Comparative Study of Fluoride Sorption Behaviour on Activated Carbon and Bone Char

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SUMMARY : Sorption behaviour of fluoride on acid washed activated carbon and deionised water washed bone char were comparatively studied in laboratory batch experiments. The experimental conditions for sorption were determined by varying the pH in the range of 3-8 and the take-up times. Both activated carbon and bone char seem to exhibit sorption behaviour independent of pH within the range 3-8. The observed saturation time was found to be 15 minutes for activated carbon, while the bone char was not at saturation even after 5 hours of contact time. At initial concentration levels of up to 1.6 mg F/L the fluoride removal is almost proportional to the dosage of sorbents. The obtained removal capacities were 0.1 and 0.4 mg/g respectively.

Key words: Activated carbon, bone char, fluoride, sorption, batch experiment.

INTRODUCTION

In general, substances with high porosity characteristics e.g. activated carbon, silica gel etc. have been used as sorbents for trapping other substances in various states. The efficiency of sorption has been widely investigated via measuring the up-taken sorbate left in solution through various techniques [1-4]. Defluoridation of water using activated alumina as a sorbent is one of sorption processes that has been used in many countries [5], while in Thailand trapping of trace pollutants from water has been done by filtering water through the activated carbon. In this work, comparative study of fluoride sorption behaviour on activated carbon and bone char were carried out for the aim of substituting bone char, a low cost and a common material from animals, to activated carbon for defluoridation of natural water for daily use in some fluorotic areas.

MATERIALS AND METHODS

All chemicals used were of analytical grade. Stock solution of 10.0 mg/L NaF was prepared in deionised water. The SPADNS, a colour-forming reagent was used for light absorption measurement [6]. Activated carbon and bone char were obtained from Carbokarn Company, Bangkok and the Intercountry Centre of Oral Health, ICOH, Chiang Mai, Thailand, respectively.

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The study of fluoride sorption behaviour was carried out on acid washed (6 M) activated carbon and deionised water washed bone char. The experimental conditions such as the saturation time and the pH that affected the sorption of fluoride were investigated by measuring the concentration of the residual fluoride spectrophotometrically (UV/VIS spectrophotometer; Hitachi Model U-2000, Japan) and confirmed by potentiometric measurement with fluoride ion-selective electrode (Fluoride combined electrode; Cole-Parmer Model 27502-19, U.S.A.). The pH adjustment in the range of 3-8 for the fluoride solution before equilibration was done by using acetic acid and ammonia. The fluoride sorption behaviour on activated carbon and bone char were then investigated by equilibrating 0.100 g of each sorbent in 50 mL NaF solutions ranging from 0.2-1.6 mg/L at the selected condition. The grain sizes of both activated carbon and bone char were in the range of 0.3-1 mm. The equilibration was normally done in a series of conical flask containing the reaction mixture. After an appropriate equilibrating time, the solution was then filtered and the residual fluoride concentration was determined.

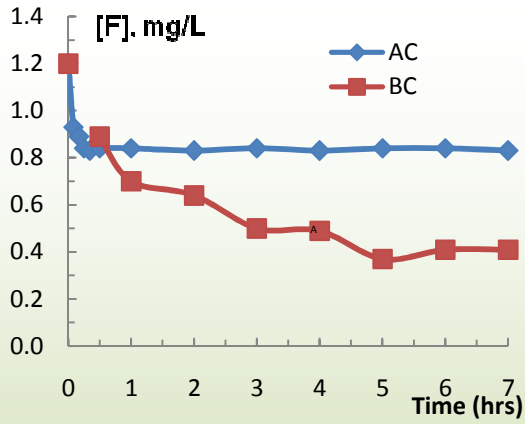

FIGURE 1. Concentration of fluoride remaining in the solution after being sorbed by activated carbon (AC) and bone char (BC) at various times. Dosage = 2.0 g sorbent/L.

RESULTS

After 0.100 g of each sorbent was separately equilibrated with 50 mL of 1.20 mg/L NaF solution at different periods of time, the residual fluoride concentration was determined. The reduction of fluoride in the solution as a function of time was shown in Figure 1. The effect of pH on the fluoride sorption by both sorbents were monitored. The result presented in Figure 2 showed that the fluoride sorption on both sorbents was independent on pH. The pH of 7.5 was selected for the sorption study. Then 0.100 g each of activated carbon and bone char was equilibrated with 50 mL of NaF solution at different initial concentrations for 15 minutes and 5 hours, respectively. The amount of fluoride sorbed in milligram per gram of sorbent was then plotted against the initial concentration of fluoride as in Figure 3.

