7-17-2018

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Water-Energy Nexus 1 (2018) 86–96

Contents lists available at ScienceDirect

Water-Energy Nexus

journal homepage: www.keaipublishing.com/en/journals/water-energy-nexus/

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A R T I C L E I N F O

Article history:
Received 3 March 2018
Revised 10 July 2018
Accepted 15 July 2018
Available online 17 July 2018

Keywords:
Water reclamation
Water regeneration
Water network
Semiconductor
Fabs
Water resource management

A B S T R A C T

The risk of water shortage has been posing as a threat to water demanding industries in Taiwan, including the high-tech industries where ultrapure water is needed for the production of microchips. Such risks are especially unpredictable in the age of climate change, where more frequent extreme climate events such as prolonged droughts have sent these industries scrambling for securing water supply at a very high cost. The national policy also mandates strict water recycling standards for these high-tech plants, while the risk of water supply shortage also forces the industry to be water-conscious. However, most plants set their water recycling strategies based on experience or "rules of thumb" practices, without implementing optimization tools that can help making decisions in a more scientific approach. In this study we applied linear programming technique to optimize the water recovery path for a microchip assembly plant. A water balance diagram was formulated and completed to determine the existing water recycling performance, and the data was converted to a water flow network. The water flow network was then derived with a mathematical model to formulate a linear optimization problem. The proposed linear programming model is composed of mass balance constraints, unit specification constraints, capacity constraints as well as water quality constraints (discharge limits). The linear programming method was effectively appplied to improve the efficiency of water reuse. With the installation of the regeneration units, an increase of ~40.1% in the volume of reused water was predicted. The results from water cost structure also indicated that, at higher water tariff, water reuses through reclaiming and generating spent effluents can alleviate the overall water consumption costs.

Introduction

Semiconductors have enabled the information age and been viewed as the key driver of the growth in global economy. A large number of these fabrication plants ("fabs") for semiconductors were built in water-stress regions such as Singapore, Taiwan, and parts of China and United States, which suggested that other factors (e.g., supply chain, logistics, labor cost, tax exemption) may outweigh the risk of water shortage. Having said that, fabs built in locations with specific water-stress concerns are inherently more conscious about securing steady source of water supply. This means that the fabs must be ready to compensate the water deficit by supplying water internally, through water-saving practices or water recycling and reuse, during drought seasons or unexpected water shortage. Internationally, the Semiconductor Industry Association (SIA) aimed to attain a short-term goal in total fab water consumption of 7.8 L/cm^2 of wafer for 300 mm and 450 mm fabs (7.6 L/cm^2 for 200 mm fabs) and a long-term goal (in 2020) of 5.5 L/cm^2 and 4.8 L/cm^2 for the respective fab categories. The overall goal is to achieve a water recycle and reclamation rate over 75% in 2020 for SIA members worldwide (ITRS, 2013).

In Taiwan, those "high-tech" fabs are perceived as water-intensive users and consequently lead to more aggressive enforcement on water reclamation rate than most of their global counterparts. For example, the fabs in the industrial science parks are expected to meet several water-saving criteria, including process

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https://doi.org/10.1016/j.wen.2018.07.004
2588-9125/

recovery rate (i.e., total volume of reclaimed water from manufacturing process loops over total water demand) over 85%, total plant recovery rate (i.e., total volume of reclaimed water fab-wide over total water demand) over 75%, and total plant discharge rate (i.e., total volume of discharged water over total water demand) less than 70%. These criteria motivate the fabs to apply various water management strategies and practices in the design phase of water network to embrace possible factors that may affect water supply-demand relationship within the water system. Multivariate statistical techniques with cluster analysis and discriminant analysis based on the existing water use related data of 70 participating plants were utilized to evaluate water resource management strategies in high-tech industries in Taiwan (Lin et al., 2015).

One effective approach for water resource management in water-intensive industries is water network design, which includes design the network in both production and secondary use levels. Design of water network, by means of graphical methodologies (Alwi et al., 2008; Manan et al., 2006), mathematical programming (Feng et al., 2007) and synthesis of mass exchange networks (Shafiei et al., 2004), has been applied to allocate streams between water-using units, due to the increased interests for sustainable development in industries (Boix et al., 2012). The purpose of water network design is to maximize water generation and reuse water into the industrial processes (El-Halwagi et al., 2003). Chew et al. (2009) investigated a mass-exchange based targeting technique for optimization of both in-plant and inter-plant water networks. In the technique, concentration cascade diagrams were constructed to determine the reuse and recycle schemes, suggesting water quality properties (e.g., conductivity, pH, turbidity) in place of mass-based chemical impurities can also be optimized with the technique. For instance, when resistivity and heavy metal content were set as the main properties in evaluating water reuse opportunity, the simulation results revealed reduction of freshwater intake from 1737 to 473 t/h, and wastewater discharge from 1430 to 166 t/h. Wang et al. (2012) proposed the concentration potential-based linear programming to optimize in-plant water network problems. Concentration potentials were inserted as objective functions and constraints in the optimization program, with network design with only reuse targets (Wang et al., 2012) and regeneration units (Zhao et al., 2016). Their results indicated comparable or reduced freshwater consumptions then that obtained in literatures.

The objective of this study is to develop an in-plant water network scheme on the basis of both volume and mass balance (i.e., water constituents) for a wafer packaging and assembly fab, and to optimize the scheme using linear programming technique. The scheme clearly defines spent water from processing units (i.e., source streams) and the points of reuse (i.e., demand points), as well as the existing and potential regeneration opportunities with cost considerations, using detailed water auditing data. Similarly, mass balance of the water constituents, including chemical oxygen demand (COD), ammonia-nitrogen (NH_4-N), suspended solid (SS), are also considered in the optimization. Additional chemical components such as boron, characteristic of the lithographic process effluent stream, and metal constituents (Cu, Zn, Pb, Ni) are regarded as constraints in the final discharge.

A microchip manufacturing fab that involves intensive water consumption is used as a case study. The scenario-based analysis first examines the water recycling opportunities and capacity with the fab's existing water network by establishing the flow data. Next, the water flow network is expanded with the incorporation of various regeneration units to reclaim process effluents, and the in-plant water reclamation scheme is revised under the assumption of various hypothetical removal efficiencies of pollutants. Finally, a generic cost component including the tap water cost and process-dependent regeneration costs is devised, and the cost-benefit of the water reclamation scheme is evaluated. The results from this study could be of interests for decision makers in water resource strategy planning.

Methodology

Water use in a fab

Semiconductor industry consists of several supply chain components, mainly including circuit design, fabrication, packaging and testing. The science industrial parks in Taiwan are clusters of semiconductor manufacturing supply chain companies, creating a symbiotic relationship with added value in adapting to swift technology evolution and market change.

Fig. 1 presents a generic water network for a typical semiconductor manufacturing fab. The water network chart was adapted from the standard version developed by the Association of Taiwan Semiconductors Industry, the leading organization made up of professional facility engineers and decision-makers of the industry. A stream of reclaimed water from the discharge of process tools is commonly segregated, screened for its water quality, and subject to different treatment processes which is contingent on whether the reclaimed water is to be reused for purposes such as UPW, facility use, and diluting wastewater. The four sources of water supply include ultrapure water (UPW) (S_3), tap water (S_4), and two regenerated water units (S_1 and S_2). The UPW unit supplies all chip-manufacturing (i.e., process) water (T_1), whereas tap water unit supplies the makeup water for the scrubber units (T_2), cooling tower units (T_3), and public uses (T_4). A portion of the spent water collected from the processes are regenerated by the two regeneration units (R); the water regenerated by Regeneration Unit I is reused to produce UPW (R_1) and that regenerated by unit II is reused for the scrubbers (R_2), cooling towers (R_3), and public use (R_4). Wastewater flows to be discharged are designated as W_1 and W_2 for those yielded from the two regeneration units, and W_3 through W_6 for those generated from processes, central scrubbers (CS), cooling towers (CT), and public uses. It is worth noting that reclaiming spent water exhibiting acceptable water quality to partially replace tap water for UPW production has become a common practice in Taiwan, where the risk of water shortage outweighs the risk of reduced yield attributable to trace impurities in the highly purified reclaimed water.

