

Review

## A review of semiconductor wastewater treatment processes: Current status, challenges, and future trends

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### ABSTRACT

Semiconductor wastewater has recently become an emerging issue with the development of semiconductor production owing to its severe toxicity and complexity. The increasing amount of semiconductor wastewater generated and discharged and the use of various chemicals in the semiconductor fabrication process highlight the necessity and importance of semiconductor wastewater treatment. This review describes various physical, chemical, biological, and hybrid or combined processes. These processes have been developed to remove and degrade fluoride, nitrogen, phosphate, turbidity, and organic compounds, including photoresist and washing solutions that are generally used in semiconductor fabrication. To properly treat semiconductor wastewater, understanding the characteristics, efficiency, effect factors, and limitations of the process is necessary. This review proposes future trends in the treatment process that can help minimize the semiconductor wastewater problem.

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### 1. Introduction

A semiconductor is classified into substances with  $10^4\text{--}10^{-10}$   $(\Omega\cdot\text{cm})^{-1}$  of metallic conductivity, typically consisting of silicon and germanium. Charge transport in a semiconductor is generated by positive or negative holes depending on the type of impurity doping (Seeger, 2013). These semiconductors can be produced in integrated circuits (IC), diodes, and transistors, so-called semiconductor products (Samsung, 2015g; Yacobi, 2003).

Along with the development of computers and the Internet, semiconductor products have been widely used to produce electric devices. Therefore, the use of semiconductors is broad and essential in the electric, medical, and military industries as well as our lives. The semiconductor market started increasing slowly in 1976, then explosively in the 1990s, and the market size in 2021 was US\$ 543,137,368 thousand (SIA, 2022b). Consequently, the annual market growth rate was expected to consciously increase along with the 4th industrial revolution, such as IoT, big data, and artificial intelligence (McClean, 2021b).

Therefore, the growth of the semiconductor industry has led to an

increase in the use of raw water for semiconductor production and in wastewater emissions. As a result, the global microelectronics industrial water quantity was US\$ 1973 million in 2017 and is expected to reach US\$ 3258 million by 2024 (Expert\_Market\_Research, 2022). In terms of the Republic of Korea, one of the leading countries in the system semiconductor industry, wastewater generation and discharge in 2019 increased by 177,937  $\text{m}^3/\text{d}$  (19.3%) and 164,833  $\text{m}^3/\text{d}$  (19.0%) from 2010, respectively (Statistics Korea, Domestic Wastewater Generation status). Therefore, appropriate treatment is required based on the increase in semiconductor wastewater generation.

The semiconductor fabrication process consists of wafer manufacturing, oxidation, photolithography, etching, deposition and ion implantation, metallization, electrical die sorting (EDS), and packaging (Tsai et al., 2002). Minuteness and delicacy are required to enhance product quality, and fabrication techniques have advanced. However, the complexity of its chemical composition makes wastewater difficult to treat.

A number of previous studies have attempted to apply physical, chemical, biological, or combined processes to semiconductor wastewater while maximizing their efficiency. However, studies on semiconductor wastewater are less developed than those on other

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<b>Nomenclature</b>	
ACF	Activated carbon filter
AEM	Anion exchange membrane
AFMBR	Anaerobic fluidized membrane bioreactor
AGMD	Air gap membrane distillation
AOB	Ammonia oxidizing bacteria
AOPs	Advanced oxidation processes
BAC	Biological activated carbon
BC	Breakpoint chlorination
BOD	Biological oxygen demand
BSR	Boron selective resin
CAGR	Compound annual growth rate
CANON	Completely autotrophic nitrogen removal over nitrite
CEDI	Continuous electro-deionization
CEMs	Cationic exchange membranes
CMP	Chemical mechanical polishing
COD	Chemical oxygen demand
CTW	Collection tank wastewater
CVP	Chemical vapor deposition
CWAO	Catalytic wet oxidation
DAC	Dodecylammonium chloride
DAF	Dissolved air flotation
DBGW	Diluted back grinding wastewater
DCMPW	Diluted chemical mechanical polishing wastewater
DEHP	Diethylhexyl phthalate
DGA	Diglycolamine
DiMAH	Dimethylammonium hydroxide
EC	Electro-coagulation
ECF	Electro-coagulation-flotation
ED	Electrodialysis
EDS	Electrical die sorting
EDX	Energy-dispersive X-ray spectroscopy
EL	Ethyl lactate
EO	Electrochemical oxidation
Fab	Fabrication plants
FBC	Fluidized bed crystallizer
FF	Fiber ball filtration
Ga	Gallium
GO	Graphene oxide
HMDS	Hexamethyldisilazane
HRT	Hydraulic retention time
HWW	High volatility wastewater
IC	Integrated circuits
IPA	Isopropyl alcohol
LbL	Layer-by-layer
LOW	Liquid organic wastes
LSW	Local scrubber water
LVW	Low volatility wastewater
MBBRs	Moving bed bioreactors
MD	Membrane distillation
MF	Microfiltration
MMA	Methyl methacrylate
MoMAH	Monomethyl ammonium hydroxide
NIPECs	Nonstoichiometric polyelectrolyte complexes
NMP	1-methyl-2-pyrrolidone
NOB	Nitrite-oxidizing bacteria
NPs	Nanoparticles
PA	Polyamide
PAC	Poly aluminum chloride
PCB	Printed circuit board
PEG	Polyethylene glycol
PEI	Polyethyleneimine
PFOS	Perfluoro octane sulfonate
PGMET	Propylene glycol methyl ether acetate
PHL	Phenol
PLC	Programmable logic controller
PR	Photoresist
PVC	Polyvinyl chloride
PVD	Physical vapor deposition
RO	Reverse osmosis
SBR	Sequencing batch reactor
SDS	Sodium dodecyl sulfate
SEM	Scanning electron microscope
SNPs	Silica nanoparticles
SOI	Sodium oleate
SS	Suspended solids
TAN	Total ammonia nitrogen
TEM	Transmission electron microscopy
TFC	Thin-film composite
TFT-LCD	Thin-film transistor liquid crystal display
TMAH	Tetramethylammonium hydroxide
TN	Total nitrogen
TOC	Total organic carbon
T-P	Total phosphorus
TriMAH	Trimethyl ammonium hydroxide
TTO	Total toxic organics
UF	Ultrafiltration
UPW	Ultrapure water
UV	Ultraviolet
VOCs	Volatile organic compounds
ZVI	Zero-valent iron

wastewaters. Therefore, studies on semiconductor wastewater treatment processes are required. This review describes the details of each treatment process along with challenges and suggests future trends. Besides, it provides a better understanding of how to treat semiconductor wastewater.

## 2. Background

### 2.1. Semiconductor industry

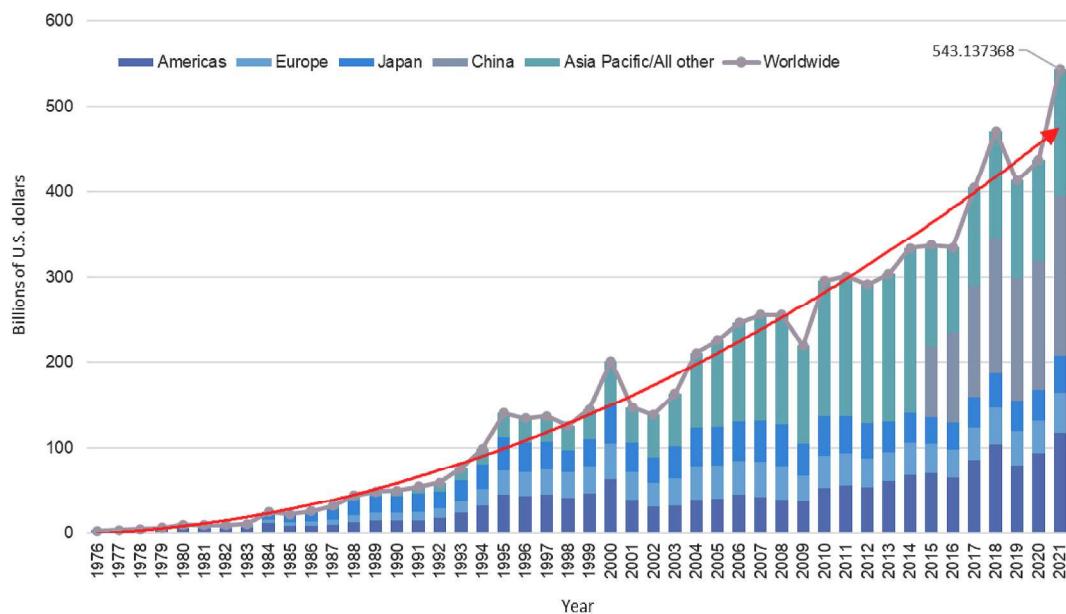
#### 2.1.1. Market status

Studies on semiconductor fabrication started when the transistor was invented by the Bell Lab in 1947, and it has been extensively developed since 1976. Therefore, it was expected that the market size would reach US\$ 543,137,368 thousand in 2022 and would show an 8.6% of compound annual growth rate (CAGR) until 2021 (McClean, 2021b; SIA,

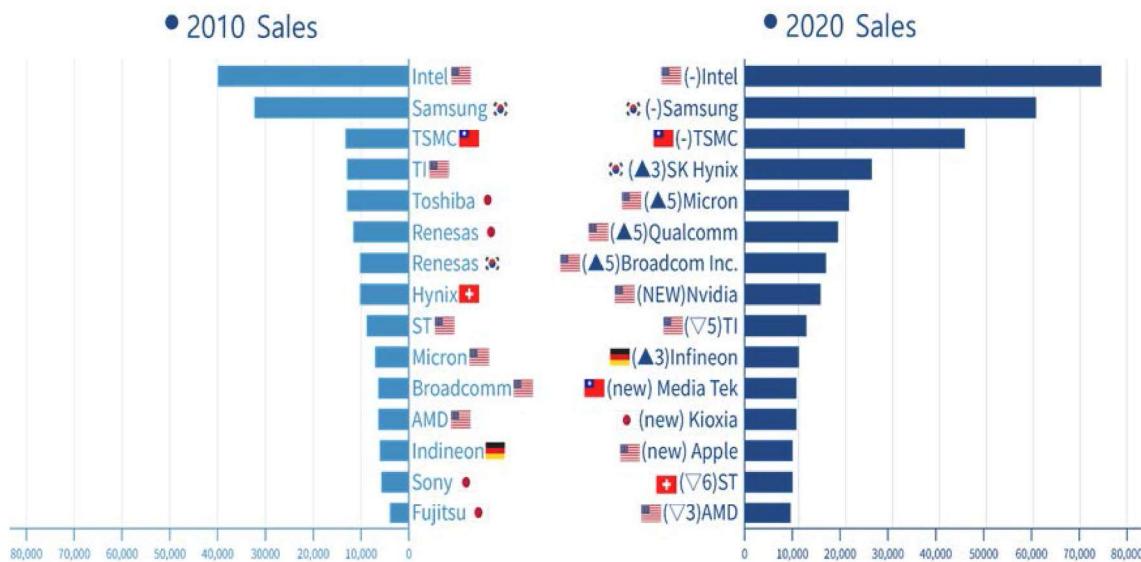
2022b) (Fig. 1 and Table S1). However, only 3% of the 2019 market size was increased in 2020 due to the recent Covid-19 pandemic across the economy. Nevertheless, it was predicted to reach US\$ 1135.3 billion in 2021, an increase of 13% annually (McClean, 2021b). Meanwhile, a steady growth of the semiconductor market was predicted along with the 4th industrial revolution, such as the IoT, big data, and AI (SIA, 2022a). Intel, Samsung, TSMC, and SK Hynix currently lead the global semiconductor market (McClean, 2021b). Specifically, Samsung, TSMC, and SK Hynix are semiconductor fabrication plants (Fab) or foundries that produce semiconductor products using the designed circuits and operate their semiconductor wastewater treatment (McClean, 2021b) (Figs. 2–4, Tables 1 and 2).

#### 2.1.2. Semiconductor manufacturing processes

Semiconductors are produced by wafer manufacturing, oxidation, photolithography, etching, deposition, ion implantation, metallization,



**Fig. 1.** Worldwide semiconductor market billings by year (McClean, 2021b).



**Fig. 2.** Semiconductor companies leading semiconductor market (McClean, 2021b).

EDS, packaging, and assembly processes.

In the wafer manufacturing process, silicon extracted from sand is manufactured in a single crystal-shaped ingot, and then the ingot is cut to manufacture a thin disk-type wafer several hundred micrometers thick. The cut wafers are cleaned and polished to improve the surface quality (Geng and CMfgE, 2018; Pei et al., 2008; Samsung, 2015a).

In the oxidation process, the slick wafers are oxidized with oxygen or water vapor to form the thin  $\text{SiO}_2$  under 900 – 1100 °C, preventing contaminating the wafer. Dry and wet are the two oxidation processes (Chao, 2001; Geng and CMfgE, 2018; Reinhardt and Reidy, 2011).

In photolithography, the designed circuit is formed on the wafer, and circuit patterns are drawn. Washing is performed to remove impurities from the wafers before the photolithography process. A photoresist (PR) is spread on the wafer surface, and a mask on which the designed circuit is drawn is placed on top of the wafer. The patterns are then exposed to light and copied. If the exposed part of the wafer is removed during the photolithography process, it is a positive photograph; conversely, the

unexposed part is called a negative photograph (Den et al., 2002; Jang et al., 2019; Samsung, 2015b).

In the etching process, unwanted parts of  $\text{SiO}_2$  on the wafer are etched (removed) by dry or wet etching processes. The reactive gas is used for etching under plasma ionization in the dry etching process, whereas an acidic or basic solution is used in the wet etching process (Samsung, 2015c; Wald and Jones, 1987). After the etching process, the remaining PR chemicals on the drawn wafer are removed using a sulfuric acid ( $\text{H}_2\text{SO}_4$ ) solution, and a layered circuit is formed by repeating the drawing circuit patterns. Consequently, several gaps between each layer are created and planarized via chemical mechanical polishing (CMP) to prevent wiring disconnection.

