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Unrecognized role of humic acid as a reductant in accelerating fluoroquinolones oxidation by aqueous permanganate

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ABSTRACT

A great concern has been raised regarding the issue of fluoroquinolones (FQs) in the environment. In this work, the transformation of FQs by commonly used oxidant permanganate (Mn(VII)) in the absence and presence of humic acid (HA), ubiquitously existing in aquatic environments, was systematically investigated. Here, the catalytic role of *in-situ* formed MnO_2 on Mn(VII) oxidation of FQs depending on solution pH and co-existing substrates was firstly reported. It was interestingly found that HA could appreciably accelerate FQs degradation by Mn(VII) at environmentally relevant pH. HA as a reductant in accelerating FQs by Mn(VII) oxidation was distinctly elucidated for the first time, where MnO_2 *in situ* formed from the reduction of Mn(VII) by HA served as a catalyst. Similar products were observed in the presence versus absence of HA. Considering that the accelerating role of HA was related to its reducing ability, an activation method based on Mn(VII) and reductant (*i.e.*, Fe(II), Mn(II) and (bi)sulfite) was proposed, which exhibited considerable potential for application in the treatment of FQs contaminated water.

Fluoroquinolones (FQs) are a class of synthetic antibacterial agents widely used in human and veterinary medicines. As a result of large usage, the ubiquitous presence of FQs in municipal wastewater effluents and affected surface waters is reported in numerous studies [1–3]. A great concern has been raised regarding the issue of FQs in the environment, since the uncertainty of constant long-term exposure to low concentrations of such compounds poses potential risk mainly to humans and aquatic fauna [4–7]. Thus, many efforts have been made to remove these pollutants in waters to avoid their possible adverse effects on aquatic ecology, animals, and human beings.

Permanganate (Mn(VII)) has been widely used for water treatment to remove taste and odor, control dissolved Mn(II) and biological growth. Mn(VII) technique shows attractive characteristics due to its relatively low cost, comparative stability, and ease of handing. In addition, numerous studies have demonstrated that Mn(VII) as a selective oxidant is fairly effective in treating emerging organic micropollutants [8–12]. For instance, in our recent work, Mn(VII) was found to readily oxidize phenols and primary anilines [13]. Interestingly, manganese dioxide (MnO_2) *in-situ* formed from Mn(VII) reduction could act as a catalyst accelerating Mn(VII) oxidation of phenolic and aniline contaminants especially at slight acid pH [8,13–15]. In the study of Hu *et al.* [16], the authors investigated the potential applicability of Mn(VII) for ciprofloxacin (CF) elimination. The authors reported that Mn(VII) mainly attacked CF at tertiary amine group. However, the role of MnO_2 in CF as well as other FQs degradation by Mn(VII) has never been characterized so far.

Dissolved organic matters (DOM) ubiquitously exists in aquatic environments and can participate in various physical, chemical, and biochemical processes [17–20]. It was reported that humic acid (HA), as a major constituent of DOM could affect Mn(VII) oxidation in markedly different ways . For instance, Jiang *et al.* [8] found that HA could significantly accelerate the oxidation of triclosan under slight acid condition, where MnO_2 generated *via* the reduction of Mn(VII) by HA acted as a catalyst and thus facilitated the reactions. Similar promoting role of HA in the degradation of phenol and chlorophenols by Mn(VII) was reported by Sun *et al.* [21], and

