

Degradation of Sulfonamides by UV/Electrochemical Oxidation and the Effects of Treated Effluents on the Bacterial Community in Surface Water

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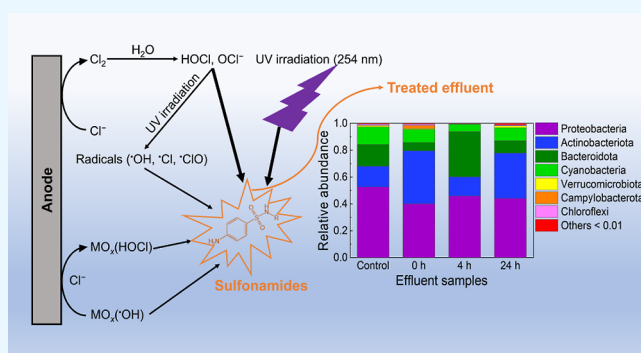
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ABSTRACT: This study evaluated the effects of ultraviolet (UV) photolysis combined with electrochemical oxidation on sulfonamides (SAs) as well as its treated effluent on the bacterial community in surface water. In terms of degradation rate, the best anode material for electrochemical oxidation was Ti/RuO₂–IrO₂, which had the highest degradation kinetic constant compared to Ti/Ta₂O₅–IrO₂ and Ti/Pt. Experiments showed the highest degradation rate of SAs at 8.3 pH. Similarly, increasing the current leads to stronger degradation due to the promotion of free chlorine production, and its energy consumption rate decreases slightly from 73 to 67 W h/mmol. Compared with tap water, the kinetic constants decreased by 20–62% for SAs in three different surface water samples, which was related to the decrease in free chlorine.

When extending the reaction time to 24 h, the concentrations of chemical oxygen demand and total organic carbon decreased by approximately 30–40%, indicating that the SAs and their products could be mineralized. The diversity analysis showed that the effluents influenced the richness and diversity of the bacterial community, particularly in the 4 h sample. Additionally, there were 86 operational taxonomic units common to all samples, excluding the 4 h sample; significant differences were derived from changes in the *Actinobacteriota* and *Bacteroidota* phyla. The toxicity of the products might explain these changes, and these products could be mineralized, as observed in the 24 h sample. Therefore, the extension of treatment time would greatly reduce the ecological harm of treated effluent and ensure that the UV/electrochemical process is a feasible treatment option. Overall, this study provides valuable insight into the optimization and feasibility of UV/electrochemical processes as a sustainable treatment option for sulfonamide-contaminated water sources, emphasizing the importance of considering ecological impacts and the need for extended treatment times that address environmental concerns and ensuring improved water quality.



INTRODUCTION

In recent decades, the widespread use of antibiotics and the risks associated with their overuse or misuse have become public concerns. Antibiotics are generally detected in the range of several ng/L to several μg/L in sewage treatment plant effluents, surface water, and groundwater.¹ Antibiotics are widely used in livestock breeding and aquaculture, as well as the treatment of human diseases, because of their suitability for controlling diseases caused by microorganisms.² Antibiotic pollution is mainly attributed to the discharge of sewage treatment plants, hospital wastewater, and industrial wastewater.³ Its prevalence and potential effects on human and ecosystem health have received considerable attention. The inhibitory effects of antibiotics on bacteria may lead to the loss of some bacterial populations and their ecological functions, which is problematic because bacterial diversity is particularly important for maintaining ecosystem balance.⁴ Antibiotics have even been detected in tap water.⁵ The prolonged

presence of antibiotics in the environment can result in the development of antibiotic-resistant bacteria.⁶ The spread of antibiotic resistance may lead to increasing antibiotic resistance in environmental microbiota,⁷ including the human microbiome, which poses a threat to the various components of the ecological cycle.

As reported, after the conventional treatment process in wastewater treatment plants, the level of tetracycline antibiotics in wastewater can be reduced by 80%, whereas the level of SAs can be reduced by approximately 20%.⁸ Thus, some antibiotics remain in the water after purification by conventional

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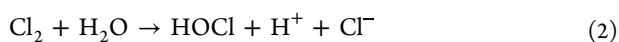
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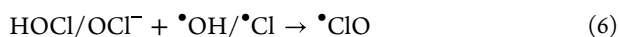
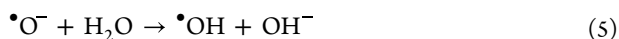
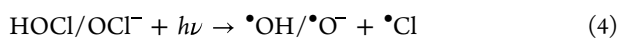


treatment processes, and they will subsequently enter the environment with the effluent.⁹ Residual antibiotics in the effluent cannot be effectively removed by conventional biological treatment.¹⁰ Because antibiotics are easily transported in the environment, almost all surface water contains trace amounts of antibiotics. SAs were the first truly effective, broad-spectrum antimicrobials in clinical use; they remain in use today.¹¹ SAs can accumulate in organisms, leading to the growth of bacterial resistance genes; the bacterial community structure may be affected, and the ecosystem could be harmed. Therefore, it is vital to find an efficient, low-cost, and easily operable treatment method.

