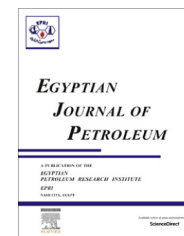




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FULL LENGTH ARTICLE

# Modeling water and hydrogen networks with partitioning regeneration units



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Received 12 April 2014; accepted 21 August 2014

Available online 17 April 2015

## KEYWORDS

Water regeneration;  
 Water network;  
 Hydrogen network;  
 Partitioning regenerator;  
 Process integration

**Abstract** Strict environment regulations in chemical and refinery industries lead to minimize resource consumption by designing utility networks within industrial process plants. The present study proposed a superstructure based optimization model for the synthesis of water and hydrogen networks with partitioning regenerators without mixing the regenerated sources. This method determines the number of partitioning regenerators needed for the regeneration of the sources. The number of the regenerators is based on the number of sources required to be treated for recovery. Each source is regenerated in an individual partitioning regenerator. Multiple regeneration systems can be employed to achieve minimum flowrate and costs. The formulation is linear in the regenerator balance equations. The optimized model is applied for two systems, partitioning regeneration systems of the fixed outlet impurity concentration and partitioning regeneration systems of the fixed impurity load removal ratio (RR) for water and hydrogen networks. Several case studies from the literature are solved to illustrate the ease and applicability of the proposed method.

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**Abbreviations:** CNHT, cracked naphtha hydrotreater; CCR, continuous catalytic reformer; DHT, diesel hydrotreater; Fuel, fuel gas system; HC, hydrocracker; MIP, mixed integer program; MILP, mixed integer linear program; MINLP, mixed integer non linear program; NHT, naphtha hydrotreater; NLP, non linear program; PSA, pressure swing adsorption; MMscfd, million standard cubic feet per day; AOC, annual operating cost; AT, annual operating time; Regen, regenerator;  $Y_F$ , impurity concentration of inlet stream of the regenerator;  $Y_P$ , impurity concentration of product stream of the regenerator;  $Y_r$ , impurity concentration of reject stream of the regenerator;  $\alpha_i$ , resource recovery of the regenerator; RR, impurity load removal ratio;  $i, j$ , from source  $i$  to sink  $j$ ;  $w_s$ , flowrate of source stream  $i$ ;  $w_{i,j}$ , flowrate from source  $i$  to sink  $j$ ;  $z_j$ , impurity concentration of sink  $j$  after mixing of sources fractions and fresh fractions;  $z_j^{\max}$ , maximum allowable impurity concentration of sink  $j$ ;  $z_j^{\min}$ , minimum allowable impurity concentration of sink  $j$ ;  $N_{\text{sources}}$ , total number of source streams;  $N_{\text{sinks}}$ , total number of sink streams;  $G_j$ , flowrate of sink  $j$  after mixing of sources fractions and fresh fractions;  $w_{\text{regen},i}$ , flowrate of regenerated source  $i$ ;  $w_{\text{regen},ip}$ , product stream flowrate of regenerator for each source  $i$ ;  $w_{\text{regen},ir}$ , reject stream flowrate of regenerator for each source  $i$ ;  $C_{\text{out}}$ , outlet concentration; DAF, dissolved air flotation tank; TSS, total suspended solids

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Peer review under responsibility of Egyptian Petroleum Research Institute.

<http://dx.doi.org/10.1016/j.ejpe.2015.02.008>

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## 1. Introduction

The efficiency with which energy and raw material are used within the process industries depends strongly on the way in which resources are distributed within a plant. The techniques for integrated design of processes can be applied to recover a process waste for reduction of water or hydrogen usage. Most works have modeled the regeneration units as either a fixed outlet concentration or fixed removal ratio (RR) [1–3]. The regeneration unit that consists of single inlet stream and single outlet stream is called a single-pass regeneration system. The regeneration unit that consists of a single inlet stream and two outlet streams is called a partitioning regeneration system. In the partitioning regeneration system, one of the two outlet streams always has a higher purity concentration (permeate stream) and the other outlet stream has a lower purity concentration (retentate stream). Examples of partitioning regeneration systems for water networks are membrane separation systems, flotation systems, gravity and settling systems, and filtration systems [4]. Examples of partitioning regeneration systems for Hydrogen networks are membrane separation systems, pressure swing adsorption, and cryogenic separation systems [18].