Though the water reclamation mandates treat each manufacturing component equally, each fab has its own distinct challenges in reclaiming spent water from processes. Most of the challenges stem from the lack of mature technologies to target contaminants unique to a manufacturing process. Other than the common water quality parameters such as COD, total organic carbon (TOC), SS, and process-specific contaminants must be removed for a stream to be reuse, regenerated, or discharged. A few examples include metal oxide (e.g., SiO_2, Al_2O_3, WO_3) nanoparticles from wafer polishing processes, heavy metals from electroplating processes, fluorides from wafer rinsing processes, metallic (e.g., Al, Ga, In, Mo) and metalloid (e.g., B) from substrate materials. Cost and space constraints may also impede the flexibility to renovate existing facilities into more an effective water reclamation system.

Water flow network in Fab A

This study presents a case of a microchip packaging and assembly fab (hereinafter referred as Fab A) converted from a wafer fabrication fab. The chip packaging and assembly processes typically involve wafer back grinding, die sawing and bonding, wire bonding, trimming, and electroplating. The waste streams generated from these processes are segregated into etching wastewater, sawing wastewater, lithographic wastewater, organic wastewater, electroplating wastewater, and rinse wastewater. Other in-plant

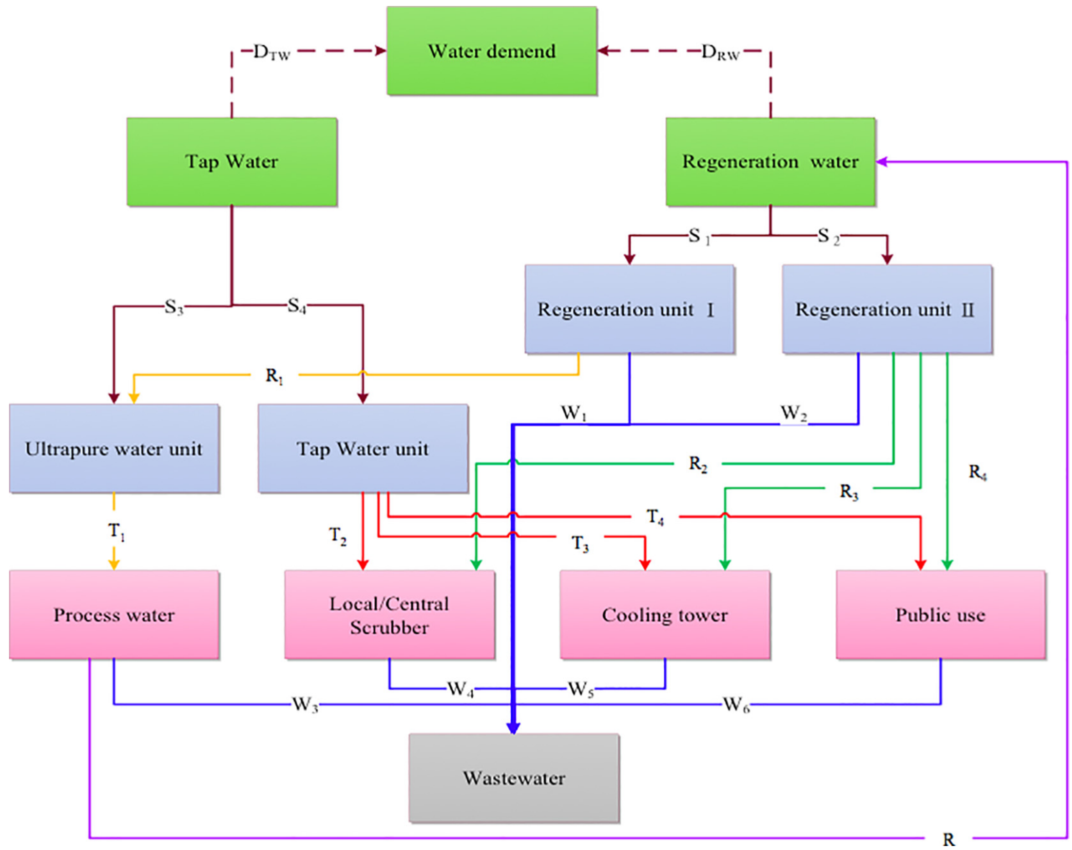

Fig. 1. Basic structure of the water flow network for a typical fab. The color-coded lines indicate the water supply (in maroon), tap water for process use (in yellow) and facility use (in red), regenerated water for process use (in yellow) and for facility use (in green). Reuse water and discharged flow are marked in black and purple, respectively.

source streams come from blowdowns by cooling towers and central air scrubbers. Fig. 2 shows the water network diagram for Fab A, expanded from the generic flow network presented in Fig. 1 to tailor the conditions for a specific fab. This network presented incorporated several forward and reverse water cycles of tap water, sanitary water use unit, ultrapure water unit, secondary usage unit, UPW regeneration wastewater unit, MMF backwash wastewater unit, ACF backwash wastewater unit, process water unit, etching wastewater unit, sawing wastewater unit, lithographic wastewater unit, organic wastewater unit, electroplating wastewater unit, rinse wastewater unit, UF unit, RO unit, secondary usage unit, cooling tower unit, neutralization tank unit, central scrubbers unit, and discharge unit.

Structure of modeling

Two objectives are proposed in this study to determine economic and environmental water reusing strategies in a wafer fab, namely, maximization of water reuse and minimization of total water operating cost (cost for tap water supply and regeneration of spent water). For cost minimization, the main consideration is to determine the difference between the cost of tap water consumption and the cost saved from using regenerated water. Consequently, one can anticipate a cost-driven motivation to reduce the use of tap water if its cost is substantially greater than that of the regenerated water. Conversely, water reclamation is likely be driven by other considerations (e.g., regulations, corporate social responsibility and risk management) if the efforts to wastewater regeneration cannot be economically justified.

The mathematical equation for the objective of water reuse maximization (E) and cost minimization (Z) can be expressed as in Eqs. (1) and (2), respectively.

$$Max\ E = \sum_{j=1}^{4}(T_j + R_j) \tag{1}$$

$$Min\ Z = \sum_{i=1}^{m} S_i c_w + \sum_{j=1}^{n} F_j c_j \tag{2}$$

Eq. (1) follows the notations in Fig. 1, whereas Eq. (2) uses the notations defined earlier in Fig. 2, where S_i are the sources of water supply and F_j are regenerated water flows; c_w is the cost of city water per unit volume, and c_j is the cost of wastewater regeneration from various sources.

The proposed linear programming model is composed of mass balance constraints, unit specification constraints, capacity constraints as well as water quality constraints (discharge limits). The detail mathematical models are introduced in the Appendix A. LINGO software (version 17, LINDO Systems, Inc., USA) was used as the optimization tool.

Model assumption on constraints

The key constraints to the modeling are water flow balance and contaminant mass balance. Additional constraints attributable to discharge limits for the regulated contaminants and the water quality requirements at the point of water reuse also take critical effect on determining the maximum quantity of wastewater to be reclaimed. For example, water regeneration units commonly function only as physical separation of contaminants from wastewater (e.g., membrane-based filtration) into concentrated streams, without changing the chemical state of the contaminants (e.g., removing contaminants by oxidation). In such cases, the contaminant mass discharge limit will dictate the quantity of “cleaner” water needed to yield sufficient dilution factor for

