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OZONATION, BIOFILTRATION AND THE ROLE OF MEMBRANE SURFACE CHARGE AND HYDROPHOBICITY IN REMOVAL AND DESTRUCTION OF ALGAL TOXINS AT BASIC PH VALUES

Joyner Eke*, Priyesh Wagh and Isabel C. Escobar
University of Kentucky
177 F. Paul Anderson Tower
Lexington, KY 40506
*Presenting Author: Joyner.Eke@uky.edu

This study was directed at the investigation of technologies for treatment of water containing algal toxins at basic pH values. Ozonation, biofiltration and membrane filtration were examined for the removal of algal toxins, specifically microcystin-LR (MC-LR). Results indicated that, as expected, ozonation completely destroyed MC-LR in water, while biofiltration using naturally-occurring bacteria did not show a significant reduction in MC-LR concentration even after 8 days of contact time. More compelling were the membrane filtration results, which showed that water affinity interaction was not the only governing factor influencing the removal of MC-LR by membranes. It was found that charge interactions between membranes and MC-LR played an important role in the rejection. MC-LR was completely removed from the feed water only by hydrophobic neutral and positively-charged membranes. Furthermore, due to charge interactions, MC-LR reversibly adsorbed to neutral hydrophobic membranes, but it irreversibly adsorbed to positive hydrophobic membranes.
IRON/PALLADIUM NANOPARTICLES IMMOBILIZED MEMBRANE PLATFORMS FOR CHLORINATED ORGANICS TREATMENT

Hongyi Wan\textsuperscript{a}, Sebastian Hernandez\textsuperscript{a}, Nicolas J. Briot\textsuperscript{a}, Anthony Saad\textsuperscript{b}, Lindell Ormsbee\textsuperscript{b}, and D. Bhattacharyya\textsuperscript{a}

\textsuperscript{a} Department of Chemical and Materials Engineering, University of Kentucky
\textsuperscript{b} Department of Civil Engineering, University of Kentucky
Hongyi Wan: (859) 447-7094
Hongyi Wan: hongyi.wan@uky.edu; Dr. Dibarkar Bhattacharyya: DB@uky.edu

Functionalized polyvinylidene fluoride (PVDF) membrane platforms were developed for environmentally benign in-situ nanostructured Fe/Pd synthesis and remediation of chlorinated contaminates (chloroform, carbon tetrachloride, TCE, PCE and polychlorinated biphenyls (PCBs)). To prevent leaching and aggregation, nanoparticle catalysts were integrated into membrane domains functionalized with poly (acrylic acid). Furthermore, nanoparticles of 24±6 nm were observed inside the membrane pores. The quantification of nanoparticles properties (size, distribution and composition) versus depth underneath the membrane surface was also investigated by cross-sectioning the membrane samples with focused ion beam (FIB). The Fe/Pd nanoparticles immobilized membrane showed excellent performance in the TCE and PCBs degradation and also the repeated degradation experiments were conducted to demonstrate the reusability of Fe/Pd immobilized membranes. The membranes have also been used in remediation of real water samples from contaminated sites.

To increase the accuracy of kinetic analysis and modeling development for further optimization of membrane performance, the correlation between nanoparticle properties and depth inside membrane pores was studied by using FIB (figure 1). Particle size was uniform inside membrane pores at different depths (particle size: 24±6 nm) but slightly smaller than those nanoparticles located on the surface (39±9 nm). Besides, the distribution of particles as well as the functionalized membrane domain were analyzed in line-scan mode by using energy dispersive spectroscopy (EDS).

Over 96% degradation of 3,3',4,4',5-pentachlorobiphenyl was achieved at a residence time of 14.7 seconds in the membrane pores. The surface area normalized reaction rate (ksa) was calculated (using LFR model) to be 0.171 L/(m\textsuperscript{2}h) in convective flow mode, which is 2.5 times the rate obtained in batch mode. The effects of temperature, pH values, as well as the morphology of Fe/Pd nanoparticles (figure 2) were also investigated. In addition to, our technology has been applied in treatment of real water samples containing chlorinated organics from a Superfund site. Dechlorination of the pollutants was achieved with our method and membrane scale up is in progress.
Figure 1. Fe/Pd nanoparticle size and distribution inside the membrane pores. Membrane thickness: 100 µm, fluorine is used as comparison because of the PVDF membrane domain.

