

Review article



Treatment technologies and mechanisms for tetramethylammonium hydroxide (TMAH) wastewater from micro-electronic industry: A review

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ABSTRACT

With the advent of Industry 4.0, the micro-electronic industry has become one of the fastest-growing industries worldwide. As a result, tetramethylammonium hydroxide (TMAH) wastewater, as the typical wastewater from the micro-electronic industry, has been dramatically increasing. The efficient treatment of TMAH wastewater has attracted growing attention due to its toxicity to the eco-environment and human health. However, the review of the mechanisms of TMAH wastewater treatment is still lacking. This paper systematically reviewed the mechanisms of TMAH treatment using various processes, including recovery processes and degradation processes. The recovery mechanisms of TMAH included cation exchange under the electrostatic attraction for adsorption, directed migration of TMA^+ and OH^- driven by electrodes for electrodialysis, etc. The degradation mechanisms of TMAH included methanogenic TMAH degradation, TMAH-oxidative methylotrophic degradation, and demethylation and oxidation. The results highlighted that both hybrid recovery and treatment processes exhibited the most efficient recovery and removal of TMAH. Future research on TMAH wastewater treatment should focus on energy metabolism, electron transfer, and co-digestion of biodegradation, efficient novel materials for TMAH recovery and degradation, and appropriate hybrid recovery and treatment processes for resource reuse of TMAH wastewater. Ultimately, the possible pathways for the resource reuse of TMAH wastewater were also proposed.

1. Introduction

With the advent of Industry 4.0, the micro-electronic industry has played a more key role all over the world. The micro-electronic industry mainly produces a series of electronic components such as high-performance chips, memory, capacitors, thin-film transistor liquid crystal display (TFT-LCD), integrated circuits, printed circuit boards (PCBs), etc. The microelectronics manufacture is a considerably high ultrapure water-consuming and wastewater-discharging industry (Frost et al., 2017). According to our statistics about official data of 28 semiconductor corporations all over the world, the total water withdrawal in 2021 reached $7.87 \times 10^8 \text{ m}^3$ in these semiconductor corporations (Wang et al., 2023). In addition, a six-generation chip manufacture factory could discharge up to $3.0 \times 10^4 \text{ m}^3$ per day wastewater, which is

equivalent to the wastewater production per day of a city with 1.0×10^6 population (Chang et al., 2008).

Meanwhile, as the chip process becomes more advanced, the water consumption and wastewater discharge of chip manufacturing have been larger and larger. At present, with the dramatic development of the micro-electronic industry, the considerable high ultrapure water-consuming and wastewater-discharging have been its obvious environmental problems. The cost of wastewater treatment has been a major part of the annual operating cost (Ferella et al., 2021).

Recently, the zero liquid discharge (ZLD) concept has been becoming more and more necessary to recover water for reuse in manufacturing processes (Lassesson et al., 2021). In other words, wastewater can also be regarded as a "misplaced resource", suitable for the recovery of energy, valuable materials, and clean water (Capocelli et al., 2019). Hence,

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the efficient treatment and resource reuse to achieve ZLD of wastewater from the micro-electronic industry, as an effective means, has to be conducted to solve its high ultrapure water-consuming and wastewater-discharging problems. It is also meaningful to lead to a better environmental impact and reduce treatment costs of the micro-electronic industry. As well, this means could fulfill the new paradigm, “Doing more with less”, proposed by the International Water Association for meeting Sustainable Development Goal 6 “Clean Water and Sanitation” by 2030 (IWA, 2016).

However, according to the latest statistics, the reclaimed water intake of these above 28 semiconductor corporations only accounted for 3.2% of its total water supply in 2021 (Wang et al., 2023). These undesirable results could mainly be attributed to a major concern about the presence of tetramethylammonium hydroxide (TMAH, $(\text{CH}_3)_4\text{NOH}$) in wastewater from micro-electronic industry in those countries where the micro-electronic industry is highly developed, such as the U.S., China, Japan, and South Korea (Ferella et al., 2021).

TMAH is the most representative chemical reagent used in the micro-electronic industry, which has been widely used as a developer in the photolithography process (Chang et al., 2008). As a result, TMAH concentration in wastewater from the micro-electronic industry generally could reach a few thousand ppm, leading to a fact that it is the highest concentration of organic pollutants in the chip manufacturing industry wastewater (Omar et al., 2008; Hu et al., 2018). In addition, the wastewater containing TMAH generally accounted for the highest proportion of 30~40% in the chip manufacturing industry wastewater (Hu et al., 2018). TMAH is a strongly alkaline solution with corrosivity and toxicity. TMAH can seriously threaten the ecosystem and be harmful to human health when exposed to the environment due to its corrosivity and toxicity.

For example, it could trigger nervous toxicity and respiratory failure even after exposure to 1% TMAH (Park et al., 2013). As a result, the TMAH concentration in wastewater from micro-electronic manufacturers has been strictly limited due to its toxicity. For instance, wastewater with TMAH below 7 mg/L only could be discharged to adequately protect aquatic eco-systems as stipulated by the EU directive 2010/75/EU (2010). TMAH in wastewater below 75 mg/L could be discharged as stipulated by Taiwan EPA (2014).

Hence, TMAH wastewater has to be properly treated in a “green”, high-efficiency, and cost-effective way. Currently, only one review (Innocenzi et al., 2022) introduced the TMAH wastewater treatment, mainly focusing on the removal effect of TMAH. However, there is still a current knowledge gap on the mechanism of TMAH treatment in wastewater due to a lack of critical reviews. It is meaningful to review the mechanisms of TMAH degradation and to discuss the resource reuse of TMAH wastewater under the background of ZLD for promoting the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater.

Based on the above considerations, this review aims to provide a comprehensive overview regarding the mechanisms of TMAH degradation, and to attempt to discuss the resource reuse of TMAH wastewater. The specific objectives were to (i) concatenate the development status of TMAH wastewater treatment according to the most recent literature data and application to better understand its status; (ii) evaluate the performance effect, influence factors, and possible mechanisms of TMAH treatment using various processes; and (iii) highlight the challenges and future prospects for TMAH wastewater treatment based on the perspective of resource reuse. The conclusions obtained in this work are available to provide new insight into the TMAH wastewater treatment, which will be helpful to better promote the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater.

2. Development status of TMAH wastewater treatment

Changes in the frequency of lectures on certain topics with publication time can reflect the development status and further trends in such

studies. Hence, we utilized “Tetramethylammonium hydroxide wastewater” as the keywords to investigate the relevant studies in the Web of Science in 2010~2022. Then the prevailing research fields of these publications were investigated for TMAH wastewater treatment by visual keyword analysis software VOS viewer 1.6.16. The minimum occurrence of a keyword was set to 2. From a total of 379 keywords, 87 keywords were selected. The co-occurrence keyword network analysis for TMAH wastewater treatment was shown in Fig. 1(a).

According to Fig. 1(a), the following fields were primarily focused on in the recent publications on TMAH wastewater over the past decade, (i) TMAH treatment technologies, including anaerobic biological treatment (Danshita et al., 2018; Lv et al., 2021; Chang et al., 2022), aerobic biological treatment (S.Y. Chen et al., 2016; Innocenzi et al., 2019; Ferella et al., 2021), activated carbon (AC) adsorption (Citraningrum et al., 2013; Chang et al., 2015; Shu et al., 2016), graphene oxide (GO) adsorption (Chang et al., 2014, 2015), ion exchange (Fu et al., 2022), hydrogen peroxide (H_2O_2) oxidation (Imtisal et al., 2019; Kim et al., 2021), persulfate ($\text{S}_2\text{O}_8^{2-}$) oxidation (Wang et al., 2014; J. Huang et al., 2017); (ii) TMAH and intermediate products identification (Whang et al., 2015; Danshita et al., 2018; Hu et al., 2018; Lv et al., 2021), kinetics (Liu et al., 2016; Hu et al., 2018; Danshita et al., 2018; Wu et al., 2020), and nitrogen removal (Murphy et al., 2012; Feng et al., 2019; Ferella et al., 2021); (iii) inhibition / toxicity of TMAH to microorganisms in biological treatments (S.Y. Chen et al., 2016; Urasaki et al., 2019; Lin et al., 2016; Wu et al., 2020). Despite it, it could be illustrated that the current focus on TMAH wastewater was mainly on the one-way linear treatment of TMAH wastewater, specifically TMAH removal and then discharge, while the close-loop treatment of TMAH wastewater, namely the multi-way resource reuse is still less focused.

In order to evaluate the mechanisms of TMAH degradation using various processes of those publications and discuss the resource reuse of TMAH wastewater, these TMAH treatment technologies could be specifically classified into two categories, including its recovery and degradation (Fig. 1(b)). The recovery technologies of TMAH mainly included adsorption, ion exchange, membrane process, electrodialysis (ED), and hybrid recovery processes, while the degradation technologies mainly included anaerobic biodegradation, aerobic biodegradation, advanced oxidation processes (AOPs), and hybrid treatment processes. In the following, the detailed mechanisms of TMAH treatment using these various technologies were evaluated respectively.

3. Recovery of TMAH from wastewater

3.1. Adsorption and ion exchange

3.1.1. Adsorption of TMAH

(1) Adsorbents for TMAH adsorption

The adsorption of TMAH in recent studies is summarized in Table 1. According to Table 1, among these adsorbents (Fig. 1(b)), AC is the most used for the adsorption of TMAH. For example, Prahas et al. (2012) investigated the adsorption of TMAH using several commercial ACs. The results demonstrated that under equal surface areas, the more micropores in ACs had, the higher adsorption capacity for TMAH was. The adsorption kinetics of TMAH using AC mainly seemed to follow the pseudo-second-order and Langmuir model under different pH and initial concentrations (Table 1). In addition, after 5 cycles of the adsorption-desorption process, the AC-saturated TMAH still could be regenerated by 0.1 N HCl. But with the increase of cycles, the separation of TMAH was gradually exhausted (Prahas et al., 2012).

Nishihama et al. (2010) reported the effect of zeolitic adsorbents on TMAH adsorption by preparing several zeolites using hydrothermal synthesis. Their results showed that zeolite X exhibited a higher adsorption capacity than zeolite A. But its maximum adsorption amount (q_e) of TMAH was only 1.0 mg/g (Nishihama et al., 2010), which was

