Journal of Hazardous Materials Advances 7 (2022) 100139

Contents lists available at ScienceDirect

Journal of Hazardous Materials Advances

journal homepage: www.elsevier.com/locate/hazadv

Advanced treatment of microplastics and antibiotic-containing wastewater using integrated modified dissolved air flotation and pulsed cavitation-impinging stream processes

Yanfeng Zhu[a], Jing Ma[b], Siyan Zeng[c,d], Xiaoxiao Li[e], Grzegorz Lisak[f], Fu Chen[b,*]

[a] School of Environmental Science and Spatial Informatics, China University of Mining and Technology, Xuzhou 221116, China
[b] School of Public Administration, Hohai University, Nanjing, 210098, China
[c] College of Environment and Resource Science, Zhejiang University, Hangzhou 310058, China
[d] Institute of Advanced Technology, Westlake Institute for Advanced Study, Hangzhou 310024, China
[e] School of Agriculture and Biology, Shanghai Jiao Tong University, Shanghai, 200240, China
[f] Residues and Resource Reclamation Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, 1 Cleantech Loop, CleanTech One, 637141, Singapore

ARTICLE INFO

Keywords:
Antibiotic resistance genes
Microplastics
Cavitation cavity
Hydroxyl radicals
Dissolved air flotation

ABSTRACT

Livestock and poultry breeding wastewater is a major reservoir of antibiotic resistance genes (ARGs) and antibiotics residues, and it is also a potential reservoir of microplastics (MPs). In this work, a novel system consisting of modified dissolved air flotation (MDAF) and self-excited oscillating pulsed cavitation-impinging (SOPC) processes was established to explore the removal performance of ARGs, antibiotics and MPs. Polydimethyldiallyl ammonium chloride (PDAC) showed better performance than cetyltrimethylammonium bromide (CTAB) in MDAF. At a saturation pressure of 0.4 MPa and 0.8 mg/L of PDAC, 88.3% of MPs was removed by MDAF. More than 97% of antibiotics and 100% ARGs were removed during the SOPC treatment, and 99.2% of antibiotics were removed by the combined system. The SOPC can stably generate hydroxyl radicals (•OH) in wide pH ranges without additional chemicals. This study demonstrates that the hybrid system has the potential to be applied for the advanced treatment of wastewater containing ARGs and MPs.

1. Introduction

In order to enhance a disease resistance and a growth of livestock and poultry, large amounts of broad-spectrum antibiotics such as aureomycin, oxytetracycline and tetracycline have been added to the feed. In China, approximately 90 000 tons of antibiotics are used in feed additives every year (Hvistendahl, 2012). Nevertheless, the antibiotics are not entirely metabolized by livestock and poultry, and about 60%–90% of antibiotics are excreted with feces and urine in the forms of parent compounds or primary metabolites (Tian et al., 2021).

Currently, antibiotics-related environmental pollution has become a major global concern; especially the abuse of antibiotics in the livestock and poultry industries (Tian et al., 2021). Subsequently, antibiotics in the environment will not only cause direct ecotoxicological hazards but may also induce antibiotic resistance genes (ARGs), which can seriously endanger the ecological safety (Zhang et al., 2020). Unfortunately, there is no restriction for antibiotics in the National Discharge Standard of Pollutants for Livestock and Poultry Breeding (GB 18,596–2001). Moreover, the existing technologies such as constructed wetlands, oxidation ponds, activated sludge processes cannot effectively remove antibiotics in livestock and poultry breeding wastewater (LPBW), resulting in various antibiotics being frequently detected in significant concentrations in the discharged effluent of LPBW treatment systems (Tullo et al., 2019). Thereupon, there is an urgent need to develop effective approaches to remove antibiotics and ARGs from LPBW.

Thompson et al. (2004) found plastic fragments in marine water and sediments, microplastics (MPs). Plastic fragments or particles with size less than 5 mm are generally defined as MPs, which are now considered as a new type of potential persistent environmental pollutants. The transport process of MPs in the environment is often accompanied by the enrichment, migration and release of organic pollutants, just like the "Trojan-Horse". The "Trojan-Horse" effect may significantly alter the potential health risks and bioavailability of MPs and organic pollutants (Zhang and Xu, 2020). Recently, the presence of MPs in livestock and poultry manure has been verified (Wu et al., 2021a; Wu et al., 2021a, b), which may originate from pipes/plastic bags for feed transport and loading, water faucets and bowls, manure scrapers, and plastic bottles for storing disinfectants and/or antibiotics. Thus, LPBW can suf-

* Corresponding author.
E-mail address: chenfu@cumt.edu.cn (F. Chen).

https://doi.org/10.1016/j.hazadv.2022.100139
Received 11 July 2022; Received in revised form 27 July 2022; Accepted 2 August 2022

fer co-contamination of MPs and antibiotics, which may exacerbate their respective ecological and environmental risks.

The antibiotics residues in wastewater are persistent and difficult to be degraded by conventional biotreatment methods (Wang and Zhuan, 2020). Physical methods such as adsorption and vacuum membrane distillation have shown good removal performance for antibiotics from water and wastewater (Yadav et al., 2021; Patel and Yadav, 2022, Yadav et al., 2022a, b). Advanced oxidation processes (AOPs) constitute a promising technology for treating wastewaters containing antibiotics. AOPs such as persulfate oxidation, ozonation, photocatalysis, ultraviolet and Fenton/photo-Fenton processes have been recommended as a good treatment process for the removal of recalcitrant pollutants including antibiotics in water and wastewater (Anjali and Shanthakumar, 2019; Wang and Zhuan, 2020). Nevertheless, photo-oxidation degradation is inefficient for high-turbidity and/or dark-color wastewaters due to the low penetration of light, whereas exogenous reagents are generally required for some AOPs during wastewater treatment. This may increase the operating costs and generate secondary pollution.

