CHEMICAL ENGINEERING TRANSACTIONS

VOL. 52, 2016

A publication of AIDIC

The Italian Association
of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Petar Sabev Varbanov, Peng-Yen Liew, Jun-Yow Yong, Jiří Jaromír Klemeš, Hon Loong Lam

ISBN 978-88-95608-42-6; **ISSN** 2283-9216

DOI: 10.3303/CET1652010

A Superstructure Based Optimisation Framework for Batch Water Network Synthesis with Multiple Wastewater Treatment Models

Shuming Wang, Xiong Zou, Hongguang Dong*, Lei Sun

School of Chemical Engineering, Dalian University of Technology, NO. 2 Linggong Road, 116024, Dalian, P.R. China
hgdong@dlut.edu.cn

The existence of numerous water treatment/regeneration technologies capable of increasing the reuse opportunities for water–using network of batch processes calls for a systematic methodology to choose between them. Hence, this article aims to propose a new framework for the synthesis of total water network in batch plants when varieties of treatment units are available. First, a state space superstructure incorporating all feasible configurations for water reuse, recycle and treatment is proposed, which also accounts for the selection of different wastewater treatment technologies. Then, a mixed-integer nonlinear programming (MINLP) model embedded with the scheduling formulation of treatment subsystemis formulated based on the proposed superstructure. Furthermore, trade-off between treatment cost and fresh water consumptionis also taken into consideration in the formulation. In addition, five rules are developed in the pre-processing procedure to eliminate redundant water treatment technologies to simplify the topological structure of the synthesis problem. Two cases are presented to compare different categories of treatment units in total water network and demonstrate the effectiveness of the proposed methodology.

1. Introduction

Increasing water requirement coupled with serious environmental crisis stimulate researchers to address the water network synthesis which aims to minimize the cost as well as exploit the opportunities for water reuse. Over the past decades, numerous methodologies have been developed to synthesize the water network for batch processes, which can be generally classed into two types: insights-based and mathematical optimization-based approaches (Pintarič et al., 2014). Exhaustive overviews of the approaches for water network synthesis can be found in review paper (Gouws et al., 2010). After the synthesis of batch water network through water reuse/recycle had been deeply delved, treatment process was also introduced to further reduce water consumption and wastewater generation. Li et al. (2010) introduced state-time-space superstructure including semi-continuous treatment/regeneration to simultaneously optimize production scheduling and water-allocation network for batch processes. Also, Adekola et al. (2011) considered semi-continuous treatment and achieved wastewater minimization and the scheduling of the batch plants through event-based model at the same time. Furthermore, Liu et al. (2009) synthesized batch water-using network with centralized semi-continuous treatment unit. However, these contributions ignored the investment of water network with different treatment units. Tokos and Pintarič (2009) developed a MINLP model to synthesize the water network in a brewery plant with batch/semi-continuous process. The addition feature of this work is the incorporation of batch/semi-continuous treatment, of which the removal ratio of contaminant and investment are different. But the research ignores the selection of various water treatment units. Cheng and Chang (2007) integrated water-using network and wastewater treatment subsystem for batch processes. However, this work fixed the beginning and end time of both batch and semi-continuous treatment units (CT), neglecting the schedule of these treatment operations. It is noteworthy that as a result of the existence of various treatment technologies, treatment facilities in different operation modes are considered in the aforementioned literatures.

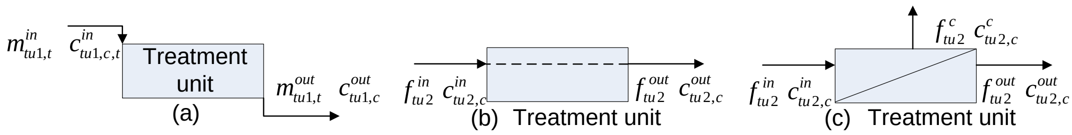

Figure 1: Three categories of regenerators: (a) BT, (b) SCT, (c) TCT

And treatment units can be classified as: batch treatment units(BT) such as membrane bioreactor (MBR) and sequential batch reactor(SBR), single outflow semi-continuous treatment unit(SCT) like adsorption system and two outflows semi-continuous treatment unit(TCT) like reverse osmosis (RO) system (Figure 1). Nevertheless, less attention has been paid to the impact of different categories of treatment units on batch water network as well as the selection of various water treatment units. Accordingly, this article presents a general superstructure of total water network with different treatment units, so as to compare different treatment technologies and determine optimal technologies in the total water network in different cases.

