我校在纳米吸附材料高效去除放射性核素研究领域取得新进展

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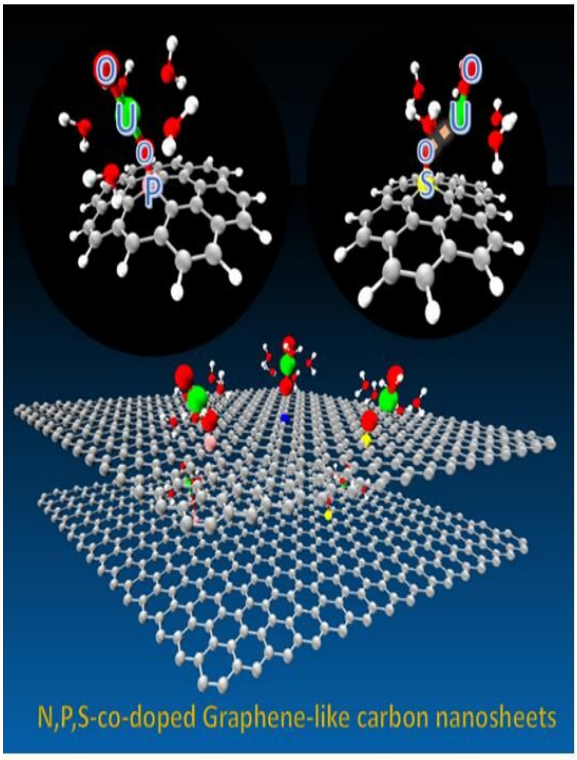
随着世界上民用和军用核技术的发展与广泛使用，部分核废物不可避免的释放到环境中，放射性污染物的治理获得了科研界的广泛关注。其中，铀元素是最常见的放射性污染物，若人类接触到水体中高浓度的放射性铀酰离子，健康将受到极大的损害。同时，将水体中的铀酰离子富集起来回收再利用，也具有很高的经济价值。因此，开发针对水体中放射性核素的吸附材料，具有重大的环境意义和经济意义。

我校环境学院王祥科教授的科研团队最近在国际著名期刊Advanced Science（影响因子12.44）上发表论文《N, P, and S Codoped Graphene-Like Carbon Nanosheets for Ultrafast Uranium (VI) Capture with High Capacity》【Adv. Sci. 2018, 5, 1800235】。此项工作中，作者制备了N, P, S三元素掺杂的类石墨烯二维纳米片层，该材料在铀酰离子的吸附中，表现出了大容量、宽pH适用范围、极快的吸附速度和高稳定性。同时，借助系统的表征和理论计算，作者提出了新颖的化学键吸附机理，为放射性核素治理材料的制备提供了创新思路。论文的第一作者是我校环境学院副教授陈哲老师，通讯作者为环境学院王祥科教授，艾玥洁副教授和中国科学院化学研究所的宋卫国研究员。



FULL PAPER

Radionuclide Capture

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N, P, and S Codoped Graphene-Like Carbon Nanosheets for Ultrafast Uranium (VI) Capture with High Capacity

Zhe Chen, Wanying Chen, Dashuang Jia, Yang Liu, Anrui Zhang, Tao Wen, Jian Liu, Yuejie Ai,* Weiguo Song,* and Xiangke Wang*

The development of functional materials for the highly efficient capture of radionuclides, such as uranium from nuclear waste solutions, is an important and challenging topic. Here, few-layered N, P, and S codoped graphene-like carbon nanosheets (NPS-GLCs) that are fabricated in the 2D confined spacing of silicate RUB-15 and applied as sorbents to remove U(VI)ions from aqueous solutions are presented. The NPS-GLCs exhibit a large capacity, wide pH suitability, an ultrafast removal rate, stability at high ionic strengths, and excellent selectivity for U(VI) as compared to multiple competing metal ions. The 2D ultrathin structure of NPS-GLCs with large spacing of 1 nm not only assures the rapid mass diffusion, but also exposes a sufficient active site for the adsorption. Strong covalent bonds such as P–O–U and S–O–U are generated between the heteroatom (N, P, S) with UO_2^{2+} according to X-ray photoelectron spectroscopy analysis and density functional theory theoretical calculations. This work highlights the interaction mechanism of low oxidation state heteroatoms with UO_2^{2+}, thereby shedding light on the material design of uranium immobilization in the pollution cleanup of radionuclides.