DISCUSSION

From the study of fluoride sorption behaviour on activated carbon and bone char, the amount of fluoride left in solution after equilibrating 0.100 g of each sorbent separately in 50.0 mL of 1.0 mg/L NaF solution at various times were determined spectrophotometrically. For the activated carbon the sorption at about 15 minutes was sufficient to obtain equilibrium, whereas in case of bone char the sorption was found to continue changing without reaching saturation even at 5 hrs. of contact time. The difference between these sorption behaviours could be rationalised simply due to the fact that bone char possessed more cavities inside the structure. In such case these pores would allow fluoride ions to wander in by diffusion. The greater the amounts of the pores present, the longer time for sorption that occurred inside the cavities would take place.Figure 2 revealed the effect of pH on the fluoride sorption measured spectrophotometrically and potentiometrically. In case of the spectrophotometric method, the colour arises from Zr-SPADNS complex formation. The presence of fluoride would fade out the colour through the competitive formation of Zr-F complex. Therefore, the fading of the colour would indicate, for both sorbents, the amount of fluoride present in the solution. In case of activated carbon, the removal of fluoride

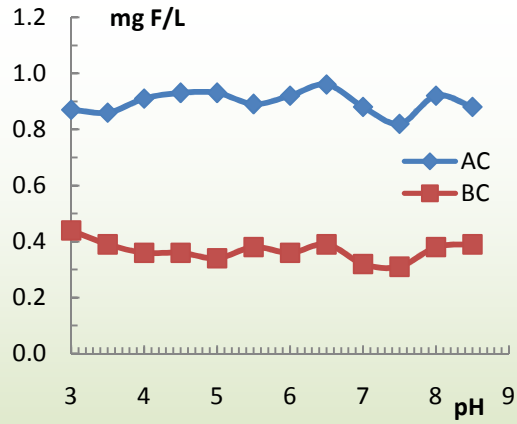

FIGURE 2. The residual fluoride concentration in the solution as a function of controlled pH after being sorbed by each sorbent. Initial concentration = 1.0 mg F/L. Dosage 2.0 g/L.

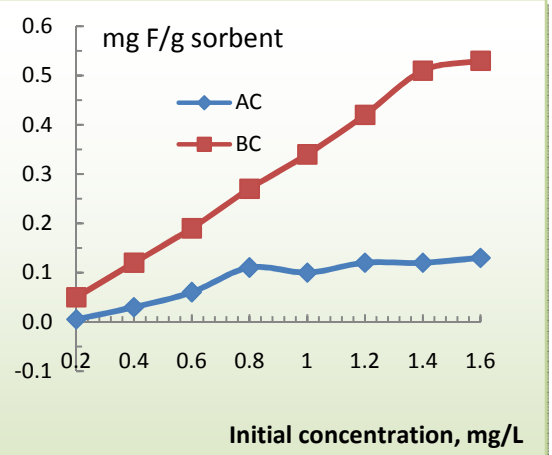

FIGURE 3. The amount of fluoride sorbed on each sorbent at different initial concentrations. Contact time = 15 minutes for AC and 5 hrs. for BC.
Dosage = 2 g sorbent /L.

from the solution occurred simply by physisorption. No matter what forms of fluoride present at the pH studied, the sorption would depend only on the availability of the active sites. Therefore, the amount of fluoride being sorbed on the activated carbon is independent of the pH of solution. But in case of bone char, because the removal of fluoride is often believed to be an ion exchange process [7], the fluoride must be in an anionic form. However, at pH in the range of 3-5, the amount of residual fluoride tended to decrease slightly due to the molecular HF form that existed at the low pH began to dissociate. Considering the pK_a value of 3.17 for HF; at pH above 5, the dissociation of HF would lead the ion exchange process to proceed and the decrease of fluoride in the solution caused the absorbance to increase. Nevertheless, the pH effect on fluoride sorption studied by potential measurement only showed slightly noticeable change in the sorption also with the bone char. So, the maximum sorption of fluoride was found to occur at the working pH in the region of pH 7-8. The sorption behaviour of fluoride, at room temperature, on activated carbon and bone char were studied through the potential measurement of fluoride solution after equilibrating each sorbent in various concentrations of NaF solutions at the optimal condition. It was observed from Figure 3 that fluoride sorption on activated carbon reached a certain degree of saturation if the initial fluoride concentration was higher than 0.8 mg/L, whereas the sorption on bone char was not conclusive to be a monolayer type. However, the amount of sorbed fluoride on activated carbon was found from this Figure to be about five times to that of bone char at the initial concentration of 1.6 mg/L fluoride solution.

ACKNOWLEDGEMENT

This work was partly funded by the Faculty of Science, Chiang Mai University under the Surface Modification and Colloids Research Laboratory. Thanks also for the Intercountry Centre of Oral Health, Chiang Mai, Thailand, for providing bone char used in this research.

REFERENCES

1. Lu HB, Campbell CT, Graham DJ, Ratner B. Anal Chem. 2000; 72: 2886-2894.
2. Cheung CW. Sepn Pur Technol. 2000; 19: 55-64.
3. Kerdpin U, Arquero O, Watanesk R, Sriyotha UJ Sci Soc Thailand 1998;24:73-80.
4. Monks CD, Nacapricha D, Taylor CG. Analyst 1993; 118: 623.
5. Karthikeyan G, Apparao BV, Meenakshi S. In: Dahi E and Neilsen JM, editors. The International Society for Fluoride Research. Proceedings of the 2nd International Workshop on Fluorosis and Defluoridation of Water; 1997 Nov 19-22; Nazreth, Ethiopia; Dunedin: New Zealand; 1998.
6. Bellack E, Schouboe PJ. Anal Chem 1958; 30: 2032-2034.
7. Williams RAD, Elliott JCE. Basic and Applied Dental Biochemistry. 2nd ed. Longman: Singapore; 1989.