An Iterative Method for Design of Water-Using Networks with Regeneration Recycling

Chunhui Pan
School of Chemical Engineering, Hebei University of Technology, Tianjin 300130, China

Dept. of Chemistry, Cangzhou Teachers' College, Hebei 061001, China

Jing Shi and Zhi-Yong Liu
School of Chemical Engineering, Hebei University of Technology, Tianjin 300130, China

DOI 10.1002/aic.12595
Published online March 22, 2011 in Wiley Online Library (wileyonlinelibrary.com).

Both freshwater consumption and wastewater discharge can be reduced significantly when wastewater regeneration recycling is introduced in the design of the water-using networks (WUNs). However, it is often difficult to design the WUNs involving regeneration recycling. This article presents an iterative method to design the WUNs involving regeneration recycling. The final designs can be obtained without iteration for the networks with known regenerated concentrations, and in a few iterations for the networks with known removal ratios of the contaminants. The method proposed can reduce the following parameters simultaneously, the consumptions of freshwater and the regenerated water, and the concentrations of the stream before regeneration, and all of them reflect the cost of the network involving regeneration recycling. The results of a few literature examples show that the designs obtained in this work are comparable to that obtained in the literature. The proposed method is simple and effective. © 2011 American Institute of Chemical Engineers *AIChE J,* 58: 456–465, 2012
Keywords: water-using network, regeneration recycling, iterative method, wastewater minimization

Introduction

Water is one of the most important resources and widely used in industries. Being one of the most efficient technologies for saving freshwater and reducing wastewater discharge, water-using system integration has become one of the research focuses recently. In water-using network design, there are three operations to minimize wastewater: wastewater reuse, regeneration reuse, and regeneration recycle. The water-using networks (WUNs) involving regeneration recycling can reduce both freshwater consumption and wastewater discharge significantly. Zero wastewater discharge might be achieved if the regeneration concentrations are low enough. Therefore, it is very important to research the design of the networks involving regeneration recycling.

In the networks involving *wastewater regeneration*, the contaminants in wastewater streams are partially or totally removed with any purification technique(s). There are two types of WUNs involving regeneration: wastewater *regeneration reuse* and *regeneration recycling*. If the regenerated water is reused in the process in which it is produced, it is called *regeneration recycling*; if the regenerated water is reused in the other water-using processes, it is called *regeneration reuse*. In the literature, the regeneration units are modeled as the fixed outlet concentration type and removal ratio (*RR*) type (Wang and Smith,[1] and Kuo and Smith[2]). In the models of the *RR* type, the regenerated concentration is calculated from the *RR* value(s) of a treatment unit as follows:

Correspondence concerning this article should be addressed to Z.-Y. Liu at liuzhiyong@hebut.edu.cn.

$$RR = \frac{f_{in}C_{in} - f_{out}C_{out}}{f_{in}C_{in}} \quad (1)$$

Many methods have been proposed for the design of the WUNs involving regeneration reuse/recycling, such as water pinch analysis methods, mathematical programming methods, and heuristic methods. Wang and Smith[1] presented a graphic method for the design of WUNs with reuse and regeneration reuse based on the water pinch analysis. They considered that the regenerated concentration was the same as the pinch concentration. Kuo and Smith[2] proposed an improved method based on the work of Wang and Smith.[1] We discuss the method of Kuo and Smith[2] later in detail. Mann and Liu[3] addressed that the regeneration concentration might be above the freshwater pinch. Hallale[4] investigated the problem of the placement of the regeneration unit by using pinch-based techniques. Feng et al.[5] proposed a new method to construct the optimal water supply line for regeneration recycling by analyzing the limiting composite curve of a single contaminant water system, from which the targets for the minimum freshwater consumption, the optimal regeneration concentration, and the minimum regeneration flowrate were considered. Castro et al.[6] considered the regeneration reuse for the WUNs with multiple pinch points. Gomes et al.[7] presented optimal solutions for reuse, multiple water sources, regeneration reuse and regeneration recycling for the WUNs of single contaminant. Deng et al.[8] investigated the optimal regeneration concentration and regenerated water flowrate for single contaminant water system. Ng et al.[9,10] also investigated the target of the WUNs of single contaminant with regeneration. Bandyopadhyay and Cormos[11] proposed an approach named source composite curve to target the regeneration flowrate. In the researches discussed above, most of them can only be used to deal with the problems of the fixed outlet regenerated concentration. The method of Kuo and Smith[2] and the work of Bandyopadhyay and Cormos[11] can be used for the problems of both the fixed outlet regenerated concentration and those of the *RR*. In the methods mentioned above, most of them can only be used for single contaminant networks. The work of Kuo and Smith[2] can deal with the networks of multiple contaminants involving regeneration.

Mathematical programming methods are very important in synthesizing the WUNs with multiple contaminants. Takama et al.[12] investigated the water allocation problems involving regeneration by using superstructure coupled with mathematical programming method. Alva-Argaez et al.[13] used superstructure-based approaches to design the WUNs involving wastewater reuse, regeneration reuse and regeneration recycling. Huang et al.[14] established an NLP method for the design of the water allocation networks. Gunaratnam et al.[15] used nonlinear mathematical programming methods to obtain the targets for the WUNs. Relvas et al.[16] developed a software named AquoMin to design and target the WUNs involving regeneration recycling/reuse. Koppol et al.[17] presented a mathematical programming approach to analyze the feasibility of zero liquid discharge option in different industries. Cao et al.[18] and Rivas et al.[19] independently developed design methodologies for the WUNs with water mains including regeneration recycle. The models generalized the methodology of Feng and Seider[20] for designing of WUNs with multiple contaminants. Feng et al.[21] utilized mathematical programming coupled with superstructure to optimize regeneration recycling water networks for grass-roots design.

Now, we discuss the work of Kuo and Smith[2] in detail. In their work, the network was decomposed into two subgroups: operations entirely below the freshwater pinch belonged with Group 1, and the processes in Group 1 were fed by freshwater. Operations across or above the freshwater pinch belonged to Group 2, and the processes in Group 2 were fed by the regenerated water. The two groups were designed separately. Operation migrations between the two groups were carried out based on the two mechanisms and seven criteria proposed by Kuo and Smith[2] to find the final design for the WUNs with regeneration.

Liu et al.[22] proposed a new method to design the WUNs involving regeneration reuse. They divided the whole network into two parts: the subnetwork before regeneration and that after regeneration, first. By designing of the subnetwork before regeneration, the flowrate and concentration(s) of the regenerated stream were obtained. Then including the regenerated stream in the sources of the network with reuse only to form the network involving regeneration reuse, the design of the networks involving regeneration reuse was obtained by using the method proposed for the networks involving reuse only (Liu et al.[23]). However, Liu et al.[22] did not consider the problem about regeneration recycling. When using the method of Liu et al.[22], the design of the WUNs involving regeneration reuse depends on the division of the network. Therefore, poor division of the network might make it difficult to obtain good design for complex networks.

