

## Defluoridation potential of jute fibers grafted with fatty acyl chain



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### ABSTRACT

Waterborne fluoride is usually removed from water by coagulation, adsorption, ion exchange, electro dialysis or reverse osmosis. These processes are often effective over narrow pH ranges, release ions considered hazardous to human health or produce large volumes of toxic sludge that are difficult to handle and dispose. Although plant matters have been shown to remove waterborne fluoride, they suffer from poor removal efficiency. Following from the insight that interaction between microbial carbohydrate biopolymers and anionic surfaces is often facilitated by lipids, an attempt has been made to enhance fluoride adsorption efficiency of jute by grafting the lignocellulosic fiber with fatty acyl chains found in vegetable oils. Fluoride removal efficiency of grafted jute was found to be comparable or higher than those of alternative defluoridation processes. Infrared and X-ray photoelectron spectroscopic evidence indicated that hydrogen bonding, protonation and C–F bonding were responsible for fluoride accumulation on grafted jute. Adsorption based on grafted jute fibers appears to be an economical, sustainable and eco-friendly alternative technique for removing waterborne fluoride.

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### 1. Introduction

Presence of elevated fluoride levels in drinking water is known to affect skeleton development and dental health. Such anionic waterborne species are usually removed from water by precipitation or coagulation by alum and lime treatment [1] or by adsorption with activated alumina [2,3]. However, the presence of leached aluminum ions in hazardous concentrations in treated water compromises the usefulness of these processes [4]. Waterborne fluoride has also been removed through ion exchange, electro dialysis and reverse osmosis [5,6]. These processes are expensive to operate and maintain, generate large volumes of toxic sludge that are difficult to dispose or lead to the wastage of large volumes of water. Carbon nanotubes and metal oxides nanoparticles have been used to adsorb waterborne fluoride [7,8]. Potential cytotoxicity and genotoxicity of these adsorbents [9,10], calls for careful evaluation of the impact of their use in water treatment.

Sustainability and economic considerations have also prompted evaluation of lignocellulosic plant matters in adsorbing waterborne fluoride [11]. Since untreated biomaterials suffer from poor fluoride removal efficacies, they were pyrolyzed [12], loaded with metal

ions [13], or chemically treated [14,15] before using in water treatment. These biosorbents require energy-intensive pre-processing or exhibit harmful leachate generation potential. Additionally, these adsorbents are efficient only over narrow pH ranges. Lanthanum [16] and aluminum [17] cross linked alginate beads have also been used for efficient adsorption of waterborne fluoride although their leachate generation potential limits their usefulness.

Among available processes for removal of waterborne fluoride, adsorption-based processes appear attractive because by and large they do not produce sludge or require frequent expert maintenance. However, adsorbents are often expensive, require energy-intensive processing, exhibit poor removal efficiencies or are potential sources of harmful waterborne chemical constituents. Cost of water purification using such adsorbents is also therefore high ([8,18–20]; Fig. 1). Recent studies have shown that affinity of biomaterials to anionic surfaces is facilitated in the presence of lipids [21,22]. It therefore appears grafting an inexpensive waste lignocellulosic plant matter, e.g., jute (*Corchorus olitorius*) fibers collected from natural fiber processing industry, with fatty acyl chains present in low-cost varieties of generic vegetable oils could lead to the development of an inexpensive yet efficient biosorbent with demonstrable fluoride removal potential. Such a biosorbent has been prepared in this study and evaluated for its fluoride removal efficiencies and leachate generation potential. Interactions

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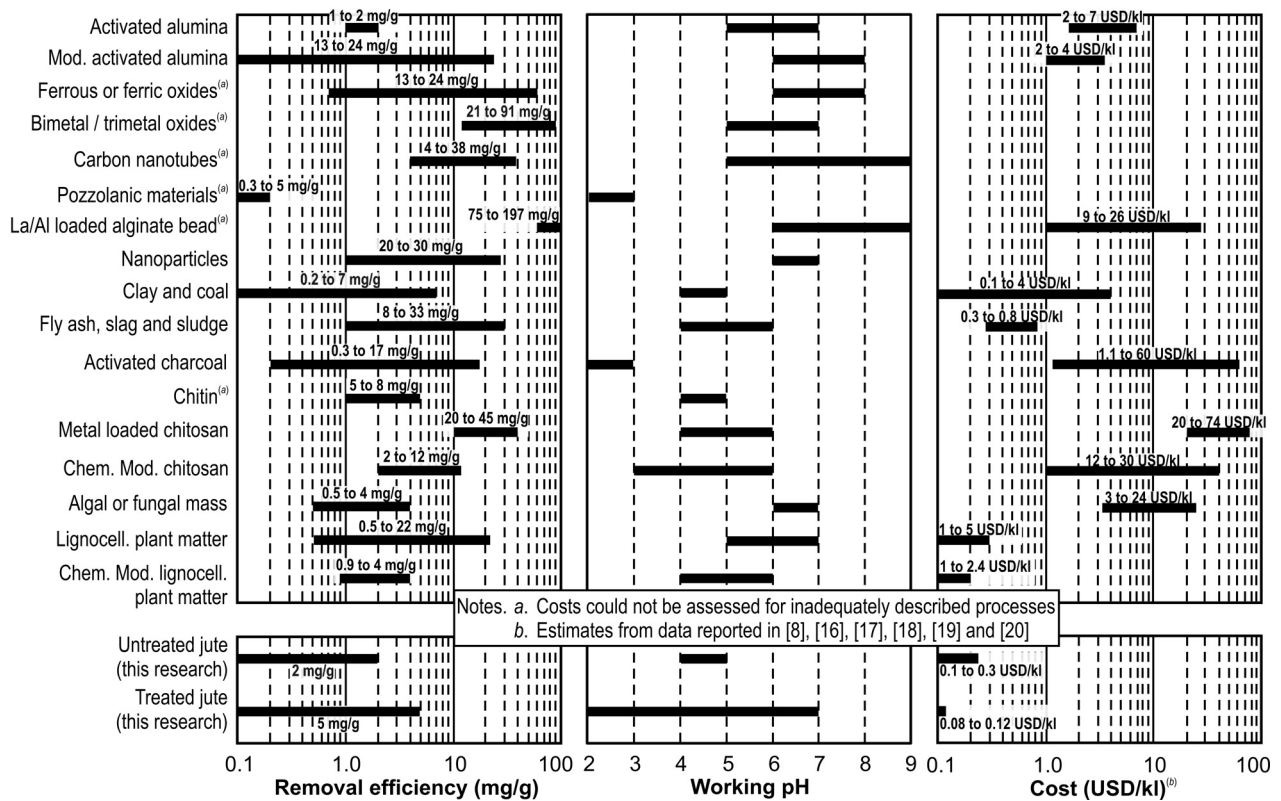