As an environmentally friendly treatment technology, the electrochemical advanced oxidation process (EAOP) has been widely used for the treatment and control of micropollutants, such as antibiotics.^{12–14} In addition, ultraviolet (UV) photolysis is also effective for these pollutants.^{15,16} UV at 254 nm is reported to induce higher free radical concentrations.^{17,18} Ultraviolet (UV) photolysis combined with electrochemical oxidation has shown a good effect.¹⁹ In the presence of chloride ions (eqs 1 and 2), free chlorine can be produced²⁰



where the HOCl/OCl[−] ratio depends on the solution pH (eq 3). When UV irradiation is applied (eqs 4–6), the free chlorine can be converted into reactive species²¹



The degradation efficiency of the UV/chlorine process can be very high because of the production of reactive substances.^{22,23} These reactive species can also react with pollutants and enhance degradation efficiency; the exact effects are dependent on the pollutant species.²⁴ Additionally, the type of anode material used for free chlorine electro-generation is important; a lower oxygen evolution overpotential usually results in better chlorine evolution, leading to more free chlorine electro-generation and conversion into reactive chlorine species (RCS). Due to their excellent catalytic activity in chlorination reactions in EAOP, dimensionally stable anodes (DSA, such as Ti/RuO₂–IrO₂, Ti/Ta₂O₅–IrO₂, etc.) are highly preferred by researchers.^{25,26} The advantages of using DSA electrodes for organic degradation include their long run time, ease of preparation, versatility, and high catalytic activity, not to mention the possibility of improving oxidation efficiency through coupling and process optimization.²⁷ Unfortunately, the presence of free chlorine in the UV/chlorine water treatment process inevitably leads to the formation of disinfection byproducts (DBPs, e.g., trihalomethanes and haloacetic acids). Even under UV, there is concern that reactive chlorine, such as $\cdot\text{ClO}/\cdot\text{Cl}$, can promote halogenated DBP production.²⁸ These substances, due to their carcinogenic risk to humans, need to be carefully evaluated before large-scale application. Concurrently, because of the diversity and complex composition of real water samples, the differences in

treatment effects can be substantial.²⁹ Therefore, it is useful to assess treatment effectiveness with different types of water samples. Additionally, the UV/EAOP method promotes the formation of DBPs in the presence of chloride ions, which may include toxic materials. Although this method may be effective for SA treatment, the toxicity of the parent materials and their products cannot be ignored.³⁰ Previous studies have demonstrated the negative effects of sulfamethazine (SMZ) and sulfamethoxazole (SMX) on bacterial diversity in drinking water, with a significant decrease in bacterial species, community richness, and community evenness upon their addition.³¹ However, there are few reports on the effects of SA on bacterial communities after treatment. Thus, there is a need to evaluate the influence of SA-containing wastewater and its treated effluent on the environment. To the best of our knowledge, the use of UV/EAOP for the treatment of SA wastewater has not been studied enough considering the effects of its effluent on bacteria. This is due to the fact that more free and active chlorine occurs in this treatment process, which has no less effect on bacteria than SAs. Also, the possible transformation, degradation, and competition between the three make the effect of the treated effluent on bacteria more complex and uncertain.

In this study, we used a combination of UV₂₅₄ photolysis and electrochemical oxidation to evaluate the effects of various factors on SA degradation, including anode material, current magnitude, pH, and UV power. Different types of water samples were also used to evaluate the treatment effect in order to make the experiment more informative. Furthermore, flat ceramic microfiltration membranes were used in the reactor to provide filtered effluent and evaluate the influence of the effluent on bacteria in the surface water environment. Therefore, this study will provide more ideas and insights for the further development and practical application of advanced oxidation processes.

MATERIALS AND METHODS

Chemical Reagents. The veterinary SAs of sulfamethazine and sulfamonomethoxine tablets (Shandong Ease Animal Pharmaceutical, China) and compound sulfamethoxazole tablets (Shandong Zhengmu Biological Pharmaceutical, China) were used as the target reactants. The standard solution of SAs (analytical reagents, Shanghai Aladdin Biochemical Technology, China) was used to determine the real concentration. Ultrapure/deionized water was prepared with an ultrapure water purification system (Chengdu Ultrapure Technology, China). The sodium hydroxide and concentrated sulfuric acid were used to adjust the pH of the solution, and sodium chloride was used to provide chloride ions (all purchased from Sinopharm Chemical Reagent with analytical reagents, China). The mobile phase was prepared using formic acid (analytical reagents, Sinopharm Chemical Reagent, China) and acetonitrile (HPLC, Tedia, USA).

Materials. The reactor (Figure 1) was custom-built from polypropylene (62 cm × 42 cm × 42 cm). Each UV lamp (TUV 8W/G8 T5, Philips, Poland) was equipped with a quartz glass water jacket. For the mesh electrode (20 cm × 21 cm; thickness, 2 mm), three different anode materials were used (Ti/RuO₂–IrO₂, Ti/Ta₂O₅–IrO₂, and Ti/Pt); titanium (Ti) was used for the cathode (all electrodes were obtained from Baoji Changli Special Metal, China), which was provided with a direct-current power supply (SD-500H-24, Hangzhou Kerui Electronics, China). The distance between the cathode

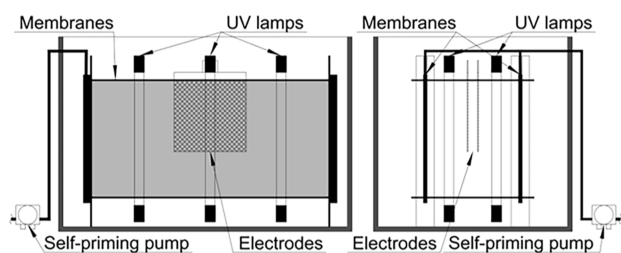


Figure 1. Schematic of the reactor (front and side views).

and anode is 2 cm. Microfiltration membranes (500 mm × 250 mm × 6 mm, strainer aperture of 0.1 μm, Shandong Sicer Membrane Materials, China) were used on both sides. The effluent was pumped with a self-priming pump (JR-7442, Ningbo Jinrui Electronic Commerce, China) from the microfiltration membrane for subsequent analysis.

