A critical review on challenges and trend of ultrapure water production process

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Abstract

The recent and vigorous developments in semiconductor technology strictly request better quality and large quantity of ultrapure water (UPW) for their production. It is crucial to secure a large amount of raw water for the future development of UPW production. Using reclaimed water as alternative raw water source to produce UPW is therefore considered the feasible trend and solution for sustainable use of water resources towards a common future practice in UPW production. The challenge of using reclaimed water is due to its higher content of organic pollutants, especially small molecule organic pollutants such as urea, which are difficult to remove through traditional UPW production process. Consequently, improving the existing UPW production process to meet the water standard desired in the semiconductor industry is essential. This paper reviewed the current traditional processes for removing organic matters in UPW production, including ion-exchange (IX) adsorption, granular activated carbon (GAC) adsorption, reverse osmosis (RO) and ultraviolet (UV) irradiation. The potential problems in the actual UPW production process were identified when using reclaimed water as raw water source. A new strategy of applying the advanced oxidation process (AOPs) to UPW production as a supplementary unit to guarantee UPW quality was proposed. Its feasibility and research focus were then analyzed and discussed in obtaining a new solution for a future development of the UPW production process.

Keywords: Ultrapure water; Organic; Urea; Advanced oxidation

1. Introduction

Ultrapure water (UPW) is of high purity and with almost no other electrolytes

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except hydrogen and oxygen ions, which is usually used in the electronics industry, photovoltaic industry, and pharmaceutical industry. Calculations based on ionic mobilities yield a specific resistance of ideal UPW is 18.3 MΩ•cm at 25℃ (Singh, 2016). Hence, ordinary distilled water, reverse osmosis water, and high-purity water (resistivity ≥ 10.0 MΩ•cm) are not considered to be UPW (Singh, 2016). In addition, UPW must be devoid of all contaminants including pathogens, suspended solids, radioactive contaminant, inorganic compounds and organic compounds in ideal condition, so it is also called "highly refined water" (Choi and Chung, 2019; Lee et al., 2016; Singh, 2016). The ultimate goal of UPW production is to completely remove all components except water molecules, but this is virtually impossible. Hence, the specific demand for UPW in different industries determines the purity level of UPW. Among them, the purity grade of UPW used in the semiconductor industry is most stringent. Especially with the booming developments in semiconductor technology, the quality requirements of UPW in the semiconductor industry are being formulated (Lee et al., 2016; Zhao et al., 2019). Thus, strictly controlling the quality of the final effluent is one of the most important challenges in the current UPW production process.

In addition, because most of the manufacturing stages of electronic products involve the use of UPW, UPW represents an important and large market. For example, a semiconductor is a miniature electronic circuit which contains a multitude of transistors. Creating an integrated circuit on a 30 cm wafer requires approximately 2,200 gallons of water. A typical electronic product manufacturing process needs about 3 to 60 million liters of UPW per day (Lee et al., 2016). Thus, the manufacture of semiconductors is highly water intensive. Along with the development of semiconductor technology, the

demand of UPW in the semiconductor industry will greatly increase, which will lead to a huge growth of the raw water demand. Therefore, how to meet a large amount of raw water demanded will be a major problem that has to be solved in UPW production. On the other hand, with rapid urbanization and population growth, water shortage is becoming more and more serious worldwide. Therefore, people have made extensive efforts to promote other water resources, such as rainwater utilization, reclaimed water utilization, seawater desalination, etc. (Charfi et al., 2017; Guo et al., 2014). Among them, reclaimed water and municipal sewage are readily available, in great quantities, and not restricted by geographical environment. These accelerate the change of raw water resource used for UPW production. Recent studies' results have shown the feasibility of using reclaimed water as raw water sources to produce UPW (Lefebvre, 2018; Wang et al., 2019), which is a feasible engineering solution to realize the sustainable utilization of water resources. For instance, Singapore's NEWater, one of that country's four national taps, is high-grade reclaimed water produced from treated effluent of sewage treatment plants utilizing membrane technology and ultraviolet disinfection. Total organic carbon (TOC) and the conductivity of NEWater are about 0.5 mg/L and 100 μm/cm, respectively, which can be employed in industries with lower UPW water quality requirements (Lefebvre, 2018; Wang et al., 2019). In general, making full use of reclaimed water to produce UPW desired in semiconductor industry can save a large quantity of tap water. Moreover, it can improve the recycled utilization rate of water resources. However, to meet the strict UPW quality of the semiconductor industry, the largest problem concerning the raw water change is probably associated with the organic compounds removal, which means the existing UPW preparation

process must be improved.

This is the first paper to critically review on challenges of the existing conventional UPW production process using reclaimed water as raw water source. The paper summarizes the technical methods of removing organic matters in the traditional UPW production process and discusses the existing problems inherent in organic matter removal while suggesting the future trend of the UPW production process.

2. Challenges

In UPW production, the existing processes for organic matter removal include ion-exchange (IX) adsorption, granular activated carbon (GAC) adsorption, reverse osmosis (RO), ultraviolet (UV) irradiation (wavelengths at 185 nm) (Choi and Chung, 2019; Choi et al., 2016; Zhao et al., 2019). However, it is difficult to remove some small molecular organic compounds using these methods. Some studies have indicated that via a series of purification processes in UPW production, small molecular organic pollutants, including isopropyl alcohol (IPA), methanol and urea, can still be detected in the final effluent. Their natural properties, for instance low molecular weight, no surface charge, and low content in UPW, result in poor degradation when implementing current UPW production techniques (Choi and Chung, 2019; Choi et al., 2016; de Abreu Domingos and da Fonseca, 2018; Lee et al., 2016). So far, some studies have shown that UV irradiation at lower wavelengths (185 nm) is effective in terms of some low molecular weight organic compounds degradation (Choi et al., 2016; Sun and Chen, 2014). Sun and Chen (2014) prepared a new type of GAC coated with photocatalyst, which can remove 85% IPA from UPW by passing through ultraviolet radiation at 254

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nm (Sun and Chen, 2014). Furthermore, Choi et al. (2016) explored the removal effect of IPA and methanol in UPW production using a 185 nm UV and ion exchange system. The results indicated all IPA (initial concentration of 50 μg/L) in raw water could be completely oxidized to acetone by UV irradiation, and then converted to organic acid and carbon dioxide (CO_2). The organic acids generated by UV oxidation can be then completely removed by the IX resin. As for methanol (initial concentration of 50 μg/L), it was converted to formaldehyde and further to formic acid by UV oxidation, and was then removed by the IX resin. As a result, all IPA and methanol in the UPW were completely removed (Choi et al., 2016).

As mentioned above, some processes have successfully improved the removal rate of methanol and IPA in UPW, however the efficient removal of low concentration of urea in UPW remains a problem to be solved. Previous study has confirmed that the residue of TOC in UPW is mainly caused by the presence of urea in the water (Choi and Chung, 2019; Rydzewski and Godec, 2002). Choi and Chung (2019) through long-term detection of the content of TOC and urea in the actual UPW production, it is found that the trend of TOC in UPW effluent is analogous to that of the urea concentration in raw water (tap water with 1 - 2 mg/L TOC and 150 - 250 μS/cm conductivity), 1 μg/L urea in the raw water may increase the TOC in the final UPW effluent by about 0.1 μg/L (Choi and Chung, 2019). Hence, it could be presumed that TOC in UPW effluent is positively correlated with the urea concentration in raw water. In addition, they found that the urea concentration in the influent was not high as 50% of the urea could not be removed through the prevailing processes. This caused TOC in the final effluent could not meet the standard (Choi and Chung, 2019). Therefore, it can be inferred that TOC in

the eventual effluent of UPW mainly depends on the concentration of urea in the raw water, which also confirms that the urea removal by the existing UPW production facility is limited. Furthermore, it has been well known that the reclaimed water has a very complex chemical and microbial composition with a substantial amount of ammonia, phosphate, microbes, suspended solids. A list of reclaimed water quality in various regions is given in Table 1.

