

2024 年 02 月 20 日

YYYY/MM/DD

独立行政法人日本学術振興会理事長 殿

To: President, Japan Society for the Promotion of Science

研究活動報告書

Research Report

1. 受入研究者/ Host researcher

受入研究機関・部局・職
Name of Host Institution, Department and Title

Kyoto University, Graduate School of Medicine, Associate Professor

受入研究者氏名
Host Researcher's Name

HARADA Koji

2. 外国人招へい研究者/ Fellow

所属研究機関・部局・職
Name of Institution, Department and Title

University of Southern Queensland, Faculty of Health, Engineering and Sciences, Senior Lecturer

外国人招へい研究者氏名
Fellow's Name

TRZCINSKI, Antoine P.

3. 採用期間/ Fellowship Period

2023 年 11 月 13 日

～

2024 年 02 月 09 日

4. 研究課題/ Research Theme

Removal of Organophosphate Flame Retardants by Combined Adsorption and Electrochemical Regeneration

5. 研究活動報告/ Research Report

(1) 研究活動の概要・成果/ Summary of Research Results

Due to their widespread use in furniture, textiles, electronics and other processing chemicals, organophosphate flame retardants (OPFRs) have been frequently detected in various environmental matrices, such as wastewater, receiving water, soil and air. The concentration of OPFRs in wastewater treatment plant (WWTP) influent, effluent and in surface and drinking water has been reported to be as high as a few tens of micrograms per liter. It has been found that some OPFRs can partially be degraded in WWTPs or adsorbed onto sludge, but they are also found in the effluent. Drinking water is a major exposure pathway for OPFRs to humans, but little research has been done to remove these chemicals from drinking water. Due to their toxicity and persistence in the environment, it is important to investigate new processes to remove these chemicals from water.

The objective was to test the removal efficiency of the organophosphate flame retardant in water using an electrochemical oxidation process. The process is based on two fundamental elements; adsorption of contaminants onto a patented graphite intercalation compound (referred to as “graphite flakes”), and electrochemical regeneration of graphite flakes both taking place within a single unit.

Eleven different OrganoPhosphate Flame Retardants (OPFR) were tested in this research: 1) Tributyl phosphate

(TBP), 2) Tris(2-chloroethyl)phosphate (TCEP), 3) Tris(1-chloro-2-propyl) phosphate (TCPP), 4) Tris(1,3-dichloro-2-propyl) phosphate (TDCPP), 5) Tris(2-butoxyethyl) phosphate (TBEP), 6) Triphenyl phosphate (TPhP), 7) 2-Ethylhexyl diphenyl phosphate (EHDPHP), 8) Tris(2-ethylhexyl) phosphate (TEHP), 9) Cresyl diphenyl phosphate, 10) Tricresyl phosphate (TCrP), 11) Tris(4-tert-butylphenyl) phosphate.

Firstly, adsorption experiments were conducted to determine the extent and rate of adsorption of these pollutants onto graphite. Then several experiments were conducted to regenerate the graphite adsorbent using electrochemical oxidation. Several parameters were tested to optimize the removal percentage.

Using 5 minutes adsorption using compressed air followed by regeneration at 3 amperes for 10 minutes, the following maximum removal percentages were obtained in the process:

Tributyl phosphate (TBP): 69%
 Tris(2-chloroethyl)phosphate (TCEP): 71.4%
 Tris(1-chloro-2-propyl) phosphate (TCPP): 67.5%
 Tris(1,3-dichloro-2-propyl) phosphate (TDCPP): 81.7%
 Tris(2-butoxyethyl) phosphate (TBEP): 80.5%
 Triphenyl phosphate (TPhP): 95.8%
 2-Ethylhexyl diphenyl phosphate (EHDPHP): 99.6%
 Tris(2-ethylhexyl) phosphate (TEHP): 100%
 Cresyl diphenyl phosphate: 98.7%
 Tricresyl phosphate (TCrP): 99.9%
 Tris(4-tert-butylphenyl) phosphate: 100%

The removal was very high with chemicals with a phenolic group, whereas organophosphate flame retardants with a chlorine atom were more difficult to adsorb on graphite and break down. This process is considered to have low operation and maintenance costs, and reduce energy consumption and waste generation in large scale applications.

(2) 主な研究発表 (雑誌論文、学会、集会、知的財産権等) / Main Research Publications

The following manuscripts will be prepared:

- 1) Removal of chlorinated organophosphate flame retardants in a combined adsorption and Electrochemical oxidation process
- 2) Removal of alkyl organophosphate flame retardants in a combined adsorption and Electrochemical oxidation process
- 3) Removal of aryl organophosphate flame retardants in a combined adsorption and Electrochemical oxidation process
- 4) Comparison of adsorption and electrochemical oxidation of chlorinated, alkyl and aryl organophosphate flame retardants

(3) その他/ Remarks

The research fellow would like to thank JSPS for supporting this research.