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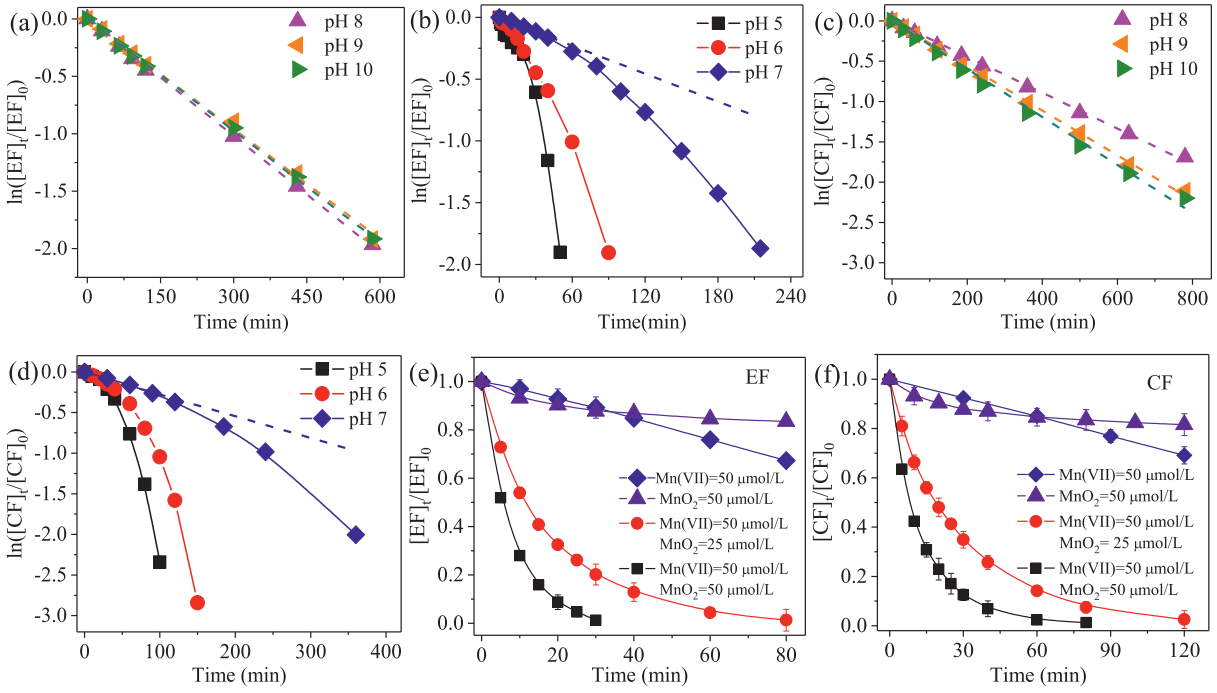

Fig. 1. The plot of $\ln(FQ)_t/\ln(FQ)_0$ *vs.* time (*i.e.*, time course of FQ oxidation) in the presence of excess Mn(VII) over the pH range of 5–10 (a–d) and the effect of additive MnO_2 on the degradation of FQs by Mn(VII) at pH 7 (e,f). Experimental conditions: $[EF]_0 = [CF]_0 = 5$ μmol/L and $[Mn(VII)]_0 = 50$ μmol/L.

these authors proposed that the MnO_2 served as oxidant rather than catalyst contributing to the acceleration. While, HA showed a limited effect on the degradation of carbamazepine by Mn(VII) and showed inhibited effect on the degradation of sulfamethoxazole by Mn(VII). Lately, Xu *et al.* [22] reported the accelerating effect of HA on the degradation of levofloxacin (LF) by Mn(VII), wherein both $^{\bullet}OH$ and HA stabilized Mn(III) were proposed to cause the accelerated degradation. So, the effect of HA on the reactions of organic compounds with Mn(VII) are far from clear which warrants further investigation.

The primary objective of this work was to evaluate the oxidation of FQs by Mn(VII), the effect of MnO_2 and the accelerating mechanism of HA. For this purpose, the transformation of FQs in the absence and presence of HA were investigated in detail. Firstly, the transformation of FQs by Mn(VII) was investigated in synthetic buffered solutions under various conditions with a focus on the role of MnO_2 *in situ* formed. Then, the effect of HA on the reaction kinetics and transformation products was comparatively investigated. Finally, an activation method based on the reduction of Mn(VII) by Fe(II), Mn(II) and bisulfate (BS) was proposed to enhance the oxidation of FQs by Mn(VII). Here, commonly detected enrofloxacin (EF) and CF were selected as representative FQs and their structures were shown in Fig. S1 (Supporting information). The details of materials used in this study and the analytical methods were listed in Supporting information.