2. Problem statement

The problem addressed in this article is stated as follows: given a specific flowsheet; fresh water source f; a set of water-using tasks $i \in I$ with contaminant $c \in C$; batch treatment units $tu1 \in TU1$ and semi-continuous treatment units $tu2 \in TU2$, before and after which storage tanks $u1/v1$ and $u2/v2$ are installed respectively; end-of-pipe treatment for effluent e . And it aims to achieve a cost-optimal total water network with treatment subsystem for the fixed-load problem of the batch processes. To simplify the formulation, several sets are defined: SR for all storage tanks, SR = $\{sr \mid u1, u2, v1, v2\}$; SO_{tu1} for water sources of batch treatment unit $tu1$, SO_{tu1} = $\{so_{tu1} \mid i, u1, v2\}$; SI_{tu1} for water sinks of batch treatment unit, SI_{tu1} = $\{si_{tu1} \mid i, v1, u2\}$; SO_{tu1} = $\{so_{sr}$ | water sources of storage tank$\}$; SI_{sr} = $\{si_{sr}$ | water sinks of storage tank$\}$; TU = TU1 $\cup$ TU2 = $\{tu$ | treatment units$\}$; T = $\{t$ | boundary points of identical time intervals achieved by dividing the time horizon$\}$. It should be noted that the capital investment for water network will be taken into consideration in the objective function. The followings are the hypotheses for the synthesis problem.

(1) Production schedule is predetermined and water-using processes operate in truly batch mode.

(2) CTs with flowrate controlled to aspecific range are available immediately, while batch treatment units with featured treatment capacity are only available when the treatment is fully completed.

(3) A pair of storage tanks is assigned, before/after the treatment unit, and stream must go through storage tanks when it is purified in the CT, while there exist direct connections between BT and water-using operations.

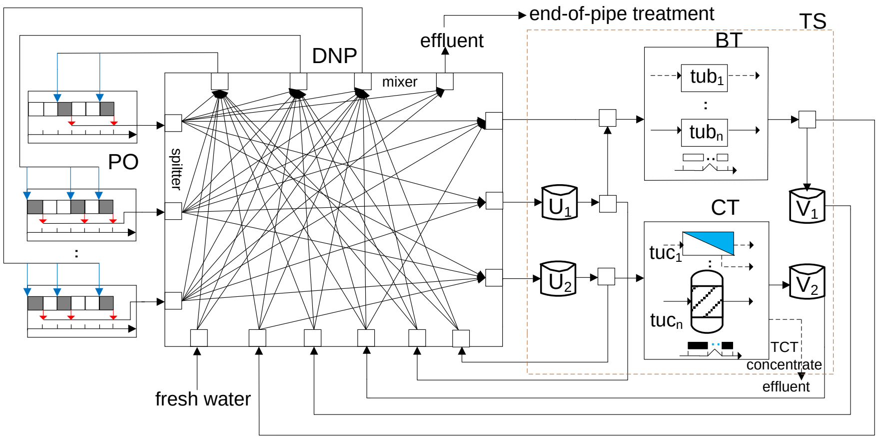

Figure2: State Space superstructure for batch water network

3. Superstructure

To design water network for batch process, a novel superstructure is proposed based on state-time-space superstructure (STS) (Li et al., 2010), as shown in Figure 2, which was consisted by process operator (PO), distribution network of water-using process(DNP) and treatment subsystem(TS). PO represents water using process in truly-batch pattern wherein water is supplied from mixers of DNP at the start and wastewater is discharged to splitters of DNP at the end of its operation. Wastewater from DNP is intercepted by RS, which includes batch treatment units (BT), semi-continuous treatment units (CT), storage tanks installed for treatment. And the superstructure also accounts for the selection of treatment technologies. According to the proposed superstructure, total water network can be synthesized with optimal treatment technologies.