In the CMP process, the polishing solution (slurry) is sprayed onto the polishing pad, and the wafer fixed to the wafer carrier is placed on the pad. When the pad rotates, the wafer flattens (Pei et al., 2008). Recently,  $\text{SiO}_2$  as a slurry, as well as metal ions such as copper (Cu), cerium (Ce), tungsten (W), and aluminum (Al), have been applied (Nishi

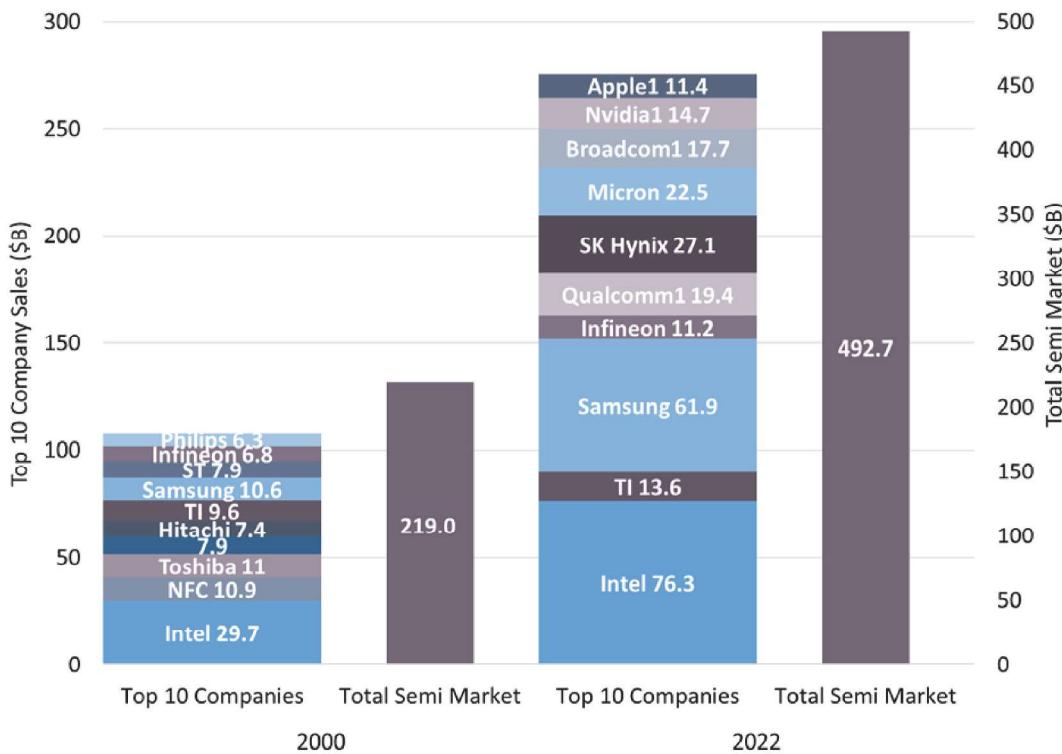


Fig. 3. Semiconductor market sales.

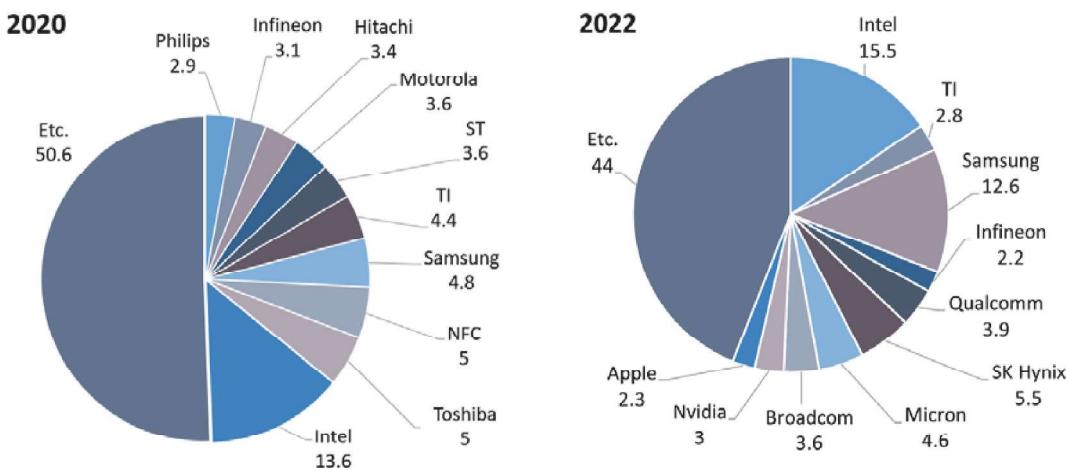


Fig. 4. Semiconductor market share (McClean, 2021a).

and Doering, 2000; Yang, 2002). After the CMP process, a washing step is required to remove the remaining impurities on the surface, which is classified as dry and wet washing. Plasma discharge or ultraviolet (UV)/O<sub>3</sub> is used for dry washing; hence, chemical solutions are used (Nishi and Doering, 2000). Furthermore, the additional washing is demanded to fully reject the remained chemicals using ultrapure water, and it can cause an increase of wastewater discharge in other words.

In the deposition process, the thin films are formed on the layered and etched wafers by chemical vapor deposition (CVD) or physical vapor deposition (PVD). The thin film is categorized into two parts: a conductive metal layer to connect the circuits electrically and an insulating dielectric to isolate the inner layers or prevent contamination electrically (Kim et al., 2011; Samsung, 2015d).

Simultaneously, ion implantation makes the wafer nonconductive and conductive. For example, an n-type semiconductor can be fabricated to generate surplus electrons when Group 15 elements (e.g., phosphorus

(P) or arsenic (As) with five outermost electrons) are added to silicon, which is a Group 14 element. However, adding Group 13 elements (e.g., boron (B) with three outermost electrons) can produce a p-type semiconductor with holes (El-Kareh and Hutter, 2012; Samsung, 2015d; Seeger, 2013).

In the metallization process, electrically conductive metals are used to connect the metal wires with the circuit pattern to conduct electricity, and then non-defective wafers are selected for the EDS process (Samsung, 2015e, 2015f). Finally, in the packaging process, high-quality semiconductor products are produced through cutting, wire bonding and molding processes for suitable use in electronic devices (Samsung, 2015g).

#### 2.1.3. Wastewater in semiconductor manufacturing processes

During the wafer manufacturing process, hydrochloric acid gas is used to extract Si, which is the main source of the wafer, and gallium

**Table 1**  
Top 10 worldwide semiconductor sales leaders (excluding pure-play foundries) (McClellan, 2021a).

Rank	1993		2000		2008		2017		2020			
	Company	Sales (\$B)	Share (%)	Company	Sales (\$B)	Share (%)	Company	Sales (\$B)	Share (%)	Company	Sales (\$B)	Share (%)
1	Intel	7.6	9.2	Intel	29.7	13.6	Intel	34.5	13.0	Samsung	65.9	14.8
2	NFC	7.1	8.6	Toshiba	11.0	5.0	Samsung	20.3	7.6	Intel	61.7	13.9
3	Toshiba	6.3	7.6	NFC	10.9	5.0	TI	11.6	4.4	SK Hynix	26.7	6.0
4	Motorola	5.8	7.0	Samsung	10.6	4.8	Toshiba	10.4	3.9	Micron	23.9	5.4
5	Hitachi	5.2	6.3	TI	9.6	4.4	ST	10.3	3.9	Broadcom <sup>a</sup>	17.8	4.0
6	TI	4.0	4.8	Motorola	7.9	3.6	Renesas	7.0	2.6	Qualcomm <sup>a</sup>	17.0	3.8
7	Samsung	3.1	3.8	ST	7.9	3.6	Qualcomm <sup>a</sup>	6.5	2.4	TI	13.9	3.1
8	Mitsubishi	3.0	3.6	Hitachi	7.4	3.4	Sony	6.4	2.4	Toshiba	13.3	3.0
9	Fujitsu	2.9	3.5	Infineon	6.8	3.1	Hynix	6.2	2.3	Nvidia <sup>a</sup>	9.4	2.1
10	Matsushita	2.3	2.8	Philips	6.3	2.9	Infineon	5.9	2.2	NXP	9.3	2.1
	Top 10 total (\$B)	47.2	57.2		108.1	49.4		119.1	44.9		259.0	58.2
	Semi Market (\$B)	82.6	100		219.0	100		265.2	100		445.2	100

<sup>a</sup> Fabless

arsenide (GaAs) is used as a substitute for Si. Therefore, wafer polishing is necessary to enhance the wafer quality. The hydrochloric acid (HCl), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), 5% sodium hypochlorite (NaOCl), ethoxylated amine, and ammonium hydroxide (NH<sub>2</sub>OH) solutions are used to remove the contaminants from the wafer surface after the wafer polishing (KOSHA, 2020). In the oxidation process, SC-1 (including NH<sub>4</sub>OH, H<sub>2</sub>O<sub>2</sub>, and H<sub>2</sub>O), SC-2 (including HCl, H<sub>2</sub>O<sub>2</sub>, and H<sub>2</sub>O), and HF solutions are used as washing solutions to remove impurities after SiO<sub>2</sub> is generated (Sparacin et al., 2005).

Typical macromolecule resins used in the photoresist process are xylene (C<sub>8</sub>H<sub>10</sub>), Novolak resin, aromatic compounds, and organic solvents (Kim et al., 2011). As In addition, tetramethylammonium hydroxide (TMAH), aliphatic compounds ethyl 3-ethoxypropionate (C<sub>7</sub>H<sub>14</sub>O<sub>3</sub>), gamma-butyrolactone (C<sub>4</sub>H<sub>6</sub>O<sub>2</sub>), and n-methyl-2-pyrrolidone (C<sub>5</sub>H<sub>9</sub>NO) are commonly used (Den et al., 2002; Jang et al., 2019; Samsung, 2015b). Moreover, an adhesion promoter is used to improve the adhesion between the wafer and photoresist, with hexamethyldisilazane (HMDS, C<sub>6</sub>H<sub>19</sub>NSi<sub>2</sub>) being the most commonly used (Horibe et al., 2005; Park et al., 2011).

The etching process can be divided into dry and wet processes. Gaseous compounds such as ammonia (NH<sub>3</sub>), argon (Ar), chlorine (Cl), hydrogen (H), sulfur hexafluoride (SF<sub>6</sub>), and fluoroform (CHF<sub>3</sub>) are used in dry etching. In contrast, compounds such as acetic acid (CH<sub>3</sub>COOH), ammonium fluoride (NH<sub>4</sub>F), ammonium hydroxide (NH<sub>4</sub>OH), hydrochloric acid (HCl), hydrofluoric acid (HF), nitric acid (HNO<sub>3</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and isopropyl alcohol (IPA, C<sub>3</sub>H<sub>8</sub>O) are used in the wet etch process (Wald and Jones, 1987). After etching, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) removes the photoresist. After drawing the circuits several times, oxides such as silicon dioxide (SiO<sub>2</sub>) and metals such as copper (Cu), cerium (Ce), tungsten (W), and aluminum (Al) are used as a slurry to chemically polish the wafer in the CMP process (KOSHA, 2020; Lee, D. et al., 2016; Lee, H. et al., 2016).

The deposition processes are categorized as CVD and PVD. Dichlorosilane (Cl<sub>2</sub>H<sub>2</sub>Si), nitrogen trifluoride (F<sub>3</sub>N), nitrous oxide (N<sub>2</sub>O), ozone (O<sub>3</sub>), phosphine (PH<sub>3</sub>), and silane (SiH<sub>4</sub>) are mainly used in CVD, whereas gaseous compounds (e.g., hydrogen (H<sub>2</sub>), hydrochloric acid (HCl) and hydrogen fluoride (HF) are used in PVD (Kim et al., 2011; KOSHA, 2020). Additionally, in the metallization process, metals such as aluminum (Al), copper (Cu), titanium (Ti), and tungsten (W) are used to connect the wafers to circuits (El-Kareh and Hutter, 2012; Samsung, 2015d; Seeger, 2013). During the packaging process, semiconductor products are assembled using benzene (C<sub>6</sub>H<sub>6</sub>) or formaldehyde (CH<sub>2</sub>O) finally (Kim et al., 2011; KOSHA, 2020).

A schematic diagram that briefly summarizes the semiconductor fabrication process, wastewater generation, treatment process, and discharge is shown in Fig. 5.

## 2.2. Status of global semiconductor wastewater (generation and discharged of wastewater, treatment process, law)

### 2.2.1. Status of USA

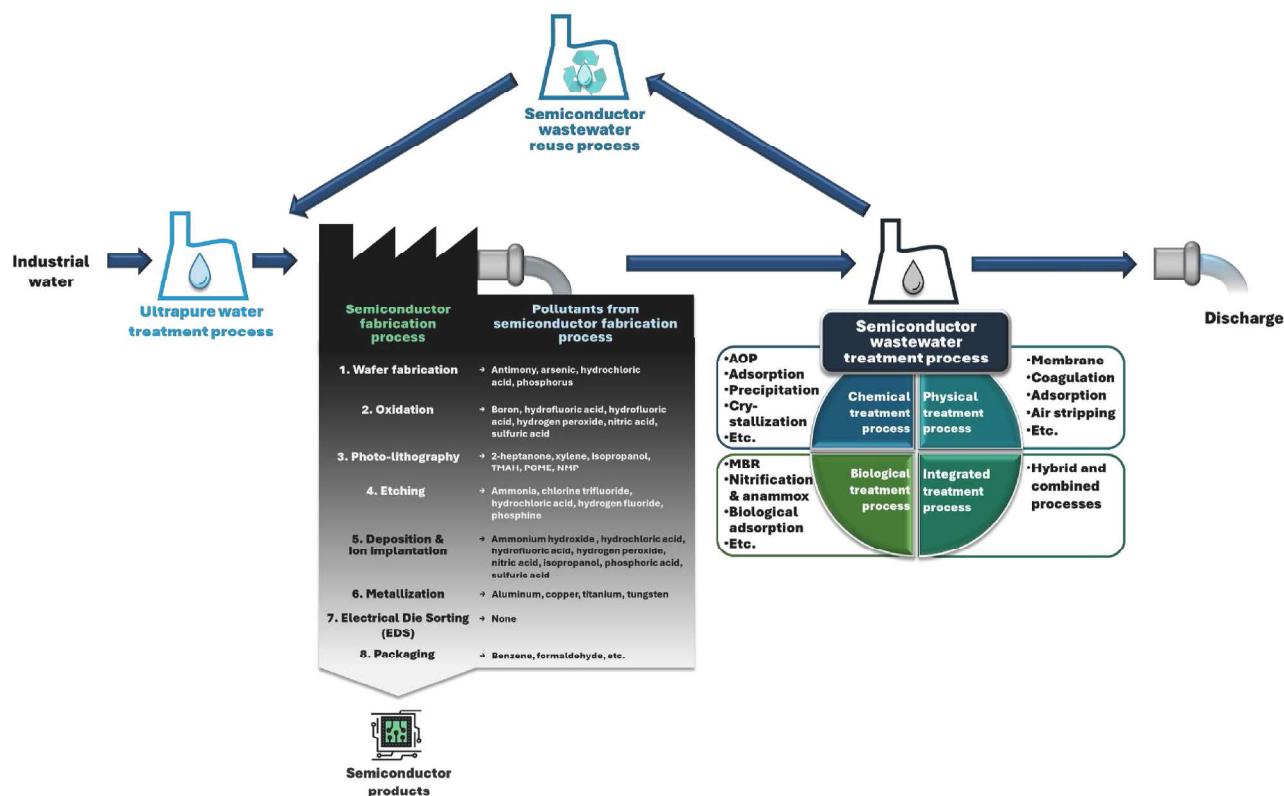
The United States Environmental Protection Agency (EPA) promulgated the Electrical and Electronic Components (E&EC) Effluent Guidelines and Standards, which address direct and indirect dischargers (40 CFR Part 469) (Table 3). The guidelines and standards are included in the National Pollutant Discharge Elimination System (NPDES), which permits direct dischargers and other control mechanisms for indirect dischargers (EPA, 2018) (Table 4). E&EC manufacturing facilities are subcategorized into four groups: semiconductors, electronic crystals, cathode ray tubes, and luminescent materials, which are covered by the regulations (EPA, 2020) (Table 5).