Table 1

As can be seen in Table 1, chemical oxygen demand (CODcr) in reclaimed water is more than 10 mg/L. It contains a large number of suspended microorganisms, dissolved organic compounds, inorganic compounds and pathogens, which is complex and diverse (Ibn Abdul Hamid et al., 2019; Liu et al., 2020; Ma et al., 2016). Conversely, the CODcr in tap water measured in actual UPW production usually does not exceed 1.0 mg/L. It is obvious in Table 1 that the TOC in reclaimed water is more than 5 mg/L, while in tap water it is usually less than 2.5 mg/L (Zhang et al., 2021). Therefore, it is certain that the quality of reclaimed water is worse than tap water. Among the pollutants in raw water for UPW production, suspended microorganisms can be removed by ultrafiltration (UF), pathogens can be deactivated by UV irradiation, dissolved macromolecular organic and inorganic compounds can be removed by IX adsorption, RO or UV irradiation, and even small molecular organic compounds such as isopropanol, methanol can be removed by UV_{185} (Choi et al., 2016; Lee et al., 2016; Malaeb and Ayoub, 2011; Matilainen and Sillanpää, 2010). However, the conventional UPW production process does not have a significant effect on the urea removal. Furthermore, the urea concentration in tap water is about 5~20 μg/L, while the urea

detected in reclaimed water as raw water for UPW production ranges from 20 μg/L to 50 μg/L (Choi and Chung, 2019). The urea is more than double that in the reclaimed water used for UPW production compared to tap water. Consequently, if reclaimed water is used as raw water for UPW production, the urea in the raw water cannot be effectively removed. More urea may lead to higher concentration of TOC in the UPW effluent, which means it is more difficult to meet the water standards required by the semiconductor industry. Consequently, to ensure UPW standard, the production process is required to effectively remove urea and other small molecule organic compounds in reclaimed water. Furthermore, the UPW production technologies need to be carefully improved to guarantee UPW quality, such as adding pretreatment or supplementary units on the basis of the current UPW production process.

To obtain a large amount of production raw water and realize the sustainable utilization of water resources, there is a pressing need to use reclaimed water as raw water to produce UPW in the future. For this reason, the effective removal of small molecule organic compounds from the reclaimed water is crucial for UPW production.

3. Traditional technologies for treating organic pollutants in the UPW system

3.1 Conventional ultrapure water production process

The UPW production system is an extremely complex process and involves many purification technologies. The conventional UPW production system mainly includes three phases: 1) pretreatment stage; 2) desalination stage; and 3) polishing stage. Due to the cumulative of the whole production system, each process will be affected by the output of the previous stage (Lee et al., 2016). Consequently, each step in the UPW production process is critical to ensure the effluent quality. Especially for the

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semiconductor industry, due to the sensitivity of precision instruments, UPW needs to meet extremely high standards before it can be put into use. It is shown from Table 2 that some of the standards for microelectronics UPW in the American Society of Testing and Materials (ASTM) and the China National Standards (CNS) (ASTM, 2018; CNS, 2013). Obviously, the microelectronic grade UPW in ASTM D5127-13(2018) is divided into seven categories, while the CNS (GB/T 11446-2013) is divided into four categories. However, no matter which standard is used, the more sophisticated devices are produced in the semiconductor industry, the higher quality UPW is needed. With the continuous development of semiconductor technology, the requirements of the electronic industry for the water quality of UPW will become more and more stringent.

Table 2

The resistivity/conductivity and TOC are two important parameters that define UPW quality, representing the concentration of ions and organic matter in UPW, respectively. Throughout the production process of UPW, almost each unit process involves the removal of ions and organic pollutants in water. As well, the requirements for the conductivity and TOC of UPW are more stringent in the semiconductor industry, and therefore, it is more important to reduce the concentration of ions and organic compounds. Compared to the production process of deionized water, RO pure water and distilled water, the UPW production line is usually the most complex and longest, in order to meet the water quality standards of UPW in the semiconductor industry. The typical flow of a conventional microelectronic UPW production system is shown in Fig. 1.

Figure 1.

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As can be seen from Fig. 1, the typical microelectronic UPW production systems consist of a variety of processes, such as IX, RO, UF, degassing, UV irradiation, and so on. Most of the pollutants in raw water, such as ions, macromolecular organic matter, suspended solids, and gases, can be effectively eliminated by a series of purification processes (shown in Fig. 1), respectively. The obtained UPW can meet the standards of the semiconductor industry. In general, the prime methods of removing suspended solids are precipitation and filtration, while the main methods to remove ions and organic pollutants are IX, RO, and UV irradiation. Almost each technology involves the removal of organic matter and ions. Hence, reducing the concentration of organic compounds and ions from water plays a decisive role in UPW production.

To sum up, the UPW quality standard in the semiconductor industry is very strict, and the typical microelectronic UPW production system is extremely complex and lengthy. The UPW quality is closely related to the influent quality and effect of each unit process. If the raw water quality used for UPW production changes, it will greatly affect the effluent quality.

3.2 Conventional technologies for organic pollutants removal in the UPW process

Conventionally, the technologies used to remove organic compounds in the UPW production mainly consist of adsorption, RO, UV irradiation, etc. Of these, the two-pass RO process can effectively reduce the concentration of organic compounds in UPW (Lee et al., 2016; Lee and Kim, 2015). Combining RO and IX or GAC is proven to have a good effect on the removal of organic pollutions and has been successfully applied in actual production activities (Lipnizki et al., 2012; Quinlivan et al., 2005; Summers et al.,

2013). In the conventional UPW production process, UV after RO is also often used to reduce the concentration of TOC. It is pointed out that different UV wavelengths have different effects on the degradation of organic compounds. The UV_{254} can directly attack organic compounds, while UV_{185} radiation can induce the dissociation of water molecules to hydroxyl radicals (HO•). The resulting HO• can oxidize a variety of organic compounds, and then achieve the reduction of the concentration of TOC in aqueous solution (Jin et al., 2018). However, it is difficult to completely remove the small molecule organic compounds in water by a single technology, so the process flow of UPW production usually adopts a combination of GAC, RO, IX, as well as UV. First of all, GAC and UV serve to remove suspended solids and some macromolecular organic pollutants in the pretreatment stage, then the UPW is desalted by IX and RO, and finally, UV and ultrafiltration are applied to improve water quality in the polishing stage. This will achieve the high removal efficiency of ions and organic compounds concentration. Throughout a series of processes above mentioned, UPW can achieve a high removal rate of ions and organic compounds to meet the needs of the semiconductor industry production line.

3.2.1 Ion-exchange (IX) adsorption

IX technology is essential for UPW production, in which IX resin is often used in the desalination stage of UPW production. In the 1950s, IX technology made the large-scale production of high purity water (HPW) a reality (Singh, 2016). The production process of UPW in the 1970s included the use of cation and anion exchangers (Lee et al., 2016). To make the final effluent quality of UPW meet the required standard, almost every different kind of UPW production system involves the

ion exchange adsorption using IX resin. The main mechanism of impurity removal by IX resin is that cationic resin has attached H^+ ions, which can exchange cations in water, while anion resin has OH^- ions that can exchange anions. As a result, the concentration of ions and organic compounds in the water is effectively removed, and at the same time, the H^+ and OH^- ions released from the resin combine to form water molecules (Bennett, 2009). However, both the experimental and actual production results reveal that the main function of IX resin is to remove charged pollutants but not neutral organic matter in water (Bolto et al., 2004; Levchuk et al., 2018; MacKeown et al., 2021). MacKeown et al. (2021) tested 4 different IX resins in terms of natural organic matter removal. They found that the humic substances removal efficiency by IX resin was 72%, but the removal efficiency of low molecular weight neutrals was only 36%. Because the charged organic compounds in the UPW are very low, the removal efficiency of neutral organic compounds by IX resin is extremely limited, while urea is a kind of uncharged small molecular organic matter which is more difficult to be removed by IX resin. Thus, IX is expected to couple with other technologies or modify IX resin to effectively reduce the organic contaminants in UPW production.

