



# Aluminum (hydr)oxide coated pumice for fluoride removal from drinking water: Synthesis, equilibrium, kinetics and mechanism

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## HIGHLIGHTS

- Removal of excess fluoride in drinking water is important to protect public health.
- The search for suitable fluoride adsorbent continues to be of interest.
- Coating of pumice surfaces with aluminum for fluoride removal was found promising.
- Approach of study was based on the HSAB concept and on use of indigenous materials.
- Aluminum oxide coated pumice could be useful in countries where pumice is indigenous.

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## ABSTRACT

Intake of excess fluoride (beyond 1.5 mg/L, WHO guideline) for long periods can result in the incidence of fluorosis. Adsorption is widely considered the most appropriate technology for water defluoridation, if a suitable adsorbent is available. Several studied adsorbents have shown certain degrees of adsorption capacity, however, applicability of most is limited either due to lack of socio-cultural acceptance, high cost or effectiveness only in extreme pH conditions. The search for alternative adsorbents thus remains of interest. Modification of pumice particle surfaces by aluminum oxide coating was found effective in creating hard surface sites for fluoride adsorption, in accordance with the hard and soft acids and bases (HSAB) concept. Aluminum oxide coated pumice (AOCP) reduced fluoride concentration in model water from  $5.0 \pm 0.2$  mg/L to 1.5 mg/L in approximately 1 h, using an adsorbent dose of 10 mg/L. Contrary to expectations, thermal treatment of AOCP aimed at further improving its performance, instead reduced the fluoride removal efficiency. The equilibrium adsorption of fluoride by AOCP conformed reasonably to five isotherm models in the order: Generalized model > Langmuir type 2 > BET > Temkin > Dubinin–Radushkevich; with a maximum capacity of 7.87 mg/g. AOCP exhibited good fluoride adsorption within the pH range, 6–9, which makes it possible to avoid pH adjustment with the associated cost and operational difficulties, especially if it is to be used in remote areas of developing countries. Based on results from kinetic adsorption experiments, it was observed that at a neutral pH of  $7.0 \pm 0.1$  which is a more suitable condition for groundwater treatment, fluoride adsorption by AOCP was quite comparable or perhaps fairly faster in the initial period of contact than that of activated alumina (AA), the commonly used adsorbent for water defluoridation. AOCP is thus promising and could also possibly be a useful fluoride adsorbent.

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## 1. Introduction

Optimum concentration of fluoride (about 1 mg/L) in drinking water is good for dental health and good bone development. Intake of excess fluoride (beyond 1.5 mg/L, WHO guideline) for long peri-

ods can, however, result in the incidence of fluorosis. Other adverse health effects include loss of mobility, changes in DNA structure, lowering of IQ of children and interference with kidney functioning [1,2]. Drinking water defluoridation is thus important to safe guard public health. The commonly used methods for fluoride removal include; coagulation with alum followed by floc separation, contact precipitation, adsorption, ion-exchange and reverse osmosis [3]. Among the available methods, adsorption process is widely considered the most appropriate, particularly for small community

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water source defluoridation. This is due to its flexibility and simplicity of design, relative ease of operation and cost-effectiveness, if a suitable adsorbent is available [4,5]. Several materials have been tested as suitable or promising adsorbents for water defluoridation including; manganese-oxide coated alumina, bone charcoal, fired clay chips, ceramic adsorbent, laterite, unmodified pumice, bauxite, zeolites, fluor spar, iron-oxide coated sand, calcite, activated quartz and activated carbon [1,4–16]. Some of these adsorbents have shown certain degrees of fluoride adsorption capacity, however, the applicability of most is limited either due to lack of socio-cultural acceptance, high cost or being effective only in extreme pH conditions. For example activated carbon is found to be effective at pH less than 3.0. This may require pH adjustment and consequently additional capital, operation and maintenance cost, and could limit feasibility of such a fluoride removal technology in remote rural areas of developing countries where more often than not the populace depend upon groundwater point sources normally with a pH range of 6.0–8.5 [7,17]. The search for alternative and suitable fluoride adsorbents therefore still remains of interest.

The aim of the present work was to modify the surfaces of pumice particles by aluminum oxide coating in order to create hard surface sites for fluoride adsorption.

The approach for the search for a fluoride adsorbent in this study was based on an application of the hard and soft acids and bases (HSAB) concept, and also on exploring the suitability of using locally available materials. Due to its characteristics,  $\text{Al}^{3+}$  is classified as a hard acid while  $\text{F}^-$  is categorized a hard base.  $\text{Al}^{3+}$  therefore has good affinity towards  $\text{F}^-$  in accordance with the HSAB concept [18,19]. The suitability of pumice as base material for the surface modification process was studied, since use of pumice could contribute to cost reductions and sustainable solutions for water defluoridation, especially in developing countries where it is indigenous. Pumice is a volcanic material with a rough surface and porous structure which may provide large number of attachment sites for aluminum oxide coating, and therefore expected to result in good fluoride removal efficiency.