### 2.2.2. Status of semiconductor industry in TSMC

The amount of discharged semiconductor wastewater from TSMC was 49.20 million metric tons in 2021, which is a 5.10% increase compared to 2020, and the amount of wastewater has been continuously

**Table 2**  
Semiconductor manufacturing process.

Steps	1	2	3	4	5	6	7	8
Major process Details	Wafer fabrication Ingot casting	Oxidation	Photo-lithography Photoresist spread	Etching Dry or wet etching	Deposition & Ion Implantation	Metallization	Electrical Die Sorting (EDS)	Packaging
	Ingot cutting		Exposure	Chemical & mechanical polishing(CMP)				
	Wafer polishing Circuit design		Develop	Cleaning				



**Fig. 5.** A schematic diagram summarized the semiconductor fabrication process, wastewater generation, treatment process and discharge.

increasing since 2017 (Table 6). Hence, the TSMC reported that different treatment processes were applied depending on the type of wastewater used for reuse and recycling. For hydrofluoric acid (HF)-or ozone ( $O_3$ )-containing semiconductor wastewater, chemical coagulation, ammonia removal, and biological processes were performed as previously described. Recycling and upcycling treatments are used for acidic, caustic, and organic wastewater, including TMAH- or ozone ( $O_3$ )-containing wastewater. Electroplating, copper-containing, cobalt-containing, and others are the four types of CMP wastewater treatments. Along with different treatment processes depending on the type of CMP wastewater, chemical coagulation recycling of CMP wastewater and upcycling for copper-containing CMP wastewater systems are operating. High-concentration liquid wastes are classified into sulfuric acid ( $H_2SO_4$ )-, copper sulfate ( $CuSO_4$ )-, and cobalt (Co)-containing wastewater, and others. Sulfuric acid ( $H_2SO_4$ )-containing wastewater is treated by a reclamation process that produces sulfuric acid ( $H_2SO_4$ ) for recycling, and copper (Cu)-or cobalt (Co)-containing electroplating wastewater, which produces copper (Cu) or cobalt (Co) bars (Tables 7 and 8).

TSMC operated subdivided treatment and recycling processes depending on the different types of wastewater sources and increased

the amount of recycled wastewater, showing that 186.3 million metric tons of wastewater had been recycled in 2021. As shown in Table 7, the total amount of wastewater was increased; however, the average process water recycling rate exceeded 85 %. The table shows that they continuously strive to reuse wastewater in preparation for the increasing amounts of wastewater. This is related to supplying water for semiconductor fabrication and minimizing the impact of water shortages due to recent droughts (TSMC, 2021).

#### 2.2.3. Status of semiconductor industry in Korea

The Korean Ministry of Environment (MOE) reported that the generation and discharge of wastewater from semiconductor manufacturing industries in 2019 increased by 177,937 m<sup>3</sup>/d (19.3 %) and 164,833 m<sup>3</sup>/d (19%), respectively, from 2010 (Fig. 6). The semiconductor wastewater in Korea is predicted to continue increasing. In Korea, the emission and treatment types of specific substances harmful to water quality, designated by relevant laws and regulations (Water Environment Conservation Act) based on chemicals used in semiconductor manufacturing processes, must be reported by each workplace (MOE).

In the case of Samsung Electronics, copper (Cu)- and selenium (Se)-containing semiconductor wastewater was discharged, and the

**Table 3**

Chemical use of semiconductor manufacturing process.

Steps	1	2	3	4	5	6	7	8
Name of process	Wafer fabrication	Oxidation	Photolithography	Etching	Deposition & Ion Implantation	Metallization	Electrical Die Sorting (EDS)	Packaging
Chemical use	Antimony (Sb)	Ammonium hydroxide	Macromolecule resin	Wet etching	Chemical vapor deposition (CVD)	Aluminum (Al)	Benzene ( $C_6H_6$ )	
	Arsenic (As) ( $NH_4OH$ )	Acrylate polymer	Acetic acid ( $C_2H_4O_2$ )	Acetylene ( $C_2H_2$ )	Copper ( $Cu$ )	Formaldehyde ( $CH_2O$ )		
	Boron (B) Arsine ( $AsH_3$ )	Novolak resin	Ammonium fluoride	Ammonia ( $NH_4$ )	Titanium (Ti)	etc.		
	Cokes Boron	Poly	( $FH_4N$ )	Chlorine trifluoride ( $ClF_3$ )	Diborane ( $B_2H_6$ )	Tungsten (W)		
	Hydrochloric acid gas (HCl) ( $BBr_3$ )	( $CH_4OSi_n$ )	Ammonium hydroxide ( $NH_4OH$ )	Dichlorosilane ( $Cl_2H_2Si$ )	Hydrogen ( $H_2$ )	Hydrochloric acid (HCl)		
	Phosphorus (P) ( $Cl_2H_2Si$ )	Polyhydroxystyrene derivative	Hydrofluoric acid (HF)	Nitric acid ( $HNO_3$ )	Hydrogen fluoride (HF)			
	Polysiloxanes (Silicone) Hydrofluoric acid (HF)	Polyisoprene ( $(C_5H_8)_n$ )	Hydrogen peroxide ( $H_2O_2$ )	Phosphoric acid ( $H_3PO_4$ )	Nitrogen ( $N_2$ )			
	Hydrogen peroxide ( $H_2O_2$ )	Polymethacrylate derivative	Isopropanol (Isopropyl alcohol, IPA) ( $C_3H_8O$ )	Polyethylene glycol mono (4-tert-octylphenyl) ether	Nitrogen trifluoride ( $NF_3$ )			
	Nitric acid ( $HNO_3$ )	Organic solvent	Nitric acid ( $HNO_3$ )	Octafluoropropane ( $(C_2H_4O_2)Cl_4H_2SiO$ )	Nitrous oxide ( $N_2O$ )			
	Phosphine ( $C_4H_{10}O_2$ )	2-Ethoxyethanol	Phosphoric acid ( $H_3PO_4$ )	Sulfuric acid ( $H_2SO_4$ )	Ozone ( $O_3$ )			
	( $H_3P$ )	2-Heptanone ( $C_7H_{14}O$ )	Polyethylene glycol	Ammonia ( $NH_3$ )	Phosphine ( $H_3P$ )			
	Silane ( $SiH_4$ )	2-Methoxy-1-propanol	mono (4-tert-octylphenyl) ether	Argon (Ar)	Silane ( $SiH_4$ )			
	Sulfuric acid ( $C_4H_{10}O_2$ )	2-Methoxy-1-propyl acetate ( $C_6H_{12}O_3$ )	Octafluoropropane ( $(C_2H_4O_2)Cl_4H_2SiO$ )	Boron trichloride ( $BCl_3$ )	Silicon tetrafluoride ( $F_4Si$ )			
	( $H_2SO_4$ )	Butyl acetate ( $C_6H_{12}O_2$ )	Carbon monoxide ( $CO$ )	Carbon tetrafluoride ( $CF_4$ )	Tetraethoxysilane ( $(C_8H_{20}O_4Si)$ )			
	Cresol ( $C_7H_8O$ )	Cyclohexanone ( $C_6H_{10}O$ )	Carbon monoxide ( $CO$ )	Carbonyl sulfide (COS)	Triethyl borate ( $C_6H_{15}BO_3$ )			
	Ethyl lactate ( $C_5H_{10}O_3$ )	Ethylbenzene ( $C_8H_{10}$ )	Carbon monoxide ( $CO$ )	Chlorine ( $Cl_2$ )	Difluoromethane ( $CF_2$ )			
	Ethylbenzene ( $C_8H_{10}$ )	Isopropyl alcohol (IPA) ( $C_3H_8O$ )	Carbon monoxide ( $CO$ )	Hexafluoro-1,3-butadiene ( $C_4F_6$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Isopropyl alcohol (IPA) ( $C_3H_8O$ )	Methyl 2-hydroxyisobutyrate ( $C_5H_{10}O_3$ )	Carbon monoxide ( $CO$ )	Hexafluoroethane ( $C_2F_6$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Methyl 3-methoxypropionate ( $C_5H_{10}O_3$ )	Methyl 2-hydroxyisobutyrate ( $C_5H_{10}O_3$ )	Hydrogen ( $H_2$ )	Hydrogen bromide (HBr)	Trimethyl borate ( $C_3H_9BO_3$ )			
	Propylene glycol monomethyl ether (PGME) ( $C_4H_{10}O_2$ )	Propylene glycol monomethyl ether (PGME) ( $C_4H_{10}O_2$ )	Hydrogen ( $H_2$ )	Methane ( $CH_4$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Propylene glycol monomethyl ether (PGMEA) ( $C_6H_{12}O_3$ )	Propylene glycol monomethyl ether (PGMEA) ( $C_6H_{12}O_3$ )	Hydrogen ( $H_2$ )	Nitrogen ( $N_2$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Xylene ( $C_8H_{10}$ )	$\gamma$ -Butyrolactone ( $C_4H_6O_2$ )	Octafluorocyclobutane ( $C_4F_8$ )	Sulfur hexafluoride ( $SF_6$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	$\gamma$ -Butyrolactone ( $C_4H_6O_2$ )	Developer	Octafluorocyclopentene ( $C_5F_8$ )	Trifluoromethane ( $CHF_3$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Developer	1-Methoxy-2-propyl acetate	Octafluoropropane ( $C_3F_8$ )	Etc.	Trimethyl borate ( $C_3H_9BO_3$ )			
	1-Methoxy-2-propyl acetate	Ozone ( $O_3$ )	Octafluorocyclobutane ( $C_4F_8$ )	Silane ( $SiH_4$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	(Propylene glycol monomethyl ether acetate, PGMEA) ( $C_6H_{12}O_3$ )	Sulfur hexafluoride ( $SF_6$ )	Octafluorocyclopentene ( $C_5F_8$ )	Tetrakis (dimethylamido) titanium (IV) (TDMA)	Trimethyl borate ( $C_3H_9BO_3$ )			
	( $C_6H_{12}O_3$ )	Trifluoromethane ( $CHF_3$ )	Octafluoropropane ( $C_3F_8$ )	( $C_8H_{24}N_4Ti$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Aliphatic hydrocarbons	Etc.	Octafluorocyclobutane ( $C_4F_8$ )	Titanium tetrachloride ( $TiCl_4$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Ethyl 3-ethoxypropionate ( $C_7H_{14}O_3$ )	Boron trichloride ( $BCl_3$ )	Octafluorocyclopentene ( $C_5F_8$ )	Tungsten hexafluoride ( $F_6W$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	N-methyl-2-pyrrolidone ( $C_6H_9NO$ )	Bromine ( $Br_2$ )	Octafluoropropane ( $C_3F_8$ )	Etc.	Trimethyl borate ( $C_3H_9BO_3$ )			
	Polyamide acid	Carbon tetrachloride ( $CCl_4$ )	Octafluorocyclobutane ( $C_4F_8$ )	Arsine ( $AsH_3$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Tetramethylammonium hydroxide (TMAH) ( $C_4H_{12}N.HO$ )	Carbon tetrachloride ( $CCl_4$ )	Octafluorocyclopentene ( $C_5F_8$ )	Boron tribromide ( $BBr_3$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	Xylene ( $C_8H_{10}$ )	Carbon tetrachloride ( $CCl_4$ )	Octafluoropropane ( $C_3F_8$ )	Phosphine ( $HP_3$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	$\gamma$ -Butyrolactone ( $C_4H_6O_2$ )	Chromium trioxide ( $CrO_3$ )	Octafluorocyclobutane ( $C_4F_8$ )	Trifluoromethane ( $CHF_3$ )	Trimethyl borate ( $C_3H_9BO_3$ )			
	( $C_4H_6O_2$ )	Trifluoromethane ( $CHF_3$ )	Octafluorocyclopentene ( $C_5F_8$ )		Trimethyl borate ( $C_3H_9BO_3$ )			

**Table 4**

40 CFR part 469 - electrical and electronic components point source category (EPA, 2018).

Pollutant or pollutant property	Maximum for any 1 day (mg/L)	Average of daily values for 30 consecutive days (mg/L)
pH	6~9	6~9
Total suspended solids (TSS)	61	23
Total toxic organics (TTO)	1.37	Not applicable
Fluoride (T)	32	17.4
Arsenic (T)*	2.09	0.83
Cadmium (Cd)	0.06	0.03
Chromium (Cr)	0.65	0.3
Lead (Pb)	1.12	0.41
Zinc (Z)	1.38	0.56
Antimony (Sb)	0.1	0.04
Total toxic organics (TTO)- Substance name (CAS Name)		CAS No.
Dioctyl phthalate ( $C_{24}H_{38}O_4$ )		117-81-7
Dibutyl phthalate ( $C_{16}H_{22}O_4$ )		84-74-2
Butyl benzyl phthalate ( $C_{19}H_{20}O_4$ )		85-68-7
Isophorone ( $C_9H_{14}O$ )		78-59-1
Anthracene ( $C_{14}H_{10}$ )		120-12-7
1,2,4-Trichlorobenzene ( $C_6H_3Cl_3$ )		120-82-1
1,2-Dichlorobenzene ( $C_6H_4Cl_2$ )		95-50-1
1,3-Dichlorobenzene		541-73-1
1,4-Dichlorobenzene		106-46-7
Ethylbenzene		100-41-4
Toluene		108-88-3
1,1,1-Trichloroethane		71-55-6
1,1,2-Trichloroethane		79-00-5
1,2-Dichloroethane		107-06-2
1,1-Dichloroethylene		75-35-4
1,2-Diphenylhydrazine		122-66-7
Bromodichloromethane		75-27-4
Dichloromethane		75-09-2
Carbon tetrachloride		56-23-5
Chloroform		67-66-3
Naphthalene		91-20-3
Phenol		108-95-2
2,4,6-Trichlorophenol		88-06-2
2,4-Dichlorophenol		120-83-2
2-Chlorophenol		95-57-8
2-Nitrophenol		88-75-5
4-Nitrophenol		100-02-7
Tetrachloroethylene		127-18-4

\*The arsenic (T) limitation only applies to manufacturers of gallium or indium arsenide crystals.

wastewater directly flowing into the water pollution prevention facilities was treated or individually treated (then directly discharged) (Table 9). The semiconductor wastewater treatment process consisted of the 1st inorganic wastewater treatment, organic wastewater treatment, and the 2nd inorganic wastewater treatment process. Chemical treatment, pH adjustment, and sedimentation were performed during the 1st inorganic wastewater treatment process. Organic wastewater treatment consists of a biological treatment process and an organic removal process. In the 2nd inorganic wastewater treatment process, semiconductor wastewater is subjected to chemical treatment and pH adjustment and discharged (Samsung, 2022).