Experimental Method. The water samples (tap water and three surface water) were prepared with each veterinary SA present at 10 μmol/L to represent simulated wastewater, which was similar to our previous work.³² The chloride ion (Cl⁻) concentration was controlled by the addition of sodium chloride (NaCl, Sinopharm Chemical Reagent, China, ≥99.8%). The UV power was provided by six UV lamps (each lamp = 8 W), and the power was controlled by the number of lamps that were in operation. The average UV fluence rate (E_p^0) was determined to be 0.250 mW/cm² by a UV radiometer (OHSP350UVS, Hangzhou Hongpu Photochromic Technology Co., Ltd, China). The constant current was regulated by the direct-current power supply. The reaction lasted for 2 h; treated effluent was filtered through the membranes with a self-priming pump, then sampled for analysis. The temperature in all experiments was in the range of 18 ± 2 °C, and the initial pH was approximately 8.3. The pH is regulated by sulfuric acid (H₂SO₄, Kelong, China, ≥95%) and sodium hydroxide (NaOH, Sinopharm Chemical Reagent, China, ≥96%).

The treated effluent used for bacterial community diversity analysis was prepared with tap water. The experimental conditions were as follows: Cl⁻ concentration, 50 mg/L; UV power, 48 W; and current, 1.8 A. The reaction time was extended to 24 h; samples were collected at 0, 4, and 24 h. The effluent samples were mixed with surface water at a volume ratio of 1:1, and deionized water was used as the control sample. Each sample was analyzed in triplicate and incubated at room temperature for >24 h. Later, the bacteria in the samples were collected with a glass fiber filter membrane (0.22 μm, Jinten, China) and cryopreservation (<-18 °C).

Analytical Method. SAs were identified by high-performance liquid chromatography (Essentia LC-16, Shimadzu, Japan) with an ACE Excel SAQ column (5 μm, 250 mm × 4.6 mm, Advanced Chromatography Technologies, UK) for separation. The column was maintained at 30 °C with a flow rate of 1 mL/min. Mobile phase A was 20% acetonitrile solution, and mobile phase B was 0.2% formic acid solution; an equal flow rate (A/B: 50%/50%) was used throughout the assay. The sample volume was 20 μL and the detection wavelength was 258 nm; measurements were made with a UV-vis spectrophotometer (UV-2550, Shimadzu, Japan). The Cl⁻ concentration was determined using an ion meter (Seven Compact S220, Mettler Toledo, China). The pH of the solution was determined by a portable pH meter (Smart Sensor PH828, Dongguan Wanchuang Electronic Products, China). Free chlorine was determined using the Hach DPD method 8021 with a spectrophotometer (DR2800, Hach Co., Ltd., Shanghai, China). Chemical oxygen demand (COD), ammonia nitrogen (NH₃-N), and suspended solids were measured by the Hach methods with a Hach spectrophotometer (DR2800, Hach, China). Total organic carbon (TOC) was determined by a TOC analyzer (TOC-2000, Shanghai Metash Instruments, China). Total nitrogen and total phosphorus were determined with a continuous flow analyzer (FlowSys, Systea S.p.A. Analytical Technologies, Italy). The bacteria in the water sample were identified by 16S rRNA gene sequencing analysis; bacterial diversity was analyzed using the Majorbio Cloud Platform (Majorbio BioPharm Technology, China).

The pseudo-first-order kinetic rate constant (k) of SA degradation was calculated as eq 7

$$-\ln(c_t/c_0) = kt \quad (7)$$

where t is the reaction time, c_0 is the initial SA concentration, and c_t is the SA concentration at sampling time t .

RESULTS AND DISCUSSION

Effect of the Anode Material on SA Degradation. As shown in Figure 2a, an anode with strong chlorine evolution had a better SA degradation efficiency. The degradation kinetic constants were in the order Ti/RuO₂-IrO₂ > Ti/Ta₂O₅-IrO₂ > Ti/Pt. The free chlorine concentrations at a reaction time of 120 min were 0.66, 0.15, and 0.07 mg/L, respectively. The Ti/RuO₂-IrO₂ anode had the best free chlorine electro-generation efficiency, with degradation kinetic constants that were 1.5–3.1-fold higher than the degradation kinetic constants of other anodes because of the strong chlorine evolution. This resulted in the generation of more free chlorine

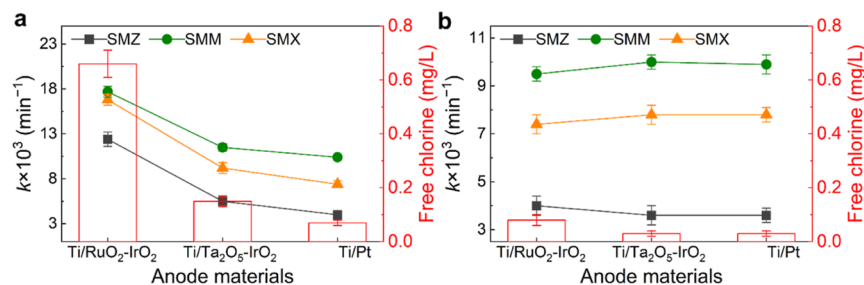


Figure 2. Effect of anode material on SA degradation and the free chlorine concentration at 120 min. The experiment was conducted using tap water and a UV power of 48 W. (a) Cl⁻ concentration, 50 mg/L; current, 1.8 A. (b) Cl⁻ concentration, 20 mg/L; current, 1.0 A.

