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## 論 文 の 要 旨 Abstract of thesis

Heavy metals including chromium (Cr) contamination is challenging the operation and pollutants removal efficiency of wastewater treatment plants (WWTPs). Biosorption is regarded as one of the promising Cr (VI) removal technologies due to its simple operation, low cost and less secondary pollution. As an emerging technology and efficient biomaterial, aerobic granular sludge (AGS) has been attempted for Cr (VI) adsorption from wastewater because of its high concentration of biomass retention and dense structure. However, up to the present, few studies focused on the bioactivity and sustainability of AGS during and after Cr (VI) adsorption. Considering the difficulty in Cr-loaded AGS disposal and a long-time requirement for AGS formation, this study for the first time explored a new operation strategy, i.e. dynamic adsorption/desorption on AGS in order to realize the reuse and recycling of AGS, and Cr removal and recovery from wastewater.

This dissertation is divided into 5 chapters. In Chapter 1, the author introduced the research background and literature review. In this chapter, the author discussed the existing problems of heavy metals pollution in water environment, especially on the toxicity and hazardousness of hexavalent chromium (Cr (VI)), its generation sources and related treatment technologies. Among these technologies, aerobic granular sludge (AGS) has a high potential for biosorption of chromium. At the end of this chapter, the author arrived at the research objectives and framework. In Chapter 2, the author investigated the desorption of Cr (VI) from active AGS, focusing on operation parameters relating to granular bioactivity and stability. Results show that among the five desorbents tested, sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) was the most efficient one for Cr (VI) desorption from Cr-loaded AGS, about  $33.65 \pm 0.76\%$  in 180 min when 1 M  $\text{Na}_2\text{CO}_3$  was applied, which increased with the increase in  $\text{Na}_2\text{CO}_3$  concentration and contact time. In comparison to the pseudo-first-order kinetic model, the desorption process

better fitted the pseudo-second-order kinetic model ( $R^2 > 0.980$ ). Meanwhile, the granular bioactivity indicated by specific oxygen uptake rate (SOUR) was detected to decrease from the initial  $22.38 \pm 1.37$  to  $5.55 \pm 1.42$  mg-O<sub>2</sub>/g-VSS·h when the desorption was processed in 1M Na<sub>2</sub>CO<sub>3</sub> for 60 min, with the granular stability indicated by integrity coefficient increased from 5.52% to 25.63%. The decline of granular bioactivity and stability during the desorption might be associated with the release of cations such as Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> in the AGS, especially from the tightly bound extracellular polymeric substances (TB-EPS). In Chapter 3, the author examined the effect of dynamic adsorption/desorption process on nutrients and Cr (VI) removal by using sequencing batch reactors (SBRs). The dynamic adsorption/desorption showed some decrease effect on cumulative Cr (VI) removal, about 1.87 mg versus 2.11 mg by the control reactor (without dynamic adsorption/desorption) up to day 35. Generally, the control reactor reflected cumulative inhibition effect on nitrification; in contrast, the test reactor demonstrated mitigated inhibition effect of Cr (VI) on NH<sub>4</sub><sup>+</sup>-N removal after operation for 13 days. In addition, the biomass desorption ratio has limited effect on the cumulative Cr (VI) adsorption amount, but greatly impacted the cumulative desorbed Cr (VI) amount with the maximum value obtained in the reactor with 75% biomass desorption ratio. And desorption frequency at 1 time/2d was conducive to the cumulative Cr (VI) adsorption amount. In Chapter 4, the author explored the effect of volumetric organic loading rate (OLR) on AGS performance during the dynamic Cr (VI) adsorption/desorption process. The AGS system operated at a higher OLR (2 kg COD/m<sup>3</sup>·day) obtained obviously higher Cr (VI) adsorption amount (2.15 mg) and desorption amount (0.46 mg) than those at lower OLRs. During the adsorption/desorption, all the indices relating to AGS properties and bioactivities worsened, including SOUR, integrity coefficient, biomass concentration, settling velocity, size and morphology, especially at low OLRs. A higher OLR (2 kg COD/m<sup>3</sup>·day tested in this study) was found to be beneficial for the maintenance of AGS stability during Cr-containing wastewater treatment by using the dynamic adsorption/desorption in AGS system. Finally, in Chapter 6, the author summarized the major conclusions, and proposed the future research directions.

## 審 査 の 要 旨

### Abstract of assessment result

Exposure to Cr (VI) can cause lung cancer, liver damage, reproductive problems and developmental harm issues. Cr (VI) removal and recovery from wastewater may challenge the sustainable operation of current WWTPs with activated sludge as the main treatment process. Aerobic granular sludge (AGS) originated from activated sludge can have high potentials for treating toxic heavy metals-containing wastewater. This study attempted a novel operation strategy on AGS for Cr (VI) removal by introducing a dynamic adsorption/desorption process into the AGS system. In this study, five different desorbent solutions were tested for Cr (VI) desorption from Cr-loaded AGS. The effects of operation parameters on the granular bioactivity and stability of AGS were investigated. Two kinetic models were used to fit the desorption data. In addition, dynamic adsorption/desorption process was introduced into the AGS-based sequencing batch reactor (SBR) system, and its effects were examined on the AGS performance and granular properties under different biomass desorption ratio, desorption frequency and volumetric OLR conditions. Results from this study suggest that dynamic adsorption/desorption process is promising for the stable operation of AGS when dealing with heavy metals-containing wastewater. Future research works are still necessary on the optimization of desorption condition and enhancement approaches for granular bioactivity and stability during desorption in order to realize its practical application in WWTPs.

The final examination committee conducted a meeting as a final examination on 13 July, 2020. The applicant provided an overview of the dissertation, addressed questions and comments raised during Q & A session. All of the committee members reached a final decision that the applicant has passed the final examination.

Therefore, the final examination committee approved that the applicant is qualified to be awarded the degree of Doctor of Philosophy in Environmental Studies.