

Research paper

# Elimination of sulfates from wastewaters by natural aluminosilicate modified with uric acid

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## Abstract

Natural aluminosilicate activated by a heat/acid treatment, followed by modification with uric acid was used to remove sulfates for treatment of wastewater effluent. Natural aluminosilicates were studied in every stage of the modification (namely activation, modification with uric acid, and after sulfates absorption) by scanning electron microscopy (SEM), spectroscopy X-ray diffraction (XRD), energy dispersive spectroscopy (EDS), surface area (BET), X-ray photoelectron spectroscopy (XPS), thermogravimetric analysis (TGA), and Z potential. More than 60% of the initial concentration of sulfates (500 mg/l) was removed with the natural aluminosilicate modified with uric acid. Absorption isotherms rendered a mechanism with contributions from both Langmuir and Freundlich mechanisms. This study opens the path for the use of natural and abundant local material to remove sulfates using a modifier already present in wastewater effluents as contaminant.

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**Keywords:** Natural aluminosilicate; Uric acid; Sulfates; Isotherms; Adsorption

## 1. Introduction

The main limitation for reusing reclaimed water is its low quality in terms of nitrogen-based nutrients (NH<sub>3</sub>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>), phosphates (PO<sub>4</sub><sup>-3</sup>) and sulfates (SO<sub>4</sub><sup>-2</sup>), which precludes its use in recharging local groundwater aquifer [1]. According to World Health Organization [2], a global network of water monitoring stations, typical sulfate levels in fresh water are in the vicinity of 20 mg/l and range from 0 to 630 mg/l in rivers (the highest values are found in Belgium and Mexico), from 2 to 250 mg/l in lakes (the highest value is found in Mexico) and from 0 to 230 mg/l in groundwater (the highest values are found in Chile and Morocco). In 1970, the US Public Health Service measured sulfate levels in the drinking-water sources of nine

geographic areas. Sulfate was found to be present in 645 of 658 groundwater supplies and in all of the 106 surface water supplies sampled. Sulfate levels ranged from <1 to 770 mg/l, with a median of 4.6 mg/l. Only 3% of the water supplies sampled had sulfate levels in excess of 250 mg/l.

With an average annual precipitation of 273 mm and a rapid population growth, the region of Tijuana–San Diego, in the Mexico–USA border, is currently experiencing a scarcity of water resources [3]. In Tijuana, local wastewater treatment plant system (WWTPS) generates 79.65 Mm<sup>3</sup>/year (2590 lps) of reclaimed water, from which 0.11 Mm<sup>3</sup>/year (85.03 lps) is used for irrigation and the rest is discharged to the Pacific Ocean and thus not utilized [4]. Therefore strategies aimed to increase the quality of reclaimed water are a must. Adsorption and ion exchange stand as the most effective technologies and economic alternatives for removing contaminants from water. Previous studies for pollutants removal of reclaimed water with aluminosilicates, zeolites [5] and activated calcite [6], showed high efficiency for nitrogen and phosphates removal; however that is not the case for sulfates.

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Technologies involving reverse osmosis [7], electrochemistry [8], biotechnology, adsorption and ion exchange [9,10] have been applied to control pollution caused by sulfates. Among the preferred absorbers, natural aluminosilicates are the most widely used due to its low cost, ample distribution and preference for specific contaminants [11–13]. During the last twenty years numerous researchers have been working on the creation of adsorption materials based on modified aluminosilicates. Common modifications include the use of acids, bases, cationic surfactants and polioxocations [14,15]. Intercalation routes, organic molecules located in the interlayer space, post-synthesis grafting of organosilane onto aluminosilicate surfaces [16,17] and one step preparation of aluminosilicates by sol–gel process [18,19] have been also used. Natural aluminosilicate (bentonite) is a widespread type of clay that can be easily modified with quaternary ammonium salts for applications in many fields, including sulfate remove from water [20]. Recent reports showed the capacity of bentonite to adsorb uric acid and creatinine in rats thus inhibiting their adsorption in the intestine [21].

In some regions (e.g. Tijuana–Otay, Mexico–USA border) soil possess high content of natural aluminosilicate (bentonite) and other aluminosilicates [22]. The aim of this study was to use a natural aluminosilicate-type material to remove uric acid and sulfate solutions that emulate treated wastewater effluent through two steps: (1) Uric acid absorption into natural aluminosilicate as modifier of the surface properties (e.g. charge), (2) adsorption of  $\text{SO}_4^{2-}$  in the modified aluminosilicate with uric acid. The method here described introduces preliminary results regarding a low cost alternative for the reuse of wastewater, which takes advantage that both the modifier and the sulfates are already present in domestic and industrial wastewater effluents.