There are two methods for the systematic design of resource recovery networks:

- (1) The first method is based on the concept of pinch analysis. Pinch analysis is a tool for the estimation of the minimum resource requirement of a resource network before the system design. Resource pinch analysis requires the flowrate and purity constraints of the network. It is developed using some assumptions as constant operating conditions of the resource network, the resource streams are considered as a binary mixture of two components, any resource source (water or hydrogen source) may supply any resource sink (water or hydrogen sink) if the resource purity is higher than that of the resource sink, assuming pressure differences are ignored [5].
- (2) The second method is based on the application of the mathematical optimization technique. It can provide systematic design methods and can deal with possible practical constraints [6]. Mathematical method depends on estimation of an objective function which is subjected to constraints. The objective function and all the constraints may be linear, non linear, mixed integer program [MIP], mixed integer linear program [MILP] or mixed integer non linear program [MINLP] [7,8].

For water, many researchers used the pinch analysis technique as a promising tool in identifying various network targets prior to detailed design to reduce the fresh resource consumption and waste discharge. Aly et al. [9] presented the load problem table method. El Halwagi et al. [10] presented the material recovery pinch diagram. Manan et al. [11] and Foo et al. [12] presented the water cascade analysis technique.

Other works have been reported for the synthesis of utility hydrogen networks. Alves and Towler [13], defined the concepts of sink and source. A sink is a stream that consumes hydrogen from the hydrogen network while a source is defined as a stream supplying hydrogen to the network. El Halwagi

et al. [10], developed a rigorous and non-iterative graphical method to minimize the fresh resource consumption. Foo and Manan [14] put forward a numerical targeting method named the gas cascade analysis (GCA) to calculate the utility target. Zhao et al. [15], takes into account impurity concentration within a hydrogen network.

For regeneration processes, fresh resource and waste flowrates can be reduced by more recovery of waste. Feng et al. [16], and Bai et al. [17], have proposed targeting approaches for minimization the regeneration costs and treatment flowrates. Ng et al. [18,19], proposed a linear model to determine the minimum resource consumption for single impurity resource conservation networks, including an extension to determine the targets for resource conservation networks with interceptors. Bandyopadhyay and Cormos [20] used a graphical representation to address water management issues of integrated processes that involve regeneration and recycle through a single treatment unit.

Tahouni et al. [21] presented an optimization mathematical model for hydrogen management in petrochemical complexes based on setting a comprehensive superstructure model. The superstructure includes a purifier and a compressor.

In this work, it is assumed that multiple partitioning regeneration systems are used to achieve minimum flowrates and costs for a single contaminant resource network. Each source is sent to an individual partitioning regeneration unit. There is no mixing of inlet streams to the regeneration unit. The number of partitioning regeneration units is based on the number of sources to be regenerated. The optimized superstructure model can be applied onto the water partitioning regeneration systems and hydrogen partitioning regeneration systems.

## 2. Problem description

- The problems of resource network designs within the refinery formulate the optimization problems with the structure in which supply can be possible from all sources to all sinks. This is to supply resources having various degrees of contaminant and produced within the plants by adjusting the necessary specification of the degree of contaminant. This can reduce the production rate of fresh resource by increasing the amount of low-contaminant resource used.
- The objective of this work is to design that resource network to minimize fresh resource requirement of the plant.
- The resource distribution network of a refinery plant consists of a set of process sources and a set of process sinks.
- The set of process sources  $i = 1, 2, \dots, N_{\text{sources}}$ . Each source  $i$ , has a flowrate of  $W_i$ , and a composition of a single contaminant,  $y_i$ .
- The set of process sinks  $j = 1, 2, \dots, N_{\text{sinks}}$ . Each sink  $j$ , has a flowrate of  $G_j$ , and a maximum composition of a single contaminant,  $z_j$ .
- There is a fresh resource  $\text{Fresh}_j$  that can be purchased to supplement the sink demand.
- There are multiple partitioning regenerators for sources to be regenerated. Each regenerator has a single inlet stream and two outlet streams as shown in Fig. 1. The two product streams are reused /recycled to the process sinks in the network or rejected to the waste sink.

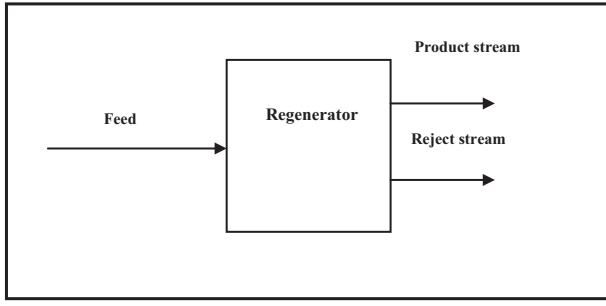


Figure 1 Schematic diagram of purifier [18].

### 3. Optimization model

A source-sink representation as described in Fig. 2, is the first step in the application of the mathematical technique. Each source is split into two fractions (of unknown flowrates). One fraction is used as a feed to the regeneration unit and the other fraction is integrated with the various sinks and waste sink. The two product streams from each regeneration unit are also integrated with all process sinks and waste sink. The fresh resource is allowed to split and integrate with all process sinks except the waste sink.