Figure 2. Dechlorination performance and H₂ production of Fe/Pd nanoparticles (different morphology) in water phase. Biphenyl is the product of PCB dechlorination. Besides Pd coated on Fe, isolate Pd nanoparticle and Fe nanoparticle were well mixed and tested as a comparison.

For membrane regeneration and reuse, both NaBH₄ and H₂ method were investigated. In XRD analysis, the Fe/Pd particle samples (which were deliberately oxidized and then reduced) exhibited the same crystalline patterns as the original samples. The membrane was tested for reactivity after four degradation cycles with regeneration between each cycle. The increase of surface particle size of 22% resulted in a decrease of 9.7% PCB conversion for the 4 hr reaction time.

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Organic solvents derived from petroleum sources, such as N-methyl-2-pyrrolidone (NMP) and dimethylacetamide (DMAc), have been traditionally used to fabricate polymeric membranes. These solvents have a negative impact on the environment and human health since most of them are volatile and hazardous; therefore, using renewable solvents derived from biomass would be of great interest in order to make membrane fabrication sustainable. In this study, methyl-5-(dimethylamino)-2-methyl-5-oxopentanoate (Rhodiasolv® PolarClean) was used because it is a bio-derived, biodegradable, nonflammable and nonvolatile solvent. Polysulfone was chosen as the polymer to fabricate membranes due to its thermal stability, strong mechanical strength, good chemical resistance and antifouling properties. The thermodynamics aspects of the polysulfone/PolarClean/water system were investigated. From cloud point curves and theoretical predictions, PolarClean showed the potential to be a solvent for polysulfone as compared to traditional petroleum-derived solvents. Polysulfone membranes prepared with PolarClean were also investigated in terms of their morphology, porosity, water permeability and protein rejection, and subsequently, compared to membranes prepared with petroleum-derived solvents. Overall, PolarClean showed a potential to replace petroleum-derived solvents in fabrication of polymeric membranes.
SELENIUM REMOVAL USING ACTIVATED ALUMINA IN A PACKED-BED REACTOR

Yuxia Ji
328 Raymond Building, Department of Civil Engineering, University of Kentucky
(859)699-6545
yuxia.ji@uky.edu

Yi-tin Wang
161 Raymond Building, Department of Civil Engineering, University of Kentucky
(859)257-5937
ywang@uky.edu

Selenium is a typical aqueous contaminant in United States as limited to 0.05 mg/L by EPA. Two dominant inorganic oxyanions, selenate (Se(VI)) and selenite (Se(IV)), can be easily found in water due to their high solubility. Selenium released from manufacture industries of glass, pigments, fossil fuel, emulsion and metal alloys is toxic to human beings as well as aquatic lives and damages the ecological environment. Treatment technologies to remove selenium including microbial reduction and adsorption using activated alumina have been studied in recent years. Both treatment processes were reported able to remove selenium effectively but with deficiencies. In the microbial selenium reduction process, Se(IV) reduction was relatively slow and required a long period to achieve complete reduction compared with Se(VI) reduction. In the process of adsorption onto activated alumina, the adsorption capacity of Se(VI) was significantly lower than Se(IV) and can be interfered by kinds of anions including bicarbonate, sulfate, phosphate, etc. Thus, in order to enhance the removal efficiency, a combined process of both microbial reduction and adsorption was designed to remove selenium complimentarily.

In this study, a packed-bed reactor packed with 40 g activated alumina and cultured with the bacterial strain *Shigella fergusonii* was applied at feeding Se(VI) concentrations of 10 and 50 mg/L, and an HRT of 3.4 days was performed. The result shows that selenium can be removed complimentarily by both adsorption with activated alumina and microbial selenium reduction leading to the improvement of removal efficiency. Approximately 4.3 mg/L and 19.7 mg/L of Se(VI) was detected in the effluents at feeding concentrations of 10 and 50 mg/L at the end of the experiments, respectively, indicating that Se(VI) has been significantly removed compared with 9.95 mg/L and 47.9 mg/L using single adsorption process. Insignificant Se(IV) accumulation was observed as Se(IV) generated from Se(VI) reduction was further absorbed by activated alumina.