Recently, a novel technology, cavitation-impinging stream (CIS) is being developed and utilized for wastewater treatment (Dhanke et al., 2019; Li et al., 2022). This technology exhibited advantages in water treatment such as multiple oxidation pathways (high temperature decomposition, free radical oxidation and supercritical water oxidation) and enhanced surface oxidation reaction (Li et al., 2022). Moreover, no additional reagents are needed by CIS. Li et al. (2022) used the combination of CIS and other methods to treat highly polluted oilfield wastewater with excellent performance. Thus, CIS is a potential technology for advanced water treatment.

In this work, a novel hybrid system consisting of a modified dissolved air flotation (MDAF) process and a self-excited oscillating pulsed CIS (SOPC) process was developed for the advanced treatment of an actual LPBW. The influences of main operational parameters on the performance were studied and optimized. The main purpose of this study was to effectively remove the residues of MPs, antibiotics and ARGs in LPBW. To the best of the authors' knowledge, this is the first research employing the MDAF-SOPC technology for advanced treatment of antibiotic-MPs containing wastewater.

2. Materials and methods

2.1. Wastewater

The LPBW studied in this study was obtained from a large-scale pig farm, which was founded in 2004 with an annual pig breeding stock of more than 10,000, covering an area of about 41 hm^2. In the farm, the rainwater-sewage diversion system is in operation in the breeding area, where the rainwater and sewage are discharged separately. Some of the feces are transferred to the stacking shed, temporarily stored and then are being sold to a local organic fertilizer plant as composting materials, while other feces are being sent to the collection tank as raw fermentation materials. Urine, flushing water and portion of feces were introduced into two-stage anaerobic fermentation tanks for fermentation treatment, followed by the aquatic plant pond treatment. The final effluent was collected, stored at 4 °C before used in this study. The wastewater had the following characteristics: pH, 6.4–6.5; chemical oxygen demand (COD), 92–106 mg/L; total suspended solids (TSS), 43–55 mg/L, tetracycline (2.103–2.535 μg/L), oxytetracycline (0.353–0.575 μg/L), chlorotetracycline (0.652–0.820 μg/L), and sulfonamides (0.856–1.106 μg/L). The total concentration of tetracycline, oxytetracycline, chlorotetracycline and sulfonamides was 3.94–4.75 μg/L. It could be found that the LPBW still contained significant amounts of various antibiotics after a series of treatments. Although current wastewater treatment methods (such as anaerobic digestion, constructed wetland, etc.) can effectively reduce the concentration of COD, nitrogen and phosphorus in LPBW, the concentration of antibiotics in the effluent can still be high (Chen et al., 2012). Thus, further treatment is needed.

2.2. Experimental apparatus

The structure diagram of the developed SOPC system is demonstrated in Fig. 1. The stainless steel-made reactor has a working volume of 3.5 L. As shown in Fig. 1, the reactor consists of six liquid inlets evenly distributed around the cylindrical impact cavity.

The reaction mechanism is as follows. Water streams enter the cavitation cavity through the inlets, and then strong cavitation is formed in the cavitation cavity. The cavitation bubbles collapse rapidly to form a high-temperature and high-pressure environment, thus the steam is cracked in this circumstance to produce hydroxyl radical (•OH) with strong oxidation capacity, and a pulsed cavitation jet will be generated at the outlets. The cavitating jet converges and collides at the center of the impact cavity. The non-collapsed bubbles in the cavitating jet split into microbubbles in the collision zone, increasing the surface area and surface energy of the bubbles and thus promoting the surface oxidation reaction. In particular, the strong micromixing in the collision zone increases the contact probability between reactants. The pressure fluctuation will alter molecular energy and its distribution, effectively strengthen the cavitation, intensify the formation, growth and collapse of cavitation bubbles, and accelerate the chemical bond scission of water molecules in the cavitation bubbles, thus leading to the increased yield of •OH.

2.3. Experimental procedures

The wastewater was first treated by dissolved air flotation (DAF) for a purpose to remove MPs. Considering the low concentration of MPs in real water body, jar tests were conducted to remove MPs in a 50 L Plexiglass barrel containing 35-L LPBW. For conventional DAF (CDAF) Water containing dissolved gas (saturation pressure 0.1–0.5 MPa) was passed into LPBW and allowed to settle down for 30 min. Then about 500 mL supernatant was withdrawn and transferred into a beaker, allowed to sit for 10 h to promote defoaming (this solution was used for MPs measurement). For MDAF, two bubble modifiers, cetyltrimethylammonium bromide (CTAB) or polydimethyldiallyl ammonium chloride (PDAC) were added to the saturator at a dosage of 0.4–1.2 mg/L, and a saturator pressure of 0.4 MPa. The other procedures were the same as those for MDAF.

To investigate the influence of operating parameters on the performance of the SOPC reactor, single-factor experiments were carried out at different values of pH (3, 4, 5, 6.4, 7, 8, and 9), reaction time (0–30 min), and pumping pressure (0.02–0.1 MPa). The reactor was operated in a batch mode at room temperature (25 ± 3 °C), wherein high-pressure airflow was continuously introduced into the reactor to retain the reaction. At regular intervals, the content of the reactor was withdrawn and allowed to settle for 30 min before analysis.

2.4. Analytical methods

A laser particle analyzer (Mastersizer 2000, Malvern Instruments, Ltd., UK) was used to determine the concentration of MPs in samples. COD and TSS were measured according to the standard methods (APHA, 1998). The type of MPs was identified by using attenuated total reflection Fourier transform infrared (ATR-FT-IR) analysis (Eqinox 55, Bruker Optics, Billerica, MA, USA). Moreover, MNPs were counted using Nano Measurer 1.2 software. The •OH radical concentration was measured by the coumarin fluorescence probing technique (Yamaguchi et al., 2018). Antibiotics were detected using high performance liquid chromatography tandem mass spectrometry (HPLC-MS/MS) as described previously (Chen et al., 2022). Seven ARGs (*tet*M, *tet*O, *tet*Q, *tet*W, *sul*I, *sul*II, and *int*I1) were identified and quantified using published primers as reported previously (Chen et al., 2022).

