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Water and Wastewater Minimization in Petroleum Refinery Through Water Pinch Analysis-Single and Double Contaminants Approach

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ABSTRACT

This research is based on finding the way to minimize water utility in the petrochemical and petroleum industries due to high rate of water consumption. One of the petroleum refineries in the center of Iran has been considered as a case study. In this research, two key contaminants including Suspended Solid (SS) and Hardness (H) have been considered to analyze the water network. These key contaminants once were analyzed separately as a single contaminant and the amount of required fresh water was calculated for both of them. In this stage, amount of freshwater was reduced about 60.9 $m^3 h^{-1}$ (17%) and 203 $m^3 h^{-1}$ (59.7%) in terms of Suspended Solids (SS) and Hardness (H), respectively. As it is seen, water minimization within three optional operations for SS is less than H. Therefore, this contaminant is a limiting contaminant and can be selected as a key contaminant. In the next stage, two mentioned contaminants were analyzed simultaneously based on their mass transfer. The results show that the amount of required water is reduced from 340 to 197.26 $m^3 h^{-1}$ that is about 42%. Analyzing both methods show that amount of required water can be determined by mass transfer of suspended solids. In addition, the method based on multiple contaminants gives more precise results rather than single contaminant. Therefore, it is suggested that more contaminants and operations are considered to study water networks and reach water utility optimization based on key contaminant as well.

Key words: Water utility, wastewater, mass transfer, water reuse, petroleum

INTRODUCTION

Generally, water is used as raw material in most of the industries and generated wastewater is discharged in to the environment. Increasing freshwater utility is due to economical and industrial growth, considerably. On the one hand, the price of water is increased and behind it, the price of products goes up, on the other hand the environmental laws do not allow discharging wastewater in to the environment. Therefore, industries have to use some strategies related to water utility minimization. Nowadays, different techniques and methods have been developed to design water allocation system so that water utility is reduced in an acceptable level (Dhole *et al*., 1996; Bagajewicz, 2000). Water pinch technology is a systematic technique for analyzing water networks and reducing expenditures related to different water using processes (Manan *et al*., 2006; Hallale, 2002; Gómez *et al*., 2001).

El-Halwagi and Srinivas (1992) propounded the theory of mass exchange networks. This theory was based on a two-stage solution; first, Mixed Integer Nonlinear Programming and then Mixed Integer Liner Programming. Wang and Smith (1994) presented a conceptual view based on maximum water reuse.

Most of the methods used in water pinch analysis are based on the mass exchange of one or several contaminants. If the mass exchange is based on mass transferring of one contaminant, the problem will be solved as a single contaminant. Nevertheless, if it includes mass transferring of two or more key contaminants, the problem will be solved as multiple contaminants. Graphical methods, mathematical and computer-based methods may be used for both cases. Each method has some advantages and disadvantages. Graphical methods are so practical to solve single contaminant problems. However, they are complicated and sometime impossible for multiple contaminants problems.

Regarding graphical methods. Wang and Smith (1994) used limiting composite curve to solve multiple contaminants problems. Kuo and Smith (1997, 1998) applied a new method to reduce complexity of graphical method. This method was based on breaking the operations. Kralj *et al.* (2005) presented the method of heat integration between processes. This method is based on energy saving. They showed that simultaneous integration between processes can be performed using MINLP algorithm. Thokozani (2005) presented a graphical technique for freshwater and wastewater minimization in completely batch operations. Water minimization was achieved through the exploitation of inter- and intra-process water reuse and recycle opportunities. The exploration and use of inherent storage in batch processes was demonstrated using a real-life case study. In addition to, Foo *et al.* (2005) presented a two-stage procedure for the synthesis of a Maximum Water Recovery (MWR) network for a batch process system, covering both mass transfer-based and non-mass transfer-based water-using processes.

Mathematical methods are more exact but sometime complicated especially in the case of multiple contaminants. There is different computer programming for users such as GAMS programming. Tan *et al.* (2007) presented a new systematic technique for the retrofit of water network with regeneration based on water pinch analysis. They developed a case study on paper making process was used to demonstrate the new methodology. Gómez *et al.* (2001) used a water source diagram method based on outlet flow-rate. Alva-Argaez *et al.* (2007) introduced a systematic methodology that empowers conceptual engineering and water-pinch with mathematical programming methods. The method focuses on petroleum refineries explaining trade-offs and savings between freshwater costs, wastewater treatment, piping costs and environmental constraints on the discharge.

Gouws *et al.* (2008) used a mathematical technique for water minimization in multipurpose batch processes. Oliver *et al.* (2008) used water pinch analysis and Mix Integer Linear Programming (MILP) to synthesize the water network for batch processes. Ulson de Souza *et al.* (2009) investigated the implementation of the Water Source Diagram (WSD) in a petroleum refinery with six operations, which consume water. They observed that with the application of the WSD method the water consumption was substantially reduced. Dakwala *et al.* (2009) undertook a case study with an aim to reduce demineralised water and freshwater flow rates and consequently the wastewater flow rate. They developed a program in MATLAB for analysis using Water Pinch. The program will formulate the optimization model and solve for the solution automatically. The improved water-using network designed for the present work consumed less demineralised and freshwater.

Mohammadnejad *et al.* (2010) studied the optimization of water and steam allocation network. They developed an algorithm to simplify the relevant calculations and applied it for reforming the network in a petroleum refinery. Mehrdadi *et al.* (2009) analyzed six method of water pinch technique to illustrate the advantages and disadvantages of each method.

In this research, two key contaminants including Suspended Solid (SS) and Hardness (H) have been considered and also the research is based on Mann and Liu (1999) research to analyze the water network. In fact This method is the completed method of Wang and Smith (1994).

There are two targets for wastewater minimization by water pinch technology in this research:

- Wastewater minimization considering single contaminant approach
- Wastewater minimization considering multiple contaminants approach

MATERIALS AND METHODS

This research has been performed for one of the petroleum refineries in the Center of Iran from 2006 to 2009. This refinery comprises two refineries and some petroleum processing manufactories. The simplified flowchart of water and steam allocation network in the refinery has been showed by Fig. 1. Currently this refinery utilizes water about 505 $m^3 h^{-1}$. As it is seen, water and steam allocation network in the refinery is well designed and amount of water utility and wastewater generation are in an acceptable level while wastewater is reused or regenerated.