In this article, we propose an iterative method based on the methodology proposed by Liu et al.[22] to design the WUNs involving regeneration recycling. For the networks with the model of the fixed outlet regeneration concentration, the final designs can be obtained without iteration. For the networks with the model of regeneration *RR*, the final design can be obtained in a few iterations. In the design, the regenerated stream is formed by the source streams, which have high reuse-possibility and are not reused by the demands. The following parameters are considered simultaneously in the design: the consumptions of freshwater and the regenerated water, and the concentrations of the stream before regeneration, which reflect the cost of the networks involving regeneration recycling. The calculation procedure proposed in this article is simple and effective. Another important feature of the proposed method is that it is suitable for computer calculation.

The new method

The method proposed in this article is based on the methodology proposed by Liu et al.[22] Liu et al.[22] pointed out that: compared with the network involving reuse only, there is an additional source stream, the regenerated stream, in the network involving regeneration reuse/recycling, if the design specifications of the network involving reuse only are the same as that involving regeneration reuse/recycling. Therefore, if the flowrate and concentration(s) of the regenerated stream can be obtained, one can include the regenerated stream in the source streams of the network involving reuse

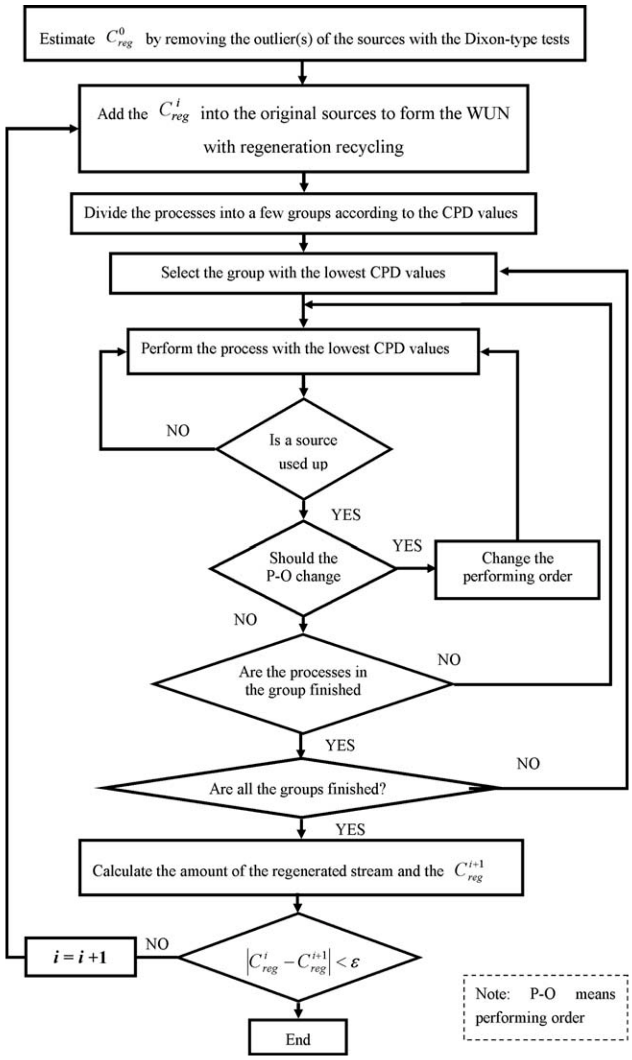

Figure 1. The design procedure proposed.

only to form the network involving regeneration reuse/recycling. The design of the network involving regeneration reuse/recycling can be obtained by using the design method proposed for the network involving reuse only (for example, the method of Liu et al.[23]). However, the flowrate and concentration(s) of the regenerated stream are unknown before detailed design. In order to start the calculation, initial regenerated concentrations should be determined first based on the features of the network and the regeneration process. The flowrate of the regenerated stream for the first iteration is to be determined. After the first iteration, the flowrate of the regenerated stream can be taken as the sum of the regenerated stream reused in the last iteration. The regenerated stream will be formed by the sources which meet the following conditions: (a) not reused by the demands in the design, (b) with high reuse-possibility after regeneration, as will be discussed later. In this article, we use a modified design procedure of Liu et al.,[23] which is based on new concepts, the concentration potentials of demands and sources, to design the water networks. When the differences of the regenerated concentrations for the two adjacent iterations are in the allowable range ε (0.1 ppm in this article), the iteration can be finished. The design procedure is shown in Figure 1. The issues discussed above and that listed in Figure 1 will be discussed in detail in the following sections.

Concepts of the concentration potentials of the source and demand streams

The determination of the concentration order of the streams is often important in the design and target of the WUNs. For a WUN of single contaminant, it is easy to determine the concentration order of the streams, but it is often difficult to determine the concentration order of the streams in a WUN of multiple contaminants. For example, for the two streams with contaminants A, B and C, $S_1 = (40, 60, 20)$ ppm and $S_2 = (30, 40, 70)$ ppm, it is difficult to tell if the concentrations of S_1 are lower than those of S_2 or not, because the concentrations of contaminants A and B in S_1 are higher than those in S_2, but the concentration of contaminant C in S_1 is lower than that in S_2. To determine the concentration order of the streams in a water-using system of multiple contaminants, Liu et al.[23] introduced the concepts of the concentration potentials of the demand (inlet) streams (*CPD*s) and that of the source (outlet) streams (*CPS*s).

The *CPD* value of a demand is a measurement of the overall possibility for the demand to reuse the source streams, as shown in Eq. (2):

$$CPD(D_j) = \sum_{i=1}^{NS} \min_{k=1,2,\ldots,NC} \left(\frac{C_{D_{j,k}}^{\lim}}{C_{S_{i,k}}} \right) \tag{2}$$

where $C_{Dj,k}^{\lim}$ is the limiting concentration of contaminant k in demand D_{j}, $C_{S_{i,k}}$ is the concentration of contaminant k in source S_{i}, NC is the number of the contaminants, and NS is the number of the source streams.

The *CPS* value of a source stream is a measurement of the overall possibility for the source to be reused by the demand streams, as shown in Eq. (3):

$$CPS(S_i) = \frac{1}{\sum_{j=1}^{ND} \min_{k=1,2\ldots,NC} \left[\frac{C_{D_{j,k}}^{\lim}}{C_{S_{i,k}}} \right]} \tag{3}$$

where ND is the number of demand streams.