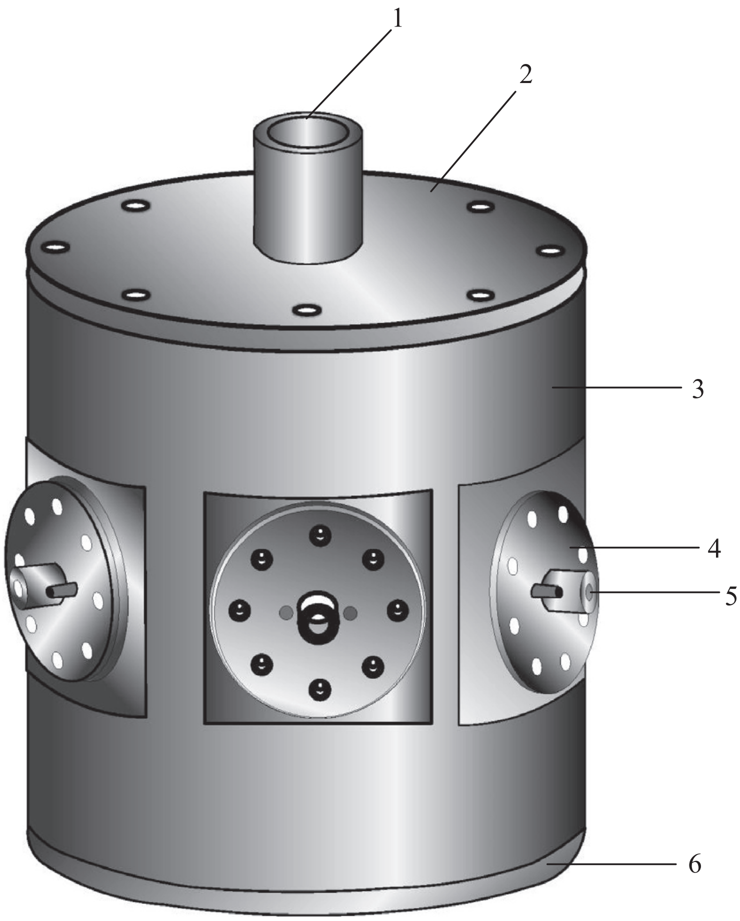

Fig. 1. Structure diagram of the developed SOPC system. (1) Liquid outlet; (2) upper cover plate; (3) wall plate; (4) flange plate; (5) liquid inlet; (6) lower cover plate.

Table 1
Overall polymer distribution (percentage ≥50 μm particles) analyzed by ATR-FT-IR spectroscopy.

Species	PP	PE	PA	PS	PET	PVF	Others
Percentage (%)	19.3 ± 2.1	14.5 ± 0.80	8.2 ± 0.75	6.3 ± 1.6	4.1 ± 0.57	2.5 ± 0.34	45.1 ± 0.55

2.5. Statistical analysis

Statistical analysis was carried out by SPSS software (version 21.0). One-way ANOVA was employed to identify differences between replicates groups with a $P < 0.05$.

3. Results and discussion

3.1. Removal of MPs by CDAF

CDAF has been widely used to remove suspended solids and particles (Pooja et al., 2021; Hasannattaj Jelodar et al., 2022), and recently was applied to MPs removal from water bodies (Ahmed et al., 2021; Esfandiari and Mowla, 2021). Thus, the performance of CDAF on MPs removal from livestock and poultry wastewater was first investigated in this work. The composition of the polymers for ≥50 μm particles is indicated in Table 1. A total of six MPs were detected: polypropylene (PP), polyethylene (PE), polyamide (PA), polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl fluoride (PVF). From the 342 particles enumerated, 187 particles were identified, with a 47.9% identification rate. Identification of all the particles was impossible because of the missing and destruction of particles during sample preparation, and the inability to visualize all the particles with the naked eye (Ahmed et al., 2021; Esfandiari and Mowla, 2021; Kankanige and Babel, 2020).

Fig. 2A shows the removal of various MPs species under different saturation pressures. As shown, the removal efficiencies of PP and PE are both maximized at 0.3 MPa (47.6% and 45.7% respectively), then remain constant with increasing saturation pressure. PA removal increases with saturation pressure until 0.5 MPa, while other three MPs reach their highest removal ratios at 0.4 MPa. Among six MPs, the removal efficiency of PA was the lowest (16.8%). PP and PE particles have good hydrophobicity with lower density compared to water, thus they are easily to adhere to microbubbles in water and float up to the water surface (Swart et al., 2022). Although the density of PET is higher than that of water, PET particles show rough surface and strong hydrophobicity, leading to moderate removal (33.2% at 0.4 MPa) by CDAF. PA contains hydrophilic amide groups (Zhang and Chen, 2020), so it is not easy to adhere to microbubbles, resulting in low removal by CDAF. It can be seen that the removal efficiency of MPs by CDAF is greatly affected by the hydrophilic/hydrophobic property of MPs particles.

