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Effect of nickel on the flocculability, settleability, and dewaterability of activated sludge

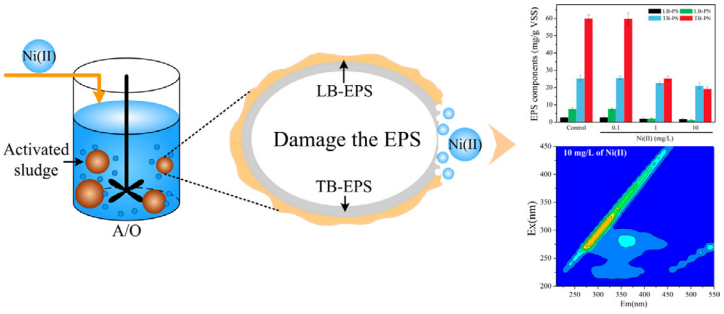
Qi Yang *, Jian Sun, Dongbo Wang, Shana Wang, Fei Chen, Fubing Yao, Hongxue An, Yu Zhong, Ting Xie, Yali Wang, Xiaoming Li, Guangming Zeng

College of Environmental Science and Engineering, Hunan University, Changsha 410082, PR China
Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, PR China

HIGHLIGHTS

- Long-term exposure to Ni(II) exhibited adverse effects on the sludge flocculability.
- The content of PN in EPS decreased after long-term exposure to Ni(II).
- EEM and FTIR showed the change of structures and functional groups in EPS.
- The hydrophobicity of sludge decreased with the increasing Ni(II) level.
- The deleterious effect of Ni(II) could be mitigated by adding the EDTA or citrate.

GRAPHICAL ABSTRACT



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ABSTRACT

Short-term and long-term effects of nickel (Ni) (0.1–10 mg/L) on the physicochemical properties of activated sludge, including the flocculability, settleability, and dewaterability, were investigated. It was found that these properties were unaffected after short-term exposure (1 day) to Ni(II) even at the level of 10 mg/L. After long-term exposure (60 days) to 1 and 10 mg/L of Ni(II), however, the sludge flocculability has seriously deteriorated, while the settleability, and dewaterability became gradually better than the control. The mechanism studies revealed that long-term exposure to Ni(II) resulted in the decrease of protein content in extracellular polymeric substances (EPS) and the damage to EPS structures. Although Ni(II) did not bring any adverse effect on the cell membrane, the relative hydrophobicity of activated sludge was significantly decreased. The negative effects on the flocculability and phosphorus removal performance of activated sludge could be completely eliminated by adding the chelator such as EDTA and citrate.

1. Introduction

Nickel (Ni) has widely been used in many consumer products such as batteries, high quality iron-based alloys, catalysts, and paints due to its unique physical and chemical properties (Gikas, 2007; You et al., 2009). The extensive applications of Ni inevitably cause its release into municipal wastewater treatment plants (WWTPs) (Gikas, 2008; Wang et al., 2013). Previous studies documented that Ni(II) concentration in the municipal wastewaters is greater than 0.1 mg/L in India, Greece, and Turkey, even greater than 0.5 mg/L (Karvelas et al., 2003; Singh et al., 2004; Üstün, 2009). In the municipal WWTPs that receive some specific

* Corresponding author at: College of Environmental Science and Engineering, Hunan University, Changsha 410082, PR China.
E-mail address: yangqi@hnu.edu.cn (Q. Yang).

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industrial wastewaters, this level is even higher. It is reported that it is generally greater than 10 mg/L in the Ni-relevant industrial wastewaters (Akbal and Camci, 2011; Malakootian et al., 2015). Therefore, the potential effect of Ni(II) on WWTPs has attracted increasing attention.

Recently, the effect of Ni on the biological treatment process has concentrated on the removal of organic pollutants and nitrogen (Gikas, 2008; Hu et al., 2002, 2003, 2004; Ong et al., 2004; You et al., 2009). These removal performances are relevant to or dependent upon the physicochemical properties of activated sludge, including the flocculability, settleability, and dewaterability (Ye et al., 2011a,b). Therefore, the variations of these properties are also used to predict the effect of toxic pollutants such as heavy metal ions on activated sludge systems (Chen et al., 2014). Some researchers have investigated the effect of Ni(II) on the settleability and dewaterability of sludge (Li et al., 2011; Ong et al., 2004; Wang et al., 2010). However, these investigated Ni (II) concentrations were usually in the range of 60–240 mg/L, which were much higher than that in real municipal wastewater. Thus, the potential effect of Ni(II) in relatively low concentration should also be studied. Moreover, to date, little is known about the short-term and long-term effect of Ni(II) on the flocculability of activated sludge.

Extracellular polymeric substances (EPS) are thought to be the glue that binds microorganisms cells together to form activated sludge flocs and play a leading role in the physicochemical properties of the flocs (Li and Yang, 2007; Ye et al., 2011b). EPS have a dynamic double-layered structure, including the loosely bound EPS (LB-EPS) and the tightly bound EPS (TB-EPS), and mainly consist of protein (PN), polysaccharide (PS), and small amounts of humic acids, lipids and nucleic acids (Luo et al., 2013; Yang et al., 2010). The structures and compositions of EPS are closely correlated with the flocculability, settleability, and dewaterability of sludge, and can be affected by heavy metal (Liao et al., 2001; Sun et al., 2009). In addition, the physicochemical properties are relevant to the surface properties of activated sludge such as the hydrophobicity, the surface charge, and the integrity of cell membrane (Jin et al., 2003). These surface properties can also be affected by heavy metal cations such as Cu(II) (Chen et al., 2014). Therefore, Ni(II), as a highly soluble heavy metal cation, may change the surface properties and the structures and compositions of EPS, further affecting the physicochemical properties of sludge. For the purpose of giving deep insight into the effect of Ni(II), these possibilities should be clarified by experiments.