It should be pointed out that, in the work of Liu et al.,[23] $i \neq j$ was taken as a limiting condition for the *CPD* and *CPS* because recycling was not considered. In this article, the condition $i \neq j$ is not needed any more because regeneration recycling is considered.

The lower the $CPD(D_{\mathrm{j}})$ value of a demand D_{j}, the lower the conventional concentration of the demand stream, as has been proved by Liu et al.[23] In the design procedure,[23] the demand streams with the lowest *CPD* will be satisfied first. The source with the largest limiting quasi-allocation ratio should be reused first. The limiting quasi-allocation ratio of S_{i} for unit amount (say, 1 ton) of D_{j} is shown in Eq. (4):

$$R_{i,j} = \min_{k=1,2\ldots,NC} \left[\frac{C_{D_{j,k}}^{\lim}}{C_{S_{i,k}}} \right] \tag{4}$$

The contaminant(s) determining the value of $R_{\mathrm{i,j}}$ is called as the reuse key contaminant(s) (*RKC*s).

Estimation of the regenerated concentrations

Estimation of the initial regenerated concentrations plays an important role in the design, because good initial regenerated concentrations will reduce iteration times. To obtain good initial values of the regenerated stream, let us analyze the sources in a WUN involving regeneration recycling first. In a WUN involving regeneration recycling, the source(s) with very high reuse-possibility will be reused by the demands in the network; the source(s) with very low reuse-possibility will be discharged; and the source(s) with medium reuse-possibility will form the regeneration stream. Therefore, when estimating initial values of the regenerated concentrations, we should not include the regenerated source(s) with very low reuse-possibility and/or that with very high reuse-possibility. Liu et al.[23] has shown that the *CPS* value of a source stream is a good indicator for its reuse-possibility: the larger the *CPS* value of a source, the lower the overall possibility for the source to be reused by the demands. In this article, we determine the reuse-possibility of a regenerated stream with its *CPS* value. The regenerated source stream(s) with very small or very large *CPS* value will not be included in the regenerated stream.

In this article, the Dixon-type tests will be used to identify the "outlier" of the initial regenerated source(s) which has very low or very high *CPS* values compared to the other ones. The reason that the Dixon-type tests are used is that it does not need to assume normality of the data. The confidence level is taken as 99%. The reader is referred to Ref. 24 for more information about the Dixon-type tests.

The initial regenerated concentration $\boldsymbol{C}^{\lim}_{reg,Si}$ of source stream S_i can be estimated from its limiting concentrations as follows:

$$\boldsymbol{C}^{\lim}_{reg,S_i} = \boldsymbol{C}^{\lim}_{S_i} \times (1 - RR_1, 1 - RR_2, \cdots, 1 - RR_{NC})^{\mathrm{T}} \qquad (5)$$

where RR_j is the *RR* for the regeneration process to remove contaminant *j* as defined in Eq. (1), $\boldsymbol{C}^{\lim}_{Si}$ is the concentration vector of S_i, and $\boldsymbol{C}^{\lim}_{reg,Si}$ is the limiting regenerated concentration vector of S_i.

The *CPS* value of $\boldsymbol{C}^{\lim}_{reg,Si}$ can be obtained from Eq. (3). The Dixon-type tests will be used to identify the "outlier" of the limiting regenerated streams according to their *CPS* values. The non-outlier sources will form the regenerated stream. The concentrations of the initial regenerated stream can be obtained as follows:

$$\boldsymbol{C}^{0}_{reg} = \sum_i \left(F^{\lim}_{S_i} \boldsymbol{C}^{\lim}_{reg,S_i} \right) \Big/ \sum_i F^{\lim}_{S_i} \qquad (i \in NOS) \qquad (6)$$

where $\boldsymbol{C}^{0}_{reg}$ is the initial concentration vector of the regenerated stream, $F^{\lim}_{Si}$ is the limiting flowrate of S_i, and *NOS* is the set of the non-outlier sources.

It should be noted that in the first iteration, the flowrate of the regenerated stream is unknown. After the first iteration, the flowrate of the regenerated stream will be taken as the sum of the regenerated streams which are reused in the last iteration. The regenerated stream will be formed by the unreused sources in ascending order of their *CPS* values after regeneration. We will show the calculation of the regenerated stream in Example 1.

The design of the networks

When the initial regenerated stream, which has known concentrations and sufficient amount, is added in the original network, the initial network involving regeneration recycling is formed. The initial network involving regeneration recycling can then be designed by using a modified method of Liu et al.[22,23] as discussed in the following:

(1) Include the regenerated stream in the sources of the original network to form the network involving regeneration recycling;

(2) The processes are divided into a few groups according to their *CPD* values. The classification of the groups will start from the lowest *CPD* values. In group G_i, if $CPD_{Gi,U}/CPD_{Gi,L} \leq EV$, processes *U* will be considered in group G_i, where $CPD_{Gi,L}$ is the lowest *CPD* value in group G_i, and the value of *EV* is taken as 1.2 in this article (It should be pointed out that *EV* can take other values as well. If the value of *EV* is 1, the processes will not be divided into groups; if the value of *EV* is taken as ∞,all the processes will be in one group and this will increase the calculation effort significantly). The smaller of the *CPD* values of the group, the earlier the processes in the group will be performed. Freshwater is often used for the group with the smallest *CPD* values;

(3) Generally speaking, for the processes in the same group, the process with the smaller *CPD* value will be performed first. However, if one source stream is used up, and the unperformed process in the group is in favor of reusing the source, the performing order can be challenged. If there are a few processes with the same inlet *CPD* value, the process with the lowest *CPS* value will be performed first, because the outlet stream with the lowest *CPS* value has the highest reuse-possibility for the downstream processes;

(4) If a source stream is used up, it will not be considered in the following steps;

(5) Return to steps (2–4), until all the processes are performed.

