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Combination of cupric ion with hydroxylamine and hydrogen peroxide for the control of bacterial biofilms on RO membranes

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Combinations of Cu(II) with hydroxylamine (HA) and hydrogen peroxide (H_2O_2) (i.e., Cu(II)/HA, Cu(II)/H_2O_2, and Cu(II)/HA/H_2O_2 systems) were investigated for the control of *P. aeruginosa* biofilms on reverse osmosis (RO) membranes. These Cu(II)-based disinfection systems effectively inactivated *P. aeruginosa* cells, exhibiting different behaviors depending on the state of bacterial cells (planktonic or biofilm) and the condition of biofilm growth and treatment (normal or pressurized condition). The Cu(II)/HA and Cu(II)/HA/H_2O_2 systems were the most effective reagents for the inactivation of planktonic cells. However, these systems were not effective in inactivating cells in biofilms on the RO membranes possibly due to the interactions of Cu(I) with extracellular polymeric substances (EPS), where biofilms were grown and treated in center for disease control (CDC) reactors. Different from the results using CDC reactors, in a pressurized cross-flow RO filtration unit, the Cu(II)/HA/H_2O_2 treatment significantly inactivated biofilm cells formed on the RO membranes, successfully recovering the permeate flux reduced by the biofouling. The pretreatment of feed solutions by Cu(II)/HA and Cu(II)/HA/H_2O_2 systems (applied before the biofilm formation) effectively mitigated the permeate flux decline by preventing the biofilm growth on the RO membranes.

1. Introduction

The matrix of extracellular polymeric substances (EPS) enclosing cells presents a difficult hurdle in the inactivation of biofilms. It has been reported that cells in biofilms are up to 1000-fold more resistant to disinfectants than planktonic cells (Kim et al., 2008; Masak et al., 2014). Biofilms are ubiquitous and problematic in many industrial fields, including water distribution systems, heat exchangers, ship hulls, and membrane filters (Flemming, 2002). In particular, the formation of biofilms on reverse osmosis (RO) membranes (biofouling) in desalination plants significantly lowers the performance of these membranes and is recognized as the largest obstacle in the RO process (Mansouri et al., 2010; Matin et al., 2011).

To alleviate the biofouling of the RO membranes, chlorine is frequently added in the pretreatment step and then quenched immediately preceding the RO process. Unfortunately, improper quenching of chlorine can cause serious damage to polyamide active layers of RO membranes (Kwon and Leckie, 2006). Furthermore, the potential health risks associated with transport and storage of chlorine, as well as the formation of toxic byproducts, are cause for concern. Alternative disinfectants such as ozone, chlorine dioxide, and chloramines have similar drawbacks (Kim et al., 2009; Bridier et al., 2011), and the quenching of these disinfectants to prevent membrane damage can lead to the regrowth of bacteria in the RO process. Several biological approaches for the control of biofouling have been investigated (e.g., quorum sensing inhibition, energy uncoupling, and enzyme treatments) (Yeon et al., 2009; Kappachery et al., 2010; Xiong and Liu, 2010; Kim et al., 2011; Xu and Liu, 2011; Yu et al., 2012), but these methods are generally not overly effective and are only applicable on a narrow spectrum of microorganism strains.

Copper has been recognized as a useful disinfectant and antimicrobial agent for inactivation of various microorganisms (Borkow and Gabbay, 2004; Dwidjosiswojo et al., 2011). Several studies have demonstrated the successful inactivation of biofilms by using copper, although the inactivation efficiency for the biofilms was not as high as for planktonic cells (Teitzel and Parsek, 2003; Harrison

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