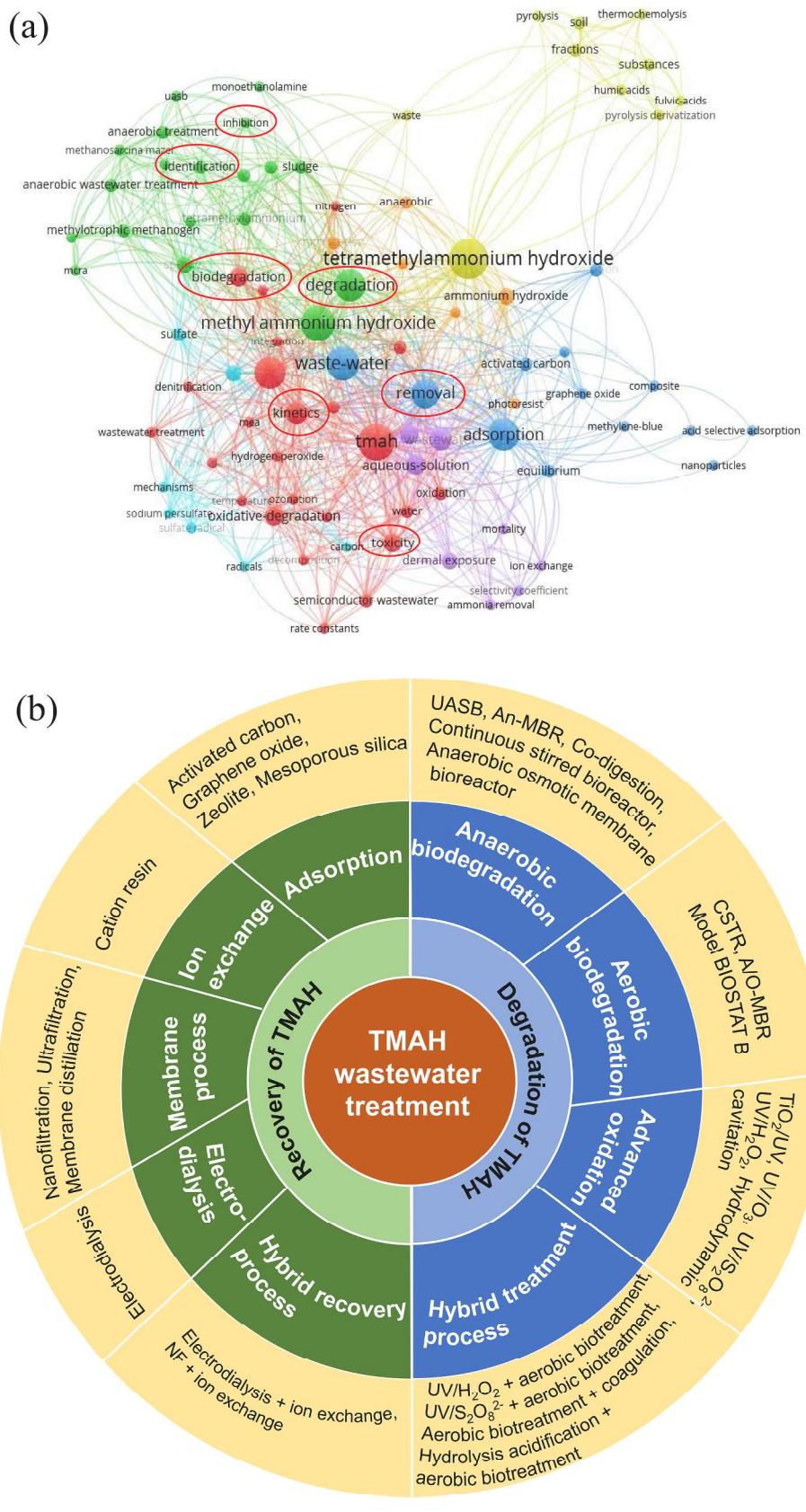


Fig. 1. Development status of TMAH wastewater treatment. (a) Cooccurrence keyword network analysis for TMAH wastewater treatment. Note: the higher diameter sphere represents higher occurrence frequency; (b) The main treatment technologies for TMAH wastewater.

Table 1
Recent studies on adsorption and ion exchange of TMAH from wastewater.

Adsorbent	Wastewater	TMAH concentration (mg/L)	HRT (h)	pH	Temperature (°C)	Optimal conditions	Kinetics	q_e max (mg/g)	Oxygen containing groups (mmol/g)	Ref.	
Commercial activated carbon	Synthetic wastewater	20~500	24	4.6 ± 0.1~10.1	25, 40, 55	pH 10.1 ± 0.1, 25°C	Pseudo-second-order model, and Langmuir model	20.50	0.179	0.134	0.160 Prahas et al., 2012
Granular activated carbon (GAC)	Synthetic wastewater	50, 200	5	3~11	25	pH 10.1 ± 0.1,	Pseudo-second-order model, and Langmuir model	21.5	0.27	0.29	0.72 Chang et al., 2014
Powdered activated carbon (PAC)	Synthetic wastewater	50, 200	5	3~11	25	pH 10.1 ± 0.1,	Pseudo-second-order model, and Langmuir model	19.7	0.34	0.46	0.55 Chang et al., 2014
Activated carbon from bamboo (BAC)	Synthetic wastewater	10~500	24	2.0 ± 0.5~12.0	25	pH 10.1 ± 0.1,	Pseudo-second-order model	23.06	0	0.120	0.361 Yamaguchi et al., 2012
Graphene oxide (GO)	Synthetic wastewater	50, 200	5	3~11	25	pH 10.1 ± 0.1,	Langmuir model	156	1.84	1.66	1.06 Chang et al., 2014
Y-type zeolite	Synthetic wastewater	25~600	5	5, 25, 45	25°C pH	Pseudo-second-order model	44.4	0.49	0.44	1.24 Chang et al., 2015	
MFI-type zeolite	Synthetic wastewater	301	24	2~12	25	pH 11	Langmuir model	54			Nishihama et al., 2013
Mesoporous silica with a hexagonal structure	Synthetic wastewater	300	2	2~12	25	pH 10	Langmuir model	94			Nishihama et al., 2012
Cubic mesoporous silica	Synthetic wastewater	150~400			7, 23, 40	23°C	Pseudo-second-order model	100			Kelleher et al., 2001
Strong acid cation resin	Synthetic wastewater	400±40		2~12		pH > 6.5	Pseudo-second-order model	177.55			Citranningrum et al., 2013
Weak acid cation resin	Synthetic wastewater	400±40		2~12		pH > 6.5	Pseudo-second-order model	186.99			Citranningrum et al., 2013
Amberlite	Synthetic wastewater	500	2	3.1, 7.0, 11.7	25	pH 11.7	Langmuir model	178.57			Shu et al., 2016

significantly lower than that (44.4 mg/g) of Y-type zeolite (Chang et al., 2015) and that (54 mg/g) of MFI-type zeolite (Nishihama et al., 2013). These results might be attributed to the different specific surface areas and pore volumes of these zeolites, such as only 727 m²/g and 0.35 cm³/g for the zeolite X (Nishihama et al., 2010), but 1614.1 m²/g and 0.419 cm³/g for the Y-type zeolite (Chang et al., 2015). It could be concluded that the adsorption capacity of zeolites to TMAH is expected to be further improved through developing the novelty zeolitic adsorbents with higher specific surface area and larger pore volume.

Mesoporous silica, as a class of inorganic adsorbents, has been paid attention to for TMAH adsorption due to its large pores. Kelleher et al. (2001) investigated the TMAH adsorption by cubic mesoporous silica. Their results showed that at the higher initial TMAH concentrations (> 400 mg/L), pore diffusion was the dominant rate-limiting step of adsorption, while at the lower (< 400 mg/L), film diffusion become the main. The adsorption amount of TMAH was related to the pore size and the pore volume of the mesoporous silica. In addition, Nishihama et al. (2012) prepared mesoporous silica with hexagonal structure adsorbents (MCM-41 s) with high selective adsorption of TMAH. The main mechanisms of selective adsorption of TMAH were the cation exchange reaction between cationic TMAH (TMA⁺) and anionic silanol groups of MCM-41 s (Nishihama et al., 2012).

Recently, GO has been proven to exhibit higher adsorption abilities than AC, zeolitic adsorbents, and mesoporous silica for organic compounds (Yang et al., 2011; Sun et al., 2021). Hence, a few studies have attempted to investigate TMAH adsorption by GO. For example, Chang et al. (2015) exploited the TMAH adsorption of GO prepared by the oxidation of graphite powders in comparison with AC and zeolite. Their results confirmed that the GO exhibited the highest TMAH adsorption capacity among these adsorbents. And the reversibility of GO was also superior, over 10 cycles of the adsorption-desorption process. In addition, the regeneration efficiency reached ~94% with 0.05 M NaCl solution (Chang et al., 2014).

(2) Adsorption conditions of TMAH

Additionally, it was worth noting that pH 10~11 and 23~25 °C were the optimal conditions for TMAH adsorption to AC, zeolite, mesoporous silica, and GO (Table 1). With the increase of pH from about 2 to about 11, the adsorption amount of TMAH gradually increased. For example, for GO, q_e increased from 20.6 to 25.1 mg/g with pH from 3 to 11 (Chang et al., 2014). For AC, at pH 4.7 ± 0.2 no adsorption of TMAH occurred (q_e was 0 mg/g), while q_e reached 22.5 mg/g at pH 11.8 ± 0.2 (Prahas et al., 2012). For mesoporous silica, at pH 2.0 ± 0.2 q_e was 0 mg/g, while at pH 10.0 ± 0.5 q_e reached the highest (94 mg/g) (Nishihama et al., 2012). In contrast, zeta potentials on the surface of the adsorbents gradually decreased with the increase of pH from about 2 to about 11 (Nishihama et al., 2012; Chang et al., 2014). For example, zeta potentials on the surface of MCM-41 s were from -5 mV to -39 mV when pH was from 2.0 ± 0.2 to 10.0 ± 0.5 (Nishihama et al., 2012). With the increase of pH from 3 to 11, zeta potentials on the surface of GO decreased from -18 mV to -42 mV (Chang et al., 2014).

(3) Mechanisms of the adsorption of TMAH

These above results revealed that the electrostatic attraction between TMA⁺ and hydroxide ions (OH⁻) was one of the main mechanisms of the adsorption of TMAH (Fig. 2(a)①). With the increase of pH, the surface charge of the adsorbent became more negative due to the more OH⁻ deposited on its surface. Consequently, the adsorbent with the negatively charged was more prone to TMA⁺ adsorption (Prahas et al., 2012).

In addition, the TMAH adsorption also could be driven by the cation exchange between TMA⁺ and H⁺ of the oxygen-containing groups under the electrostatic attraction between TMA⁺ and the negatively-charged polar groups of AC, zeolite, and GO, such as carboxylic, lactonic, and phenolic like groups (Prahas et al., 2012; Hui et al., 2013; Ledezma