Therefore, the amount of discharged semiconductor wastewater in 2021 of Samsung Electronics increased by 19.92% (30,955,000 tons) compared with 2020, showing that the progress of discharged semiconductor wastewater has continuously increased. In contrast, discharged water pollutants decreased in 2021. Samsung Electronics is currently reusing semiconductor wastewater to protect and replace water resources. The process for reusing wastewater into ultrapure water is in operation, and the reuse rate has been steadily increasing since 2015 (Samsung, 2022) (Tables 10 and 11).

In the case of the SK Hynix, copper-, selenium-, or formaldehyde-containing semiconductor wastewater was discharged and treated via

water pollution prevention facilities, individual treatment (then direct discharge), or commissioned treatment (Table 12). In 2020, the amount of discharged wastewater increased by 56.8% compared with 2015, which was related to the amount of wastewater reuse that had increased approximately twice (Table 13). The semiconductor wastewater treatment process of SK Hynix was composed of three steps: physicochemical treatment via aggregation and precipitation, biological treatment using membrane bioreactor (MBR), activated carbon (AC), and TP removal, and an advanced treatment process using ozone ( $O_3$ ). In addition, UF (ultrafiltration) and RO (reverse osmosis) processes were added for wastewater reuse to increase water quality (hynix (2022)) (Table 14).

Common things of U.S., TSMC, Samsung electronics and SK Hynix which are reading the semiconductor industry are that the generated wastewater is increasing and the chemical, physical and biological treatment process have been used complexly. In U.S. and TSMC, the TMAH removal and reuse are focused in the semiconductor wastewater treatment. In TSMC and Korea companies, the semiconductor wastewater process has been developed to remove the heavy metals such as copper, cobalt and selenium. The wastewater reuse process has been applying world widely, however, it seems that the studies are needed to enhance the wastewater treatment process efficiency and reuse process rate.

### 2.3. Research status of semiconductor wastewater treatment processes

The studies for semiconductor wastewater treatment process have been conducted from end of 1990 and around 60 papers were published, which are comparably few (Fig. 7). Currently, the global demand of the semiconductor wastewater treatment process has been expanded with the growth of the semiconductor market size. In addition, the semiconductor wastewater became the issue due to the complexity to treat it and its toxicity. Thus, as much as the fabrication process which became complicated, there is a need to understand the intricate semiconductor wastewater treatment process.

A number of review papers about the industrial wastewater treatment have been published, however, we have not found yet any review paper dealing with the semiconductor wastewater treatment process. For example, research has been conducted globally on the removal of fluorine compound (e.g. polyfluorinated alkyl substances or fluoroquinolones) in wastewater, but the research on fluoride removal in semiconductor wastewater is limited (Moneta et al., 2023; Orimolade et al., 2023). Therefore, it deemed that up-to-date review of the semiconductor wastewater treatment process will suggest the future trend.

## 3. Overview of research status in semiconductor wastewater treatment processes

### 3.1. Physical treatment

Physical processes can be classified into technologies such as adsorption, coagulation, flocculation, aggregation, precipitation, flotation, air stripping, membrane separation or combination with electric and magnetic force based on physics such as gravity, electric, magnetic or fluid dynamics (Table S2).

Adsorption is technologically simple (simple equipment), adaptable to many treatments and widely used to target contaminants with generally lower cost (Crini and Lichfouse, 2019). However, a process of the study is nondestructive and non-selective methods. Also, rapid saturation and clogging of the reactors of adsorption are the one of disadvantages. Yang et al. investigated the effects of calcined alkaline residue to remove phosphate of wastewater from etching. It showed that phosphate adsorption rate reached 99% within 75 min of equilibrium. The maximum phosphate adsorption capacity was 139 mg/g of adsorbent process (Yan et al., 2014). Hu et al. applied micro-bubble floatation with dodecyl dimethyl betaine ( $BS-12, C_{12}H_{25}N^+(CH_3)_2CH_2COO^-$ ) as adsorbent to adsorb and aggregate silica nanoparticles (SNPs). The

**Table 5**

Electrical and electronic components industry of USA (EPA) (EPA, 2020).

INDUSTRY	TREATMENT_TECH_DESCRIPTION	Waste-water Use	Media Type	pH Range	Chemical Addition	Scale
Electrical and electronic components	The effluent from the inorganic wastewater treatment process (lime addition for the removal of fluoride and phosphate) was used as the influent of this pilot test. The influent was fed to the seed activation tank and completely mixed with the activated calcite seeds and a sodium carbonate solution (soda ash) to precipitate calcium in the form of calcite (a calcium carbonate mineral). The calcite seed activation was achieved using the controlled hydrodynamic cavitation (CHC) unit. The wastewater then flowed into the clarifier equipped with a scrapper, where the settled calcite slurry was then returned to the seed activation tank for activation of the calcites.				Sodium carbonate (soda ash) dose rate: 940 mg/L (based on influent flowrate)	Pilot
Electrical and electronic components	The high- and low-concentration printed circuit board (PCB) industrial wastewaters were introduced to the electrochemical-coagulation (EC) reactor alternatively every 20 min over the course of the 80-min operation time. The EC reactor was constructed from polyvinyl chloride (PVC) (capacity = 12 L) with aluminum electrodes (surface area = 0.3025 m <sup>2</sup> ) that are arranged in a monopolar configuration. The EC reactor also included a programmable logic controller (PLC) that was developed in-house to provide the optimal current signal according to the pre-measured conductivity value identified during the lab-scale tests.			5.8–5.8		Pilot
Electrical and electronic components	This system treats the liquid organic wastes (LOW) waste stream from the semiconductor manufacturing facility and employs activated sludge and advanced oxidation processes. The system begins with a pre-anoxic treatment for degradation of organics, followed by treatment in an aerobic reactor for TMAH breakdown into ammonia. Then the waste stream goes through a clarifier followed by advanced oxidation processes (AOPs) where ammonia is oxidized to nitrate and finally a post anoxic treatment to denitrify the nitrate produced during AOPs followed by a clarifier.			6.9–7.1	Concentrated HCl for pH adjustment. The influent was blended with essential micro/macro nutrients (metals, alkalinity, and phosphorus) for proper microbial metabolism.	Pilot
Electrical and electronic components	TMAH wastewater undergoes aerobic activated sludge treatment for TMAH breakdown into ammonia, after which it goes through a clarifier and then undergoes AOPs to oxidize ammonia to nitrate. After this step the nitrate is denitrified via anoxic activated sludge and finally goes through another clarifier.			6.9–7.1	Concentrated HCl for pH adjustment. The influent was blended with essential micro/macro nutrients (metals, alkalinity, and phosphorus) for proper microbial metabolism.	Pilot
Electrical and electronic components	Wastewater was pumped via peristaltic pump into a PVC column packed with 150 polyester fiber balls for filtration. The water then flowed through a spiral-wound ultrafiltration membrane cartridge. To minimize the fouling of the UF membrane, intense aeration, intermittent effluent discharge (15 min ON/1 min OFF), and continuous NaOCl (12%) and citric acid (50%) dosing was used. The water then flowed through a spiral-wound polyamide reverse osmosis (RO) membrane (LFC1 4040). Each unit consisted of a high-pressure feed pump, pressure vessel, and a recirculation loop with a pressure gage and a flow meter. This system was operated in continuous-feed mode with a recycle of concentrate flow to maintain the minimum concentrate flow requirements. Flows and pressures were controlled by adjusting the feed, recycle, concentrate, and permeate valves.	Recycled /Reused	Filtration: polyester fiber balls; UF: spiral-wound polyether sulfone membrane; RO: spiral-wound polyamide membrane	7.4–7.4	To minimize UF membrane fouling, continuous NaOCl (12%) and citric acid (50%) dosing was used (in addition to other processes).	Pilot
Electrical and electronic components	An existing wastewater treatment plant was retrofitted by adding a struvite precipitation process. After the existing equalization tank, water entered the chemical mix tank where MgCl <sub>2</sub> ·H <sub>2</sub> O as an alternate source of magnesium ion was added to reach at 1:1:1 in the molar ratio of NH <sub>4</sub> <sup>+</sup> : Mg: PO <sub>4</sub> <sup>3-</sup> for struvite formation	Unknown		around 9	Chemical mix tank: MgCl <sub>2</sub> ·H <sub>2</sub> O as magnesium source (at a 1:1:1 M ratio), 5N NaOH added continuously to adjust pH to 9. CaCO <sub>3</sub> was added to final settler to precipitate fluoride.	Full

(continued on next page)

**Table 5 (continued)**

INDUSTRY	TREATMENT_TECH_DESCRIPTION	Waste-water Use	Media Type	pH Range	Chemical Addition	Scale
<p>and then, the pH adjustment of the wastewater to 9 was achieved by the continuous addition of 5N NaOH. The liquid stream then moved to the struvite reaction tank where struvite was formed. The solution was allowed to settle in the intermediate settler. The settled sludge was recycled to the chemical mixing tank in order to provide seeding material. The water then continued into the rest of the existing system, which included a fluoride removal tank, where fluoride was precipitated with <math>\text{CaF}_2</math> by adding <math>\text{CaCO}_3</math>.</p>						

**Table 6**

Water recycling and usage efficiency of TSMC (TSMC, 2021).

			2017	2018	2019	2020	2021
Wastewater	Total	million metric tons	29.4	33.7	38.67	46.69	49.2
	Increase rate of year-on-year	%		12.76	12.85	17.18	5.1
	Wastewater qualities						
	$\text{NH}_4\text{-N}$	ppm	25.2	21	17.31	unknown	unknown
	Chemical oxygen demand (COD)	ppm	112.5	177.5	185.5	194	180
	Suspended solids (SS)	ppm	33.6	29.4	unknown	unknown	unknown
	TMAH	ppm	12.9	13.1	7.9	6.3	5.5
	Copper ion ( $\text{Cu}^{2+}$ )	ppm	0.22	0.18	0.09	0.07	0.07
Recycled wastewater	Total	million metric tons	103.4	129	133.6	173	186.3
	Increase rate of year-on-year	%		19.84	3.44	22.77	7.14
	Average process recycling rate	%	87.5	87.5	86.7	86.5	85.4

**Table 7**

Wastewater treatment and resource system of TSMC (TSMC, 2021).

38 Different Types of Liquid Classified	13 Types of Wastewater Treatment	9 Recycling Systems	9 Products Recycled
Hydrofluoric (HF) Acid Wastewater	<ul style="list-style-type: none"> <li>Ozone-containing HF acid wastewater</li> <li>Manufacturing process scrubbing water</li> </ul>	<ul style="list-style-type: none"> <li>Chemical coagulation treatment of HF acid</li> <li>HF (including ammonia) treatment</li> <li>Biological treatment of HF acid wastewater</li> </ul>	<ul style="list-style-type: none"> <li>Hydrofluoric (HF) acid wastewater</li> <li>Local scrubber wastewater recycling</li> </ul>
Acidic, Caustic, and Organic Wastewater	<ul style="list-style-type: none"> <li>Deionized water</li> <li>Ozone-containing deionized water</li> <li>Acidic wastewater (2 types)</li> <li>Ozone-containing acidic wastewater</li> <li>Caustic wastewater</li> <li>Ozone-containing Caustic wastewater</li> <li>TMAH wastewater</li> <li>Organic wastewater (2 types)</li> </ul>	<ul style="list-style-type: none"> <li>Acidic and caustic wastewater treatment</li> <li><math>\text{NH}_4\text{-N}</math> wastewater treatment and recycling</li> <li>TMAH liquid waste recycling</li> <li>Biological treatment system for organic wastewater</li> </ul>	<ul style="list-style-type: none"> <li>Acidic wastewater recycling</li> <li>Ozone-containing acidic wastewater recycling</li> <li>Caustic wastewater recycling</li> <li>Caustic wastewater upcycling</li> <li>Organic wastewater recycling</li> </ul>
CMP Wastewater	<ul style="list-style-type: none"> <li>Electroplating wastewater</li> <li>Copper-containing CMP wastewater</li> <li>Cobalt-containing CMP wastewater</li> <li>CMP wastewater (2 types)</li> </ul>	<ul style="list-style-type: none"> <li>CMP wastewater treatment</li> <li>Copper-containing CMP wastewater treatment</li> <li>Cobalt-containing CMP wastewater treatment</li> </ul>	<ul style="list-style-type: none"> <li>Chemical coagulation recycling of CMP wastewater</li> <li>Copper-containing CMP wastewater upcycling</li> </ul>
High-concentration Liquid Waste	<ul style="list-style-type: none"> <li>Waste sulfuric acid</li> <li>Waste copper sulfate</li> <li>Cobalt-containing liquid</li> <li>Other wastewater (16 types)</li> </ul>	<ul style="list-style-type: none"> <li>Waste-<math>\text{H}_2\text{SO}_4</math> reclamation</li> <li>Copper-containing electroplating wastewater recycling</li> <li>Cobalt-containing electroplating wastewater recycling</li> </ul>	<ul style="list-style-type: none"> <li>Sulfuric acid</li> <li>Copper bar</li> <li>Cobalt bar</li> </ul>

enrichment ratio and recovery percentage of SNPs were around 30.4% and 90.8% respectively (Hu et al., 2018).