Fig. 2B shows the removal of total MPs by CDAF under different saturation pressures. It can be found that the removal of total MPs is highest (40.1%) at 0.4 MPa. The removal efficiency of various sized MPs increases with particle sizes at 0.4 MPa (Fig. 2C). For example, 2–5 μm MPs was removed by 21.3% relative to 53.8% removal of 20–25 μm MPs (Fig. 2C). Previous studies have shown that the closer the size of floc particle is to that of bubbles, the higher the probability of collision and adhesion between them (Qi et al., 2018; Wang et al., 2022). The size

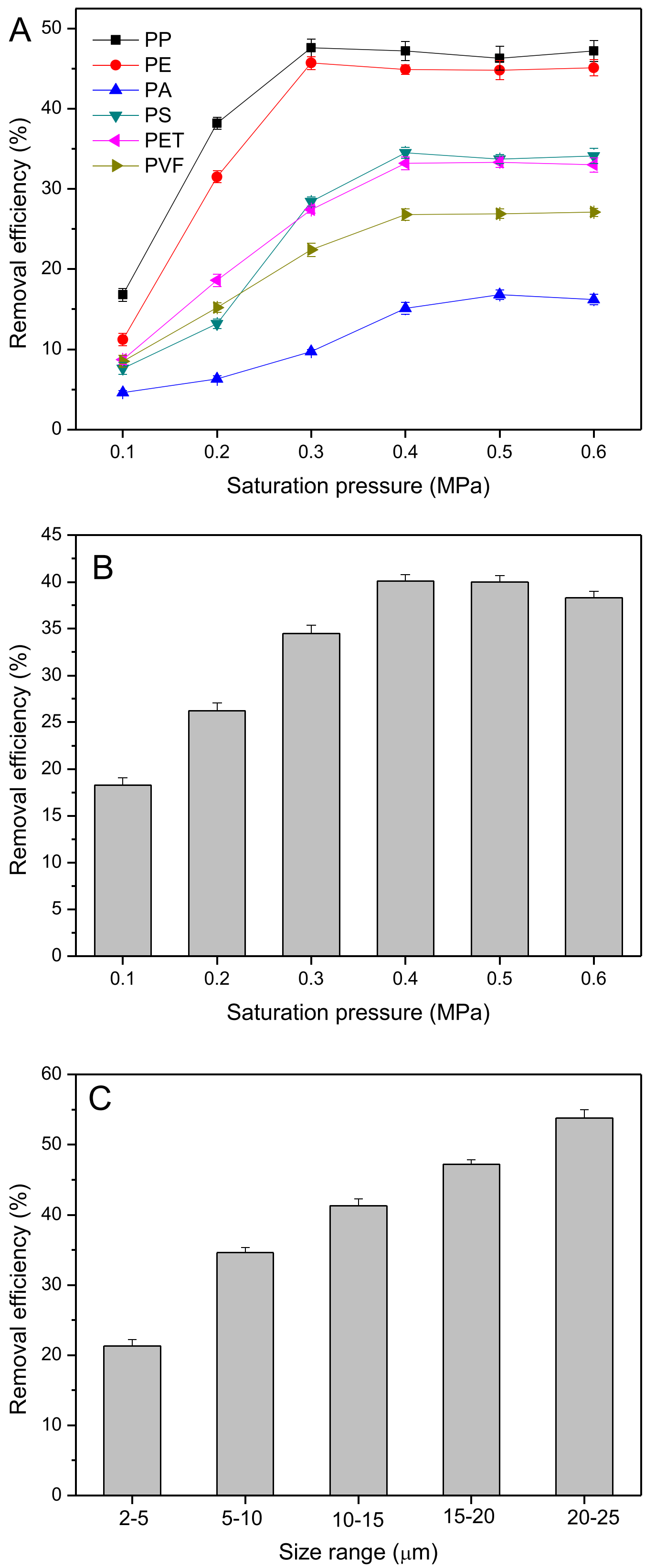

Fig. 2. Removal efficiency of various types of MPs (A), total MPs (B) and various sized MPs at 0.4 MPa by CDAF.

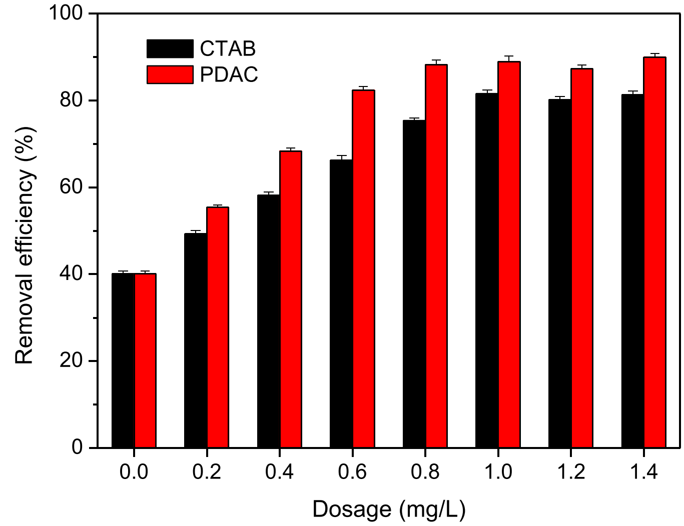

Fig. 3. Effect of the dosage of CTAB and PDAC on the removal of total MPs by MDAF at a saturation pressure of 0.4 MPa.

of microbubbles is generally 20–40 μm. Therefore, the removal ratio of 20–25 μm MPs is higher than that of other groups due to their similar sizes with microbubbles.

3.2. Removal of MPs by MDAF

Although a part of MPs could be removed in water by MDAF, the removal efficiency was not ideal, thus MDAF was used to improve the removal performance. Previous research demonstrated that CTAB and PDAC had good modification effect on microbubbles (Wang et al., 2021). Thus, these two agents were used to enhance the removal of MPs through the MDAF process at 0.4 MPa based on total removal ratio and the removal of individual size ranges of MPs. It should be noted that the removal ratio of some sized MPs particles had reached 100% during both CDAF and MDAF processes, thus these MPs were not mentioned.