The procedure of allocating the source streams to the demand streams is as follows:

(a) The usage-priority order of the water streams is: the internal sources, the regenerated source, and freshwater, because the cost order is often as follows: freshwater > the regenerated stream > the internal streams;

(b) If the *RKC* and *FKC* (*FKC* is the freshwater key contaminant which determines the minimum freshwater amount when freshwater is used only[23]) are the same contaminant, and if the limiting outlet concentration of the key contaminant of the process is lower than that of the source, the source will not be reused. Instead, freshwater is used solely. Otherwise, allocate the sources to the demand, and go to step (c);

(c) The internal source with the largest limiting quasi-allocation ratio (see Eq. 4) will be reused first. If the limiting quasi-allocation ratios of a few internal sources are the same, the source with the highest *CPS* value will be used first to reduce the consumption of freshwater or the regenerated water for the downstream processes;

(d) When satisfying a demand stream, if the limiting quasi-allocation ratio of a source (say, S_1) is smaller than unity, and the quasi-allocation ratio of the *RKC* of another

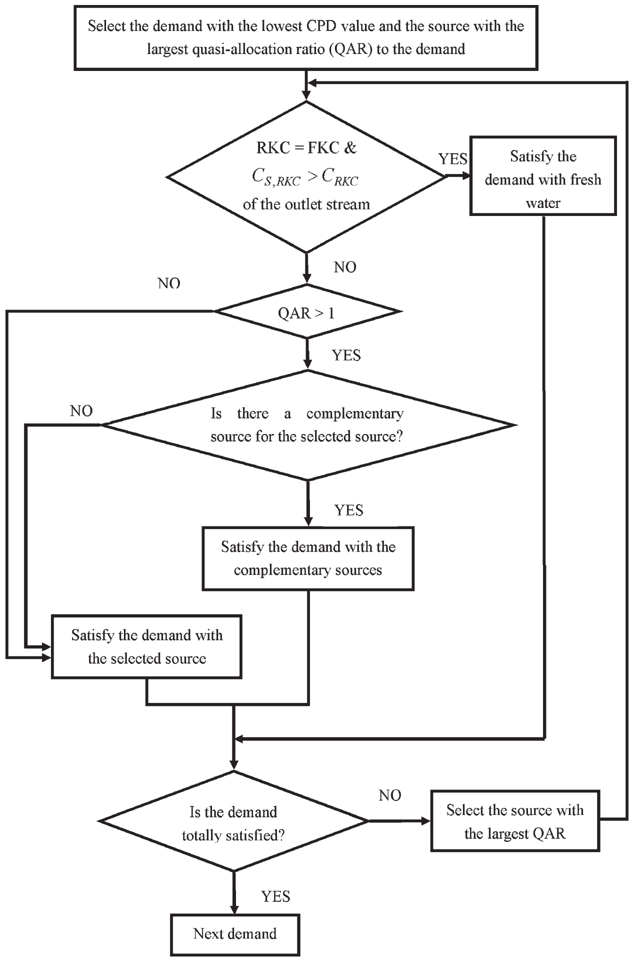

Figure 2. Allocating procedure from the sources to the demands.

source (say, S_2) is larger than unity, the two sources are complementary. If the two complementary sources are reused simultaneously to satisfy the demand, the total reusing amount will often be increased, and the consumption of freshwater or the regenerated water will be reduced. The amounts of the sources can be obtained based on mass balance of the key contaminant and the flowrate of the process;

Table 1. The Limiting Data for Example 1 From Kuo and Smith[2]

Process	Contaminant	C_{in}^{lim} (ppm)	C_{out}^{lim} (ppm)	F^{lim} (t/h)
1	A	0	15	
	B	0	400	50
	C	0	35	
2	A	20	120	
	B	300	12,500	34
	C	45	180	
3	A	120	220	
	B	20	45	56
	C	200	9500	
4	A	0	20	
	B	0	60	8
	C	0	20	
5	A	50	150	
	B	400	8000	8
	C	60	120	

Table 2. The Initial Concentrations of the Regenerated Streams and Their *CPS* Values in Example 1

	$C_{reg,Si}^{lim}$ (ppm)				
Streams	A	B	C	*CPS*	To be Regenerated?
$S_{reg,S1}^{lim}$	15	0.4	35	0.11	Yes
$S_{reg,S4}^{lim}$	20	0.06	20	0.11	Yes
$S_{reg,S2}^{lim}$	120	12.5	180	0.67	Yes
$S_{reg,S5}^{lim}$	150	8	120	0.79	Yes
$S_{reg,S3}^{lim}$	220	0.05	9500	31.15	No

(e) If a demand is not totally satisfied, select the next source by using the similar procedure as mentioned in steps (a) to (c), until the demand is totally satisfied.

It should be pointed out that in the design procedure, the *CPD* values should be calculated based on the current available sources, which include the outlet streams of the performed processes and the regenerated stream. The allocation of the sources to the demands is shown in Figure 2.

Case Studies

Example 1

This example is taken from Kuo and Smith,[2] with the data shown in Table 1. The *RR* values for the regeneration process to remove contaminants *A*, *B*, *C* are (0, 99.9%, 0), respectively.

In the first iteration, the initial regenerated concentrations should be estimated first. The limiting regenerated concentrations for source S_i, $C_{reg,Si}^{lim}$, can be calculated with Eq. (5). The *CPS* values of the limiting regenerated streams for the sources are listed in Table 2, which can be calculated with Eq. (3), where $C_{Dj,k}^{lim}$s are the limiting inlet concentrations in Table 1, and the $C_{S_i,k}$s are the regenerated concentrations listed in Table 2 and the limiting outlet concentrations in Table 1. By using the Dixon-type test, it is found that the source with the largest *CPS* value of 31.15 (for $S_{reg,S3}^{lim}$) is an outlier. The calculation of the Dixon-type test is given in the Appendix. The other sources, S_1, S_2, S_4, S_5, will be mixed and regenerated. The initial regenerated concentrations are (61.90, 5.09, 89.90) ppm.

The procedure for the first iteration is as follows: Arrange the processes in ascending order of the *CPD* values, which are calculated from the limiting concentrations of all the source streams (including the original sources S_1, S_2, S_3, S_4, S_5 and the initial regenerated stream). The processes with the lowest limiting inlet concentration potentials, P_1 and P_4, will use freshwater solely, and the freshwater consumption amounts are 50 t/h and 8 t/h, respectively.

Table 3. The Quasi-Allocation Ratios for the Streams of S_1, S_4, and the Initial Regenerated Stream to Satisfy D_2

Contaminant	S_1	S_4	S_{reg}^0
A	1.33	1.00	0.32
B	0.75	5.00	58.88
C	1.29	2.25	0.50

The shadowed elements corresponding to the limiting quasi-allocation ratios.