The objective function is to minimize the annual operating cost of the fresh and regenerated sources [18].

$$AOC = (A \text{ Fresh} + B \text{ wregen})AT \quad (1)$$

where:  $wregen$  is the flowrate of the regenerated sources,  $A$  and  $B$  are the unit costs of fresh and regenerated sources and  $AT$  refers to the annual operating time (8000 h/yr).

For the regeneration unit, the two concentrations in parts per million of the product ( $Y_p$ ) and residue stream ( $Y_r$ ) can be calculated by using Eqs. (2) and (3) [18]:

$$Y_p = \frac{Y_F(1 - RP)10^6}{\alpha_i(10^6 - Y_F) + Y_F(1 - RP)} \quad (2)$$

where:

$\alpha_i$  is the fluid recovery factor. The resource recovery factor is defined as the fraction of the feed stream that passes through the regeneration system into the higher-quality stream and is assumed to be constant for a given regeneration process [18].

RP is the removal ratio.

$Y_F$  is the concentration of source stream used in the regeneration.

$$Y_r = \frac{[Y_F - \left(\frac{10^6 - Y_F}{10^6 - Y_p}\right)\alpha_i Y_p]}{[1 - \left(\frac{10^6 - Y_F}{10^6 - Y_p}\right)\alpha_i]} \quad (3)$$

For both Eqs. (2) and (3), the term  $(10^6)$  can be changed depending on the concentration units used (100 for mass percentage and 1 for mass fraction) [18].

Each source  $i$ , is split into two fractions. The first flowrate is described as  $w_i$ , and the other fraction is described as  $wregen_i$ , as shown in Fig. 2.

$$w_i = \sum_{j=1}^{N_{sinks}} w_{ij} + waste_i \quad \text{for } i = 1, 2, 3, \dots, N_{sources} \quad (4)$$

$$wregen = \sum_{i=1}^{N_{sources}} wregen_i \quad (5)$$

$$wregen_i = wregen_{ip} + wregen_{ir} \quad (6)$$

$$wregen_{ip} = \sum_{j=1}^{N_{sinks}} wregen_{ipj} + wastewregen_{ip} \quad \text{for } i = 1, 2, 3, \dots, N_{sources} \quad (7)$$

$wregen_{ipj}$  is the flowrate sent from the top product of the regenerator to the  $j^{\text{th}}$  sinks.

$$wregen_{ir} = \sum_{j=1}^{N_{sinks}} wregen_{irj} + wastewregen_{ir} \quad \text{for } i = 1, 2, 3, \dots, N_{sources} \quad (8)$$

$wregen_{irj}$  is the flowrate sent from the residue product of the regenerator to the  $j^{\text{th}}$  sinks.

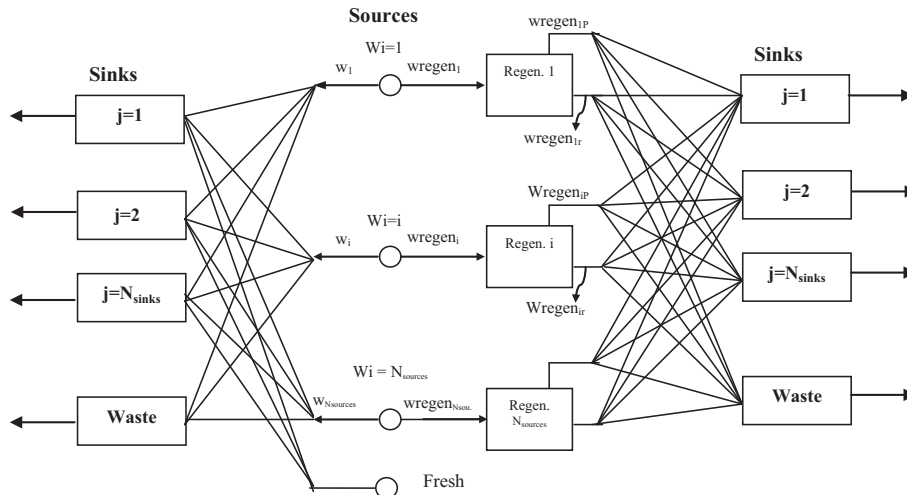


Figure 2 Structural representation.

$$waste = \sum_{i=1}^{N_{sources}} waste_i + \sum_{i=1}^{N_{sources}} wasteregen_{ip} + \sum_{i=1}^{N_{sources}} wasteregen_{ir} \quad (9)$$

The following step is the mixing of the split fractions into a feed to the  $j^{\text{th}}$  sinks.