Figs. 1a–d showed the time course of EF and CF decay in the presence of excess Mn(VII). As can be seen, at pH 8–10, the decay of EF and CF followed pseudo-first-order kinetics, indicating the reactions were first-order with respect to EF in alkaline solution. However, the degradation of EF and CF at pH 7 displayed interesting autocatalysis: an initial lag phase followed by a secondary rapid stage. In addition, more evident autocatalysis was observed with solution pH decreasing from 7 to 5. Similar autocatalysis was observed during the oxidation of triclosan, 2,4-dichlorophenol, aniline and primary anilines (*e.g.*, bromo-aniline) by Mn(VII) especially under acid conditions, and these enhancements were previously ascribed to the gradual accumulation of MnO_2 *in situ* formed [8,14,15]. To shed light on the role of MnO_2, the effect of MnO_2 *ex-situ* prepared on the reactions of FQs with Mn(VII) was examined. As can be seen in Figs. 1e and f, the addition of MnO_2 appreciably accelerated the degradation of FQs by Mn(VII). Slight or negligible FQs were degraded by 50 μmol/L MnO_2 alone during the investigated time scale. Therefore, MnO_2 was inert to the reaction with FQs. These considerable enhancements could not be resulted from the contribution of MnO_2 as a co-oxidant. The catalytic role of MnO_2 well reported in literature especially in acid and neutral solutions might rationalize the enhancement in FQs degradation by Mn(VII).

It was well reported that complexing ligands (*e.g.*, pyrophosphate (PPP)) could effectively suppress the generation of MnO_2 *in situ* formed from Mn(VII) reduction [23]. As can be seen in Fig. S2a (Supporting information), the presence PPP considerably inhibited the degradation of EF over the pH range of 5–7, while indiscernible difference was observed under alkaline conditions in the presence *versus* absence of PPP (data not shown). Interestingly, in the presence of PPP, the decay of EF in the pH range of 5–7 followed pseudo first-order kinetics without autocatalysis. These findings further suggested the important role of MnO_2 especially under acid and neutral conditions, where gradual accumulation of MnO_2 as a catalyst resulted in the autocatalytic kinetics. Similar to PPP, co-existing cations Ca^{2+} and Mg^{2+} considerably inhibited the degradation of EF as well as CF at acid and neutral pH, where the loss of FQs followed pseudo-first-order rate law without autocatalysis (Fig. S2b in Supporting information). In previous work, a surface complex catalytic mechanism was proposed to explain the role of MnO_2 in accelerating Mn(VII) oxidation [24–28]. It was proposed that absorbed contaminants on MnO_2 surface facilitated its electron transfer with Mn(VII), where a precursor complex of organic compound and MnO_2 initially formed was necessary. So, it was not difficult to understand the inhibitory effect of cations that Ca^{2+} and Mg^{2+} competitively occupied adsorptive sites on negatively charged MnO_2 *via* electrostatic interactions, and accessible sites for FQs and amount of the precursor complex according to decreased leading to the suppressive degradation. In addi-

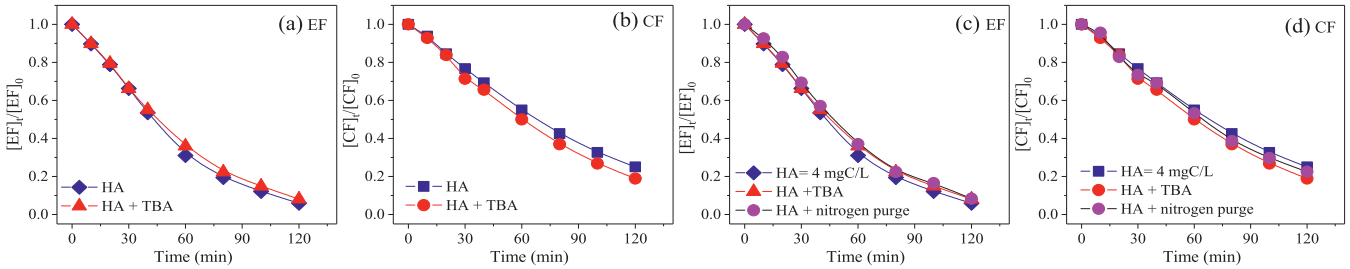

Fig. 2. (a,b) Effect of HA on the degradation of FQs by Mn(VII). Effect of TBA and nitrogen purge on the degradation of FQs by Mn(VII)/HA (c,d). Experimental conditions: $[EF]_0 = [CF]_0 = 5$ μmol/L, $[Mn(VII)]_0 = 50$ μmol/L, $[HA]_0 = 1$ or 4 mg C/L, $[TBA]_0 = 5$ mmol/L, pH 7.

