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学位論文題目 Investigation on the Application of Active Algal-bacterial Granular Sludge for Hexavalent Chromium Removal from Wastewater  
(廃水から六価クロムの除去のための活性藻類-細菌グラニュール汚泥の応用に関する研究)

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## 論 文 の 要 旨 Abstract of thesis

Heavy metals (HMs) are regarded as a great challenge to wastewater treatment plants (WWTPs) due to their toxic effects on aquatic organisms and human beings. As one of the typical HMs, hexavalent chromium (Cr(VI)) has significant carcinogenic and mutagenic effects to humans. Among the various methods for HMs removal, biosorption is considered as a cost-effective and environmentally friendly process. Conventional bacterial aerobic granular sludge (AGS) system has been reported to have potential applications for simultaneous organics, nutrients and HMs removal from wastewater. As a new type of AGS, algal-bacterial AGS is expected to largely reduce operation costs due to much less consumption of carbon source and energy in comparison to the bacterial AGS process. However, little information is available on the application of algal-bacterial AGS for Cr(VI) removal or recovery from wastewater. This research for the first time investigated the feasibility of active algal-bacterial AGS for Cr(VI) removal from synthetic wastewater in comparison to bacterial AGS. Additionally, the influences of external environmental factors and the underlying mechanisms were also explored.

This dissertation is divided into 5 chapters. In Chapter 1, the author introduced the research background and literature review. The issue of Cr(VI) pollution, conventional treatments for Cr(VI)-containing wastewater, current challenges and advantages of biosorption were discussed. Specifically, the author addressed the great potentials of AGS as biosorbent and stated the possible mechanisms, then arrived at the objectives and framework of this research. In Chapter 2, the author compared algal-bacterial AGS with the conventional bacterial AGS. Batch tests revealed that Cr(VI) biosorption onto algal-bacterial AGS was highly pH dependent and the maximum Cr(VI) biosorption

capacity of 51.0 mg g<sup>-1</sup> occurred at pH2. Compared to the bacterial AGS, algal-bacterial AGS demonstrated higher biosorption capacity (9.60 mg/g vs. 8.49 mg/g under the same test conditions;  $p=0.0014 \ll 0.05$ ) and better granular stability. In Chapter 3, the author researched the influences of environmental factors on Cr(VI) removal by algal-bacterial AGS. Results show that microbial activity and granular stability of algal-bacterial AGS were maintained after 6 h biosorption at an initial Cr(VI) concentration of 5 mg/L. The highest Cr(VI) reduction (99.3%) and total Cr removal (89.1%) were achieved within 6 h at pH2 and pH6, respectively. Metal cations enhanced Cr(VI) reduction but suppressed total Cr removal. In addition, the test natural organics promoted Cr(VI) removal, especially on Cr(VI) reduction; while salinity > 5 g/L severely inhibited both Cr(VI) reduction and total Cr removal. In Chapter 4, the author studied the underlying mechanisms for Cr(VI) removal by algal-bacterial AGS. Results show that 85.1% of total Cr was removed by active algal-bacterial AGS after 6 h under pH6 at an initial Cr(VI) concentration of 5 mg/L, which decreased dramatically to 29.6% when sterilized algal-bacterial AGS was used. Glucose promoted Cr(VI) removal by active AGS while no effect was noticed by the sterilized one. Besides, total Cr removal via bacterial AGS was inhibited by 11.2% but maintained in the case of algal-bacterial AGS when antibiotic levofloxacin was added. Analysis of the secretion of proteins and polysaccharides showed that more soluble microbial products (SMP) and loosely bound extracellular polymeric substances (EPS) were secreted by algal-bacterial AGS under Cr(VI) exposure. Moreover, results from Cr distribution and fractionation revealed that around 17.3% of the loaded Cr was EPS bounded and 69.2% was intracellularly accumulated. And 61.7% of the loaded Cr was organic bound fraction with totally around 90.5% of Cr being immobile, indicating the safety of using algal-bacterial AGS for hazardous heavy metals removal. Finally, in Chapter 5, the author summarized the major conclusions, and proposed the future research directions.

## 審 査 の 要 旨

### Abstract of assessment result

This research investigated the feasibility of a new type of AGS, algal-bacterial AGS for Cr(VI) removal in comparison with conventional bacterial AGS. Results show that algal-bacterial AGS possesses superiorities than bacterial AGS and its biosorption capacity is comparable with other reported efficient biosorbents. Besides, the performance of Cr(VI) removal by algal-bacterial AGS could be maintained under the co-existence of various environmental factors. Analysis of mechanisms indicate that cell viability and the existence of algae are crucial for the high efficiency Cr(VI) removal by algal-bacterial AGS. More importantly, the loaded Cr in algal-bacterial AGS is mainly in a less toxic form of Cr(III) with a low mobility, indicating the safety of using algal-bacterial AGS for hazardous heavy metals removal. To achieve more efficient and practical application of algal-bacterial AGS for Cr(VI) remediation, further research is still necessary regarding the treatment of Cr(VI)-containing wastewater using sequencing batch reactors and the re-utilization of Cr-loaded algal-bacterial AGS after Cr(VI) biosorption.

The final examination committee conducted a meeting as a final examination on 15 January, 2021. The applicant provided an overview of the dissertation, addressed questions and comments raised during Q & A session. All 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.