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Communication

# Comparative study on ciprofloxacin removal in sulfur-mediated biological systems

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ABSTRACT

The removal of ciprofloxacin (CIP) in sulfur-mediated bioprocesses, *e.g.*, sulfate-reducing bacteria (SRB)-mediated process and sulfur-oxidizing bacteria (SOB)-mediated process, was examined for the first time. The results showed that the SRB-mediated process had more efficient CIP removal than that in SOB-mediated process. Adsorption was the primary removal pathway of CIP in SRB-mediated process and SOB-mediated process with the specific adsorption removal rate of $131.4 \pm 1.1$ μg/g-SS/d and $30.1 \pm 1.4$ μg/g-SS/d, respectively, at influent CIP concentration of 500 μg/L. In addition, extracellular polymeric substances (EPS) also played an important role on CIP migration and removal in both types of sludge. Further study was conducted to specify the different adsorption of CIP in these two sludge systems from the perspective of sludge properties. The results indicated that there are more potential adsorption sites exist on the SRB-mediated sludge for CIP adsorption than SOB-mediated sludge since the higher protein (PN) content and more kinds of aromatic amino acid substances in EPS, more negative zeta-potential and stronger and more numbers of functional groups in SRB-mediated sludge compared to SOB-mediated sludge. The findings of this study provide insights into the sludge properties affecting CIP removal in sulfur-mediated bioprocesses, and are of guiding significance to employ sulfur-mediated biological systems for treating CIP-containing wastewaters.



In recent years, antibiotics have become a kind of contaminants in aquatic environments and received increasing public attention and scientific interest [1]. As a representative broad-spectrum antibiotic, ciprofloxacin (CIP) has been widely used in the treatment of animal and human disease, and frequently detected in various natural water bodies since incomplete assimilation and metabolism in organism [1,2]. The wastewater-treatment plants (WWTPs), as the main points of collection and subsequent release of antibiotics into the environment, are less efficient in CIP removal [3]. The residual CIP and its derivatives would accumulate in the environment and further bring a serious threat to ecosystem and human health by inducing antibiotic resistance genes (ARGs) and bacteria (ARB) and producing the physiological and genetic toxicity for organisms [4,5].

Currently, activated sludge (AS) and anaerobic methanogenic sludge (AnMS) processes are the main processes in WWTPs [3,6]. However, the processes have high energy consumption, large sludge production, low antibiotics removal efficiency and poor tolerance against pharmaceuticals [6,7]. In recent years, sulfur-mediated bioprocesses, such as sulfate-reducing bacteria (SRB)-mediated process and sulfur-oxidizing bacteria (SOB)-mediated process have drawn significant attention since the potential for pharmaceutical wastewaters treatment due to several inherent merits, such as low sludge production and energy consumption, high tolerance to antibiotics, and high efficiency for antibiotics and pollutants removal [8,9]. Antibiotics, such as fluoroquinolones and sulfonamides were effectively removed by SRB-mediated process *via* adsorption and biodegradation [10]. Moreover, some SOB genera (*e.g.*, *Thiobacillus delicatus*, *Thiothrix*, *Sulfurimonas denitrificans*, *Thiobacillus thyasiris*, and *Thiosphaera pantotropha*) can secrete some key enzymes for organics biodegradation [11]. Thus, these studies clearly highlighted the potential of sulfur-mediated bioprocesses for pharmaceutical compounds removal.

Adsorption and biodegradation are the main removal pathways for antibiotics in biological sludge systems, and adsorption was found to be the primary removal pathway of CIP in AS and AnMS systems [1,3]. The CIP adsorption occurred mainly *via* multiple

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adsorption mechanisms including electrostatic interaction (electrostatic attraction, and cation exchange and bridging), $\pi$-$\pi$ interaction, hydrogen bond and surface complexation (coordination and chelation) in biological sludge systems [12,13]. And these adsorption processes were strongly correlated with the properties of the sludge, such as the compositions and structure of extracellular polymeric substances (EPS), zeta-potential and surface functional groups of biological sludge [7,12–14]. EPS, a mixture of high molecular weight polymers produced by microorganisms, play an important role in pollutants removal in biological sludge system [15]. The functional groups, such as C—OH, C—O—C, N—H, O—H and COOH in sludge can provide sites for CIP adsorption [12,16,17]. In addition, the sludge surface potential also has a significant effect on the electrostatic interaction between sludge and antibiotics [12,18]. Thus, there is a critical need to specify the sludge properties affecting CIP removal in sulfur-mediated bioprocesses to better understand the removal mechanisms of CIP in sulfur-mediated bioprocesses.