Fig. 1. Comparison of fluoride removal processes.

between the surface of grafted jute and fluoride ions were also assessed with spectroscopic measurements.

## 2. Experimental

### 2.1. Processing and characterization of jute

Waste fiber of jute washed with distilled water, dried at  $85 \pm 2^\circ\text{C}$ , and chopped mechanically in a mixer grinder. Ground fibers, with nominal lengths smaller than  $300 \mu\text{m}$ , were treated with a dilute alkaline solution or an alkaline aqueous emulsion of neem (*Azadirachta indica*) oil and used in this research. For alkali treatment chopped jute (UJ) powder was immersed within 0.5% (w/v) NaOH (Merck) aqueous solution for 30 min at  $30 \pm 2^\circ\text{C}$  and subsequently steamed at 103 kPa for 30 min at  $121^\circ\text{C}$ . Thereafter, the ground plant matter was washed with distilled water to bring down the pH to 7.0. A portion of alkali treated jute (AJ) powder was treated further by keeping it immersed for 15 min within an alkaline aqueous emulsion of neem oil and phenolic resins (NOPR) and squeezing out excess emulsion and leachate from recovered powder. The resin soaked jute fibers were then cured at  $105 \pm 5^\circ\text{C}$  for 1 h. The NOPR treated jute (NJ) powder was then washed with ethanol and water to remove unused reagents and oven dried at  $70 \pm 2^\circ\text{C}$  for 24 h. The emulsion used was prepared by mixing sodium hydroxide, cashew nut shell liquid, resorcinol, neem oil and formaldehyde in 0.5:1:2:5:2 proportion by weight with 1000 ml of distilled water. The treated biomaterial does not leach any of the chemicals used in the treatment in toxic concentration. Surface area of UJ, AJ and NJ powders obtained following [23] using an air permeability apparatus (AIM390, Aimil Ltd., India) were  $1.9 \text{ m}^2/\text{g}$ ,  $2.4 \text{ m}^2/\text{g}$  and  $2.1 \text{ m}^2/\text{g}$ , respectively.

#### 2.1.1. Fourier transform infrared spectroscopy (FTIR)

Pellets prepared from potassium bromide (KBr) and dried jute (1–2 mm long) in ratio 100:1 (w/w) were subjected to FTIR spectroscopy in a spectrophotometer (Thermo Nicolet, Nexus 870) for assessing jute chemistry. Wave numbers between  $3500 \text{ cm}^{-1}$  and  $500 \text{ cm}^{-1}$  were covered using 32 scans in the investigation. The ratio of absorption intensities representing OH vibration (at wavenumber of  $3300 \text{ cm}^{-1}$ ),  $I_{3300}$ , and CH vibration (at  $2980 \text{ cm}^{-1}$ ),  $I_{2980}$ , was used as an index of mean hydrogen bond strength (MHBS) [24].

#### 2.1.2. X-ray photoelectron spectroscopy (XPS)

A Thermo Scientific K-Alpha XPS spectrometer (ThermoFisher, E. Grinstead, UK) available at the University of Toronto was used for characterizing treated jute powders before and after exposure to fluoride solutions. Samples were affixed to the sample platen by spreading a spatula tip of biosorbent powder to double-sided adhesive C tape such that the tape was completely covered with powder and there was no cross-contamination. Excess powder was blown away from the tape before placing the platen in the load-lock. After subjecting it to pumping for about 15 min the platen was loaded into the analysis chamber. The samples were run at a take-off angle (relative to the surface) of  $90^\circ$ . A monochromatic Al  $K_{\alpha}$  X-ray source was used, with a spot area (on a  $90^\circ$  sample) of  $400 \mu\text{m}$ . Charge compensation was provided using combined  $e^-/\text{Ar}^+$  flood gun supplied with the instrument, with the position of the energy scale adjusted to place the main C1s feature (C–C bonding) at  $285.0 \text{ eV}$ . A survey spectrum (200 eV pass energy, 5 scans, 1 eV step size) was first collected followed by scans of the regions of the elements (2 scans for C1s, 2 scans for O1s, 10 scans for Si2p and 10 scans for F1s) at low resolution (150 eV pass energy, 0.2 eV step). Relative atomic percentage was taken from these spectra using sensitivity factors supplied with the instrumentation. Finally, high resolution spectra

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