Table 1 shows flow-rate and stream constraints in the water network. Based on these constraints, the limiting water flow-rates are determined for the optional operations. The water flow-rate is needed for achieving the mass transfer of contaminants is required for water minimization. Selecting contaminants depends on the industry and its water requirements. In addition, it is very important to select processes, which have high rate of water consumption.

According to these considerations, Suspended Solids (SS) and Hardness (H) were selected as key contaminants and three processes, which use vast amount of water such as desalter, cooling towers as well as portable; plant and fire were selected to be analyzed.

These key contaminants once were analyzed separately as a single contaminant and the amount of required fresh water was calculated for both of them so in which case that amount of

Table 1: Flow-rates and stream constraints for the optional water network

No.	Flow-rate ($M^3 h^{-1}$)	Stream constraints (ppm)
1	505	TH = 150, M-ALK = 140, SiO_2 = 9.3, SS = 1, TSS = 2.15, TFe<0.05, CL_2<0.05
17	104	TH = 270, M-ALK = 66, SiO_2 = 9.87, SS = 2, TSS = 2.66, TFe<0.05, CL_2<0.05
10	20	TH = 0, PO_4 = 20, TFe<0.05
13	113	TH = 150, M-ALK = 140, SiO_2 = 9.3, S.S = 1, TSS = 2.15, TFe<0.05, CL_2<0.05
18	168	TH = 150, M-ALK = 140, SiO_2 = 9.3, SS = 1, TSS = 2.15, TFe<0.05, CL_2<0.05
19	160	TH = 241, M-ALK = 23, SS = 22
21	17	TH = 12, M-ALK = 44, SiO_2 = 6.6, SS = 13, TSS = 24.3, TFE = 0.83, CL_2<0.05, H_2O = 3.4, NH_3 = 46
22	59	TH = 12, M-ALK = 44, SiO_2 = 6.6, SS = 13, TSS = 24.3, TFE = 0.83, CL_2<0.05, H_2O = 3.4, NH_3 = 46
23	59	TH = 160, M-ALK = 40, SiO_2 = 1.4, SS = 20, TSS = 25, TFe = 3.12, CL_2<0.05
15	37	TH = 1250, M-ALK = 30, SiO_2 = 48.9, S.S = 1, TSS = 2.95, TFe = 0.35, CL_2 = 2.5

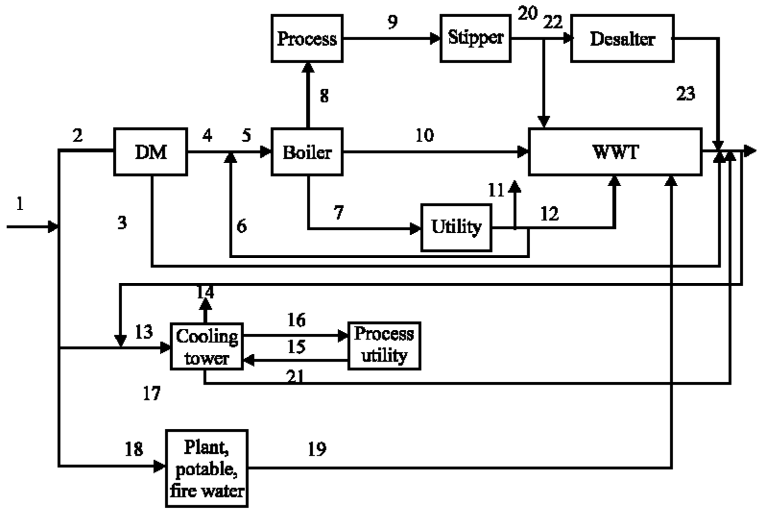

Fig. 1: Flowchart of water and steam allocation network in the refinery

minimization is less than another one, we can select it as a limiting contaminant for processes. This method can be applied easily for different industries and mathematical calculations are not complicated as well.

After that, two mentioned contaminants were analyzed simultaneously based on their mass transfer. In other words, mass transfer of a contaminant was analyzed with respect to another one. Firstly, limiting water profile is drawn based on inlet and outlet concentrations of one of the contaminants then the concentration of second one is calculated in each concentration interval. Here, fraction $\theta_{i,n}$ is defined as a ratio of the actual flow-rate to operation i at concentration interval boundary n to the limiting flow-rate of operation i. This fraction is used to design the water network and according to this, total flow-rate of network is obtained. Finally, the results of two methods are compared.

RESULTS

Single contaminant approach: To minimize wastewater by water pinch technique, it is necessary to calculate minimum water flow-rate required to reduce the contaminant concentration of the operation in an acceptable level. Therefore, it must be taken some steps. The first step is providing limiting process data table. This table includes minimum inlet and outlet flow-rates, maximum inlet and outlet concentrations as well as transferred mass by processes. Table 2 and 3 show the limiting process data for the processes in terms of suspended solids and hardness, respectively.