4. Mathematical model

For water-using processes in PO, mass balances around the inlet/outlet and overall balances can refer to the constraints in water reuse/recycle module in the literature of Chen et al. (2011).

4.1 Constraints in TS

Resources balance of batch treatment unit:

$$R_{tu1,t} = \mathrm{R0}_{tu1}|_{t=1} + R_{tu1,t-1}|_{t>1} + \sum_{\substack{t'\in T \\ t-bt\le t'<t}} y_{tu1,t',t} - \sum_{\substack{t'\in T \\ t<t'\le t+bt}} y_{tu1,t,t'} \quad tu1\in \mathrm{TU1}, t\in \mathrm{T} \tag{1}$$

where $\mathrm{R}_{tu1,t}$ is the resource of $\mathrm{tu1}$ at time point t ; $\mathrm{y}_{tu1,t,t'}$ is a binary variable that denotes the usage of $\mathrm{tu1}$.

Wastewater mass and contaminant balances around inlet and outlet ofbatch treatment unit:

$$m^{in}_{tu1,t} = \sum_{so_{tu1}\in SO_{tu1}} m_{so_{tu1},tu1,t} \quad \forall tu1\in \mathrm{TU1}, t\in \mathrm{T} \tag{2}$$

$$m^{out}_{tu1,t} = \sum_{si_{tu1}\in SI_{tu1}} m_{tu1,si_{tu1},t} \quad \forall tu1\in \mathrm{TU1}, t\in \mathrm{T} \tag{3}$$

$$m^{in}_{tu1,t} c^{in}_{tu1,c,t} = \sum_{so_{tu1}\in SO_{tu1}} m_{so_{tu1},tu1,t} c^{out}_{so_{tu1},c,t} \quad \forall tu1\in \mathrm{TU1}, c\in \mathrm{C}, t\in \mathrm{T} \tag{4}$$

where $\mathrm{m}^{in}_{tu1,t}$ / $\mathrm{m}^{out}_{tu1,t}$ are the amount of inlet/outlet water in $\mathrm{tu1}$, $\mathrm{m}_{so_{tu1},tu1,t}$ amount of water flow from water source so_{tu1} to $\mathrm{tu1}$, $\mathrm{m}_{tu1,si_{tu1},t}$ amount of water flow from $\mathrm{tu1}$ to water sink si_{tu1}; $\mathrm{c}^{in}_{tu1,c,t}$, $\mathrm{c}^{out}_{so_{tu1},c,t}$ denote inlet and outlet concentration of contaminant c in $\mathrm{tu1}$ and so_{tu1}.

Overall water and contaminant balances for batch treatment unit:

$$m^{out}_{tu1,t'} - \mathrm{cap}^{max}_{tu1}(1-y_{tu1,t,t'}) \le \alpha_{tu1} m^{in}_{tu1,t} \le m^{out}_{tu1,t'} + \mathrm{cap}^{max}_{tu1}(1-y_{tu1,t,t'}) \quad \forall tu1\in \mathrm{TU1}, t,t'\in \mathrm{T}, t<t'\le t+\mathrm{bt} \tag{5}$$

$$c^{out}_{tu1,c,t'} - \mathrm{c}^{max}_{tu1,c}(1-y_{tu1,t,t'}) \le \gamma_{tu1,c} c^{in}_{tu1,c,t} \le c^{out}_{tu1,c,t'} + \mathrm{c}^{max}_{tu1,c}(1-y_{tu1,t,t'}) \quad \forall tu1\in \mathrm{TU1}, t,t'\in \mathrm{T}, t<t'\le t+\mathrm{bt} \tag{6}$$

Where: $\mathrm{c}^{out}_{tu,c,t}$ represents contaminant concentration in $\mathrm{tu1}$ at time point t ; cap^{min}_{tu1} / cap^{max}_{tu1} ,minimum /maximum amount of wastewater purified by $\mathrm{tu1}$, $\mathrm{c}^{max}_{tu1,c}$ maximum concentration of contaminant c in $\mathrm{tu1}$, $\gamma_{tu1,c}$ removal ratio for contaminant c , α_{tu1} recovery ratio of water in $\mathrm{tu1}$.