1. Introduction

The annual development of nuclear power plants is intensive and widespread around the world. As a result, the prevention of radionuclide release to the environment and reduction of human exposure to radiation has become increasingly important. Uranium, which may come from uranium mining, nuclear fuel production, and even nuclear accidents, is one of the dominant components of nuclear wastes. Uranium can exist in a variety of valence states, and the uranyl cation (UO_2^{2+}) that dissolves in water is the most common and important species in nature.[1] Therefore, research attention has been focused on the development of functional materials to clean these radionuclides from contaminated water and soil.

Among the different methods for uranium removal from nuclear waste, such as liquid–liquid extraction,[2] ion exchange,[3] adsorption,[4] and reductive precipitation,[5] adsorption is a popular method due to its high efficiency, convenient operation, and low cost. Various kinds of adsorbent material have been developed to efficiently remove and recover radionuclides, including traditional adsorbent such as clay minerals,[6] nanoscale zero-valent iron,[7] and layered double hydroxides (LDHs).[8] However, clays and LDHs often suffer from slow adsorption kinetics, limited selectivity, and low adsorption capacity. At the same time, novel adsorbents are emerging, such as layered metal sulfides,[9] graphene oxides (GO), porous organic polymers,[10] metal–organic frameworks (MOFs),[11] and covalent organic frameworks (COFs). Wang and co-workers reported the amidoxime appended metal–organic framework UiO-66-AO for rapid and efficient extraction of uranium from seawater, which forming strong interaction between amidoxime ligands and uranyl (VI) ions.[12] Ma and co-workers designed the amidoxime-functionalized COFs, and proved it highly efficiency in the U extraction from spiked seawater with capacity of 127 mg g^{-1}.[13] These results broaden the adsorbent material types, supplied insight between highly reticular/topological structure with chelating groups and uranyl (VI) ions.

The adsorption performance of graphene, GO, and their composite materials has also been investigated in the removal of radionuclides.[14] Generally, graphene produced from mechanical exfoliation or chemical vapor deposition exhibits an unsatisfying radionuclides adsorption performance due to the absence of functional groups on the surface. Meanwhile, GO nanosheets, which contain abundant carboxyl (–COOH) groups and hydroxyl (–OH)/epoxy groups (–O–) possess good prospect for the application of radionuclide pollution