Therefore, the objectives of this study were to investigate the CIP removal in sulfur-mediated bioprocesses (SRB-mediated process and SOB-mediated process). The sludge properties (compositions and structure of EPS, zeta-potential and surface functional groups) affecting CIP adsorption in these two sludge systems were determined using a series chemical and spectral analyses. The findings of this study are helpful to better understand the removal mechanisms of CIP in sulfur-mediated bioprocesses and thus opens-up a new opportunity in employing sulfur-mediated bioprocesses for treating CIP-containing wastewaters.

Two sludge systems, namely anaerobic SRB-mediated process and anoxic SOB-mediated process were set up in two different reactors, *i.e.*, sulfate-reducing up-flow sludge bed (SRUSB) reactor and sulfur-driven autotrophic denitrification (SAD) reactor, respectively (Fig. S1 in Supporting information). The two reactors were operated in parallel for 120 days in different stages to examine the CIP removal (the details of inocula and operating conditions for each reactor are presented in Supporting information). The two reactors were fed with synthetic wastewaters (Tables S1 and S2 in Supporting information) with different influent CIP concentrations at different stages: CIP of 0 μg/L (Control) in Stage 1, CIP of 100 μg/L in Stage 2 and CIP of 500 μg/L in Stage 3. The CIP concentration ranges were selected based on CIP concentrations detected in municipal sewage and hospital wastewaters [3,19]. The CIP removal efficiency and specific removal rate *via* adsorption and biodegradation at different operation stages of SRUSB and SAD reactors were determined, respectively (details in Supporting information).

There are three groups of batch experiments were conducted in serum bottles to investigate the adsorption and biodegradation of CIP in sulfur-mediated bioprocesses (details in Supporting information). Group I designated as abiotic control (without sludge) was conducted to examine the possible hydrolysis and photolysis of CIP; Group II was designed to examine the CIP biodegradation by SRB-mediated sludge and SOB-mediated sludge. Both the batch experiments of Groups I and II were conducted for 5 days at initial CIP concentration of 500 μg/L. In Group III, the batch experiments were conducted to investigate the adsorption kinetics and isotherms of CIP by SRB-mediated sludge and SOB-mediated sludge, and the adsorption experiments were carried out for 24 h at different initial CIP concentrations of 50, 100, 200, 300, 400 and 500 μg/L (details in Supporting information). During the experiments, 2 mL of mixed liquor sample was regularly withdrawn from each serum bottle for analyzing CIP concentration in aqueous, EPS and Sludge (after EPS extraction) phases to examine the CIP removal *via* biodegradation and adsorption (EPS + sludge) (details in Supporting information).

The influent, effluent and sludge samples from SRUSB and SAD reactors were regularly collected for routine chemical analyses (details in Supporting information). CIP content in aqueous phase, EPS and Sludge samples were determined by an ultra-performance liquid chromatography (UPLC) (details of CIP analysis are given in Supporting information). The SRB-mediated sludge and SOB-mediated sludge samples were collected from SRUSB and SAD reactors on Day 40, 80 and 120 for EPS content (polysaccharide (PS), PN and humic acid) and structure (fluorescence components in EPS) analyses, and zeta-potential and Fourier transform infrared (FTIR) analyses (details in Supporting information).