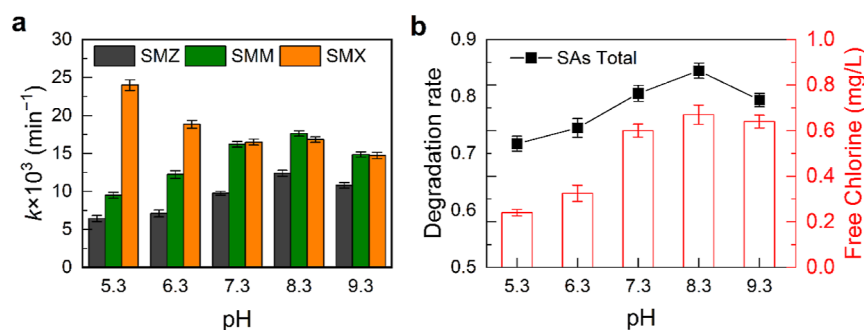


Figure 3. Effect of pH on the degradation of SAs. Experiment using tap water with a Cl^- concentration of 50 mg/L, a UV power of 48 W, and a current of 1.8 A. (a) Degradation rates of different types of SAs. (b) Free chlorine concentration and total degradation rate of SAs at 120 min.

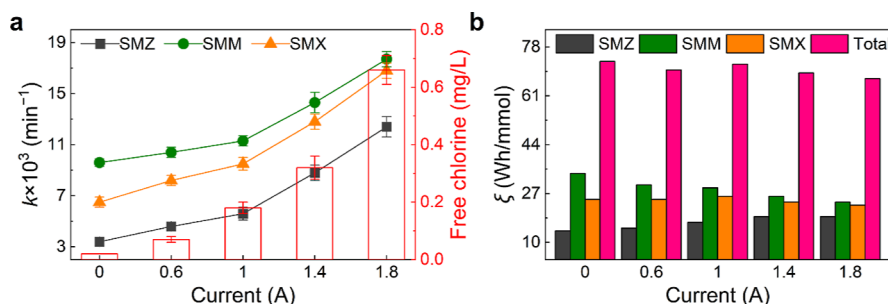


Figure 4. Effect of current on SA degradation. (a) SA degradation rate and free chlorine concentration at 120 min. (b) Energy consumption rate for SA removal. The experiment was conducted using tap water, a Cl^- concentration of 50 mg/L, a UV power of 48 W, and a current that ranged from 0 to 1.8 A (the electrode area available was 376 cm^2).

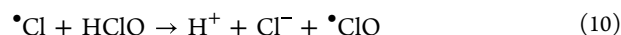
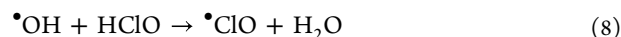
for use in the SA degradation process; therefore, it achieved the best SA degradation efficiency.

Chlorine evolution occurs through intermediates, such as RuO_2 and IrO_2 ; for oxides with specific oxygen adsorption energies, the potential necessary for chlorine evolution is always smaller than the potential necessary for oxygen evolution.³³ This has been demonstrated by experimental studies,³⁴ a lower oxygen evolution potential was associated with a better effect on chlorine evolution in the presence of chloride ions. The oxygen evolution potentials of some base oxides have been reported ($\text{RuO}_2\text{-TiO}_2 = 1.4\text{--}1.7 \text{ V}$, $\text{IrO}_2\text{-Ta}_2\text{O}_5 = 1.5\text{--}1.8 \text{ V}$, and $\text{Pt} = 1.7\text{--}1.9 \text{ V}$),³⁵ and the chlorine evolution potential of $\text{Ti/RuO}_2\text{-IrO}_2$ is better than the chlorine evolution potential of $\text{Ti/Ta}_2\text{O}_5\text{-IrO}_2$.³⁶ These differences explain the chlorine evolution performances of the electrodes in our experiment.

Furthermore, to evaluate the contributions of indirect oxidation for each of the anodes, the experiments were conducted under low chlorine evolution conditions (Figure 2b). The three anodes had an almost identical effect, indicating that the process of indirect oxidation could substantially promote degradation for a specific amount of Cl^- . Therefore, a $\text{Ti/RuO}_2\text{-IrO}_2$ anode and a Cl^- concentration of 50 mg/L were used in subsequent experiments.

Effect of Different pH on the Degradation of SAs. As shown in Figure 3a, the three antibiotics exhibited different degradation kinetic constants under different initial pH conditions. Among them, the degradation of SMZ and SMM showed a consistent change in pH: as the pH increased from 5.3 to 8.3, the degradation rate continued to increase; when the pH continued to increase from 8.3 to 9.3, the degradation rate started to slow down. As seen in Figure 3b, the SA drug's overall degradation exhibits the same trend as that of SMZ and SMX, and the solution's residual free chlorine concentration

performs similarly. We have two speculations about this phenomenon. First, it may be due to the fact that more free chlorine is produced for the degradation of SAs at a pH close to 8.3 after the addition of SAs, and therefore, the degradation rate is highest and the kinetic rate of degradation is maximum at this time. Another possible reason is due to the fact that free chlorine in solution will be more present in solution as OCl^- when the solution is weakly alkaline, and HOCl/OCl^- will be more photolytically converted to $\cdot\text{Cl}$, $\cdot\text{OH}$ under UV.³⁷ At the same time, both HOCl and OCl^- continue to generate more active chlorine species with free radicals, but the kinetic rate of OCl^- generation of active chlorine species is significantly faster (eqs 8–11). Their rate constants are 2.0×10^9 , 8.8×10^9 , 3.0×10^9 , and $8.2 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$, in that order.