Table 4. The Iterative Calculation Results for Example 1

Iteration		0	1	2	3	4
F_{fresh}, t/h		58	58	58	58	58
F_{reg}, t/h		55.61	56.09	56.08	56.08	56.08
C_{reg} (ppm)	A	61.90	88.98	88.35	89.14	89.16
	B	5.09	8.46	8.39	8.39	8.39
	C	89.90	120.89	120.16	121.07	121.09

To determine the performing order of the unperformed processes, the *CPD* values of D_2, D_3 and D_5 are calculated based on the current available sources, S_1, S_4 and S^0_{reg}. The quasi-allocation ratios for S_1, S_4 and S^0_{reg} to D_2 are shown in Table 3, which are calculated with Eq. (4). The shadowed elements correspond to the limiting quasi-allocation ratios for the sources to satisfy D_2. The sum of the limiting quasi-allocation ratios, $0.75 + 1 + 0.32 \approx 2.1$, is the $CPD(D_2)$ value according to Eq. (2). Likewise, the *CPD* values for the other demands can be obtained. The *CPD* values of D_2, D_3 and D_5 are 2.1, 2.3 and 4.2, respectively. Processes 2 and 3 can be classified in the same group, because $CPD(D_3)/CPD(D_2) = 1.1 < 1.2$. From the *CPD* values, P_2 should be performed before P_3. From Table 3, it can be seen that S_4 should be reused first for D_2, because the limiting quasi-allocation amount of S_4 to D_2 is the largest. Then, S_1 is considered. From the data in Table 3, contaminant *B* is the *RKC* when S_1 is reused for D_2, and the limiting quasi-allocation ratio is 0.75. On the other hand, for contaminant *B*, the quasi-allocation ratio for S^0_{reg} to D_2 is 58.8. Therefore, S_1 and S^0_{reg} are complementary and can be used simultaneously to satisfy D_2. The amounts for S_1 and S^0_{reg} allocating to D_2 can be obtained from mass balance of contaminant *B*, and they are 24.28 and 1.72 t/h, respectively.

The performing order of P_3 and P_5 is considered based on their *CPD* values. The *CPD* values for P_3 and P_5 are 1.99 and 1.69, respectively, which are calculated from the current available sources S_1, S_2 and S^0_{reg}. Therefore, P_5 is performed before P_3. For P_5, 8 t/h of S_1 can be reused. Freshwater is not required. Because no source is used up, the performing order will not be challenged, although $CPD(D_3)/\ CPD(D_5) < 1.2$.

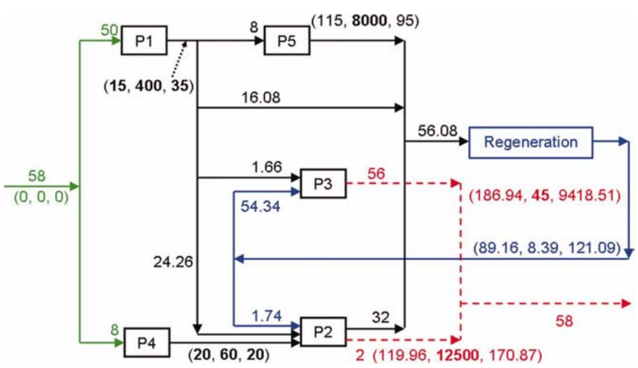

Figure 3. Design for Example 1 (where the numbers is the flowrate in t/h, and the concentrations reaching the limiting values are printed in bold).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table 5. The Final Data of the Streams for Example 1

	C_{in} (ppm)			C_{out} (ppm)			
Streams	A	B	C	A	B	C	*F* (t/h)
1	0	0	0	15	400	35	50
2	19.96	300	35.87	119.96	12,500	170.87	34
3	86.94	20	118.51	186.94	45	9418.51	56
4	0	0	0	20	60	20	8
5	15	400	35	115	8000	95	8

The concentrations reaching their limiting values are shadowed in the Table.

Now, P_3 is performed. For P_3, 2.11 t/h of S_1 and 53.89 t/h of the regenerated stream can be used. The first iteration is finished.

From the above design, it can be seen the amount of the regenerated stream reused is 55.61 t/h ($53.89 + 1.72 = 55.61$). This amount will be taken as the flowrate of the regenerated stream to calculate the regenerated concentration for the next iteration. The amounts of the unreused sources, S_1, S_2, S_3, and S_5, are 15.61, 34, 56 and 8 t/h, respectively. Some of them will form the regenerated stream. If the sources unreused (S_1, S_2, S_3 and S_5) are regenerated, their *CPS* values will be 0.39, 2.45, 105.67, and 1.86, respectively. The regenerated stream will be formed by the sources in ascending order of their *CPS* values. Therefore, the regenerated stream is formed by the following streams: the total amounts of S_1 (15.61 t/h) and S_5 (8 t/h), and part of S_2 (32 t/h, where 32 = 55.61-15.61-8). The concentrations of the regenerated stream will be (88.98, 8.46, 120.89) ppm for contaminants *A*, *B* and *C*, respectively. The regenerated stream obtained can be used for the next iteration.

The next iteration can be carried out similarly as discussed above. Table 4 shows the iteration results for iterations 0 to 4. It should be pointed out that in iterations 1 to 4, after P_1 and P_4 are performed, P_2 and P_3 can be classified in the same group because $CPD_2/CPD_3 < 1.2$. According to the *CPD* value, P_3 should be performed first. However, if P_3 is performed first, S_4 will be used up and it is more suitable for P_2, because the limiting quasi-allocation ratio for S_4 to D_2 is unity. Therefore, the performing order should be challenged. When P_2 is performed first, freshwater consumption can be reduced. Therefore, P_2 should be performed before P_3.

From Table 4, it can be seen that, the results for iteration 3 are very close to that for iteration 4. This means that the final design can be obtained with five iterations. The freshwater consumption is 58 t/h, the regenerated flowrate is 56.08 t/h, and the regenerated concentrations for *A*, *B* and *C* are (89.16, 8.39, 121.09) ppm, respectively. The final design is shown in Figure 3. The concentrations and flowrate data of the streams in Figure 3 are shown in Table 5. From Table 5, it can be seen that at least one contaminant in each demand stream reaches the limiting inlet concentration(s), and at least one contaminant in each source stream reaches

Table 6. Comparison of Different Methods for Example 1

	This Work	Kuo and Smith[2]	Liu et al.[22]
F_{fresh}, t/h	58	59.7	59.7
F_{reg}, t/h	56.08	55.48	55.48
Connection number	13	12	12

Table 7. The Limiting Data for Example 2 from Xu[25]

Process	Contaminant	C_{in}^{lim} (ppm)	C_{out}^{lim} (ppm)	M_{rem} (kg/h)	F^{lim} (t/h)
1	A	300	500	0.18	0.9
	B	5000	11,000	0.75	0.13
	C	1500	3000	0.1	0.07
2	A	10	200	3.61	19
	B	0	500	0.25	0.5
	C	0	1000	0.8	0.8
3	A	10	1000	6	6.061
	B	0	2000	15	7.5
	C	0	3500	10	2.86
4	A	100	400	2	6.67
	B	50	2000	0.8	0.41
	C	1000	3500	1	0.40
5	A	100	350	3	12
	B	50	1800	1.9	1.09
	C	1000	3500	2.1	0.84
6	A	85	350	3.8	14.34
	B	300	6500	1.1	0.18
	C	200	1000	2	2.50

