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Fluoride Adsorption from Water on Waste Materials

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Abstract

The occurrence and concentrations of fluoride in surface and groundwater depend on pH, total dissolved solids, alkalinity, hardness, and geochemical composition of aquifers. However, in many countries, elevated fluoride concentration values are the result of fluoride-contaminated wastewater discharges. Because of fluoride pollution and the health problems that it causes, the World Health Organization (WHO) has set a maximum permissible fluoride content in drinking water of 1.5 $\text{mg}\,\text{l}^{-1}$.

There are different ways of removing elevated concentrations of fluoride from water, such as coagulation and precipitation, membrane processes, electrochemical treatments, ion-exchange and its modification, but the adsorption process is generally accepted as the cheapest and most effective method for removing fluoride from water.

Organic waste is increasing every day, especially in developed countries, and is generated in both industries and households. One of the ways to reduce such waste is the production of adsorbents for water defluorination. Adsorbents, most often prepared as activated carbon, can be obtained from various materials such as egg shells, fruit and vegetable peel, various leaves, stems, trunk bark, grain shells, legume shells, and many others.

The aim of this paper is to provide an overview of the latest research on the use of adsorbents obtained from organic waste materials in order to remove elevated concentrations of fluoride from water.

Keywords

Fluoride, water, adsorption, waste materials, fluoride removal

1 Introduction

The daily intake of an optimal concentration of fluoride through water, food, and various supplements in the body causes increased mineralisation of teeth and an increase in bone density, as well as reduces the risk of caries.[1] Intake of excessive fluoride concentration in the body causes numerous health problems such as dental fluorosis,[2] bone fluorosis,[3] central nervous system disorders.[4]

Fluoride concentration in drinking water greater than 1.5 $\text{mg}\,\text{l}^{-1}$ is a global health problem, as more than 250 million people worldwide consume contaminated water.[5] In groundwater, the concentration of fluoride is primarily determined by the geological composition of the aquifer, but the influence of various industries, which significantly increase their concentration through the discharge of wastewater and gases, is increasing.[6–8] Research has shown that 80 % of all diseases in the world are caused by the reduced quality of drinking water, while the presence of increased fluoride concentrations in drinking water is responsible for 65 % of endemic fluorosis in the world.[9] *Viswanathan et al*.[10], investigating the presence of fluoride in the waters of South India, found that 50 % of groundwater sources are contaminated with fluorides and more than 90 % of rural settlements obtain their drinking water from these sources.

2 Adsorption

There are many treatment technologies used for fluoride removal, including precipitation, membrane separation, ion exchange, reverse osmosis, advanced oxidation processes, electrocoagulation, photocatalytic disinfection, and electrodialysis. The disadvantages of these treatment methods are that they are energy-intensive, expensive, require high operational and capital inputs, and require advanced technology and skilled labour. On the other hand, adsorption is a flexible, efficient, easy-to-design and cost-effective treatment process.[11–13]

Adsorption is one of the procedures for removing high concentrations of fluoride from water, which has been increasingly used in recent years. The method is extremely economically profitable and efficient. Adsorption on solid adsorbents is the most effective when purifying water with different degrees of pollution. With their use, the water is not additionally burdened by the dosing of chemicals as it is with some other conventional methods of water treatment. Common to research on fluoride adsorption is the emphasis on the importance of the characteristics of the adsorbent. Its suitability for practical application is determined by adsorption capacity, selectivity for fluoride ions, regenerability, compatibility, particle and pore size, and price. Fluoride removal efficiency always depends on the initial fluoride concentration, pH, temperature, contact time, and adsorbent dose.[14]

In recent years, there has been increased research in the direction of applying materials for the production of activated carbon from the biomass of different plant species.

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Sivasankar et al.[15–17] examined the possibility of adsorption of fluoride ions on activated carbon obtained from the shell of the tamarind fruit. The mentioned raw material is very rich in calcium-containing components. During the carbonisation process, calcium is incorporated into the structure of the adsorbent and favours the adsorption of fluoride ions. Similar research was conducted by *Ramírez-Montoya et al*.[18] comparing the adsorption properties of pecan shell and plum stone without carbonisation, after carbonisation at 800 °C, and after impregnation with calcium acetate and carbonisation. Activated carbon obtained by impregnation and carbonisation has the highest adsorption capacity, while pecan shell has slightly better properties than plum stone.