Mass load is calculated as follows:

$$\Delta m_{opi} = \frac{(C_{out} - C_{in}) f_{opi}}{1000} \quad (1)$$

Since, operation 1 and 3 lose freshwater, which is discharged as wastewater, it is necessary to separate water losses from utilized water within processes. In the next step, pinch point of operations is determined as some operations with the concentration lower than freshwater are

Table 2: Limiting data for Suspended Solids (SS) (single contaminant approach)

Operations i	Q_{in}	Q_{out}	C_{in}	C_{out}	Δm	Cumulative Δm
	($m^3\ h^{-1}$)		(ppm)		($kg\ h^{-1}$)	
Cooling tower1a	37	37	1	2	0.037	0.037
Desalter	59	59	13	20	0.413	0.450
Potable, fire, plant water	160	160	1	22	3.360	3.810

Table 3: Limiting data for hardness (H) (single contaminant approach)

Operations i	Q_{in}	Q_{out}	C_{in}	C_{out}	Δm	Cumulative Δm
	($m^3\ h^{-1}$)		(ppm)		($kg\ h^{-1}$)	
Desalter1	59	59	12	160	8.732	8.732
Potable, fire, Plant water2	160	160	400	500	16.00	24.70
Cooling Tower3	37	37	150	1250	40.70	65.40

Table 4: The results for minimum required water regarding SS

Operation i	f_{min} ($m^3\ h^{-1}$)	C_{out} (ppm)
1a	37.0	2.0
2	21.7	20.0
3a	160.0	22.0
Overall	218.7	18.4

Table 5: The results for minimum required water regarding H

Operation i	Q_{min} ($m^3\ h^{-1}$)	C_{out} (ppm)
1	54.5	160
2a	37.0	1250
3a	45.7	500
Overall	137.2	567

supplied, but reach operations do not need freshwater. The minimum freshwater flow-rate is called the water pinch. The pinch point is important to minimize wastewater because the system does not require freshwater above this point. The water streams can be reused above this point from elsewhere in the system. Therefore, minimum flow-rate with water reuse is the flow-rate required to reach pinch point.

In this research, a graphical method named concentration composite curve has been used to determined pinch point. Figure 2a and 3a show the concentration composite curves for optional operations. To draw these curves, it is necessary to calculate minimum required water flow-rate without water reuse for each process according to below equation:

$$f_{min}^{opi} = \frac{\Delta m_i}{C_{out}^{*} - C_{in}^{w}} \times 1000\ m^3\ h^{-1} \tag{2}$$

Table 4 and 5 give the calculation related to minimum required flow-rate. Here, we did not consider the water losses between processes. After that, minimum water loss is calculated for two mentioned operations as below:

Accordingly, the concentration composite curve regarding SS and H will change as Fig. 2b and 3b. As it is seen, total required freshwater for operations in terms of SS and H will be 300.8 and 194.1 $m^3\ h^{-1}$, respectively.

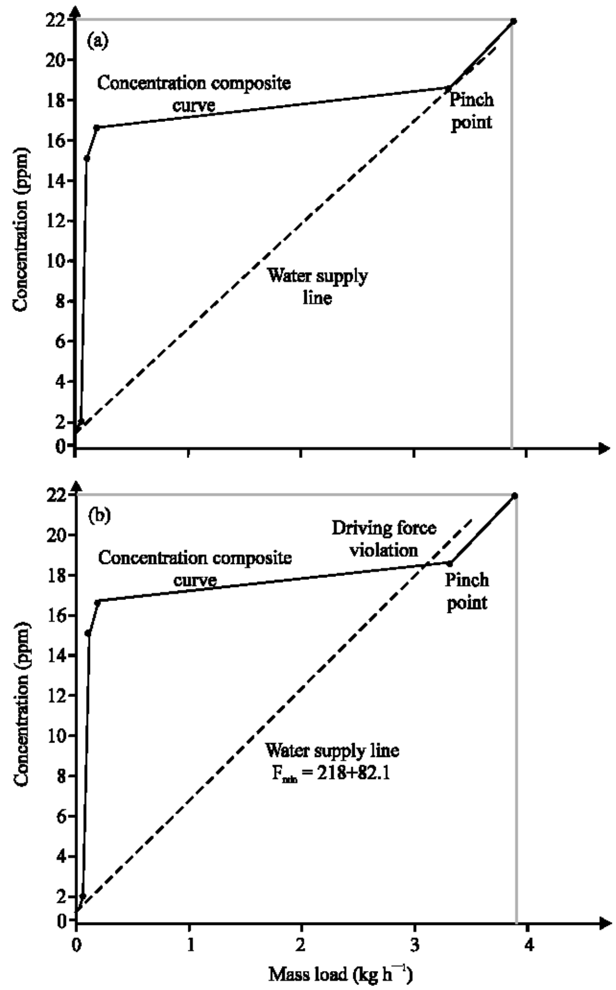

Fig. 2: The concentration composite curves (a) without water losses for SS and (b) considering water losses for SS

$$f = f_{loss}\left(\frac{C_{pinch} - C_{in}}{C_{pinch}}\right) \tag{3}$$

According to Fig. 1, two new water streams are capable for reuse. Their constraints are as below:

Stream 10: Boiler blow down (C_{out}= 4 ppm and f_{12} = 20 $m^3 h^{-1}$)
Stream 12: Outlet utility (C_{out} = 2 ppm and f_{12} = 45 $m^3 h^{-1}$)

Since, these water streams supply 65 $m^3 h^{-1}$ water so total required water is:

SS : 300.8- 65=235 $m^3 h^{-1}$
H : 194.1- 65=129.1 $m^3 h^{-1}$

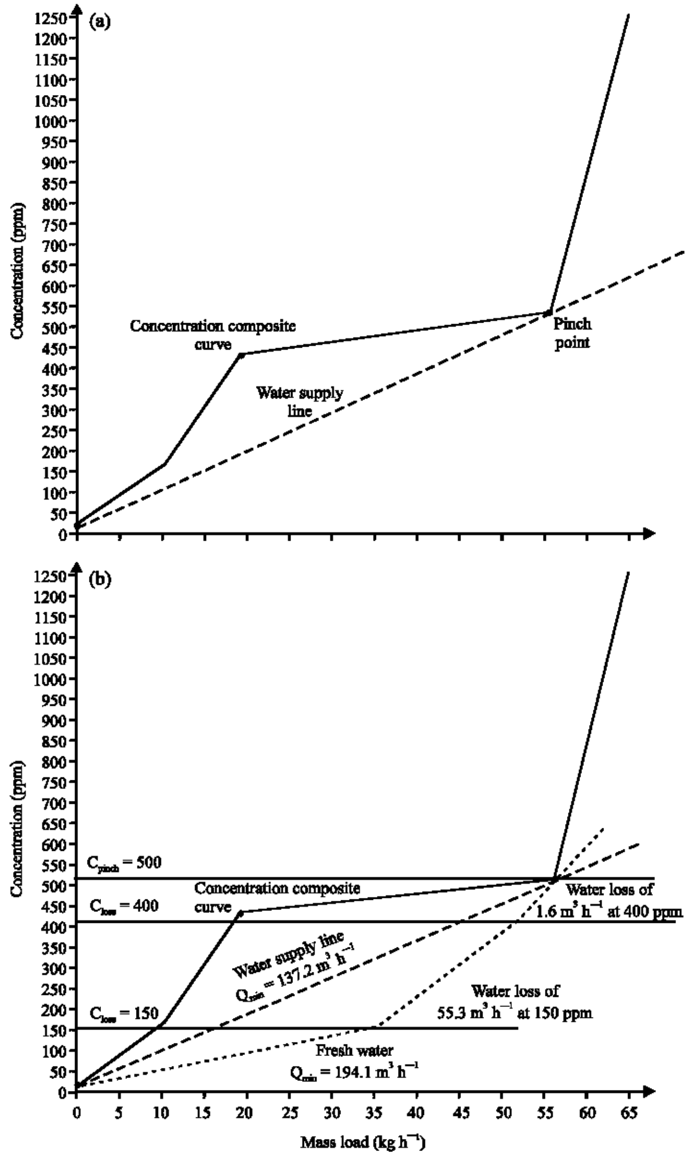

Fig. 3: The concentration composite curves (a) without water losses for H and (b) considering water losses for H

James G. Mann's method is so easy and efficient for designing networks with minimum freshwater required in the different industries. In this method, at first concentration interval boundaries are selected from limiting process data tables for all operations. These interval boundaries are drawn as horizontal lines and different operations are drawn as upward-directed arrows and water streams as downward-directed arrows. In this research, three water stream sources are considered including freshwater, boiler blow down and outlet utility. Transferred mass load of contaminant for each interval boundary is calculated as follows (Mann and Liu, 1999):

$$m_{i,k}(\text{kg h}^{-1}) = \Delta m_{i,tot}(\text{kg h}^{-1})\left[\frac{C^{*}_{k+1} - C^{*}_{k}}{C^{lim}_{i,out} - C^{lim}_{i,n}}\right] \quad (4)$$

Then required water flow-rate is calculated for each transferred mass load according to below equation:

$$f_{i,k}^{tot}(m^3\ h^{-1}) = \frac{m_{ik}(kg\ h^{-1})}{[C_{k+1}^{*} - C_{i,k}^{w}]\ ppm} \times 10^3 \tag{5}$$

is average contaminant concentration of the water $C_{i,k}^{w}$ is required flow-rate for each interval boundary and $f_{i,k}^{tot}$ is average contaminant concentration of the water stream for operation i entering interval boundary k. The calculated mass loads and required flow-rates are shown in Fig. 4a and b.

Loop breaking is a method applied to reduce number of required operations and simplify the network (Mann and Liu, 1999). A loop includes:

- One water stream
- One entire water using unit
- One process stream
- A second entire water using unit

In this method, mass load is transferred from an operation to another one in a same loop. Then outlet concentration of water stream is recalculated for the combined unit according to below equation.

$$c_{out}^{w}(ppm) = c_{in}^{w}(ppm) + \frac{\sum \Delta m_{i,tot}(kg\ h^{-1})}{f_{min}(m^3\ h^{-1})} \times 10^3 \tag{6}$$

If the outlet concentration is equal to or less than total outlet concentration of the system, mass load will be transferred. For example, the outlet concentration for transferring from E to B according to Fig. 5a is calculated as bellow:

$$C_{out}^{w} = 1 + {[0.36 + 1.12 + 1.76 + 0.16]}\big/{160} \times 10^3 = 22\ ppm \equiv 22\ ppm$$

That is not greater than total outlet concentration, so it is possible to be transferred. Final diagram and flowchart have been represented in Fig. 5a,b and 6a, b. As it is seen, in the single contaminant method, amount of freshwater was reduced about 60.9 $m^3\ h^{-1}$ (17%) and 203 $m^3\ h^{-1}$ (59.7%) in terms of Suspended Solids (SS) and Hardness (H), respectively. Also water minimization within three optional operations for SS is less than H. Therefore, this contaminant is a limiting contaminant and can be selected as a key contaminant. In some cases it would be incorrect to assume that greater of the two flow-rate would be sufficient to transfer both contaminants.

Double contaminant approach: In this method, limiting water profile is drawn based on inlet and outlet concentration of one of the contaminants as a reference contaminant according to

