

# Adsorption of Fluoride on Limestone-Derived Apatite: Equilibrium and Kinetics

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## ABSTRACT

Fluoride in drinking water above permissible levels is responsible for human dental and skeletal fluorosis. Adsorptive based defluoridation is the most popular technique with several end-user applications. Consequently, this paper describes the fluoride removal potential of a novel sorbent, limestone-derived apatite from drinking water. The adsorbent was prepared by calcining limestone followed by reacting with orthophosphoric acid. Batch sorption studies were performed as a function of contact time, pH, initial fluoride concentration, temperature and adsorbent dose. Sorption of fluoride was found to be pH dependent with a maximum occurring in the pH range of 5-9. The authors also observed that the material had a buffering effect on the same pH range. Meanwhile, the adsorption capacity was found to increase with temperature, depicting the endothermic nature of the adsorption process and decreases with adsorbent mass. The equilibrium data was well described by the conventional Langmuir isotherm, from which isotherm the maximum adsorption capacity was determined as 22.2 mg/g. From the kinetic perspective, the fluoride adsorptive reaction followed the pseudo-second order mechanism.

Keywords: Adsorption, Apatite, Endothermic, Fluoride, Limestone

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## NOMENCLATURE

$C_0$  initial concentration of adsorbate in solution (mg/L)

$C_e$  equilibrium concentration of the adsorbate in the solution (mg/l)

$C_t$  concentration of the adsorbate in the solution at any time  $t$  (mg/L)

$Q_e$  amount of adsorbate removed from aqueous solution at equilibrium (mg/g)

$Q_t$  amount of adsorbate sorbed on the adsorbent surface at any time  $t$  (mg/g)

$m$  mass of adsorbent (g)

ppm parts per million

$R^2$  linear regression coefficient

$v$  volume (mL)

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## 1. INTRODUCTION

Fluoride is one of the very few chemicals that have been shown to cause significant health problems in people through drinking-water. Fluoride has beneficial effects on teeth at low concentrations in drinking-water (0.5 to 1.0 mg/l), but excessive exposure to fluoride in drinking-water (above 1.5 mg/l), or in combination with exposure to fluoride from other sources, can give rise to a number of adverse effects. These range from mild dental fluorosis to crippling skeletal fluorosis as the level and period of exposure increases (Fawell, Bailey, Chilton, Dahi, Fewtrell, & Magara, 2006; Hichour, Persin, Sandeaux, & Gavach, 2000; Mehmet et al., 2007). There are four probable mechanisms of human exposure to the fluoride ions. This can occur through air, dental products, food and beverages other than water and drinking water (Daia, Rena, & Maa, 2004; Ponikvar, Stibilj, & Emva, 2007; Cao, Zhao, Li, Deng, & Yi, 2006). Drinking-water is typically the largest single contributor to daily fluoride intake (Maliyekkal, Shukla, Philip, & Nambi, 2008). Fluoride poisoning can be prevented or minimized by using alternate water sources, improving the nutritional status of population at risk or removing excess fluoride (defluoridation). Defluoridation of drinking water is the only practicable option to overcome the problem of excessive fluoride in drinking water, where alternate source is not available (Maheshwari, 2006).

Fluoride removal from drinking water can be achieved by chemical precipitation (Pinon-Miramontes, Bautista-Margulis, & Perez-Hernandez, 2003), adsorption (Lounici et al., 1997; Ghorai & Pant, 2005), membrane process (Arora, Maheshwari, Jain, & Gupta, 2004; Wisniewski, Rozanska, & Winnicki, 2005) and ion exchange (Meenakshi & Viswanathan, 2007). Among these processes, membrane and ion exchange processes are not very common due to their high installation and maintenance cost. Chemical precipitation by addition of alum and

lime mixture into fluoride-contaminated water (Nalgonda technique) is a common method for defluoridation after its inception in India. However, the associated problems like generation of acid/alkali water, residual aluminium and soluble aluminium fluoride complexes, generation of sludge, and relatively higher residual fluoride concentrations are of major concern (Onyango, Kojima, Aoyi, Bernado, & Matsuda, 2004; Maliyekkal, Sharma, & Philip, 2006). Adsorption onto solid surface is a simple, versatile and appropriate process for treating drinking water system, especially for small communities. Adsorption technique is considered as economical and can remove ions over a wide pH range and to a lower residual concentration than precipitation (Maliyekkal et al., 2008).

Several materials have been identified as potential adsorbents for the removal of excess fluoride in water. However, the fact that most of the affected are the rural population with little or no income poses the need for the continued search of low cost and efficient adsorbents. The present study therefore explores the potentiality of using apatite impregnated locally sourced limestone as a drinking water defluoridation media.

## 2. MATERIALS AND METHODS

### 2.1 Modification of Limestone

The limestone used in this work was obtained from Continental Cement Pty in South Africa. The modification process involves calcination at a temperature of 800°C for 24 h. Table 1 shows the chemical composition of the calcined phase of the limestone. The calcined material was air cooled to room temperature and reacted with 50% orthophosphoric acid. The acid was added drop wise till excess to ensure that the reaction goes to completion. During the reaction, the mixture was under constant agitation and temperature kept constant at about 50°C. The resultant precipitate was left in the mother

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