The pollutants removal in SRB-mediated process and SOB-mediated process were investigated. As shown in Fig. 1, the average removal efficiency of chemical oxygen demand (COD) at all operational stages in SRUSB reactor was over 85% with the average specific removal rate of 170.6 ± 4.4 mg/g-SS/d. And over 60% of the influent sulfate was reduced into dissolved sulfide with the average specific removal rate of 67.3 ± 1.7 mg/g-SS/d and no sulfur intermediates (*e.g.*, sulfite, thiosulfate, and elemental sulfur) were detected in the effluent of reactor (the performances of SRUSB and SAD reactors are summarized in Table S3 in Supporting information). Similar results were observed that high influent CIP concentration of 5000 μg/L had no inhibitory effect on the acclimatized microbes in SRB-mediated process; instead facilitated *Desulfobacter* enrichment [20]. However, the removal efficiencies of $NO_3^-$ and $S^{2-}$ decreased significantly in SOB-mediated process under CIP exposure. The specific removal rate of $NO_3^-$ and $S^{2-}$ decreased significantly from (7.0 ± 0.5 mg/g-SS/d) and (32.3 ± 0.8 mg/g-SS/d) to (4.6 ± 0.4 mg/g-SS/d) and (26.5 ± 0.8 mg/g-SS/d) with increasing CIP concentration from 0 to 500 μg/L. The results suggested that the SRB-mediated sludge showed higher tolerance against CIP than SOB-mediated sludge, and has significant potential in treating wastewater containing sulfate and FQs (*e.g.*, CIP).

The abiotic control experiments showed almost no CIP removal *via* volatilization and hydrolysis during the experiment (Fig. S2 in Supporting information). Thus, the CIP removal in sulfur-mediated bioprocesses was mainly attributed to adsorption and biodegradation. As shown in Fig. 2A, CIP was effectively removed in SRB-mediated process with the adsorption and biodegradation removal efficiencies of 81.1% ± 0.7% and 7.3% ± 0.6% at Stage 2, and 70.3% ± 1.1% and 13.5% ± 0.9% at Stage 3, respectively. However, nearly no CIP biodegradation (biodegradation removal efficiency < 0.3%) was observed and only adsorption was contributed to CIP

