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学位論文題目	A Study on Separation Media for Sustainable Water Treatment Technologies （持続可能な水処理技術のための分離媒体の研究）
論文審査委員	主査　教授　齋藤　徹 教授　駒井　克昭 教授　南　尚嗣 教授　大津　直史 教授　小西　正朗

学位論文内容の要旨

A novel separation medium was developed for designing simple, highly efficient, and eco-friendly remediation technologies for treating water containing highly bioactive organic pollutants such as pharmaceuticals, personal care products, and pesticides. The separation medium, didodecyldimethylammonium bromide-montmorillonite (DDAB-MT) organoclay, was readily prepared by mixing a dialkylated cationic surfactant, didodecyldimethyl-ammonium bromide (DDAB) and a layered clay mineral, montmorillonite K30 (MT), in the aqueous solution. The possibility of DDAB-MT organoclay for treating water contaminating highly bioactive organic pollutants was studied.

DDAB-MT organoclay was used for the sorption and degradation of an organophosphate pesticide, fenitrothion, in water. The degree of sorption increased by the modification of MT with DDAB, because of the formation of hydrophobic DDAB assemblies for the sorption of hydrophobic fenitrothion. Fenitrothion ([M+1] = 278) sorbed on the organoclay rapidly converted to the degraded product anion ([M] = 262) followed by the further degradation into 3-methyl-4-nitrophenol ([M−1] = 152). The activation energy for the first-order reaction of the primary degradation of fenitrothion (79.9 kJ mol−1) in the organoclay was lower than the value (94.1 kJ mol−1) in water. Organoclay-mediated catalytic activity expressed over a wide pH region (pH 5−9), being advantageous for the application to different wastewaters and environmental waters. Continuous sorption and degradation of fenitrothion in contaminated water was examined by using a laboratory-scale organoclay-packed column. Fenitrothion-free effluent containing its degraded product, 3-methyl-4-nitrophenol, having lower toxicity to aquatic organisms outflowed from the bottom of the column.

The sorption of various antibiotics, pharmaceuticals and personal care products in water onto DDAB-MT organoclay was also studied. The extent of the sorption on the organoclay was largely dependent on the DDAB content; the effect of DDAB modification on the sorption was also influenced by hydrophobicity and/or net charge of pharmaceutical. The binding constants of β-lactam antibiotics were determined from their interaction with the DDAB molecules on the organoclay and were correlated with their aqueous-octanol distribution coefficients. Additionally, it was also influenced by the net charge of the antibiotic. A wide range of β-lactam antibiotics including penicillin- and cephalosporin-type antibiotics were sorbed on the organoclay and rapidly degraded under mild conditions (pH 7, 25ºC). The continuous sorption and degradation of penicillin G in a buffer solution and synthesized hospital wastewater were demonstrated by using an organoclay-packed column. The resulting effluent was free of penicillin, and containing only penicillin degraded products.

In conclusion, DDAB-MT organoclay was useful separation medium for decontaminating water containing bioactive organic pollutants such as pharmaceuticals, personal care products, and pesticides. Because of high water permeability and hydrophobic property, the organoclay sorbed a wide range of organic pollutants and efficiently decontaminated polluted water. Moreover, β-lactam antibiotics and fenitrothion sorbed on the organoclay degraded into less toxic compounds. Continuous sorption and degradation in the contaminated water was successfully demonstrated, suggesting the usefulness of DDAB-MT organoclay as a barrier material for controlling diffusion of bioactive organic pollutants.

論文審査結果の要旨

近年、抗生物質や農薬等の高生理活性物質の水環境汚染防止のための吸着材に関する研究が盛んに行われている。粘土鉱物に界面活性剤を吸着させたオルガノクレーは環境汚染物質の吸着材として有力な候補とされているが、界面活性剤の漏出が多く、実用化の道筋が立っていなかった。

本論文は、自己会合性の極めて高い二本鎖アルキル基を有するカチオン界面活性剤を吸着させたオルガノクレーを新たに調製し、水中抗生物質や農薬の除去を試みたところ、新規オルガノクレーが高い安定性を有するとともに、抗生物質はじめ様々な薬物や農薬の捕集への有効性が確認された。さらに、オルガノクレー中で抗生物質や有機リン系農薬の分解が促進されることを発見し、オルガノクレーが高生理活性物質汚染水の連続処理可能な吸着材としての可能性を見出した。抗生物質や農薬の水環境汚染は広範囲に亘っており、実態把握については数多くの報告があるが、有効な対策技術に欠ける。本論文は研究段階にあったオルガノクレーの高生理活性物質のための実用的吸着材としての道筋をつけたものである。

これを要するに、申請者は、オルガノクレーについて環境浄化媒体としての新知見を得たものであり、水環境汚染対策技術の低環境負荷化に対して学術的に貢献するところ大なるものがある。

よって、申請者は、北見工業大学博士（工学）の学位を授与される資格があるものと認める。