

論文 / 著書情報
Article / Book Information

題目(和文)	水中からの有機汚染物質除去を目的としたカーボンナノチューブの持続可能な活用
Title(English)	Sustainable Application of Magnetic Carbon Nanotubes to Remove Organic Pollutants from Water
著者(和文)	
Author(English)	Mohamed Ateia Ibrahim
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種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

専攻 : Department of	Civil and Environmental Engineering	専攻	申請学位 (専攻分野) : Academic Degree Requested	博士 Doctor of	(Philosophy)
学生氏名 : Student's Name	Mohamed Ateia Ibrahim		指導教員 (主) : Academic Advisor(main)	YOSHIMURA Chihiro	
			指導教員 (副) : Academic Advisor(sub)		

要旨 (和文 2000 字程度)

Thesis Summary (approx.2000 Japanese Characters)

Adsorption is a facile, eco-friendly and low-energy requiring technology that aims to separate diverse compounds using a wide variety of adsorbent materials. Among them, carbon nanotubes (CNTs), which is characterized by large specific surface areas, hydrophobicity, porosity, and rapid sorption kinetics, have been explored as one of the next-generation adsorbents. Over the past decade, extensive studies have investigated applications of CNTs to adsorb several organic and inorganic water contaminants. It has been also proposed that magnetic CNTs (MCNTs) could be used to reduce the cost of environmental remediation as they could be easily separated from a solution using magnets without the need for centrifugal separation or filtration. However, adsorption technologies are nondestructive and the pollutants do not degrade or disappear when transferred from aqueous to solid phase. Thus, regeneration and management of spent adsorbents are of utmost importance for the sustainability of adsorption systems. This study aimed to improve the understanding and the applicability of CNTs for sustainable applications to water treatment.

Current methods to prepare MCNTs include extensive use of treatment with strong acids and result in massive losses of CNTs. Chapter 3 explored the potential of direct use of the magnetic properties associated with the metal (alloy or oxide) acting as catalyst for CNT formation. The as-received CNTs were graded by applying a permanent magnet to a suspension of CNTs to collect the high-magnetic fraction and decanting the low-magnetic fraction. The collected MCNTs were carefully characterized using eight different diffraction and spectroscopic techniques. A key insight was that metallic nano-clusters of Fe and/or Ni located in the interior cavities of the nanotubes give MCNTs their ferromagnetic character. After application of this straightforward method, the MCNTs showed saturation magnetization up to 10 times the value of the as-received materials. The resulting MCNTs showed high adsorption capacities (> 40 mg/g), high magnetic response, and regeneration. This method is simpler, faster, and substantially reduces chemical waste relative to current techniques and the resulting MCNTs are promising adsorbents for organic/chemical contaminants in environmental waters.

CNTs have been found to interact with natural organic matter (NOM), which is a heterogeneous

mixture of organic compounds and omnipresent in aquatic environments, modifying characteristics of both NOM and CNTs. Further, NOM compete with micropollutants (e.g., atrazine) on adsorption sites of CNTs. Yet, our understanding of NOM components' adsorption by carbon nanotubes (CNTs) is limited. In Chapter 4, eleven standard NOMs from various sources were characterized, and their adsorption on four different CNTs were examined side-by-side using total organic carbon, fluorescence, UV-visible spectroscopy, and high performance size exclusion chromatography (HPSEC) analysis. Adsorption was influenced by chemical properties of NOM, including aromaticity, degree of oxidation, carboxylic acidities and hydrophobicity. Fluorescence excitation-emission matrix analysis showed preferential adsorption of decomposed and terrestrial-derived over freshly produced and microbial-derived NOM. HPSEC analysis revealed preferential adsorption of fractions in the molecular weight (MW) range of 0.5–2 kDa of humic acids, while in MW range of 1–3 kDa for all fulvic acids and reverse osmosis isolates. However, the smallest characterized fraction (MW < 0.4 kDa) in all samples did not adsorb on CNTs. The results indicate that FI can be an easy, practical, and sensitive tool to describe and track the changes of NOM during the adsorption process. Moreover, the MW selectivity revealed in the present study will support future investigations of NOM interaction with CNTs and/or effect of NOM on the adsorption of other organic compounds by CNTs.

Finally, Chapter 5 investigated the feasibility of regenerating magnetic carbon nanotubes (MCNTs) using ozone degradation for organic pollutants removal from water. Experimentally, we MCNTs to remove atrazine and run multiple regeneration cycles (i.e., adsorption → collection → ozone regeneration → washing with ethanol). Theoretically, we used density functional theory (DFT) to calculate the free energy of adsorption and the free energy of solvation of atrazine and its byproducts in different solvents (i.e., water and ethanol). The removal capacity of MCNTs capacity decreased from 57.8 to 27.6 mg/g after three ozonation cycles. However, the removal capacity was recovered after washing MCNTs with ethanol and the removal capacity was maintained at 85-93% of its original value after ten consecutive regeneration cycles. Key findings by X-ray photoelectron spectra (XPS) and DFT calculations were that π - π interactions of MCNTs were not affected by ozonation and ethanol could remove degradation byproducts that were attached on the surface. Altogether, the proposed approach is facile and efficient to regenerate the spent MCNTs and to degrade the organic pollutants using ozone.

備考：論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

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