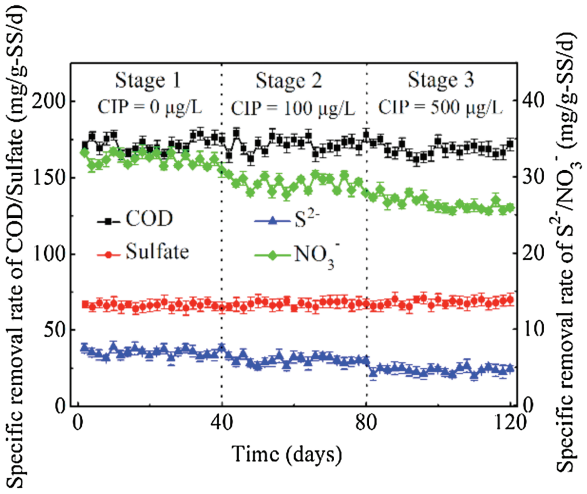


**Fig. 1.** COD and sulfate removal in SRB-mediated process, and nitrate and sulfide removal in SOB-mediated process.

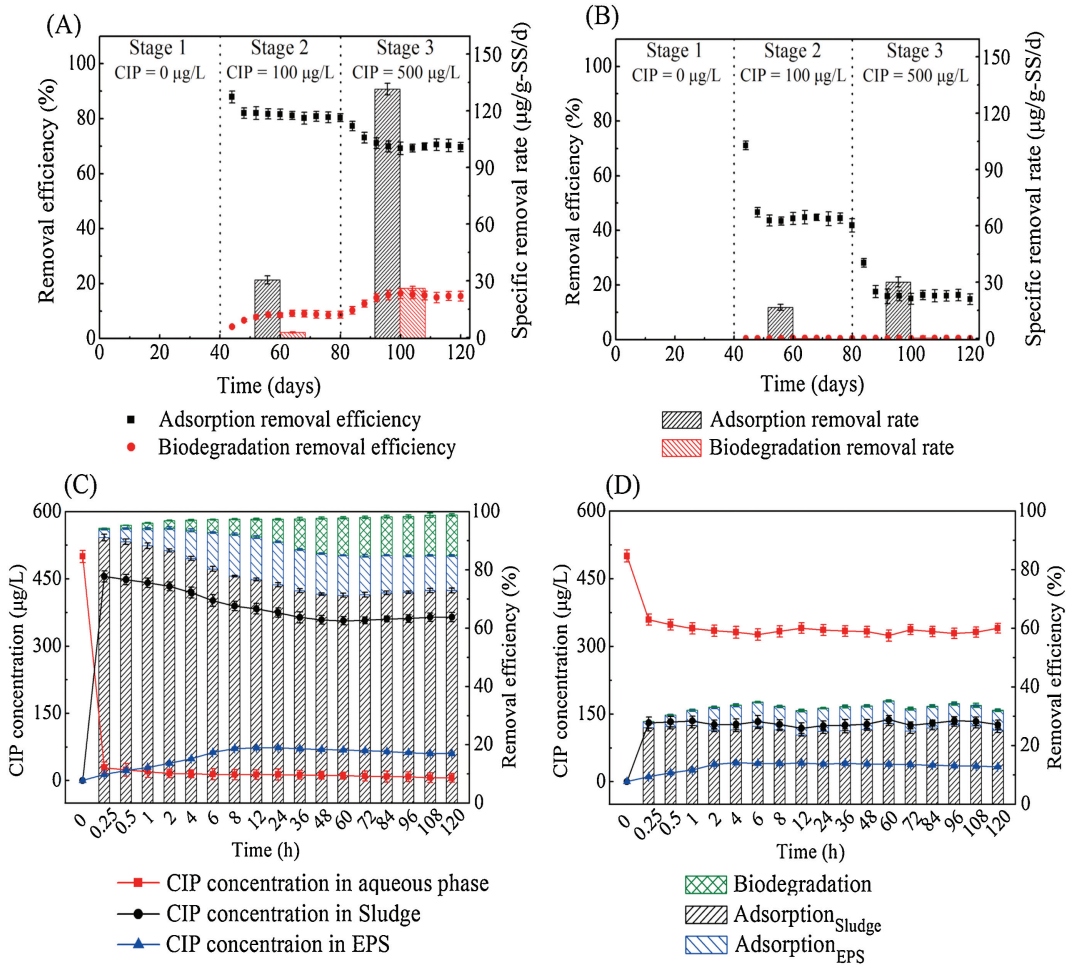


**Fig. 2.** CIP removal by SRB-mediated process (A) and SOB-mediated process (B) in bioreactors at different operation stage, and CIP removal by SRB-mediated sludge (C) and SOB-mediated sludge (D) at initial CIP concentration of 500 μg/L in batch tests.

removal in SOB-mediated process with the removal efficiency of 44.1% ± 1.6% at Stage 2 and 16.4% ± 1.3% at Stage 3 (Fig. 2B). It is apparent that adsorption was the primary pathway for CIP removal in sulfur-mediated bioprocesses. And SRB-mediated process showed higher CIP adsorption than SOB-mediated process with the specific adsorption removal rate of 131.4 ± 1.1 μg/g-SS/d and 30.1 ± 1.4 μg/g-SS/d, respectively, at Stage 3. Biodegradation also played an important role in CIP removal in SRB-mediated process, the specific biodegradation mechanisms and pathways of CIP have been reported in our previous study [20].