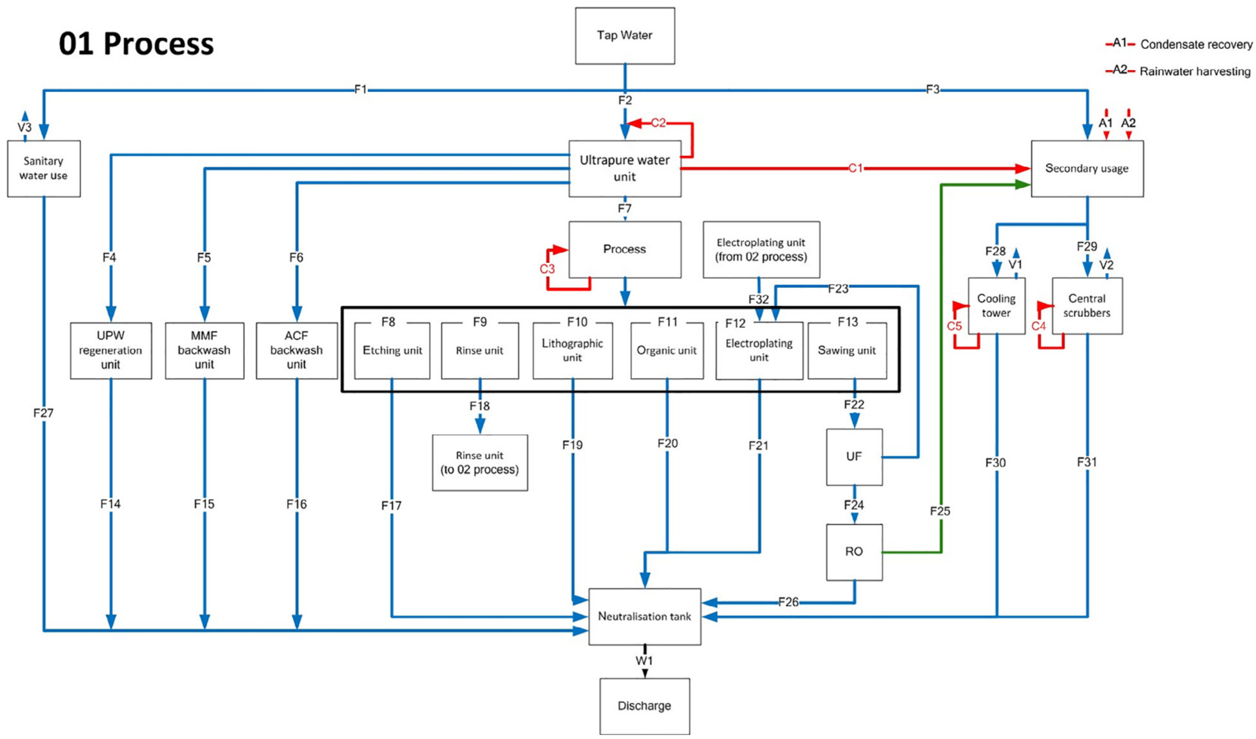

Fig. 2. Water flow network for Fab A (F1-F3 represent, respectively, the flow of tap water to sanitary use, ultrapure water production, and facility uses; F4–F7 the ultrapure water to its regeneration unit, MMF backwash unit, ACF backwash unit, and process units; F8–F13 the process water to segregated wastewater units of etching, sawing, lithographic, organic, electroplating, rinse; F14–F21 the flows to the neutralization tank from the wastewater units of UPW regeneration, MMF backwash, ACF backwash, etching, sawing, lithographic, organic, electroplating; F22 and F24 the flow of rinse wastewater unit to UF unit and to RO unit; F23 the reuse from UF unit to electroplating wastewater unit; F25 and F26 the flow from RO unit to facility units and to neutralization tank; F27 the quantity from domestic wastewater to neutralization tank; F28 and F29 the flow from facility unit to cooling tower and central scrubbers; F30 and F31 the discharge from cooling towers and central scrubbers to neutralization tank; F32 the flow from Electroplating wastewater unit in process II to electroplating wastewater unit; V1 and V2 the water evaporated from cooling towers and central scrubbers; W1 the neutralized wastewater to discharge tank.

discharge, instead of being reclaimed and reused. Contrarily, if contaminants are purified or removed from waste streams in any of the water regeneration units, then the mass of contaminants to be discharged becomes lesser, hence creating more options to reuse the "cleaner" which otherwise would have been used for diluting the discharging stream.

For simplicity, assumptions for ruling the water reuse for a fab are as the following:

– Contaminants of concern in the effluents include COD, SS, NH_3-N, Cu, Ni, Pb, Zn, B, and F. Other water quality parameters such as electrical conductivity, which is a critical parameter specific only to cooling towers and central air scrubbers, are not considered in the present study.
– Wastewater can only be reclaimed from the segregated process streams. Wastewater collected in the neutralization tank is not considered recyclable.
– For any water regeneration unit, contaminants are removed at efficiencies indiscriminately of its type.
– A fixed cost rate (i.e., dollars/volume) is applied regardless of the quantity of supply and reclamation. The base-scenario uses a fixed tap water cost of US\$0.40/m^3. Other cost components associated with water supply and treatment for final discharge are not considered.
– The backwash water collected from the regeneration processes of UPW and ACF units are assumed to be ten times concentrated from the tap water, whereas that from the MMF unit is twenty times concentrated.
– Recirculating water for cooling towers and central air scrubbers were not considered in the model, as inclusion of these recirculating water in the calculation often misleads a plant's efforts on water reclamation and reuse.

Water flow network scenarios

In the present study, three scenarios are proposed to understand the influence of factors on water reuse performances:

1. Installation of process effluent regeneration units: Under the circumstances where contaminants undergoing no chemical changes, the enhanced water reclamation performance attributable to the newly installed water regeneration units. These units mainly include regeneration from process effluents such as electroplating, sawing, lithographic, etching, and rinsing. Fig. 3 highlights the units added to the expanded flow network for Fab A. However, effluents from etching (F17) and lithographic (F19) processes, which comprise only 4.3% of the total process effluent flow volume, are not considered cost-effective to be a viable source of water regeneration.
2. Change in tap water tariff: With consideration of cost components, the cost effectiveness (cost per unit volume of water reclaimed) of the water reclamation options. Specifically, we compare the cost effectiveness of maximizing the quantity of water reclamation *vis-a-vis* the minimal total cost. Fixed rates of water regeneration units are adopted from literature (Table 1), which strongly depend on the water quality to be regenerated and the technology applied. In the present case,

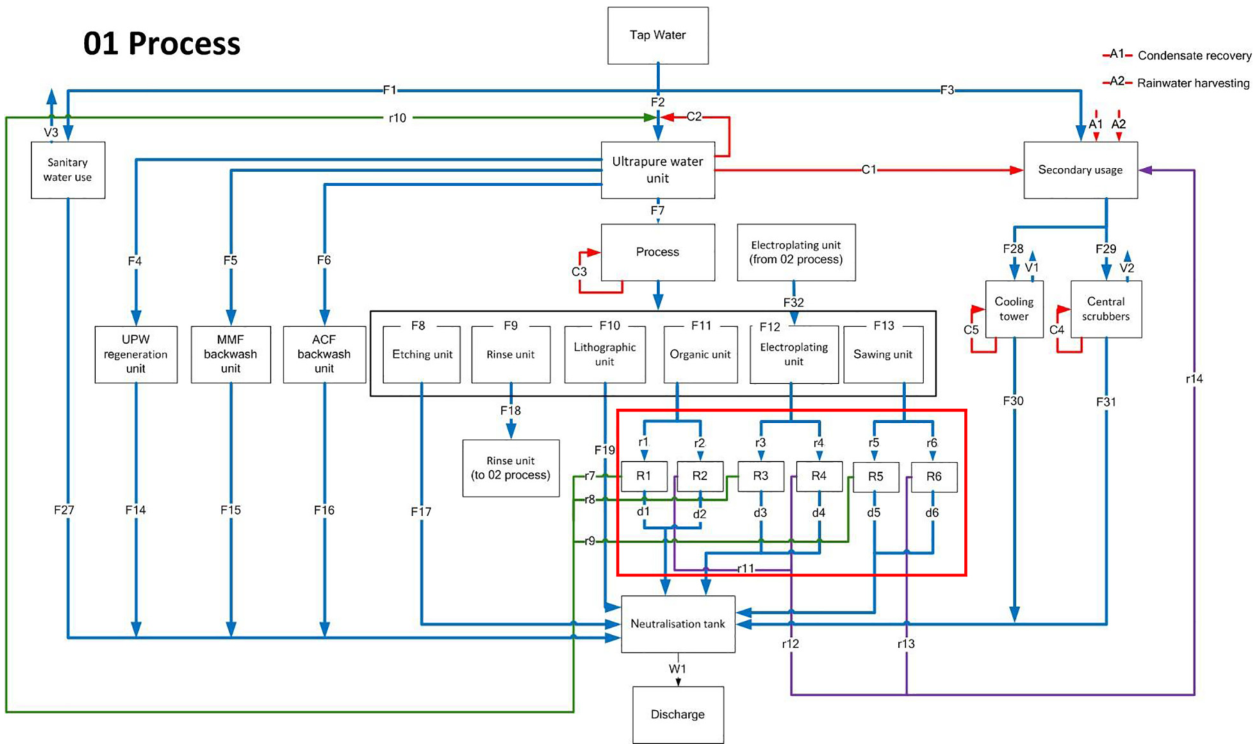

Fig. 3. Scenario with installation of process effluent regeneration units (highlighted in box area). R1, R3, and R5 are regenerated and reused at the process level, whereas R2, R4, and R6 are regenerated and reused at the facility level. d1 through d6 are concentrated discharges from the regeneration units.

we assume that regenerated streams from segregated process effluents are reused both for microchip processes that entail a water quality better than tap water, and for facility processes (i.e., CT and CS) which require less stringent water quality.

