearized form is shown in Equation [4].

Box 5

Table 1

Results of adsorption isotherm experiments

Co	Ce	qe
10	2.265	7.735
25	7.3	17.7
50	34.95	15.05
75	64.5	10.5
100	90	10
125	113.5	11.5
150	142	8

Hanese-Woolf linearization of the Langmuir model [Type 1]:

$$\frac{C_e}{q_e} = \left(\frac{1}{Q^0_{max}}\right) C_e + \frac{1}{Q^0_{max}K_L} \qquad [4]$$

where Q^0_{max} [mg/g] represents the maximum adsorption capacity of a saturated monolayer on the adsorbent, C_e [mg/L] is the equilibrium fluoride concentration, q_e [mg/g] is the amount of fluoride adsorbed at equilibrium, and K_L [L/g] is a constant related to the affinity between the adsorbent and the adsorbate.

The Langmuir model assumes that adsorption results in the formation of a monolayer with no interactions between adsorbate molecules and that all adsorption sites on the solid surface are equivalent.

This model further assumes that no strong bonds develop on the adsorbent surface, and that all specific sites possess a constant binding energy [10-11]. The Langmuir coefficients, Q^0_{max} and K_L, can be determined from the slope and intercept of a linear plot of C_e/q_e versus C_e [Figure 5]. The maximum monolayer adsorption capacity obtained from the linear model for the synthesized adsorbent was 8.2 mg/g, with a K_L value of –0.13 L/g.

Box 6

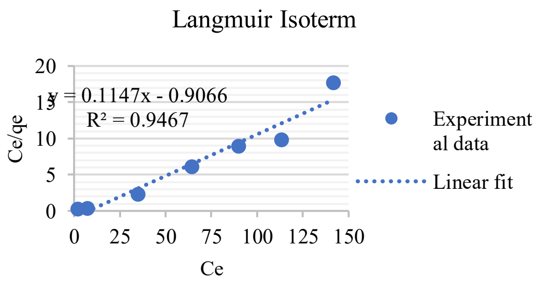

Figure 5

Linear fit of the Langmuir model on aluminum-acetylacetonate-based material.

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Conclusions

The findings of this research confirm that aluminum-acetylacetonate-based materials synthesized through a solvothermal process exhibit remarkable efficiency in the adsorption of fluoride from aqueous media, achieving removal rates above 90% within relatively short contact times. Comprehensive characterization by FTIR and XRD established the successful structural integration of the acetylacetonate ligand into the aluminum framework, while adsorption kinetics and isotherm modeling indicated a favorable interaction between the adsorbent and fluoride ions, with a maximum monolayer capacity of 8.2 mg/g.

Overall, these results underscore the potential of this material as a sustainable and cost-effective alternative for water purification, advancing the development of functional adsorbents designed to address pressing environmental and public health challenges.

Declarations

Conflict of interest

The authors declare no interest conflict. They have no known competing financial interests or personal relationships that could have appeared to influence in this chapter.

Author contribution

Ramirez-Contreras, Yeairim Elena: Contributed to experimental development of the project, writing of the document and methodology research.

Ramírez-de-Alba, Daniel: Contributed to the project idea and experimental development of the project.

Maldonado-Ríos, Juan José: Contributed to the project idea and experimental development of the project.

Pérez-Tavares, José Antonio: Contributed in the methodology research, project idea and follow-up of the proyect.

Availability of data and materials

The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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