It is proposed that the IX resin can be used as a supplement to the other technology systems (Choi et al., 2016). The concentration of organic compounds can be effectively reduced by the UV_{185} firstly, and then the organic acids (such as compounds containing carboxyl groups), which are oxidative by-products of organic compounds in the UV irradiation system, can be removed by the following IX system. When the initial TOC was 20 μg/L, the TOC removal efficiency can reach more than 80% after the UV oxidation and ion exchange reactions. However, when the initial TOC increased from

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20 μg/L to 100 μg/L, the TOC removal efficiency decreased to 40% (Choi et al., 2016).

Lee et al. (2020) prepared a new type of IX resin by silanized magnetic nanoparticles

(mNPs) and then embedded them in IX resin, which greatly improved the ability of IX

resin to remove TOC. The TOC level was greatly reduced from 228 μg/L to 7 μg/L by

this way. This is mainly due to the fact that silanized mNPs have a very large specific

surface area, which enables more organic compounds to be adsorbed (Lee et al., 2020).

Therefore, the effective combination of IX technology with other processes, the

modification of existing IX resin materials and the exploration of new adsorbents with

selectivity for organic compounds will be the research focus of using IX technology to

remove organic pollutants in the future.

On the other hand, some studies have shown that the removal efficiency of organic

matter varies from the IX resin with different structures. Additionally, the metal

components (Na, Ca, Fe, Zn, etc.) in IX resin may weaken the removal of organic matter,

and the moisture-holding capacity of IX resin also has a significant effect on the

removal of organic matter (Apell and Boyer, 2010; Bolto et al., 2002; Zhao et al., 2019).

Thus, the selection of appropriate IX resin is of great significance for the removal of

organic matter. Nonetheless the regeneration of resin requires the consumption of a

large number of chemicals which results in a large amount of acid and alkali wastes. In

general, this method of removing organic compounds will greatly increase the

production costs.

3.2.2 Granular activated carbon (GAC) adsorption

GAC is often used in the pretreatment stage and desalination stage in UPW

production to remove suspended solids, macromolecular organic compounds, and ions

in water, owing to its large adsorption capacity. For instance, the adsorption capacities of GAC for phenol, methylene blue and Pb(II) could reach 192 mg/g, 131.8 mg/g and 21.88 mg/g, respectively (Aygün et al., 2003; Goel et al., 2005; Liu et al., 2013). However, it has been proved that GAC can only remove some large non-polar molecules, while some small polar molecules with high solubility (such as isopropyl alcohol, urea, etc.) cannot be effectively adsorbed on GAC (Safwat and Matta, 2018; Schmotzer et al., 2002). Safwat and Matta (2018) explored the removal efficiency of urea (1000 mg/L) in synthetic wastewater by two different adsorbents (granular activated alumina and GAC). They found that the maximum removal efficiency was 31% for GAC at pH =9.0. Clearly, the removal efficiency of urea by GAC is not ideal. Moreover, the adsorption capacity of GAC relies greatly on the molecular polarity, size, charge, as well as solubility of organic compounds. Thus, it is not ideal that GAC is used to remove small molecular organic pollutants in UPW, far from reaching the requirements of the microelectronics industry. To effectively remove the small molecular organic compounds in UPW production, it is necessary to use other technologies to assist GAC adsorption or develop new adsorbents.

A large number of studies and practical production have shown that IX resin or GAC alone cannot effectively remove the organic compounds in water. Compared with the use of GAC or IX resin alone, the combination of GAC-IX can significantly increase the adsorption removal of dissolved organic compounds in water (Humbert et al., 2008). On the other hand, Ooi et al. (2017) improved the urea removal efficiency by utilizing activated carbon fiber (ACF). They prepared ACF from oil palm empty fruit bunch fiber via physicochemical activation using sulfuric acid as an activating reagent.

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They found that these ACF samples have well developed micropore structures and high surface area, and the adsorption capacity of urea (2310 mg/L) could reach 878 mg/g. However, the urea concentration in synthetic wastewater was much higher than that in reclaimed water, and its removal efficiency of low concentration urea has not been verified yet. It is also pointed out that zeolites and mesoporous silica with a high specific surface area have superior adsorption ability for urea in dialysate over GAC, which are two feasible urea adsorbents (van Gelder et al., 2020). Studies have shown that the adsorption capacities of zeolites and mesoporous silica for urea in dialysate (about 38 mM) could reach around 165 mg/g and 500 mg/g, respectively (Cheah et al., 2016, Wernert et al., 2005). Moreover, various surface functional groups can be introduced into mesoporous silica to enhance the interaction between adsorbents and adsorbates, to improve the adsorption capacity. For instance, Cheah et al. (2016) found that the urea adsorption capacity of amine-functionalized mesoporous silica was further improved, and its adsorption capacity for urea in dialysate (about 38 mM) could reach 550 mg/g (Cheah et al., 2016). Therefore, the selective adsorbents with enhanced adsorption capacity will be sequentially developed for organic compounds adsorption removal in the following exploration.

3.2.3 Reverse osmosis (RO)

In the 1980s, reverse osmosis (RO) appeared in UPW production, which replaced the traditional two-bed IX system to reduce total dissolved solids (TDS), particulates, and TOC. Many studies have confirmed the high pollutants removal performance of RO (Lee and Kim, 2015; Malaeb and Ayoub, 2011). For instance, the IPA removal efficiency could exceed 93 % (initial concentration of 683 μg/L). This result was mainly

due to the steric hindrance of solutes by pore structure of membrane (Lee and Kim, 2015). In the past decade, with the continuous development of membrane technology, RO has gradually occupied the core of UPW production technology (Lee et al., 2016). Utilizing RO is considered effective in removing organic compounds from water in the UPW production process. In a series of processes, RO is usually used in the desalination stage. Most of the impurities can be removed by RO to obtain water with a maximum resistivity of 2 MΩ•cm (Lee et al., 2016).

It was found that RO has better repulsion of high molecular weight compounds than low molecular weight compounds, which can reject more than 80% of organic matter with higher molecular weight such as 2-(2-butoxyethoxy) ethanol (162.23 g/mol). However, the rejection rate of some smaller molecular organic compounds such as urea (60.06 g/mol) and methanol (32.04 g/mol) is less than 22% (Yoon and Lueptow, 2005). Zhao et al. (2019) used the two-pass RO process to optimize the UPW production system. Their results further confirmed that the two-pass RO process can effectively reduce the concentration of TOC, and the removal efficiency of target organic compounds can reach 60-95% (Zhao et al., 2019). However, some studies demonstrated RO cannot efficiently remove small molecular organic compounds such as urea, and urea was rejected ranging from 20% to 50%. This is because urea is a small molecule without charge, and it can be removed neither by size repulsion nor by charge repulsion (Singh, 2016; Yoon and Lueptow, 2005).

On the other hand, the performance of RO is closely related to the influent quality. When reclaimed water is used as raw water to produce UPW, the change of the raw water quality affects the treatment effect of RO. It will be more difficult to rely on RO

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alone to make TOC concentration in the final effluent of UPW reach the required standard. When the influent quality changes, it is necessary to adjust the configuration of the RO process to improve the UPW production system's performance, including adding another process for small molecule removal before RO, improving the performance of RO membrane and so on. Recently, a study has proposed an innovative process combining the moving bed ceramic membrane bioreactor (MBCMBR) and RO, which can effectively regenerate municipal wastewater into UPW products, providing an innovative method for the production of UPW (Wang et al., 2019). MBCMBR can remove more than 95% of COD under different dissolved oxygen (DO) conditions, while RO can remove more than 95% of TOC and inorganic salts. However, the ceramic microfiltration membrane can only retain about 20% of small molecular organic compounds, and the removal performance of the small molecular organic compounds by RO is also very limited with the highest removal rate of 36%. The obtained UPW employing this combined process only reaches the Type E-4 in the ASTM, and further polishing is needed to meet the Type E-1. On the other hand, at low DO concentration, more low molecular weight organic compounds and biopolymers are produced, which leads to higher scaling tendency of MBCMBR and RO.