$$G_j = \sum_{i=1}^{N_{sources}} w_{i,j} + \sum_{i=1}^{N_{sources}} w_{regen_{ipj}} + \sum_{i=1}^{N_{sources}} w_{regen_{irj}} + Fresh_j$$

for  $j = 1, 2, 3, \dots, N_{sinks}$  (10)

$$G_j z_j = \sum_{i=1}^{N_{sources}} w_{i,j} y_i + \sum_{i=1}^{N_{sources}} w_{regen_{ipj}} y_{ip} + \sum_{i=1}^{N_{sources}} w_{regen_{irj}} y_{ir} + Fresh_j x_j \quad \text{for } j = 1, 2, 3, \dots, N_{sinks} \quad (11)$$

$$Fresh = \sum_{j=1}^{N_{sinks}} Fresh_j \quad \text{for } j = 1, 2, 3, \dots, N_{sinks} \quad (12)$$

$$w_i = W_i - w_{regen_{ip}} - w_{regen_{ir}} \quad (13)$$

$$w_i y_i = W_i y_i - w_{regen_{ip}} y_{ip} - w_{regen_{ir}} y_{ir} \quad (14)$$

$$w_{ij} \geq 0 \quad (15)$$

$$w_{regen_{ipj}} \geq 0 \quad (16)$$

$$w_{regen_{irj}} \geq 0 \quad (17)$$

$$z_j^{\min} \leq z_j \leq z_j^{\max} \quad (18)$$

$$fresh_j \geq 0 \quad (19)$$

$$\text{for } i = 1, 2, 3, \dots, N_{sources}$$

$$\text{for } j = 1, 2, 3, \dots, N_{sinks}$$

Regeneration systems are generally categorized as fixed –  $C_{out}$  and fixed removal ratio (RR) model [18]. The proposed method is applied on these two regeneration systems.

In the regeneration system of the fixed –  $C_{out}$  model, the product stream is always at a constant concentration. For instance a water source of 100 ppm that is fed to the regeneration system of a fixed –  $C_{out}$  model (with  $C_{out} = 20$  ppm) will always exit at 20 ppm [18]. Examples of this kind of regeneration system include dead end filtration, membrane separation and pressure swing adsorption (PSA).

The product stream of the regeneration system of the fixed removal ratio model is always at a constant removal ratio. For such model, if the water source fed to the regeneration unit with  $RR = 0.9$ , the product stream will exit at 10 ppm [18]. Example of this kind of regeneration system is a dissolved air flotation (DAF) tank that can reduce the total suspended solid (TSS) concentration in wastewater.

#### 4. Case studies

Three case studies are solved to illustrate the application of the proposed method. Two case studies are solved to the partitioning regeneration system with fixed outlet concentrations and the third case study is solved to the partitioning regeneration system with a fixed removal ratio.

##### 4.1. Case study 1 (partitioning regeneration system of the fixed – $C_{out}$ model)

This case study consists of four water sources and four water sinks [3,22] with contaminant concentrations and flowrates as described in Table 1. A regenerator with liquid phase recovery  $\alpha_l = 0.5$  and fixed lean stream outlet concentration  $Y_p = 30$  mg/l have been used for the regeneration process. The existing fresh water and wastewater flowrates in this water network are 300 and 280 t/h, respectively. The fresh water demand limit is 50 t/h.

By applying El Halwagi et al. method [10] for reuse/recycle system it is found that:

The minimum fresh water and wastewater flowrate for this case study were determined to be 70 and 50 t/h, respectively and these results agree with the results reported in other earlier works [3]. The unit costs of fresh water (A) and regenerated water (B) are taken as \$ 1/ton and \$ 0.6/ton respectively [3,22]. The impurity concentration of the reject stream ( $Y_r$ ), for each source is determined by using Eq. (3).

By applying the proposed method and using Lingo program v.11 general solver to get the minimum objective function Eq. (1) subjected to the constraints in Eqs. (4)–(19), and a new constrain related to this example ( $Fresh_{max} \leq 50$ ), it is found that: the minimum fresh water flowrate is targeted as 50 t/h, the wastewater discharge flowrate is 30 t/h and the annual operating cost (AOC) is \$ 640,038.5/yr. The sources used in the regeneration are 30 t/h from source 3 sent to the first regenerator and 20 t/h from source 4 sent to the second regenerator as shown in Fig. 3. Waste water is targeted as 30 t/h and sent to the effluent. These results agree with the previous work by Tan et al. [3].

From the results, it is noted that the same fresh water and regenerated water are achieved in the two cases, mixing sources as described by Tan et al. [3] and without mixing the sources by the proposed method.