Water and contaminant balance in semi-continuous treatment unit:

$$\alpha_{tu2} f^{in}_{tu2} = f^{out}_{tu2} \quad \forall tu2\in \mathrm{TU2} \tag{7}$$

$$\gamma_{tu2,c} c^{out}_{u2,c,t} = \gamma_{tu2,c} c^{in}_{tu2,c,t} = c^{out}_{tu2,c,t} \quad \forall tu2\in \mathrm{TU2},, c\in \mathrm{C}, t\in \mathrm{T} \tag{8}$$

Where: f^{in}_{tu2} / f^{out}_{tu2} are inlet/outlet flow rate, $\mathrm{c}^{in}_{tu2,c,t}$ / $\mathrm{c}^{out}_{tu2,c,t}$ inlet/outlet concentration, $\mathrm{mu}_{tu2,t,t+1}$ amount of wastewater purified in $\mathrm{tu2}$, $\gamma_{tu2,c}$ removal ratio for contaminant, α_{tu2} recovery ratio of water. For semi-continuous treatment unit with fixed outlet concentration $\mathrm{C}^{fx,out}_{tu2,c}$: $\mathrm{c}^{out}_{tu2,c,t} = \mathrm{C}^{fx,out}_{tu2,c}$

$$\int_{T_t}^{T_{t+1}} f^{in}_{tu2} dt - \int_{T_t}^{T_{t+1}} \mathrm{F}^{max}_{tu2} dt (1-ux_{tu2,t,t+1}) \le mu_{tu2,t,t+1} \le \int_{T_t}^{T_{t+1}} f^{in}_{tu2} dt + \int_{T_t}^{T_{t+1}} \mathrm{F}^{max}_{tu2} dt (1-ux_{tu2,t,t+1}) \tag{9}$$

where: T_t time at time point t , $\mathrm{ux}_{tu2,t,t+1}$ binary variable denoting the usage of $\mathrm{tu2}$, F^{max}_{tu2} maximum flow rate. Installation of the treatment units:

$$\chi_{tu1} \geq \frac{1}{\text{nmax}} \sum_{t \in T} \sum_{\substack{t' \in T \\ t < t' \leq t+bt}} y_{tu1,t,t'} \quad \forall tu1 \in \text{TU1} \qquad \chi_{tu2} \geq \frac{1}{\text{nmax}} \sum_{t < T0} ux_{tu2,t,t+1} \quad \forall tu2 \in \text{TU2} \tag{10}$$

where χ_{tu1}, χ_{tu2} denote binary variables that denote the selection of treatment unit.

The mass balances around specific storage tank $sr \in \text{SR}$ in TS:

$$q_{sr,t} = q_{sr,t-1} + m_{sr,t}^{in}\Big|_{sr \neq v2} - m_{sr,t}^{out} - \sum_{tu2 \in TU2} mu_{tu2,t-1,t}\Big|_{sr=u2} + \sum_{tu2 \in TU2} \alpha_{\text{tu2}} mu_{tu2,t-1,t}\Big|_{sr=v2} \ \forall sr \in \text{SR}, t \in \text{T}, t > \text{t1} \tag{11}$$

$$\begin{aligned} q_{sr,t} c_{sr,c,t}^{out} = {} & q_{sr,t-1} c_{sr,c,t-1}^{out} + m_{sr,t}^{in} c_{sr,c,t}^{in}\Big|_{sr \neq v2} - m_{sr,t}^{out} c_{sr,c,t}^{out} - \sum_{tu2 \in TU2} mu_{tu2,t-1,t} c_{sr,c,t}^{out}\Big|_{sr=u2} \\ & + \sum_{tu2 \in TU2} \alpha_{\text{tu2}} mu_{tu2,t-1,t} c_{tu2,c,t-1}^{out}\Big|_{sr=v2} \qquad \forall sr \in \text{SR}, c \in \text{C}, t \in \text{T}, t > \text{t1} \end{aligned} \tag{12}$$

$$m_{sr,t}^{in} = \sum_{so_{sr} \in S_{sr}} m_{so_{sr},sr,t} \quad sr \in \text{SR}, sr \neq v2, t \in \text{T} \tag{13}$$