In contrast, for the degradation of SMX, there was a general trend toward a gradual decrease with increasing pH. We infer that the cause is due to the selectivity of RCS, such as $\cdot\text{Cl}$ and $\cdot\text{ClO}$, to the contaminants,³⁸ indicating that the selectivity of RCS to SMX decreases as the pH increases. The RCS responded to pH changes in Fang et al.'s experiments on benzoic acid, which are consistent with this phenomenon.³⁹

Effects of Different Currents on SA Degradation. Current density can indirectly influence oxidation efficiency by affecting the amount of free chlorine produced by an EAOP.⁴⁰ As the current increases (i.e., greater current density), the kinetic rate constant of the SAs increases (Figure 4a). This relationship results from the higher yield of free chlorine

the anode surface at higher current densities. Higher degradation kinetic constants were accompanied by higher residual free chlorine concentrations, implying that more free chlorine was also produced and involved in the degradation of SAs. The degradation efficiency was evaluated by the energy consumption rate as eq 12¹⁹

$$\xi = (P_{UV} + P_E)/(\Delta c_{SA} \times V) \quad (12)$$

where ξ is the energy consumption rate of SA removal (W h/mmol), P_{UV} is the power consumption of the UV lamps (W h), P_E is the power consumption of the electrolytic process (W h), Δc_{SA} is the total SA removal in the system (mmol), and V is the volume of the solution (70 L).

Figure 4b shows the energy consumption rates of SA removal at different currents. The total energy consumption rate slightly decreased with increasing current (from 73 to 67 W h/mmol), which implied that the increased power consumption for electrolysis promoted reaction efficiency in this experiment. A higher current (greater current density) implied a higher overvoltage of chlorine evolution and promoted the electro-generation of free chlorine at the anode.¹⁹ Therefore, for the same electrolysis time, an increase in current results in a higher rate of free chlorine production, which also means that more RCS are produced under UV conditions, resulting in a lower energy consumption rate at higher current densities in the experimental results.

As the concentration of free chlorine increases, more RCS ($\cdot\text{Cl}$, $\cdot\text{ClO}$, and $\cdot\text{Cl}_2^-$) are also generated under UV light, while the oxidation of active chlorine species is more selective.⁴¹ Therefore, the different energy consumption rates for different SAs' removal with increasing current can be attributed to the selective oxidation of active chlorine and the complex competition between SAs for free chlorine and RCS.^{42,43} However, how this competitive relationship evolves may be further investigated in our future work due to the complexity of the solution composition.

Effect of UV Power on SA Degradation. The photolysis process generally consists of direct and indirect photolysis. The direct photolysis of SAs considerably varies according to the irradiation source, substrate, and pH, whereas indirect photolysis is mediated by $\cdot\text{OH}$. Direct photolysis generally occurs easily; with high-energy UV (254 nm) irradiation, direct photolysis dominates the process.

The degradation kinetics of SAs under different UV irradiation intensities were investigated by changing the number of UV lamps in operation. As shown in Figure 5,

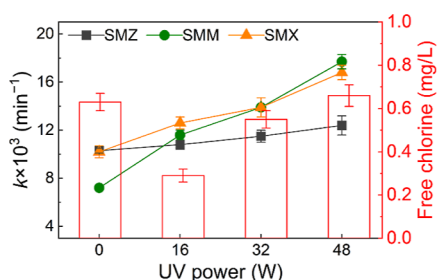


Figure 5. SA degradation rates under different UV irradiation intensities and free chlorine concentrations at 120 min. The experiment was conducted using tap water with a Cl^- concentration of 50 mg/L, a UV power that ranged from 0 to 48 W, and a current of 1.8 A.

the magnitude of UV power affected SA degradation in the order of $\text{SMM} > \text{SMX} > \text{SMZ}$. The reason for this difference may be related to solution pH, irradiation intensity, molecular structure, and SA concentration. Meanwhile, with the increase in UV power, we found that the concentration of free chlorine remaining in the solution first decreased and then increased. This may be due to the photolysis of free chlorine to $\cdot\text{Cl}$ and $\cdot\text{OH}$ with increasing UV intensity (eq 4), leading to a decrease in free chlorine concentration at 16 W. Subsequently, the free chlorine concentration showed a slightly increasing trend when the UV intensity continued to increase. This variation may have been caused by a complex interaction between free chlorine species, reactive intermediates, equilibrium reactions, and specific characteristics of the wastewater matrix.⁴² For example, generated hydroxyl radicals ($\cdot\text{OH}$) and other reactive species can undergo radical-mediated reactions with chloride ions in wastewater. These reactions can lead to the formation of free chlorine species, such as hypochlorous acid (HOCl) and hypochlorite ions (OCl^-), produced from the chlorination of chloride ions. This phenomenon can in turn be attributed to the indirect chlorination process facilitated by the active species produced during UV photolysis. The subsequent $\cdot\text{ClO}$ reaction tends to generate ClO_x that can regenerate free chlorine (eqs 13 and 14).⁴⁴