This work is different from the previous work of Tan et al. [3] in two main points:

- (1) Tan et al. work [3] used one partitioning regeneration unit with mixing the regenerated sources. The two product streams of the regeneration unit are allowed to reuse/recycle in the network.

In this work, two individual partitioning regeneration units are used for the two regenerated sources. There is no mixing of the regenerated sources. The product streams for each partitioning regeneration unit are allowed to reuse/recycle in the network.

- (2) The formulation of the previous Tan et al. work [3] is nonlinear and that increases the computational problems.

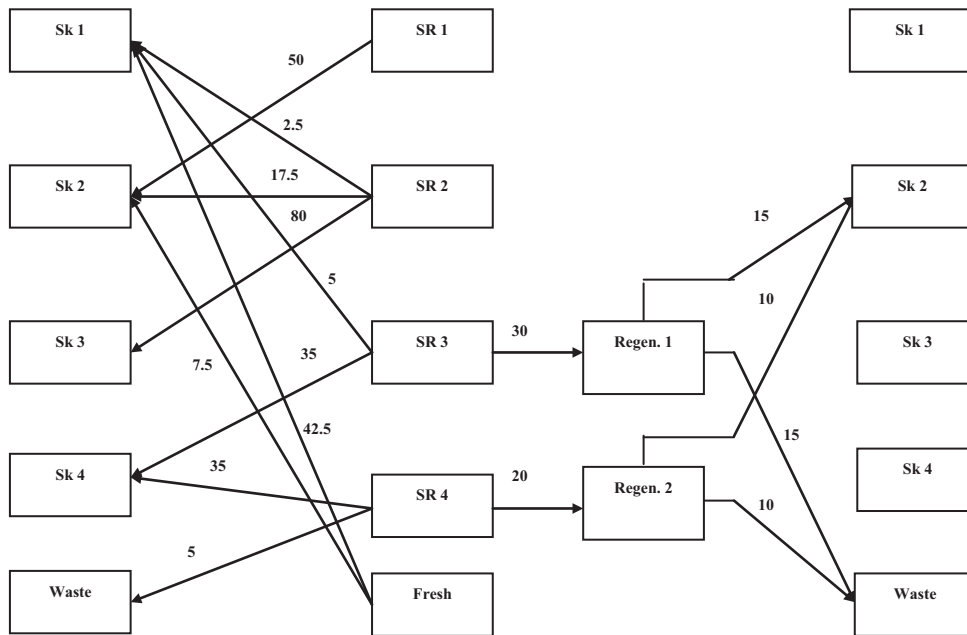
In this work, the proposed superstructure model formulation is linear and easy to apply.

##### 4.2. Case study 2 (partitioning regeneration system of the fixed – $C_{out}$ model)

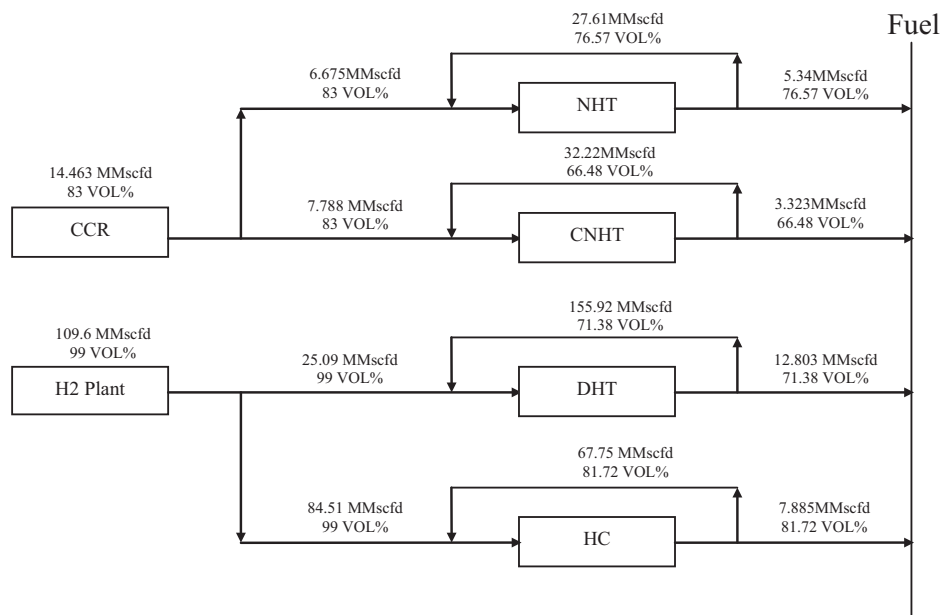
This case study is represented by Jia and Zhang [6] for hydrogen network. Each stream is assumed to be a mixture of two

