

NANOFILTRATION AND REVERSE OSMOSIS FOR DEFLUORIDATION: THE ROLE OF INORGANIC CARBON

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Fluoride (F) concentrations above the World Health Organization (WHO) guideline value of 1.5 mg/L in drinking water can lead to serious health problems such as dental fluorosis and skeletal fluorosis. High F levels are often associated with carbonaceous (i.e. high inorganic carbon (IC)) type waters. The high fluoride concentrations in natural waters often occur in arid regions where no sufficient quantity of alternative water is readily available due to scarcity of water, consequently, treatment is the best option to provide safe drinking water.

Nanofiltration (NF) and reverse osmosis (RO) are promising and appropriate membrane technologies for defluoridation due to their high fluoride removal efficiency and their ability to simultaneously remove a wide range of other inorganic and organic contaminants¹. Different ions can have various effects on F removal by NF/RO². IC in natural waters is present as carbonate ion (CO_3^{2-}), bicarbonate ion (HCO_3^-), carbonic acid (H_2CO_3), and carbon dioxide (CO_2) depending on the pH. Due to the different characteristics of these species it is important to study the impact of IC on F retention mechanisms at different pH. In this study the mechanisms of IC species impact on F retention by NF/RO has been investigated as a function of pH. Two commercial NF and RO membranes, BW30 and NF270 respectively from DOW Chemicals (USA) were used. Synthetic waters were prepared using realistic ranges of F and IC for carbonaceous waters found for example in the fluoride rich waters in Tanzania. Feed concentration of F and IC were 50 mgF/L as NaF and 500 mgC/L as NaHCO_3 respectively. Visual MINTEQ software was used to predict the speciation of IC and F at various pH.

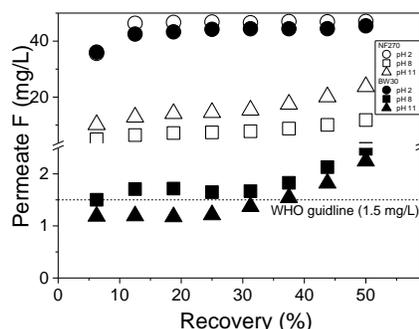


Figure 1: Influence of IC on permeate F. (IC concentration is 500 mg/L and F is 50 mg/L).

Figure 1 indicates that the permeate F concentrations were high (35-47 mg/L) at pH 2 where F existed mainly as uncharged HF. At pH 8 and 11, when there was a change in speciation to F ion and the membranes were negatively charged, permeate F concentrations decreased drastically. Permeate F concentrations for the RO BW30 membrane were lower than that of the relatively open NF270. BW30 removed fluoride to meet WHO guideline value of 1.5 mg/L at pH 11 up to about 40% recovery, while NF270 did not achieve the guideline value at any pH studied. In the past, BW30 had achieved the guideline value at pH 8 but with relatively low electrical conductivity (EC) of about 2000 $\mu\text{S}/\text{cm}$. However, in the current study the high IC concentration resulted in high EC (3600 $\mu\text{S}/\text{cm}$) and high osmotic pressure. This led to a decrease in the net driving pressure thus facilitating the diffusion of F through the membrane³.

The predominant IC species at pH 2 was H_2CO_3 . This resulted in very low feed IC concentrations (1-5 mg C/L, less than intended 500 mgC/L) at pH 2 due to degassing (H_2CO_3 decomposes to CO_2). Monovalent HCO_3^- predominated at pH 8 and easily permeated the NF270 membrane. At pH 11, MINTEQ predicted divalent CO_3^{2-} as the predominant IC species and permeate IC was lower than at pH 8 for the NF270. For the BW30 membrane, IC concentrations in the permeates were lower than the NF270 and impact of IC speciation was not observed. This suggests that the main retention mechanism of IC by the NF270 is charged repulsion and that of the RO BW30 membrane is size exclusion.

IC species influenced F retention by the NF270 membrane. Permeate F concentrations at pH 8 was lower than pH 11 (see Figure), when there was a shift of IC speciation from HCO_3^- to CO_3^{2-} . The decrease in fluoride retention at pH 11 can be explained by the rejection of CO_3^{2-} ion which created an electric field and forced F ion to penetrate the membranes. At pH 8, IC existed as HCO_3^- and was easily permeating the NF270 membrane compensate. The results obtained in this study indicate the complexity of F and IC retention by NF/RO.

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