DOCTORAL THESIS

Catalytic advanced oxidation processes for degradation of environmental emerging contaminants

Law, Cheuk Fung Japhet

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Abstract

In recent years, the increasing release of trace organic chemicals to the aquatic environment have been problematic to both the ecosystem and the human society. These trace organic chemicals, regarded as emerging contaminants, include different categories of chemicals, which are either deemed to be safe for human consumption or they are naturally occurring compounds. As a newly recognized class of emerging contaminant, artificial sweeteners are proven to be one of the most ubiquitous classes of emerging contaminants in environmental waters. Its transformation to different suite of TPs during water treatment processes generated more toxic influence than the parent compound is problematic.

The realization of the widespread of emerging contaminants, together with their ambiguous fate and impact to the environment have led to the development of advanced oxidation processes that can effectively attenuate this wide range of contaminants. In this work, several catalytic advanced oxidation processes were studied. On one hand, it aimed to evaluate their effectiveness on the removal of the artificial sweetener – acesulfame; and on the other hand, to shed lights on the future development of catalytic advanced oxidation processes.

In the first part of this thesis, the photo-Fenton treatment was evaluated on its potential to effectively remove acesulfame together with the produced transformation products, and the post-treatment toxicity screening. The photo-Fenton treatment was found to be effective in removing both the parent compound and the transformation

products, without leading to an increase in toxicity, which is largely related to the effective removal of the transformation products.

In attempt to reduce the reliance on UV irradiation, newly synthesized carbon and nitrogen co-doped TiO_2-based photocatalyst was applied to capture the simulated sunlight for the degradation of acesulfame. The heterogenous photocatalytic treatment was found to involve several different oxidative reactive species for both degradation and transformation by using several scavengers to alter the degradation profile. Unexpected transformation product was also formed upon treatment in actual water matrix, suggesting the impact of water constituents to the transformation of emerging contaminants. Toxicity results indicated the inability to achieve detoxification, suggesting that a more effective degradation process was needed.

To accelerate the degradation process, and enhance the performance at neutral pH, the use of redox mediators for Fenton/Fenton-like system was evaluated. Developed novel Fenton-like system involving copper(II) as transition metal ion, persulfate as oxidant and mercaptosuccinic acid as redox mediator led to effective removal of different contaminants. Elucidation of the proposed oxidation mechanism suggested the role of each components of the system, and the generation of different reactive species for degradation as indicated by the different acesulfame transformation profile obtained. The implementation of redox mediators to Fenton/Fenton-like system was beneficial and an effective approach.

In short, this work presents several kinds of catalytic advanced oxidation process and shed lights on improving the degradation performance with directions for the future development of better and more effective water treatment processes.

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