3. Increase in contaminant removal efficiency: In the present scenario water purification processes are introduced. The processes allow chemical removal of contaminants from the wastewater stream. A removal rate is hypothesized to assess the effect of water recycling processes. For simplicity, we assume that all water quality parameters are purified invariably at a designated removal rate by a given water regeneration unit. This assumption is obviously not realistic considering different water purification technologies are needed to target for the reduction of specific types of contaminants. The assumption nevertheless permits us to avoid complication of problems by decoupling the extent of contaminant reduction from the cost components.

Table 1
Unit cost for tap water supply (S1) and regeneration of spent water (R1–R6) in the water flow network considering regeneration. The cost for regeneration units include running cost but exclude capital cost.

Unit	Suggested regeneration treatment technology	Unit cost (USD/m^3)
S1	–	0.40
R1	MBR	0.67
R2	MBR + RO	1.07
R3	ERD	0.53
R4	ERD	1.00
R5	UF	0.30
R6	UF + RO	0.63

Results and discussion

Water flow network optimizations

Existing status of Fab A

The water reuse mandates set by the local environmental regulatory agency requires a three-level measure, namely process-level reuse efficiency (RP), fab-level reuse efficiency (RT) and fab-level discharge rate (DT), as shown in Table 2.

At present, Fab A is able to comply with the stringent mandates by attaining a RP value at 84.6%, RT value at 83.5%, and DT value at 62.9%. Most of the reuse water is attributed to C1, C2, and C3 as indicated in Fig. 2. Water can be reclaimed from these sources with relatively low cost because little or no additional purification process is needed. In particular, C3 represents a water circulation

Table 2
Enhanced water reuse and discharge performances through installation of regeneration units. The calculations include reuse of circulated cascade rinsing water (C3) from process units.

Performance indicator[a]	Base-scenario (%)	Installation of regeneration units (%)	Percent change (%)
Process-level reuse (RP)	84.6	88.4	+3.8
Fab-level reuse (RT)	83.5	87.2	+3.7
Fab-level discharge (DT)	62.9	57.5	−5.4
Total amount of recovered water (CMD)	703.1	984.2	+40.1

[a]Definition of the indicators are:
- Process-level reuse efficiency (RP) $= \frac{C1+C2+C3+F23+F25}{C2+C3+F2} \times 100\%$
- Fab-level reuse efficiency (RT) $= \frac{A1+A2+C1+C2+C3+C4+C5+F23+F25}{S1+A1+A2+C1+C2+C3+C4+C5+F23+F25-V1-V2} \times 100\%$
- Fab-level discharge rate (DT) $= \frac{W1}{S1+A1+A2} \times 100\%$

loop designed for cascade-type rinsing processes. Rinsing water is used for consecutive steps in a rinsing procedure which requires progressively lesser purity of water for the downstream cycle. The rinsing water becomes progressively contaminated in the cascade tank and is eventually discharged when it no longer meets a pre-determined level of water quality. Oftentimes the value of C3 is estimated by measuring the volume of the rinsing chamber and the number of cascade in the rinsing process, without an actual measuring gauge. Further, C1 and C2 are direct reuses of the reject portion of ultrapure water production process. The water quality of the reject streams, though inherently worse than the feed water, is consistent and acceptable for secondary reuses as backwash water for various filter media, cooling water, and air scrubber inflows (C1). In the present case, a fraction of the reject water is also recycled back to replace feed water for the UPW production unit (C2).

One should note that C3, C4, and C5 each represents a reuse loop (circulating of used water) that substantially contributes to the fab's water reuse efficiency, but it does not play any role in the water reuse allocation and distribution. It is pertinent to also examine the water reuse efficiency by excluding them from the calculation, especially in the case of Fab A for having a C3 in remarkable reused volume (5800 m^3/d). This further suggests that regeneration of used water needs to be considered to boost the in-plant water reclamation and reuse capacity (Lin et al., 2016). Regeneration of used water entails a holistic approach to assess the availability of purification technologies tailoring the type of contaminants to be removed, to identify the allocation of the points of reuse and the minimum water quality needed, and the cost associated with the installation, operation, and maintenance of the regeneration units (Yang et al., 2014; Ruiz-Rosa et al., 2016)

Installation of regeneration units

To enhance the water recycling efficiency, a more complicated scenario with introduction of reverse units of R_1 and R_2 from organic wastewater unit, electroplating wastewater unit, and sawing wastewater unit, as shown in Fig. 3.

Table 3 lists the water flow rate and the concentrations of contaminants of concern in the flow streams. The discharge limits for the contaminants are also included in the table. These values reflect a realistic case scenario, although water quality of the actual flow streams can significantly fluctuate, and it is contingent on the operating mode of the corresponding fabrication processes. The change of chemicals applied to the processes, often involving proprietary chemicals to meet the needs of new fabrication techniques or products, also complicates the chemical composition of the spent water. In this case scenario, flow streams from key fabrication lines include etching (F8), rinsing (F9), lithography (F10), organic (F11), electroplating (F12), and sawing (F13). As shown in Table 3, F11 through F13 predominate the total flow volume. The key challenges concerning water quality mainly include: the organic stream (F11) contains COD concentration more than four times of the discharge limit; the sawing spent stream (F13) contains SS concentration that is nearly twice the discharge limit; and the lithographic waste stream, despite small in volume, contains boron concentration that is consistently over fifty times over boron's discharge limit. Therefore, meeting the discharge limits generally imposes a limitation to the volume of water to be regenerated or reuse.

With the installation of the regeneration units defined in Fig. 3, the RP value can be enhanced to 88.4%, RT to 87.2%, and DT is reduced to 57.5%, as shown in Table 2. This scenario, with an increase of 40.1% in the volume of reuse water, also represents the threshold of the water reuse capacity for fab A, without consideration of discharge limits and the cost components. Realistically, the various classes of contaminants in the process effluents may not be sufficiently removed to meet the discharge limits, and thus one needs to evaluate both the technical viability and the cost benefit to treat the contaminants in the effluents for enhancing water reusability and reducing discharge liability. The ensuing sections discuss the impacts of the aforementioned factors.

In the case scenarios, the regeneration technology can involve either a physical process or a (bio)chemical process. While both processes can remove contaminants, physical processes merely separate contaminants from the purified streams, hence the total mass of a contaminant remains unchanged in the water flow network. In contrast, biological or chemical processes convert contaminants into other forms, therefore mass reduction of a contaminant via various pathways (e.g., oxidation, complexation, precipitation) can be expected. For example, the SS concentration in F13 stems primarily from wafer backgrinding process in which the backside of the silicon wafers are thinned prior to wafer dicing, in order to separate the wafer components into individual microchips. The solid particles comprise mostly silicon residues (via course grinding) and polishing slurry particles (fine grinding) that typically in the size range of micrometers. These particles can be effectively separated from water by ultrafiltration preceded by either microfiltration or coagulation/flocculation step (Huang et al., 2011). It is also noteworthy to mention that fluoride ions in the effluent of etching process can occasionally exceed the discharge limit. Fluoride ions can be precipitated via reaction with various calcium salts ($Ca(OH)_2$, $CaSO_4$, $CaCl_2$) to form insoluble calcium fluoride (CaF_2) which is often reused as cement ingredient (Won et al., 2012).

Increase in contaminant removal efficiency

In this presented case, boron is a critical component of proprietary chemical agent applied in the lithographic process of wafer fabrication. The stringent discharge limit for boron necessitates its removal from the waste stream to create room for water

Table 3
Input values of water flow and water quality parameters for the major flow streams in the water flow network in Fab A.