Effect of Water Quality on SA Degradation. Experiments were conducted with tap water and three types of surface water (SW1, SW2, and SW3); the results are shown in Figure 6, and the water quality data are shown in Table 1. The

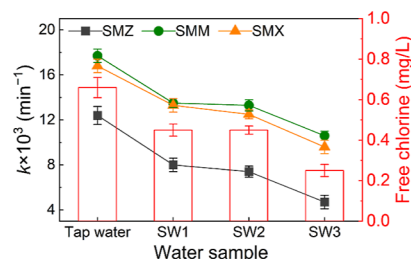


Figure 6. Effects of different water samples on SA degradation and the free chlorine concentration at 120 min. The experiment was conducted using four water samples with different water qualities (Table 2), a Cl^- concentration of 50 mg/L, a UV power of 48 W, and a current of 1.8 A.

degradation kinetic constants in the three surface water samples were all lower than in tap water and decreased by 20–62% for the three SAs, which could be attributed to the interference of organic compounds in the reactions.⁴⁵ Additionally, the kinetic rates corresponded with the residual free chlorine concentration but were the opposite of the $\text{NH}_3\text{-N}$ concentrations in surface water samples. These findings might be related to the involvement of $\text{NH}_3\text{-N}$ in reactions with active species in the UV electrochemical system; this involvement decreases the SA degradation rate. Xiao et al. studied the ammonia nitrogen removal effect in an electrochemical cell combined with a UV irradiation process,⁴⁶ which was similar to our system. The results indicated the free chlorine and chlorine radical ($\cdot\text{Cl}$) in the system could react with NH_4^+ , resulting in a decrease in the quantity of active

Table 1. Water Qualities of Different Water Samples^a

water quality	tap water	SW1	SW2	SW3
total nitrogen (mg/L)	2.6 ± 0.2	2.1 ± 0.2	2.3 ± 0.1	5.2 ± 0.2
total phosphorus (mg/L)	ND	0.05 ± 0.01	ND	0.12 ± 0.02
NH ₃ -N (mg/L)	ND	0.24 ± 0.05	0.44 ± 0.08	2.58 ± 0.15
pH	8.0 ± 0.1	8.2 ± 0.1	8.2 ± 0.2	7.8 ± 0.1
suspended solids (mg/L)	ND	18 ± 2	18 ± 2	19 ± 1
COD (mg/L)	ND	11 ± 1	11 ± 1	11 ± 2
TOC (mg/L)	2.4 ± 0.2	4.2 ± 0.2	4.3 ± 0.2	3.9 ± 0.3

^aNote: ND indicates below the detection limit.

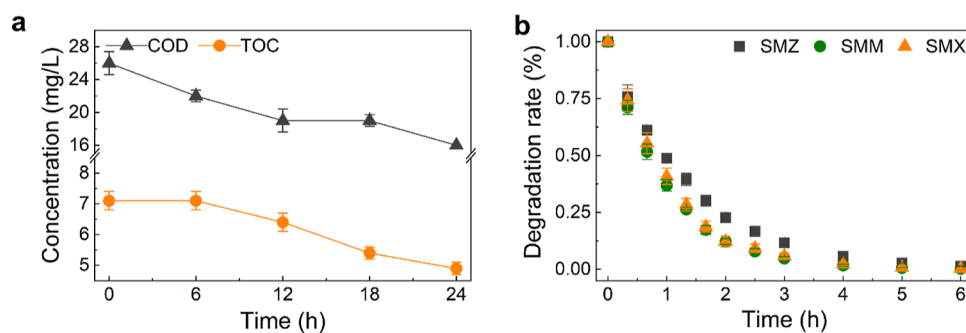


Figure 7. Variations in effluent quality and SA degradation rate. (a) Variations in the COD and the concentration of TOC. (b) SA degradation rate in the reaction. The experiment was conducted using tap water with a Cl⁻ concentration of 50 mg/L, a UV power of 48 W, and a current of 1.8 A.

Table 2. Alpha Diversity Analysis Indices at the OTU (Operational Taxonomic Unit) Level^a

sample	community richness indices		community diversity indices	
	Sobs	Chao1	Shannon	Simpson
control	360 ± 7	451 ± 49	4.04 ± 0.04	0.033 ± 0.002
0 h	348 ± 34	444 ± 10	3.74 ± 0.13	0.049 ± 0.011
4 h	266 ± 31*	327 ± 25**	3.33 ± 0.21***	0.075 ± 0.016***
24 h	396 ± 60	467 ± 52	4.00 ± 0.10	0.035 ± 0.007

^aNotes: One-way analysis of variance with Fisher's least significant difference test compared with the control sample, * $P < 0.05$, ** $P < 0.01$, and *** $P < 0.001$.