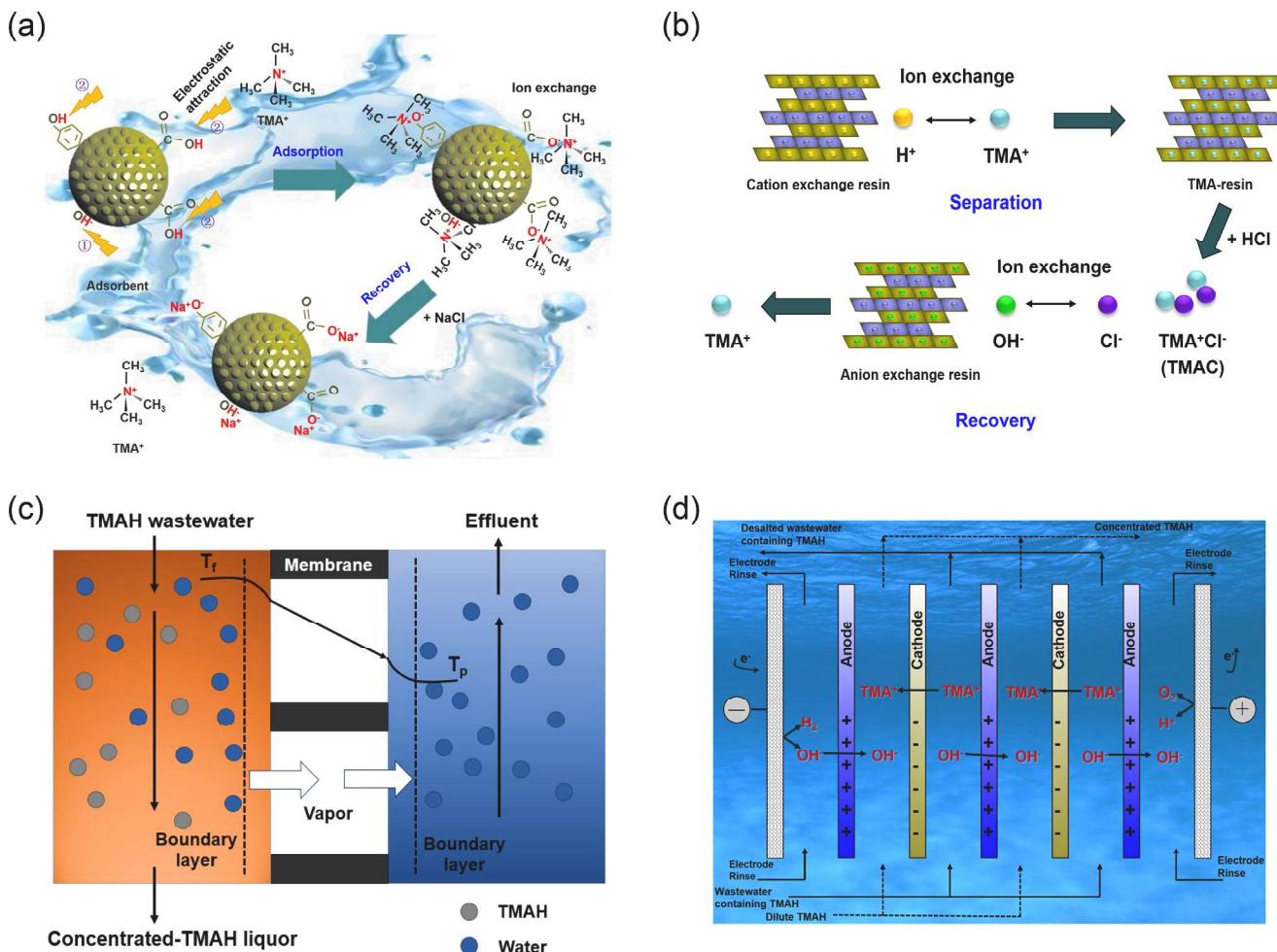


Fig. 2. The main recovery mechanisms of TMAH. (a) The main adsorption/recovery mechanisms of TMAH; (b) The main separation and recovery mechanism of TMAH by ion exchange; (c) The main recovery mechanism of TMAH by MD; (d) The main recovery mechanism of TMAH by ED.

et al., 2013; Chang et al., 2014; 2015) (Fig. 2(a)②). Chang et al. (2014) clarified that a strong interaction existed between TMAH and functional groups containing C = O bonds (i.e., carbonyl and carboxylate groups). In addition, since the pH of 10~11 was higher than the point of zero charges, the maximum surface charge density might be generated by the complete ionization process of these polar functional groups. As a result, more TMA⁺ was attracted by these polar functional groups at the pH of 10~11 (Lopez-Ramon et al., 1999; Prahas et al., 2012).

(4) Comparison of adsorbents for TMAH adsorption

Even though the order of specific surface areas and specific pore volumes was Y-type zeolite > GAC > GO, the adsorption capacity of TMAH was GO > Y-type zeolite > GAC (Chang et al., 2015). In contrast, the order of total surface acidity was consistent with its adsorption capacity (GO > Y-type zeolite > GAC) (Table 1). And GO exhibit a higher binding affinity for TMA⁺ than the GAC and PAC due to the higher Langmuir constant of GO (0.0165) than that of the GAC (0.0023) and PAC (0.0025) (Chang et al., 2014). Meanwhile, from the perspective of adsorption thermodynamics, the adsorption enthalpies of GO, zeolite, and AC were lower than the limit of that of adsorptions associated with ion exchange (8.4 kJ/mol) (Chang et al., 2015). These results could further prove that the TMAH adsorption could be primarily driven by the ion exchange driven by electrostatic attraction rather than the physisorption process.

In addition, in the recovery of TMAH, the positive ion from the desorbent, such as H⁺, Na⁺, K⁺, and Mg²⁺, could replace TMA⁺ on the

adsorbent to achieve the regeneration of adsorbent and recovery of TMAH (Prahas et al., 2012; Citraningrum et al., 2013; Chang et al., 2014, 2015; Citraningrum et al., 2016) (Fig. 2(a)).

3.1.2. Ion exchange for TMAH

(1) Cation resins used for TMAH

Recently, ion exchange has been used for the separation and recovery of TMAH from wastewater due to its low-cost, efficient, and easy regeneration (Fu et al., 2022). For example, Shibata et al. (2006) reported that the separation could be achieved by a cation exchange reaction between TMA⁺ and H⁺ of cation resin. The recovery rate of TMAH was 71.4%. In addition, Shu et al. (2016) demonstrated that the ion exchange capacity of cation resin (amberlite) reached the highest (178.57 mg/g) at a pH of 11.7 (Table 1). These results could be attributed to the fact that under the alkaline condition ion exchange sites of TMA⁺ were more abundant, and the competition between TMA⁺ and OH⁻ was less.

(2) Recovery mechanism of TMAH by ion exchange

The main separation and recovery mechanism of TMAH by ion exchange was summarized in Fig. 2(b). The separation of TMAH from wastewater could be achieved by a cation exchange reaction between TMA⁺ and H⁺ of cation resin. Generally, TMA⁺ on the resin could be eluted by an HCl solution, generating TMAC (Shibata et al., 2006). Then

the recovery of TMAH could be conducted by a cation exchange reaction between Cl^- of TMAC and OH^- of anion resin.

It could be concluded that the adsorption capacity of TMAH was cation resin > GO > mesoporous silica > zeolite > AC (Table 1). Adsorption and ion exchange could be used as the recovery of TMAH from wastewater before the biological treatment unit, or as the advanced treatment at the end-of-pipe within a process plant. So far, although the high-efficiency separation of TMAH has been achieved by the adsorption and ion exchange, the investigation of the recovery rate of TMAH by these various technologies is still less. In addition, the predictive model of TMAH adsorption should be also developed (Al-asad et al., 2022). Meanwhile, due to the complexity of actual wastewater components, the adsorbents with higher adsorption capacity, lower cost, stronger selectivity and easier recovery used for TMAH also should be developed through collaboration with other fields, such as materials and chemistry.

3.2. Membrane process

3.2.1. NF and UF for TMAH recovery

Recently, the membrane process has been employed for the separation of TMAH from wastewater (Nishihama et al., 2013; Tortora et al., 2018; Noor et al., 2019). For example, Nishihama et al. (2013) prepared a new nanofiltration membrane (NF) by coating MFI-type zeolite on the membrane for TMAH separation from the synthetic TMAH wastewater. Their results showed that the rejection of TMAH in wastewater reached the highest (100%) at a pressure of 0.080 MPa when the influent contained TMAH of 301 mg/L. The mechanisms for TMAH separation by the NF coated with the MFI-type zeolite mainly included molecular sieve and adsorption, with the former contributing 75% and the latter 25%.

Tortora et al. (2018) investigated the effect of TMAH wastewater treatment from the micro-electronic industry using a micellar-enhanced ultrafiltration (MEUF) process. Their results demonstrated that the rejection coefficient increased from ~80% to ~100% using the MEUF.

3.2.2. MD for TMAH recovery

(1) Conditions for TMAH recovery by MD

Membrane distillation (MD) is a phase change process thermally driven by low-grade waste heat utilization. It can use the micro-porous hydrophobic membrane to separate two different aqueous solutions but allows only volatile (i.e., water vapor) permeation (Lu et al., 2019; Sun et al., 2022). MD not only could employ simple heat transfer equipment and a low areal footprint, but also could provide approximately 100% rejection of contaminants (Wang et al., 2015; Sun et al., 2022). Noor et al. (2019) investigated the performance of MD for TMAH separation from wastewater in the micro-electronic industry. The results demonstrated that under 646.1 mg/L of TMAH in influent, TMAH in the effluent of MD all was below 1.7 mg/L when the inlet temperature was from 60 °C to 70 °C. In this study the optimal inlet temperature was chosen as 65 °C, considering that TMAH thermal decomposition temperature is over 65 °C. Under the optimal inlet temperature, the recovered effluent only contained 1.2 mg/L of TMAH. Meanwhile, the cost of water treatment was as low as about 16 \$/m³, which could account for only about 32% of the unit wastewater treatment cost.

(2) Recovery mechanism of TMAH by MD

The main recovery mechanism of TMAH by MD was shown in Fig. 2 (c). Under a certain different temperature (T_f and T_p) on both sides of the membrane, water vapor could pass through the microporous membrane and condense on the other side due to the different vapor pressure. In contrast, TMAH could not pass through since the provided temperature was lower than its thermal decomposition temperature, thus making TMAH gradually concentrated.

So far, although the high-efficiency separation of TMAH has been achieved by MD, NF, and UF, the investigation of the recovery rate of TMAH by these various technologies is still less. In addition, generally, the membrane process is an inevitable unit for advanced treatment and reclamation (Wu et al., 2021). For example, in the last decades, over 25% of the studies about water reclamation reported that UF + reverse osmosis (RO) was used for water reclamation (Tong et al., 2022). Therefore, membrane processes, especially MD, UF, NF, and RO, could be recommended to use for water reuse from TMAH wastewater. In addition, since fouling and aging have been unavoidable issues in the operation of the membrane process (TAN et al., 2022), the fouling characteristics, cleaning methods, regeneration approaches, and membrane materials for TMAH recovery and water reuse also should be investigated in the future. Meanwhile, the coexisting substances in the TMAH wastewater might impact the recovery of TMAH. Hence, the effect of coexisting substances in wastewater on TMAH recovery should be also investigated.

3.3. Electrodialysis

Electrodialysis (ED), as a membrane process combined with electrochemistry, has obtained major attention in recent years, since it usually can yield a concentrated and relatively pure product under lower energy consumption than other (membrane-based) technologies (Wei et al., 2022; Yuzer et al., 2022). Some lectures have employed ED for the TMAH recovery from wastewater in recent years (Wang et al., 2013; Sugawara et al., 2002; Ahn et al., 2023).

(1) Conditions of recovery of TMAH by ED

Wang et al. (2013) investigated the recovery of TMAH from developer wastewater using the ED process. Their results demonstrated that the TMAH concentration increased first and then decreased with the current density from 20 mA/cm² to 50 mA/cm². The recovery rate (93.7%) and concentrate ratio (3.50) of TMAH reached the highest at the current density of 30 mA/cm². These results showed that a higher current density could not produce a higher recovery of TMAH from wastewater, which could be mainly attributed to the following two aspects. On one hand, water electro-osmosis could cause ions to move across the ion exchange membrane, dragging water molecules from the inlet chamber to the concentration chamber. On the other hand, the volume of the concentration chamber could be gradually increased by the different pressures.

(2) Recovery mechanism of TMAH by ED

The main recovery mechanism of TMAH by ED is proposed in Fig. 2 (d). TMAH could be generally decomposed into TMA^+ and OH^- . During the recovery of TMAH from wastewater using ED, driven by the negative electrode, TMA^+ could directly move through the cation exchange membrane but not the anion exchange. Similarly, driven by the positive electrode, OH^- could directly pass through the anion exchange membrane but not through the other kinds (Sugawara et al., 2002; Wang et al., 2013; Innocenzi et al., 2022), causing that TMAH in wastewater be concentrated. The recovery rate of TMAH using ED reached 88.7%, and the corresponding cost was only 0.0364 \$/L (Wang et al., 2013). It could be concluded that ED, as a cost-effective and environmentally friendly technology, could be recommended to use as the pre-concentration and recovery of TMAH.

3.4. Hybrid recovery process

As a rule, it is possible to improve recovery efficiency through the hybrid use of various technologies. Sugawara et al. (2002) reported the recovery of TMAH from wastewater through an ED + ion exchange process. Their results demonstrated that the recovery rate of TMAH was

over 90%, and the reclaimed TMAH could be directly reused in the photolithography process, leading to a 55~75% decrease in the operation cost. Govardhan et al. (2021) reported the recovery of TMAH using an NF + ion exchange process, achieving a TMAH recovery rate of over 85%.