tion, cations could accelerate the aggregation of MnO_2, leading to the increase of particle size and decrease of specific surface area of MnO_2, which resulted in the decrease of adsorptive sites of MnO_2.

In previous work, the important role of MnO_2 on the transformation of FQs by Mn(VII) was somewhat overlooked. So the reactivity of Mn(VII) direct oxidation should be reinvestigated. In the pH range of 5–7, complexing ligands or divalent metal cations were added to inhibit the contribution of MnO_2 and the *pseudo*-first-order rate constants (k_{obs} in s^{-1}) of FQs oxidation by Mn(VII) alone could be obtained. The apparent second-order rate constants (k_{app} in L mol^{-1} s^{-1}) could be determined as shown in Fig. S3 (Supporting information). As can be seen, pH had negligible effect on the Mn(VII) direct oxidation rates, where solution pH increased from pH 5 to 10, the values of $k_{app,EF}$ varied form 1.23 L $mol^{-1}s^{-1}$ to 1.06 L $mol^{-1}s^{-1}$, and $k_{app,CE}$ varied from 0.93 L $mol^{-1}s^{-1}$ to 0.99 L $mol^{-1}s^{-1}$, respectively. Previous work have demonstrated that fluoroquinolone moiety was inert toward Mn(VII) [16]. So, the comparable reactivity of Mn(VII) towards EF and CF reactivity indicated that Mn(VII) might primarily attack EF and CF at their tertiary aromatic amine group (*i.e.*, N1 atom see Fig. 1). The weak pH dependency of k_{app} also precluded aliphatic amine as the dominant target moiety, because the secondary amine (N4 nitrogen) exhibited higher reactivity after deprotonated while the species of N1 nitrogen remained unchanged over a wide pH range.

In the absence of inhibitors (*i.e.*, complexing ligands and cations), the oxidation kinetics of EF and CF at acid and neutral solution was accelerated and the degradation rates of FQs were increased as solution pH decreased from 7 to 5, as described in the precedent section. Given that FQs showed similar reactivity towards Mn(VII) regardless of solution pH, this pH-dependency was due to the fact that the catalytic role of MnO_2 was highly related to solution pH [14]. Such pH-dependence might be related to the electrostatic repulsion that affected the surface-complex formation between MnO_2 and FQs, which was necessary for the subsequent electron transfer. The increase of the pH would increase the negatively charged sites of MnO_2. Moreover, with the increase of solution pH, the speciation of FQs changes from cationic to anionic species, less susceptible to adsorbing to the negative charged MnO_2, resulting in stronger electrostatic repulsion. Thus, the amount of the precursor complex would accordingly decrease as a function of increasing pH. Given the surface complex catalytic mechanism, the decrease of precursor complex amount would result in the inhibition of oxidation process.