Fig. 3 shows total removal of MPs with different dosages of CTAB or PDAC at a saturation pressure of 0.4 MPa. It can be observed that MPs removal increases with increasing the dosage of CTAB or PDAC up to a certain level. Specifically, the highest removal efficiencies of MPs were 81.6% and 88.3% obtained at 1.0 mg/L of CTAB and 0.8 mg/L of PDAC, respectively (Fig. 3), which are 41.5 and 48.2 percentage points higher than that in CDAF, respectively. Further increasing the dosage of the bubble modifiers could not apparently improve the performance. Obviously, PDAC exhibits better effect and lower dosage than CTAB. In CDAF, the surfaces of microbubbles and MPs are negatively charged but hydrophobic, which is similar to the adsorption of hydrophobic polymers on hydrophobic surface (Jamadagni et al., 2009; Pooja et al., 2021; Hasannattaj Jelodar et al., 2022). For MDAF, the hydrophobic tail of CTAB molecules is easy to closely adhere to the gas-liquid interface, CTAB may be located closer to the surface of microbubbles, and its hydrophilic head is located at the outer edge of microbubbles and thus can generate a positively charged region. Because of its low molecular weight (365 g/mol), CTAB adsorption will not significantly increase the sweeping volume of microbubbles (Henderson et al., 2010). Under the action of electrostatic attraction, PDAC is loosely attached to the surface of microbubbles (Zhao et al., 2021). PDAC has a longer molecular chain and is more hydrophilic than CTAB (Hankins et al., 2018). Thus PDAC molecules are prone to extend from the surface of microbubbles to the solution, bridging between MPs particles and microbubbles and forming a larger sweeping volume, thus leading to better treatment performance than CTAB.

Table 2
Average effluent concentration of each unit and total removal efficiency of pollutants by the MDAF-SOPC system.

Parameter	Effluent value		Total removal(%)
	MDAF	SOPC	
COD_{cr} (mg/L)	91.3	24	77.1%
TSS (mg/L)	3.6	ND	100%
Antibiotics (μg/L)	4.35	0.04	99.2

ND: not detected.

On the basis of the above results, the LPBW was treated under the conditions of 0.4 MPa saturation pressure and 0.8 mg/L PDAC by MDAF for a purpose to provide the SOPC process with influent stream.

3.3. Antibiotics degradation by SOPC

The effect of pumping pressure on the degradation ratio of antibiotics was investigated at pH 6.4 and reaction time of 20 min. The results are demonstrated in Fig. 4A. It can be found that higher degradation efficiency was achieved at higher pressure up to 0.06 MPa. The reason for this phenomenon is that higher pressure leads to higher injection speed to swirling cavitation chamber and then causes more strongly pressure drop in vortex center, resulting in the production of more cavitation bubbles (Li et al., 2022). Antibiotics degradation has reached as high as 98.3% at 0.06 MPa of pumping pressure (Fig. 4A), thus further improvement is difficult due to the low residue level.

The effect of reaction time on the degradation ratio of antibiotics was investigated at pH 6.4 and 0.06 MPa of pumping pressure. As shown in Fig. 4B, rapid degradation of antibiotics occured within the first 5 min and, thereafter, the degradation rate slows down gradually and reaches equilibrium in about 20 min. At this point, 98.3% of antibiotics was degraded, indicating an excellent performance of the SOPC system. Although 100% removal could be achieved in 30 min, 20 min was selected as the optimum reaction time for the SOPC system or for the consideration of operation cost savings. For AOPs including CIS, the degradation rate of organic pollutants generally presented a “fast first, slow later” pattern (Li et al., 2022). Several oxidation routes such as themolysis, free radical oxidation and supercritical water oxidation are involved in CIS.

Fig. 4C shows the influence of solution pH on the degradation of antibiotics at reaction time of 20 min and 0.06 MPa of pumping pressure. The influent pH was 6.4. It can be found that solution pH has slight effect on the degradation efficiency of antibiotics (Fig. 4C). Specifically, 99.5% and 97.6% degradation efficiency were obtained at pH 3 and 9, respectively. This is of practical significance since some AOPs, such as Fenton oxidation, have strict requirements for solution pH. In general, the decrease in pH increases the degradation efficiency as pH affects the oxidation potential by directly influencing Fe^{2+} concentration and the amount of •OH generated via the Fenton’s reagent. In the present study, the production mechanism of •OH is ascribed to the collapse of cavitation bubbles in the SOPC reactor (Dhanke et al., 2019; Li et al., 2022). Thus, the yield of •OH was less influenced by solution pH in this study.

As shown in Table 2, a small portion of COD and antibiotics were removed by MDAF, which can be mainly ascribed to TSS carrying. The SOPC process removed not only most of antibiotics but also a large portion of COD. The final effluent satisfied the national discharge standard of China for LPBW (Class I).

3.4. ARGs removal by the MDAF-SOPC process

Currently, ARGs are a major environmental concern in the wastewater treatment for their potential hazards to human health and ecological safety. All seven gene targets (*tet*M, *tet*O, *tet*Q, *tet*W, *sul*I, *sul*II,