Flow stream	Q (m^3/d)	COD (mg/l)	NH_3-N (mg/l)	SS (mg/l)	Cu (mg/l)	Ni (mg/l)	Pb (mg/l)	Zn (mg/l)	B (mg/l)	F (mg/l)
F1	260	25.4	10.4	16.2	0.195	0	0	0.135	0	0
F4	96	78	1	0	0	0	0	0	0	2
F5	18	156	2	0	0	0	0	0	0	4
F6	11	156	2	0	0	0	0	0	0	4
F8	0.6	12.8	0	22.9	0	0	0	0	0	11.7
F10	20	103	19.53	3	0.151	0.047	0.011	0.156	50	0
F11	339	462.3	2.2	1.35	0.052	0.059	0.119	0.315	0.061	3.74
F12	520	12.4	0.05	37	0.128	0.139	0.116	0.54	0.173	0.1
F13	180	70	4.5	96	0	0	0	0	0	0
F30	92	35	0	0	0	0	0	0	0	0
F31	10	5	0	9	0	0	0	0	0	48
C3	5800	0	0	0	0	0	0	0	0	0
Discharge limit		100	30	30	3	1	1	5	1	15

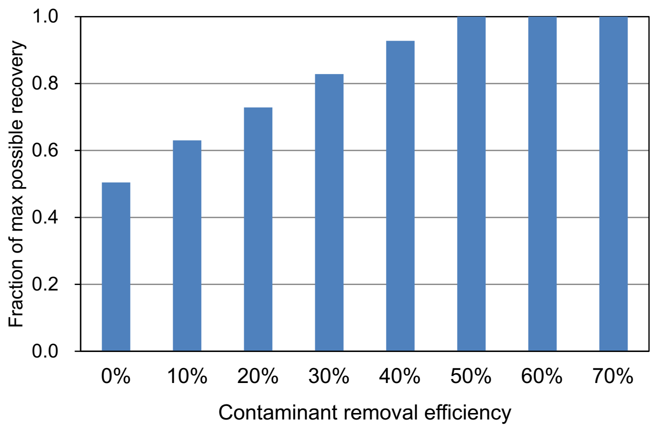

Fig. 4. Effects of the boron removal efficiency for the regeneration units on the degree of water reuse, expressed in the fraction of maximum possible recoverable water. The fraction is defined as Volumatic ratio between the water regenerated at a specific contaminant removal efficiency and that of the base scenario. The maximum of the fraction is 1.

reclamation. Chemical precipitation involving the addition of an oxidant to convert boric acid into perboric acid in alkaline condition, followed by using $Ba(OH)_2$ as the precipitation agent, has been demonstrated as a viable method to remove boron from low ppm level (Lin et al., 2016). Alternatively, boric acid being a Lewis acid in that they readily form adducts with electron-pair donors, can easily complexes with dihydroxyl functional groups in commercial ion exchange resins (Wang et al., 2014). Ion exchange is a more prudent method when dealing with low level of boron concentration. In both purification methods, boron is considered "removed" from the waste streams because it is converted into forms of a solid (e.g., chemical sludge, spent resin) that are not part of the water flow network.

Fig. 4 shows the effects of boron removal efficiency on the amount of water to be reclaimed and reuse. In the figure the degree of water regeneration is expressed as the normalized reused rate, defined as the volumetric ratio between the water regenerated at a contaminant removal efficiency and that of the "base" scenario (i.e., R10 and R14 in Fig. 3 are both nil). The results indicate that, with no mass removal of the boron, the normalized maximum recovery rate is capped at approximately 50%. This value progressively increases as the boron are removed. With a removal efficiency greater than 50%, the normalized rate reaches its threshold. For Fab A, the limiting factor is the aforementioned boron contamination which necessitates an optimized rate of its removal to maintain sufficient level of dilution and to regenerate a fraction of the process effluent. The results from sensitivity analysis in Fig. 5 also support the findings that the water recovery rate is strongly influenced by wastewater discharge limit. Part (a) of the figure indicates the influence of tap water cost to the potential of reclaimable water. For example, a + 50% refers to an increase of 50% in tap water cost; the corresponding increase in the reclaimed water would be 54% as compared to that at the base cost. Part (b) indicates the influence of discharge limit to the potential of reclaiming water. For example, a 50% relaxation of the limit (in concentration, −50%) would yield an increase of 30.7% more water to be reclaimed. Conversely, a tighter limit of 20% (+20%) would yield a 28.2% reduction of water to be reclaimed.

Influences of cost structure

Regenerating process effluents typically involve various cost levels that play a critical role in the decision making pertaining to the level of decontamination and the points of reuse. Conversely, the cost of tap water becomes the primary factor influencing whether an investment on water regeneration can bring financial benefits. Theoretically, when the tap water cost is lower than any of the regeneration cost level, then regenerating process effluents would yield no cost benefit and a decision would base solely on complying regulatory requirements. In contrast, when the tap water cost is greater than any of the regenerating costs, then regenerating and reuse all process effluents would be the most prudent decision. When the tap water cost is in the range of the regenerating cost levels, then regenerating only the process effluents with their costs lower than that of tap water would make sense. Consequently, the volume of reused water will form a step-wise increase with the tap water cost.

Fig. 6 presents the overall water consumption cost for Fab A as a function of tap water cost. The tap water cost of US\$0.40/m^3 is used as the based value against which all other scenario costs are normalized. Each junction of slope changes represents one of the cost levels for regenerating process effluents (Table 1). The increase in the water tariff inherently raises the overall water consumption cost, however the slope of the consumption cost gradually lessens with increasing water tariff. This indicates that, at higher water tariff, water reuses through reclaiming and generating spent effluents can alleviate the overall water consumption costs. However, if the water tariff includes discharge fees and sludge treatment and disposal fees, then the actual cost of water supply can be significantly greater than the fixed flat rate. But regenerating wastewater for reuse still remains as an economic option comparing to other

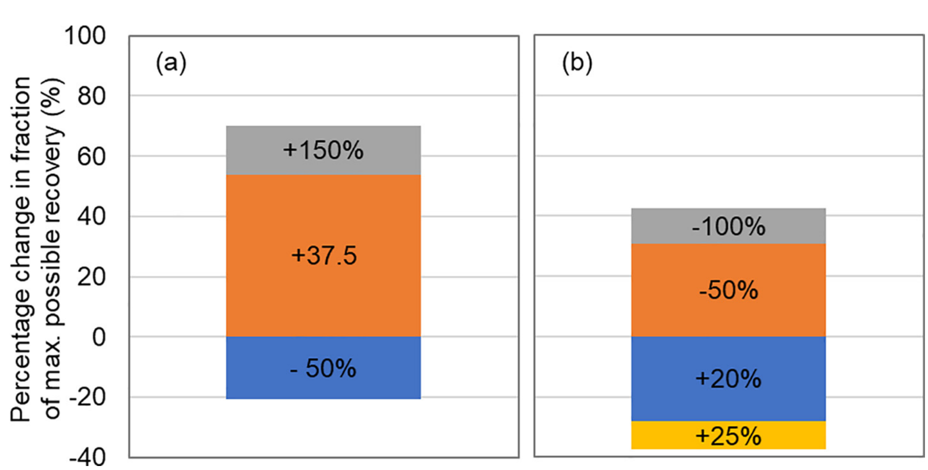

Fig. 5. Sensitivity analysis on influences of (a) tap water cost and (b) wastewater discharge limit.

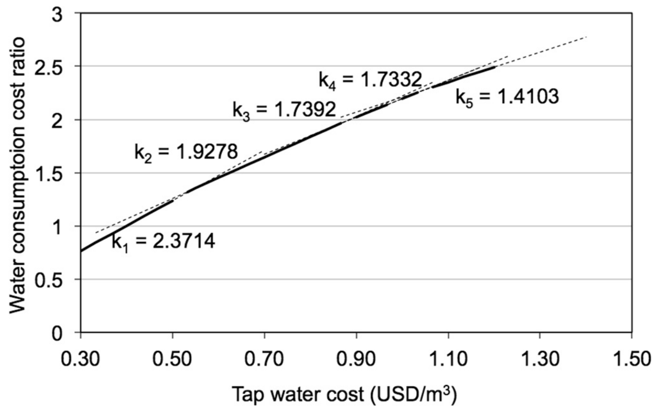

Fig. 6. The effect of tap water price on the overall water supply cost in Fab A. The water consumption cost ratio is normalized against the cost corresponding to the existing tap water cost at USD 0.40/m^3. k_1 through k_5 represent the values of slopes corresponding to the various levels of effluent regeneration cost.

existing water sources such as desalinated seawater (Ruiz-Rosa et al., 2016) Therefore, the cost benefit of reducing water intake for the plant operation by attaining improved water reuse efficiency can be markedly greater than the scenario demonstrated thus far. For example, if the water tariff were US$1.00/m^3 instead of the fixed flat rate at US$0.40/m^3, then the overall water consumption cost would have been reduced by 27%, on the basis of the difference in slopes reflected in the values of k_1 and k_3.