$$m_{sr,t}^{in} c_{sr,c,t}^{in} = \sum_{so_{sr} \in S_{sr}} m_{so_{sr},sr,t} c_{so_{sr},c,t}^{out} \quad sr \in \text{SR}, c \in \text{C}, t \in \text{T} \tag{14}$$

$$m_{sr,t}^{out} = \sum_{si_{sr} \in SI_{sr}} m_{sr,si_{sr},t} \quad sr \in \text{SR}, t \in \text{T} \tag{15}$$

where: $q_{sr,t}$ represents the amount of water stored, $m_{sr,t}^{in}$ / $m_{sr,t}^{out}$ amount of inlet/outlet water, $c_{sr,c,t}^{in}$ / $c_{sr,c,t}^{out}$ inlet/outlet concentration of contaminant c in sr, $m_{so_{sr},sr,t}$ amount of water flow from water source so_{sr} to sr, $m_{sr,si_{sr},t}$ amount of water flow from sr to water sink si_{sr}, $c_{so_{sr},c,t}^{out}$ outlet concentration of contaminant c in so_{sr}.

4.2 Objective function

$$\begin{aligned} Cost = {} & [\sum_{t \in T} \sum_{i \in I} m_{f,i,t} \text{cf} + \sum_{t \in T} \sum_{i \in I} m_{i,e,t} \text{ce}]\ \text{NTC} + \sum_{tu \in TU} \chi_{tu} \text{IC}_{tu} + \sum_{tu1 \in TU1} \sum_{t \in T} \sum_{\substack{t' \in T \\ t < t' \leq t+bt}} y_{tu1,t,t'} \text{mav}_{tu1} \text{OC}_{tu1} \text{NTC} \\ & + \sum_{sr} \text{ICS}_{sr} n_{sr} + \sum_{tu2 \in TU2} \sum_{t < T0} f_{\text{tu2}}^{\text{in}} (\text{T}_{t+1} - \text{T}_t) \text{OC}_{tu2} \text{NTC} + \sum_{t < T0} (1\text{-}\alpha_{tu2})(\text{T}_{t+1} - \text{T}_t) f_{tu2}^{\text{in}} \text{ce NTC} \end{aligned} \tag{16}$$

where: $Cost$ is total annualized cost, n_{sr} represents the installation of storage tank, cf/ce fresh water/end-of-pipe treatment cost, IC_{tu} / OC_{tu} installation/operational cost, ICS_{sr} annualized investment of storage tank; NTC is total number of batch cycle every year.

5. Rules in pre-processing procedure

$$\text{Cp}_{tu1} = \frac{\text{IC}_{tu1} \Delta t}{\text{H} \cdot \text{cap}_{\text{tu1}}^{\text{max}} \cdot \text{NTC}} + \text{OC}_{tu1} \quad \forall tu1 \in \text{TU1} \qquad \text{Cp}_{tu2} = \frac{\text{IC}_{tu2}}{\alpha_{\text{tu2}} \cdot \text{F}_{\text{tu2}}^{\text{max}} \cdot \text{H} \cdot \text{NTC}} + \text{OC}_{tu2} \quad \forall tu2 \in \text{TU2} \tag{17}$$

To reduce the model complexity, five rules in the pre-processing procedure for the selection of proper technologies are developed when various treatment technologies are available, shown as the following.

Rule 1: If one contaminant can only be purified by a certain treatment technology, choose this technology.

Table 1: Comparison results for case I

Treatment units	BT(Δt = 1 h)	BT(Δt = 0.5 h)	SCT	TCT(α = 0.7)	BT & SCT
Fresh water consumption(t/cycle)	71.594	68.594	68.594	71.266	63.03

Rule 2: For CT/BT, treatment units with less costs for treated water per hour Cp_{tu} ($\$/(t \cdot h)$) (shown as Eq(17)) would be preferred when the contaminant concentrations of their outlet flows are identical.

Rule 3: For CT/BT, if the costs for treated water per hour Cp_{tu} ($\$/(t \cdot h)$) are equal, treatment units with lower outlet concentration would be preferred.