Coagulation, flocculation, aggregation and precipitation are physical treatments with simplicity of process and cost-effectiveness, required the injection of coagulants, flocculants and acid chemicals which are usually unable to reuse though. These processes were known that these technics can reduce turbidity but also organic contaminants, fluoride

etc. Lin and Yang employed poly aluminum chloride (PAC) and polymer (coagulant aid) as coagulants to treat CMP wastewater. The coagulation reduced 99.92% of turbidity at the optimum dose of PAC and polymer. However, for rapid flocculation process, the adjustment of pH of final wastewater was recommended to naturalize (Lin and Yang, 2004). Hence, Won et al. used lime coagulant to reduce fluoride concentration and showed 95.5% of removal rate. However, the fluoride concentration

**Table 8**

Water quality items and limits of discharge from the wafer and semiconductor manufacturing industry in Taiwan (EPA, 2017).

Item		Limit	Remarks
Water temperature	Discharge into non-marine surface water bodies		
	Direct discharge to the ocean		Lower than 38 °C (for May to September) Lower than 35 °C (for October to next April) Water temperature at discharge point ≤42 °C; temperature difference of surface water 500m from discharge point ≤4 °C
Hydrogen ion concentration index		6.0–9.0	
Villiaumite		15	
Nitrite nitrogen		50	
Ammonia nitrogen	Discharged into tap water quality and volume protection area Discharged into places outside tap water quality and volume protection area	10 30	Constructed, under construction or tendering procedures completed before Dec. 1, 2011 Tendering procedures not completed before Dec. 1, 2011
Orthophosphate (calculated based on trivalent phosphate ion)	Discharged into tap water quality and volume protection area	4	
Phenols		1	
Anion surfactant		10	
Cyanide		1	
Grease (Hexane extracts)		10	
Dissolved iron		10	
Dissolved manganese		10	
Cadmium	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	0.03	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.02	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.02	
Lead	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	1	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.5	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.5	
Total chromium	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	2	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	1.5	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	1.5	
Hexavalent chromium	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	0.5	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.35	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.35	
Copper	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	3	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	1.5	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	1.5	
Zinc	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	5	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	3.5	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	3.5	
Nickel	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	1	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.7	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.7	
Selenium	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	0.5	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.35	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.35	
Arsenic	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	0.5	
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.35	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.35	
Tin	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	2	In effect from Jan. 1, 2021.
	Constructed, under construction or tendering procedures completed before Dec. 25, 2017 with an approved discharge volume more than 500m <sup>3</sup> /day	0.35	
	Tendering procedures not completed before Dec. 25, 2017	0.35	
Total mercury		1	
Silver		0.005	
		0.5	

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**Table 8** (continued)

Item		Limit	Remarks
Boron	Discharged into tap water quality and volume protection area	1	
	Discharged into places outside tap water quality and volume protection area	5	
Molybdenum	Constructed, under construction or tendering procedures completed before Dec. 25, 2017	0.6	In effect from Jan. 1, 2021.
	Tendering procedures not completed before Dec. 25, 2017	0.6	
Sulfide		1	
Chemical oxygen demand		100	
Suspended solids		30	
Total toxic organics		1.37	
N-methylpyrrolidone	Approved discharge volume more than 10,000 m <sup>3</sup> per day except for those only engaging in polishing, cutting, testing or packaging.	1	In effect from Jan. 1, 2021.
2-Methoxy-1- propanol		0.1	
Dimethylacetamide		0.1	
Cobalt		1	
Antimony		1	

was 15 mg/L after coagulation, it means that additional fluoride treatment was needed to satisfy water quality from US EPA (Won et al., 2012). Alberto Lopez-Maldonado et al. improved the efficiency of coagulation-flocculation by zeta potential measurement. Coagulant and flocculant were Polydadmac (OPTIFLOC C-1008) and Trident 27,506 which were nonstoichiometric polyelectrolyte complexes (NIPECs) (Alberto Lopez-Maldonado et al., 2014).

The ballasted flocculation process was employed to remove fluoride of semiconductor wastewater. Calcium hydroxide was used as coagulant forming calcium fluoride in coagulation sedimentation. The average removal rate of fluoride and turbidity were 96.3% and 98.1% respectively under the optimum conditions (Wang, B.-y. et al., 2013).

The combined precipitation process with flotation process was demonstrated to reduce the high concentrated fluoride wastewater from semiconductor industry. When the molar concentration ratio of  $[Ca_2^+]/[F]$  was greater than 1.0, the removal rate of fluoride was greater than 97% (Huang and Liu, 1999). Chuang et al. conducted similar study with Huang and Liu (Chuang et al., 2002). Furthermore, several studies contributed to decrease  $NH_4^+$ -N and  $PO_4^{3-}$ -P by precipitation process. Ryu et al. applied struvite precipitation on the treatment using magnesium chloride ( $MgCl_2 \cdot 6H_2O$ ) as precipitation reagent. The performance of  $PO_4^{3-}$  removal rate (83%) was higher than  $NH_4^+$  (78%) (Ryu et al., 2008). Warmadewanthi and Liu also conducted recovery of phosphate and ammonium as struvite from semiconductor wastewater with same precipitation reagent. It showed 92.5% of  $PO_4^{3-}$  removal rate, while removal rate of  $NH_4^+$  was much lower (33.5%) at the suitable conditions (Warmadewanthi and Liu, 2009). The hybrid precipitation microfiltration (MF) process was introduced to not only remove but also recover phosphate and fluoride in thin-film transistor liquid crystal display (TFT-LCD) wastewater. Calcium chloride which has high solubility and produces less sludge than lime was employed as a chemical. The removal rates of  $PO_4$  and F were 96% and 54% at  $Ca:PO_4:F = 2.5:1.0:0.7$  and pH 8.5 by the hybrid process (Lu and Liu, 2010). Huang et al. investigated the precipitation process by magnesium chloride ( $MgCl_2 \cdot 6H_2O$ ) to remove phosphate and fluoride but also the total ammonia nitrogen (TAN). In pilot-scale, it performed 97% of phosphate removal rate and 91% of fluoride removal rate, while TAN was relatively less (58%) by a two-stage precipitation process (Huang et al., 2017) (Fig. 8) (Fig. 9). Meanwhile, to remove heavy metals such as copper, nickel, and tin of wastewater which was generated the production of PCBs, sodium trithiocarbonate ( $CH_4Na_2O_2S_3$ ) precipitate was tested (Thomas et al., 2018).

For semiconductor wastewater treatment, the combination of electrical or magnetic forces with traditional physical treatment was attempted by several researchers. The researchers attempted to minimize the amount of discharged sludgy by chemical coagulation, flocculation and aggregation using the combined process. Most of researcher applied Al-anode and Fe-cathode as the electrode pairs which showed the best performance to reduce the concentration of COD,

turbidity, F, Cu,  $NH_4^+$ , sodium dodecyl sulfate (SDS) (Aoudj et al., 2013, 2015, 2016, 2017; Chou et al., 2009, 2010; Chung et al., 2020b; Drouiche et al., 2008; Hu et al., 2005; Lai and Lin, 2003, 2004, 2006; Liu et al., 2016; Wang et al., 2009). They revealed that the current density, voltage power and pH affected to the performances. Lai & Lin tried to reduce COD, copper and turbidity of semiconductor wastewater and figured out the effects of the type of electrode pair. Al-anode/Fe-cathode pair showed the best performance in electro-coagulation (Lai and Lin, 2003). Also, they optimized the best voltage power condition with NaCl electrolyte dosage of yielded. Additionally, they applied the complex models to the sludge settling characteristics (Lai and Lin, 2004). Hu et al. evaluated the effects of cationic and anionic surfactants to treat CMP wastewater and figured out that the addition of cation in CMP wastewater can reduce the sludge volume and the flotation/sedimentation time in electro-coagulation-flotation (ECF) process (Hu et al., 2005). Lai and Lin optimized the conditions for continuous treatment for CMP wastewater and reduce the sludge volume by freeze-thaw method (Lai and Lin, 2006). Drouiche et al. employed electro-coagulation for removal fluoride from semiconductor wastewater with characterizing the sludge and calculating the operation cost (Drouiche et al., 2008). Wang et al. identified the relationships with the zeta potential of the silica particles, solution turbidity, and the corresponding mean particle size of three operating stages (lag, reactive, and stabilizing) for removal of silica and turbidity of oxide-CMP wastewater (Wang et al., 2009). Similarly, Chou et al. identified the relationships which were same with Wang et al. to remove COD and turbidity by iron electro-coagulation (Chou et al., 2009). Chou et al. used the iron hydroxides which were generated during electro-coagulation process to remove COD of oxide-CMP wastewater by adsorption with iron hydroxides (Chou et al., 2010). Aoudj et al. targeted the removal of fluoride of semiconductor wastewater after neutralizing step. They figured out that Al-anode/Al-cathode pair for that wastewater showed the best performance and the current density was the most important factor to control the performance (Aoudj et al., 2013). Aoudj et al. developed the electro-coagulation-flotation process for semiconductor wastewater treatment. They investigated the effects of electrode characteristic and arrangement, current intensity, initial pH, initial concentration, type and concentration of supporting electrolytes (Aoudj et al., 2015). Liu et al. especially investigated the effects of aeration for removing particles in electro-coagulation for semiconductor wastewater treatment (Liu et al., 2016). Aoudj et al. suggested the combined electro-coagulation and electro-flotation process with hybrid anode composed of iron and aluminum plates to remove SDS, fluoride and ammonia from semiconductor wastewater showing relatively less performance for ammonia removal (Aoudj et al., 2017). Chin et al. and Shen et al. conducted the removal of turbidity by magnetic seeding aggregation with chemical co-precipitation of  $FeCl_2$  and  $FeCl_3$  (Chin et al., 2006; Shen et al., 2013). However, additional studies are needed to scale up.

Membrane separation process is one of the advanced water treatment

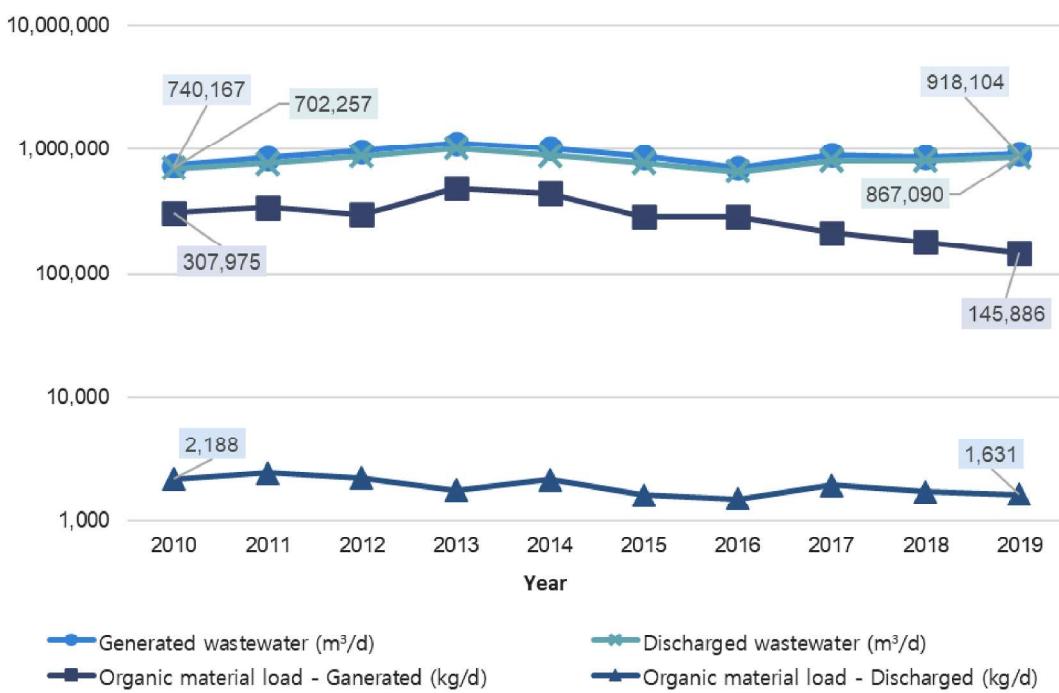


Fig. 6. Status of semiconductor manufacturing industrial wastewater of the Republic of Korea (Korea, 2022).

**Table 9**  
Status of specific water pollutants emission in Samsung Electronics in 2019.

Place	Specific substance harmful to water quality	Discharged (kg/yr)	Type of treatment
Giheung campus	Hexavalent chromium compounds	0	Water pollution prevention facilities
	Copper and its compounds	192.2	
	Lead and its compounds	0	
	Diethylhexyl phthalate (DEHP)	0	
	Dichloromethane	0	
	Bromoform	0	
	Arsenic and its compounds	0	
	Selenium and its compounds	0	
	Styrene	0	
	Cyanide	0	
Hwaseong campus	Cadmium and its compounds	0	
	Chloroform	0	
	Formaldehyde	0	
	Hexavalent chromium compounds	0	Individual treatment (then direct discharge)
	Copper and its compounds	136.23	
	Lead and its compounds	0	
	Dichloromethane	0	
	Bromoform	0	
	Arsenic and its compounds	0	
	Selenium and its compounds	16.14	
	Cyanide	0	
	Cadmium and its compounds	0	
	Chloroform	0	
	Formaldehyde	0	

technologies and has been employed to treat high-concentrated industrial wastewater. The membrane is to separate certain species in a fluid by a selective barrier. This membrane process requires small space to be occupied producing high quality of permeate water without any chemical additives. However, trade-off between permeability and salt rejection and maintenance problems to solve the fouling and pore blocking still leave a challenge. Furthermore, the operation cost for high pressure

and initial installation cost are obstacles with used membrane disposal problems (Obotey Ezugbe and Rathilal, 2020). Many researcher proved the possibility of membrane to treat semiconductor wastewater showing good performance of removal rate of contaminants. Furthermore, membrane is the dominant treatment process for semiconductor wastewater among all process, because membrane processes are easily applied for wastewater reuse and reclaim. Lin and Yang applied RO membrane to remove COD after coagulation process (Lin and Yang, 2004). RO could degrade over 95% of COD from 221 mg/L of initial concentration. It is tried to reject perfluoro octane sulfonate (PFOS) employing four commercial thin-film composite (TFC) polyamide (PA) RO membranes (ESPA3, LFC3 (Hydranautics, California, USA), BW30 (Dow FilmTec, Michigan, USA), SG (GE Osmonics Minnesota, USA)) (Tang et al., 2006) (Fig. 10). These membranes performed over 90% of PFOS rejection (BW30 > LFC3 > SG > SPA3), while SPA3 membrane showed the highest flux. Huang et al. treated semiconductor wastewater by coagulation/flocculation, fiber ball filtration (FF), UF, RO in order (Huang et al., 2011). The FF was installed as a pretreatment unit (first stage) for particle removal. UF (spiral wound type module, polyether sulfone) and RO membrane (polyamide) were applied in the second and third treatment stages. Removal rate of SS reached 95.8% by FF with 88 m/h of filtration velocity. UF process removed over 98% of turbidity (less than 0.6 NTU). RO process reduced 97.3% of conductivity and removed 99.8% of turbidity and 97.5% of SS. Won et al. evaluated reuse process consisting of coagulation and RO membrane process by treating fluoride-concentrated wastewater based on pilot scale (Won et al., 2012) (Fig. 11). Spiral wound and disc tube type of RO membrane were evaluated, spiral wound type of RO membrane showed 97.3% of reduction of conductivity and excellent chemical cleaning cycles. An et al. employed three different reverse osmosis (RO) membranes (ESPA2-LD, RE4040-BE, and TMG10D) to treat semiconductor wastewater (An et al., 2022). A ceramic membrane with ozone injection was evaluated as pre-treatment of RO process. The ceramic membrane removed SiO<sub>2</sub> reducing turbidity to below 0.4 NTU and conductivity wasn't reduced regardless of the present of ozone. TMG10D RO membrane exhibited the highest permeability, removal rate of total organic carbon (TOC) and power consumption, whereas EPA2-LP showed the highest salt rejection. It was concluded that EPA2-LP was suitable