According to these lectures, the recovery efficiencies of TMAH from wastewater using various technologies, including adsorption, ion exchange, ED, and hybrid recovery process were statistically analyzed in Fig. 3. The order of the TMAH recovery rate was hybrid recovery process > ED > ion exchange > adsorption. Unlike the single, when the hybrid recovery process was conducted, they allowed to achieve greater recovery efficiencies and often at a lower cost. For example, for the NF + ion exchange process, macromolecular organic matters, such as photoresist (> 5000 Da) in TMAH wastewater could be pre-removed by NF to reduce interference with TMAH recovery by ion exchange, improving TMAH recovery efficiency and resin regeneration efficiency (Govardhan et al., 2021). In addition, to further improve TMAH recovery efficiency and reduce operation cost, the pre-concentration technologies of TMAH also should be used before TMAH recovery technologies, such as ion exchange, ED, etc.

Therefore, the hybrid recovery process, as a “green”, high-efficiency, and cost-effective process, should be recommended to use in the recovery and purification of TMAH from wastewater. But so far, the studies about the hybrid recovery process for TMAH are still fewer. Hence, the appropriate hybrid recovery process needs to be further exploited in the future.

4. Degradation of TMAH in wastewater

4.1. Biodegradation method

4.1.1. Anaerobic biodegradation

The performance of TMAH wastewater treatment by anaerobic biotreatment process in recent studies is summarized in Table 2. According to Table 2, most removal rates of TMAH from 10 mg/L to 3000 mg/L could be over 90% using various anaerobic biotreatment processes. For example, Chang et al. (2008) reported that the removal rate of THAM from actual wastewater was over 95% using an up-flow anaerobic sludge

bed (UASB) under the maximum volumetric loading rate of 7.30 kg TMAH/m³·d (4.1 kg TOC/m³·d). Hu et al. (2018) showed that the THAM removal rate reached 99% under a TMAH concentration of 3000 mg/L using a continuous stirred anaerobic bioreactor. To date, the lectures on TMAH biodegradation by the anaerobic mainly involved in the degradation pathways (Whang et al., 2015; Danshita et al., 2018; Hu et al., 2018; Lv et al., 2021), functional bacteria and kinetics (Hu et al., 2012; Whang et al., 2015; Liu et al., 2016; Hu et al., 2018; Danshita et al., 2018) as well as influence factors (Chang et al., 2008; Hu et al., 2012; Urasaki et al., 2019; Lv et al., 2021; Choi et al., 2021).

(1) Degradation pathways of TMAH by anaerobic biodegradation

According to the previous lectures (Whang et al., 2015; Danshita et al., 2018; Hu et al., 2018; Lv et al., 2021; Innocenzi et al., 2022), under anaerobic conditions, TMAH could be successively degraded into trimethylamine (TMA), dimethylamine (DMA), monomethylamine (MMA), and finally methane (CH₄), ammonia (NH₃), and carbon dioxide (CO₂). Tanaka et al. (1994) found that a methanogen from Tokyo Bay in Japan could use TMAH as the substrate to metabolize 1 M TMAH to 3 M CH₄, 1 M NH₃, and 1 M CO₂. Hu et al. (2018) also reported that the generated gas included 70% CH₄, 15% NH₃, and 15% CO₂ by analyzing the gas composition and effluent NH₃. Liu et al. (2014) found that the CH₄ conversion yield of TMAH reached 90% under TMAH concentration of 340 mg/L at room temperature using UASB.

(2) Functional bacteria and kinetics

Methanogens have been proven to play a key role in TMAH degradation (Hu et al., 2012; Whang et al., 2015; Liu et al., 2016; Hu et al., 2018; Danshita et al., 2018). The reported functional methanogens capable of degrading various TMAH concentrations were shown in Table 3. According to Table 3, these functional methanogens mainly were genus *Methanomethylovorans*, *Methanosaeta*, and *Methanosarcina*. The main mechanism of TMAH degradation by these methanogens was methanogenic TMAH degradation (Whang et al., 2015; Danshita et al., 2018; Hu et al., 2018; Lv et al., 2021), which was shown in Fig. 4(a). Specifically, first, the methyl group of TMAH was directly mobilized and

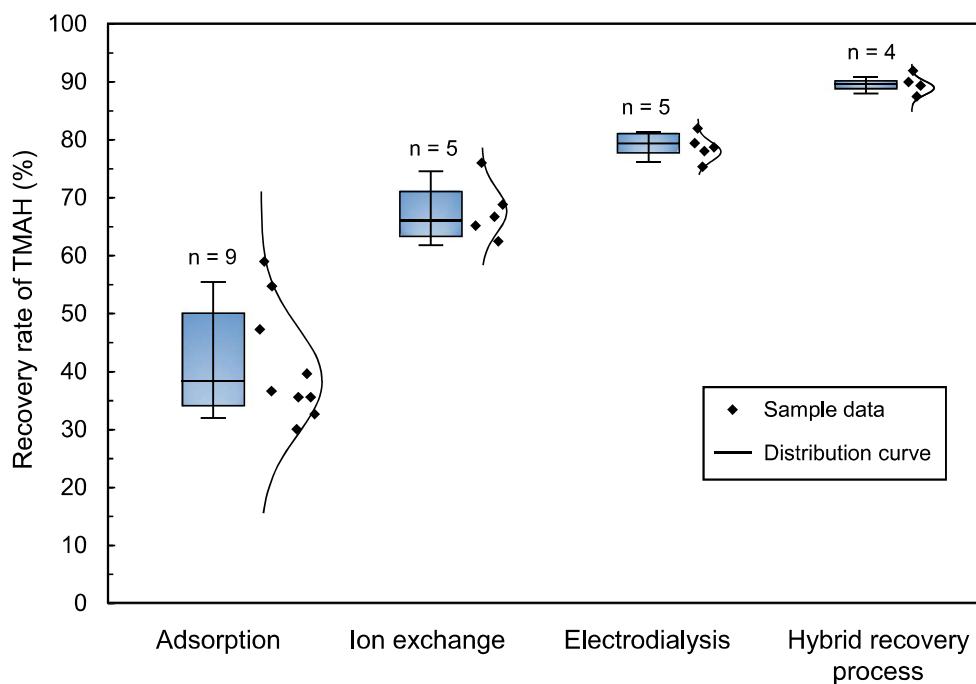


Fig. 3. Recovery efficiencies of TMAH from wastewater using adsorption, ion exchange, electrodialysis, and hybrid recovery process; n = number of samples.

Table 2

Recent studies on TMAH treatment in wastewater using biological process.

Biodegradation method	Biological process	Wastewater	TMAH concentration (mg/L)	Operation conditions	Performance effect	Ref.
Anaerobic biodegradation	UASB (10 L)	Synthetic wastewater	500 ~ 3000	pH 7.50, 37°C, VLR of 0.21 ~ 4.10 kg TOC/m ³ ·day	The maximum VLR was 7.30 kg TMAH/m ³ ·day (4.0 kg TOC/m ³ ·day). The removal rate of THAM was over 95%.	Chang et al., 2008
	UASB	TMAH wastewater from TFT-LCD manufacture	1144, 1528	HRT was 15 h, pH 7.0 ± 0.2, 35±1°C	The UASB could perform up to 92% of TMAH degradation efficiency and achieve more than 90% in gas composition for CH ₄ .	Hu et al., 2012
	UASB (2.0 L)	Synthetic wastewater	777 ~ 975	HRT was 3.0 ~ 10.0 h, pH 7.50, 35±2°C, OLR of 6.2 ~ 11.5 kg COD/m ³ ·day	The removal rate of THAM was over 99% at OLR of 11.5 kg COD/m ³ ·day.	Urasaki et al., 2019
	Continuous stirred bioreactor (10 L)	Synthetic wastewater	1000	HRT was 30 d, pH 7 ± 0.1, 30°C	The removal rate of THAM was more than 99% under a TMAH concentration of 3000 mg/L. And the gas composition mainly included CH ₄ (about v/v 70%), CO ₂ (about v/v 15%), and NH ₃ (about v/v 15%).	Hu et al., 2018
	UASB (2.0 L)	Synthetic wastewater	360	HRT was 4 h, pH 7.5, 30°C, OLR of 7.5 ± 0.78 kg COD/m ³ ·day	The removal rate of COD was 90% at 7.5 ± 0.78 kg COD/m ³ ·day.	Danshita et al., 2018
	Continuous stirred bioreactor (2.5 L)	Synthetic wastewater	100 ~ 1000	HRT was 48 h, pH 6.8 ~ 7.2, 38±1°C, ORP -150 mV, MLSS of 3 g/L	TOC concentration in the effluent declined from 520 mg/L to below 25 mg/L.	Lv et al., 2021
	Anaerobic osmotic membrane bioreactor (2.5 L)	TMAH wastewater from TFT-LCD manufacture	100±10	HRT was 48 h, pH 6.8 ~ 7.2, 35°C, ORP < -300 mV	The removal rate of THAM reached 99.99% in the anaerobic osmotic membrane bioreactor and MD hybrid system. And the effluent could be reused.	Chang et al., 2022
	CSTR (1.1 m ³)	TMAH wastewater from TFT-LCD manufacture	2161±187	HRT was 160 h, 25°C, DO 2.0 ± 0.5 mg/L	In the first bioreactor, the TMAH was reduced by around 80%, and in the second and third bioreactors, it was completely reduced (100%).	Ferella et al., 2021
Aerobic biodegradation	Model BIOSTAT B (5 L)	TMAH wastewater from TFT-LCD manufacture	1800 ~ 2500	HRT was 13 d (312 h), 25°C, pH 7, air flux of 2 L/min	After 12 d (288 h), TMAH concentration was 0.4 mg/L with a 99.95% of removal rate.	Innocenzi et al., 2019
	A/O-MBR	TMAH wastewater from TFT-LCD manufacture		HRT was 32 h, SRT was 40 d, DO 3.0 mg/L, pH 7 ± 0.1, 27±2°C	The TN removal efficiency was 59%.	Whang et al., 2012
	A stirring up-flow reactor (5 L)	Synthetic wastewater with TMAH	200 ~ 1000	HRT was 4.34 d, DO 0.5 mg/L, pH 7.0 ~ 8.0	TMAH was almost completely biodegraded in a CANON process. The average removal rates of total nitrogen were higher than 90%.	S.Y. Chen et al., 2016

Table 3

The reported functional bacteria for TMAH degradation in wastewater.