Table 8. The Initial Concentrations of the Limiting Regenerated Streams and Their *CPS* Values for Example 2

Streams	$C_{reg,Si}^{lim}$ (ppm) A	B	C	*CPS*	To be Regenerated?
$S_{reg,S1}^{lim}$	50	1100	300	0.20	Yes
$S_{reg,S2}^{lim}$	20	50	100	0.05	Yes
$S_{reg,S3}^{lim}$	100	200	350	0.25	Yes
$S_{reg,S4}^{lim}$	40	200	350	0.19	Yes
$S_{reg,S5}^{lim}$	35	180	350	0.36	Yes
$S_{reg,S6}^{lim}$	35	650	100	0.12	Yes

Table 9. The Iterative Calculation Results for Example 2

		Iteration 0	1	2	3	4
F_{fresh}, t/h		34.88	25.55	25.55	25.55	25.55
F_{reg}, t/h		10.64	20.50	20.89	20.86	20.86
C_{reg} (ppm)	A	39.13	28.48	31.68	31.46	31.48
	B	269.08	9.94	7.27	7.02	7.01
	C	211.27	13.08	13.51	13.33	13.33

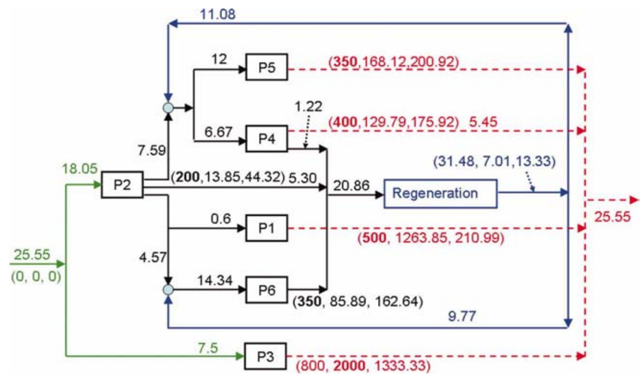

Figure 4. Design for Example 2 (where the numbers is the flowrate in t/h, and the concentrations reaching the limiting values are printed in bold).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table 10. The Final Data of the Streams for Example 2

Streams	C_{in} (ppm) A	B	C	C_{out} (ppm) A	B	C	*F* (t/h)
1	200	13.85	44.32	500	1263.85	210.99	0.60
2	0	0	0	200	13.85	44.32	18.05
3	0	0	0	800	2000	1333.33	7.50
4	100	9.78	25.92	400	129.79	175.92	6.67
5	100	9.78	25.92	350	168.12	200.92	12
6	85	9.18	23.16	350	85.89	162.64	14.34

The concentrations reaching their limiting values are shadowed in the Table.

Table 11. The Limiting Data for Example 3

Process	Contaminant	C_{in}^{lim} (ppm)	C_{out}^{lim} (ppm)	Mass load (g/h)
1	A	0	50	1000
	B	0	100	2500
	C	0	50	1500
2	A	10	100	5000
	B	30	300	20,000
	C	40	600	5000
3	A	20	200	5000
	B	50	400	15,000
	C	50	100	10,000
4	A	50	600	20,000
	B	110	450	15,000
	C	200	400	10,000
5	A	500	1100	30,000
	B	300	3500	15,000
	C	600	2500	25,000

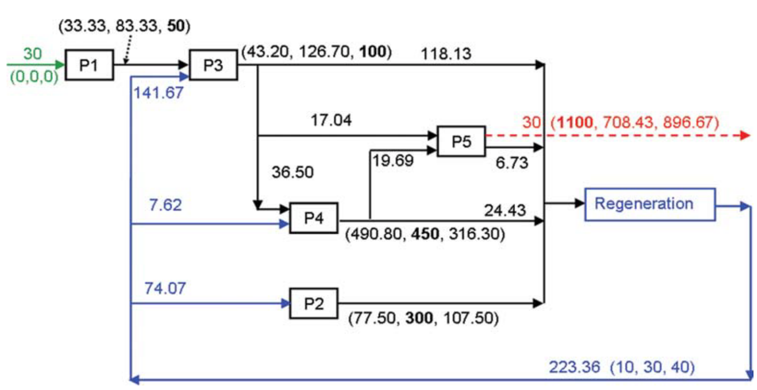

Figure 5. Design for Example 3 when the regenerated concentrations are fixed as Feng et al.[21] (where the numbers is the flowrate in t/h, and the concentrations reaching the limiting values are printed in bold).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table 12. The Final Concentrations of the Streams in Example 3 When the Regenerated Concentrations are Fixed as Feng et al.[21]

	C_{in} (ppm)			C_{out} (ppm)			
Streams	A	B	C	A	B	C	F (t/h)
1	0	0	0	33.33	83.33	50	30
2	10	30	40	77.50	300	107.50	74.07
3	14.08	39.32	41.75	43.20	126.70	100	171.67
4	37.47	110	89.64	490.80	450	316.30	44.12
5	283.13	300	215.95	1100	708.43	896.67	36.73

The concentrations reaching their limiting values are shadowed in the table

the limiting outlet concentration(s). The design has the feature of regeneration recycling.

Kuo and Smith[2] and Liu et al.[22] investigated this example and their design has the feature of regeneration reuse. Table 6 lists the results of different methods for this example. From the results, it can be seen that results obtained proposed in this work is comparable to that obtained in the literature. From the design procedure, it can be seen that the method proposed in this article is simpler than that of Kuo and Smith,[2] and is more suitable for computer calculation compared to the method of Liu et al.[22]

Example 2

This example is taken from Xu,[25] with the data shown in Table 7. The *RR* values for contaminants *A*, *B*, *C* are 90%.

The *CPS* values of the initial limiting regenerated streams are listed in Table 8. From the Dixon-type test results, it can be seen that there is no outlier in the *CPS* values in Table 8. All the source streams will be used to estimate the initial concentrations of the regenerated stream, which are (39.13, 269.08, 211.27) ppm for *A*, *B* and *C*, respectively.

The iteration calculation procedure is similar as that in Example 1, and the iterative results are listed in Table 9. From Table 9, it can be seen that the results for iteration 3 are nearly the same as that for iteration 4. This means that the final design can be obtained with five iterations. From Table 9, it can be seen that although the initial values of the concentrations of *B* and *C* are not very good compared to the converged values, the final result can still be obtained in a few iterations.

The total freshwater consumption is 25.55 t/h, the regenerated stream consumption is 20.86 t/h, and the regenerated concentrations for *A*, *B* and *C* are (31.48, 7.01, 13.33) ppm, respectively. The final design is shown in Figure 4. The flowrate and concentration data for the streams in Figure 4 are shown in Table 10. From Table 10, it can be seen that at least one contaminant reaches the limiting inlet and outlet concentrations except the inlet stream of process 1.