**Table 10**Status of water usage, wastewater discharge, water reuse in Samsung Electronics ([Samsung, 2022](#)).

		Unit	2015	2016	2017	2018	2019	2020	2021
Water usage	Industrial water	Million ton	58.44	62.99	67.71	78.84	81.98	75.24	0.00
	Municipal water (surface water)	Million ton	32.83	40.15	51.92	54.43	51.84	66.47	157.69
	Underground water	Million ton	1.14	1.12	0.99	0.96	0.66	0.59	5.97
	Total	Million ton	92.41	104.25	120.62	134.23	134.48	142.29	163.66
Wastewater	Discharge	Million ton	72.58	81.72	95.92	107.70	108.46	109.20	130.96
	Increase rate of year-on-year	%		12.58	17.38	12.28	0.71	0.68	19.92
	Total	Million ton	46.20	48.60	56.15	62.37	68.56	70.18	93.95
Reused water <sup>1)</sup>	Increase rate of year-on-year	%		5.20	15.54	11.07	9.91	2.37	33.87
	Rate	%	50	46.6	46.6	57.9	63.2	64.3	71.7
	Supply	Million ton	34.40	37.77	44.58	52.61	55.04	57.23	61.99
Ultra-pure water reused	Increase supply rate of year-on-year	%		9.81	18.04	18.00	4.62	3.97	8.32
	Recovery	Million ton	14.63	15.53	16.36	17.51	15.01	19.69	22.54
	Recovery rate	%	42.5	41.1	36.7	33.3	27.3	34.4	36.4

**Table 11**Status of water pollutants emissions in Samsung Electronics ([Samsung, 2022](#)).

Unit (ton)	2015	2016	2017	2018	2019	2020	2021
Total water pollutants	1936	3055	3190	3797	2128	2162	2098
COD	970	1639	1771	2407	1047	1033	906
BOD	277	520	521	392	415	315	266
SS	436	598	497	649	315	377	393
F	240	253	386	337	342	428	520
Heavy metals	13	45	15	12	9	9	13

**Table 12**Status of specific water pollutants emission in SK hynix in 2019 ([hynix, 2022](#)).

Place	Specific substance harmful to water quality	Discharged (kg/yr)	Type of treatment
Cheongju site	Hexavalent chromium compounds	0	Treatment via water pollution prevention facilities,
	Copper and its compounds	42.55	individual treatment
	Lead and its compounds	0	(then direct discharge)
	Dichloromethane	0	or commissioned treatment
	Arsenic and its compounds	0	
	Selenium and its compounds	1.9	
	Mercury and its compounds	0	
	Cyanide	0	
	Acrylonitrile	0	
	Cadmium and its compounds	0	
	Chloroform	0	
	Tetrachlorethylene	0	
	Phenol	10.4	
	Formaldehyde	444.01	

membrane for semiconductor wastewater in this paper. Teow et al. studied the performance of commercial UF ceramic and polymeric membranes to treat three types of semiconductor wastewaters; diluted back grinding wastewater (DBGW), diluted chemical mechanical polishing wastewater (DCMPW), and collection tank wastewater (CTW) ([Teow et al., 2022](#)). Even though three different membrane showed satisfactory results, a ceramic membrane was better than two polymeric membranes in terms of water qualities and permeability. Depending of each wastewater qualities, the cleaning methods by suitable chemicals were study to recover the flux of ceramic membranes. Eng et al. tried to reuse and reclaim two wastewaters from semiconductor industries; one was treated by continuous electro-deionization (CEDI) and the other was local scrubber water (LSW) ([Eng et al., 2019](#)). Due to water qualities of wastewater treated by CEDI were relatively good, be used to supply as ultrapure water. The reclaim system for wastewater treated by CEDI consisted of boron selective resin (BSR), activated carbon filter (ACF) sequentially, and removed up to 83.33% of TOC and 86.49% of boron. To reuse local scrubber water (LSW), controlled decomposition and oxidation (CDO) reclaim system consisted of the 1st pH adjustment, ACF, UF, RO process and the 2nd pH adjustment sequentially and supplied to cooling tower. It performed that TOC removal rate reached 79.59%.

Su et al. assessed the performance of combined UF and electrodialysis (ED) processes to treat and reclaim Cu-CMP wastewater ([Su et al., 2014](#)). This study assesses the efficiency of a combining UF and ED processes to treat and reclaim wastewater, a discharge of the copper chemical mechanical planarization (Cu-CMP) process typical among semiconductor manufacturing procedures. Cationic exchange membranes (CEMs) in the ED reactor removed  $\text{Cu}^{2+}$  as a pre-treatment but also recover  $\text{Cu}^{2+}$ . Cha et al. demonstrated the fabrication graphene oxide (GO) assembled ceramic NF membranes to control fouling ([Cha et al., 2022](#)). The GO-ceramic NF membranes were fabricated following method that layer-by-layer (LBL) assembly of GO and polyethyleneimine (PEI) on a ceramic UF substrate. It showed removal rate of polyethylene

**Table 13**

Status of water usage, wastewater discharge, water reuse in SK hynix, 2022.

		Unit	2015	2016	2017	2018	2019	2020	2021
Water intake	Municipal water	Million ton	40.11	79.49	43.29	43.21	53.69	57.43	64.40
	Surface water	Million ton	16.01	5.48	30.53	33.72	30.95	30.96	32.34
	Gray water	Million ton	0.00	0.00	0.00	3.91	5.92	7.32	7.30
	Total	Million ton	56.12	84.97	73.82	80.83	90.56	95.72	104.04
Water usage	Consumption	Million ton	11.08	29.36	11.61	13.85	14.71	12.79	16.44
	Reuse	Million ton	17.49	19.10	21.72	27.29	32.62	40.49	47.57
	Reuse rate	%	28	26	26	29	30	33	35
Wastewater	Increase reuse rate of year-on-year	%		9	14	26	20	24	17
	Discharge	Million tons	45.04	55.61	62.21	66.98	75.84	82.92	87.60
	Increase rate of year-on-year	%		23	12	8	13	9	6
Ultrapure water	Usage	Million tons		25.20	27.44	30.36	34.30	37.37	40.18
	Increase rate of year-on-year	%		9	11	13	9	8	

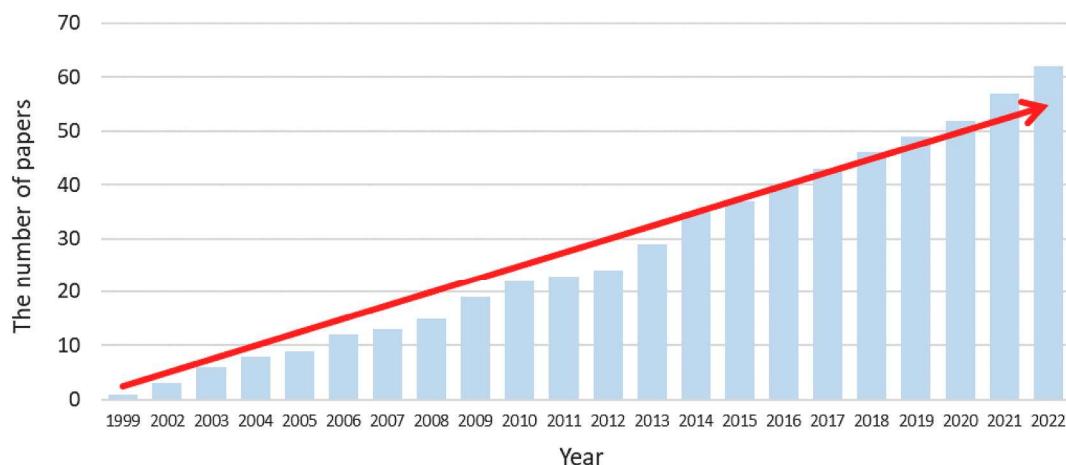
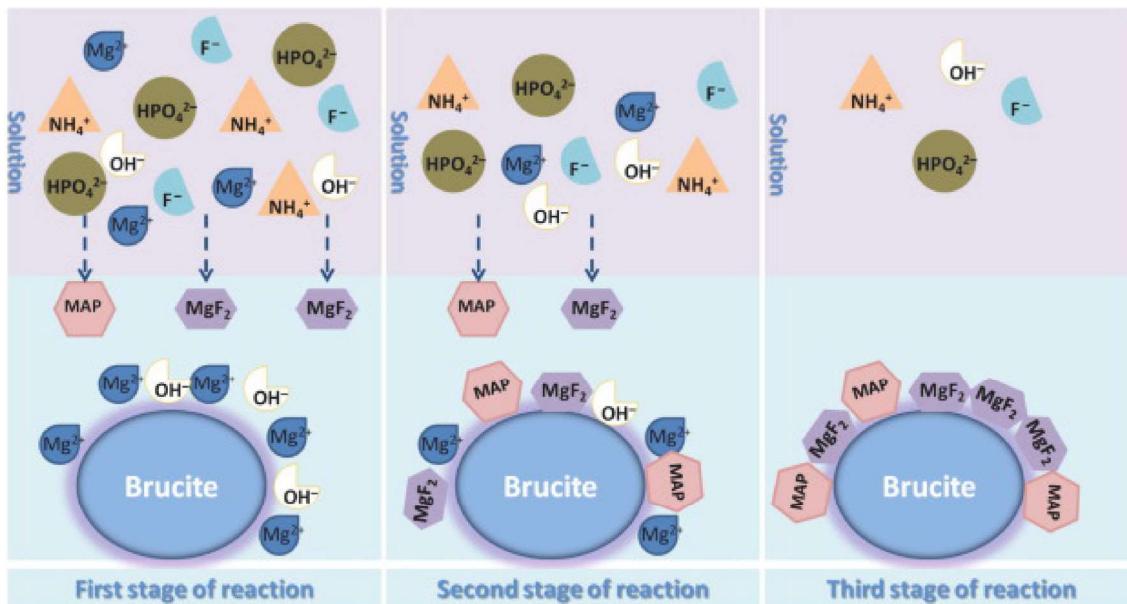
**Table 14**

Status of water pollutants emissions in SK hynix, 2022.

Unit (ton)	2015	2016	2017	2018	2019	2020	2021
COD	180.99	211.07	203.8	204.8	235.6	234.2	203.9
BOD	206.13	154.02	166.38	183.6	172.5	140.7	116.6
Total nitrogen (TN)	696.37	1670.69	944.47				
F	141.36	152.87	162.61				
Total phosphorus (TP)				2	1.6	1.6	1
SS		3.31		117.1	106	113.8	97.1

It was reported that the TN and F from 2018 were not revealed but maintained internally.

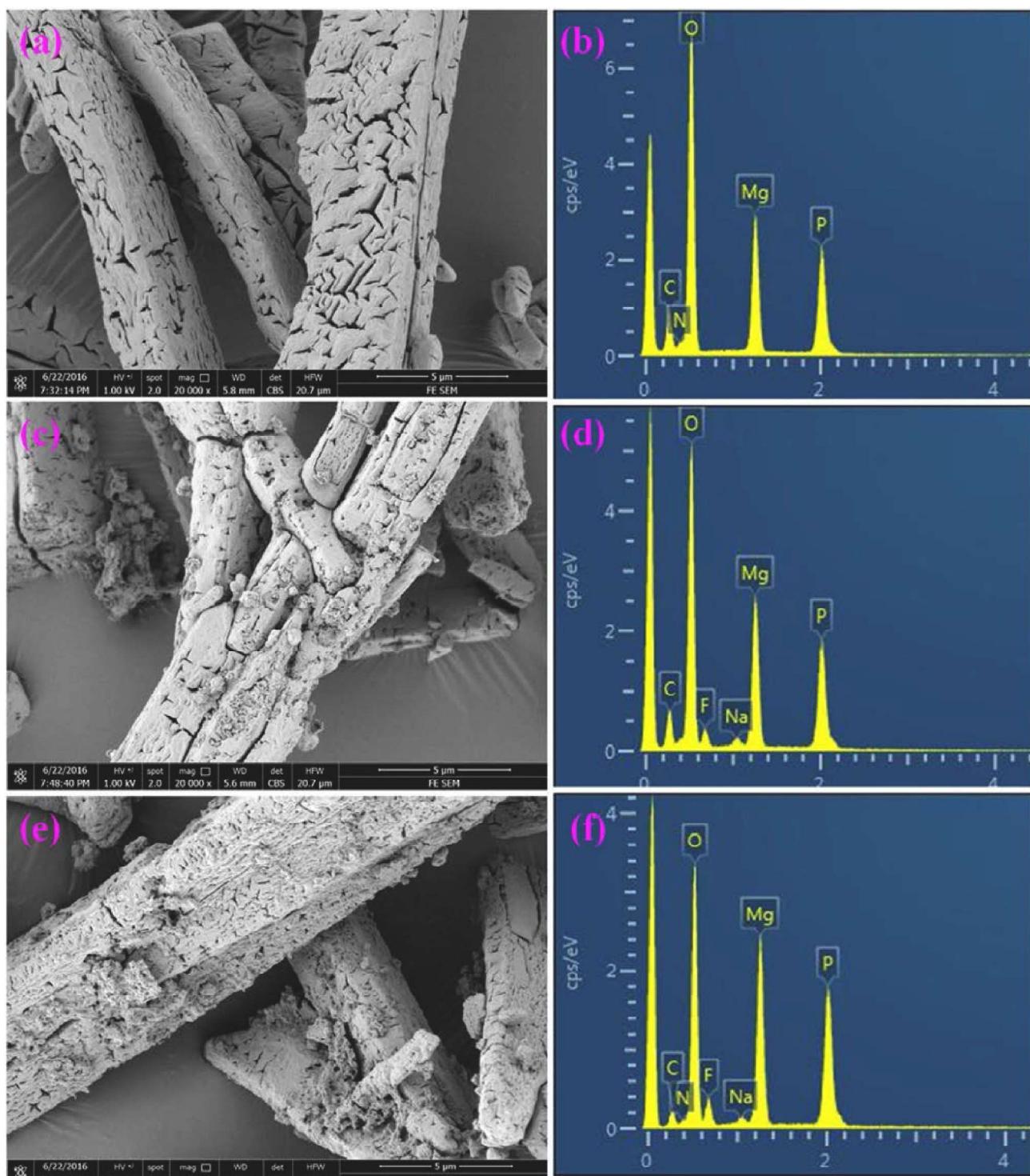
T-P was reported since 2018, SS was reported since 2017.

