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## Review article Clay and clay minerals for fluoride removal from water: A state-of-the-art review

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## ABSTRACT

The presence of fluoride in water has become a matter of great concern around the world due to its chronic human carcinogenic behavior. Developing easily accessible and environmentally sustainable removal strategies is therefore a challenge for the scientists. Among the different treatment technologies, adsorption process for fluoride removal is considered cost-effective, flexible, and easy to design and operate. This review discusses the recent trends in scientific research and development on the exploitation of clay and clay minerals for fluoride removal from water, focusing on the effect of various factors on the adsorption, mechanism, isotherms and kinetics of the adsorption process.

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## 1. Introduction

Fluoride is one of the hazardous chemicals known to cause largescale health problems by exposure through drinking water (Ayoob and Gupta, 2006; Maiti et al., 2011). It has both beneficial and detrimental effects on human health (Fan et al., 2003; Jagtap et al., 2012; Meenakshi and Maheshwari, 2006; Mohapatra et al., 2009). For example, a small amount of fluoride in water is generally considered to have a beneficial effect on the occurrence of dental caries, particularly among children while excess intake of fluoride leads to various diseases viz. osteoporosis, arthritis, and cancer (Bhatnagar et al., 2011; Wambu et al., 2013). The maximum acceptable fluoride concentration set by the World Health Organization is less than 1.5 mg/L (Nagendra Rao and Karthikeyan, 2011; Singh et al., 2013).

Fluoride releases into the groundwater by slow dissolution of fluorine-containing rocks (Bhatnagar et al., 2011) and consumption of the contaminated groundwater is the major route of human exposure (Agarwal et al., 2003; Bhatnagar et al., 2011; Sujana et al., 2009). In addition, various industries involved in glass and ceramic production, semiconductor manufacturing, electroplating, coal fired power stations, beryllium extraction plants, brick and iron works, and aluminum smelters can add up the fluoride burden in groundwater (Bhatnagar et al., 2011; Sun et al., 2011).

al of fluoride has been demonstrated to be an effective strategy (Liu et al., 2010; Mandal and Mayadevi, 2009; Tripathy et al., 2006). Though activated carbon derived from various biomass resources has traditionally been exploited as adsorbent for fluoride removal, research is warranted to find out alternative adsorbents, such as clav and clav minerals that are naturally abundant, renewable, and environmentally sustainable (Bergaya and Lagaly, 2013; Crini, 2006; Srinivasan, 2011). Due to their low cost, high sorption properties and potential for ionexchange, clay materials are considered as robust adsorbents (Crini, 2006). Natural clay minerals with high surface area, molecular sieve structure, chemical and mechanical stability, variety of surface and structural properties are very effective adsorbents (Sparks, 2003; Srinivasan, 2011). The potential application of clay and clay minerals in removal of fluoride from water is well documented. With substantial amounts of gibbsite or aluminum oxides, clays exhibit immense potential for fluo-

Fluoride removal from water can be achieved by various physiochemical and biological methods (Mekonen, 2001). Biological methods

alone may not be effective, rather an integrated approach with physio-

chemical treatment is appropriate (Mekonen, 2001). Adsorptive remov-

gibbsite or aluminum oxides, clays exhibit immense potential for fluoride adsorption (Agarwal et al., 2002a; Bhatnagar et al., 2011; Sujana and Anand, 2011; Tomar and Kumar, 2013). Positively charged surface of the clay minerals explains high affinity uptake of negatively charged ions at near neutral pH. The fluoride removal capacity of clay and clay minerals is influenced by many factors, such as pH, ionic strength and thermodynamic conditions of the medium (Gitari et al., 2015; Kamble

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# et al., 2009; Kim et al., 2013; Maiti et al., 2011; Sarkar et al., 2006b; Srinivasan, 2011; Thakre et al., 2010; Tor, 2006).

This paper aims at providing a state-of-the-art review on the use of clay and clay minerals for fluoride removal from water. It gives a brief description about the effect of various factors viz. adsorbent dose, contact time, particle size, initial fluoride concentration, pH, co-existing ions on adsorption, mechanism, isotherms and kinetics of the adsorption process. This review intends to offer a starting point for future studies that should be able to confirm or complete the actual findings, and improve knowledge in this field.

#### 2. Clay and clay mineral based adsorbents for fluoride removal

An array of clay and clay minerals has been evaluated for their potential defluoridation properties (Table 1). However, non-implementation of reference material and inherent compositional variance of the "natural" clay minerals used across different studies do not permit their parallel comparison. Influence of different experimental conditions (fluoride concentration, adsorption time, flowrate, and adsorbent dose) can only be interpreted when included as experimental subset in a single study design. The raw clay or clay minerals are rarely utilized and they are often used after physico-chemical modifications such as metal oxide amendment, acid treatment, and thermal activation. Such modifications offer better stability at the optimal operating pH, accessibility of active sites, adsorption capacity, and surface area (Gitari et al., 2015; Kamble et al., 2009; Maiti et al., 2011; Meenakshi et al., 2008; Thakre et al., 2010).

In mechanochemical activation of kaolinites using an oscillatory disc mill, Meenakshi et al. (2008) reported an enhancement of surface area from 15.11  $m^2/g$  (raw kaolinite) to 32.43  $m^2/g$ , which ultimately led to formation of new active surfaces resulting in increased fluoride adsorption capacity (from 0.096 mg/g to 0.106 mg/g). In another study, Gogoi and Baruah (2008) reported effective defluoridation with acid-activated kaolinite in comparison to raw kaolinite, where maximum sorption capacity of the acid activated clay ranged between 0.0450 mg/g and 0.0557 mg/g at different temperatures. Hamdi and Srasra (2007) reported fluoride adsorption capacity as high as 93.45 mg/g for a Tunisian clay containing kaolinite and a small amount of smectite.