**Table 1** Water process data for case study 1.

Water sinks	Flowrates (t/h)	Contaminant concentration (mg/l)	Water sources	Flowrates (t/h)	Contaminant concentration (mg/l)
1	50	20	1	50	50
2	100	50	2	100	100
3	80	100	3	70	150
4	70	200	4	60	250



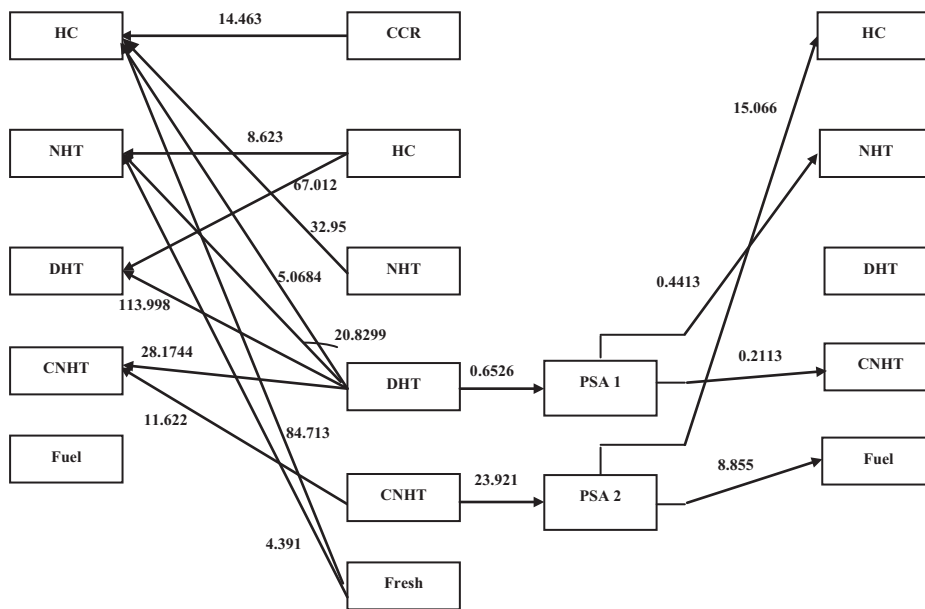
**Figure 3** Optimal water network for case study 1 (flowrate in t/h).



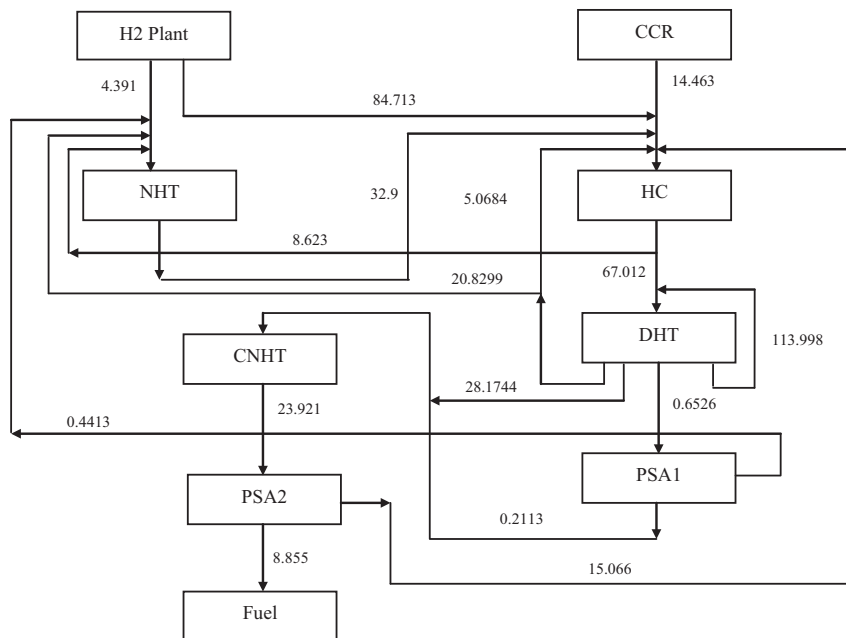
**Figure 4** Hydrogen network for case study 2.