In the design of Xu,[25] which is obtained by using an MINLP approach, the total freshwater consumption is 27.10 t/h, the regenerated flowrate is 24.75 t/h, and the regenerated concentrations are (37.61, 59.69, 42.27) ppm. In addition, the connection number for the design of this work is the same as that of Xu.[25] From the above results, it can be seen that the design obtained in this work is better than that of Xu.[25]

Table 13. The Iterative Calculation Results for Example 3 When All the *RR* Values are 95%

		Iteration				
		0	1	2	3	4
F_{fresh}, t/h		37.22	30	30	30	30
F_{reg}, t/h		154.39	174.28	175.21	175.18	175.18
C_{reg} (ppm)	A	11.14	7.45	8.41	8.41	8.41
	B	18.04	14.19	13.88	13.80	13.80
	C	12.14	6.77	7.46	7.46	7.47

Table 14. The Final Data of the Streams for Example 3 When All the *RR* Values are 95%

	C_{in} (ppm)			C_{out} (ppm)			
Streams	A	B	C	A	B	C	F (t/h)
1	0	0	0	33.33	83.33	50	30
2	10	18.24	10.18	80.44	300	80.62	70.98
3	19.00	50	21.66	58.17	167.51	100	127.65
4	39.55	110	65.38	492.89	450	292.04	44.12
5	262.06	300	190.07	1100	718.97	888.36	35.80

The concentrations reaching their limiting values are shadowed in the table.

Example 3

This example is taken from Feng et al.[21], with the data shown in Table 11. The regenerated concentrations for contaminants *A*, *B* and *C* are fixed: (10, 30, 40) ppm, respectively.

For this example, the regenerated concentrations are given. Therefore, the final design can be obtained without iteration. The key is to determine the flowrate of the regenerated stream. The design of this example is shown in Figure 5. The total freshwater consumption is 30 t/h, and the regenerated stream flowrate is 223.36 t/h.

When all the processes are performed, the amounts of the unreused sources, S_2, S_3, S_4, and S_5, are 74.07, 118.13, 24.43, and 36.73 t/h, respectively. If the unreused streams are regenerated, the order of the *CPS* values is: $CPS(S_{reg,S_3}) < CPS(S_{reg,S_2}) < CPS(S_{reg,S_4}) < CPS(S_{reg,S_5})$. Then, the regenerated stream will be formed by the following streams: the total amount of S_3 (118.13 t/h), S_2 (74.07 t/h), S_4 (24.43 t/h), and part of S_5 (6.73 t/h). From the above results, it can be obtained that the concentrations before regeneration are (135.35, 237.05, 150.13) ppm. The flowrate and concentration data for the design are shown in Table 12. From Table 12, it can be seen that at least one contaminant reaches the limiting inlet and outlet concentrations for all the streams, except the inlet stream of P_3.

Feng et al.[21] addressed that the optimization of the WUNs involving regeneration recycling was a multiobjective problem with several important parameters. They used a sequential nonlinear optimization method to solve this multiobjective problem. Freshwater consumption, regenerated water flowrate and contaminant regeneration load, were in turn minimized, which correspond to three mathematical models. They also considered the complexity of water network configuration by minimizing the number of interconnections among the processes. In the optimal design of Feng et al.[21],

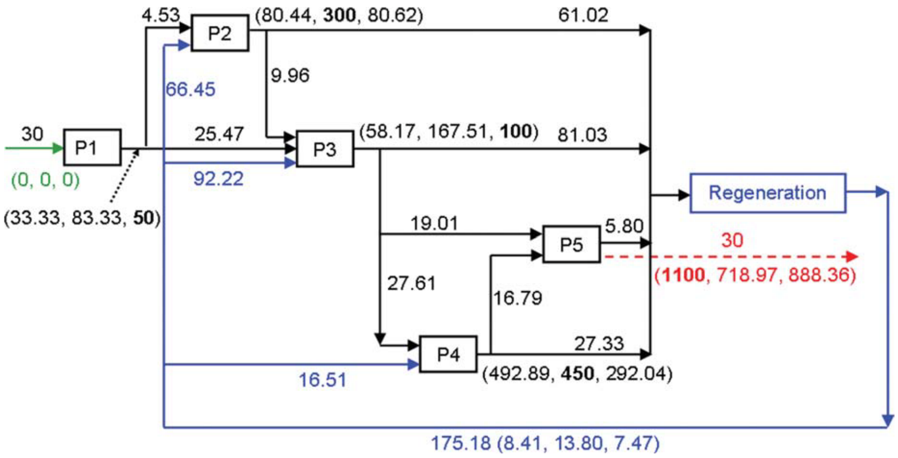

Figure 6. Design for Example 3 when all the *RR* values are 95% (where the numbers is the flowrate in t/h, and the concentrations reaching the limiting values are printed in bold).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the consumptions of freshwater and the regenerated stream are the same as that obtained in this work. The concentrations before regeneration are (135.4, 224.8, 142.6) ppm for *A*, *B* and *C*, respectively. The connection number for the design of this work is the same as that of Feng et al.[21] From the above results, it can be seen that the design obtained in this work is comparable to that of Feng et al.[21]

For this example, when all the *RR* values are 95%, the iteration results and the final stream data are listed in Tables 13 and 14, respectively. The final design is shown in Figure 6. The regenerated concentrations are (8.41, 13.80, 7.47) ppm, which are lower than the fixed regenerated concentrations of Feng et al.,[21] (10, 30, 40) ppm. The amount of the regenerated stream is 175.18 t/h, which is smaller than that of Feng et al.,[21] 223.36 t/h. This means that when the regenerated concentrations are reduced, the consumption of the regenerated stream (even freshwater consumption) can be reduced.

Discussion and Conclusions

In this article, an iterative design procedure is proposed for the WUNs involving regeneration recycling based on a modified design procedure of Liu et al.[22,23] In the design, the regenerated stream is treated as an additional source stream of the WUN involving reuse only. For the networks with given regenerated concentrations, the final design can be obtained without iteration. For the networks with known *RR*s for the contaminants, the final design can be obtained in a few iterations. For the problem with known *RR*s, in the first iteration, the initial regenerated concentrations are determined first based on the features of the network and the regeneration process. In the design, the freshwater consumption, regenerated water flowrate, and the before-regeneration concentrations, which can reflect the total cost of the network, are considered simultaneously. A few case studies are investigated. The results show that the results obtained in this work are comparable to that obtained in the literatures, and the method proposed is simple and effective.

