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Cost-effective hybrid filter for remediation of water from fluoride

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Abstract. Incidence of fluoride concentrations in drinking water above the allowable limit (1.5 mg/L) leads to varied health issues. However, fluoride concentration below 1.5 mg/L in drinking water is useful for teeth and bones health. A considerable concentration of fluoride is naturally released to the sources of freshwaters from the geological environment, specifically the groundwater, because of the weathering and seepage of water phenomena influences. Unfortunately, nowadays world population depend on groundwater as the main drinking water source, which means those people are exposed to fluoride contaminations. As a result, contamination of groundwater with fluoride has been lately considered as a universal grave issue. Although fluoride could be removed from drinking water using efficient methods, such as reverse osmosis and filtrations, there is a challenge to develop a cost-effective practical removal method. This study examines the efficiency of an economically-efficient a hybrid filtration cell (HFC), which utilizes limestone and activated carbons, for fluorides removal from water. Batch flow experiments were conducted using HFC to remediate artificial water from fluorides. Additionally, the influences of initial pH, initial concentration of fluoride (IFC), water temperature (WT) and adsorbent dosage (AD) were optimized, using Box–Behnken approach, to reach the highest removal of fluorides. The results demonstrated that fluoride could be completely removed from artificial water when the HFC is run at pH of 5.0, IFC of 30 mg/L, AD of 30 mg/L and WT of 313 K.

1. Introduction

Water availability and purity determine its suitability for human needs and essential activities; including the demands for industry and agriculture, which means water determines the sustainability of human civilization [1-3]. In fact, although safe drinking water should be available to sustain the civilisations of human and well-beings, it is considered one of the human rights [4-6]. One of the main piles to achieve the overall world development, which includes food-security, educations and specially decreasing poverty, is insuring safe drinking water availability [4, 7-11]. Contamination level of water sources and the treatment efficiency are the core issues that determine the availability of safe drinking water in developing countries [12-14]. In addition, accessibility of safe drinking water in many nations (especially those with low national income) faces the following issues [7, 15-17]:

1- Cost of advanced treatment methods is high in comparison with the national income.
2- Lack of skills, sources of energy and the essential equipment for operating advanced treatment methods.

3- Disposal of the generated by-products from water treatment methods.

The fundamental and preferable source for drinking water on a global-scale is the groundwater, which represents 0.60% of the worldwide water resources. In addition, some parts of the developing countries completely depend on groundwater as drinking water source [18]. Ground water is preferable as a source of freshwater not only because of its usual better quality than surface water, but also because it is less contaminated due to the insulation from the external exposure by the geological cove [19]. Although construction of boreholes and wells requires high investment cost, the operational cost for groundwater treatment is relatively lower than that of surface water treatment. For example, some studies demonstrated that the required operational cost for treating the groundwater, in low population density areas (≤5,000 people), is half that of surface water [16].

One of the challenging issues that face groundwater treatment industry is fluorides contamination [17-19]. Human health is seriously threated by the long-term exposure to high fluorides concentration as it causes a number of serious diseases, such as nauseas and diarrhoeas [7, 20-22]. Thus, remediation of drinking water from fluorides becomes a persistent need [16]. Several methods for fluoride removal from drinking water or wastewaters were remarkably developed during the late decades, such as chemical coagulation, reverse osmosis, electrocoagulation and ion exchange [7, 23-27]. Despite the proven efficiency of these methods for the removal of various pollutants, the need for constant power supply, production of toxic intermediates, high investment cost or their low ability for the removal of organic matter determine their uses, especially in rural areas [1, 28-32].

The current research therefore was allocated to construct and apply an economically-efficient hybrid filtration cell (HFC) for water de-fluoridation. Additionally, this study examines the impacts of the initial pH, initial concentration of fluoride (IFC), water temperature (WT) and adsorbent dosage (AD) on the removal of fluoride by the HFC. The latter utilized limestone and activated carbons as filtration medium. The latters were chosen as adsorption media as they are affordable and worldwide available [19]. Additionally, they have a considerable affinity for fluorides, and their raw material are naturally available (limestone is a kind of natural rocks, an the activated carbons could derived from natural or waste materials) [19]. Furthermore, the adsorption methods produce small amounts of sludge, and the depleted limestone could be recycled in concrete production that enhances the cost-effectiveness of this method [33-38]. The last advantage of this method also reduces need for landfills for disposal of sludge of depleted media, which requires expensive transportation and management arrangements [2, 39-41].