Biodegradation method	TMAH (mg/L)	Enriched microorganisms	Inoculated sludge	Key functions	Ref.
Anaerobic biodegradation	100	<i>Methanomethylovorans</i>	UASB treating TMAH wastewater	Methanogenic TMAH degradation	Hu et al., 2012; 2018
	100	<i>Methanomethylovorans</i> (3.1%), <i>Methanosaeta</i> (41%)	Anaerobic sludge from municipal WWTP	Methanogenic TMAH degradation	Chang et al., 2022
	360	<i>Methanomethylovorans</i> (49%), <i>Methanosaeta</i> (38%)	UASB treating TMAH wastewater	Methanogenic TMAH degradation	Danshita et al., 2018
	343	<i>Methanomethylovorans</i> (60.3%), <i>Methanosarcina</i> (36.3%)	Anaerobic sludge from municipal WWTP	Methanogenic TMAH degradation	Liu et al., 2016
	620	<i>Methanomethylovorans</i>	UASB treating TMAH wastewater	Methanogenic TMAH degradation	Whang et al., 2015
	1200	<i>Methanosarcina</i>	UASB treating TMAH wastewater	Methanogenic TMAH degradation	Hu et al., 2012; 2018
	1400	<i>Methanosarcina</i>	UASB treating TMAH wastewater	Methanogenic TMAH degradation	Whang et al., 2015
	90	<i>Hypomicrobium</i> , <i>Methyloversatilis</i>	MBR treating TMAH wastewater	TMAH-oxidative methylotrophic degradation	Wu et al., 2008
Aerobic biodegradation	200 ~ 800	<i>Hypomicrobium</i> (25.2%), <i>Mycobacterium</i> (11.5%), <i>Methyloversatilis</i>	SBR treating TMAH wastewater	TMAH-oxidative methylotrophic degradation	Wu et al., 2020
	2161±187	<i>Methyloversatilis</i> (6%), <i>Delftia</i> (28%), <i>Mycobacterium</i> (13%), <i>Serratia</i>	CSTR treating TMAH wastewater	TMAH-oxidative methylotrophic degradation	Ferella et al., 2021

transferred to the thiol group of coenzyme M (H-S-CoM) under methyltransferase (Mtr), generating a one-carbon carrier Coenzyme M (CH₃-S-CoM), and promoting adenosine triphosphate (ATP) synthesis through the formation of the ionic gradient inside and outside the cell membrane (Chang et al., 2008; Hu et al., 2018; Shima et al., 2022). Then

CH₃-S-CoM interacted with coenzyme B to produce CH₄ and heterodisulfide (CoB-S-S-CoM) under methyl coenzyme M reductase (Mcr) (Wagner et al., 2017; Shima et al., 2022). Subsequently, the CoB-S-S-CoM was again reduced to H-S-CoM and CoB-S-H by electron disproportionation under the heterodisulfide reductase / [NiFe] -

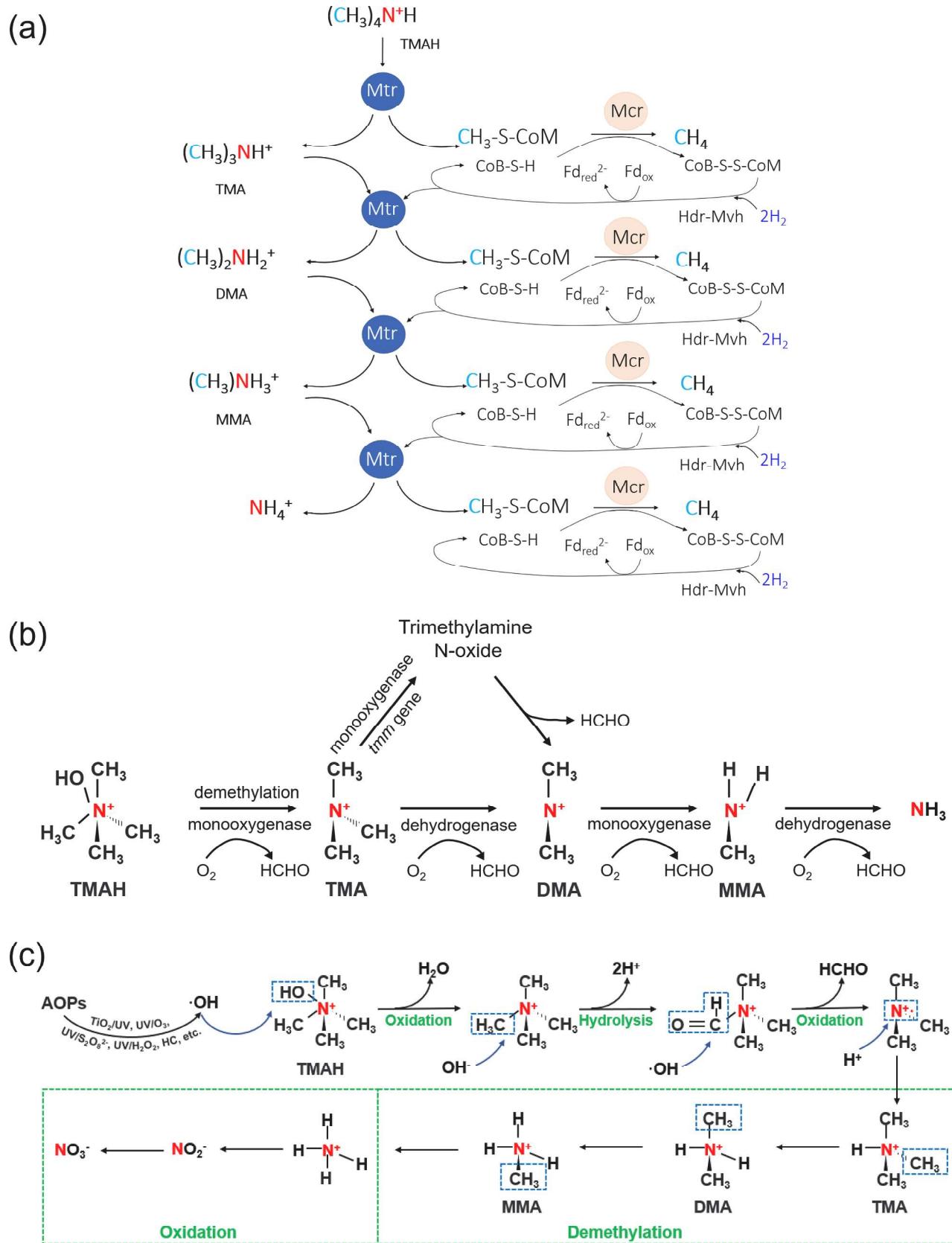


Fig. 4. The main degradation mechanisms of TMAH in wastewater. (a) The main mechanisms of methanogenic TMAH degradation; (b) The proposed pathways of TMAH-oxidative methylotrophic degradation; (c) The proposed pathways of TMAH degradation by AOPs.

hydrogenase (Hdr-Mvh) (Wagner et al., 2017). The main characteristics of methanogenic TMAH degradation were that methyl groups were successively transferred from TMAH to H-S-CoM under Mtr to generate CH₃-S-CoM, and then CH₃-S-CoM entered the methanogenic metabolism process (Wagner et al., 2017; Conrad., 2020).

The characteristics of these functional methanogens have been investigated in a few lectures (Hu et al., 2012; Whang et al., 2015). For example, Whang et al. (2015) investigated the methanogenic communities involved in TMAH degradation in a full-scale UASB. Their results revealed that at higher TMAH concentrations (> 1000 mg/L), the *mcr* alpha subunit (*mcr A*) mRNA was obviously expressed by *Methanosarcina*, while at lower TMAH concentrations (< 1000 mg/L), it was considerably expressed by *Methanomethyllovorans*. As a result, *Methanosarcina* might play a dominant role in methanogenic TMAH degradation at higher TMAH concentrations (> 1000 mg/L), while *Methanomethyllovorans* might be important at a lower TMAH condition (< 1000 mg/L). These similar results were also reported by Hu et al. (2012; 2018). It is generally known that *Methanosaeta* could use acetic acid as the sole source of energy to generate CH₄ (Nedwell et al., 2016). Hence, the acetic acid during the TMAH wastewater treatment might be produced mainly by the degradation of other organic compounds (Chang et al., 2022).

The kinetics of the methanogenic TMAH degradation were also analyzed (Liu et al., 2016; Hu et al., 2018). The kinetic and stoichiometric coefficients were summarized in Table 4. Hu et al. (2018) reported that under 1000 mg/L TMAH, the kinetic of methanogenic TMAH degradation followed the Monod-type model, with a TMAH degradation rate of 10.4 mmol/g VSS/d, and a half-saturation coefficient of 820 mg TMAH/L. Liu et al. (2016) reported the kinetic of methanogenic TMAH degradation followed the Monod-type model, with a biomass yield of 0.14 g COD/g TMAH, a maximum specific growth rate of 0.372 d⁻¹, and a specific decay rate of 0.017 d⁻¹.

(3) Influence factors on TMAH degradation

A few influence factors on methanogenic TMAH degradation also have been investigated, such as TMAH concentration (Hu et al., 2012; Urasaki et al., 2019; Lv et al., 2023), sulfate (Hu et al., 2012, 2018; Urasaki et al., 2019), surfactants (Hu et al., 2012), and co-digestion substances (Choi et al., 2021), etc. For example, Hu et al. (2018) investigated the effect of TMAH, sulfate, and sulfide on methanogenic TMAH degradation. Their results demonstrated that TMAH below 2000 mg/L showed no significant influence on the TMAH degradation rate, while with sulfate from 0 to 300 mg/L, the maximum TMAH degradation rate decreased from 40.8 to 35.6 mg-TMAH gVSS⁻¹ h⁻¹, and adding

sulfide from 0 to 20 mg/L caused its rate to drop from 35.5 to 15.1 mg-TMAH gVSS⁻¹ h⁻¹.

Urasaki et al. (2019) also investigated the effect of TMAH and sulfate shock load on methanogenic TMAH degradation. Their results indicated that under TMAH of 11,000 mg-COD/L and HRT of 5.0 h (52.8 kg-COD/m³ d), after 10 days, TMAH removal was significantly weakened, just 4%, and sulfate reduction also stopped. Hu et al. (2018) also noted that methanogenesis began to be inhibited by 9450 mg-COD/L of TMAH.

The mechanisms for inhibition of excess TMAH to methanogenesis might be attributed to excess ammonia produced during the degradation of TMAH (Lei et al., 2010; S.Y. Chen et al., 2016; Urasaki et al., 2019). Ammonia inhibition to methanogenesis could occur when total ammonia exceeds 1000 mg-N/L and free ammonia exceeds 100 mg-N/L (Li et al., 2015). Under the TMAH of 11,000 mg-COD/L shock load, ammonia of over 800 mg-N/L could be produced, which was obviously higher than the inhibition level (100 mg-N/L) (Urasaki et al., 2019).

The possible mechanisms for the inhibition of sulfide on methanogenesis might be explained from two aspects. On one hand, the competition for common substrates between the sulfate-reducing bacteria (SRB) and methanogens could decrease CH₄ production (Harada et al., 1994; Hu et al., 2012). On the other hand, the sulfide produced during sulfate reduction could cause the inhibition of methanogens in anaerobic bioreactors (Hu et al., 2012).

In addition, Choi et al. (2021) investigated the performance of TMAH wastewater treatment using anaerobic biotreatment via co-digestion with ethyl lactate (EL) wastewater. These two wastewaters are significant organic wastewaters from micro-electronic manufacture. Their results demonstrated that the CH₄ production rate and yield were obviously increased under the co-digestion with TMAH and EL wastewaters, indicating that this co-digestion effect was synergistic. The optimal mixing ratio of TMAH and EL wastewaters for co-digestion was between 50:50 and 25:75 (total carbon basis).

Although TMAH wastewater could be efficiently treated by the anaerobic biotreatment process, the research could be further carried out in the following aspects. Firstly, currently, the mechanisms of methanogenic TMAH degradation mostly focused on degradation pathways, kinetics, etc., while the energy metabolism and electron transfer mechanism are still unclear. It is meaningful for the development of enhanced anaerobic biotreatment to reveal the energy metabolism and electron transfer mechanism.

Secondly, since a large amount of wastewater from the micro-electronic industry is generated, it also could be a cost-effective option that the anaerobic biotreatment for TMAH wastewater was performed via co-digestion with other organic wastewaters from the micro-

Table 4

The kinetic and stoichiometric coefficients under anaerobic and aerobic TMAH degradation.