Maiti et al. (2011) reported enhanced adsorption capacities in laterite when subjected to sequential acid–base treatment. Due to the acid– base treatment on laterite, there was an increase in surface area from nearly 17.5  $m^2/g$  to178  $m^2/g$  and the maximum adsorption capacity of fluoride on treated laterite was found to be 39.1 mg/g. Heat treatment (400 °C, 2 h) can increase fluoride adsorption capacity in laterite, possibly from de-hydroxylation of minerals during heat treatment (Osei et al., 2015).

Chemical modification with different metal oxides (e.g. lanthanum, magnesium, manganese) has often been comparatively tested for enhanced fluoride removal capacity of bentonite. However, no metal oxide had been identified to be uniquely optimal. For example, Kamble et al. (2009) reported lanthanum modified bentonite to have the best efficiency. However, Thakre et al. (2010) have found better fluoride adsorption capacity of MgCl<sub>2</sub> incorporated bentonite than the unmodified one. Gitari et al. (2015) found nearly complete fluoride removal from aqueous phase system using Fe<sup>3+</sup>-modified bentonite at an adsorption capacity of 2.91 mg/g. In another study, fluoride removal by Al<sup>3+</sup> modified bentonite was about 5.7 mg/g at room temperature (Vhahangwele et al., 2014). However, chemical modification has not always proven to be effective. For example with granular acid treated bentonite (specific area 24.5 m<sup>2</sup>/g), Ma et al. (2011) reported equilibrium fluoride adsorption capacity of just 0.07 mg/g.

Organic modification of Tunisian smectite with cationic surfactants (hexadecylpyridinium bromide and hexadecyltrimethylammonium bromide) has been shown to offer better fluoride removal capacity (17.9, 11.95 mg/g respectively at pH 5.9) than the unmodified clay

(<2 mg/g) (Gammoudi et al., 2013). The authors explained the increased fluoride removal capacities to changes in the surface property of the clay during their modification where surfactant cations can attract and electrostatically hold fluoride anion, which replaces bromide ions associated with the surfactant head groups in an anion exchange process. Using calcium rich and calcium free natural montmorillonite, Ramdani et al. (2010) studied the influence of chemical modification (sodium exchange) and thermal activation (200 and 500 °C) on fluoride removal efficiencies. Results suggested chemical activation (with RE of 88%) to be more effective than the thermal activation (RE of only 5%). The authors attributed the decreased fluoride adsorption after thermal pretreatment to significant decrease in surface hydroxyl groups, and surface area. Moriyama et al. (2014) reported decreasing fluoride sorption density of bimetallic oxides prepared from hydrotalcite under increasing calcination temperatures. Increased proportion of MgAl<sub>2</sub>O<sub>4</sub> phase, not contributing to fluoride immobilization, at higher calcination temperature was attributed for the behavior.

## 3. Fluoride removal from water by clay and clay minerals

## 3.1. Effect of adsorption parameters

## 3.1.1. Contact time

Contact time is the agitation time required for the adsorbent–adsorbate system to reach equilibrium which strongly depends on factors, such as the pore structure of the adsorbent, adsorbent particle size, type of the adsorbate, and adsorbate concentration (Goswami and Purkait, 2011; Guo and Reardon, 2012; Meenakshi et al., 2008; Ramdani et al., 2010; Thakre et al., 2010). With pyrophyllite, Goswami and Purkait (2011) reported that fluoride removal can reach about 85% within the first 20 min but then slows down reaching equilibrium after 120 min for an initial fluoride concentration of 4 mg/L. This behavior was attributed to the ionic interaction of fluoride ion with pyrophyllite particles. The authors have explained that aggregation of fluoride molecules on pyrophyllite as mesopores get filled up and start offering resistance to diffusion. A similar change in the removal efficiency (RE) with bentonite was attributed to progressive loss of solute

#### Table 1

Clay minerals used for fluoride removal from water.

Clay minerals	References
Clay minerals without modification	
Allophane	Kaufhold et al. (2010)
Bentonite	Srimurali et al. (1998)
Chlorite and illite	Du et al. (2011)
Kaolinite	Agarwal et al. (2002a) and
	Srimurali et al. (1998)
Laterite	Sarkar et al. (2007), Sarkar et al.
	(2006a), and Vithanage et al.
	(2012)
Montmorillonite	Agarwal et al., 2002b, Ramdani
	et al. (2010), and Tor (2006)
Pyrophyllite	Goswami and Purkait (2011) and
	Kim et al. (2013)
Clay minerals with modification	
Calcined meixnerite	Guo and Reardon (2012)
Calcined Layered Double Hydroxides	Lv et al. (2006)
Chemically modified bentonite (10%	Kamble et al. (2009)
lanthanum bentonite)	
Chemically treated laterite	Maiti et al. (2011)
Double layered hydroxides of Mg-Al-CO <sub>3</sub>	Batistella et al. (2011)
Туре	
Fe <sup>3+</sup> -modified bentonite	Gitari et al. (2015)
Hydrotalcite (anion clay)	Guo and Guo (2013)
Magnesium incorporated bentonite	Thakre et al. (2010)
Mechanochemically activated kaolinite	Meenakshi et al. (2008)

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