3.2.4 Ultraviolet (UV) irradiation

Since the TOC concentration required in UPW is extremely strict, UV irradiation plays an irreplaceable role in the production of UPW (Bennett, 2009). UV radiation can directly photolysis the organic matter and induce the dissociation of water molecules into HO• (see Eq (1)-(3)) (Vallejo et al., 2015). Thus the production of HO• can greatly improve the ability to degrade organic compounds.

$$\text{Target Pollutants} + h_v \rightarrow \text{Intermediate Products} + H_2O + CO_2 \qquad (1)$$

$$H_2O + h_v \rightarrow HO\bullet + H\bullet \qquad (2)$$

$$\text{Target Pollutants} + HO\bullet \rightarrow \text{Intermediate Products} + H_2O + CO_2 \qquad (3)$$

Generally, the HO• can react strongly with most organic substances by hydrogen abstraction, electrophilic addition to double bonds, or electron transfer, which can degrade organic compounds and eventually transform into water and a series of intermediates. At the same time, the reaction rate between HO• and organic compounds is very rapid, which is much faster than that of organic compounds with chlorine (Cl_2) and ozone (O_3) (Zhao et al., 2019). Therefore, to improve the removal rate of organic pollutants in UPW, UV is needed in both the pretreatment and polishing stages.

It is found that the efficiency in removing organic compounds by UV is closely related to UV wavelength. When the aqueous solution passes through ultraviolet radiation at 254 nm, although the concentration of TOC in the aqueous solution can be reduced, the removal efficiency is only 4.8%. While the aqueous solution goes through ultraviolet radiation at 185 nm, the removal rate can be as high as 68% (Zhao et al., 2019). The results show that the removal rate of TOC under a low and medium voltage lamp is larger. Kutschera et al. (2009) further explained why UV_{185} has high removal efficiency, as ultraviolet radiation at 185 nm can not only directly oxidize organic matter, but produce HO• through the synergistic action with water molecules, thus accelerating the oxidation of organic compounds (Kutschera et al., 2009). In fact, the use of multiwavelength $UV_{185/254}$ can lead to a rapid fall in TOC concentration. Some experiments show that multiwavelength $UV_{185/254}$ can sharply reduce the TOC concentration in the final effluent of UPW, and the removal rate is as high as 86%

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(Kutschera et al., 2009; Zhao et al., 2019; Zoschke et al., 2014). Also, the removal efficiency of organic matter by UV is positively correlated with temperature. High power multiwavelength can heat the water, which increases the quantum yield of HO• (Xie et al., 2018). Thus, this results in a great enhancement in the removal rate of organic compounds (Zhao et al., 2019). Thus, $UV_{185/254}$ wields the best effect on the removal of TOC in UPW.

Although the concentration of TOC in water can be greatly reduced by using UV in the pretreatment stage and polishing stage, UV radiation cannot destroy the N-bonds in urea, due to the weak absorption capacity of urea to UV radiation. Even if the best operating conditions are adopted, it is difficult to remove urea in water. As shown in previous studies, the maximum removal efficiency of urea by using UV alone is less than 10% (Choi and Chung, 2019; Singh, 2016; Zare Afifi and Blatchley, 2016). Therefore, if the source of raw water is changed with the influent quality of UPW declining, it is difficult to ensure the effluent reaches the standard of microelectronics even through UV polishing.

4. Feasibility and trends of advanced oxidation processes (AOPs) in the UPW production process

To sum up, in the past the UPW production process, the combination of IX, RO, UV, and other processes have helped to improve the removal rate of organic pollutants. Nonetheless, when changing the raw water source, it is difficult to use the above processes to remove urea and make the effluent TOC meet the requirements of the semiconductor industry. How to perfect or improve the existing process to meet the strict water standards continues to be an issue of some concern. Generally, many

technologies have been used for urea removal including nanoparticles, enzymatic hydrolysis, combined technologies etc. (El-Lateef et al., 2019; Pillai et al., 2014; van Gelder et al., 2020). While some processes suffer from high cost (e.g., enzymatic hydrolysis), others suffer from complicated (e.g., combined technologies). In this section, a new route is proposed for UPW production adding the advanced oxidation processes (AOPs) (except UV irradiation) on the basis of the original process to remove the small molecular organic compounds completely.

The proposal of AOPs can be traced back to the 1980s when it was defined as all oxidation processes involving the production of large amounts of HO• to purify water (Deng and Zhao, 2015; Fernandes et al., 2019), including photocatalytic oxidation, electrocatalytic oxidation, Fenton oxidation, etc. Subsequently, the concept of AOPs was gradually extended to the oxidation processes of producing sulfate radical ($SO_4^-•$). In recent years, AOPs have attracted increasing attention in wastewater treatment and drinking water disinfection (Bilinska et al., 2016; Wacławek et al., 2017). A large number of experimental studies and practical applications of wastewater treatment have confirmed that AOPs have an excellent effect on the removal of organic pollutants (Yang et al., 2019). Thus, AOPs are promising approaches to remove small organic compounds such as urea for UPW production. It is expected to apply AOPs to fully remove small molecular organic pollutants in UPW and transform them into basically non-toxic products, which will provide a new solution for the future development of UPW production. However, there are only a few cases of introducing AOPs (except UV irradiation) into UPW production at present, and the overall research and application are still in their initial stages (Humbert et al., 2008).

In this section, some of the recent research is presented on small molecule organic compounds removal from water using AOPs, namely photocatalytic oxidation, sulfate radical-based advanced oxidation processes (SR-AOPs), electrocatalytic oxidation (EO) and some other AOPs. This section focuses on understanding the removal mechanisms of these technologies and their feasibility in the UPW production process. Additionally, the benefits and limitations of the introduction of AOPs as a supplementary unit are addressed and potential new research trends are speculated.

4.1 Photocatalytic oxidation

With regard to photocatalytic oxidation (the process of photoactivation of photocatalyst), the light sources and photocatalysts play a crucial role in the occurrence of photocatalysis. Because semiconductor photocatalysis only needs to be excited by visible light or ultraviolet light, it is widely used in water treatment. By far, as the most widely studied and commonly used photocatalyst, TiO_2 has the characteristics of low cost, safety, stability, and high catalytic activity. Hence, TiO_2 is often used as a photocatalyst in many waste water treatments and drinking water disinfection (Matoh et al., 2019). The reaction principle of semiconductor photocatalysis for the degradation of urea is shown in Fig. 2.

Figure 2.

It can be seen that after the semiconductor catalyst is exposed to light, it will cause the photon to absorb enough energy and excite the electron (e^-) from the valence band to the conduction band, thereby generating a positive electron hole in the valence band (h^+). Since h^+ is a strong oxidant and e^- is a good reductant, the electron-hole charge carriers can form many strongly active substances (such as $HO \bullet$, $O_2^- \bullet$) by redox. This

functions by degrading organic pollutants (Banerjee et al., 2014; Matoh et al., 2019; Schneider et al., 2014). Therefore, urea in water can be degraded into NH_4^+, NO_3^- and CO_2 by free radicals (Kim et al., 2019). Because HO• has high reactivity and non-selectivity, they can degrade most of the organic compounds in water, and in some cases, the organic compounds can be fully converted into CO_2, H_2O, and inorganic salts (Fernandes et al., 2019). It is obvious that no new contaminants are produced in this process. At present, a large number of studies have shown that a variety of organic pollutants in water can be effectively removed by photocatalytic oxidation technology, such as phenolic compounds, azo dyes, and so on (Zangeneh et al., 2015). Recently, a photocatalyst was developed by being modified with both palladium and fluoride, which can efficiently degrade and mineralize urea in urine (Kim et al., 2019). Pd supported on the surface of TiO_2 can promote the transfer of h^+ from the inside of TiO_2 to its surface. In addition, fluoride ions promote the movement of h^+ to water molecules and react with them. The synergistic effect of the two steps greatly enhanced the formation of HO•, and urea in urine can be degraded into NH_4^+ and NO_3^- by HO•, which greatly improves the degradation rate. 67% of urea (initial concentration of 12 mg/L) was degraded after 3 hours of UV irradiation by using this photocatalyst. Thus, the prepared F-TiO_2/Pd showed great potential for urea removal with both high degradation rate and removal efficiency. Therefore, it can be seen that it shows great potential for the application of photocatalytic oxidation technology to the removal of urea and other organic substances in UPW production. However, to generate a large number of stable HO•, the catalysts still need to be further explored. In addition, the photocatalytic oxidation technology has not been put into practical application in UPW

production. Therefore, it is necessary to focus on the concrete implementation of practical application in UPW production.