**Table 2** Hydrogen process data for case study 2.

Hydrogen sources	Flowrates (MMscfd)	Impurity fraction	Hydrogen sinks (Demands)	Flowrates (MMscfd)	Impurity fraction
NHT	32.95	23.43	NHT	34.285	22.178
CNHT	35.543	33.52	CNHT	40.008	30.304
DHT	168.723	28.62	DHT	181.01	24.792
HC	75.635	18.28	HC	152.26	8.689
CCR	14.463	17			
Fresh	-	1			



**Figure 5** Optimal hydrogen network for case study 2 (flowrate in MMscfd).



**Figure 6** Optimized Hydrogen network flowsheet for case study 2 (flowrate in MMscfd).

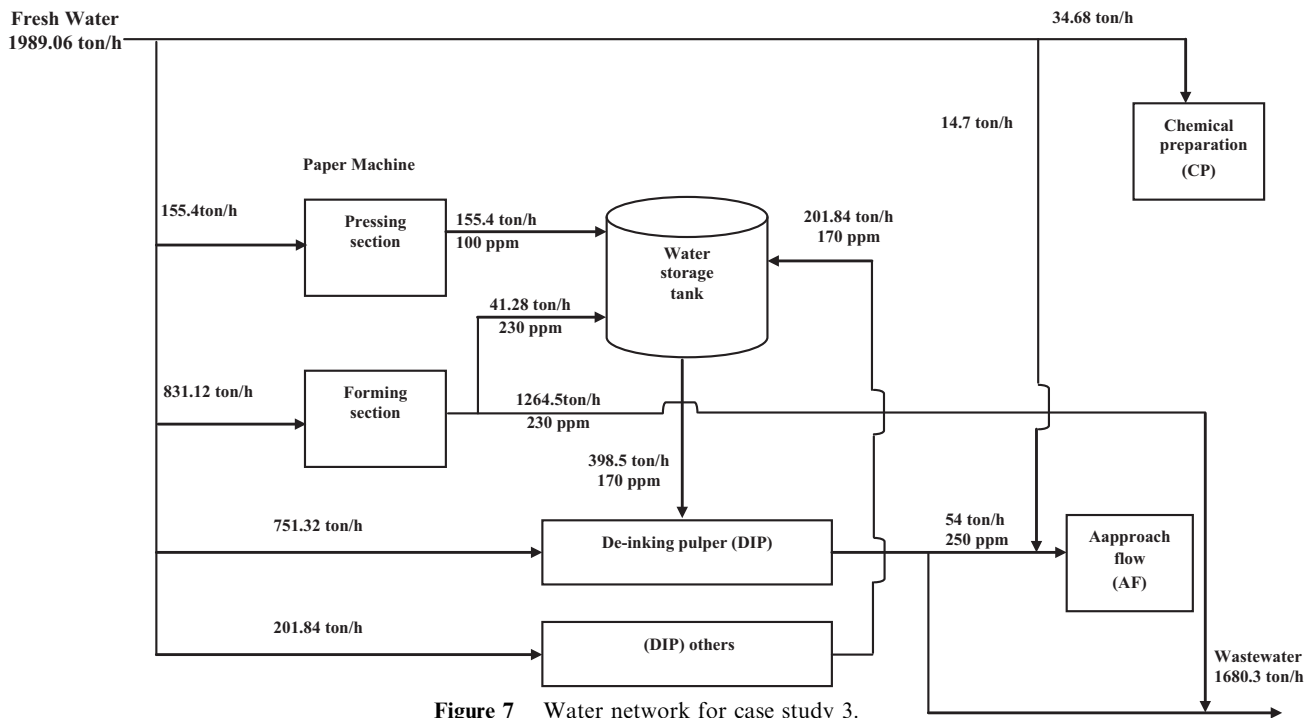


Figure 7 Water network for case study 3.

Table 3 Limiting data for case study 3.

j	Water sink (SK <sub>j</sub> )	Flowrates (ton/s)	Impurity concentration (ppm)	Water source (SR <sub>i</sub> )	Flowrates (ton/s)	Impurity concentration (ppm)
1	Pressing section	155.4	20	Pressing section	155.4	100
2	Forming section	831.12	80	Forming section	1305.78	230
3	DIP-others	201.84	100	DIP-others	201.84	170
4	De-inking pulper (DIP)	1149.84	200	De-inking pulper (DIP)	469.8	250
5	Chemical preparation (CP)	34.68	20			
6	Approach flow (AF)	68.7	200			

components hydrogen and methane. Fig. 4 shows the hydrogen network. The hydrogen plant and the CCR are the main two hydrogen producers in the network. Also, there are three different hydrotreaters and one hydrocracker. The hydrocracker is the largest hydrogen consumer. Table 2 shows the relevant data. The regeneration in this hydrogen network is achieved through a pressure-swing adsorption (PSA). The PSA product purity  $Y_p$  is specified as 95% and the recovery  $\alpha_r = 90\%$ . The minimum fresh hydrogen and purge gas flowrate for this case study were determined to be 102.365 MMscfd and 22.116 MMscfd, respectively. The unit cost of the fresh hydrogen (A) and the regenerated Hydrogen (B) are taken as \$1/MMscfd and \$0.01/MMscfd, respectively. The impurity concentration of the reject stream ( $Y_r$ ), for each regenerator is determined by using Eq. (3). For source 1,  $Y_r = 61.15$ , for source 2,  $Y_r = 63.81$ , for source 3,  $Y_r = 72.116$ , for source 4,  $Y_r = 77.953$ , and for source 5,  $Y_r = 82.042$  vol%.