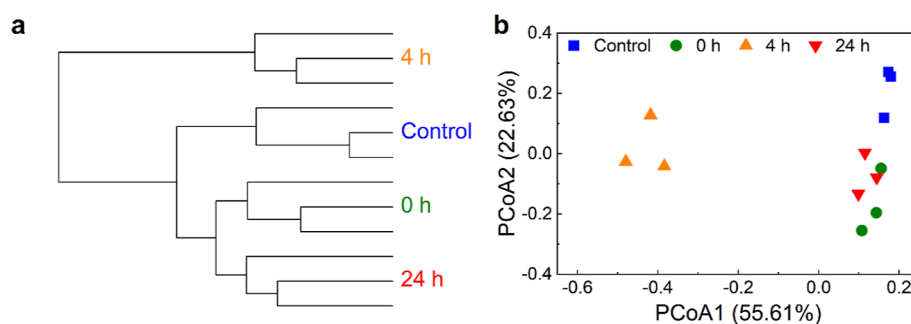


Figure 8. Beta diversity analysis (three replicates) at the OTU level. (a) Hierarchical cluster analysis with Bray–Curtis distances. (b) Principal coordinate analysis with Bray–Curtis distances (analysis of similarities, $r = 0.95$, $P < 0.001$).

species that could react with SAs. The residual free chlorine concentration at 120 min confirmed this suspicion. Additionally, suspended solids could decrease the photo-permeability, which might weaken the effect of UV irradiation in surface water.

Variations in Effluent Quality and SA Degradation Rate. The water quality of SA-containing effluent was evaluated; the COD and the concentration of TOC are shown in Figure 7a. The TOC exhibited a minimal decrease in the first 6 h, indicating that most SAs were in the degradation stage (Figure 7b) and had not been mineralized. The COD

and the concentration of TOC both decreased by approximately 30–40% after 24 h, which indicated that the products of SAs could be mineralized.

Effects of Effluent on Bacterial Diversity and Composition in Surface Water. An alpha diversity analysis was conducted to assess the potential ecological effects of the effluent on surface water. As listed in Table 2, high values of Sobs and Chao1 represent high species richness, whereas high values of Shannon and low values of Simpson represent high species diversity. According to one-way analysis of variance, the richness indices did not statistically differ from the control

sample, except for the 4 h sample. In the 4 h sample, the Sobs and Chao1 indices clearly decreased, indicating that community richness had been affected. However, the Shannon diversity index decreased and the Simpson diversity index increased because SAs were present (0 h sample). This result was consistent with the previous finding that SAs decreased bacterial community diversity.³¹ The change in bacterial diversity was profound at 4 h in the SA-treated samples. Because the three target SAs decreased by >95% after a 4 h reaction (Figure 7b), the results demonstrated that the effects of SA transformation intermediates on bacterial diversity were greater than the effects of the parent SAs. The effects of the treated effluent on bacterial diversity became marginal as the treatment time increased to 24 h, and the bacterial richness and diversity did not change.

The similarities of various samples were determined from the beta diversity analysis (Figure 8). According to the hierarchical cluster analysis (Figure 8a), the 0 and 24 h samples had the highest similarity; both were close to the control sample, and the 4 h sample exhibited the greatest difference from the other samples. According to principal coordinates analysis (Figure 8b), two principal factors (PCoA1 and PCoA2) were dominant among the samples with a ratio of 79.24%. The results clearly showed the distinct nature of the 4 h sample, as well as the higher similarities of the 0 and 24 h samples; their similarity (0 and 24 h samples compared with the 4 h sample) was comparable with the similarity between the control and 4 h samples.

The Venn diagram in Figure 9 reflects the differences in sample compositions. There were 302 OTUs common to all

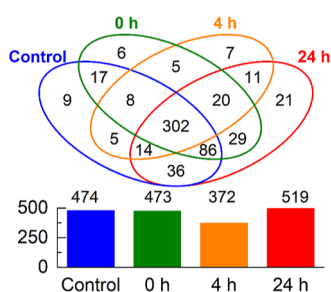


Figure 9. Venn diagram community composition analysis at the OTU level.

samples; only a small number of OTUs were unique to each sample. The three SA-treated effluent samples (0, 4, and 24 h) had 413, 329, and 438 OTUs in common with the control sample, respectively, indicating the 0 and 24 h compositions

still had higher similarity compared to the control sample, whereas the 4 h sample was more different in composition. There were 86 OTUs common to all samples, excluding the 4 h sample; this was presumably because some bacteria were inactivated by SA products in the effluent after 4 h of reaction.

As shown in Figure 10, there were significant differences in Actinobacteriota, Bacteroidota, Campilobacterota, and Bdellovibrionota at the phylum level. The significant differences were derived from changes in Actinobacteriota and Bacteroidota because of the lower relative abundances of the other two phyla. These phyla had similar relative abundances in the 0 and 24 h samples, which were both distinct from the 4 h sample. At the genus level, *Limnohabitans*, *Aurantimicrobium*, *Rhodoluna*, *Pseudomonas*, *Rhodoferax*, and *Saprospiraceae* exhibited significant differences in the four samples; all of these differences were derived from changes in the top four phyla. It has been reported that *Limnohabitans* is the most common genus in wastewater from backyard-based aquaculture systems, which can contain high levels of antibiotic residues (e.g., SAs) and organic nutrients.⁴⁷ Nutrient levels are positively correlated with *Limnohabitans* abundance,⁴⁸ which could be related to the presence of some nutrient ingredients in veterinary SAs, and the SAs/SA products (0 and 4 h samples) could have caused some interference; the 24 h sample had the highest relative abundance. Additionally, high abundances of *Flavobacterium* and *Pseudomonas* were unique to the control sample, with both substantially decreasing in the three SA effluent samples. Similar results have been reported in other studies of wastewater containing SAs,⁴⁹ indicating that these bacteria could be inhibited by SAs and SA products. The abundance of *Pseudomonas* reportedly decreased because of the presence of active species when using an EAOP for water disinfection, which suggests that the slightly residual free chlorine might also be responsible for the significant decrease in this genus. *Aurantimicrobium* and *Rhodoluna* had higher relative abundances in the 0 and 4 h samples, indicating that the high concentrations of SAs and SA products did not have a strong influence on these genera. The antibiotic resistance genes present in river water are not negligible;⁵⁰ *Aurantimicrobium* and *Rhodoluna* may carry antibiotic resistance genes,⁵¹ indicating their potential for greater resistance than other bacteria in our SA effluent samples. The relative abundance of *Rhodoferax* was correlated with the concentrations of SAs and SA products. There was a lower abundance in the three SA samples than in the control sample, particularly in the 4 h sample, which indicated the inhibitory effects of SAs and SA products on *Rhodoferax*. *Saprospiraceae* reportedly has a high relative abundance in domestic wastewater and a lower relative