## 2. Materials and methods

### 2.1. Materials

#### 2.1.1. Natural aluminosilicate

The natural aluminosilicate was collected by gravimetric sedimentation and passed through No. 635 mesh ASTM Standard Testing Sieve vibrator E-11, model 150 [23]. Then, the sample was washed with distilled water and dried at 100 °C. The sample was labeled NA, which stands for natural aluminosilicate.

#### 2.2. Activation of natural aluminosilicate (NA)

Twenty-five grams of natural aluminosilicate (NA) was dispersed into 250 ml of hydrochloric acid 2N, at 90 °C for 2 hours, afterwards it was washed with 3 l water and dried at 250 °C for 4 h [24]. The sample was labeled AA (activated aluminosilicate). The cation exchange capacity of NA was determined by the copper ethylene diamine complex ( $\text{Cu}(\text{EDA})_2^{+2}$ ) method [25].

#### 2.3. Adsorption kinetic of uric acid into activated aluminosilicate (AA)

A sample of 600 mg of AA was added to 50 ml of uric acid solution ( $113 \text{ mg l}^{-1}$ ) under magnetic stirring in a 250 ml Pyrex glass flask at 25 °C; hydrochloric acid solution (2N) was used to

adjust the pH to 2.0 (Orion type PHS-25C). Uric acid absorption on the filtrates was measured after 5, 15, 30, 60, 90 and 120 min on different samples. The resultant sediment was filtered and centrifuged. The remained uric acid in solution was determined by an enzymatic–colorimetric method (diagnostic ELITECH) at  $\lambda = 550 \text{ nm}$  with a spectrophotometer DR.5000 HACH, thus the uric acid uptake was calculated by the difference between the initial and final concentrations of uric acid in solution after absorption on the aluminosilicate.

#### 2.4. Effect of pH on the uric acid absorption on AA

Two hundred fifty milliliters of a uric acid solution with an initial concentration of  $400 \text{ mg l}^{-1}$  was added to different masses of AA (from 0.1 to 1.0 g) in stopped Pyrex glass flasks under magnetic stirring at 25 °C. Hydrochloric acid solution (2N) was used to adjust the pH to 2 followed by agitation by 30 min at 89 rpm. The absorption of uric acid at different pH was calculated by measuring the concentration of uric acid remained in the solution after the filtration of the aluminosilicate (enzymatic–colorimetric method using diagnostic ELITECH at  $\lambda = 550 \text{ nm}$  with a spectrophotometer DR.5000 HACH). The same approach was used with pH 7 and 10, adjusting the pH with a sodium hydroxide solution 2N.

#### 2.5. Preparation of modified aluminosilicate with uric acid (AA-U)

Five grams of AA was dispersed into 250 ml of uric acid solution ( $600 \text{ mg l}^{-1}$ ) and heated to 80 °C for 1 h. Then the aluminosilicate, now modified with uric acid (AA-U), was washed with 3 l of deionized water followed by addition of 200 ml of hydrochloric acid (7N). The modified aluminosilicate was finally centrifuged and dried at 60 °C for 24 hours [21,24]. The sample was labeled AA-U.

#### 2.6. Adsorption kinetic of sulfates on AA-U

A sample of 1 g of AA-U was added to 200 ml of sodium sulfate solution ( $500 \text{ mg l}^{-1}$ ) under magnetic stirring in a 250 ml Pyrex glass flask at 25 °C; sodium hydroxide (2N) was used to adjust the pH to 7.0 (Orion type PHS-25C). Sulfates absorption on the filtrates was measured after 20, 40, 60, 80, 100, and 120 min on different samples. The resultant sediment was filtered and centrifuged. The amount of sulfates removed by the AA-U was calculated measuring the concentration of sulfate ion remained in the supernatant after centrifugation and concentration of sulfate ion initial.

#### 2.7. Absorption of sulfates on AA-U

Five grams of AA-U was used as absorbent material in 200 ml of sodium sulfate with concentration ranging from 200 to  $500 \text{ mg ml}^{-1}$  at pH 7. The samples were stirred in a rotatory equipment Jar Tester Unit programmable Phipps & Bird series PB-900 at 89 rpm and  $25 \text{ °C} \pm 2 \text{ °C}$ , for 120 minutes. The samples were then filtered and centrifuged (VWR clinical 50 centrifuge). The amount of sulfates removed by the AA-U was calculated measuring the concentration of sulfate ion remained in the supernatant after centrifugation. The method employed

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