The results of batch experiments showed that the CIP concentration in aqueous phase decreased significantly from 500 ± 4.2 μg/L to 28.8 ± 1.8 μg/L and 358.8 ± 3.6 μg/L within 15 min in SRB-mediated sludge and SOB-mediated sludge, respectively (Figs. 2C and D). The rapid decrease in CIP concentration at such a short period of time was attributed to adsorption onto the sludge [21]. The results are in consistent with the batch adsorption experiments that about 97.4% ± 0.7% and 33.1% ± 0.4% of CIP (initial CIP concentration of 500 μg/L) in aqueous phase were adsorbed onto inactivated SRB-mediated sludge and SOB-mediated sludge, respectively, at the end of adsorption experiments (24 h) (Fig. S3 in Supporting information). Similar CIP removal was found in aerobic AS and AnMS systems [7]. As shown in Figs. 2C and D), EPS also play a critical role on CIP migration and removal, the CIP in EPS increased in the first 24 h of the batch tests and about 14.7% ± 0.3% and 8.2% ± 0.3% of CIP were bound to EPS of SRB-mediated sludge and SOB-mediated sludge, respectively. The higher CIP removal efficiency of EPS in SRB-mediated sludge than that in SOB-mediated sludge was due to the higher content of EPS produced by SRB-mediated sludge than SOB-mediated sludge as shown in Figs. 3A and B.

Further, the adsorption behavior of CIP onto SRB-mediated sludge and SOB-mediated sludge were determined based on the adsorption kinetics (*pseudo*-first-order and *pseudo*-second-order models) and isotherms (Freundlich, Linear and Langmuir) analyses (the relevant constants are summarized in Tables S4 and S5 in Supporting information). The results showed that the *pseudo*-second-order kinetics model provided good fit for CIP adsorption in both types of sludge (correlation coefficient, $R^2 > 0.999$) with very low deviation (with relative error ≤ 5%) between calculated and experimental data (Table S4). It indicated the chemisorption of CIP on both types of sludge which resulted from strong interaction [22]. The *pseudo*-second-order adsorption rate constants ($K_2$) of CIP on SOB-mediated sludge (4.1∼0.9 g-SS/μg-CIP/h) were higher than those on SRB-mediated sludge (3.1∼0.6 g-SS/μg-CIP/h), suggested that the adsorption equilibrium of CIP on SOB-mediated sludge is faster than that on SRB-mediated sludge [23].

The adsorption of CIP by the two types of sludge conformed well to Langmuir ($R^2 > 0.999$) and Freundlich ($R^2 > 0.99$) isotherms at the CIP concentration range of 50∼500 μg/L (Table S5). It suggested that the adsorption of CIP by these two kinds of sludge is nonlinear, while a complex heterogeneous process, which was

