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Review

Removal of fluoride and uranium by nanofiltration and reverse osmosis: A review

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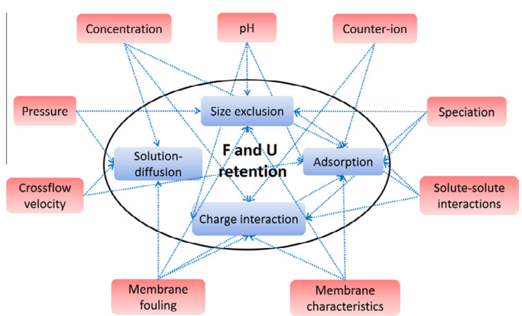
HIGHLIGHTS

- F and U occurrences and health implications are comprehensively summarized.
- Up-to-date progress on F and U removal by NF and RO are critically reviewed.
- F and U removal under various conditions are illustrated with mechanistic schematics.

GRAPHICAL ABSTRACT



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ABSTRACT

Inorganic contamination in drinking water, especially fluoride and uranium, has been recognized as a worldwide problem imposing a serious threat to human health. Among several treatment technologies applied for fluoride and uranium removal, nanofiltration (NF) and reverse osmosis (RO) have been studied extensively and proven to offer satisfactory results with high selectivity. In this review, a comprehensive summary and critical analysis of previous NF and RO applications on fluoride and uranium removal is presented. Fluoride retention is generally governed by size exclusion and charge interaction, while uranium retention is strongly affected by the speciation of uranium and size exclusion usually plays a predominant role for all species. Adsorption on the membrane occurs as some uranium species interact with membrane functional groups. The influence of operating conditions (pressure, crossflow velocity), water quality (concentration, solution pH), solute–solute interactions, membrane characteristics and membrane fouling on fluoride and uranium retention is critically reviewed.

Contents

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1. Introduction

Global demands for safe drinking water are increasing and inability to meet requirements is leading to increasing water conflicts (Mbonile, 2005; Sivakumar, 2011). Natural waters including groundwater, surface water (rivers and lakes) and rainwater are the main drinking water sources, while desalination of brackish and seawater is playing an increasing role. Groundwater, which constitutes 97% of global freshwater, is consumed for drinking purpose by more than 50% of the world population (Schmoll et al., 2006). In many remote and developing communities where basic water distribution systems are unavailable, groundwater serves as the most economically viable option (Ayoob and Gupta, 2006). However, groundwater often contains inorganic contaminants such as fluoride, uranium, arsenic, and boron amongst many others. Long term exposure to such contaminants causes health effects in humans. For example, excessive fluoride ingestion leads to dental and skeletal fluorosis (Fawell et al., 2006), and continuous uranium intake from drinking water has toxic effects on kidneys (Zamora et al., 1998). The occurrence of inorganic contaminants is highly geology-dependent. Therefore the practical approach to remove such ions/species/contaminants is to develop appropriate and flexible technologies for local use (Schwarzenbach et al., 2006).

Conventional water treatment methods involve a combination of adsorption, coagulation, flocculation, clarification, filtration and disinfection (Binnie and Kimber, 2009). The main drawback of conventional methods is that they are generally less effective for removing trace contaminants (Schwarzenbach et al., 2006). Nanofiltration (NF) and reverse osmosis (RO) are very promising techniques compared with conventional methods, in particular for drinking water applications. NF/RO can achieve high inorganic removal as they involve a mixture of separation mechanisms including solution diffusion, size exclusion, charge repulsion and adsorption. Suitable membrane characteristics can be selected to match particular water qualities. Inorganic contaminants in groundwater are often accompanied by bacteria, viruses and micropollutants such as pesticides which are also undesirable. NF/RO can simultaneously remove those contaminants in one single process, while the removal of micropollutants depends on specific characteristics. Furthermore, NF/RO is modular in design and flexible in implementation, making it a suitable choice for remote communities. Nevertheless, the possible drawbacks of NF/RO cannot be denied, those include membrane fouling and scaling, concentrate disposal and relatively high energy consumption. Membrane fouling and scaling are inherent in the separation process but can be significantly reduced by optimizing the operation conditions while scaling depends on the likelihood of precipitate formation in a particular water. High energy consumption can be compensated by introducing renewable energy technologies (Schäfer et al., 2006; Richards et al., 2008). Given the very good water quality produced by NF/RO, it is a good practice to use such water predominantly for potable purposes. Feed water quality permitting usage of concentrates for non-potable purposes provides near zero discharge opportunities.

However, inorganic contaminants in water involve a wide range of chemical characteristics and the removal mechanisms by NF/RO are not well understood for all. Inorganics such as fluoride and uranium are rapidly emerging issues of water quality which are of increasing concern worldwide. Their removal mechanisms differ significantly and in consequence these two contaminants are chosen for this critical review. The occurrence of fluoride and uranium and their health implications, reported removal by NF/RO along with specific removal mechanisms are investigated in this paper.

2. Worldwide occurrence of fluoride and uranium

It is evident that fluoride contamination is a worldwide issue (Table 1). Amini et al. (2008) provides a global overview of groundwaters with fluoride concentration exceeding the WHO guideline of 1.5 mg L^{-1}. The results show that areas most severely affected include East Africa, Middle East, Argentina, the United States, India, and China.

The occurrence of fluoride in natural waters is closely linked to the local geology. The chemical element fluorine is abundant in the Earth's crust (625 mg kg^{-1}) as a result of volcanic activity and fumarolic gases (Edmunds and Smedley, 2005). Fluorides are naturally released into water by the dissolution of fluoride-containing rocks and soils. The dissolution process is affected by various factors including rock chemistry, groundwater age, residence time, well depth and conditions of the pathways (Kim and Jeong, 2005). Fluoride concentration in water is strongly controlled by the solubility of minerals, especially calcium fluorite (CaF_2) which has the lowest solubility of 15 mg L^{-1} at 18 °C (Kwasnik, 1963). Therefore high fluoride concentrations are associated with minerals with low calcium contents, or high alkaline and carbonous conditions where sodium instead of calcium dominates the water composition (Amini et al., 2008). One typical example arises in the East African Rift Valley. Fluoride concentration in local soda lakes is up to 2800 mg L^{-1} and in groundwater is as high as 330 mg L^{-1} (Smedley et al., 2002; Fawell et al., 2006). Such concentrations are extremely high even compared to other elevated-fluoride areas in the world.

In addition to natural dissolution of minerals, industrial operations, such as metallurgical industries, fertilizer plants, and semiconductor production, generate effluents with high fluoride contents (Ndiaye et al., 2005; Dolar et al., 2011). In the case of phosphate production, fluoride in the effluent can reach up to 3000 mg L^{-1} (Ndiaye et al., 2005).