氏　　名	Farook Ahmed
授与した学位	博　士
専攻分野の名称	環境学
学位授与番号	博甲第４６５１号
学位授与の日付	平成２４年　９月２７日
学位授与の要件	環境学研究科　社会基盤環境学専攻 （学位規則第５条第１項該当）
学位論文の題目	Fundamental Study on Adsorption and Interaction of Anionic Surfactant and Volcanic Ash Soil （アニオン界面活性剤の火山灰土への吸着と相互作用に関する基礎的研究）
論文審査委員	教授　赤江　剛夫　　教授　諸泉　利嗣　　准教授　森　也寸志

学位論文内容の要旨

Sodium dodecylbenzene sulfonates (DBS) are widely used anionic surfactants for many domestic (detergent, shampoo, surface cleaners) and industrial purposes. DBSs are used in large quantities and when disposed they behave as one of the most common pollutant found in almost all environmental settings. When DBSs are discharged to the environment they come in contact with the soils and immediately adsorbed by soils and soil sediments. The adsorption of DBS by soil may affect the DBS movement and may also influence on the behavior of the toxic substances. Therefore, knowledge on the adsorption behavior is very important to understand the fate of discharged DBS and to apply in remediation of contaminated soil and groundwater. But very few information is available to understand the overall processes of the DBS adsorption.

In this study, we investigated adsorption characteristics of DBS in a volcanic ash soil (non-allophanic Andisol) which is unique soil in Japan and contain huge amount of organic matter and is only negatively charged. That is why this soil has been chosen for the experiment. Since the surfactant DBS is anionic, the electrostatic interaction between the soil particle and the surfactants is basically repulsive.

The experiments are designed to consist of two parts. The first is systematic experimental study on the effects of pH, DBS concentration, carbon chain structure, time and electrolyte on DBS adsorption. The DBS concentration was measured by using the anionic surfactant selective electrode membrane. In addition, the zeta potential of the DBS adsorbed particles was obtained by measuring the electrophoretic mobility. The adsorption isotherm was examined using the Langmuir-Freundlich-Hill equation. The second part is on the role of the organic matter on DBS adsorption and interaction among the soil, organic matter and surfactants. The DBS adsorption of organic matter free system was compared with that of the control, then influence of concentration and adsorbed amount of the surfactant, electrolytes concentration and pH on the amount of the dissolved humus was analyzed. The amount of the dissolved organic matter was determined by measurement of the spectroscopic absorbance of the soil solution.

The experimental results indicated that adsorption of DBS was strongly influenced by pH, surfactant concentration, electrolyte concentration, carbon chain structure of DBS and humic substances. The adsorption amount become higher at a lower pH and become lower at high pH. When the soil pH is higher, electrostatic repulsion between the soil and the surfactant increases, thus adsorption decreases. The adsorbed amount increases with the increase in DBS concentration and that in the low-concentration range is outstandingly sharp. The sharp increase is accused to cooperative adsorption caused by hydrophobic interaction among the carbon chains of adsorbing surfactants. The increase in electrolyte concentration promoted adsorption because the increase of the electric field around the particle. The increase of the shielding effects was supported by the observed increase in the negative zeta potential. The adsorption of DBS with a linear carbon chain was larger than that with a branched chain DBS due to the larger interaction between the linear carbon chain structures. The second experiment revealed that organic matter was found to enhance the DBS adsorption under all pH condition tested. The dissolution of humic substance from the soil complex increases at lower electrolyte concentration and with the increase of pH. With the increase of surfactant concentration the dissolution of humic substances increases. The results satisfactorily revealed main mechanisms and interactions on DBS adsorption in the volcanic ash soil-organic matter complex.

The obtained results on DBS adsorption and interaction of the soil complex give us a new thought which can be applied in understanding its movement and remediation of the soil and water by the DBS.

論文審査結果の要旨

ドデシルベンゼン硫酸ナトリウム（DBS）は、家庭用および産業用に広く使われている陰イオン性界面活性剤である。廃棄されると、環境汚染物質として振る舞うが、DBSの土壌への吸着に関する研究は少なく、その要因と吸着機構について不明な点が多く残されている。本研究は、多量の有機物を含み、負荷電のみを有する火山灰土中におけるDBSの吸着特性に関連する要因の効果を調べ、吸着機構の解明を試みたものである。実験は、大きく分けて2つの内容から成る。第1は、pH、電解質濃度、DBS濃度、炭素鎖の構造、時間がDBSの吸着に与える影響を系統的に検討する実験である。界面活性剤の測定には、自作の選択性電極膜法を用い、ラングミューア・フロインドリヒ・ヒルの吸着等温式で分析した。併せて、電気泳動法でDBS吸着土粒子のゼータ電位を測定した。実験結果によると、低濃度領域でDBS濃度が増大すると吸着量が急激に増大した。この急激な増大は表面活性剤の炭素鎖間の疎水的相互作用に基づく協同吸着による。吸着量は、低pHで多く、高pHで少なかった。土壌pHが高い時、土粒子の負荷電が増大するため界面活性剤との静電的反発力が増大し、吸着量は減少する。また、吸着量は電解質濃度の増大とともに増大したが、これは対イオン吸着のために、電場の遮へいが生じたことによる。電場の遮へい効果は、ゼータ電位の増大からも裏付けられた。直鎖状DBSの吸着量は分枝状DBSよりも多かったが、これは、両者の構造形態の違いから説明された。第2は、DBSの吸着における有機物の役割と、土壌と有機物及び界面活性剤間の相互作用に関する実験である。まず有機物を除いた試料と処理しない試料のDBS吸着量を比較し、土壌中の腐植物質がDBSの吸着に寄与することを示した。ついで、界面活性剤の濃度と吸着量、電解質濃度、pHが溶解有機物量に与える影響を分析した。土壌複合体からの腐植物質の溶解は、低い電解質濃度でpHの上昇とともに増大し、界面活性剤濃度の増加によっても促進された。実験結果は、吸着の原動力としての疎水性相互作用と静電的反発力の効果から説明できることを明らかにしている。

以上のように、本研究は非火山灰土のDBS吸着と腐植との相互作用の主要な機構を明らかにすることに成功しており、学術的な貢献が認められ、応用面での貢献が期待されるため、博士の学位を授与するに値するものと判定する。