Rule 4: For BT, when their unit costs Cp_{tu1} and outlet concentrations are equivalent, choose the facility with shorter operation duration of single batch.

Table 2: Process parameters for TU in the RS of the example

Technologies		Outlet concentration	Water recovery ratio	Installation cost ($/y)	Operation cost ($/t)	Operation capacity	Duration (h)
TU1	Situ reactor	150	1	5,000	0.1	[5,12]t	1
	SBR	70	1	15,000	0.15	[7.5,18]t	1.5
	MBR	70	1	10,000	0.2	[5,12]t	1
TU2	RO	20	0.7	23,000	0.6	[5,12]t/h	-
	Ultrafiltration	50	0.7	14,000	0.5	[5,12]t/h	-
	Adsorption	50	1	15,000	0.4	[5,12]t/h	-

Rule 5: If unit cost of $BTCp_{tu1}$ is consistent with the counterpart of $CTCp_{tu2}$ and their outlet concentrations for contaminant are controlled to a similar range, the latter is preferentially selected to remove contaminant due to that the scheduling of batch treatment unit may limit the opportunities for further water reuse.

6. Example study

An illustrative example from Liu et al. (2009) is introduced, which consists of five water-using processes merely involving single contaminant. Maximum capacity of storage tanks are determined as 70 t. Limiting data for water-using processes is taken from scenario 2 in the work of Liu et al. (2009). Two cases will be discussed to illustrate the proposed method for the synthesis of total network. The problem is solved by DICOPT in GAMS 23.4, using CPLEX as the MILP solver and CONOPT as the NLP solver.

6.1 Case I: Comparison of different types of treatment units

Irrespective of economic cost for treatment, this case is intended to explore the discrepancies of different categories of treatment units that are incorporated into the total water network, as well as verify rules in the pre-processing procedure. Maximum inlet flow rates of both SCT and TCT are set to be 11.875 t/h, and the capacity of BT is 11.875 t/batch. The duration of BT is defined as one hour. Outlet concentration of the all the treatment units are set to be 100 μg /g, and the recovery ratio of TCT is determined as 0.7.

The results are shown in Table 1. Optimal fresh water requirement of 68.594 t/cycle is attained when only SCT is involved in the water network, the same as the result in the paper of Liu et al. (2009). Note that fresh water consumption for batch water network involving BT& SCT is the lowest, as a result of greater capacity of this treatment. SCT has advantage over BT (Δt = 1 h) in fresh water consumption with identical average treatment capacities, which tests the pre-processing rule 5. Similarly, BT (Δt = 0.5 h) is superior to BT (Δt = 1 h) in fresh water consumption while their average treatment capacities are equal, verifying the pre-processing rule 4. Since rules1, 2 and 3 are apparent, it is unnecessary to validate them in this paper.

6.2 Case II: Selection of proper treatment technologies

This case aims to apply the proposed methodology to determine proper treatment units and optimize the total water network. Economic parameters for available treatment units are presented in Table 2. The annual cost for each storage tank is assumed to be 1,000 $/y. Process parameters for treatment units are shown in Table 2, and costs of fresh water and wastewater treatment are 1 $/t and 5 $/t, respectively. Total number of batch cycles every year is perceived as 1,000. To decrease computational efforts, pre-processing rules are exploited to eliminate redundant treatment technologies before solving the problem.