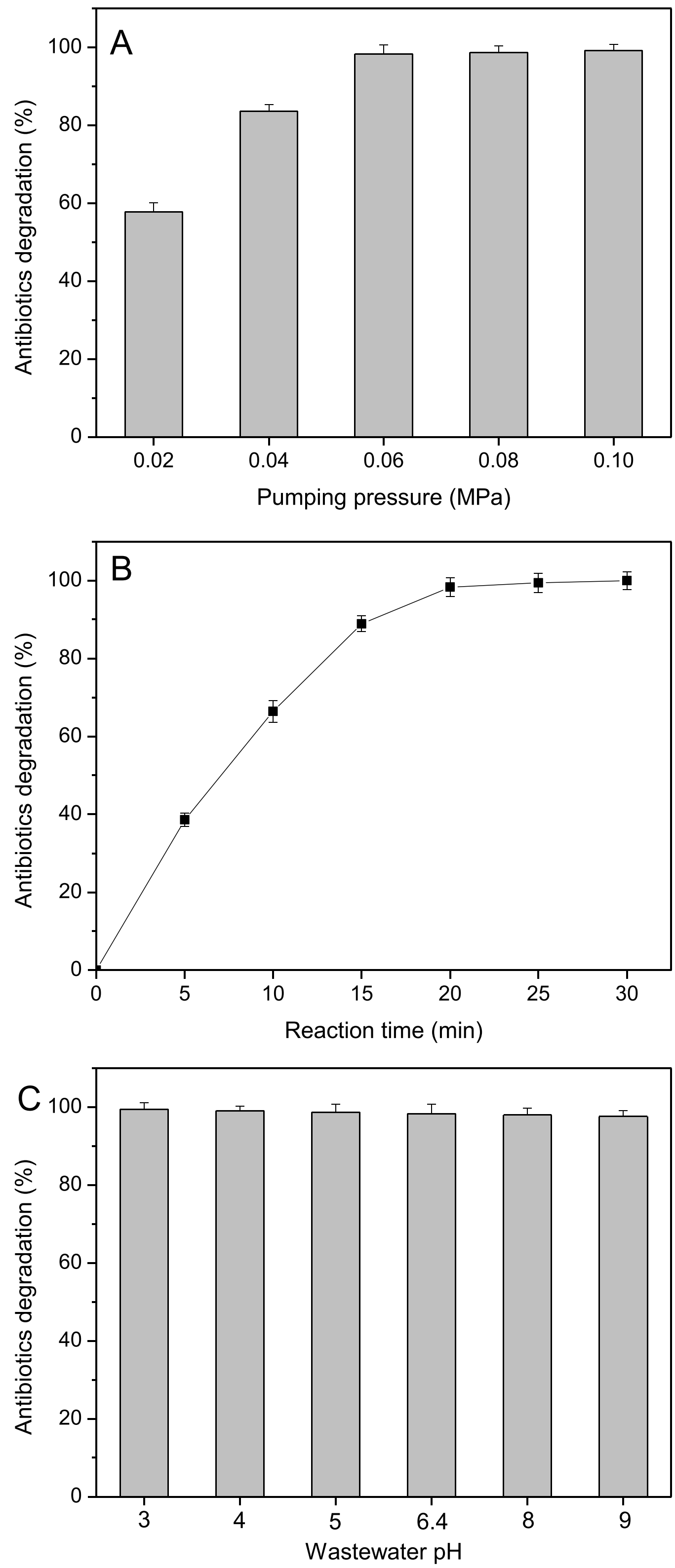

Fig. 4. Effect of pumping pressure (A), reaction time (B) and solution pH (C) on the degradation ratio of antibiotics by SOPC.

Table 3
16S rRNA gene abundance (gene copy number/mL) of targeted ARGs in the raw influent and effluents of different units of MDAF-SOPC systems for LPBW treatment.

	*int*Il	*sul*I	*sul*II	*tet*M	*tet*O	*tet*Q	*tet*W
Raw wastewater	6.3×10^{8}	9.4×10^{9}	7.3×10^{9}	1.9×10^{11}	3.4×10^{10}	2.1×10^{11}	5.8×10^{11}
MDAF effluent	2.4×10^{6}	4.6×10^{7}	6.1×10^{6}	2.7×10^{8}	1.6×10^{9}	4.5×10^{9}	3.3×10^{8}
SOPC effluent	ND	ND	ND	ND	ND	ND	ND

ND: not detected.

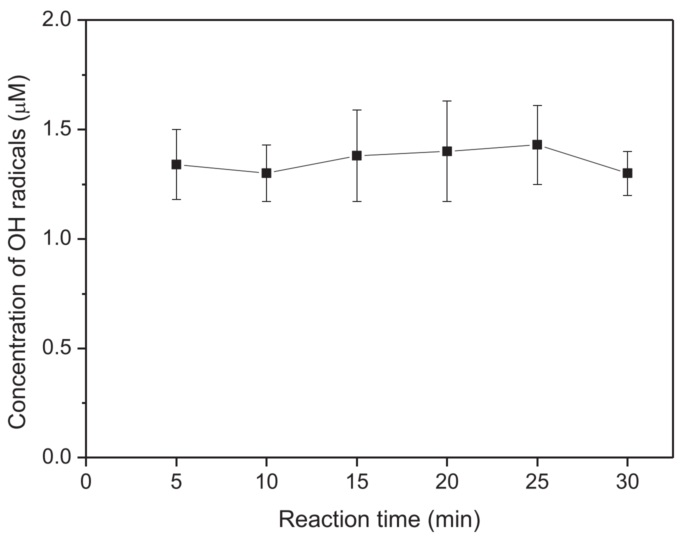

Fig. 5. The concentration of •OH radicals generated by the SOPC process at pH 6.4 and 0.06 MPa of pumping pressure.

and *int*Il) were detected in the raw LPBW (Table 3). The concentrations of tetracycline-related ARGs (including *tet*M, *tet*O, *tet*Q and *tet*W) were significantly higher than that of others, which can be ascribed to the relatively high levels of tetracycline in the wastewater. ARGs abundance values were reduced by 2–3 orders of magnitude in the MDAF unit (Table 3), indicating that suspended solids (including MPs particles) could carry ARGs through adsorption of antibiotic-resistant bacteria and the related ARGs (Zhang and Xu, 2020; He et al., 2022). Interestingly, ARGs were almost completely removed in the SOPC unit (Table 3), which was of special significance since one of the main purposes of the present treatment work was to effectively remove ARGs from the wastewater. SOPC involved high temperature decomposition, free radical oxidation and supercritical water oxidation (Li et al., 2022), thus it could strongly destroy ARGs. When selecting used technology for disinfection of ARGs, not only operational cost and ARGs removal efficiency but also possible secondary contamination should be considered. AOPs can damage the cell surface and DNA structure of microbes through free radical reactions (Li et al., 2021). In general, ARGs can be effectively destroyed by various AOPs. For example, Gao et al. (2020) showed 99.99% removal of *int*I1 through persulfate oxidation; Zheng et al. (2017) reported that the removal efficiency of ARGs in secondary sewage ranged from 34.5% to 49.2% at 2 mg/L of O_3. Contrastly, no additional agents were required in the present study, demonstrating the environmental friendliness of the SOPC process.