**Fig. 7.** Cumulative number of publications of semiconductor wastewater treatment process.**Fig. 8.** Diagrammatic sketch of simultaneously removing the phosphate, ammonia nitrogen and fluoride with brucite (adapted from (Huang et al., 2017)).

glycol (PEG) solutions of 600 g/mol and 1000 g/mol exceeded 83.9% and 90.1% each.

Wang, Y. et al. introduced the TMAH recovery process from developer wastewater using ion exchange membrane (Wang, Y. et al., 2013). Anion exchange membrane (AEM, JAM-II-07, Beijing TingRun Membrane, China) and cation exchange membrane (CEM, JCM-II-07, Beijing TingRun Membrane, China) were used for electrodialysis process. However, Ohanessian et al. introduced simulations using ultrafiltration

to nanoparticles (NPs) of CMP wastewater (Ohanessian et al., 2020). This study is focused on a performance assessment of membrane distillation (MD) technology for CMP wastewater treatment. Imtisal e et al. estimated the feasibility of air gap membrane distillation (AGMD) module to treat CMP wastewater (Imtisal e et al., 2020). Except fluoride, nitrate, magnesium, gallium, nickel, strontium, tungsten and tantalum, removal rates of components were over 90%. Eng et al. suggested the treatment removing TOC, boron to reclaim the wastewater as ultrapure

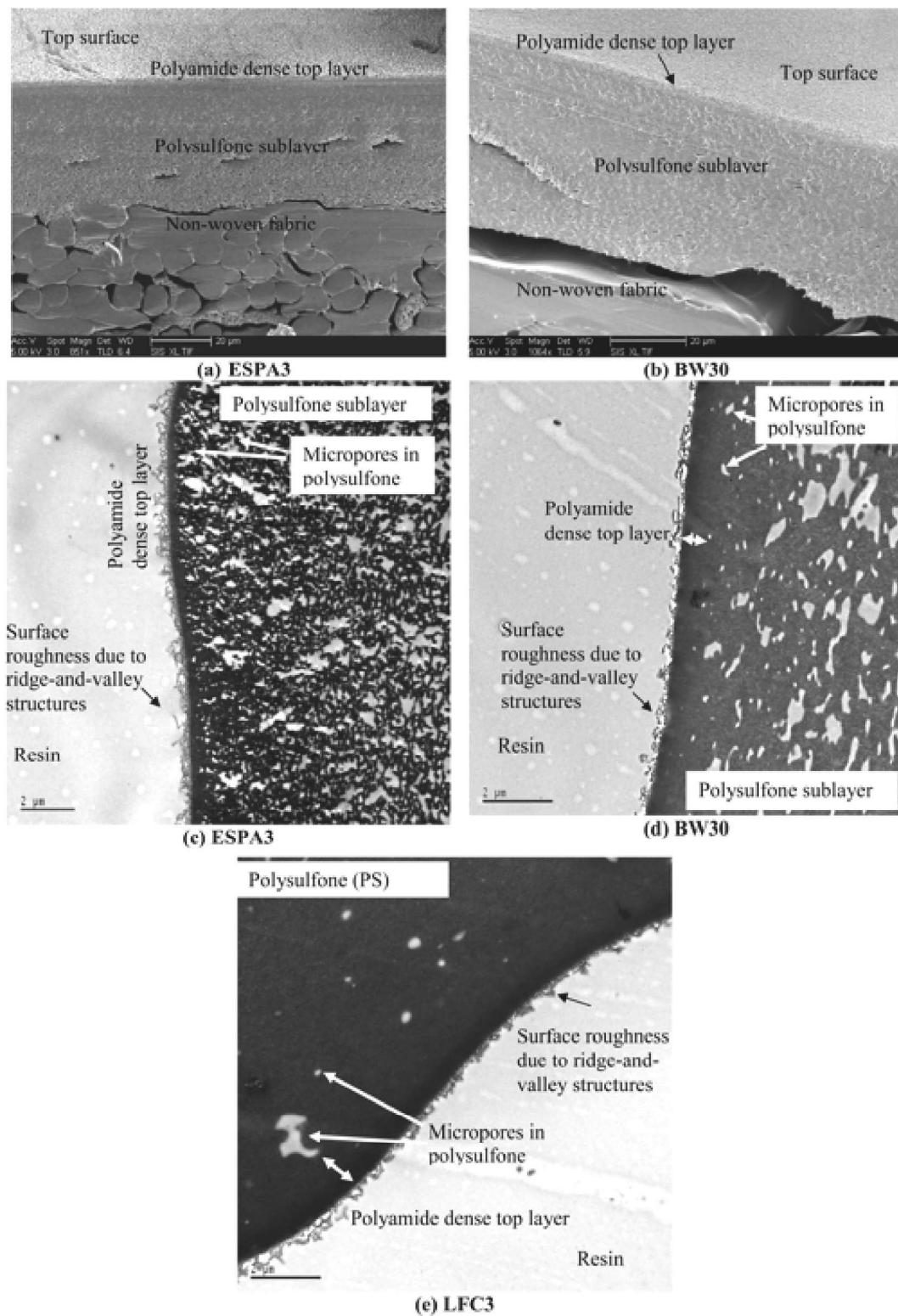


**Fig. 9.** SEM-EDX characterization results of the struvite crystals obtained at pH 9.5 and different fluoride concentrations: (a) and (b) are the SEM micrograph and the EDX pattern at a fluoride concentration of 0 mg/L, respectively; (c) and (d) are the SEM micrograph and the EDX pattern at a fluoride concentration of 600 mg/L, respectively; (e) and (f) are the SEM micrograph and the EDX pattern at a fluoride concentration of 1500 mg/L, respectively.(adapted from (Huang et al., 2017)).

water (UPW) (Eng et al., 2019). Lu and Liu employed hybrid precipitation–MF process to treat TFT-LCD wastewater (Lu and Liu, 2010). MF was affected by precipitation conditions causing significant fouling at pH 8.5–10.5.

Flootation is a separation process using gas bubbles as transport medium to efficiently remove small particles, organic compounds, volatile compounds and etc. The severe disadvantage of the process is a cost for initial installation or operation who high energy. The dispersed air

fлотation process combined with the precipitation was investigated to reject the fluoride and the  $\text{CaF}_2$  removal rate of the flotation was affected by the types and concentrations of surfactants as well as the types of calcium ions. It was reported that SDS show the best performance compared with sodium oleate (SOL) and dodecylammonium chloride (DAC) as surfactants and increased the removal rate of  $\text{CaF}_2$  (Huang and Liu, 1999). On the other hand, the dissolved air flotation (DAF) was investigated to remove fluoride in semiconductor wastewater,



**Fig. 10.** SEM and TEM images of virgin membranes. (a) ESPA3 and (b) BW30 SEM images with 20  $\mu\text{m}$  scale bars. Both SEM samples were inclined at 45° to the mounting stage. (c) ESPA3, (d) BW30, and (e) LFC3 TEM images with 2  $\mu\text{m}$  scale bars (adapted from (Tang et al., 2006)).

It was reported that air stripping can eliminate the volatile compounds in liquid. The compounds are transferred by the gas (usually air) into the air, then separated from the liquid. The design and operation of air stripping are simple with taking small pace. However, the air pollution can be accrued by the process, so it caused another problem (Srinivasan et al., 2008). Air stripping treatment was tired to reject

volatile organic components such as IPA and resulted 90% of removal or more (Den et al., 2002; Lin and Jiang, 2003; Lin and Kiang, 2003).

Fluidized bed crystallizer (FBC) and fluidized-bed homogeneous granulation that deviate from the existing traditional process were attempted to introduced by Bayon et al., and Priambodo et al. beside (Bayon et al., 2021; Priambodo et al., 2017).

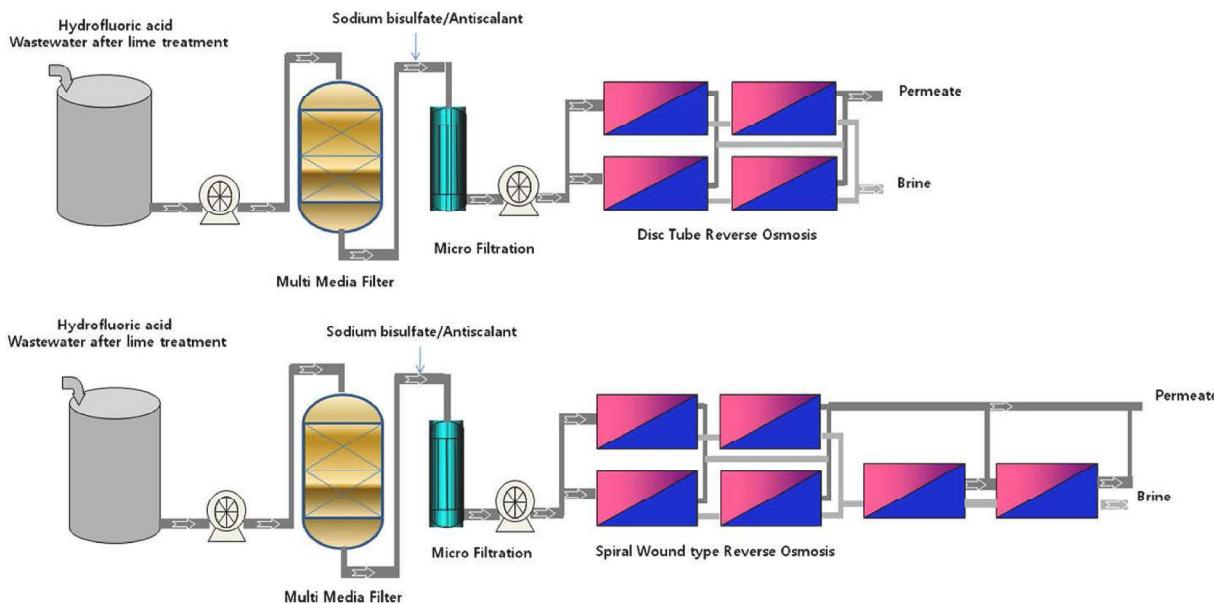


Fig. 11. Schematics of the pilot-scale plants (adapted from (Won et al., 2012)).

### 3.2. Chemical treatment

The chemical treatment processes use treatment processes which use chemical reactions or mechanisms and have traditionally been widely used in water treatment processes (Table S2). Coagulation, precipitation, membrane separation, crystallization, and modified processes are described in Chapter 3.1 for representative processes of chemical as well as physical treatment.

The chemical treatment processes are classified into AOPs including catalytic wet oxidation (CWAO), electrochemical oxidation (EO) and combination with zero-valent iron (ZVI) process, crystallization and physicochemical treatment process such as membrane process and chemisorption. AOPs is defined the chemical treatment by oxidants generating hydroxyl radicals which have powerful oxidation power (Glaze et al., 1987). Oxidants such as ozone,  $H_2O_2$  have been collaborated with Fenton, UV, etc. to maximize their performance. AOPs have been applied to remove organic compounds, non-biodegradable compounds, micropollutant with high reactivity and low selectivity of hydroxyl radicals. Furthermore, these processes react rapidly with target compounds without any sludge production. However, hydroxyl radicals are non-selective. Moreover, the generation of by-products, injection of oxidant reagents, installation of ozone off-gas treatment facility are hurdles to overcome. Crystallization is a separation technique which separates the solute crystallizes in liquid solution by creating solid struvite crystals. This process is recently studied to remove and recover phosphate, magnesium, calcium and heavy metals in wastewater at the same time. Nevertheless, drawbacks such as the complicated system and complex operation, high energy consumption and capital costs are challenges to broaden the applications in wastewater treatments (Lu et al., 2017; Suzuki et al., 2002). Main targets of AOPs to remove were COD (or TOC), organic compounds (propylene glycol methyl ether acetate (PGMEA), ethyl lactate (EL), TMAH, 1-methyl-2-pyrrolidone (NMP), IPA, phenol (PHL), methyl methacrylate (MMA), pyrazole and diglycolamine (DGA)), refractory TMAH etc.

Den et al. used two-stage of UV/ $H_2O_2$  process combining the biodegradation (Den et al., 2002). More than 95% concentration of TOC, PGMEA, EL, TMAH, IPA, and PHL were decreased, hence removal rate of NMP reached around 35% because of its stable ring structures. Moreover, it is considered to increase UV intensity to fully degrade NMP and its by-product (UV intensity was 15.6 mW/cm<sup>2</sup>). Li et al. conducted UV/ $H_2O_2$  process which exhibited 99.99% of pyrazole and DGA removal

performance (Li et al., 2021). On the other hand, removal rates of TOC by Fenton and UV/Fenton were less than 90%. Kim et al. demonstrated the degradation mechanism of TMAH under the different wavelengths of UV (265, 310 and 365 nm) (Kim et al., 2022) (Fig. 12). The chemicals degraded by the UV/ $H_2O_2$  were investigated trimethyl ammonium hydroxide (TriMAH), dimethylammonium hydroxide (DiMAH), mono-methyl ammonium hydroxide (MoMAH).

Ozone and ozone/UV process were employed to remove MMA and COD of semiconductor wastewater (Shang et al., 2007). Ozone/UV process was better than ozone process for reduction of MMA and COD (Kim et al., 2021).

In addition, A nano-ozone bubble was used to enhance the efficiency of the ozone/ $H_2O_2$  process for the degradation of TMAH containing semiconductor wastewater (Kim et al., 2021). Under the optimal conditions, TMAH was removed to 95 % of initial concentration, whereas TOC was decreased to 65% of initial concentration in 90 min. Meanwhile, integrated ceramic membrane with ozone which was injected through pore of ceramic membrane was tested for RO pre-treatment (An et al., 2022). It was concluded that the ozone reduced the flux of membrane, while the concentration of TOC was generally decreased but not effective.