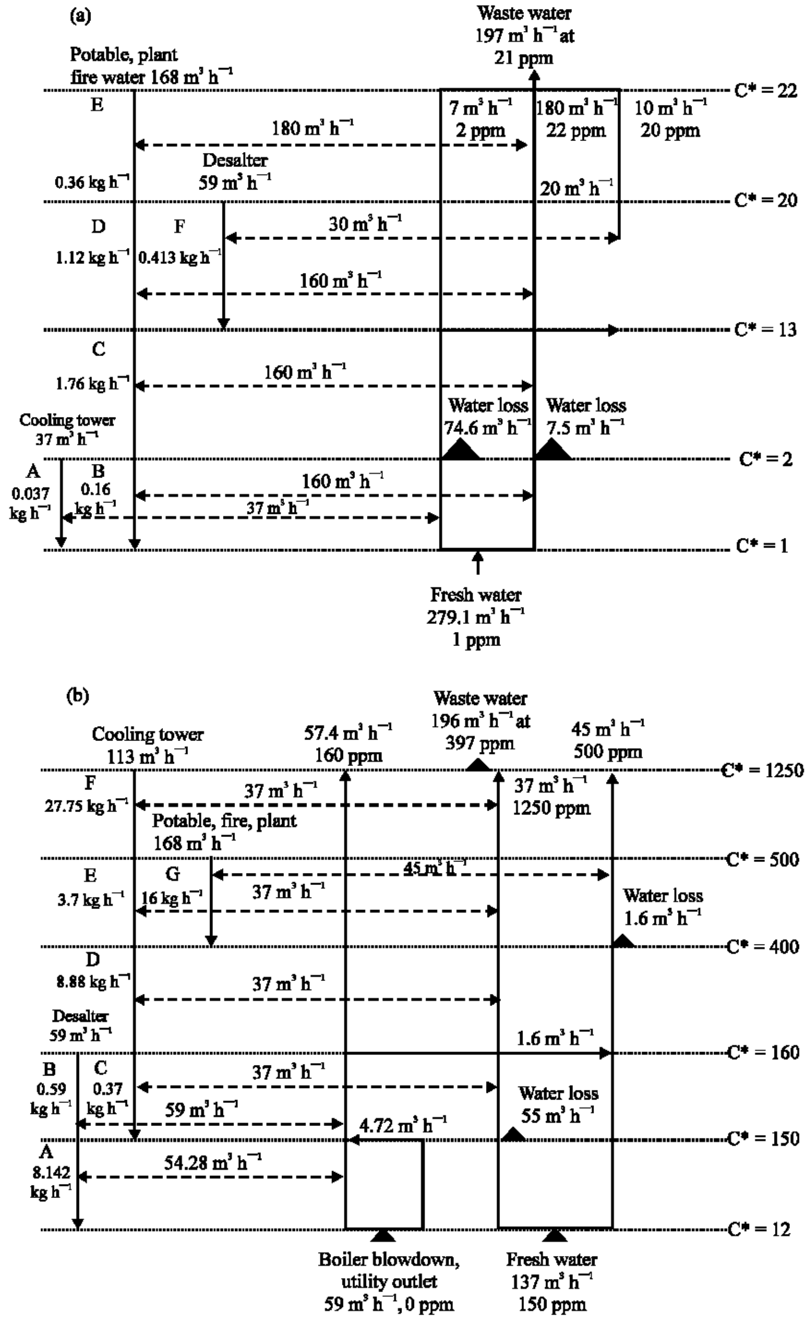

Fig. 4: Water network diagram before loop breaking in terms of (a) SS and (b) H

limiting process data shown in Table 2 and 3. Then the concentration of second contaminant is calculated based on the first one by below equations. Figure 7 shows limiting water profile for three operations. In this profile, the concentrations of two key contaminants at each concentration interval boundary have been shown in the brackets for each operation. For example, [12,13] means that the concentration of reference contaminant and second one are 13 and 12 ppm, respectively.

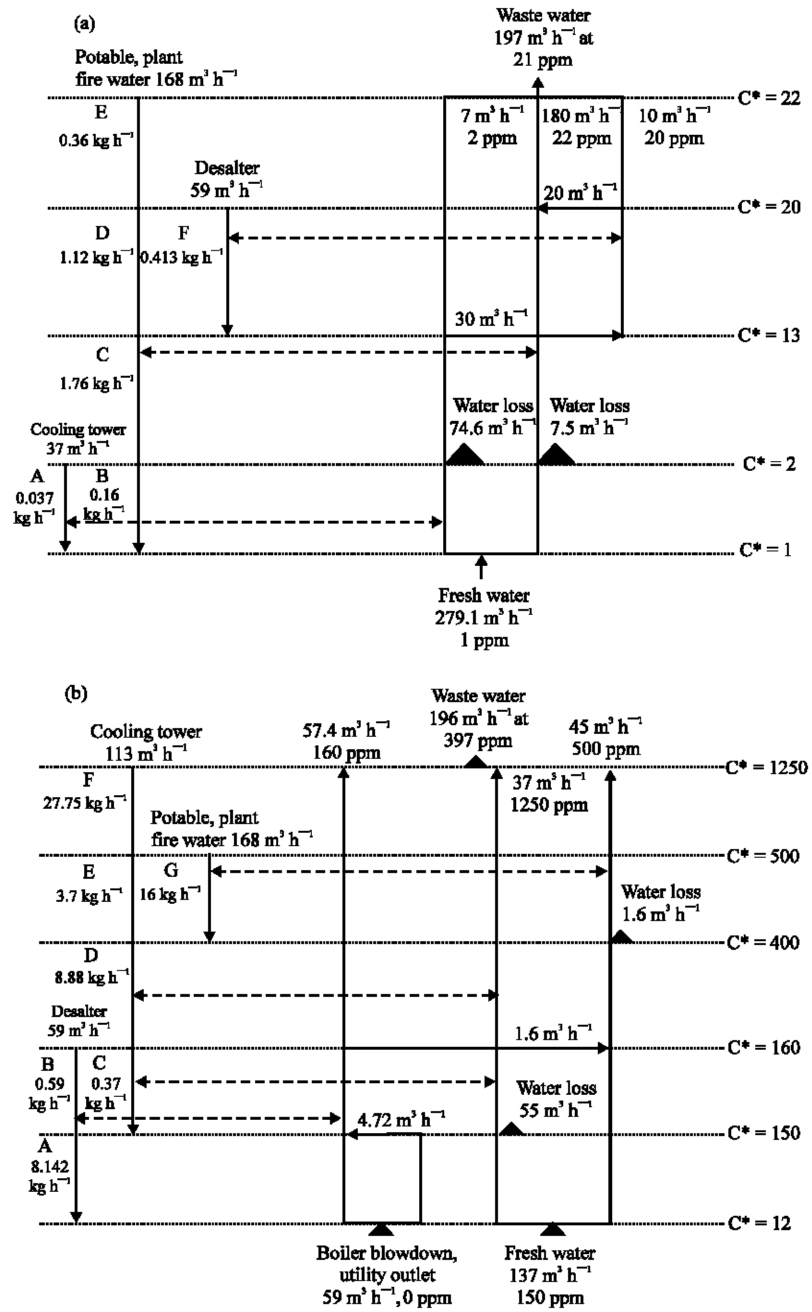

Fig. 5: Water network diagram after loop breaking in terms of (a) SS and (b) H

$$\frac{C_{i,H,n} - C_{i,H,in}}{C_{i,H,out} - C_{i,H,in}} = \frac{C_{i,SS,n} - C_{i,SS,in}}{C_{i,SS,out} - C_{i,SS,in}} \tag{7}$$