In most cases, the cost of a treatment process exhibits an exponential-like function of pollutant concentrations (or mass and volumetric loadings), though the cost functions are highly site-specific and difficult to acquire from literatures other than those disclosed in technical reports of regulatory agencies concerning the cost assessment of municipal wastewater treatment works. Removal of nutrients (nitrogen and phosphorous) through biological treatment processes and the associated costs have been the focus of investigations in these reports (Jiang et al., 2004; Hartman and Cleland, 2007), which are of limited value to the treatment of industrial effluents. Nevertheless, from an operational point of view, having a cost increasing with removal efficiency makes sense for the following reasons: (1) lowering the mass of a contaminant (i.e., increasing removal efficiency) inherently reduces its mass transfer rate when a heterogeneous phase is introduced in a treatment process. Higher energy is entailed to compensate for the reduced mass transfer and perpetuate the process functionality. (2) In homogeneous reactions with fixed reaction kinetic rates, the degree of absolute mass reduction as a function of reaction time diminishes. Longer reaction time is thus needed to reach a high removal efficiency, thus elevating the cost of operation. (3) While the marginal pollution control costs increases with removal efficiency, the marginal environment benefit (i.e., environmental damage cost) decreases. This counteraction of the costs generally indicates the existence of an "optimum" value of removal efficiency with respect to cost of operation. Reducing energy intensity of treatment processes and system as a whole (treatment, transport, reuse application) is widely regarded as the best approach to reduce the operating cost. For example, Gabarrón et al. (2014) compared the energy intensities of several municipal facilities in Spain and studied the energy-saving strategies and operational costs of stand-alone, hybrid, and dual stream full-scale membrane bioreactors (MBRs). An Australia study examined the energy intensity of treatment and distribution systems and strategies to reduce energy consumption based on a participatory study involving eight diverse water recycling schemes across Australia (Institute for Sustainable Futures, 2013). They concluded that recycled water energy intensity is generally high because the source water quality is low, especially when schemes managing risk perceptions require treating beyond the level required. With the intention in the present study to demonstrate the effect of removal efficiency of pollutants on the potential volume of recoverable water for reuse, we have assumed a fixed cost for specific treatment processes irrespective of their capacities in removal efficiency due to the lack of cost-function data for these treatment technologies. This assumption obviously does not reflect the likely increase in the process cost at an improved removal efficiency, which would have been an important decision-making factor and could be a major source of uncertainty on the predictive outcome of the model.

Recommendation of selecting water reclamation process

Incentives of promoting water recycle and reuse in manufacturing plants rely on whether the benefits of reuse water worth the costs and risks. Educated managers for fabs with large volume of water consumption generally recognize that the risks of short water supply and unreliable source water quality far exceed the cost of implementing water recovery systems. The economic viability, however, is still the determining factor whether a plant is committed to enhance its water use efficiency. Several models have been proposed to evaluate the economically "optimal" water recovery rate (WRR) using different assumptions. The common cost analyses include: tap water costs, utility (operating) costs, equipment and maintenance costs of water recovery system, wastewater treatment and discharge costs. Among these factors, the cost-benefits are mainly driven by the water cost (tariff) and wastewater discharge cost.

Fig. 7 shows the cost-benefits estimated by the semiconductor industry in Taiwan, using the total costs at 0% WRR (no recycle) as the baseline level. These results show that the economically optimal WRR was near 75%, but further implicate that the optimal WRR increases with water tariff. If the water tariff were to increase at levels greater than US$2.5/m^3, then strong incentive of cost-reduction is still present to drive toward further WRR.

To raise the water use efficiency, plants are encouraged to develop advanced processes which are more environmental benign, exemplified by using processes such as ozonated clean solution and supercritical CO_2 cleaning to replace conventional chemical cleaning methods. These advanced processes can reduce the discharge of hazardous chemical waste and the use of UPW for rinsing. Additionally, plants should consider the current status of

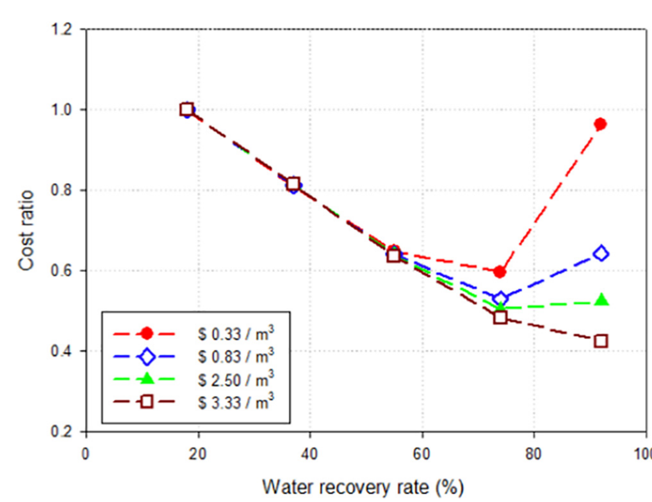

Fig. 7. Cost-benefits of water recovery at various water recovery rates for the semiconductor industry in Taiwan.

their WRRs and adapt a strategy that best suits their goals, economically and productively. For example, direct recycling of UPW reject water and part of processing water by simple re-routing would attain a WRR as high as 60%. The next option would be recycling high-purity processing water through treatment, followed by end-of-pipe water reclamation through extended treatment. The costs of the last option, however, needs to be carefully examined depending on factors such as the potential reclaimed water volume and designation of uses. In Fab A's situation, through the reuse of cascade rinsing water, UPW reject stream and other filtration backwash water, its water use efficiency has already met the stringent requirements. However, meeting the discharge limits for boron, SS and COD necessitates the purification of process effluents discretely collected and treated. The costs associated with the regeneration of process effluents are highly contingent on the level of purification needed for each regulated contaminants. The purity of the regenerated water, in turn, shapes the water network in the fab by determining the requirements of the points-of-reuse.

Notably, even though the cost of effluent regeneration has accounted for the energy (electricity and steam) consumed, the growing awareness of energy consumption relating to water consumption can play an increasing role in the deployment of regeneration strategy and the selection of water treatment technology. With the availability of viable data on the energy intensity or carbon intensity for each of the potential water reclamation and regeneration processes, these criteria can be easily applied in the optimization programming as an additional objective function.

Conclusions

Semiconductors are enabling technologies for the global economy. Semiconductor manufacturing technologies have also advanced over the last four decades, but the environmental burden associated with the technology advancements has also raised awareness of the natural resources consumed and the hazardous waste produced by the life-cycle of semiconductor devices. Securing safe and clean water supply is therefore regarded as one major challenge to the corporates. This work introduces several water reuse strategies to enhance the water use efficiency of a wafer manufacturing fab using linear programming approach. The results from the optimization revealed that enhanced in water reuse efficiency could be greatly improved, while both water and constitute mass balances were considered in the model. Our findings also suggested the need for collaboration between policy makers and plant managers for having a maximum water reuse at maximum environmental and economic benefits.

Acknowledgement

The authors acknowledge the funding support by the Administration Office of the Central Taiwan Science Park (CTSP) and Siliconware Precision Industries Co., Ltd. (SPIL).

Appendix A.