Biotreatment process	Biochemical Process	Maximum specific rate	Half-saturation coefficient	Process rate expression	Ref.
Anaerobic biodegradation	CH ₄ production from TMAH	10.4 mmol/g VSS/d	820 mg TMAH/L	Monod-type	Hu et al., 2018
	TMA production from TMAH	13.0 mmol/g COD/d (2.0 g COD/g COD/d)	15 mg COD/L	Monod-type	Liu et al., 2016
	DMA production from TMA	14.0 mmol/g COD/d (2.0 g COD/g COD/d)	0.06 mg COD/L	Monod-type	Liu et al., 2016
	MMA production from DMA	26.0 mmol/g COD/d (2.5 g COD/g COD/d)	0.025 mg COD/L	Monod-type	Liu et al., 2016
	MeOH production from MMA	29.8 mmol/g COD/d (1.4 g COD/g COD/d)	0.15 mg COD/L	Monod-type	Liu et al., 2016
	Growth of TMAH-degraders	0.37 d ⁻¹	0.2 mg COD/L	Monod-type	Liu et al., 2016
	Decay of TMAH-degraders	0.017 d ⁻¹		First order-type	Liu et al., 2016
	Decay of acetotrophic methane-producing organisms	0.017d ⁻¹		First order-type	Liu et al., 2016
	Decay of hydrogenotrophic methane-producing organisms	0.017 d ⁻¹		First order-type	Liu et al., 2016
Aerobic biodegradation	TMAH-oxidative methylotrophic degradation	3.74 mmol/g VSS/d	1140 mg TMAH/L	Monod-type	Wu et al., 2020
	Growth of TMAH-degraders	1.008 d ⁻¹		Monod-type	Innocenzi et al., 2019

electronic industry. Meanwhile, the mechanisms of co-digestion for enhancing TMAH treatment could be also further studied.

Thirdly, it was recommended that the anaerobic biotreatment process should be started-up with a steady feeding and then a gradual increase of TMAH loading due to the slow growth and sensitivity of methanogens to the external environment. In addition, potential inhibitors such as surfactants and sulfates have to be also avoided during the methanogenic TMAH degradation for the stability of the anaerobic biotreatment process. Hence, the pre-treatment process has to be operated before the anaerobic biotreatment process.

Furthermore, the granulation of the methanogens population is also the key to the stable methanogenic TMAH degradation, which is closely associated with temperature, seed sludge, and hydraulic characteristics of reactors (Rajakumar et al., 2012). It is also necessary to investigate the formation and stability mechanism of granules for methanogenic TMAH degradation for stable operation in the future.

4.1.2. Aerobic biodegradation

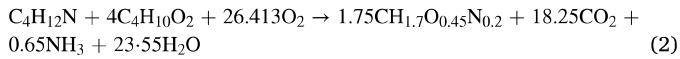
Recently, some studies have investigated the TMAH wastewater treatment by aerobic biotreatment process, which is summarized in Table 2.

(1) Degradation pathways of TMAH by aerobic biodegradation

TMAH could be biodegraded by an aerobic process, in which the methyl groups from methylated amines could be oxidized to formaldehyde, then the formaldehyde could be assimilated (Lin et al., 2016). The main mechanism of TMAH aerobic biodegradation was TMAH-oxidative methylotrophic degradation (Lv et al., 2021). The proposed pathways of TMAH-oxidative methylotrophic degradation were shown in Fig. 4(b).

According to Fig. 4(b), under aerobic conditions, firstly, TMAH could be degraded to TMA via the demethylation pathway under tetramethylammonium monooxygenase (Chistoserdova et al., 2013). Secondly, TMA could be further oxidized to DMA and formaldehyde through two different pathways. One was that TMA was directly oxidized to DMA under trimethylamine dehydrogenase. The other was that TMA was first oxidized to trimethylamine N-oxide under trimethylamine monooxygenase encoded by the *tmm* gene, then to DMA (Chen et al., 2012). Thirdly, DMA was oxidized to MMA and formaldehyde under dimethylamine monooxygenase. MMA was then further degraded to NH₃ and formaldehyde through three different pathways, including from MMA directly to NH₃ under methylamine dehydrogenase (MADH), through the conversion of MMA to N-methyl glutamate and then to methylene-H₄F under N-methyl glutamate dehydrogenase, as well as through blue copper amine oxidase (Chistoserdova et al., 2013).

The stoichiometry of aerobic TMAH degradation reaction using TMAH and photoresist (C₄H₁₀O₂) as substrates was also determined. The details were as follows (Eq. (2)) (Innocenzi et al., 2019),



(2) Functional bacteria and kinetics

The main functional bacteria for TMAH-oxidative methylotrophic degradation also were investigated, which was summarized in Table 3. According to Table 3, there are some differences in the functional flora in various aerobic biological processes. For example, the predominant genera for TMAH-oxidative methylotrophic degradation in the membrane bioreactor (MBR) were *Hypomicrobium* and *Methyloversatilis* (Wu et al., 2008). Wu et al. (2020) reported that the genera *Hypomicrobium*, *Mycobacterium*, and *Methyloversatilis* were the main functional bacteria for TMAH-oxidative methylotrophic degradation in the sequencing batch reactor (SBR). Ferella et al. (2021) revealed that the genera *Methyloversatilis*, *Delftia*, *Mycobacterium*, and *Serratia* were predominant in the continuous stirred tank reactor (CSTR) for treating TMAH

wastewater. These different results might be attributed to various process types, operation conditions, etc.

Genera *Methyloversatilis*, belonging to the Actinobacteria phylum, is a well-known methylophiles, and has been found to grow on methanol and methylamine. Its presence could suggest the community-driven TMAH degradation, and that methanol and MMA, DMA and TMA are the products of TMAH-oxidative methylotrophic degradation by *Methyloversatilis* (Chang et al., 2019). *Serratia*, belonging to the Proteobacteria phylum, as the methylophiles, could use TMAH as the sole nitrogen and carbon source (Mahlen et al., 2011). *Hypomicrobium* belongs to the Proteobacteria phylum, and has been reported to be capable of growing on TMAH under aerobic condition (Chistoserdova et al., 2013). *Delftia*, belonging to the Proteobacteria phylum, includes several facultative methylotrophic species capable of degrading organic compounds (Khaila et al., 2019).

In addition, some ammonium- (AOB) and/or nitrite-oxidizing (NOB) bacteria, such as *Achromobacter*, *Aeromonas*, *Comamonas*, *Desulfovibrio*, *Flavobacterium*, *Microvirogula*, *Rhizobium*, were also observed in the aerobic systems for TMAH treatment (J. Huang et al., 2017; Chen et al., 2015; S.Y. 2016; Zhang et al., 2016; Feng et al., 2019; Murphy et al., 2012). These bacteria could furtherly promote the oxidation of NH₄⁺-N into nitrate (NO₃⁻). Therefore, a further denitrification stage has to be required to achieve TN removal after the aerobic biotreatment process.

It was worth noting that the biochemical oxygen demand (BOD)/total kjeldahl nitrogen (TKN) ratio of TMAH wastewater generally was only 0.30~1.03 (Omar et al., 2008), causing a negative impact on microbial growth and denitrification due to the inadequate supply of carbon sources to microbial cells (J. Huang et al., 2017; Ferella et al., 2021). Recently, S.Y. Chen et al. (2016) employed a continuous anoxic up-flow bioreactor using a completely autotrophic nitrogen removal over nitrite (CANON) technique for TMAH wastewater treatment. Their results showed that under TMAH of 200 ~ 1000 mg/L, the removal rate of TMAH and total nitrogen (TN) reached about 100% and over 90%, respectively. And in the CANON process, TMAH was first degraded into NH₄⁺-N by TMAH-oxidative methylotrophic degradation, and then sequentially transformed into NO₂⁻, NO₃⁻, and nitrogen gas. Although the CANON process could achieve efficient TMAH removal, in fact, due to the long generation cycle of autotrophic bacteria, the technology not only requires a larger volume of equipment, but also is usually accompanied by N₂O emissions (Schultheiss et al., 2009).

In addition, the kinetics of TMAH-oxidative methylotrophic degradation were also investigated (Table 4). This kinetic followed the Monod-type model well ($R^2 = 0.9002$). The maximum specific TMAH degradation rate was 3.74 mmol/g VSS/d, and the half-saturation coefficient for TMAH was 1140 mg TMAH/L (Wu et al., 2020). According to Table 4, it is worth noting that the maximum specific TMAH degradation rate under aerobic conditions is obviously lower than that (10.4 mmol/g VSS/d) under the anaerobic (Hu et al., 2018), suggesting that methanogenic TMAH degradation could be favored over TMAH-oxidative methylotrophic degradation. These results might be attributed to the molecular structures and properties of TMAH. Due to its high electrostatic potential energy (LP (+100.22–100.31 kcal/mol); HP (+128.55 kcal/mol)), TMAH is generally more difficult to be degraded by oxidation than by reduction. Therefore, the efficiency of TMAH degradation by the aerobic is lower than that by the anaerobic. In addition, TMAH and its decomposition products, including DMA and MMA could cause obvious inhibition to the aerobic communities. Under TMAH of 150 mg/L, the activity of activated sludge could start to be inhibited (Hu et al., 2012), and under TMAH of 500 mg/L, the activity of AOB could be obviously reduced (Lin et al., 2016). The inhibition of TMAH to nitrification could be attributed to the inactive expression of amoA gene. Specifically, TMAH and its intermediates, including DMA, and MMA, might compete with the enzyme ammonia monooxygenase (AMO), causing enzyme structure destruction (Arp et al., 2003; Wu et al., 2020).

The disadvantages of aerobic biodegradation were the problems

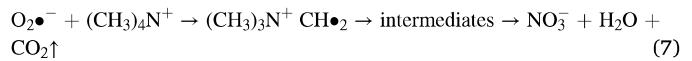
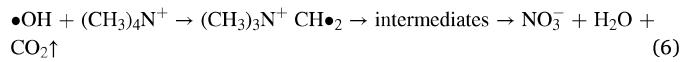
connected with low removal efficiency and long HRT compared to anaerobic biodegradation (Table 2). And the high TMAH concentrations could cause a significant inhibition to the aerobic communities, especially nitrobacteria. Therefore, this method could be more suitable for degrading low-concentration TMAH wastewater or could serve as a follow-up process to the anaerobic biotreatment process. And a further denitrification step has to be required after this method. In addition, the inhibition mechanism of TMAH to nitrification still needs further clarification using state-of-the-art techniques, such as meta-omic analyses and stable isotope labeling, etc.

4.2. Advanced oxidation process

In recent years, advanced oxidation processes (AOPs) have been conducted for the TMAH wastewater treatment, including TiO_2/UV , UV/O_3 , $\text{UV}/\text{S}_2\text{O}_8^{2-}$, $\text{UV}/\text{H}_2\text{O}_2$, hydrodynamic cavitation (HC), etc. (Fig. 1(b)), which is summarized in Table 5.

(1) Degradation mechanisms of TMAH by AOP

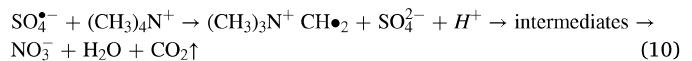
The ability of these AOPs to degrade TMAH is mainly dependent on generating radicals with strong oxidation potentials, such as hydroxyl radicals ($\bullet\text{OH}$), superoxide radicals ($\text{O}_2^{\bullet-}$), and sulfate radicals ($\text{SO}_4^{\bullet-}$). For example, the generation of $\bullet\text{OH}$ and mechanisms of TMAH degradation by the TiO_2/UV process were shown as follows (Eq. (3) ~ (7)) (Kim et al., 2002; Riaz et al., 2020),



Wang et al. (2014) and J. Huang et al. (2017) have demonstrated that $\text{UV}/\text{S}_2\text{O}_8^{2-}$ could be capable of TMAH degradation under acidic conditions (Table 5). Under UV irradiation, 2 mol of sulfate radicals could be generated per mole of $\text{S}_2\text{O}_8^{2-}$. The generation of sulfate radical was shown as follows (Eq. (8) ~ (9)),



The reactions mechanisms of TMAH degradation by the $\text{UV}/\text{S}_2\text{O}_8^{2-}$ process were shown as follows (Eq. (10)) (Liang et al., 2009; Wang et al., 2014; J. Huang et al., 2017),



HC could primarily occur through the dissociation of water molecules into hydroxyl radicals under high temperature and pressure conditions (Rajoriya et al., 2016). Innocenzi et al. (2018) demonstrated that the removal rate of TMAH was around 44% by the HC process under a pressure of 4 bar and pH of 3.00 (Table 5). The generation of $\bullet\text{OH}$ by HC process was shown in Eq. (11) ~ (12) (Rajoriya et al., 2016; Innocenzi et al., 2018, 2022). And the reactions mechanisms of TMAH degradation by the HC process were also similar to Eq. (6) (Innocenzi et al., 2018, 2022).



