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Feasibility of hard acid–base affinity for the pronounced adsorption capacity of manganese(II) using amino-functionalized graphene oxide

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The present study reveals the feasibility of using graphene oxide (GO) functionalized with 3-mercaptopropyl-trimethoxysilane (APTMS) for the removal of Mn(II) from aqueous solution. The APTMS bound on GO's surface was successfully confirmed by FTIR and EDX. For an optimal adsorption study, the effects of pH (pH 1–6), incubation time (5–80 min), temperature (30–60 °C) and initial concentration of Mn(II) (0.1–60 mg L^{-1}) were investigated in association with Mn measurement by AAS. The optimum conditions for Mn(II) removal included a 20 mg L^{-1} initial concentration with 0.02 mg adsorbent at pH 5.5, and complete adsorption equilibrium was reached within 50 min. Their adsorption models are well described by both Langmuir and Freundlich isotherms. The maximum adsorption capacity of GO-NH_2 for Mn(II) was 161.29 mg g^{-1}, about 10 and 4 times higher than those of GP (16.45 mg g^{-1}) and GO (41.67 mg g^{-1}) according to their relevant functional groups. For the GO-NH_2 adsorbent, the adsorption process of Mn(II) follows a pseudo-second-order kinetics model, indicating that the overall rate of Mn(II) uptake is controlled by external mass transfer at the initial stage of the adsorption. Later, the driving forces that control the adsorption rate are attributable to an intra-particle diffusion. A thermodynamic adsorption process reveals mainly an exothermic spontaneous reaction. Therefore, the present study provides an excellent adsorbent for Mn(II) from aqueous solution. This adsorbent can also potentially be used for wastewater treatment.

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Introduction

Manganese (Mn) is an abundant element in the Earth's crust and its presence in drinking water is a result of leaching processes depending on rock types.[1] Although, it is an essential nutrient element in living systems that is found most often in the +2 valence among various oxidation states, elevated levels of Mn content can result in toxic neurological effects.[2] There are many neurotoxic effects that cause a series of symptoms, such as adynamia, reduced coordination, masklike face, gait changes, sleep disturbances, muscular pain, hallucinations, and mental irritability, finally leading to a manganese-induced Parkinson like disease, called manganism.[3] Therefore, it is necessary to remove Mn ion contaminants from wastewater before release into the environment.

Recently, numerous approaches have been studied for the development of effective technologies for pollutant removals.[4–14] Several common wastewater treatments for Mn(II) have been studied, such as solvent extraction,[15] biological aerated filter,[16] precipitation,[17] coagulation/flocculation,[18] flotation,[19] ion-exchange[20] and adsorption.[21] Of these above techniques, adsorption technique provides an attractive alternative one to other removal techniques because it is more economical and readily available.[22] This technique relies on interactions between adsorbent and adsorbate. Thus, a key factor in the adsorption process is selective adsorbent with its intrinsic functional groups. Several adsorbents that have been studied for Mn removal include fly ash,[21] manganese oxide coated zeolite,[22] white rice husk ash,[23] natural and modified zeolites,[24] *Pleurotus ostreatus* nanoparticles,[25] glycine modified chitosan resin[26] and phosphine-functionalized electrospun poly(vinyl alcohol)/silica nanofibers.[27] However, some of these adsorbents are still either low efficiency or low adsorption capacity, thus new adsorbents have been developed to maximize their adsorption property with higher capacity and selectivity.

Graphene oxide (GO) is an alternative choice of the carbon-based adsorbents due to various functional groups (carbonyl, hydroxyl and epoxide) on their surface providing anchor sites for metal ion complexation. Thus, there are many applications of GO for toxic metal adsorption such as Hg(II),[28] U(VI),[29] Pb(II),[30] Cd(II),[31] Cu(II) and Zn(II),[32] Au(III), Pd(II) and Pt(IV).[33] Moreover,

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