Furthermore, most of the photocatalytic oxidation technologies using unmodified semiconductor catalysts (such as TiO_2, carbon nitride) are activated by UV irradiation rather than visible light (Stylidi et al., 2004; Zhou et al., 2019). This is because most semiconductor catalysts have a weak response to visible light, so it is difficult for visible light to activate their electrons. If it can be activated by visible light, it can be put into the actual production on a large scale and the cost will be greatly reduced. Therefore, the photocatalytic reaction under visible light will be a major challenge for the development of photocatalytic technology in the future. In most existing studies, there are two main ways to enhance the catalytic ability of visible light: one is to modify the existing photocatalyst, and the other is to develop new photocatalyst materials (Kalantari et al., 2016; Moreira et al., 2019; Samadi et al., 2016). In the production of UPW, if the small molecular organic compounds in water can be removed by photocatalytic oxidation under visible light, it will greatly reduce the production costs while improving the effluent quality. This is of great significance to the low-cost and high-quality production of UPW.

4.2 Sulfate radical-based advanced oxidation processes (SR-AOPs)

The redox potential of $SO_4^-\bullet$ is about 2.5-3.1V, which is higher than that of $HO\bullet$ (1.9V-2.7V), and its life span is longer than $HO\bullet$ (Xie et al., 2019). Therefore, the new type of AOPs based on $SO_4^-\bullet$ has attracted wide attention in recent years. The common methods to obtain $SO_4^-\bullet$ are to activate persulfate or sulfite by adding transition metal, UV irradiation, and heating (He et al., 2014; Lebik-Elhadi et al., 2020; Matzek and
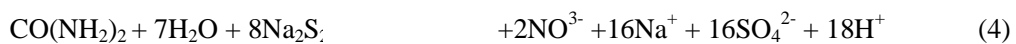
Carter, 2016). Although persulfate anions will react with some organic chemicals without activation, the removal rate is much lower than the $SO_4^-\bullet$ produced after activation (Lee et al., 2009). Therefore, using $SO_4^-\bullet$ to oxidize organic pollutants is highly effective and feasible in practical application, and the activation is a key step to produce a large amount of $SO_4^-\bullet$.

Activation methods such as heating, UV irradiation, ultra-sonication mainly occur through energy transfer to obtain $SO_4^-\bullet$, while the addition of transition metal catalysts is mainly done through a redox reaction to obtain $SO_4^-\bullet$ (Matzek and Carter, 2016). Different activation methods have a different mechanism for removing organic pollutants, along with the varied removal efficiency and reaction rate. Therefore, the activation method is critical for removing organic pollutants. Some other studies have shown that the $SO_4^-\bullet$ produced after activation can effectively oxidize many kinds of organic compounds in sewage, such as trichloroethylene, amitriptyline, imipramine, etc. (Devi et al., 2016; Xie et al., 2019). Therefore, the application of this technology to the removal of organic matter in UPW will become a major research field with future advances made in UPW production technology.

In order to improve the performance in removing small molecular compounds such as urea, a facile SR-AOPs is introduced into the UPW production method. In this technology, small molecule organic pollutants such as urea, are difficult to be oxidized by UV irradiation, but can be chemically converted to nontoxic products by adding ammonium persulfate (Singh, 2016). The key to this technology is the introduction of ammonium persulfate (APS), which is then activated by UV radiation, and a large amount of $SO_4^-\bullet$ produced can oxidize urea. The ammonia produced by urea

decomposition can be removed by the mixed-bed ion exchange (MBIX), and the DO released after the water enters the MBIX unit is removed by the membrane contactors. Moreover, Choi and Chung (2019) monitored urea concentration of the operation of a UPW production facility in South Korea. Its unit processes include GAC, degasifier (CO_2 removal), UV Sterilizer, micro filtration, 1st RO, 2nd RO, primary degasifier (O_2 removal), primary TOC reduction UV, electro deionization, mixed bed ion exchanger, secondary TOC reduction UV, H_2O_2 removal ion exchanger, secondary mixed bed ion exchanger, secondary degasifier (O_2 removal), polishing mixed bed ion exchanger and ultrafiltration. In the UPW production system, tap water as raw water was supplied to the UPW production. Through more than a year of monitoring, they found approximately 50% of the urea in tap water was removed by the UPW production process and urea concentration in raw water had a significant effect on the UPW quality. Hence, they directly added persulfate to the TOC reduction UV unit in the UPW production process and explored the removal effect of urea removal by persulfate with UV irradiation. Results show that the persulfate can be activated by UV, and the generated $SO_4^-\bullet$ is sufficient to oxidize the urea in UPW. The reaction mechanism is as follows (see Eq. (4) and (5)) (Choi and Chung, 2019):

$$CO(NH_2)_2 + 7H_2O + 8Na_2S_2 \qquad\qquad +2NO^{3-} +16Na^+ + 16SO_4^{2-} + 18H^+ \qquad (4)$$

$$S_2O_8^{2-} \quad \rightarrow \quad SO_4^-\bullet + SO_4^-\bullet \qquad (5)$$

In this system, the persulfate is added to the UV irradiation process and can be activated by UV to form $SO_4^-\bullet$, which will oxidize urea to nitrate and CO_2 under the action of $SO4^-\bullet$. The removal rate of urea (initial concentration of 1.65 μmol/L) reached up to 90% when 20 μmol/L persulfate was added in the UV reactor (Choi and Chung,

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2019). In the meantime, the sulfate radicals will be converted to sulfate ions after the oxidation (Pi et al., 2019). Therefore, it is necessary to add another step to capture the sulfate ions. In general, RO can reject more than 99% of ions in the water, which is usually used in the typical microelectronic UPW production systems (Lee et al., 2016). Consequently, through the desalination stage in the subsequent UPW production process, the sulfate ions generated by the SR-AOPs can be effectively removed. Thus, this method greatly reduces the complexity of the device, and is easy to operate and has good practicability. It is more suitable for use in the actual production process, but this method is still at the laboratory research stage. Whether the ideal treatment effect can be achieved in the complex actual environment remains to be verified.

The use of SR-AOPs mentioned above in the production of UPW is all activated by UV. Based on the original process (UV irradiation), a large amount of organic matter can be removed, and the small molecular organic compounds in water can be further oxidized by $SO_4^-\bullet$. Whether other activation methods can be used for targeted removal of urea and other small molecular organic compounds to achieve a better removal outcome and improve the removal efficiency has not been demonstrated. It has been found that using zero-valent iron to activate sulfite can effectively remove organic pollutants in water (Xie et al., 2017). After adding simulated solar radiation, the photo/zero-valent iron/sulfite system greatly improves the removal efficiency of organic matter (Xie et al., 2019), but the system heavily favors the removal of organic compounds. Whether it can be used in UPW to remove urea and other small molecular organic compounds is yet to be confirmed. Therefore, the activation method with the best removal rate and removal efficiency of urea and other small molecular organic

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compounds in water will be the focus of UPW research in the future.

4.3 Electrocatalytic oxidation (EO)

Electrocatalytic oxidation (EO) is also a form of AOPs, and its working mechanism is to remove organic matter through HO• produced in the EO process, which has been widely used in the treatment of sewage wastewater. Reasons for the application of EO may include its high efficiency, cleanliness, and easy automation. This process is gradually becoming a kind of water purification technology with development potential (Sirés et al., 2014; Zhang et al., 2020). Therefore, it has a certain development prospect to apply EO to the production of UPW to remove the small molecular organic pollutants. However, no systematic research has yet confirmed the feasibility of applying it to UPW, and this is also a major problem that must be explored in the future.