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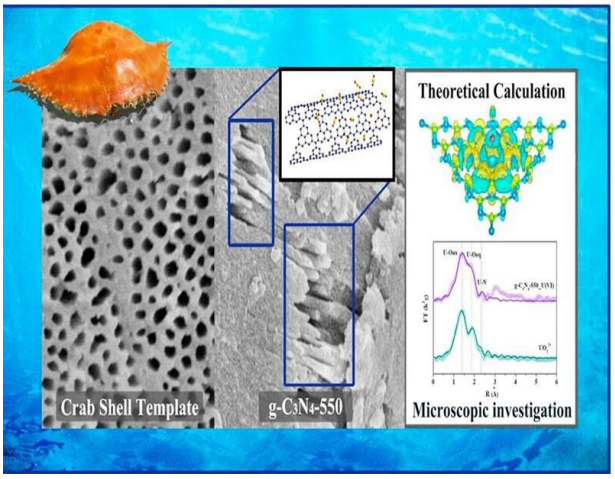
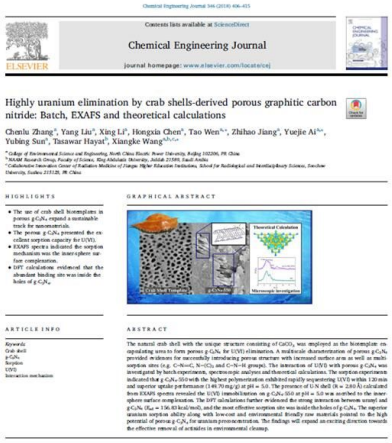
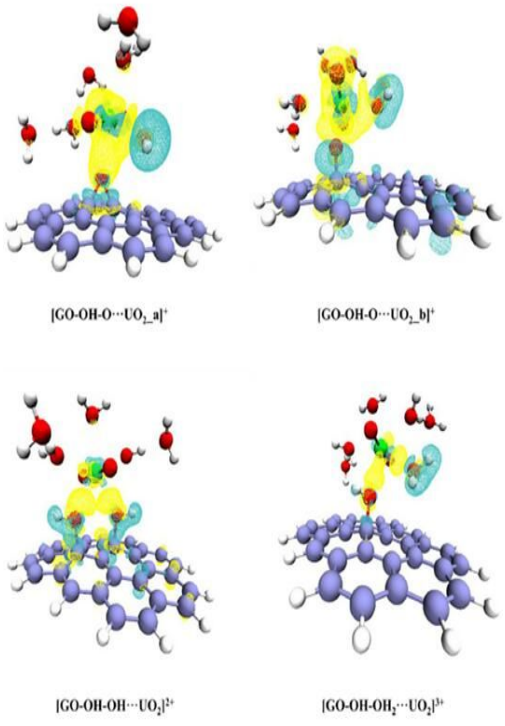
环境学院在放射性核素的纳米吸附材料设计方面做了大量的前沿研究工作。例如，他们利用螃蟹壳为模版，制备了具有规则孔道的石墨化氮化碳结构，材料对于放射性核素铀酰离子表现出了149.70 mg/g的吸附容量，理论计算证明，主要吸附位点在氮化碳的孔道内部。文章《Highly uranium elimination by crab shells-derived porous graphitic carbon nitride: Batch, EXAFS and theoretical calculations》发表在著名SCI一区期刊Chemical Engineering Journal上。【Chemical Engineering Journal 346 (2018) 406－415】。

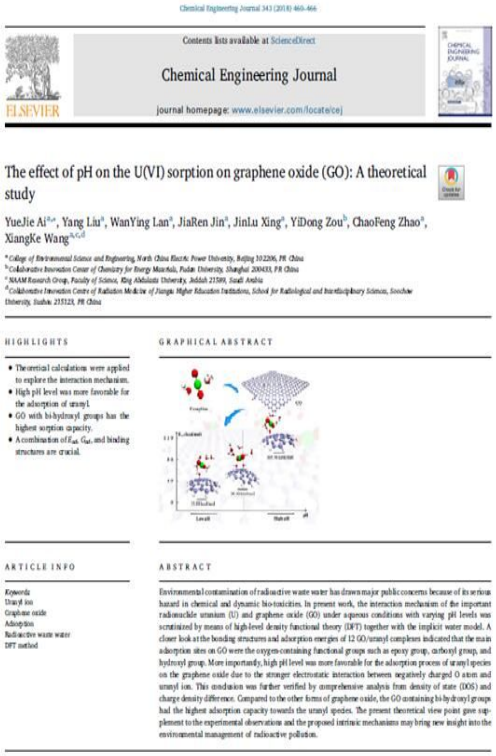



Chemical Engineering Journal

Highly uranium elimination by crab shells-derived porous graphitic carbon nitride: Batch, EXAFS and theoretical calculations

除了材料设计，系统的理论计算模拟也将大大促进材料性能的优化和作用机理的揭示。环境学院科研团队使用密度泛函理论，在石墨烯材料吸附铀酰离子的作用机理方面做了大量的研究。研究揭示了氧化石墨烯对于铀酰离子的吸附位点主要为含氧官能团；并系统研究了pH变化对于作用力改变和强度的影响。研究工作以《The effect of pH on the U(VI) sorption on graphene oxide (GO): A theoretical study》发表在著名期刊Chemical Engineering Journal上。【Chemical Engineering Journal 343 (2018) 460－466】。




Chemical Engineering Journal

The effect of pH on the U(VI) sorption on graphene oxide (GO): A theoretical study

上述研究工作表明了我校环境学院强劲的科研实力，提高了我校在环境领域的科研声望。研究工作得到了国家自然科学基金，科技部重大研究计划和中央高校经费的支持。