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論　文　の　要　旨

Abstract of thesis

Decabromodiphenyl ether (BDE-209), as one of the famous poly-brominated diphenyl ethers (PBDEs), possesses excellent physical and chemical properties due to its full-brominated structure. Therefore, BDE-209 is regardedas necessary anthropogenic chemicals, which has been widely used in the textile and plastics industries to improve the fire resistance ability of the corresponding products. Considering the contact frequency and potential toxicity of nerve inhibition, endocrine disruption, and carcinogenicity, it is of significance to degrade BDE-209. Taking advantage of the strong degradation ability in anaerobic conditions, manyresearchers reported that BDE-209 could be degraded through reductive debromination by anaerobic microorganisms under thermophilic and mesophilic conditions. However, the difference between specific degradation characteristics underboth conditions is not examined. Therefore, this dissertation focused on the effects of temperature on the distribution, degradation rate, and pathway of PBDEs during anaerobic digestion. The dissertation is divided into 5 parts.

In chapter 1, the author introduced the currently existing problems concerning PBDE contamination and compared several treatment methods for solving the problems. Specifically, in this chapter a literature review on the currentsituation, toxicity, pollution, degradation of PBDE was conducted. And the objectives of thisdissertation were identified.

In chapter 2, the author investigated the partition coefficient (log K_{DOC}) for PBDE in dissolved organic carbon solution to understandthe pollutant dispersing behavior in unnatural water systemsuch as organic waste stream/anaerobic digestate, which has never been reported before. Four typically slow-degrading PBDEs and four different filtrates from anaerobic digestion sludges including samples from two laboratory-scale and two full-scale anaerobic digesters were used for experiment. The results showed that for the laboratory samples, log K_{DOC}sfor BDE-153 was6.38 in thermophilic conditionand5.46in mesophilic condition, suggesting that a thermophilic environment might promote the solubility of PBDEs to a greater extent than mesophilic

conditions. On the other hand, the log K_{DOC}s for BDE-153at thermophilic condition for two full-scale samples were 6.62 and 6.45,suggesting the composition of dissolved organic carbon directly influenced the PBDE solubility.A linear regression between log K_{DOC} and DOC composition was established with a R^2> 0.98, which indicatedthat the specific category of organic carbon protein, polysaccharide, and lipids, and PBDE molecular structure had a direct impact on the value of log K_{DOC}. It is suggested that the log K_{DOC} for PBDE in anaerobic sludge DOC solution, and the equation from regression results couldbe used to simulate the log K_{DOC} for PBDEs in the similar unnatural water system.

In chapter 3, the author researchedthe degradation rate of PBDEs under both thermophilic and mesophilic conditions. Because BDE-209 was regarded as the most representative PBDE, a 200-day activity degradation experiment was carried out by using a commodity curtain containing BDE-209 as the substrate.The experimental phenomenon revealed that high temperature could enhance the degradation rate of PBDEs, attributable to the high reaction activity. Andthe BDE-209 degradation efficiency was less than 5% with an average mass reaction rate of 0.8 $\mu g \cdot d^{-1}$. Theseresults also showed that with the increase of the initial BDE-209 dose, the degradation rate of BDE-209 increasedsharply under the optimal pH of 7.

In chapter 4, the author concentratedon the anaerobic degradationrate and kinetics of BDE-209 in two continuously stirred tank reactors (CSTRs) under thermophilic and mesophilic conditions for 210 days. The results indicated that the degradation rate of BDE-209 was higher under thermophilic condition than that under mesophilic one, the maximum degradation rate was 1.1 $\mu g \cdot d^{-1}$. Meanwhile, the degradation of BDE-209 was associated with the replacement of bromines from PBDE with hydrogen atom, and the formation of nona-, octa- and hepta- BDEthrough multiple-step reaction gradually. It was found that the decrease of BDE-209 was following the first-order kinetic model. The above findings could provide an assistance for application of BDE-209 degradation in anaerobic digestion-based techniques.

In chapter 5, the author summarized the major conclusions for the whole study. It was clarified that compared with mesophilic condition, thermophilic one accelerated the solubilization effect and degradation rate in the anaerobic digestion. And the future researchprospective for applying the results from this study was proposed.

This study offered an opportunity to realize distribution, degradation rate, and pathway of PBDEs under anaerobic conditions. These findings in this research could raise the prospects of the application of anaerobic digestion-based techniques for bromide retardant degradation.

審 査 の 要 旨

Abstract of assessment result

In this dissertation, the distribution, degradation rate, and pathway of PBDEs under anaerobic conditions were investigated.The author summarized that the specific category of organic carbon and PBDE molecular structure has a direct impact on the value of log K_{DOC}.The partition coefficients for PBDE were calculated and a linear regression between log K_{DOC} and DOC composition was established, which could be used to simulate the log K_{DOC} in similar unnatural water system. Furthermore, the 200-day activity experiment results showed that BDE-209 degradation rate underthe thermophilic conditionwas significantly higher than that underthe mesophilic one, from which the optimal degradation conditions for BDE-209 debromination were obtained.The CSTR experiment resultsshowed that through multiple-step reaction, BDE-209 was degraded intonona-, octa- and hepta-BDEs by debromination process following the first-order kinetics, and the maximum degradation rate reached 1.1 $\mu g \cdot d^{-1}$.These findings in this research could raise the prospects of the application of anaerobic digestion-based technology for bromide retardant degradation.This study also offers an opportunity to realize distribution, degradation rate, and pathway of PBDEs under anaerobic conditions.

The final examination committee conducted a meeting as a final examination on 14thJanuary2020. The applicant provided an overview of dissertation, addressed questions and comments raised during Q&A session. All of the committee members reached a final decision that the applicant has passed the final examination.

Therefore, the final examination committee approved that the applicant is qualified to be awarded the degree of Doctor of Philosophy in Environmental Studies.