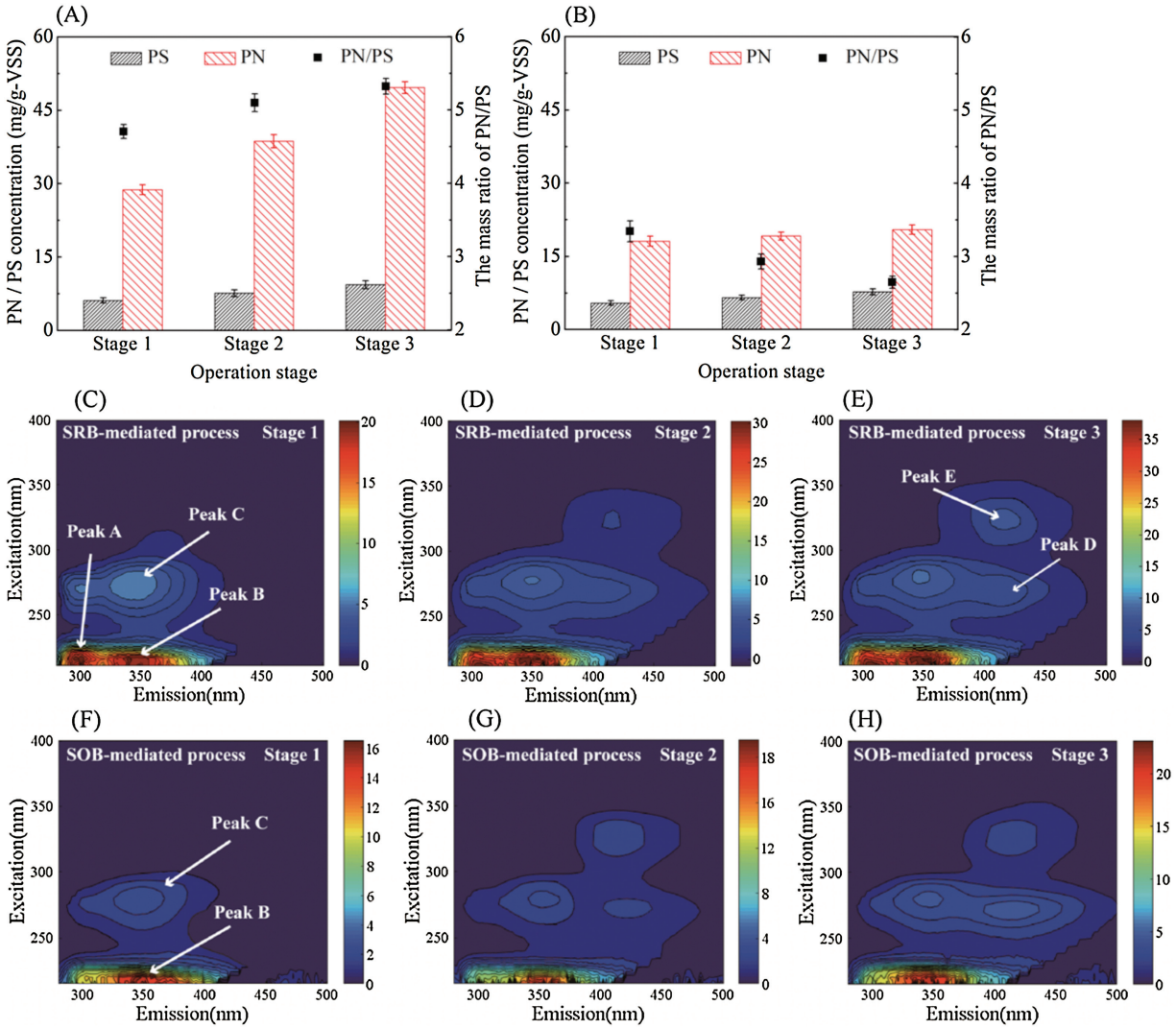


**Fig. 3.** Compositions of EPS in SRB-mediated sludge (A) and SOB-mediated sludge (B) samples, and three-dimensional excitation-emission matrix (3D-EEM) fluorescence spectra of EPS in SRB-mediated sludge (C)-(E) and SOB-mediated sludge (F)-(H) samples on Days 40 (Stage 1), 80 (Stage 2) and 120 (Stage 3).

attributed to the presence of heterogeneous structure of EPS distributed throughout the sorption surface [24]. The $q_m$ (theoretical maximum adsorption capacity) of CIP on SRB-mediated sludge ($476.2 \pm 10.4$ μg/g-SS) was significantly higher than that on SOB-mediated sludge ($82.0 \pm 5.3$ μg/g-SS), indicated that the SRB-mediated sludge showed more excellent adsorption potential for CIP than SOB-mediated sludge.

EPS played an important role in CIP removal in sulfur-mediated bioprocesses in this study. The variations of the composition of EPS in both types of sludge at each operation stage were examined. As shown in Figs. 3A and B, the EPS content of the two types of sludge samples increased significantly with increasing influent CIP concentration from 0 to 500 μg/L, suggesting the secretion of EPS was stimulated under CIP exposure. EPS is always regarded as natural barriers to protect cells against the unfavorable circumstances, and microbial consortia tend to secrete more EPS under antibiotics exposure [25,26]. No humic acid was detected in EPS, the PN and PS are the primary components in EPS of the two types of sludge. However, the PN content was significantly higher than PS in EPS with the PN/PS ratio range of 4.7∼5.3 and 3.3∼2.6 in SRB-mediated sludge and SOB-mediated sludge samples, respectively. The PN such as tryptophan and tyrosine can provide binding sites for the micropollutants adsorption [7,27]. The higher PN content and the PN/PS ratio in EPS of SRB-mediated sludge than that of SOB-mediated sludge, indicated that EPS of SRB-mediated sludge provided more adsorption sites for CIP than that of SOB-mediated sludge.