To elucidate the effect of HA on the degradation of FQs by Mn(VII), experiments were conducted in the presence of varying HA concentrations at environmentally relevant pH 7. As can be seen in Figs. 2a and b, the presence of HA significantly accelerated the degradation of FQs and the accelerating effect was enhanced with the elevated HA concentration. For instance, with the increase of HA concentration from 0 to 4 mgC/L, the percentage of EF degradation considerably increased from ~50% to ~100% at 120 min. The promoting effect of HA on levofloxacin oxidation by Mn(VII) was also observed in a previous study [22]. Therein, hydroxyl radical (•OH) were proposed to be responsible for the enhancement, which was originated from the reaction of manganese intermediates with oxygen. In order to discern the role of HA during Mn(VII) oxidation of CF and EF, the effect of *tert*-butanol (TBA; scavenger for •OH) was examined. Moreover, experiments were also conducted in oxygen-free atmosphere (by bubbling nitrogen) since oxygen was reported to play a significant role in the formation of •OH. Interestingly, it was found that TBA and nitrogen-atmosphere had negligible inhibiting effect on the degradation of EF and CF in Mn(VII)/HA system (Figs. 2c and d). These findings indicated that the promoting effect of HA on Mn(VII) oxidation of CE and EF was not attributed to •OH. It was well known that HA with abounding functional groups (*e.g.*, phenolic, alkene groups) could act as a reductant to consume Mn(VII) and the accompanied formation of MnO_2 was expected. The enhancement induced by HA observed at pH 7 might be ascribed to contribution of MnO_2 *in situ* formed from the reduction of Mn(VII) by HA. In order to confirm this, one common reducing agent As(III) was comparatively used in replacement of HA. As expected, the oxidation kinetics of EF were also conspicuously accelerated (Fig. S4 in Supporting information).

The transformation of EF by Mn(VII) in absence and presence of HA were analyzed by HPLC-QTOF-MS. A total of thirteen products were identified in the absence of HA, and their detailed information including retention time, accurate mass, molecular formula and proposed molecular structure were shown in Table S1 (Supporting information). For clear display, these products were numbered sequentially on the basis of retention time. As can be seen, the fluoroquinolone structure retained intact in all the proposed products, in consistent with the kinetic results that fluoroquinolone was unreactive towards Mn(VII). Multiple changes at piperazine ring were occurred with the formation of hydroxyl group (II, IV, VI, and X) and carbonyl group (VIII), and even dealkylation and ring cleavage products (I, III, V, VII, IX, XI, XII and XIII). In the presence of HA, identical products were detected indicating that MnO_2-catalysis did not affect the transformation of FQs by Mn(VII). On the basis of identified products as well as the literatures on the reactions of Mn(VII) with amines, the transformation pathway for the reactions of Mn(VII) with EF in the absence and presence of HA was tentatively proposed (Fig. S5 in Supporting information). The presence of the piperazine ring on the quinolone ring structure provides FQs with a considerably more potent antibacterial activity. Kinetics and products identification results in this study demonstrated that Mn(VII) could selectively oxidize the piperazine moiety of FQs and the antibacterial activity of FQs was expected to be reduced. Similar finding was also reported in the study of Hu *et al.* [29] that ciprofloxacin (one of the FQs) was oxidized by Mn(VII) *via* the attack of piperazine substituent, resulting in a significant reduction of antibacterial activity. Thus, Mn(VII) and Mn(VII)/MnO_2 treatment would cause efficient deactivation of FQs.

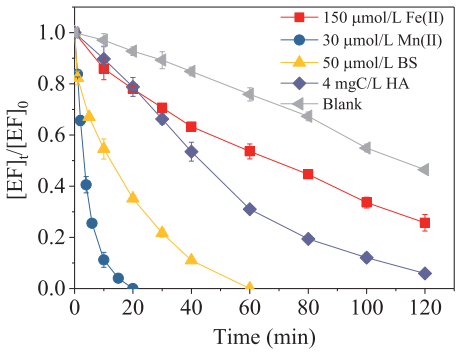

Fig. 3. Degradation of EF by Mn(VII) in the presence of reducing agents. Experimental conditions: $[EF]_0$ = 5 μmol/L, pH 7, Fe(II) group: $[Fe(II)]_0$ = 150 μmol/L, $[Mn(VII)]_0$ = 100 μmol/L; Mn(II) group: $[Mn(II)]_0$ = 30 μmol/L, $[Mn(VII)]_0$ = 70 μmol/L; BS group: $[BS]_0$ = 50 μmol/L, $[Mn(VII)]_0$ = 50 μmol/L; HA group: $[HA]_0$ = 4 mgC/L $[Mn(VII)]_0$ = 50 μmol/L; blank group: $[Mn(VII)]_0$ = 50 μmol/L.