There are two forms of degradation of organic pollutants by EO. One is known as anodic direct oxidation where organic pollutants are removed by direct transfer of electrons, or HO• reacts with the anode to form a higher valence metal oxide (MO), then oxidizes the pollutants through MO. The other is indirect oxidation of pollutants by the free HO• around the electrode (Tan et al., 2020). Therefore, the treatment performance of EO is closely related to free HO•, and the number of free HO• largely depends on the activity of anode materials. Thus, the selection of anode materials has a significant impact on the electrocatalytic degradation of organic compounds. Fig. 3 illustrates the degradation pathway of urea when using different anode materials. For negative anodes, most of HO• is free, which can react with urea to form NH_4^+, NO_3^- and CO_2, while for active anodes, HO• reacts with the electrode first. Then the reaction products react with organic pollutants to oxidize target pollutant (Tan et al., 2020; Wu et al., 2014). In

addition, urea can be directly oxidized directly by active anode. Furthermore, urea can be oxidized to the production of nontoxic products in an alkaline medium, such as H_2O, CO_2 and nitrogen (N_2) (Urbańczyk et al., 2020).

Figure 3.

According to the analysis by Comninellis et al. (2008), almost no HO• accumulates on the surface of the active anode, while a large amount of HO• will appear on the surface of the negative anode, especially the boron-doped diamond (BDD) electrode (Comninellis et al., 2008). Tan et al. (2020) further confirmed that the produced HO• when using the BDD electrode can be regarded as "quasi-free" by comparing many kinds of anode electrodes, which has high reactivity and activity, and performs the best when removing organic matter (Comninellis et al., 2008; Martínez-Huitle et al., 2015; Tan et al., 2020). It is also confirmed that when BDD is used as the anode, the free HO• produced can mineralize urea (initial concentration of 2 g/L) completely (Cataldo Hernández et al., 2014), however, the cost of the BDD electrode is more expensive. Whether it can be widely used in UPW production remains to be discussed.

Recently, a Co decorated graphene composite electrode has been proposed, which has high electrocatalytic activity, and can effectively oxidize urea and methanol in an alkaline solution. The anodic reaction is as follows (see Eq. (6)) (Urbańczyk et al., 2020):

$$CO(NH_2)_2 + 6OH^- \rightarrow N_2 + 5H_2O + CO_2 + 6e^- \qquad (6)$$
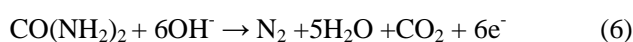
This composite electrode has high activity in urea solution (initial concentration of 0.33 mol/L) and provides a feasible scheme for the removal of urea in UPW. However, this method decomposes urea through direct oxidation, which cannot

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completely oxidize urea in water and requires high anodic activity. It is limited about

the HO• forming during the electrochemical oxidation process, while H_2O_2, O_3 and even

chlorine could be formed. Thus, if urea is removed by direct oxidation, future studies

need to focus on improving the activity of the anode.

With the currently available information, there is no research on the use of EO in

UPW production. Therefore, the feasibility of this technology, coupled with more

suitable electrode materials and a specific implementation scheme are worthy of further

research. Furthermore, the main problem of EO (except high energy cost) is the removal

of electrolytes (Shu et al., 2016; Urbańczyk et al., 2020). It is a crucial problem for the

practical application since they need an additional step for the removal of electrolytes.

Additionally, using EO technology may lead to rising production costs, with the results

that it is important to explore ensuring the economic feasibility of the new process at the

same time.

4.4 Other AOPs

As mentioned above, the AOPs can remove small molecular organic compounds

such as urea from water by generating a large number of highly oxidative free radicals.

Therefore, it is theoretically feasible to remove urea by generated free radicals (such as

HO• and SO_4^-•), which means that other AOPs or the combinations of AOPs may be

also used to remove urea, such as ozonation, UV/O_3, UV/H_2O_2 etc.. Some studies have

indicated that UV/H_2O_2 can transform 75% of urea (initial concentration of 4.5 mg/L) in

distilled water within 24h, The possible reason is that UV irradiation and H_2O_2 can work

synergistically to degrade urea by generating HO• (Bronk et al., 2000; Rosa et al., 2020).

However, through comparing several UV-based processes (Table 3) to remove urea (initial concentration of 5 mg/L) from swimming pools, it is found that the urea content was still high after UV/H_2O_2 treatment; the urea removal rate can only reach 12.8% in 3 hours (Long et al., 2019). Long et al. (2019) deduced that it probably was related to the power input, H_2O_2 dose, irradiation time etc. A higher power input, dosage of H_2O_2 and a longer irradiation time may be more beneficial to remove urea, but the cost will also be higher (Long et al., 2019; Rosa et al., 2020). Therefore, its use in the production of UPW and the optimal conditions in the actual operation remain to be further studied and verified.

At present, it has been confirmed that UV/H_2O_2 can remove urea from swimming pools (about 5 mg/L) effectively, but no information is currently available for removing low concentration urea from reclaimed water by UV/H_2O_2. If reclaimed water is used as raw water for UPW production, with the lower urea concentration (20 ~50 μg/L), it is not clear whether it can reach the same effluent quality. In addition, the effect and influencing factors of removing urea by UV/H_2O_2 have not been determined, and whether UV/H_2O_2 can be used in the production of UPW appropriately is yet to be confirmed. Furthermore, there is no research currently available for removing urea by ozonation, UV/O_3 or other AOPs. As such, in-depth study of all kinds of AOPs for urea removal, such as removal performance, operating cost, operation difficulty, side effects and so on, is of great significance to the application to the actual UPW production.

4.5 Remark and recommendation

When the raw water changes, the great challenge in UPW production is to remove urea effectively. Some AOPs such as photocatalyst, activation of persulfate and electrocatalysis have been explored, but these methods have some specific disadvantages. Therefore, we discussed the effective ways in AOPs for urea removal. A summary of the urea removal efficiencies by AOPs in various water types is given in Table 3. As shown in Table 3, urea can be effectively oxidized by SR-AOPs in UPW with a removal rate of up to 90%. Furthermore, when BDD was used as anode, urea in artificial simulated wastewater can be completely mineralized through EO. However, AOPs such as UV alone and UV/H_2O_2 are not very effective in removing urea from swimming pools, especially when using UV alone, the removal rate is only 0.7%. Table 3 also showed that the removal rate of urea in urine by photocatalytic oxidation is 67%, which may be due to the high urea content in urine. Overall, AOPs can be introduced into the UPW production in the future for effective removal of low concentration urea.

Table 3

Both photocatalytic oxidation and EO (negative anodes used) remove urea through the production of free radicals (mainly HO•). However, some major disadvantages associated with urea removal need to be further explored: (1) need for a relatively great amount of free radicals, (2) requirement for modification and development of photocatalysts and electrode materials, (3) high energy cost for EO, and (4) additional steps required removing the electrolytes through using electrocatalytic oxidation. On the other hand, when using active anodes in electrocatalytic oxidation, HO• reacts with the

electrode first, and then the reaction products react with urea. However, it cannot oxidize urea completely and requires the anodes with high activity. Further studies are necessary to overcome the existing shortcomings of these methods in the actual UPW production. Overall, laboratory studies have confirmed that urea removal by photocatalytic oxidation and EO is effective and offers prospects for UPW system. All these methods are still at the laboratory research stage, the removal effect of these methods has not been verified in the complex actual UPW production.

SR-AOPs seems promising for application in UPW production as the redox potential of $SO_4^-\bullet$ is is higher than that of $HO\bullet$ and its life span is longer than $HO\bullet$. Furthermore, some studies have proved the effectiveness of SR-AOPs. Nevertheless, the SR-AOPs used in UPW production is all activated by UV presently, and other more cost-effective activation methods has not been studied. In addition, these UPW systems that successfully adopted SR-AOPs used tap water as raw water rather than reclaimed water. If reclaimed water is used as the raw water, it is not clear whether it can reach the same effluent quality. Besides, as for other AOPs, laboratory studies have only verified that UV/H_2O_2 can remove urea, while the removal effect is not ideal. Moreover, there is no information currently on the removal of urea by other AOPs such as ozone and UV/O_3. Thus, it is also of great significance to deeply analyze the performance, operating cost, operation difficulty, and side effects of various AOPs on urea removal.