Mathematical model 1. Maximization of water reuse

Maximize: F25 + C1 + C2 + A1 + A2
Subject to

- Flow constraints
  1. S1 − F1 − F2 − F3 = 0
  2. F2 − F4 − F5 − F6 − F7 − C1 = 0
  3. C1 − 0.05F2 = 0
  4. C2 − 0.2F2 = 0
  5. F7 − F8 − F9 − F10 − F11 − F12 − F13 = 0
  6. F1 − F27 − V3 = 0
  7. F4 − F14 = 0
  8. F5 − F15 = 0
  9. F6 − F16 = 0
  10. F8 − F17 = 0
  11. F9 − F18 = 0
  12. F10 − F19 = 0
  13. F11 − F20 = 0
  14. F12 + F23 + F32 − F21 = 0
  15. F13 − F22 = 0
  16. F22 − F23 − F24 = 0
  17. F25 − 0.8F24 = 0
  18. F3 + F25 + C1 + A1 + A2 − F28 − F29 = 0
  19. F28 + C5 − F30 − C5 − V1 = 0
  20. F29 + C4 − F31 − C4 − V2 = 0
  21. F27 + F14 + F15 + F16 + F17 + F19 + F20 + F21 + F26 + F30 + F31 − W1 = 0
- Water quality constraints: (COD, NH_3 − N, SS, Cu, Ni, Pb, Zn, B, F)

  1. $a_{27}^{COD} F_{27} + a_{14}^{COD} F_{14} + a_{15}^{COD} F_{15} + a_{16}^{COD} F_{16} + a_{17}^{COD} F_{17} + a_{19}^{COD} F_{19} + a_{20}^{COD} F_{20} + a_{21}^{COD} F_{21} + a_{26}^{COD} F_{26} + a_{30}^{COD} F_{30} + a_{31}^{COD} F_{31} < w^{COD} W_1$
  2. $a_{27}^{SS} F_{27} + a_{14}^{SS} F_{14} + a_{15}^{SS} F_{15} + a_{16}^{SS} F_{16} + a_{17}^{SS} F_{17} + a_{19}^{SS} F_{19} + a_{20}^{SS} F_{20} + a_{21}^{SS} F_{21} + a_{26}^{SS} F_{26} + a_{30}^{SS} F_{30} + a_{31}^{SS} F_{31} < w^{SS} W_1$
  3. $a_{27}^{Cu} F_{27} + a_{14}^{Cu} F_{14} + a_{15}^{Cu} F_{15} + a_{16}^{Cu} F_{16} + a_{17}^{Cu} F_{17} + a_{19}^{Cu} F_{19} + a_{20}^{Cu} F_{20} + a_{21}^{Cu} F_{21} + a_{26}^{Cu} F_{26} + a_{30}^{Cu} F_{30} + a_{31}^{Cu} F_{31} < w^{Cu} W_1$
  4. $a_{27}^{Ni} F_{27} + a_{14}^{Ni} F_{14} + a_{15}^{Ni} F_{15} + a_{16}^{Ni} F_{16} + a_{17}^{Ni} F_{17} + a_{19}^{Ni} F_{19} + a_{20}^{Ni} F_{20} + a_{21}^{Ni} F_{21} + a_{26}^{Ni} F_{26} + a_{30}^{Ni} F_{30} + a_{31}^{Ni} F_{31} < w^{Ni} W_1$
  5. $a_{27}^{Pb} F_{27} + a_{14}^{Pb} F_{14} + a_{15}^{Pb} F_{15} + a_{16}^{Pb} F_{16} + a_{17}^{Pb} F_{17} + a_{19}^{Pb} F_{19} + a_{20}^{Pb} F_{20} + a_{21}^{Pb} F_{21} + a_{26}^{Pb} F_{26} + a_{30}^{Pb} F_{30} + a_{31}^{Pb} F_{31} < w^{Pb} W_1$
  6. $a_{27}^{F} F_{27} + a_{14}^{F} F_{14} + a_{15}^{F} F_{15} + a_{16}^{F} F_{16} + a_{17}^{F} F_{17} + a_{19}^{F} F_{19} + a_{20}^{F} F_{20} + a_{21}^{F} F_{21} + a_{26}^{F} F_{26} + a_{30}^{F} F_{30} + a_{31}^{F} F_{31} < w^{F} W_1$
  7. $a_{27}^{Zn} F_{27} + a_{14}^{Zn} F_{14} + a_{15}^{Zn} F_{15} + a_{16}^{Zn} F_{16} + a_{17}^{Zn} F_{17} + a_{19}^{Zn} F_{19} + a_{20}^{Zn} F_{20} + a_{21}^{Zn} F_{21} + a_{26}^{Zn} F_{26} + a_{30}^{Zn} F_{30} + a_{31}^{Zn} F_{31} < w^{Zn} W_1$
  8. $a_{27}^{B} F_{27} + a_{14}^{B} F_{14} + a_{15}^{B} F_{15} + a_{16}^{B} F_{16} + a_{17}^{B} F_{17} + a_{19}^{B} F_{19} + a_{20}^{B} F_{20} + a_{21}^{B} F_{21} + a_{26}^{B} F_{26} + a_{30}^{B} F_{30} + a_{31}^{B} F_{31} < w^{B} W_1$

Mathematical model 2. Maximization of water reuse with addition of regeneration installations

Maximize: R10 + R14 + C1 + C2 + A1 + A2
Subject to

- Flow constraints
  1. S1 − F1 − F2 − F3 = 0
  2. F2 − F4 − F5 − F6 − F7 − C1 + R10 = 0
  3. C1 − 0.05F2 = 0
  4. C2 − 0.2F2 = 0
  5. F7 − F8 − F9 − F10 − F11 − F12 − F13 = 0
  6. F1 − F27 − V3 = 0
  7. F4 − F14 = 0
  8. F5 − F15 = 0
  9. F6 − F16 = 0
  10. F8 − F17 = 0
  11. F9 − F18 = 0
  12. F10 − F19 = 0
  13. F11 − R1 − R2 = 0
  14. F12 + F32 − R3 − R4 = 0
  15. F13 − R5 − R6 = 0
  16. R7 + R8 + R9 − R10 = 0
  17. R7 − 0.8R1 $\leq$ 0
  18. R8 − 0.8R3 $\leq$ 0
  19. R9 − 0.8R5 $\leq$ 0
  20. R11 + R12 + R13 − R14 = 0
  21. R11 − 0.8R2 $\leq$ 0
  22. R12 − 0.8R4 $\leq$ 0

23. R13 – 0.8R6 ≤ 0
24. F3 + C1 + R14 + A1 + A2 – F28 – F29 = 0
25. F28 + C5 – F30 – C5 – V1 = 0
26. F29 + C4 – F31 – C4 – V2 = 0
27. F27 + F14 + F15 + F16 + F17 + F19 + F30 + F31 + D1 + D2 + D3 + D4 + D5 + D6 – W1 = 0
28. R1 – R7 – D1 = 0
29. R2 – R11 – D2 = 0
30. R3 – R8 – D3 = 0
31. R4 – R12 – D4 = 0
32. R5 – R9 – D5 = 0
33. R6 – R13 – D6 = 0

- Water quality constraints: (COD, NH3-N, SS, Cu, Ni, Pb, Zn, B, F)