For example:

$$\frac{150-12}{160-12} = \frac{C_{i,SS,n} - 13}{20-13} \rightarrow C_{i,SS,n} = 19.5\text{ppm}$$

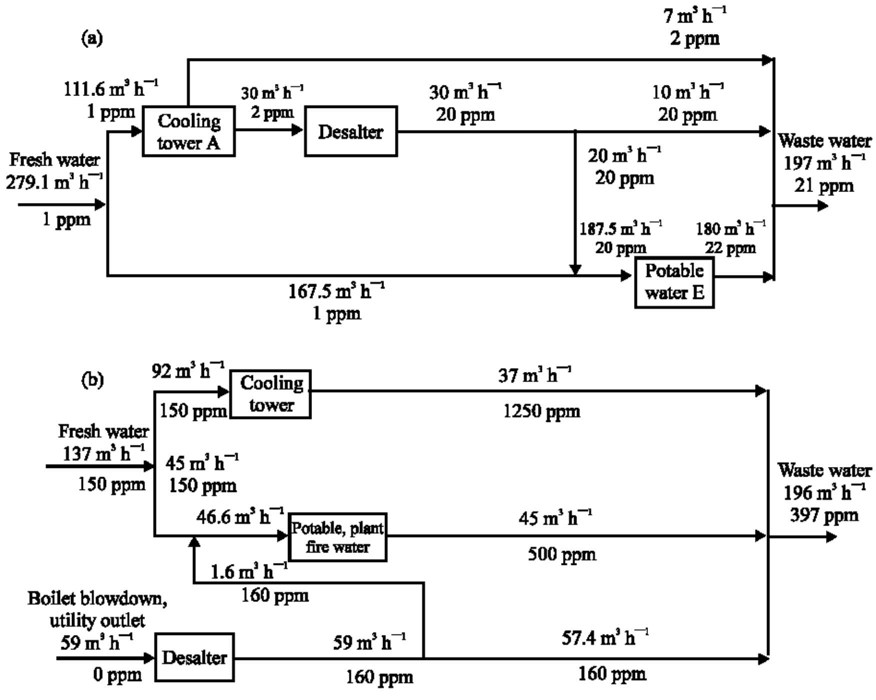

Fig. 6: Final Water network flowchart in terms of (a) SS and (b) H

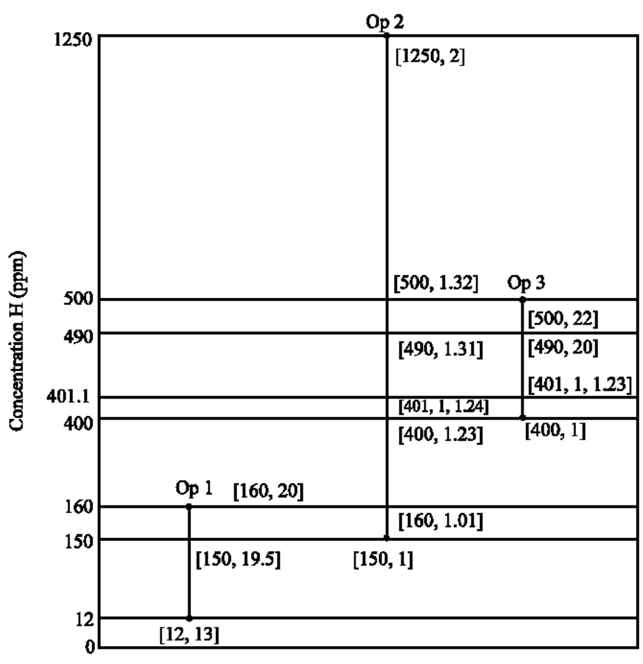

Fig. 7: Limiting water profile

After that, the actual flow-rate is determined for operations based on ratio $\theta_{i,n}$.

$$f_{i,n} = f_i \times \theta_{i,n} \tag{8}$$

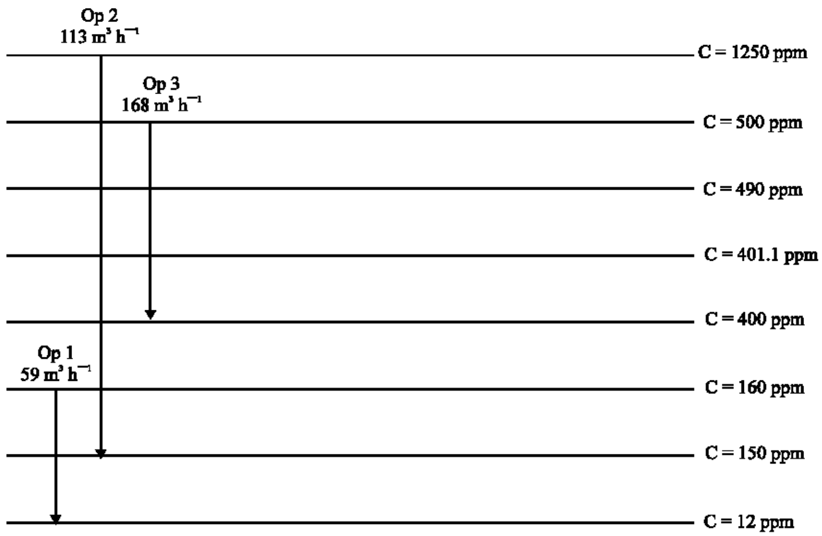

Fig. 8: The concentration interval boundaries and optional operations based on hardness

where, $f_{i,n}$ is the actual flow-rate and f_i is inlet flow-rate. In addition, the actual flow-rate can be calculated as follow:

$$f_{i,n} = T_{i,n} + q_{li,m\le n} + F_{i,n} = f_i \times \theta_{i,n} \tag{9}$$

where, $T_{i,n}$ is water flow-rate available for reuse within operation i at interval boundary n. $Q_{l,i,m\le n}$ is water flow-rate from operation i at interval boundary n that is supplied by (or reused from) operation l at interval boundary m smaller than n and $F_{i,n}$ is required freshwater for each operation in each interval boundary.