Inspired by the accelerating role of HA as a reductant in Mn(VII) oxidation, addition of reducing agents might be a promising method to enhance the oxidation efficiency of FQs by Mn(VII). It was well known that Mn(II) and Fe(II) always exist in natural waters. The application of Mn(VII) oxidation as pre-oxidation technology was expected to readily oxidize Mn(II) and Fe(II) in real waters with the formation of MnO_2, which could in turn activate Mn(VII) for the treatment of organic contaminants. Recent studies demonstrated that the use of bisulfite (BS) could activate Mn(VII), where reactive Mn intermediates (*i.e.*, Mn(V) and Mn(III)) and sulfate radical *in-situ* generated were proposed to responsible for the enhancement of organics degradation [30–33]. However, the use of excess BS intending to maximize the activation efficiency could result in the release of Mn(II), which was undesirable in drinking water treatment processes. According to the results in this study, relatively low dosage of BS could be used in Mn(VII) activation process where MnO_2 *in situ* formed could also accelerate Mn(VII) oxidation.

The effects of Mn(II), Fe(II) and BS on the degradation of EF by Mn(VII) were investigated at environmentally relevant pH 7. In this section, different initial concentrations of Mn(VII) and stoichiometric concentrations of Mn(II) and Fe(II) (Eqs. S3-S5 in Supporting information) were used to ensure the preparation of identical Mn(VII) and *in-situ* formed MnO_2 ($[Mn(VII)]_0 = [MnO_2]_0 = 50$ μmol/L) in these two cases. In the case of BS, the [Mn(VII)]/[BS] ratio was set at 1:1 to maximum the activation of Mn(VII) by BS and prevent the release of Mn(II). As shown in Fig. 3, about 54% of EF was degraded by Mn(VII) alone within 120 min, while the degradation of EF increased to 74% in the presence of Fe(II). In the cases of Mn(II) and BS, EF was completely degraded within 20 min and 60 min, respectively. The accelerating effect of Mn(II) was more efficient than Fe(II). Possible explanations for the less efficient of Fe(II) includes (i) undesired precipitation of Fe(II) and coacervation of *in-situ* formed colloid MnO_2 caused by the ferric oxyhydroxides at pH 7 [34] and (ii) the lower activating efficiency of Fe(II)-induced MnO_2 than Mn(II)-induced MnO_2 by the loss of adsorption active sites. In the case of BS, the degradation of EF showed biphasic kinetics. About 17% EF was degraded within 1 min and as the reaction proceed EF was completely degraded in 60 mins. The rapid degradation of EF in 1 min was attributed the activation of Mn(VII) by BS where reactive Mn intermediate (*e.g.*, Mn(V)) and sulfate radical were involved [30]. Moreover, MnO_2 was formed from the rapid reduction of Mn(VII) by BS, which activated residual Mn(VII) for the degradation of EF in the following reaction time. The above results suggested that Mn(II), Fe(II), and BS inducing the formation of MnO_2 could activate Mn(VII) for the degradation FQs. For BS/Mn(VII) process, although the activation of Mn(VII) by BS was not quite efficient at neutral pH, a relative high ratio of [Mn(VII)]/[BS] ratio could be set so that the *in-situ* formed MnO_2 could act as catalyst for Mn(VII) after the consumption of BS.

In conclusion, the transformation of FQs by Mn(VII) in the absence and presence of HA was systematically investigated. Here, the catalytic role of *in situ* formed MnO_2 was reported, which was always overlooked in previous studies. Also, HA as a reductant in accelerating FQs degradation by Mn(VII) oxidation was distinctly elucidated for the first time, where MnO_2 *in situ* formed from the reduction of Mn(VII) by HA served as a catalyst. Considering that the accelerating role of HA was related to its reducing ability, an activation method based on Mn(VII) and reductant (*i.e.*, Fe(II), Mn(II) and (bi)sulfite) was proposed, which exhibited considerable potential for application in the treatment of FQs contaminated water.

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.

Acknowledgments

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.cclet.2021.06.036.