2. Methodology

2.1. Fluoride solution

The grade of all the chemical materials utilised in this research is analytical and used directly without any processing. The supplier of these materials was Sigma Aldrich, UK. The preparation of fluoride artificial solution was by adding the desire weight of NaF (to have IFC of 300 mg/L) deionised water and stirred it to achieve a complete dissolve of NaF powder. This solution was used later to create samples with a lower IFC (30, 40 and 60 mg/L) by diluting it with the required volume of water.

1M HCl or NaOH solutions were utilised to adjust the initial pH of the diluted samples to the required value (4, 7.5 and 11).

2.2. Adsorption studies

Experiments of fluoride removal, using the HFC, were conducted in batch flow system. The adsorption media, which is consisted of equal weights of activated carbons and limestone, was placed in a plastic container with an active volume of 1.73 L, then fluoride solution was decanted into this container and left to react for 90 minutes. During the reaction period, 90 minutes, tiny volumes of solution were taken from HFC to determine the residual fluorides concentration. Whatman No.2 paper filters was utilised to filter the collected samples, then standard fluoride cuvette test (LCK-323) and spectrophotometer were applied to measure the residual fluorides concentration in the filtrate. The following equation was adopted to calculate the capacity of the filtration media to adsorb fluoride (mg/g) [42]:

$$Adsorption\ capacity\ \left(\frac{mg}{g}\right) = \frac{(C_i - C_e) \times V}{M} \quad (1)$$

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where,
C_i: IFC (mg/L)
C_e: fluoride equilibrium concentration
V: sample volume (L)
M: adsorbents dosage (g)
Fluorides removal, using HFC, was optimized for the impacts of the initial pH, IFC, AD and WT. The selection of these factors was in accordance with literature [4, 16]. A water bath (Clifton Ne1 Series) was utilised for controlling the water temperature. Table 1 presents the studied values of these factors.

Table 1. Studied values of IFC, AD, WT and initial pH

Parameters	Unit	Coded name	Studied levels		
			Low	Mid	High
IFC	mg/L	P1	30	45	60
Initial pH		P2	4	7.5	11
AD	g/L	P3	10	20	30
WT	K	P4	293	303	313

2.3. Optimization process
As it was mentioned, Box-Behnken design was utilized for optimizing of the effects of IFC, AD, WT and initial pH on fluoride removal by the HFC. Minitabe 19.2 package was used in this study to perform Box-Behnkendesing. As stated in Table 1, the fed values to the optimization process were INF of 30-60 mg/L, AD of 10-30 g, initial pH of 4-10 and WT of 293-313 K, while the the response was the observed adsorption (in mg/g). Table 2 lists the yielded experiments' design, which has been run in the laboratory.

Table 2: Experiments' design.

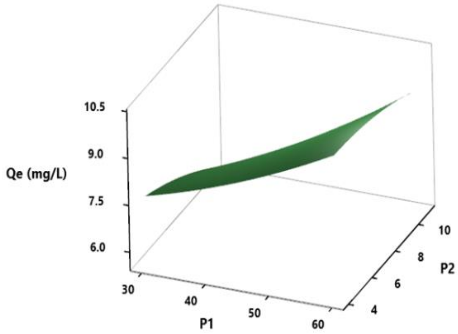
Runs	P1	P2	P3	P4	Runs	P1	P2	P3	P4
1	30	4	20	303	**15**	45	4	30	303
2	60	4	20	303	**16**	45	11	30	303
3	30	11	20	303	**17**	30	7.5	10	303
4	60	11	20	303	**18**	60	7.5	10	303
5	45	7.5	10	293	**19**	30	7.5	30	303
6	45	7.5	30	293	**20**	60	7.5	30	303
7	45	7.5	10	313	**21**	45	4	20	293
8	45	7.5	30	313	**22**	45	11	20	293
9	30	7.5	20	293	**23**	45	4	20	313
10	60	7.5	20	293	**24**	45	11	20	313
11	30	7.5	20	313	**25**	45	7.5	20	303
12	60	7.5	20	313	**26**	45	7.5	20	303
13	45	4	10	303	**27**	45	7.5	20	303
14	45	11	10	303					

3. Results
All the required 27 experiments, presented in Table 2, were conducted in the laboratory under controlled conditions. Human errors were avoided by repeating each experiment twice. Table 3 and Figures 1 list the calculated adsorptions for each experiment.