(2) Degradation pathways of TMAH by AOP

In addition, the degradation pathways of TMAH by AOPs were also investigated by analyzing the intermediates. For example, Kim et al.

Table 5
Recent studies on TMAH treatment in wastewater using AOPs.

AOP methods	TMAH (mg/L)	Conditions	Reaction time (min)	pH	Temperature (°C)	k_{obs} (min ⁻¹)	Performance effect	Ref.
TiO_2/UV (90 mL)	9.12	TiO_2 0.5 g/L,	360	11.00	Room		The removal rate of TMAH was 100%, but the intermediates, such as DMA, MMA, NO_2^- still were remained.	Kim et al., 2002
UV/O_3	40	UV intensity 8 W (254 nm)	60	10.00	Room	0.005	The mineralization rate of TMAH was $\text{UV}/\text{O}_3 > \text{UV}/\text{H}_2\text{O}_2/\text{O}_3 > \text{H}_2\text{O}_2/\text{SiO}_2/\text{Fe}_3\text{O}_4/\text{O}_3 > \text{H}_2\text{O}_2/\text{O}_3 > \text{SiO}_2/\text{Fe}_3\text{O}_4/\text{O}_3 > \text{O}_3 > \text{UV}/\text{H}_2\text{O}_2$. The mineralization rate of TMAH was 87.6% by UV/O_3 .	Chiou et al., 2013
$\text{UV}/\text{S}_2\text{O}_8^{2-}$ (3 L)	100.27	UV intensity 15 W (254 nm), $\text{S}_2\text{O}_8^{2-}$ 50 mM	130	2.00	30	0.0331 ± 0.0031	The removal rate of TMAH was 100%, and NO_3^- and NH_4^+ was still the final products.	Wang et al., 2014
$\text{UV}/\text{S}_2\text{O}_8^{2-}$ (3 L)	100	UV intensity 15 W (254 nm), $\text{S}_2\text{O}_8^{2-}$ 50 mM	98	2.00	24 ± 2	0.00986	The removal rate of TMAH was 80%. And the LC ₅₀ and TU _a of effluent was 70.69% and 1.41.	J. Huang et al., 2017
$\text{AC/Ag-O}_3/\text{H}_2\text{O}_2$	500	1.5 g/L AC/Ag, H_2O_2 8.8 mM,	60	11.00	Room	0.0191	The removal rate of TMAH was 81%, and NO_3^- and NH_4^+ were the final products, containing 167 mg/L and 102 mg/L.	Lu et al., 2018
$\text{MnO}_2/\text{H}_2\text{O}_2$	646	175 g of 60 μm mesh powdered MnO_2 , H_2O_2 3000 mg/L	480		Room		The catalyst MnO_2 improved the decomposition rate of H_2O_2 by about 1000 times compared to the uncatalyzed reaction.	Imtisdal et al., 2019
nano- $\text{O}_3/\text{H}_2\text{O}_2$	190	H_2O_2 100 mg/L	90	10.00	25	0.0146	The removal rates of TMAH and TOC were 95% and 65%. And about 80% of TN was transformed to NO_3^- .	Kim et al., 2021
$\text{UV}/\text{H}_2\text{O}_2$	5.01	UV 265 nm, H_2O_2 59.3 mg/L	30	7.00	35	0.0064	Although the removal rate of TMAH was 100%, NO_3^- and NH_4^+ were the final products. And these nitrogen byproducts could cause toxicity to <i>Alivibrio fischeri</i> .	Kim et al., 2022
Hydrodynamic cavitation	2000	A pressure of 4 bar	20	3.00	20		The removal rate of TMAH was around 44% under optimal condition.	Innocenzi et al., 2018

(2022) identified that TMA, DMA, and MMA were the dominant demethylated by-products, and NH_4^+ was the nitrogen by-product during the TMAH degradation by the UV/ H_2O_2 process. These results were consistent with that of UV/ $\text{S}_2\text{O}_8^{2-}$ (Wang et al., 2014; J. Huang et al., 2017), TiO_2/UV (Kim et al., 2002), and AC/Ag-O₃/ H_2O_2 (Lu et al., 2018).

According to these previous lectures (Kim et al., 2002; Wang et al., 2014; J. Huang et al., 2017; Lu et al., 2018; Kim et al., 2022), the proposed degradation pathways of TMAH by AOP included demethylation and oxidation, which is summarized in Fig. 4(c). The degradation of TMAH could be mainly attributed to the attack of radicals with strong oxidation potentials. First, hydrogen (H) could be transferred from the methyl moiety bound to the nitrogen atom of TMAH, followed by the generation of a carbon-centered radical. Second, $(\text{CH}_3)_3\text{N}^+\text{CHO}$ could be produced by the hydrolysis of carbon-centered radicals. Subsequently, TMA radicals ($(\text{CH}_3)_3\text{N}^+\bullet$) and formaldehyde could be generated through $\bullet\text{OH}$ -induced oxidation of $(\text{CH}_3)_3\text{N}^+\text{CHO}$. Then $(\text{CH}_3)_3\text{N}^+\bullet$ could be stabilized by forming TMA ($(\text{CH}_3)_3\text{N}^+\text{H}$). Then, sequential TMA demethylation could result in the production of DMA and MMA via radicals and/or oxidation of oxidizing agents, such as UV, H_2O_2 , and O₃, etc. Finally, MMA could be transformed to NH_4^+ by demethylation, followed by the sustained oxidation of NH_4^+ to NO₃⁻.

Although AOPs could achieve a high removal rate of TMAH in wastewater, the high cost of their implementation should not be ignored. Hence, this method could be suitable for degrading low-concentration TMAH wastewater or could be recommended as an advanced treatment. In addition, the investigation of the enhanced efficiency of radical generation, namely increasing kinetic rate constant k_{obs} , by methods such as developing low-cost and high-efficiency catalytic materials, is also one of the aspects to promote the AOPs application for TMAH wastewater.

4.3. Hybrid treatment process

To enhance the treatment efficiency of TMAH wastewater, some hybrid treatment processes have been employed in some lectures, such as the UV/ H_2O_2 + aerobic biotreatment process (Den et al., 2002), UV/ $\text{S}_2\text{O}_8^{2-}$ + aerobic biotreatment process (J. Huang et al., 2017), aerobic biotreatment + coagulation process (Innocenzi et al., 2019), hydrolysis acidification + aerobic biotreatment process (Lv et al., 2021), etc.

Den et al. (2002) investigated the feasibility of a two-stage process combining a UV/ H_2O_2 and a biological fluidized-bed process to treat TMAH wastewater. Their results showed that under TMAH of 500 mg/L in wastewater, the TMAH removal rate was > 95% by the combined process, which was higher than that (29%) by the single UV/ H_2O_2 process and that (68%) by the single biological fluidized-bed process.

J. Huang et al. (2017) employed a UV/ $\text{S}_2\text{O}_8^{2-}$ combined with CSTR process to treat TMAH wastewater. Their results demonstrated that the TMAH removal rate was about 80% by the combined process under TMAH of 100 mg/L. And the “bio-acute toxicity unit” (TU_a) to *Cyprinus carpio* was only 1.41, which was less than that (1.43) of the discharge standard.

Innocenzi et al. (2019) took an attempt to implement a sequence combined process using a continuous model BIOSTAT B process associated with a batch coagulation of aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$) for TMAH wastewater treatment. The results showed that about 99% of degradation to TMAH was achieved under TMAH of 1800 ~ 2500 mg/L. And the effluent could be directly reused and/or discharged to surface waters.

Lv et al. (2021) investigated the effect of TMAH wastewater treatment using hydrolysis acidification + aerobic biotreatment process. Their results demonstrated that the TMAH removal rate was over 91%. The pre-treatment by hydrolytic acidification could be favored over that by the anaerobic methanogenic process to prevent the inhibition of TMAH in the influent to following aerobic biotreatment processes.

According to these lectures, the removal efficiencies of TMAH from wastewater using various technologies, including anaerobic biodegradation, aerobic biodegradation, AOPs, and hybrid treatment process were statistically analyzed in Fig. 5. According to Fig. 5, the TMAH concentration treated by anaerobic biodegradation, aerobic biodegradation, and hybrid treatment process was from 100 mg/L to over 1000 mg/L, while that by AOPs mostly was < 200 mg/L. In addition, the order of HRT was aerobic biodegradation > hybrid treatment process > anaerobic biodegradation > AOPs, while the average removal rate of TMAH was hybrid treatment process (95.7%) > anaerobic biodegradation (89.4%) > AOPs (82.4%) > aerobic biodegradation (76.2%). It could be concluded that among these treatment technologies for TMAH wastewater, the hybrid treatment process not only could most efficiently treat high-concentration TMAH wastewater (> 1000 mg/L), its HRT but also could be moderate. But so far, the studies about the hybrid treatment process for TMAH are still fewer. Hence, an appropriate hybrid treatment process should be developed for the high-efficiency treatment of TMAH wastewater. Meanwhile, the coordinated control and optimization of the hybrid treatment process need to be further expanded by digital and intelligent technologies.

5. Challenges and perspectives for TMAH wastewater treatment

With the rapidly growing demand for micro-electronic products worldwide, TMAH wastewater generated by the micro-electronic manufacturing industry has been increasing dramatically. The efficient treatment of TMAH wastewater has received growing attention due to its toxicity to the eco-environment and human health. However, several challenges to the development of TMAH wastewater treatment still exist. Therefore, further research should be necessarily carried out in the following aspects to promote the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater.