The 3D-EEM fluorescence spectra of EPS in two types of sludge samples were examined and shown in Figs. 3C–H. Three obvious fluorescence peaks were identified from the EEM fluorescence spectrum of EPS in SRB-mediated sludge sample collected from Stage 1. The Peak A (excitation/emissions (Ex/Em): 230/295∼305 nm) is identified as tyrosine in PN [7]. The Peak B (Ex/Em: 215∼225/335∼350 nm) and Peak C (Ex/Em: 270∼280/335∼350 nm) are associated with tryptophan [27]. But, only Peaks B and C (tryptophan) were detected in EEM fluorescence spectrum of EPS in SOB-mediated sludge sample at Stage 1. The fluorescence intensities of EPS in both types of sludge samples increased with increasing influent CIP concentration, especially the amino acid substances (tyrosine and tryptophan), suggesting that tyrosine- and tryptophan-like PN in EPS played an important role in the adsorption of CIP [7]. The Peak D (Ex/Em: 268∼273/410∼420 nm) and Peak E (Ex/Em: 320∼330/410∼420 nm) identified as CIP were detected after CIP addition and intensified with increasing CIP concentration, which was caused by the accumulation of CIP in EPS. The results indicated that there were more kinds of aromatic

amino acid substances involved in CIP adsorption in EPS of SRB-mediated sludge (tyrosine and tryptophan) than that in SOB-mediated sludge (tryptophan).

The zeta potentials of SRB-mediated sludge and SOB-mediated sludge were determined and showed in Fig. 4. The surfaces of the both types of sludge were negatively-charged. It may be due to the ionization of functional groups of carboxyl and amino existing in the PN in EPS of sludge flocs were negative at neutral pH values [28]. Therefore, the increase of PN in EPS resulted in the decrease of the zeta potential of both types of sludge. Apparently, the surface of SRB-mediated sludge had more negative zeta-potential (-21.7 ~ -25.4 mV) than that of SOB-mediated sludge (-12.0 ~ -13.5 mV), suggesting that more available sites for CIP adsorption *via* electrostatic attraction and cation exchange in SRB-mediated sludge than that in SOB-mediated sludge at neutral pH values [12]. The difference of surface charge between SRB-mediated sludge and SOB-mediated sludge may be caused by the different content and composition of EPS [29].

The chemical groups on SRB-mediated sludge and SOB-mediated sludge were identified with the results being shown in Fig. 5. At first glance, the positions of some FTIR peaks of the two types of sludge are quite close, indicating that certain types of chemical groups are similar. The peak at 1034 $cm^{-1}$ corresponds to stretching vibration of C—OH in PS [30]. The peaks at about 1385 $cm^{-1}$ and 1454 $cm^{-1}$ are due to the symmetric deformation and asymmetric bending vibration of COOH group in amide III, respectively, suggesting the presence of the PN [12,31]. A relatively sharp peak at 1643 $cm^{-1}$ is assigned to C=O and C—N stretching of amide I group in the PN [31]. The peak at 1404 $cm^{-1}$ is assigned to C—O group in hydroxyl acid lipids [32]. The peak at 2926 $cm^{-1}$ is attributed to the stretching vibration of C—H in the alkenes class [30]. The broad band (around 3417 $cm^{-1}$) indicates the presence of O—H group [7,30]. All of these functional groups were observed on the surface of the both types of sludge samples. However, the peak at 1113 $cm^{-1}$ related to C—O—C group [12] and the peak at 1543 $cm^{-1}$ corresponded to the stretching of N—H and C=N in amide II linkage of PN [31], were only appeared on the surface of SRB-mediated sludge samples, but not on the surface of SOB-mediated sludge. The number of FTIR peaks of SRB-mediated sludge is more than that of SOB-mediated sludge, indicating that more chemical groups on the surface of SRB-mediated sludge than that on SOB-mediated sludge. Furthermore, the relative intensity of each peak on SRB-mediated sludge was stronger than that on SOB-mediated sludge, which may be because the SRB-mediated sludge generated more PN, PS and other polymers than SOB-mediated sludge. The intensities of the peaks of C—OH, C—O—C, COOH, C—O, N—H/C = N, C=O/C—N and O—H decreased with increasing influent CIP concentration, suggesting that these functional groups on sludge surface were involved in CIP adsorption [7]. Several studies also found that the functional groups, such as C—O, C—O—C, N—H, O—H and COOH in sludge can provide sites for CIP adsorption [12,16,17]. The results indicated that the SRB-mediated sludge can provide more available sites for CIP adsorption than SOB-mediated sludge.