The fluoride adsorption potential of the aluminum oxide coated pumice (AOCP) was investigated. The effect of thermal treatment of AOCP on its fluoride removal efficiency was also investigated. The effects of adsorbent dose and pH on fluoride removal by AOCP were examined. The physico-chemical characteristics of AOCP were determined. Additionally initial results from this study, i.e., the fluoride adsorption kinetics of AOCP, were compared with that of activated alumina (AA) which is commonly used for water defluoridation [20,21].

## 2. Materials and methods

### 2.1. Synthesis of Al-oxide coated pumice (AOCP)

#### 2.1.1. Coating of aluminum oxides onto pumice

Pumice samples used in this work as base material for the aluminum oxide coating were obtained from Aqua-TECHNIEK bv, The Netherlands. The samples were sieved to a particle size range of 0.8–1.12 mm, thoroughly washed with demineralized water and air-dried before being used in the coating process. To coat the pumice, a sufficient amount of 0.5 M  $\text{Al}_2(\text{SO}_4)_3$  was added to completely soak about 150 g of the dried pumice in a beaker. The mixture was stirred mechanically for 1.5 h at 150 rpm and pumice was after that drained and air dried at room temperature. The dried pumice was subsequently soaked in 3 M  $\text{NH}_4\text{OH}$  to neutralize it and complete the coating process. The coated sample was sieved again and washed several times with demineralized water buffered at pH  $7.0 \pm 0.1$ , in order to remove any loosely bound aluminum

oxides. AOCP was finally dried and stored for fluoride adsorption studies.

#### 2.1.2. Thermal treatment of AOCP

Samples of AOCP were calcined at temperatures between 200 °C and 1000 °C, in a muffle furnace for 2 h. The thermal treatment was aimed at further enhancing the fluoride removal capacity [22].

### 2.2. Characterization techniques

The aluminum contents of both uncoated pumice and AOCP samples were extracted by acid digestion at a temperature of 200 °C using reagent grade concentrated  $\text{HNO}_3$  acid, and was measured by colorimetric methods using a UV-VIS Recording spectrophotometer (SHIMADZU, Japan).

The specific surface area of both uncoated pumice and AOCP were determined by  $\text{N}_2$  gas adsorption-desorption method at 77 K with TriStar 3000 gas adsorption analyzer (Micromeritics, USA), and the Brunauer-Emmett-Teller (BET) method was used in the calculation. The Barrett-Joyner-Halenda (BJH) pore size model was used for determining pore size distribution. The empirical *t*-plot methodology was used to discriminate between contributions from microspores and remaining porosity (i.e. mesoporosity, macroporosity and external surface area contributions). Prior to the  $\text{N}_2$  adsorption measurements, the samples were pre-treated by degassing in a vacuum at 1500 °C [23].

Chemical compositions were obtained from X-ray Fluorescence (XRF) analysis provided by Axiosmax-Advance (from PANalytical Spectris, UK). Spot elemental analysis of AOCP was carried out using a scanning electron microscope (SEM NovaNano, FEI company, Netherlands) coupled with an energy dispersive X-ray spectroscopy (EDX) (EDAX, AMETEX, USA). Point of zero charge (pHpzc) of AOCP was determined by a mass titration method [24].

### 2.3. Fluoride adsorption experiments

A 1000 mg/L stock fluoride solution was prepared by dissolving reagent grade NaF in demineralized water. Model fluoride water used for adsorption experiments was prepared by diluting the stock solution with Delft (The Netherlands) tap water with the following composition (mg/L); Ca = 49.7, Mg = 9.0,  $\text{SO}_4^{2-}$  = 78.8, Cl = 72.2, Na = 35, K = 5.5 to obtain a fluoride concentration of  $5 \pm 0.2$  mg/L, similar to that found in groundwater in Northern region of Ghana. The bicarbonate concentration was adjusted to 260 mg/L by spiking with sodium bicarbonate in order to simulate similar concentrations in groundwater in the Northern region of Ghana.

Batch adsorption experiments were conducted using the completely mixed bath reactor method for studying the fluoride removal behavior of both AOCP and activated alumina (AA). The adsorption experiments were conducted at a neutral pH  $7.0 \pm 0.1$  and room temperature (20 °C), using 0.5 L of model water in 500 ml P.E. bottles. HCl and/or NaOH were used for pH adjustment after which the P.E. bottles were tightly closed to avoid exchange of  $\text{CO}_2$  in order to stabilize pH during the experiments. A control adsorption experiment using uncoated pumice was conducted. In addition a blank fluoride adsorption experiment, without adsorbent, was carried out to evaluate experimental uncertainties. All adsorption experiments were carried out in duplicate in order to check reproducibility. The experiments were carried out on a shaker (Innova 2100 Platform shaker, New Brunswick Scientific, USA), at a shaking speed of 100 rpm. Aqueous samples taken at pre-determined times during the adsorption experiments were immediately filtered through a microfilter paper (0.45  $\mu\text{m}$ ) to separate adsorbent from fluoride model water and prevent any further reactions.

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