3.5. *•OH radical detection*

In the SOPC process, the extreme conditions of high temperature, high pressure and high micro jet can produce •OH radicals with strong oxidation capacity through the collapse of cavitation bubbles (Dhanke et al., 2019; Li et al., 2022). Fig. 5 shows the content of •OH radicals generation during the SOPC process. The experiments were conducted at pH 6.4 and 0.06 MPa of pumping pressure. It can be found from Fig. 5 that the amount of •OH radicals remained stable over time during the treatment period. This shows the superiority of the SOPC process over Fenton oxidation namely wide pH range and stable generation of •OH radicals. During Fenton oxidation, the yield of •OH radicals would gradually decline if no agents were supplemented continuously.

4. Conclusions

A novel system combining MDAF and SOPC was developed for the advanced treatment of LPBW. The operating conditions were optimized by using batch tests. The system exhibited outstanding performance on the removal of antibiotics and ARGs, namely 77.1% of COD, 100% of TSS, 99.2% of antibiotics and 100% of ARGs. MDAF could remove MPS more effectively than CDAF, and a large quantity of ARGs were removed in the MDAF unit through flotation removal of suspended solids. ARGs were completely removed in the SOPC unit. The hybrid system is characterized by high effectiveness, environmental friendliness and mild conditions. Especially, the system can be adapted to wide pH range. Future study is required to further optimize the design of the system to combine the MDAF and SOPC reactors into one unit for a purpose to simplify operation. The present study is beneficial for the further development of effective and efficient system aiming at removing ARGs and MPs from co-contaminated wastewater.

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

This work was funded by the National Natural Science Foundation of China (grant no. 51974313, 41907405).

References

Ahmed, M.B., Rahman, M.S., Alom, J., Hasan, M.S., Johir, M.A.H., Mondal, M.I.H., Lee, D.Y., Park, J., Zhou, J.L., Yoon, M.H., 2021. Microplastic particles in the aquatic environment: a systematic review. Sci. Total Environ. 775, 145793.

Anjali, R., Shanthakumar, S., 2019. Insights on the current status of occurrence and removal of antibiotics in wastewater by advanced oxidation processes. J. Environ. Manage. 246, 51–62.

APHA. Standard methods for the examination of water and wastewater. 20th ed. Washington (DC): American Public Health Association, American Water Works Association, Water Environment Federation; 1998.

Chen, Y., Zhang, H., Luo, Y., Song, J., 2012. Occurrence and dissipation of veterinary antibiotics in two typical swine wastewater treatment systems in east China. Environ. Monit. Assess. 184, 2205–2217.

Chen, F., Ma, J., Zhu, Y., Li, X., Yu, H., Sun, Y., 2022. Biodegradation performance and anti-fouling mechanism of an ICME/electro-biocarriers-MBR system in livestock wastewater (antibiotic-containing) treatment. J. Hazard. Mater. 426, 128064.

Dhanke, P., Wagh, S., Patil, A., 2019. Treatment of fish processing industry wastewater using hydrodynamic cavitational reactor with biodegradability improvement. Water Sci. Technol. 80, 2310–2319.

Esfandiari, A., Mowla, D., 2021. Investigation of microplastic removal from greywater by coagulation and dissolved air flotation. Process Saf. Environ. Protect. 151, 341–354.

Gao, J.F., Duan, W.J., Zhang, W.Z., Wu, Z.L., 2020. Effects of persulfate treatment on antibiotic resistance genes abundance and the bacterial community in secondary effluent. Chem. Eng. J. 382, 121860.

Hankins, C., Duffy, A., Drisco, K., 2018. Scleractinian coral microplastic ingestion: potential calcification effects, size limits, and retention. Mar. Pollut. Bull. 135, 587–593.

Hasannattaj Jelodar, A., Amini Rad, H., Borghei, S.M., Vossoughi, M., Rouhollahi, R., 2022. Particle removal optimization in rotating dissolved air flotation used in paper-recycling wastewater treatment. Water Environ. J. 36 (1), 3–17.

Henderson, R.K., Parsons, S.A., Jefferson, B., 2009. The potential for using bubble modification chemicals in dissolved air flotation for algae removal. Sep. Sci. Technol. 44, 1923–1940.

He, Y., Bai, M., He, Y., Wang, S., Zhang, J., Jiang, S., Wang, G., 2022. Suspended particles are hotspots for pathogen-related bacteria and ARGs in coastal beach waters of northern China. Sci. Total Environ. 817, 153004.

Jamadagni, S.N., Godawat, R., Garde, S., 2009. How surface wettability affects the binding, folding, and dynamics of hydrophobic polymers at interfaces. Langmuir 25, 13092–13099.

Kankanige, D., Babel, S., 2020. Smaller-sized micro-plastics (MPs) contamination in single-use PET-bottled water in Thailand. Sci. Total Environ. 717, 137232.