In addition, the feasibility and cost-effectiveness of using reclaimed water as raw water should also be been brought into focus in the real application of UPW production. Although reclaimed water is considered as alternative water resource (normally for non-potable applications) due to lower cost compared to tap water, more complicated

treatment technologies and safety barrier system should be applied for UPW production, as reclaimed water serves as raw water source. Hence, the specific operation of introducing AOPs as a supplementary unit into real applications to maximize cost-effectiveness requests further explored. Besides, how to determine urea at extremely low concentrations is also a challenge.

5. Conclusions and Perspectives

(1) Using reclaimed water for UPW production will not only help to obtain a large amount of produced water but also achieve a sustainable utilization of water resources, which will become the trend of UPW production in the future. However, the content of organic compounds in reclaimed water is often higher than that in tap water, especially small molecular organic pollutants such as urea, which is difficult to remove through the traditional production process. To ensure the TOC concentration of effluent meets the requirements of the semiconductor industry, accordingly, the present UPW preparation process does have to deal with improvement and modification issues.

(2) The adsorption and efficiency in removing organic pollutants using IX resins are very limited. In addition, the GAC can only adsorb and remove some large non-polar molecules, and furthermore it is not effective in the removal of small molecular organic compounds. As for RO and UV irradiation, they can greatly reduce the TOC concentration of the effluent, while they are not the most efficient means of removing urea and other small molecular organic compounds. Consequently, it is difficult to completely remove small molecular organic compounds in water with a regular UPW production process.

(3) To resolve the influence of changing in raw water sources on UPW production, introducing novel and effective technologies on the basis of the existing production line to remove small molecule organic compounds in UPW systems is essential and helpful to meet the water standard desired in the semiconductor industry. AOPs have not been introduced into UPW production on a large scale. However, AOPs such as photocatalyst, activation of persulfate and electrocatalysis, provide a new strategy for the actual UPW production in the future.

Acknowledgement

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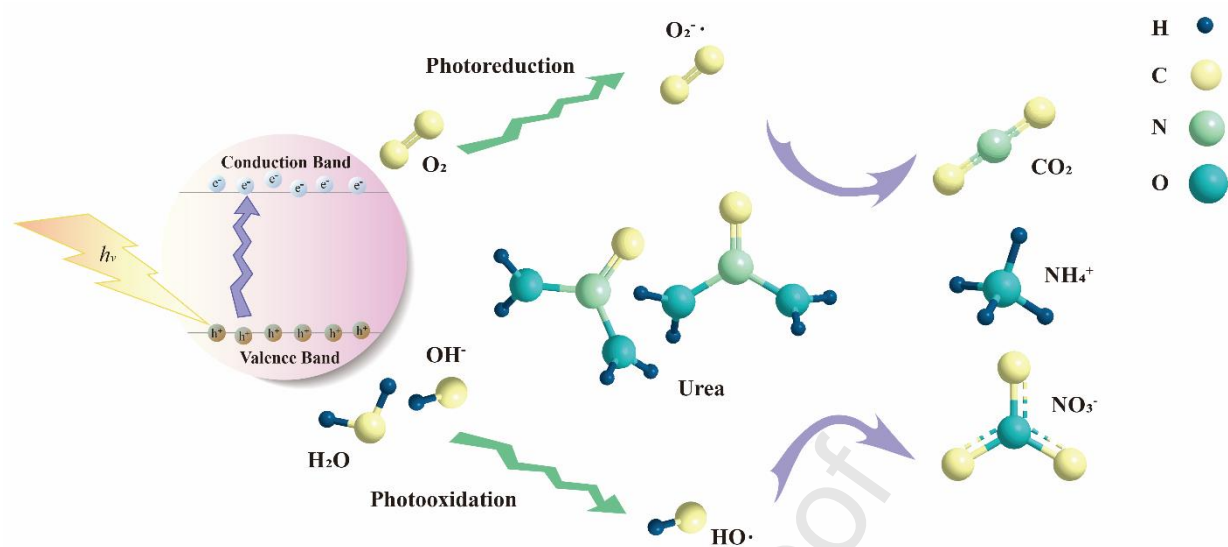
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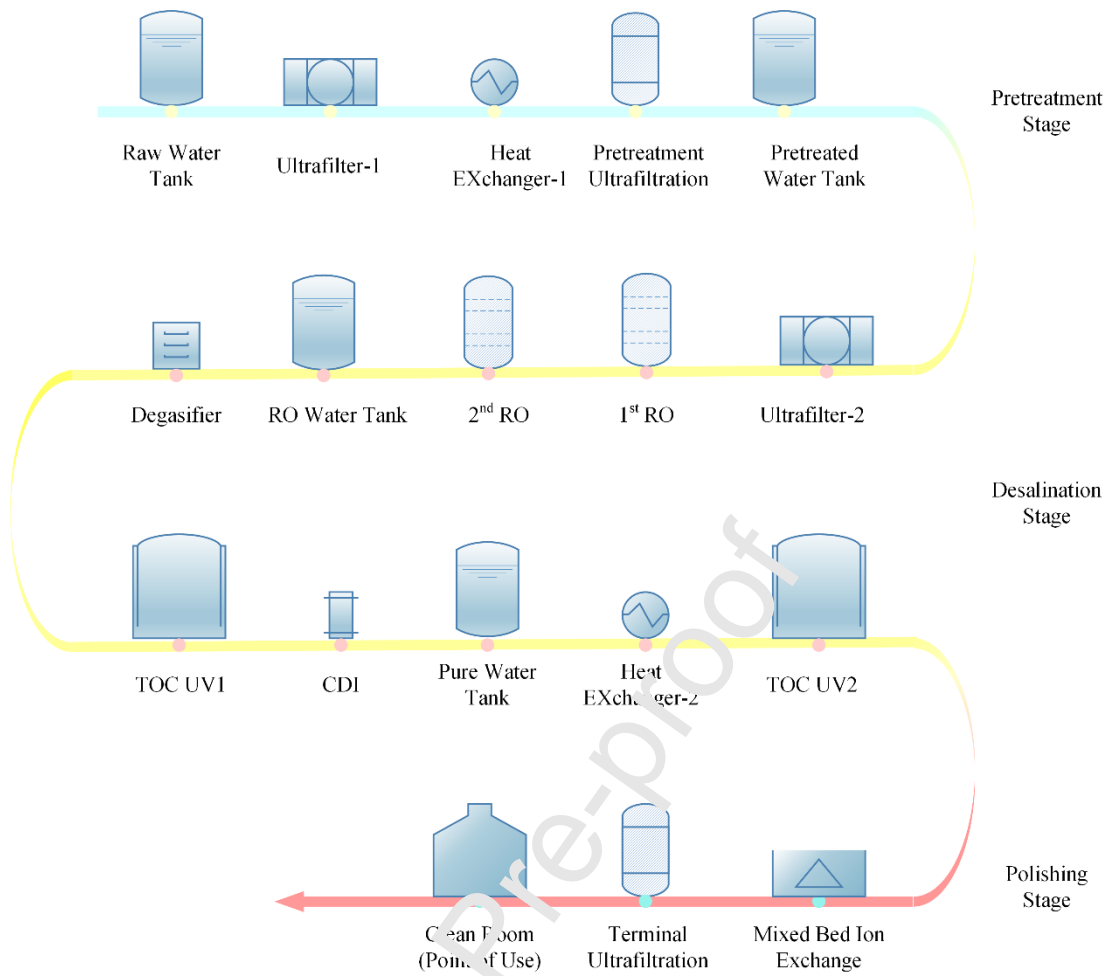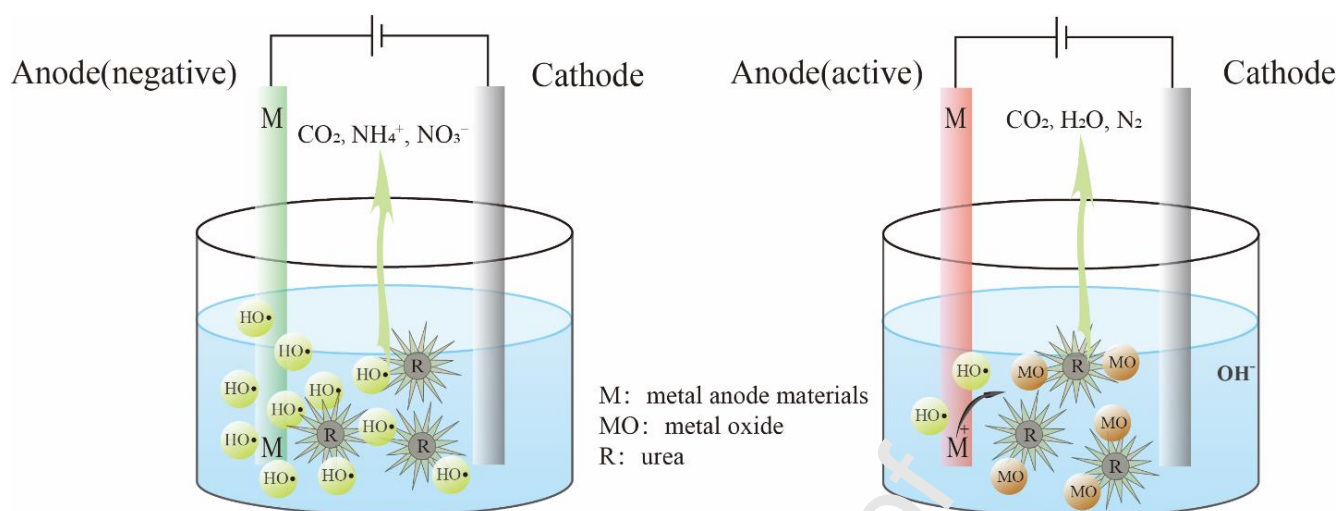
Fig. 1. Typical microelectronic UPW production system flow