When Ni(II) enters the WWTPs, another important job is to explore an effective method to reduce its deleterious effect on activated sludge. Some publications point out that the heavy metal toxicity to activated sludge is strongly correlated with the activities of free metal ions. Their activities can significantly be altered by adding the strong complexing agent (e.g., EDTA) (Hu et al., 2002). Accordingly, the effect of Ni(II) on the physicochemical properties and removal performance may be mitigated by adding the chelator, and yet this assumption needs to be confirmed.

The objectives of this work are therefore to (i) investigate the short-term and long-term effects of Ni(II) ranging from 0.1 to 10 mg/L on the flocculability, settleability, and dewaterability of activated sludge in enhanced biological phosphorus removal (EBPR) system; (ii) explore the details of how Ni(II) affects these properties from the aspects of the compositions and structures of EPS by using excitation-emission matrix (EEM) fluorescence spectroscopy and Fourier-transform infrared (FTIR) spectroscopy, and the surface properties of activated sludge; (iii) find a valid strategy to mitigate the adverse effect of Ni(II).

2. Materials and methods

2.1. Start-up of parent sequencing batch reactor (SBR)

A parent SBR with 80 L working volume was operated under alternate anaerobic-aerobic model to culture the EBPR activated sludge. The seed sludge was collected from the return sludge in Guozhen Wastewater Treatment Plant, Changsha, China. The reactor was operated with three cycles daily at controlled room temperature (21 ± 1 °C). Each cycle (8 h) comprised 20 min of feeding, 2 h of anaerobic and 3 h of aerobic stage, followed by 80 min of settling, 20 min of decanting and 60 min of idle stage. After settling period, 50 L supernatant was discharged from the reactor, and replaced with the same volume of fresh feeding medium (component detailed as below) at the feeding stage. The influent pH value was maintained at 7.5 ± 0.2 by using 2 M HCl or 2 M NaOH. At the anaerobic stage, the SBR was continuously mixed with a mechanical stirrer, and then air was provided to keep the dissolved oxygen (DO) level between 2 and 3 mg/L at the aerobic stage. In order to control the sludge retention time (SRT) at about 10 days, 2.66 L sludge mixture was wasted at the end of aerobic stage per cycle but before settling. After 60 days of cultivation, the relatively stable effluent suspended solids (ESS) and P removal efficiency (>98%) were achieved in the parent SBR.

The feeding medium was prepared daily, and its components were as follows (mg/L): 255.4 CH_3COONa, 49.03 $K_2HPO_4{\cdot}3H_2O$ and 14.62 KH_2PO_4, leading to a theoretical ratio of chemical oxygen demand (COD) to P (≈20 mg COD/mg P), which was thought to be favorable for biological P removal (Wang et al., 2012). The other nutrients in the medium consisted of (per liter): 57.2 mg NH_4Cl, 10 mg $MgSO_4{\cdot}7H_2O$, 5 mg $CaCl_2$, and 0.5 mL of trace metals solution. This trace-element solution comprised (g/L): 0.03 $CuSO_4{\cdot}5H_2O$, 0.06 $Na_2MoO_4{\cdot}2H_2O$, 0.12 $MnCl_2{\cdot}4H_2O$, 0.12 $ZnSO_4{\cdot}7H_2O$, 0.15 $CoCl_2{\cdot}6H_2O$, 0.15 H_3BO_3, 0.18 KI, 1.50 $FeCl_3{\cdot}6H_2O$, and 10 ethylenediamine tetra-acetic acid. Allylthiourea (10 mg/L) was provided in this medium to inhibit nitrification.

2.2. Batch Ni(II) toxicity experiments

Three Ni(II) levels (i.e., 0.1, 1, and 10 mg/L, from environmentally relevant level to high level) were set to assess the short-term and long-term effect of Ni(II) on the physicochemical properties. The 12 L biomass obtained from the parent SBR, was firstly washed for 3 times with distilled water, and then divided evenly to four identical SBRs with 3 L working volume. All reactors operated according to the procedure as same as the parent SBR. After settling period, 2 L supernatant was taken out from each SBR, and replaced with 2 L fresh feeding mediums containing 0 (i.e., the control reactor), 0.1, 1, and 10 mg/L of Ni(II) at the feeding stage, respectively. The feeding mediums were prepared by diluting 0, 2, 20, and 200 mL nickel sulfate stock solution (100 mg/L), respectively. Except for Ni(II) concentrations, the other components were identical to the parent reactor. After the addition of Ni(II), the effluent P concentrations, ESS, sludge volume index (SVI) and capillary suction time (CST) values in all SBRs were measured every day to indicate the effect of Ni (II). In this study, the short-term exposure was defined as the exposure of activated sludge to Ni(II) only 1 day. After 60-days exposure, these determined data reached relatively stable. So the 60-days exposure to Ni(II) was defined as the long-term exposure. At the day 60, the compositions and structures of EPS, the surface hydrophobicity, the cell surface charge, and the integrity of cell membrane were determined to reveal the long-term effects of Ni(II).