（様式 3）

学 位 論 文 要 旨

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論 文 名

Removal of nonylphenolic compounds from water and sewage sludge using ferrate

学位論文要旨

Nonylphenolic compounds such as nonylphenol (NP) and nonylphenol monoethoxylate (NP1EO) are partially degraded in conventional wastewater treatment processes and are persistent in sludge over a long period. These compounds have toxic and estrogenic effects on humans and animals. However, sewage sludge can be reused as a raw material for fertilizers. Consequently, the removal of these compounds is necessary before it is recycled and/or discharged into the environment. In this study, potassium ferrate (K_2FeO_4, Fe(VI)) was used as an environment-friendly oxidizing agent to mediate NP and NP1EO removal. K_2FeO_4 can be prepared by wet chemical or electrochemical methods.

In Chapter 2, the K_2FeO_4 preparation by the modified wet chemical method increased the purity of K_2FeO_4 and the recovery of total iron from 30% to 70% and from 50% to 80%, respectively. On the other hand, the purity of K_2FeO_4 was around 30% by the electrochemical method.

In Chapter 3, the removal of NP and NP1EO from water and sewage sludge using K_2FeO_4 was investigated. The application of K_2FeO_4 produced by the wet chemical method (Purity 30%) has good potential to remove NP and NP1EO from water and sludge samples. The NP removal efficiency was enhanced by increasing the mass ratio to 5:1 (Fe(VI):NP) for water samples. Conversely, increasing of K_2FeO_4 dosage did not always improve the removal efficiency of NP from sludge samples because K_2FeO_4 probably reacted with organic matter eluted from sludge solid by alkalization. The removal efficiencies of NP and NP1EO varied with initial and final pH values, which depended on the K_2FeO_4

dosage. The optimum pH range differed between NP and NP1EO for water samples. The optimum initial pH values for the removal of NP and NP1EO were acidic condition (pH 3-4) in water samples. The final pH should be between 3 and 6 for the optimal NP1EO removal from water and sludge samples. Therefore, acidifying samples before adding K_2FeO_4 precipitate is necessary. Additionally, NP and NP1EO removals were found to be due to degradation by Fe(VI).

In Chapter 4, characterizations of degradation products from NP with Fe(VI) were investigated. The biodegradability of NP and its degradation products was evaluated using NP labeled with ^{14}C as a tracer. Adding Fe(VI) to the NP solution spiked with the tracer resulted in a slight decrease in the concentration of ^{14}C, demonstrating partial mineralization of NP and formation of degradation products. The estrogenic activity of NP treated with Fe(VI) in water samples was studied using a yeast estrogen screen (YES). The estrogen activity of NP decreased with increasing the mass ratio of Fe(VI) to NP. Furthermore, the degradation products from NP with Fe(VI) were estimated based on mass spectra, which detected a unique peak at a low intensity. It is likely that four hydrogen atoms were added to NP through the degradation with Fe(VI).

In Chapter 5, the bioavailability of phosphorus in sludge treated with Fe(VI) was evaluated using different extraction reagents such as water, 2% formic acid, 2% critic acid and neutral ammonium citrate. These results demonstrated that sludge treated with Fe(VI) improved phosphorus bioavailability and could be utilized as a raw material for organic fertilizer, although the treated sludge needs to be neutralized before its use as fertilizer.

In conclusion, the addition of K_2FeO_4 can enhance nonylphenolic compounds removal efficiency from water and sewage sludge. NP treated with K_2FeO_4 was transformed into more biodegradable and less estrogenically active products. These would help reduce the adverse effects of nonylphenolic compounds in environments. Also, sludge treated with K_2FeO_4 could be beneficially utilized as organic fertilizers.