1. $a_{27}^{COD}\ F_{27} + a_{14}^{COD}\ F_{14} + a_{15}^{COD}\ F_{15} + a_{16}^{COD}\ F_{16} + a_{17}^{COD}\ F_{17} + a_{19}^{COD}\ F_{19} + a_{20}^{COD}\ F_{20} + a_{21}^{COD}\ F_{21} + a_{26}^{COD}\ F_{26} + a_{30}^{COD}F_{30} + a_{31}^{COD}F_{31} < w^{COD}W_1$
2. $a_{27}^{SS}F_{27} + a_{14}^{SS}F_{14} + a_{15}^{SS}F_{15} + a_{16}^{SS}F_{16} + a_{17}^{SS}F_{17} + a_{19}^{SS}F_{19} + a_{20}^{SS}F_{20} + a_{21}^{SS}F_{21} + a_{26}^{SS}F_{26} + a_{30}^{SS}F_{30} + a_{31}^{SS}F_{31} < w^{SS}W_1$
3. $a_{27}^{Cu}F_{27} + a_{14}^{Cu}F_{14} + a_{15}^{Cu}F_{15} + a_{16}^{Cu}F_{16} + a_{17}^{Cu}F_{17} + a_{19}^{Cu}F_{19} + a_{20}^{Cu}F_{20} + a_{21}^{Cu}F_{21} + a_{26}^{Cu}F_{26} + a_{30}^{Cu}F_{30} + a_{31}^{Cu}F_{31} < w^{Cu}W_1$
4. $a_{27}^{Ni}F_{27} + a_{14}^{Ni}F_{14} + a_{15}^{Ni}F_{15} + a_{16}^{Ni}F_{16} + a_{17}^{Ni}F_{17} + a_{19}^{Ni}F_{19} + a_{20}^{Ni}F_{20} + a_{21}^{Ni}F_{21} + a_{26}^{Ni}F_{26} + a_{30}^{Ni}F_{30} + a_{31}^{Ni}F_{31} < w^{Ni}W_1$
5. $a_{27}^{Pb}F_{27} + a_{14}^{Pb}F_{14} + a_{15}^{Pb}F_{15} + a_{16}^{Pb}F_{16} + a_{17}^{Pb}F_{17} + a_{19}^{Pb}F_{19} + a_{20}^{Pb}F_{20} + a_{21}^{Pb}F_{21} + a_{26}^{Pb}F_{26} + a_{30}^{Pb}F_{30} + a_{31}^{Pb}F_{31} < w^{Pb}W_1$
6. $a_{27}^{F}F_{27} + a_{14}^{F}F_{14} + a_{15}^{F}F_{15} + a_{16}^{F}F_{16} + a_{17}^{F}F_{17} + a_{19}^{F}F_{19} + a_{20}^{F}F_{20} + a_{21}^{F}F_{21} + a_{26}^{F}F_{26} + a_{30}^{F}F_{30} + a_{31}^{F}F_{31} < w^{F}W_1$
7. $a_{27}^{Zn}F_{27} + a_{14}^{Zn}F_{14} + a_{15}^{Zn}F_{15} + a_{16}^{Zn}F_{16} + a_{17}^{Zn}F_{17} + a_{19}^{Zn}F_{19} + a_{20}^{Zn}F_{20} + a_{21}^{Zn}F_{21}+$
8. $a_{26}^{Zn}F_{26} + a_{30}^{Zn}F_{30} + a_{31}^{Zn}F_{31} < w^{Zn}W_1$
9. $a_{27}^{B}F_{27} + a_{14}^{B}F_{14} + a_{15}^{B}F_{15} + a_{16}^{B}F_{16} + a_{17}^{B}F_{17} + a_{19}^{B}F_{19} + a_{20}^{B}F_{20} + a_{21}^{B}F_{21} + a_{26}^{B}F_{26} + a_{30}^{B}F_{30} + a_{31}^{B}F_{31} < w^{B}W_1$

Mathematical model 3. Minimization of total water cost

Minimize: P0*S1 + P1*R11 + P2*R7 + P3*R12 + P4*R8 + P5*R13 + P6*R9

Subject to

- Flow constraints

1. S1 – F1 – F2 – F3 = 0
2. F2 – F4 – F5 – F6 – F7 – C1 + R10 = 0
3. C1 – 0.05F2 = 0
4. C2 – 0.2F2 = 0
5. F7 – F8 – F9 – F10 – F11 – F12 – F13 = 0
6. F1 – F27 – V3 = 0
7. F4 – F14 = 0
8. F5 – F15 = 0
9. F6 – F16 = 0
10. F8 – F17 = 0
11. F9 – F18 = 0
12. F10 – F19 = 0
13. F11 – R1 – R2 = 0
14. F12 + F32 – R3 – R4 = 0
15. F13 – R5 – R6 = 0
16. R7 + R8 + R9 – R10 = 0
17. R7 – 0.8R1 ≤ 0
18. R8 – 0.8R3 ≤ 0
19. R9 – 0.8R5 ≤ 0
20. R11 + R12 + R13 – R14 = 0
21. R11 – 0.8R2 ≤ 0
22. R12 – 0.8R4 ≤ 0
23. R13 – 0.8R6 ≤ 0
24. F3 + C1 + R14 + A1 + A2 – F28 – F29 = 0
25. F28 + C5 – F30 – C5 – V1 = 0
26. F29 + C4 – F31 – C4 – V2 = 0
27. F27 + F14 + F15 + F16 + F17 + F19 + F30 + F31 + D1 + D2 + D3 + D4 + D5 + D6 – W1 = 0
28. R1 – R7 – D1 = 0
29. R2 – R11 – D2 = 0
30. R3 – R8 – D3 = 0
31. R4 – R12 – D4 = 0
32. R5 – R9 – D5 = 0
33. R6 – R13 – D6 = 0

- Water quality constraints: (COD, NH3-N, SS, Cu, Ni, Pb, Zn, B, F)

1. $a_{27}^{COD}\ F_{27} + a_{14}^{COD}\ F_{14} + a_{15}^{COD}\ F_{15} + a_{16}^{COD}\ F_{16} + a_{17}^{COD}\ F_{17} + a_{19}^{COD}\ F_{19} + a_{20}^{COD}\ F_{20} + a_{21}^{COD}\ F_{21} + a_{26}^{COD}\ F_{26} + a_{30}^{COD}\ F_{30} + a_{31}^{COD}\ F_{31} < w^{COD}W_1$
2. $a_{27}^{SS}F_{27} + a_{14}^{SS}F_{14} + a_{15}^{SS}F_{15} + a_{16}^{SS}F_{16} + a_{17}^{SS}F_{17} + a_{19}^{SS}F_{19} + a_{20}^{SS}F_{20} + a_{21}^{SS}F_{21} + a_{26}^{SS}F_{26} + a_{30}^{SS}F_{30} + a_{31}^{SS}F_{31} < w^{SS}W_1$
3. $a_{27}^{Cu}F_{27} + a_{14}^{Cu}F_{14} + a_{15}^{Cu}F_{15} + a_{16}^{Cu}F_{16} + a_{17}^{Cu}F_{17} + a_{19}^{Cu}F_{19} + a_{20}^{Cu}F_{20} + a_{21}^{Cu}F_{21} + a_{26}^{Cu}F_{26} + a_{30}^{Cu}F_{30} + a_{31}^{Cu}F_{31} < w^{Cu}W_1$
4. $a_{27}^{Ni}F_{27} + a_{14}^{Ni}F_{14} + a_{15}^{Ni}F_{15} + a_{16}^{Ni}F_{16} + a_{17}^{Ni}F_{17} + a_{19}^{Ni}F_{19} + a_{20}^{Ni}F_{20} + a_{21}^{Ni}F_{21} + a_{26}^{Ni}F_{26} + a_{30}^{Ni}F_{30} + a_{31}^{Ni}F_{31} < w^{Ni}W_1$
5. $a_{27}^{Pb}F_{27} + a_{14}^{Pb}F_{14} + a_{15}^{Pb}F_{15} + a_{16}^{Pb}F_{16} + a_{17}^{Pb}F_{17} + a_{19}^{Pb}F_{19} + a_{20}^{Pb}F_{20} + a_{21}^{Pb}F_{21} + a_{26}^{Pb}F_{26} + a_{30}^{Pb}F_{30} + a_{31}^{Pb}F_{31} < w^{Pb}W_1$
6. $a_{27}^{F}F_{27} + a_{14}^{F}F_{14} + a_{15}^{F}F_{15} + a_{16}^{F}F_{16} + a_{17}^{F}F_{17} + a_{19}^{F}F_{19} + a_{20}^{F}F_{20} + a_{21}^{F}F_{21} + a_{26}^{F}F_{26} + a_{30}^{F}F_{30} + a_{31}^{F}F_{31} < w^{F}W_1$
7. $a_{27}^{Zn}F_{27} + a_{14}^{Zn}F_{14} + a_{15}^{Zn}F_{15} + a_{16}^{Zn}F_{16} + a_{17}^{Zn}F_{17} + a_{19}^{Zn}F_{19} + a_{20}^{Zn}F_{20} + a_{21}^{Zn}F_{21}+$
8. $a_{26}^{Zn}F_{26} + a_{30}^{Zn}F_{30} + a_{31}^{Zn}F_{31} < w^{Zn}W_1$
9. $a_{27}^{B}F_{27} + a_{14}^{B}F_{14} + a_{15}^{B}F_{15} + a_{16}^{B}F_{16} + a_{17}^{B}F_{17} + a_{19}^{B}F_{19} + a_{20}^{B}F_{20} + a_{21}^{B}F_{21} + a_{26}^{B}F_{26} + a_{30}^{B}F_{30} + a_{31}^{B}F_{31} < w^{B}W_1$

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