Abstract No: PS-29

Fabrication of reduced graphene oxide – nano iron (rGO-nZVI) anode for electrocoagulation treatment to regulate excess fluoride in water

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Due to excess fluoride in drinking water, over millions of people are being affected globally with dental and bone problems. The World Health Organization (WHO) guidelines specify 1.5 mg /L fluoride in drinking water as the maximum permissible contaminant limit (MCL). However, in dry climatic zones and tropical countries, the water consumption is high, therefore in a lower fluoride value than WHO MCL may suitable for the dry zone of Sri Lanka. The situation aggravates further in dry zone (Sri Lanka) as the fluoride in groundwater sometimes exceeds 5 mg/L. Presently water treatment methods based on adsorption, co-precipitation, membrane technology and ion exchange are used to mitigate enriched fluoride water with limited success. Briefly, both adsorption and co-precipitation generate excess sludge. The reverse osmosis methods remove ions in treated water than required. Most of the electrocoagulation (EC) methods used aluminum scarifying anode, which may pause additional threat of leaching neuro-toxic free Al^{3+} into the treated water stream. Therefore, we developed a novel anode composites using nano Fe and reduced graphene oxide (rGO) to be used in EC cells to regulate excess fluoride. rGO was synthesized by the modified Hummers method at ambient temperature. Natural green tea leaves polyphenols were used to reduce Fe^{2+}/ Fe^{3+} into metallic Fe. The widely used $NaBH_4$ method was also used to produce metallic Fe. The resulted composites are designated as polyphenol derived reduced graphene oxide-nano Fe (rGO–nZVI-P), and $NaBH_4$ derived reduced graphene oxide-nano Fe (rGO–nZVI-B). Before their application, conventional and spectroscopic methods extensively characterized rGO-nZVI composites. 2D Raman bands appeared at 2715 cm^{-1} for rGO-nZVI-P confirm the presence of multilayer structure while the bands at 2680 cm^{-1} for rGO-nZVI-B confirm the presence of the single-layer structure of graphene. XRD diffraction peaks confirm the presence of BCC, α-phase Fe (0) in the core of both nZVI particles. Our results suggest that 1 mg/L fluoride in water can be removed within 1 hr. to 75 µg/L and 3 µg/L respectively using rGO-nZVI-P and rGO-nZVI-B composites with minimal waste generation. Following pseudo-second-order kinetics, at pH 5.6, both composite materials adsorbed over 90 % of 1 mg/L fluoride within 1 hr. The rGO-nZVI-P based EC has the potential to treat fluoride-rich waters efficiently.

Keywords: Fluoride, Groundwater, Reduced graphene oxide, World Health Organization

Acknowledgment
This work was supported by National Research Council under the research grant NRC-TO-16-015