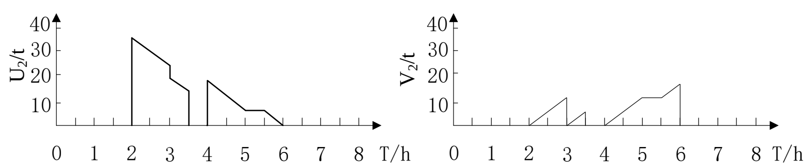

Figure 3: Storage profiles of the storage tanks

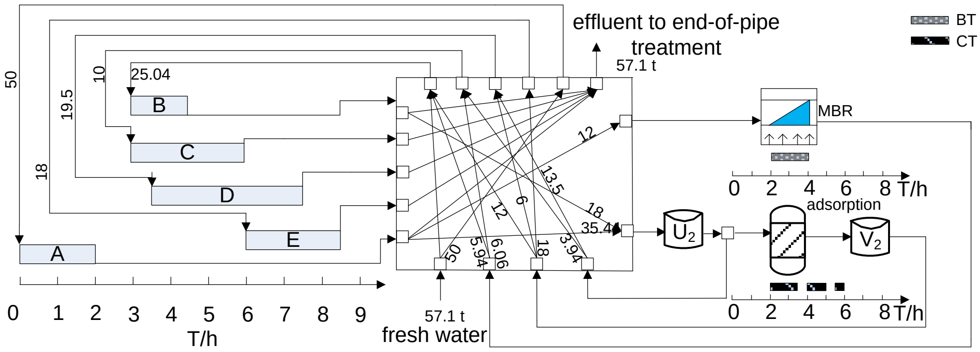

Figure 4: Resulting total water network for the example

According to the pre-processing rules, ultrafiltration and SBR are firstly removed from the synthesis framework. Then, the other four types of technologies are incorporated into the superstructure, based on which the problem can be efficiently solved. The total annual cost of 386,400 \$/y and fresh water consumption of 57,100 t/y for the total water network are achieved, which corresponds to a 20 % reduction in annualized investment cost and a 29.07 % reduction in fresh water consumption as compared to the water-using network with central storage tank. And compared with water network with adsorption, it yieldsa 9.3 % reduction in annual cost and a 14.8 % reduction in freshwater consumption. The total water network structure is depicted as Figure 4, indicating MBR and adsorption are selected for treatment. Also, the variations of the amount of residual water in storage tank u2 and v2 are depicted in Figure 3. In addition, it can be noted that storage tank u1 and v1 designated as buffer vessels for BT are not installed, which can decrease the investment cost of storage tanks.

7. Conclusions

This article introduces a superstructure to incorporate varieties of treatment facilities (BT, SCT/TCT) into the optimization framework of batch water network. On the basis of the proposed superstructure, a MINLP model is formulated which consists of the scheduling formulation of batch treatment units. Then, five heuristic rules are developed to eliminate redundant treatment technologies, which can simplify the topological structure of the synthesis problem. Two cases are introduced todemonstrate the application of the proposed method. Form case I, pre-processing rule 4 and 5 are clearly verified, while the result of case II illustrates that the proposed approach can achieve a good trade-off between treatment cost and fresh water consumption.

Acknowledgments

This work is supported by the National Natural Science Foundation of China (No.21276039).

Reference

Adekola O., Majozi T., 2011, Wastewater minimization in multipurpose batch plants with a treatment unit: Multiple contaminants, Computers & Chemical Engineering, 35(12), 2824-2836.

Chen C.L., Chang C.Y., Lee J.Y., 2010, Resource-task network approach to simultaneous scheduling and water minimization of batch plants, Industrial & Engineering Chemistry Research, 50(7), 3660-3674.

ChengK.F., ChangC.T., 2007, Integrated water network designs for batch processes, Industrial & Engineering Chemistry Research, 46,1241-1253.

GouwsJ.F., Majozi T., Foo D.C.Y., Chen C.L., Lee J.Y., 2010, Water minimization techniques for batch processes, Industrial & Engineering Chemistry Research, 49(19), 8877-8893.

Li L.J., Zhou R.J., Dong H.G., 2010, State− time− space superstructure-based MINLP formulation for batch water-allocation network design, Industrial & Engineering Chemistry Research, 49(1), 236-251.

Liu Y.Z., Li G.H., Wang L., Zhang J.Z., Shams K, 2009, Optimal design of an integrated discontinuous water-using network coordinating with a central continuous treatment unit, Industrial & Engineering Chemistry Research, 48(24), 10924-10940.

Pintarič Z.N., Ibrić N., Ahmetović E., Grossmann I.E., Kravanja Z., 2014, Designing optimal water networks for the appropriate economic criteria, Chemical Engineering Transactions, 39, 1021-1026.

Tokos H., Pintarič Z.N., 2009, Synthesis of batch water network for a brewery plant, Journal of Cleaner Production, 17(16), 1465-1479.