Sinha et al.[19] investigated the possibility of fluoride removal by adsorption on carbonised and non-carbonised biomass of the plant *Eichhornia crassipes*. It was found that carbonisation favoured the adsorption capacity of the adsorbent, because in this way the proportion of inorganic substances increased thus enabling easier access of fluoride ions to the active surface of coal. Chemical and thermal activation of coconut bark and fibres and rice husk for the purpose of producing activated carbon was carried out by *Mohan et al*.[20] The mentioned agricultural by-products in India represent a significant environmental problem. The most suitable adsorbent was activated carbon prepared from coconut bark fibres, which removed more than a quarter of the fluoride from the solution. In the absence of certain functional groups on the surface of activated carbon obtained from materials of plant origin, and with the aim of increasing the adsorption capacity, impregnation methods with different chemical agents have been applied. One of the examples that confirm this statement is the activated carbon of the cashew shell impregnated with zirconium, which removed 80.33 % of fluoride ions. By using non-impregnated material, it was possible to remove 72.67 % of fluoride ions.[21] Similar tests were carried out by *Ramos et al*.[22] by impregnation of activated carbon of coconut bark with aluminium ions, which increased the adsorption capacity of the adsorbent four times.

3 Waste materials as adsorbents

Numerous authors have focused their research on developing and modifying adsorbents that will be economically profitable and effective (Table 1). For this purpose, a wide variety of materials, created as by-products in industrial production, have been used. There are a number of publications related to the use of polysaccharide materials that have a high affinity for fluoride ions. *Zhao et al*.[23] developed an iron-modified highly selective cellulose adsorbent obtained from cotton that successfully removed fluoride ions from drinking water in almost 100 % proportion, while *Tian et al*.[24] applied N,N-dimethyl aminoethyl methacrylate-modified form of cellulose as an adsorbent for fluoride and arsenic removal, where the adsorbent showed a better affinity for arsenic. In addition to cellulose, starch can also be used as an adsorbent for fluorides. For this purpose, *Sivasankar et al*.[25] impregnated starch obtained from potatoes with ammonium cerium(IV) sulphate hydrate, the maximum adsorption capacity of which was 29.1 mg l^{-1} at an optimal pH of 7.75. Orange peel and juice residues also contain numerous polysaccharides as the basis for modification with various multivalent metal ions such as Al^{3+}, La^{3+}, Ce^{3+}, Ti^{3+}, Sn^{4+} and V^{4+}, which create very stable compounds with fluorides and increase the adsorption effect of the adsorbent for the purpose of their removal.[26–29] In addition to the aforementioned, there is a number of adsorbents obtained from different waste materials such as bones,[30] eggshells,[31,32] sludge,[33,34] ash, and others.[35,36]

Tefera et al.[12] examined the possibility of fluoride adsorption on avocado seeds. Avocado-based activated carbon was a successful adsorbent, and the maximum fluoride removal from aqueous solution was found to be 86 %, whereas the fluoride removal from groundwater was 72 %. Adsorption on alternative materials such as agri-food residues can be a valid treatment for fluoride removal considering their optimal removal yield, high availability, and low cost. *Collivignarelli et al*.[8] and *Dehghani et al*.[37] pointed out that the interest in this issue is rapidly growing, over 50 % of the total research has been published in the last 10 years. There is also a great interest in the use of palm residues as an adsorbent for fluoride removal.

4 Conclusion

The problem of elevated fluoride concentrations in drinking water is present in developed and underdeveloped countries. Adsorption is a cheap and easily available method of fluoride removal from drinking water through different adsorption materials. People's awareness of the potential offered by waste materials that can be used for the preparation of adsorbents is increasing. There is also more and more research that provides clear guidelines and possibilities for the preparation of adsorbents that will effectively remove fluoride from water. Biomass from plants and agricultural waste by-products can be used for efficient fluoride absorption as well as solving the problem of their disposal. These cheap materials help to replace expensive commercial adsorbents like activated carbon, which again have a problem in regeneration. Agricultural waste and plant materials are available in huge quantities, biodegradable or non-biodegradable in nature, they are cheap and environmentally friendly.