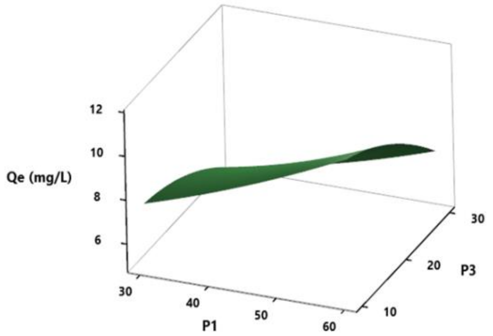
Table 3. The measured and predicted adsorptions of fluoride.

Runs	P1 (mg/L)	P2	P3 (g/L)	P4 (K)	Experimental adsorption (mg/g)	Predicated adsorption (mg/g)
1	30	4	20	303	7.119	7.849
2	60	4	20	303	9.656	10.249
3	30	11	20	303	6.125	5.807
4	60	11	20	303	9.780	9.325
5	45	7.5	10	293	8.382	8.639
6	45	7.5	30	293	4.701	5.162
7	45	7.5	10	313	10.681	10.495
8	45	7.5	30	313	7.288	7.307
9	30	7.5	20	293	6.340	6.274
10	60	7.5	20	293	9.274	8.960
11	30	7.5	20	313	7.099	8.002
12	60	7.5	20	313	10.579	11.234
13	45	4	10	303	10.743	11.098
14	45	11	10	303	6.209	7.416
15	45	4	30	303	6.189	5.567
16	45	11	30	303	6.044	6.282
17	30	7.5	10	303	8.312	7.871
18	60	7.5	10	303	11.587	11.472
19	30	7.5	30	303	5.079	5.180
20	60	7.5	30	303	7.072	7.497
21	45	4	20	293	7.140	7.294
22	45	11	20	293	6.851	7.113
23	45	4	20	313	10.869	10.597
24	45	11	20	313	7.980	7.811
25	45	7.5	20	303	7.731	8.308
26	45	7.5	20	303	9.076	8.308
27	45	7.5	20	303	7.691	8.308

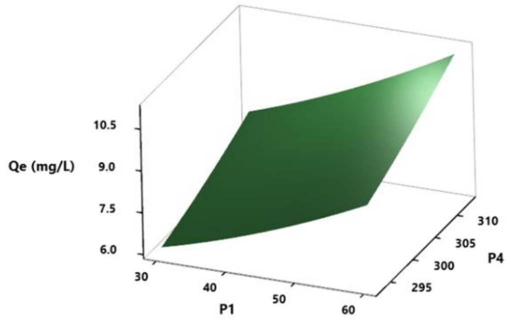
Generally, It could be noticed from these results that when the initial pH was in the moderate acidic range (<7), the performance of the HFC was better than it in the alkaline range (>7.0). The developed charges on the adsorbents' surface could be the reason behind this variation in the fluorides adsorption with the pH of solution [7, 19], where the negative fluoride ions are attracted by the positive charges of the adsorbents when pH is below 7. At the alkaline range, the developed charges on the adsorbents' surfaces are negative, so they repel fluoride ions that decreases the adsorption efficiency [7, 19]. The positive influence of AD on fluoride removal is because the number of active sites increases with increasing of AD that significantly maximizing the adsorption efficiency [42]. The relation between the fluoride adsorption and WT followed the similar trend to that in AD experiments, where increasing WT gives more thermal energy for ions of fluoride that increases the movements of the ions in the direction of the absorption sites, and thereby improve the adsorption efficiency [43]. However, ions of fluoride could be escaped from the surfaces of the adsorbent when temperatures are very high (>50 ^{o}C) because significant thermal energy is delivered to the ions of fluorides, so these temperatures must be avoided [43]. There are two possible perspectives to explain the influence of IFC on the adsorption efficiency of fluorides using the HFC. The first prospective is high IFC leads to a quick depletion of the available active sites, which results in a decrement in the adsorption efficiency [44]. The second prospective is the adsorption per unit of surface area increases with the increase of IFC, i.e., the increment in the IFC results in increasing the adsorption per unit of surface area due to the accumulation of high number of fluoride ions at the same active site [19].