Acknowledgments

This work is financially supported by the Research Foundations for Returned Scholars from Overseas of the Human Resources Department of Hebei Province, China, and the National Natural Science Foundation of China (Grant No. 20776036).

Notation

$C^{\lim}_{Dj,k}$ = limiting concentration of contaminant k in demand D_j
$\boldsymbol{C}^{\lim}_{Si}$ = limiting concentration vector of source S_i
$\boldsymbol{C}^{\lim}_{reg,Si}$ = initial regenerated concentration vector of source S_i
$C_{S_{i,k}}$ = concentration of contaminant k in source S_i
CPD = concentration potential of demand, defined in Eq. (2)
CPS = concentration potential of source, defined in Eq. (3)
FKC = freshwater key contaminant which determines the minimum freshwater amount when freshwater is used only
NC = number of the contaminants
ND = number of demand streams
NS = number of source streams
$R_{i,j}$ = limiting quasi-allocation ratio from S_i to D_j, as shown in Eq. (4)
RKC = reuse key contaminant
RR = removal ratio, as defined in Eq. (1)
$\boldsymbol{C}^0_{reg}$ = initial regenerated concentration vector

Literature Cited

1. Wang YP, Smith R. Wastewater minimisation. *Chem Eng Sci*. 1994;49:981–1006.
2. Kuo W, Smith R. Design of water-using systems involving regeneration. *Trans Inst Chem Eng*. 1998;76(B):94–114.
3. Mann JG, Liu YA. *Industrial Water Reuse and Wastewater Minimization*. New York: McGraw-Hill, 1999.
4. Hallale N. A new graphical targeting method for water minimisation. *Adv Environ Res*. 2002;6:377–390.
5. Feng X, Bai J, Zheng XS. On the use of graphical method to determine the targets of single-contaminant regeneration recycling water systems. *Chem Eng Sci*. 2007;62:2127–2138.
6. Castro P, Matos H, Fernandes, MC, Nunes CP. Improvements for mass exchange networks design. *Chem Eng Sci*. 1999;54:1649–1665.
7. Gomes JFS, Queiroz EM, Pessoa FLP. Design procedure for water/wastewater minimization: single contaminant. *J Clean Prod.*. 2007;15:474–485.
8. Deng C, Feng X, Bai J. Graphically based analysis of water system with zero liquid discharge. *Chem Eng Res Des*. 2008;86:165–171.
9. Ng DKS, Foo DCY, Tan RR, Tan YL. Ultimate flowrate targeting with regeneration placement. *Chem Eng Res Des*. 2007;85:1–15.

10. Ng DKS, Foo DCY, Tan RR, Tan YL. Extension of targeting procedure for ultimate flowrate targeting with regeneration placement by Ng et al., Chem Eng Res Des. 85(A9):1253–1267. *Chem Eng Res Des*. 2008;86:1182–1186.
11. Bandyopadhyay S, Cormos CC. Water management in process industries incorporating regeneration and recycle through a single treatment unit. *Ind Eng Chem Res*. 2008;47:1111–1119.
12. Takama N, Kuriyama T, Shiroko K, Umeda T. Optimal water allocation in a petroleum refinery. *Comput Chem Eng*. 1980;4:251–258.
13. Alva-Argaez AA, Kokossis C, Smith R. Wastewater minimisation of industrial system using an integrated approach. *Comput Chem Eng*. 1998;22:S741–S744.
14. Huang CH, Chang CT, Ling HC, Chang CC. A mathematical programming model for water usage and treatment network design. *Ind Eng Chem Res*. 1999;38:2666–2679.
15. Gunaratnam M, Alva-Argaez A, Kokossis A, Kim J-K, Smith R. Automated design of total water systems. *Ind Eng Chem Res*. 2005;44:588–599.
16. Relvas S, Matos HA, Fernandes MC, Castro P, Nunes C. AquoMin: A software tool for mass-exchange networks targeting and design. *Comput Chem Eng*. 2008;33:1085–1105.
17. Koppol A, Bagajewicz M, Dericks B, Savelski M. On zero water discharge solutions in the process industry. *Adv Environ Res*. 2003;8:151–171.
18. Cao D, Feng X, Duan X. Design of water network with internal mains for multi-contaminant wastewater regeneration recycle. *Chem Eng Res Des*. 2004;82:1331–1336.
19. Rivas A, Irizar I, Ayesa E. Model-based optimisation of wastewater treatment plants design. *Environ Model Softw*. 2008;3:435–450.
20. Feng X, Seider WD. New structure and design method for water networks. *Ind Eng Chem Res*. 2001;40:6140–6146.
21. Feng X, Bai J, Wang H, Zheng XS. Grass-roots design of regeneration recycling water networks. *Comput Chem Eng*. 2008;32:1892–1907.
22. Liu Z-Y, Li Y-M, Liu Z-H, Wang Y-J. A simple method for design of water-using networks with multiple contaminants involving regeneration reuse. *AIChE J*. 2009;55:1628–1633.
23. Liu Z-Y, Yang Y, Wang L-Z, Wan X, Hou K-H. A heuristic design procedure for water-using networks with multiple contaminants. *AIChE J*. 2009;55:374–381.
24. Walfish S. *A Review of Statistical Outlier Methods*. *Pharmaceutical Technology*. Available at: http://www.pharmtech.com/pharmtech/content/printContentPopup.jsp?id=384716 last visited Dec. 10, 2010.
25. Xu DM. Study on the optimization design of water-using network. Ph.D. thesis, Ocean University of China, Shandong, China, 2004.

Appendix

The Dixon-type test results for Example 1

According to the Dixon-type test, we can use the following equations to determine whether the largest or smallest *CPS* values are the potential outliers.

The ratio r_{10} is used when the suspected outlier is the largest observation:

$$r_{10} = \frac{x_{(n)} - x_{(n-1)}}{x_{(n)} - x_{(1)}} \tag{A1}$$

The ratio r'_{10} is used when the suspected outlier is the smallest observation:

$$r'_{10} = \frac{x_{(2)} - x_{(1)}}{x_{(n)} - x_{(1)}} \tag{A2}$$

To determine the potential outlier, the *CPS* values obtained in this example should be arranged in ascending order, which are listed in Table 2. From the data in Table 2, for the largest observation, r_{10} = (31.15–0.79)/(31.15–0.11) = 0.978, which is greater than the tabled value of 0.780.[24] Therefore, for this system, the largest observation, 31.15, is a statistical outlier. For the smallest value of *CPS*, r'_{10} = (0.11–0.11)/(31.15–0.11) = 0. Therefore, the smallest value, 0.11, is not a statistical outlier.

Manuscript received Sep. 9, 2010, and revision received Jan. 21, 2011.