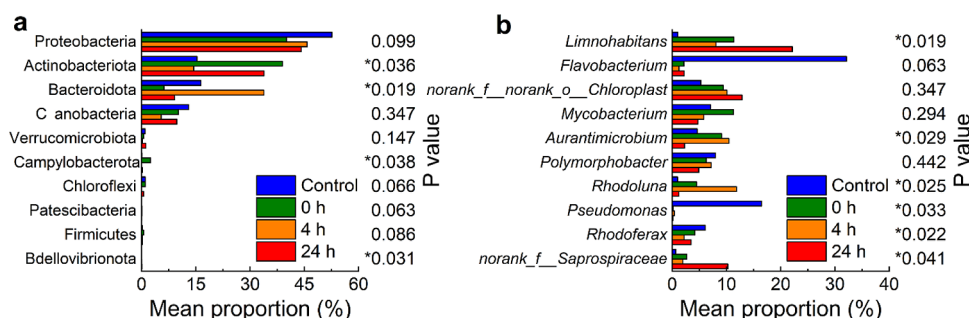


Figure 10. Statistically significant differences in relative abundance (top 10 species) as determined by the Kruskal–Wallis H test. (a) Differences among samples at the phylum level. (b) Differences among samples at the genus level, * $P < 0.05$.

abundance in hospital wastewater, which may be related to high antibiotic concentrations.⁵² Additionally, *Saprosiraceae* is commonly found in various active sludge samples,⁵³ which suggests that nutrients and organic matter could promote its growth. These findings may explain its highest relative abundance in the 24 h sample; all SA samples had higher levels of nutrients associated with veterinary SAs compared with the control sample.

CONCLUSIONS

In this study, we explored the various factors influencing the degradation of SAs using the UV/EAOP approach. Among the anode materials tested, Ti/RuO₂-IrO₂ exhibited the highest degradation kinetic constant, indicating its superior performance in electrochemical oxidation for SA removal. This material outperformed Ti-Ta₂O₅-IrO₂ and Ti/Pt, showing a 1.5- to 3.1-fold increase in the degradation kinetic constant. Furthermore, strong chlorine evolution was achieved in the presence of chloride ions. The optimal pH for SA degradation was found to be 8.3, where the free chlorine concentration was maximized, resulting in the most effective degradation. Increasing the current during the electrochemical oxidation process improved the degradation effect while slightly reducing the energy consumption rate. Notably, different SAs exhibited selective oxidation behavior, which led to distinct degradation patterns at different energy consumption rates. By adjusting the UV power in the UV/electrochemical oxidation process, the degradation order of SAs was determined as SMM > SMX > SMZ. However, the degradation efficiency was observed to be lower in surface water samples compared to tap water, suggesting the influence of surface water organic compounds and ammonia nitrogen (NH₃-N) on the reaction with active species, particularly free chlorine. Assessing the effluent quality and degradation rate over a 24 h reaction period, it was observed that the majority of SAs were still in the degradation stage and had not been fully mineralized. However, at the end of the 24 h period, a significant decrease in COD and TOC concentrations indicated mineralization of the SA products. By investigating the bacterial community, alpha and beta diversity analyses showed that sewage had a significant effect on the abundance and diversity of the bacterial community, with the 4 h sample showing the least diversity. This decrease in diversity could be attributed to the toxicity of the SA products, leading to the inactivation of certain bacteria. Notably, extending the treatment time was found to reduce the ecological harm of the treated effluent and ensure the feasibility of the UV/electrochemical process.

In conclusion, our study proved the success of UV/EAOP as a treatment for SA wastewater. However, surface water samples exhibited lower degradation efficiency due to the presence of organic compounds and ammonia nitrogen. Our findings emphasize the importance of extending treatment time to minimize ecological harm and achieve complete mineralization of SA products. Future research should focus on further optimizing the treatment processes and investigating the long-term effects on the bacterial community in the treated effluent. By advancing our understanding of SA removal methods, we can contribute to the development of sustainable and efficient water treatment strategies.

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▽X.J. and J.Y. contributed equally to this work. Julin Yuan was in charge of literature data collection. The first draft of the manuscript was written by Zhijie Zheng. Xiaogang Wu provided guidance on the conception and writing of the article. Xinwei Jiang was responsible for reviewing and summarizing literature in related fields, and Julin Yuan and Xinwei Jiang made major contributions to writing the manuscript. Grammar corrections were performed by Yufang Tao. All authors read and approved the final manuscript.

Notes

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