- (1) Due to the complexity of actual wastewater components, firstly, the adsorbents with higher adsorption capacity, lower cost, stronger selectivity and easier recovery used for TMAH should be developed through collaboration with other fields, such as materials and chemistry. Secondly, the membrane processes, especially MD, UF, NF, and RO, could be recommended to use for the recovery and reuse of TMAH wastewater. Meanwhile, the fouling characteristics, cleaning methods, and membrane materials for TMAH recovery and water reuse also should be investigated.
- (2) From the perspective of environmental protection, the biological treatment process should be recommended as the high-efficiency and cost-effective treatment technology for TMAH. However, firstly, so far the energy metabolism and electron transfer mechanism of methanogenic TMAH degradation are still unclear. Secondly, the potential inhibitors, such as surfactants and sulfates, have to be pre-treated for the stability of the anaerobic biotreatment process. In addition, it also could be a cost-effective option that the anaerobic biotreatment for TMAH wastewater was performed via co-digestion with other wastewaters from the micro-electronic industry. Thirdly, the inhibition mechanisms of TMAH to nitrification still need further clarification using state-of-the-art techniques, such as meta-omic analyses and stable isotope labeling, etc.
- (3) Although AOPs could achieve a high removal rate of TMAH in wastewater, the high cost of their implementation should not be ignored. Hence, this method should be more suitable for degrading low-concentration TMAH wastewater or be recommended as an advanced treatment. In addition, the investigation of the enhanced efficiency of $\bullet\text{OH}$ radical generation is also one of the aspects to promote the AOPs application for TMAH wastewater.
- (4) Currently, these studies about TMAH wastewater mainly focused on one-way linear treatment of TMAH wastewater, while the

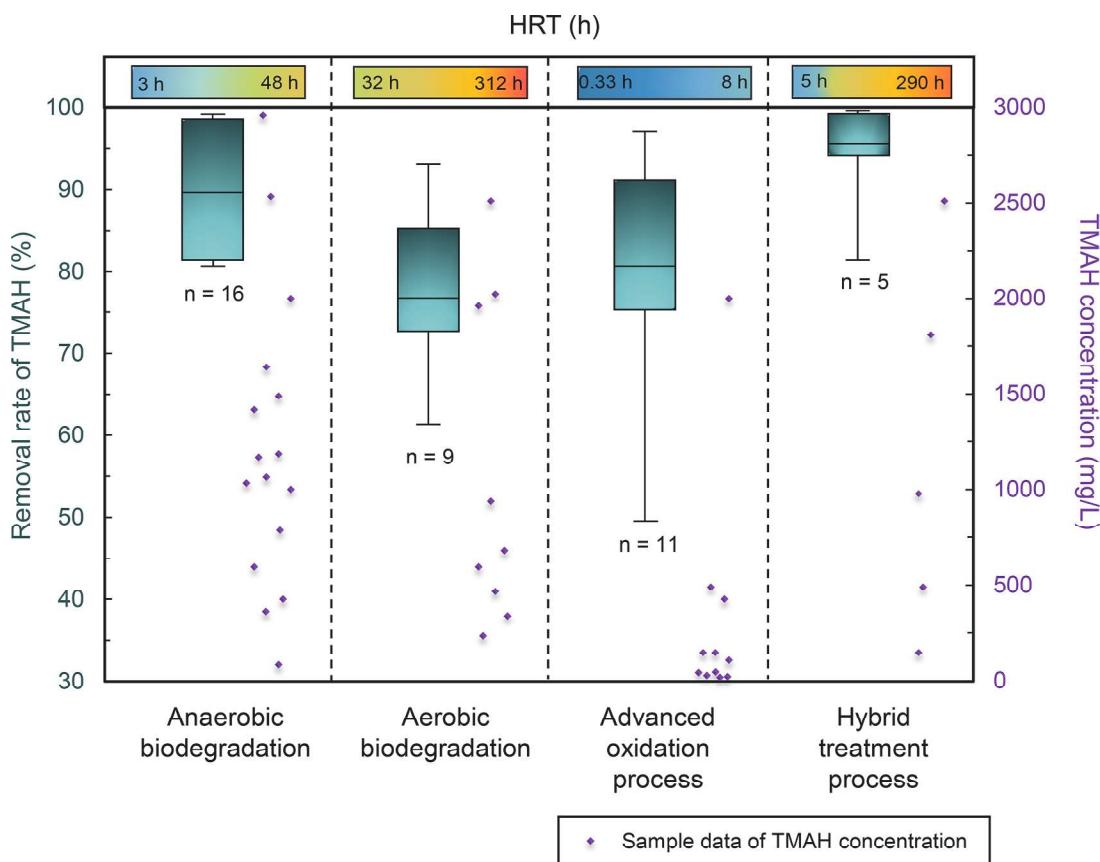


Fig. 5. Removal efficiencies of TMAH in wastewater using anaerobic biodegradation, aerobic biodegradation, advanced oxidation process, and hybrid treatment process; n = number of samples.

close-loop treatment of TMAH wastewater, namely the multi-way resource reuse is also still less focused. In order to promote the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater, three aspects could be its main driven forces, including recovery, resource and reuse (“Three Re”) of TMAH wastewater, core equipment upgrade, as well as digital and intelligent operation and maintenance (O&M) (Fig. 6(a)). “Three Re” was the main action to drive the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater. The innovation in hybrid processes, including hybrid recovery and hybrid treatment, is one of its keys. The possible implementation pathways of “Three Re” for TMAH wastewater were shown in Fig. 6(b).

- (5) The appropriate hybrid recovery and treatment processes should be developed for “Three Re” of TMAH wastewater. Firstly, due to the complexity of actual wastewater components, the interference of other organics in the wastewater with TMAH recovery should not be ignored. Hence, to improve TMAH recovery efficiency and reduce operation cost, the pre-removal of other organic matters in wastewater, such as photoresist, and the pre-concentration of TMAH should be considered in the hybrid recovery processes. The pre-removal technologies of other organic matters included UF, NF, etc. The pre-concentration technologies of TMAH were ion exchange, MD, ED, etc. (Fig. 6(b)). As a result, the hybrid recovery process, i.e., ED + ion exchange, ED + adsorption, NF + ion exchange, etc., was recommended to use for the recovery and purification of TMAH from wastewater. Then the high-efficiency treatment of residual TMAH in wastewater should be conducted by the appropriate hybrid treatment processes. During the treatment, resource recovery, i.e., CH₄, N, hydrogen, etc., and water reuse should be carried out (Fig. 6(b)).

(6) The core equipment upgrade, and digital and intelligent O&M were the important guarantees to drive the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater. For example, membrane regeneration should be recommended to use for the aging issue of the membrane process, decreasing the cost of new membrane preparation and waste membrane disposal. The digital and intelligent O&M could achieve the co-ordinated control and optimization of the hybrid recovery / treatment process by artificial intelligence (AI) modeling and diagnosis, real-time monitoring, intelligent feedback, optimization, etc.

6. Conclusions

With the rapidly-growing demand for micro-electronic products worldwide, TMAH wastewater generated by micro-electronic manufacture has been increasing significantly. TMAH wastewater has to be efficiently treated to avoid its toxicity to the eco-environment and human health. To date, the treatment technologies for TMAH wastewater included recovery methods and degradation methods. The recovery methods of TMAH from wastewater included adsorption, ion exchange, NF, MD, ED, and the hybrid, while its degradation methods included anaerobic biodegradation, aerobic biodegradation, AOPs, and the hybrid. The hybrid recovery and treatment processes should be recommended for the TMAH wastewater treatment. The early works mainly focused on the one-way linear treatment of TMAH wastewater, while the multi-way resource reuse of TMAH wastewater is still poorly focused. Further scope of the study includes efficient novel materials for TMAH recovery and degradation, biodegradation mechanisms including energy metabolism, electron transfer, co-digestion, and inhibition of TMAH to nitrification, as well as an appropriate hybrid recovery and

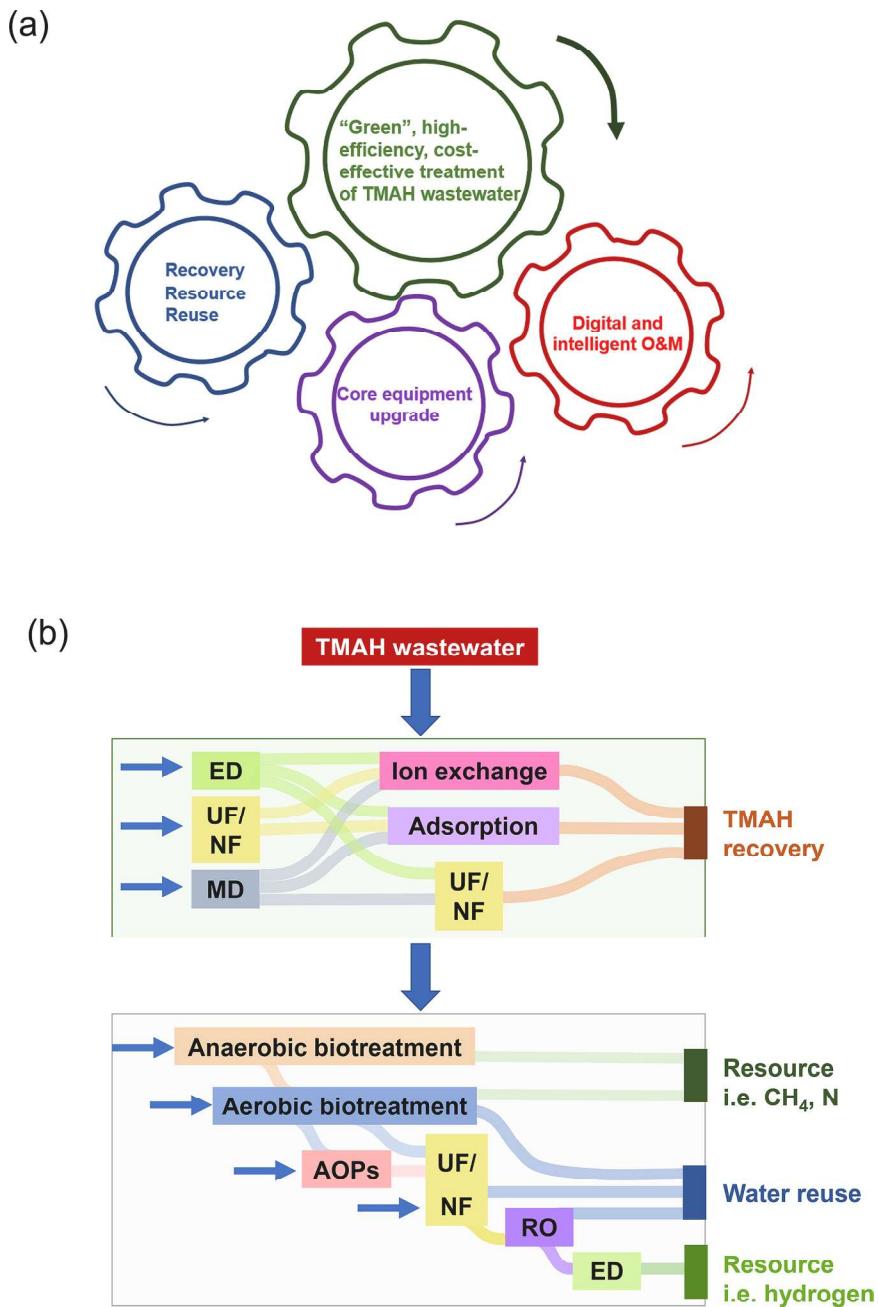


Fig. 6. Possible pathways for the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater. (a) The main driven forces to promote the “green”, high-efficiency, and cost-effective treatment of TMAH wastewater; (b) The possible implementation pathways of “Three Re” for TMAH wastewater.

treatment processes for the resource reuse of TMAH wastewater. Ultimately, the possible pathways for the resource reuse of TMAH wastewater were proposed. The present work is expected to promote the “green”, high-efficiency and cost-effective treatment of TMAH wastewater.

CRediT authorship contribution statement

Zhuo-Wei Zhang: Methodology, Formal analysis, Writing – original draft, Data curation. **Yin-Hu Wu:** Formal analysis, Resources, Supervision, Writing – review & editing. **Zhuo-Yuan Xiao:** Methodology, Formal analysis, Writing – review & editing. **Zhao Zhang:** Methodology, Formal analysis, Writing – review & editing. **Qi Wang:** Methodology, Formal analysis, Writing – review & editing. **Yu-Qing Xu:** Methodology, Formal analysis, Writing – review & editing. **Jia-Hao Luo:** Formal

analysis, Resources, Supervision. **Jiang-Lei Xiong:** Formal analysis, Resources, Supervision. **Hong-Ying Hu:** Writing – review & editing, Supervision, Funding acquisition.

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.

This work has not been published before in whole or in part or in any language, and it is not under consideration for publication anywhere else. Its publication has been approved by all co-authors as well as by the corresponding author.

Data availability

No data was used for the research described in the article.

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