Table 1 – Fluoride adsorption conditions on different waste materials
Tablica 1 – Uvjeti adsorpcije fluorida na različitim otpadnim materijalima

Adsorbent	Parameters of adsorption					Maximum fluoride removal / %	Ref.
	Concentration / mg l^{-1}	Adsorbent dose / g l^{-1}	Temperature / K	pH	Contact time / min		
Palmyrah (*Borassus flabellifer*) nut shells carbon	0.5–2	0.05–0.25	303–353	7	15–120	61	38
Oyster shell	2–12	0.1–0.5	283–313	3–9	10–90	97.26	39
Orange peel powder	10	0.5–2	298	7.5	1440	28	40
Citrus limetta peels	5–30	0.5–3	298–318	4–10	30–300	94.8	41
Waste peanut hull biochar	10–50	2–14	25–45	2–11	0–150	88.21	42
Cow dung carbon	2–8	1–6	298–318	7.5	30	99	43
Fish scale biochar	2–14	2–12	298	2–12	30–240	97.41	44
Citron peel	1.5–20	2–24	298	2–10	20–180	89.5	45
Ultrafine tea powder	5–200	0.4–4.8	298	3–12	0–350	93	46
Coconut fibre dust	1.5–15	0.05–2	303–333	2–10	20–240	97.2	47
Enterolobium saman fruit carbon	1–7	1–7	303	5–9	10–90	90.8	48
Prosofis juliflora fruit carbon	1–7	1–7	303	5–9	10–90	80.2	
Kenaf core fibre	5–80	0.2–1.6	298	2–9	4320	65	49
Tea waste	5–200	0.4–3.2	298	2–11	0–300	89,7	50
Nutshells carbon	2–100	10	298–318	7	120	46	51
Citrus limonum leaves	2–15	20–200	303	2–8	5–145	70	52
Sawdust of Indian Rosewood *(Dalbergia sissoo)*	2.5–15	1–10	301	4–9	60–75	49.8	53
Wheat straw *(Triticum* spp.*)*	2.5–15	1–10	301	4–9	60–75	40.2	
Tamarindus indica fruit shells	2–8	2	298	4–12	30	91.6	16
Rice husk	5	1–10	302	2–12	5–240	83	54
Orange waste	9.5–152	0.5–15	303	1–12	0–960	98	26
Coconut shells	507	40	283–313		0–1152	13	20
Coconut shell fibres	507	40	283–313		0–1152	27	
Rice husks	507	40	283–313		0–1152	2	

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SAŽETAK

Adsorpcija fluorida iz vode na otpadnim materijalima

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Pojavnost i koncentracije fluorida u površinskim i podzemnim vodama ovise o pH, ukupnoj otopljenoj krutoj tvari, alkalnosti, tvrdoći i geokemijskom sastavu vodonosnika, no u mnogim zemljama svijeta povišene vrijednosti koncentracije fluorida rezultat su ispuštanja otpadnih voda onečišćenih fluoridima. Zbog onečišćenja fluoridima i zdravstvenih problema koje ono uzrokuje Svjetska zdravstvena organizacija (WHO) odredila je najveći dopušteni sadržaj fluorida u pitkoj vodi od 1,5 mg l^{-1}. Postoje različiti načini uklanjanja povišenih koncentracija fluorida iz vode, kao što su koagulacija i taloženje, membranski procesi, elektrokemijski tretmani, ionska izmjena i njezina modifikacija, ali je adsorpcijski proces općenito prihvaćen kao najjeftinija i najučinkovitija metoda. Količina organskog otpada svakim je danom sve veća, posebice u razvijenim zemljama, a stvara se kako u industriji tako i u kućanstvima. Jedan od načina smanjenja takvog otpada je proizvodnja adsorbensa za defluorizaciju vode. Adsorbensi, najčešće pripremljeni kao aktivni ugljen, mogu se dobiti od raznih materijala, kao što su ljuske jajeta, kore voća i povrća, razni listovi, stabljike, kora debla, ljuske žitarica, ljuske mahunarki i mnogi drugi.

Cilj ovog rada je dati pregled najnovijih istraživanja o uporabi adsorbensa dobivenih iz organskog otpada za uklanjanje povišenih koncentracija fluorida iz vode.

Ključne riječi
Fluoridi, voda, adsorpcija, otpadni materijali, uklanjanje fluorida

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