In case of Fenton, researchers focused to reduce the COD of IPA containing semiconductor wastewater (Lin and Jiang, 2003; Lin and Kiang, 2003). They showed the similar results which the removal rate of COD and ADMI (color) were over 95 % and 99 % each at the similar conditions.

Ji et al. introduced catalytic wet oxidation (CWAO) using Pt/Al<sub>2</sub>O<sub>3</sub> catalyst to treat MMA containing semiconductor wastewater (Ji et al., 2010). Pt/Al<sub>2</sub>O<sub>3</sub> catalyst is one of powerful oxidants, insoluble and solid, so it is easily separated from solution after reaction, it means that the amount of generated wastewater can be reduced. Chung et al. EO process to decompose ammonia in wastewater from semiconductor industry (Chung et al., 2020b). Following the paper, the oxidant scavenger such as hydroperoxide which was contained in semiconductor wastewater affected negatively on ammonia removal efficiency. AC was applied as a pre-treatment to remove hydroperoxide. EC process with pre-treatment was compared with breakpoint chlorination (BC). It exhibited the effective performance of ammonia removal the BC process without additional chemical injection and economical than BC process.

Chen et al. investigated cryolite crystallization to remove fluoride from etching solution (Chen et al., 2014). Experiments showed that pH

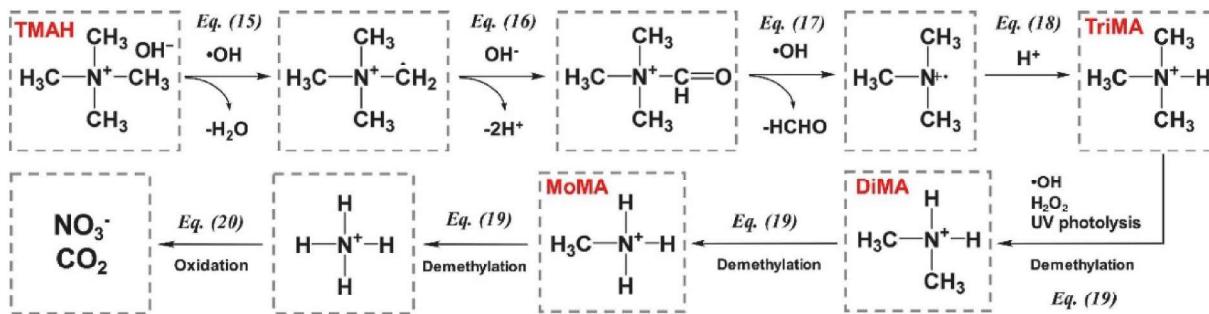


Fig. 12. Proposed degradation pathway of TMAH during the UV-LED/H<sub>2</sub>O<sub>2</sub> reaction (adapted from (Kim et al., 2022)).

3–7 and Al: F moral ratio (1:6) were the major key to produce cylolite compared with temperature.

ZVI is a reactive metal which has  $-0.44\text{V}$  standard redox potential and reacts with the oxidized contaminants with nitrate and organic compound (Fu et al., 2014). Yoshino et al. proposed ZVI treatment to remove nitrate, hydrogen peroxide and phosphate from semiconductor wastewater (Yoshino et al., 2014). Fe<sup>0</sup> was used in this paper and oxidized to ferrous ion primarily producing electrons due to zero-valent iron metal corrosion. The released electrons reacted with NO<sub>3</sub><sup>-</sup>, and Fe<sup>2+</sup> and Fe<sup>3+</sup> reacted with H<sub>2</sub>O<sub>2</sub>, PO<sub>4</sub><sup>3-</sup> from wastewater. However, more studies are required to scale up.

### 3.3. Biological treatment

Biological treatment is not only used to remove compounds which are difficult to degrade by physical and chemical treatment but also to enhance the whole treatment by combination with other treatments (Table S2). The biological treatment is known to effectively reject organic matters, NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup> and TN. Meanwhile, there are several disadvantages of biological treatments. It is required to make the suitable environments which are difficult to control for microorganism growth along with slowly removal rate of pollutants. Furthermore, the additional injection of culture medium as a feed could be needed to maintain its growth during the process. The critical drawback of the biological treatment is the generation of sludges and by-products to treat. Nevertheless, this process has been developed by researchers because hardly-degradable compounds are decomposed.

Some researchers applied biological treatment to degrade organic compounds including VOCs employing different processes combined with AOP and air stripping (Den et al., 2002; Lin and Jiang, 2003; Lin and Kiang, 2003). The performance of the combined biological fluidized-bed process which UV/H<sub>2</sub>O<sub>2</sub> was investigated. The degradation path way for each compound by UV/H<sub>2</sub>O<sub>2</sub> were proposed, then remove TOC which were produced by not only IPA but also PGMEA, EL, TMAH, NMP and PL. Most of TOC and each compound except NMP were removed by the combined process (Den et al., 2002). The combined process consisting of air stripping, modified Fenton oxidation and sequencing batch reactor (SBR) in order was employed to treat IPA-containing semiconductor wastewater. The organic compounds in the wastewater were degraded by air stripping and Fenton oxidation, then the SBR reduced over 85% of COD concentration of effluent of Fenton oxidation (Lin and Jiang, 2003; Lin and Kiang, 2003).

It was reported that biological activated carbon (BAC) reactors and biofilter ware applied to directly remove IPA, 2-propanone which are VOCs in the semiconductor wastewater without any collaboration. This work showed over 92 % removal rate of COD at 24 h of total HRT. For removal of remaining organic vapors, the biofilter was employed to prevent serious air pollution (Hsu et al., 2009).

Chung et al. and Yang et al. tried to degrade TMAH by bacterium (*Mycobacterium* sp. TMAH-W0418), these paper showed similar results; over 95 % of removal rate of TMAH (Chung et al., 2020a; Yang et al.,

2015). Innocenzi et al. tried to remove TMAH by biological treatment (detailed information wasn't mentioned) performing 99% of removal rate (Innocenzi et al., 2021).

Algae-based biosorption process was introduced to remove gallium from semiconductor wastewater. Ga (gallium) biosorption capacity was 38.5 mg gallium/g sorbent at pH 2.8. However, few studies for algae-based biosorption process was conducted to apply for scale-up (Li et al., 2018).

Two research teams targeted to reduce NH<sub>4</sub><sup>+</sup>-N, NH<sub>4</sub><sup>+</sup>, TN using ammonia oxidizing bacteria (AOB) activity in SBR process. Completely autotrophic nitrogen removal over nitrite (CANON) process in SBR exhibited 85.5% of NH<sub>4</sub><sup>+</sup>-N, 75.7% TN reduction by AOB and anammox bacteria (Liang et al., 2016). Song et al. performed 94.1–99.8% of NH<sub>4</sub><sup>+</sup> reduction by AOB and nitrite-oxidizing bacteria (NOB) activities of partial nitrification (PN) process (Song et al., 2019).

MBR was combined with RO, 4-stage Bardenpho or specific conditions (aerobic MBR and anaerobic fluidized MBR (AFMBR)) to reduce COD concentration. They showed over 90 % of reduction rate of COD (Cheng et al., 2019; Chung et al., 2014; Xiao et al., 2014) (Fig. 13). Hence, for removal of VOCs in semiconductor wastewater, the feasibility of two lab-scale moving bed bioreactors (MBBRs) using commercial biocarrier was investigated. The biological treatment process was evaluated under aerobic and anoxic conditions for treating low volatility wastewater (LVW) and high volatility wastewater (HVW). It performed 81% of COD reduction in 1 day of HRT (Cheng et al., 2021).

### 4. Future aspects and concluding remarks

The semiconductor wastewater becomes a threat to water safety, and to safely treat the semiconductor wastewater should be accompanied by the development of semiconductor industry. As the global semiconductor industry market continues to increase, the generation and discharge of semiconductor wastewater grow steadily, the study and research on proper semiconductor wastewater treatment processes are inevitable and unavoidable for mankind and the environment. The semiconductor manufacturing process is composed various detailed processes, and it has been investigated that chemicals are not only toxic but also complex to remove. This review involves a literature investigation on the current status of global semiconductor wastewater treatment such as the generation and discharge of semiconductor wastewater, the semiconductor wastewater treatment process, and regulation. This literature review on semiconductor wastewater treatment processes was classified into physical, chemical, biological, and integrated processes, then the composition, efficiency, target material, and characteristics of the processes were investigated. Additionally, the features of each process were introduced in Table S2.

Coagulation, flocculation, aggregation and precipitation mainly targeted reduction of turbidity, SS, fluoride, CaF<sub>2</sub>, COD and few of metal ions such as copper. These processes were used as pre-treatment to prevent membrane fouling and easily combined with electric, magnetic force or flotation as well. The combined processes had been

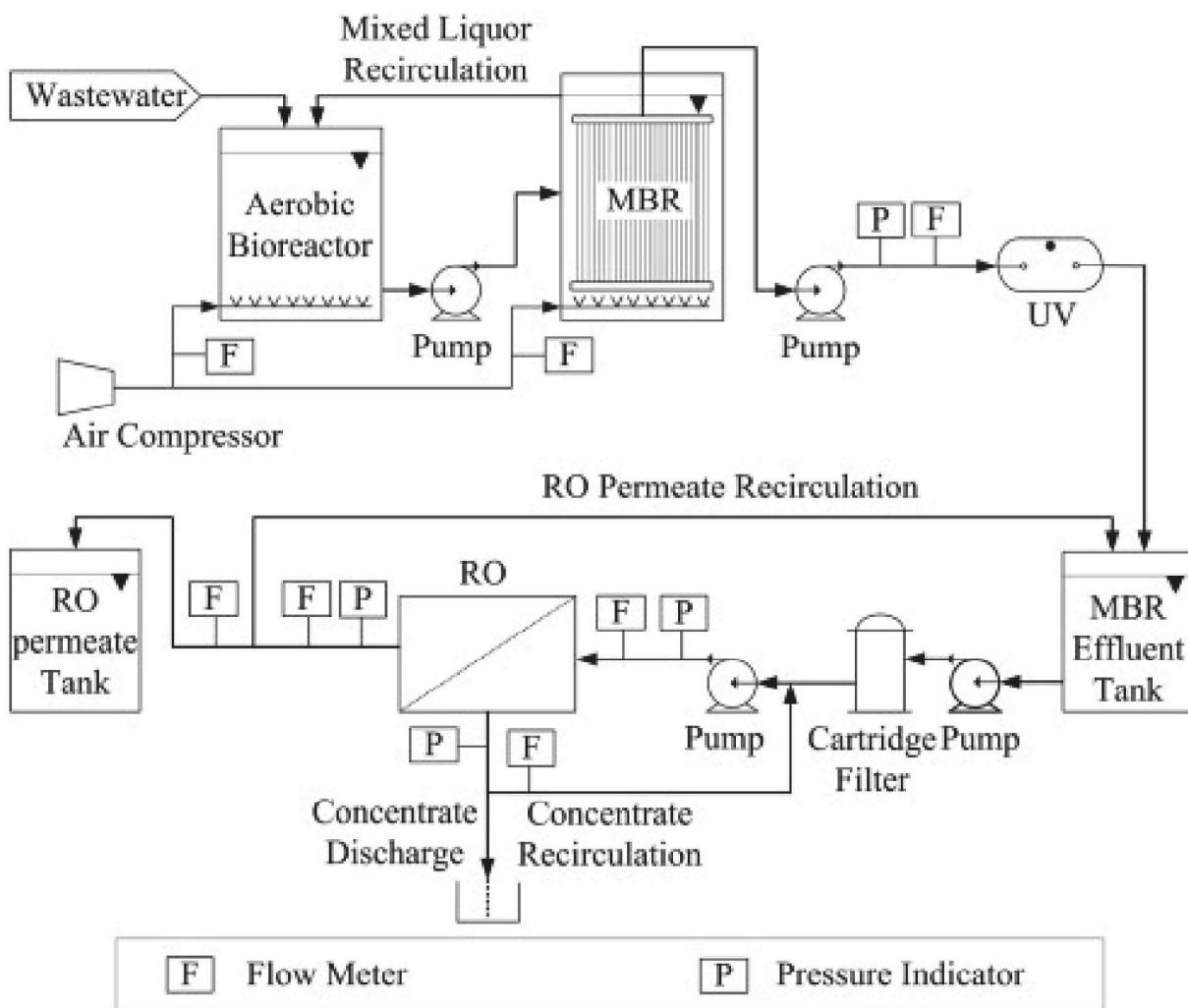


Fig. 13. Diagram of the pilot-scale MBR-RO process (adapted from (Xiao et al., 2014)).

continuously attempted. Al-anode/Fe-cathode was mainly used as the pairs of electrodes of electrical coagulation. Air stripping was employed for reduction of volatile compounds such as IPA. Air stripping and adsorption were mostly applied as pre-treatment to support the next processes. Target compounds of AOP were mainly COD or TOC along with complex contaminates such as PGMEA, EL, TMAH, IPA, PHL, NMP, MMA, Pyrazole and DGA, which are difficult to break their chemical bonding. Nevertheless, studies for unreactive target compounds still remain.

Biological treatment is primarily designed to be located at the end of the entire process and can remove COD, TOC, TN,  $\text{NH}_3^+$  and TMAH which were not fully removed from the previous processes or difficult to remove physically or chemically.

Membrane processes including MF, UF, and RO membranes but also MD, ED, and MBR Aldo It has been applied for wastewater treatment and reuse. Membranes were studied to improve water quality by reducing conductivity, turbidity, COD, TOC, etc., and tested to satisfy water quantity by permeability and flux. Some papers resulted that the treated concentrations of semiconductor wastewater reached the water quality for wastewater reuse and reclaim. However, the brine still has been remaining as challenges to safely remove. Thus, additional studies for membrane fouling are required in the future.

On the other hand, several pilot studies were reported to have been conducted so far, and studies are somewhat insufficient due to concerns over the leakage of confidential information of semiconductor companies.

In addition, the study and research on the semiconductor wastewater reuse and reclaim, which is valuable as an alternative water resource in the future, seems to require research that can meet water quality standards suitable for the purpose of using wastewater while securing water. Furthermore, in terms of resource reclaim, the semiconductor wastewater could be a chance. For that, the process for resource recovery from the semiconductor wastewater should be developed.

Understanding the address on studies and researches is the first step to solve the semiconductor wastewater problem. Through this review, it is possible to understand the proceeding study and research on semiconductor wastewater treatment process, and this review presented the research direction for future semiconductor wastewater processes. It deemed that this review sufficiently helps to the industry or research that studies semiconductor wastewater processes in the future.

#### 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.

#### Data availability

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2023.139570>.

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