References

[1] W. Xu, G. Zhang, X. Li, et al., Water Res. 41 (2007) 4526–4534.
[2] Q.W. Yiruhan, Q. Wang, C. Mo, et al., Environ. Pollut. 158 (2010) 2350–2358.
[3] Q. Wang, Q. Xue, T. Chen, et al., Chin. Chem. Lett. 32 (2021) 609–619.
[4] M. Goñi-Urriza, M. Capdepuy, C. Arpin, et al., Appl. Environ. Microbiol. 66 (2000) 125–132.
[5] T. Iwane, T. Urase, K. Yamamoto, Water Sci. Technol. 43 (2001) 91–99.
[6] A.M. Christensen, F. Ingerslev, A. Baun, Environ. Toxicol. Chem. 25 (2006) 2208–2215.
[7] B. Quinn, F. Gagné, C. Blaise, Sci. Total Environ. 389 (2008) 306–314.
[8] J. Jiang, S. Pang, J. Ma, Environ. Sci. Technol. 43 (2009) 8326–8331.
[9] J. Jiang, S. Pang, J. Ma, et al., Environ. Sci. Technol. 46 (2012) 1774–1781.
[10] J. Jiang, Y. Gao, S. Pang, et al., Environ. Sci. Technol. 48 (2014) 10850–10858.
[11] S. Pang, J. Jiang, Y. Gao, et al., Environ. Sci. Technol. 48 (2013) 615–623.
[12] R.H. Waldemer, P.G. Tratnyek, Environ. Sci. Technol. 40 (2006) 1055–1061.
[13] S. Pang, J. Duan, Y. Zhou, et al., Chemosphere 235 (2019) 104–112.
[14] J. Jiang, Y. Gao, S. Pang, et al., Environ. Sci. Technol. 49 (2014) 520–528.
[15] S.Y. Pang, Q. Wang, J. Jiang, Environ. Sci. Technol. 48 (2014) 6518–6519.
[16] L. Hu, H.M. Martin, T.J. Strathmann, Environ. Sci. Technol. 44 (2010) 6416–6422.
[17] C. Yang, M. Zheng, Y. Zhang, et al., Chin. Chem. Lett. 31 (2020) 2693–2697.
[18] A. Zhu, Y. Guo, G. Liu, et al., Chin. Chem. Lett. 30 (2019) 2241–2244.
[19] Y. Chen, Y. Guo, G. Liu, et al., Chin. Chem. Lett. 31 (2020) 1970–1973.
[20] X. Liu, H. Lv, H. Xu, Chin. Chem. Lett. 26 (2015) 205–209.
[21] B. Sun, J. Zhang, J. Du, et al., Environ. Sci. Technol. 47 (2013) 14332–14340.
[22] K. Xu, W. Ben, W. Ling, et al., Water Res. 123 (2017) 67–74.
[23] J. Jiang, S. Pang, J. Ma, Environ. Sci. Technol. 44 (2010) 4270–4275.
[24] W. Chen, Y. Ding, C.T. Johnston, et al., Environ. Sci. Technol. 44 (2010) 4486–4492.
[25] K. Lin, W. Liu, J. Gan, Environ. Sci. Technol. 43 (2009) 3860–3864.
[26] K. Lin, C. Yan, J. Gan, Environ. Sci. Technol. 48 (2013) 263–271.
[27] H. Zhang, C. Huang, Environ. Sci. Technol. 39 (2005) 4474–4483.

[28] H. Zhang, W.R. Chen, C.H. Huang, Environ. Sci. Technol. 42 (2008) 5548–5554.
[29] L. Hu, A.M. Stemig, K.H. Wammer, et al., Environ. Sci. Technol. 45 (2011) 3635–3642.
[30] Y. Gao, Y. Zhou, S. Pang, et al., Environ. Sci. Technol. 53 (2019) 3689–3696.
[31] Y. Gao, J. Jiang, Y. Zhou, et al., Chem. Eng. J. 327 (2017) 418–422.
[32] B. Sun, X. Guan, J. Fang, et al., Environ. Sci. Technol. 49 (2015) 12414–12421.
[33] Y. Zhu, J. Ling, L. Li, et al., Chin. Chem. Lett. 31 (2020) 1545–1549.
[34] H. Kim, H. Lee, H. Kim, et al., Water Res. 86 (2015) 66–73.