Fig. 2. Schematic of semiconductor photocatalysis oxidation of urea in UPW

Fig. 3. Schematic of degradation of urea in UPW by Electrocatalytic Oxidation

Table 1 Reclaimed water quality in various regions

Location	Parameters					Reference
	COD (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	TOC (mg/L)	Turbidity (NTU)	
Melbourne Water's Western Treatment Plant (WTP), Australia	27.9 ± 1	13 ± 0.5	0.218 ± 0.02	–	0.9 ± 0.1	(Ibn Abdul Hamid et al., 2019)
Everbright Water, Shandong Province, China	–	16.42 ± 0.07	0.097 ± 0.009	–	1.07 ± 0.14	(Liu et al., 2020)
Jinan Urban Wastewater Treatment Plant, Shandong Province, China	–	6.058 –7.489	0.125 – 0.168	–	2.56 – 4.06	(Zhang et al., 2013)
Reclaimed Water Treatment Plant	–	–-	–	5.2-7.7	–	(Wu et al., 2016)
Truckee Meadows Water Reclamation Facility (TMWRF), Nevada, United States	–	–	-	6.67	–	(Yanala and Pagilla, 2020)
Xili Wastewater Treatment Plant, Shenzhen, China	20.5 ± 4	7.5 ± 1	0.158 ± 0.04	–	3.65 ± 1	(Liu et al., 2018)
Municipal Wastewater Treatment Plant, Harbin, China	–	11.5 ± 0.1	0.230 ± 0.002	–	–	(Tang et al., 2021)
Municipal Wastewater Treatment Plant, Mostoles, Spain	49.7 ± 3.5	–	0.247 ± 0.014	17.6 ± 1.9	2.4 ± 0.3	(Acero et al., 2010)
Municipal Wastewater Treatment Plant, Serbia	33.4 ± 2.7	–	–	–	–	(Bogunović et al., 2020)
Municipal Wastewater Treatment Plant, Ciudad Real, Spain	22.1	–	0.152	–	0.75	(Acero et al., 2016)
Municipal Wastewater Treatment Plant, Montpellier, France	50.3 ± 8.1	–	–	17.2 ± 1.2	–	(Ouarda et al., 2020)
Municipal Wastewater Treatment Plant, Montpellier, France	19.1 ± 2.7	–	–	7.4 ± 0.8	–	(Ouarda et al., 2020)

Wenquan Water Reclamation Plant, Beijing, China.	30.6 ± 15.5	–	0.199 ± 0.078	8.5 ± 1.8	4.3 ± 3.6	(Xiao et al., 2013)
Water Reclamation Plant, Beijing, China.	–	8.0 – 11.1	0.139 – 0.168	–	2.3 – 4.3	(Shi et al., 2021)
Municipal Wastewater Treatment Plant, Texas	21 ± 3.4	7.75 ± 0.101	0.165 ± 0.007	–	2.1 ± 0.5	(Gupta and Chellam, 2021)
Wastewater Treatment Plant, Shanghai, China	–	–	–	10.7 ± 1.3	2.6 ± 0.7	(Lin et al., 2011)

−: not mentioned.

Table 2 Microelectronic grade UPW classification

Standards		ASTM D5127-13（2018）						
Parameter		Type E-1	Type E-1.1	Type E-1.2	Type E-1.3	Type E-2	Type E-3	Type E-4
Linewidth (microns)		1.0-0.5	0.35-0.25	0.18-0.09	0.065-0.032	5.0-1.0	>5.0	-
Resistivity, 25°C (on-line)		18.1	18.2	18.2	18.2	16.5	12.0	0.5
TOC (ug/L) (on-line for <10 ppb)		5	2	1	1	50	300	1000
On-line particles/L (micron range）	>0.05 μm				500			
	0.05 ~0.1 μm		1000	200	N/A	-	-	-
	0.1~0.2 μm	1000	350	<100	N/A	-	-	-
	0.2~0.5 μm	500	<100	<10	N/A	-	-	-
	0.5~1.0 μm	200	<50	<5	N/A	-	-	-
	1.0 μm	<100	<20	<1	N/A	-	-	-

Standards		GB/T 11446.1-2013			
Parameter		EW- I	EW- II	EW- III	EW-IV
Resistivity, 25°C/ mΩ•cm		≥18 (5% of the time ≥17)	≥15 (5% of the time ≥13)	≥12.0	≥0.5
TOC (ug/L)		≤20	≤100	≤200	≤1000
Particles/L	0.05 ~0.1 μm	500	-	-	-
	0.1 ~0.2 μm	300	-	-	-
	0.2 ~0.3 μm	50	-	-	-
	0.3~0.5 μm	20	-	-	-
	>0.5 μm	4	-	-	-

N/A: not detected.

Table 3 Summary of the urea removal efficiencies by AOPs in various water types

Water type	Initial urea concentration	Final urea concentration	Urea removal efficiency	Method	Reference
Urine	12 mg/L	3.96 mg/L	67%	Photocatalytic oxidation	(Matoh et al., 2019)
UPW	1.65 μmol/L	0.17 μmol/L	90%	Sulfate radical-based advanced oxidation processes	(Choi and Chung, 2019)
Wastewater	2 g/L	-	100%[a]	Electrocatalytic oxidation	(Urbańczyk et al., 2020)
Distilled water	4.5 mg/L	1.13 mg/L	75%	UV/H_2O_2	(Bronk et al., 2000)
Swimming pool water	5 mg/L	4.97 mg/L	0.7 %	UV alone	(Long et al., 2019)
Swimming pool water	5 mg/L	4.36 mg/L	12.8%	UV/H_2O_2	(Long et al., 2019)
Swimming pool water	5 mg/L	> 4.60 mg/L	< 8%	UV/Na_2SO_3	(Long et al., 2019)
Swimming pool water	5 mg/L	3.40 mg/L	67.8%	UV/$K_2S_2O_8$	(Long et al., 2019)

[a] The BDD anode was used.

-: not detected.