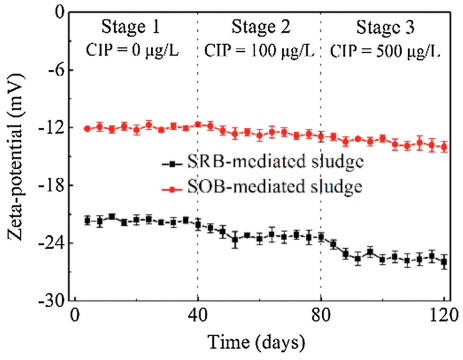


**Fig. 4.** The zeta-potential of SRB-mediated sludge and SOB-mediated sludge samples at different operation stage.

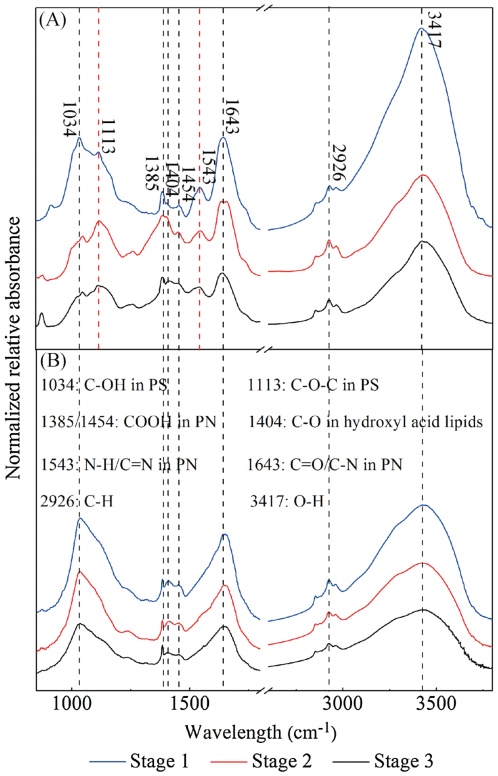


**Fig. 5.** FTIR spectra of SRB-mediated sludge samples in SRUSB reactor (A) and SOB-mediated sludge samples in SAD reactor (B) on days 40 (Stage 1), 80 (Stage 2) and 120 (Stage 3).

In this work, we investigated the CIP removal in sulfur-mediated bioprocesses. The results indicated that SRB-mediated process showed higher tolerance against CIP than SOB-mediated process with effective removal of COD and sulfate in SRB-mediated process, but suppression effect on $NO_3^-$ and $S^{2-}$ removal in SOB-mediated process under CIP exposure. The SRB-mediated process had more efficient CIP removal than that in SOB-mediated process with the removal efficiency of 84.1% ± 1.3% and 16.4% ± 1.3%, respectively, at influent CIP concentration of 500 μg/L (Stage 3). Adsorption was the primary removal pathway of CIP, and EPS also played an important role on CIP migration and removal in both types of sludge. Further studies indicated that there are more potential adsorption sites exist on the SRB-mediated sludge for CIP adsorption than SOB-mediated sludge since the higher protein (PN) content and more kinds of aromatic amino acid substances in EPS, more negative zeta-potential and stronger and more numbers of functional groups in SRB-mediated sludge compared to SOB-mediated sludge. The findings of this study are helpful to understand the adsorption mechanisms of CIP in sulfur-mediated bioprocesses, and have guiding significance for the treatment of CIP-containing wastewaters with sulfur-mediated bioprocesses.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.cclet.2020.04.048.

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