$\theta_{i,n}$ is obtained by following equation:

$$\theta_{i,n} = \max_j\left[\frac{C_{i,j,n+1} - C_{i,j,n}}{C_{i,j,n+1} - \overline{W}_{i,j,n}}\right] \tag{10}$$

where, $\overline{W}_{i,j,n}$ is flow-rate weighted average concentration of the current water sources and is calculated as:

$$\overline{W}_{i,j,n} = \frac{T_{i,n} \times W_{i,j,n} + \sum_l q_{li,m\le n} \times W_{lj,m\le n}}{T_{i,n} + \sum_l q_{li,m\le n} + F_{i,n}} \tag{11}$$

$$W_{i,j,n+1} = \overline{W}_{i,j,n} + \frac{f_i \times (C_{i,j,n+1} - C_{i,j,n})}{T_{i,n+1}} \tag{12}$$

where, $W_{i,jn+1}$ is outlet concentration of each operation and inlet concentration of next one.

To design the water network, at first concentration interval boundaries are drawn. Figure 8 shows concentration interval boundaries and optional operations based on Hardness. As it is represented, seven concentration interval boundaries have been plotted for three operations.

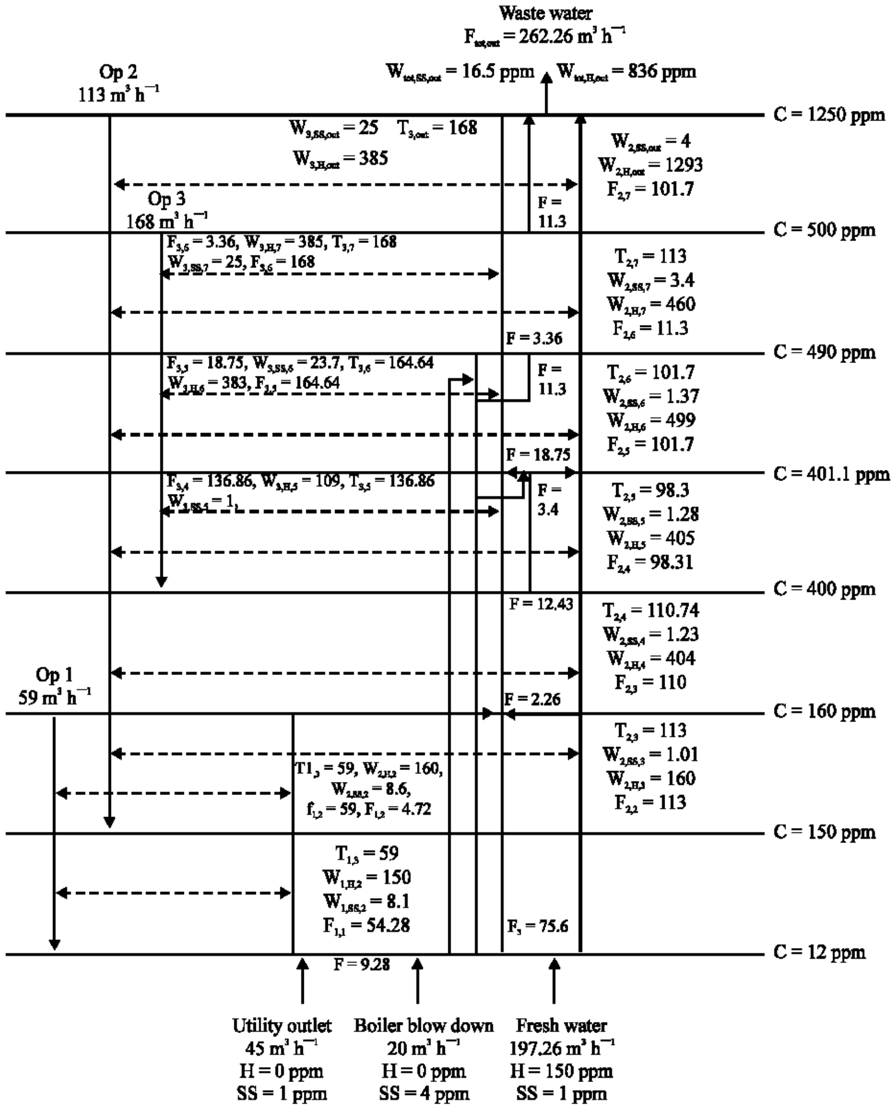

Fig. 9: Final water network diagram

Then water flow-rate is calculated for each operation in each interval boundary based on mass transfer of key contaminants (SS and H) by above-mentioned equations.

For example, water flow-rate for interval boundary 1 and operation 1 is calculated according to above equations as follow:

- Determining $\theta_{i,n}$

$$\theta_{1,1} = \max\ [\frac{150-12}{150-0}, \frac{19.5-13}{19.5-1}] = \max[0.92, 0.3] = 0.92$$

- Calculating required flow-rate

$$f_{i,1} = 0.92 \times 59 = 54.28 \text{ m}^3 \text{ h}^{-1}$$

For example, water flow-rate for interval boundary 1 and operation 1 is calculated according to above equations as follow:

- Determining $\theta_{i,n}$

$$\theta_{1,1} = \max\left[\frac{150-12}{150-0}, \frac{19.5-13}{19.5-1}\right] = \max[0.92, 0.3] = 0.92$$

- Calculating required flow-rate

$$f_{i,1} = 0.92 \times 59 = 54.28 \text{ m}^3 \text{ h}^{-1}$$

- Calculating outlet concentration

$$W_{1,SS,2} = 1 + \frac{59 \times (19.5-13)}{54.28} = 8.1 \text{ ppm}$$
$$W_{1,H,2} = 1 + \frac{59 \times (150-12)}{54.28} = 150 \text{ ppm}$$

Likewise, water flow-rate and outlet concentration are calculated for the rest of the operations in each interval boundary. Therefore, the final water diagram is drawn based on above calculations as Fig. 9.