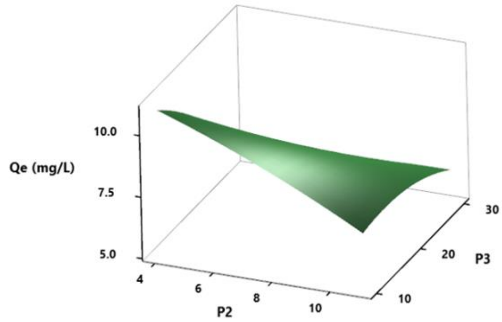
A) Fluoride adsorption vs IFC and initial pH.



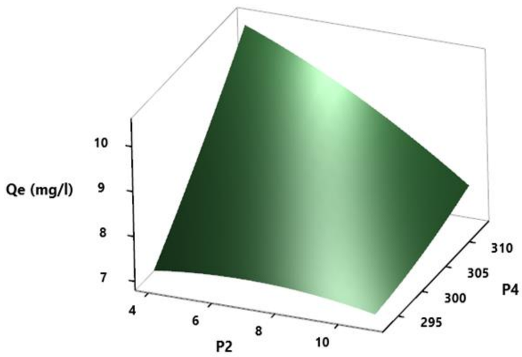
B) Fluoride adsorption vs IFC and AD.



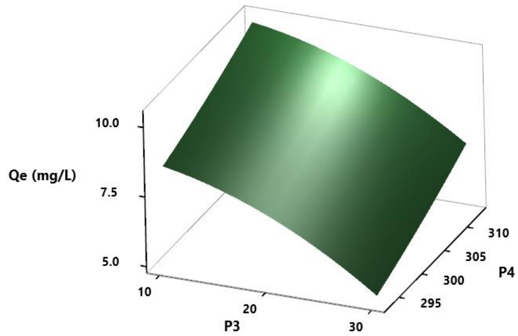
C) Fluoride adsorption vs IFC and WT.



D) Fluoride adsorption vs initial pH and AD.



E) Fluoride adsorption vs initial pH and WT.



F) Fluoride adsorption vs AD and WT.

Figure 1. Fluoride adsorption as a function of the studied parameters.

The outcomes of experimental runs have been fed to the Box-Behnken design to generate a model that gives an expectation about the effect of the studied factors; as following:

Adsorption (mg/L) $= 51.0 - 0.257P1 + 4.81P2 - 0.32P3 - 0.44P4 + 0.92 \times 10^{-3}P12 - 16.9 \times 10^{-3}P22 - 5.1 \times 10^{-3}P32 + 1.030 \times 10^{-3} \times P2 + 5.320 \times 10^{-3}P1 \times P2 - 2.140 \times P1 \times P3 + 0.91 \times 10^{-3}P1 \times P4 + 31.40 \times 10^{-3}P2 \times P3 - 18.60 \times 10^{-3}P2 \times P4 + 0.720 \times 10^{-3}P3 \times P4$
(2)

The determination coefficient of this model is (0.9329), so it could be considered as reliable one. This gives an idea that this model could reliably predict the outcomes of 93% of conducted experiments for fluorides removal from water using HFC. The predicted adsorption values, using the developed model, are well agreed with real ones as shown in Table 3.

Finally, the authors recommends, for future researches, to use sensing technology [45-49] to monitor the depletion of the filtration media that provides an accurate estimation for the life span of the filtration unit.

4. Conclusions

This research has been conducted to examine the adsorption of fluoride on hybrid filtration cell (HFC), which was made from activated carbon and limestone, in batch experiments, and it also examined the fundamental effects of the PH, dosage of adsorbent, water temperature and concentration of fluoride on the removal process. The outcomes of the experiments demonstrated that the HFC could be used efficiently for minimizing concentration of fluoride in drinking water in relatively short time (to ≤1.5 mg/L within 1.5 hours). In general, it was obvious that increasing the adsorbents dosage and water temperature resulted in an increment in fluoride removal, while increasing the initial fluoride concentration and initial pH resulted in a decrement in fluoride removal. The results obtained from this research could evident the applicability of the HFC to remove fluorides from drinking water in the rural areas in those countries who cannot afford the advanced treatment methods.
Future researches, some studies should investigate the suitability of the HF to remove hazardous pollutants and nutrients, such as arsenic and phosphate, from freshwaters.

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