Li, S., Zhang, C., Li, F., Hua, T., Zhou, Q., Ho, S.H., 2021. Technologies towards antibiotic resistance genes (ARGs) removal from aquatic environment: a critical review. J. Hazard. Mater. 411, 125148.

Li, M., Zhang, H., Tong, X., Wang, B., Shang, J., Lu, M., 2022. Advanced treatment of hydraulic fracturing flowback water using a novel combined process. J. Environ. Chem. Eng., 107178.

Hvistendahl, M., 2012. Public health. China takes aim at rampant antibiotic resistance. Science 336, 795.

Patel, R.V., Yadav, A., 2022. Photocatalytic MIL101 (Fe)/ZnO chitosan composites for adsorptive removal of tetracycline antibiotics from the aqueous stream. J. Molecul. Structur. 1252, 132128.

Pooja, G., Kumar, P.S., Prasannamedha, G., Varjani, S., Vo, D.V.N., 2021. Sustainable approach on removal of toxic metals from electroplating industrial wastewater using dissolved air flotation. J. Environ. Manage. 295, 113147.

Qi, J., Lan, H., Liu, R., Liu, H., Qu, J., 2018. Fe (II)-regulated moderate pre-oxidation of Microcystis aeruginosa and formation of size-controlled algae flocs for efficient flotation of algae cell and organic matter. Water Res. 137, 57–63.

Swart, B., Pihlajamäki, A., Chew, Y.J., Wenk, J., 2022. Microbubble-microplastic interactions in batch air flotation. Chem. Eng. J., 137866.

Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., Mcgonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304 838-838.

Tian, M., He, X., Feng, Y., Wang, W., Chen, H., Gong, M., Liu, D., Clarke, J.L., Eerde, A.V., 2021. Pollution by antibiotics and antimicrobial resistance in livestock and poultry manure in china, and countermeasures. Antibiotics 10, 539.

Tullo, E., Finzi, A., Guarino, M., 2019. Review: environmental impact of livestock farming and precision livestock farming as a mitigation strategy. Sci. Total Environ. 650, 2751–2760.

Wang, J., Zhuan, R., 2020. Degradation of antibiotics by advanced oxidation processes: an overview. Sci. Total Environ. 701, 135023.

Wang, Y., Li, Y.N., Tian, L., Ju, L., Liu, Y., 2021. The removal efficiency and mechanism of microplastic enhancement by positive modification dissolved air flotation. Water Environ. Res. 93, 693–702.

Wang, Y., Ma, H., Wang, X., Ju, L., Tian, L., Qi, H., Yu, H., Li, J., 2022. Study on the operation performance and floc adhesion mechanism of dissolved air flotation equipment. Environ. Sci. Pollut. Res. doi:10.1007/s11356-022-19359-9.

Wu, R.T., Cai, Y.F., Chen, Y.X., Yang, Y.W., Xing, S.C., Liao, X.D., 2021a. Occurrence of microplastic in livestock and poultry manure in South China. Environ. Pollut. 277, 116790.

Wu, R.T., Cai, Y.F., Xing, S.C., Yang, Y.W., Mi, J.D., Liao, X.D., 2021b. A novel method for extraction of polypropylene microplastics in swine manure. Environ. Sci. Pollut. Res. 28, 13021–13030.

Yadav, A., Yadav, P., Labhasetwar, P.K., Shahi, V.K., 2021. CNT functionalized ZIF-8 impregnated poly (vinylidene fluoride-co-hexafluoropropylene) mixed matrix membranes for antibiotics removal from pharmaceutical industry wastewater by vacuum membrane distillation. J. Environ. Chem. Eng. 9, 106560.

Yadav, P., Yadav, A., Labhasetwar, P.K., 2022a. Sustainable adsorptive removal of antibiotics from aqueous streams using Fe3O4-functionalized MIL101(Fe) chitosan composite beads. Environ. Sci. Pollut. Res. 29, 37204–37217.

Yadav, A., Dindorkar, S.S., Ramisetti, S.B., 2022b. Adsorption behaviour of boron nitride nanosheets towards the positive, negative and the neutral antibiotics: insights from first principle studies. J. Water Process Eng. 46, 102555.

Yamaguchi, R., Kurosu, S., Suzuki, M., Kawase, Y., 2018. Hydroxyl radical generation by zero-valent iron/Cu (ZVI/Cu) bimetallic catalyst in wastewater treatment: heterogeneous Fenton/Fenton-like reactions by Fenton reagents formed in-situ under oxic conditions. Chem. Eng. J. 334, 1537–1549.

Zhang, S., Pang, X., Yue, Z., Zhou, Y., Duan, H., Shen, W., Li, J., Liu, Y., Cheng, Q., 2020. Sulfonamides removed from simulated livestock and poultry breeding wastewater using an in-situ electro-Fenton process powered by photovoltaic energy. Chem. Eng. J. 397, 125466.

Zhang, M., Xu, L., 2020. Transport of micro-and nanoplastics in the environment: trojan-Horse effect for organic contaminants. Crit. Rev. Environ. Sci. Technol. doi:10.1080/10643389.2020.1845531.

Zhang, Z., Chen, Y., 2020. Effects of microplastics on wastewater and sewage sludge treatment and their removal: a review. Chem. Eng. J. 382, 122955.

Zhao, C., Zhou, J., Yan, Y., Yang, L., Xing, G., Li, H., Wu, P., Wang, M., Zheng, H., 2021. Application of coagulation/flocculation in oily wastewater treatment: a review. Sci. Total Environ. 765, 142795.

Zheng, J., Su, C., Zhou, J., Xu, L., Qian, Y., Chen, H., 2017. Effects and mechanisms of ultraviolet, chlorination, and ozone disinfection on antibiotic resistance genes in secondary effluents of municipal wastewater treatment plants. Chem. Eng. J. 317, 309–316.