Figure 10 and 11 show final stream flowchart for three optional operations and the whole water network, respectively. As it is seen, two mentioned contaminants were analyzed simultaneously based on their mass transfer. The results show that the amount of required water is reduced from 34 to 197.26 m^3 h^{-1} that is about 42%. As well as that, about 65 m^3 h^{-1} water is reused from boiler blow down and utility outlet into operation 2 and 3.

Analyzing both methods makes clear that amount of required water can be determined by mass transferring of suspended solids.

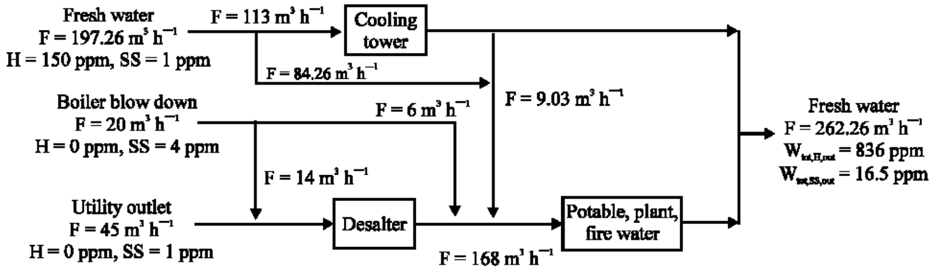

Fig. 10: Stream flowchart within three optional operations

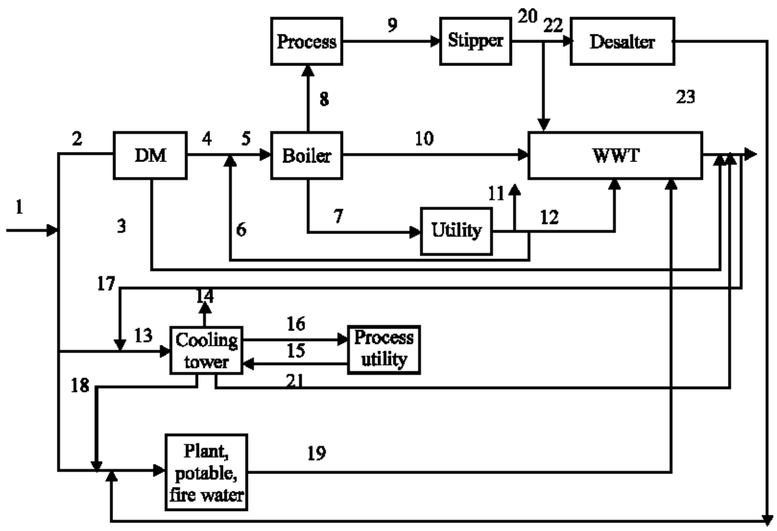

Fig. 11: Final flowchart for the whole water system

CONCLUSIONS

Nowadays, the crisis of water storage, discharging wastewater into the environment as well as expenditures of water supply and wastewater treatment are the main reasons for finding new methods to minimize freshwater utility in the different industries. Since, water is intensively used in petrochemical and allied industries especially petroleum refineries, water pinch technique is introduced as an efficient method to minimize water and wastewater.

In this research, two key contaminants including Suspended Solid (SS) and Hardness (H) have been considered to analyze the water network. These key contaminants once were analyzed separately as a single contaminant and the amount of required fresh water was calculated for both of them. In this stage, amount of freshwater was reduced about 60.9 $m^3 h^{-1}$ (17%) and 203 $m^3 h^{-1}$ (59.7%) in terms of Suspended Solids (SS) and Hardness (H), respectively. As it is seen water minimization within three optional operations for SS is less than H. So, this contaminant is a limiting contaminant and can be selected as a key contaminant. In the next stage, two mentioned contaminants were analyzed simultaneously based on their mass transfer so that mass transfer of a contaminant was analyzed with respect to another one. The results show that the amount of required water is reduced from 340 to 197.26 $m^3 h^{-1}$ that is about 42%. Analyzing both methods show that amount of required water can be determined by mass transfer of suspended solids. In addition, the method based on multiple contaminants gives more precise results rather than single contaminant. Therefore, it is suggested that more contaminants and operations are considered for study of water networks and reach water utility optimization based on key contaminant as well. Besides, mathematical optimization methods and computer programming can be used to obtain results that are more exact.

NOMENCLATURE

SS = Suspended solid
H = Hardness
Q = The value of a parameter according to the standards of power ministry
W = The weight of a parameter according to the standards of power ministry

$C_{i,in}^{lim}$ = Limiting inlet concentration
$C_{i,out}^{lim}$ = Limiting outlet concentration
$\Delta m_{i,tot}$ = Mass load of contaminant
$C_{i,in}^{lw}$ = Inlet water concentration
$m_{i,k}$ = Transferred mass load in interval boundary k for each operation i
$f_{i,k}$ = Required flow-rate for mass transferring in interval boundary k for each operation i
C_{k+1}^{*} = Concentration of upper interval boundary
C_{k-1}^{*} = Concentration of lower interval boundary
$C_{i,H,n}$ = Concentration of H within operation i in interval boundary n
$C_{i,H,in}$ = Inlet concentration of H in operation i
$C_{i,H,out}$ = Outlet concentration of H in operation i
$C_{i,SS,n}$ = Concentration of SS within operation i in interval boundary n
$C_{i,SS,in}$ = Inlet concentration of SS in operation i
$C_{i,SS,out}$ = Outlet concentration of SS in operation i
$f_{i,n}$ = Required flow-rate
f_i = Inlet flow-rate
$T_{i,n}$ = Outlet flow-rate
$q_{lim \le n}$ = Flow-rate from operation l within operation i with flow-rate q in previous interval boundary m
$F_{i,n}$ = Required water for each operation in each interval boundary
$\overline{W}_{i,j,n}$ = Average concentration of weighted flow-rate for current water sources
$W_{ij,n+1}$ = Outlet concentration of any operation as inlet concentration of next operation

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