Chemical Engineering Doctoral Defense

Advancing the Technology Readiness of Membrane Catalyst-film Reactors for Nitrate Removal

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Abstract

Due to the use of nitrogen-based fertilizers, concentrations of oxidized nitrogen species, particularly nitrate, have increased in groundwater and surface waters globally in the last century. This presents a drinking water treatment challenge because human consumption of nitrate in water can cause blue baby syndrome and there is increasing correlative evidence between nitrate ingestion and certain cancers. Water treatment plants are currently able to mitigate nitrate contamination in drinking water by separating or diluting the nitrate in water. However, with increasing concentrations of nitrate in water and waste streams globally, there is a growing need to focus on nitrate destruction rather than separation. A technology capable of transforming nitrate can meet this need while also providing a nitrate treatment option to rural communities that are currently without water treatment technology to remove nitrate.

This dissertation focuses on catalytic hydrogenation of nitrate, an emerging technology capable of reducing nitrate to nitrogen gas using hydrogen gas (H2) as the reductant and a catalytic surface as the site of reaction. This technology is an improvement over technologies at water treatment plants, because the nitrate is transformed with harmless hydroxide, dinitrogen, and water molecules as the byproducts. Prior studies have found catalytic hydrogenation to be capable of selectively reducing nitrate to nitrogen gas at a 95% removal rate. However, there are significant barriers to overcome in order to scale-up and commercialize this technology outside of a laboratory setting.

The research presented in this dissertation centers on identifying and resolving barriers to scale-up in order to improve the commercialization potential of catalytic hydrogenation of nitrate technology. One of the limitations of past nitrate hydrogenation research is uncontrolled H2 gas delivery. This limitation is resolved by a scalable reactor setup with controlled hydrogen delivery, the Membrane Catalyst-film Reactor (MCfR), and is the configuration used for all experiments presented in this dissertation. In the first stage of research, a method was identified to attach the nitrate hydrogenation catalyst to a surface to enable catalyst reusability. The second stage of research focused on optimizing bimetallic nano catalyst structures on a membrane to maximize nitrate removal in the MCfR. The final stage of research involved long-term nitrate removal experiments in the H2-MCfR to simultaneously resolve operational issues of continuous operation and identify sources of catalyst deactivation. Completing these objectives resulted in increased technology readiness of MCfRs for commercialization and scale-up, which is supported by the background, research outcomes, and conclusions presented herein.

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