When the proposed method is applied, it is found that: the minimum fresh hydrogen flowrate is targeted as 89.104 MMscfd, the hydrogen discharge flowrate is 8.855 MMscfd and the annual operating cost (AOC) is \$714,799/yr. The sources used in the regeneration are source 4 and source 5.

0.6526 MMscfd from source 4 is sent to the first PSA and 23.921 MMscfd from source 5 is sent to the second PSA as shown in Figs. 5 and 6.

#### 4.3. Case study 3 (partitioning regeneration system of the fixed-removal ratio (RR) model)

The water network of a paper-milling process is shown in Fig. 7, with limiting data given in Table 3 [18]. The figure shows that: the fresh water and wastewater flowrates are 1989.06 and 1680.3 ton/h, respectively. The minimum fresh water and wastewater flowrates were determined as 848 and 539 ton/h, respectively and these results are the same as previous work [12]. For more reduction of fresh water, a regeneration unit can be added to the network. A dissolved air flotation (DAF) tank that can reduce the total suspended solid (TSS) concentration is added. A DAF unit can be modeled as having a single inlet stream and two outlet streams with constant impurity removal ratio [18].

In this case study, a DAF tank with  $RR = 0.9$  and  $\alpha_r = 0.98$  is used. The unit cost of fresh water (A) and regenerated water (B) are taken as \$1/ton and \$0.001/ton, respectively [18]. The impurity concentration of the product stream

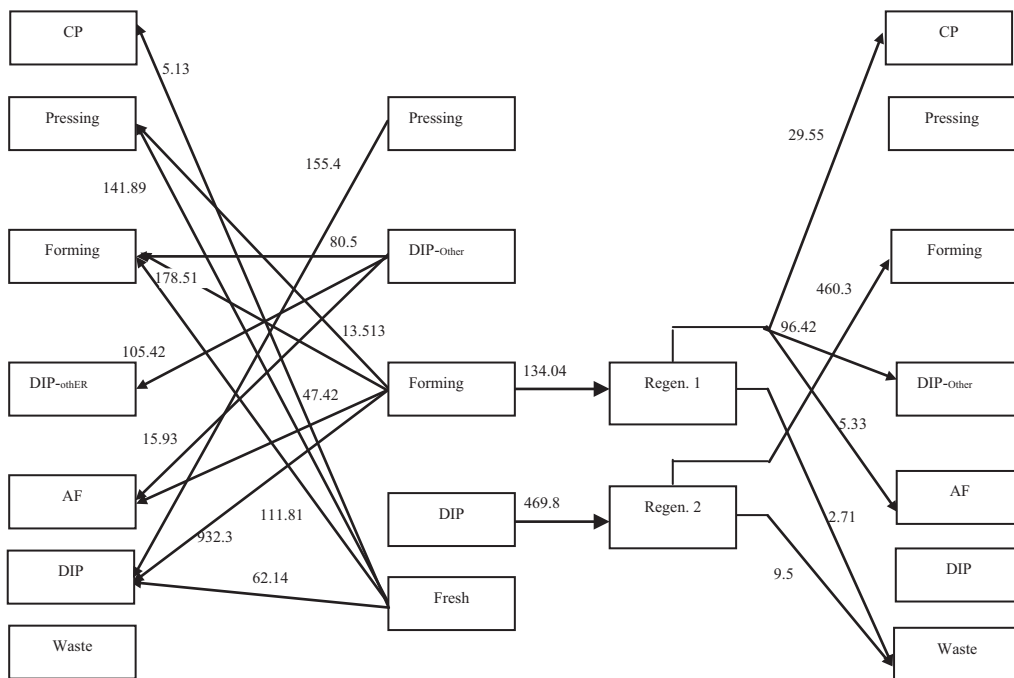


Figure 8 Optimal water network for case study 3 (flowrate in t/h).

$Y_p$  and the retentate (reject) stream,  $Y_r$ , for each source is determined by using Eqs. (2) and (3).

By applying the proposed method it is found that:

The minimum fresh water flowrate is targeted as 320.97 ton/h, the wastewater discharge flowrate is 12.21 ton/h and the annual

operating cost (AOC) is \$ 2,572,568/yr. The sources used in the regeneration are 134.04 ton/h from forming section (source 2) and all DIP (source 4) flowrate 469.8 ton/h are sent to two different DAF tanks as described in Figs. 8 and 9. These results agree with the previous work by Ng et al. [18].

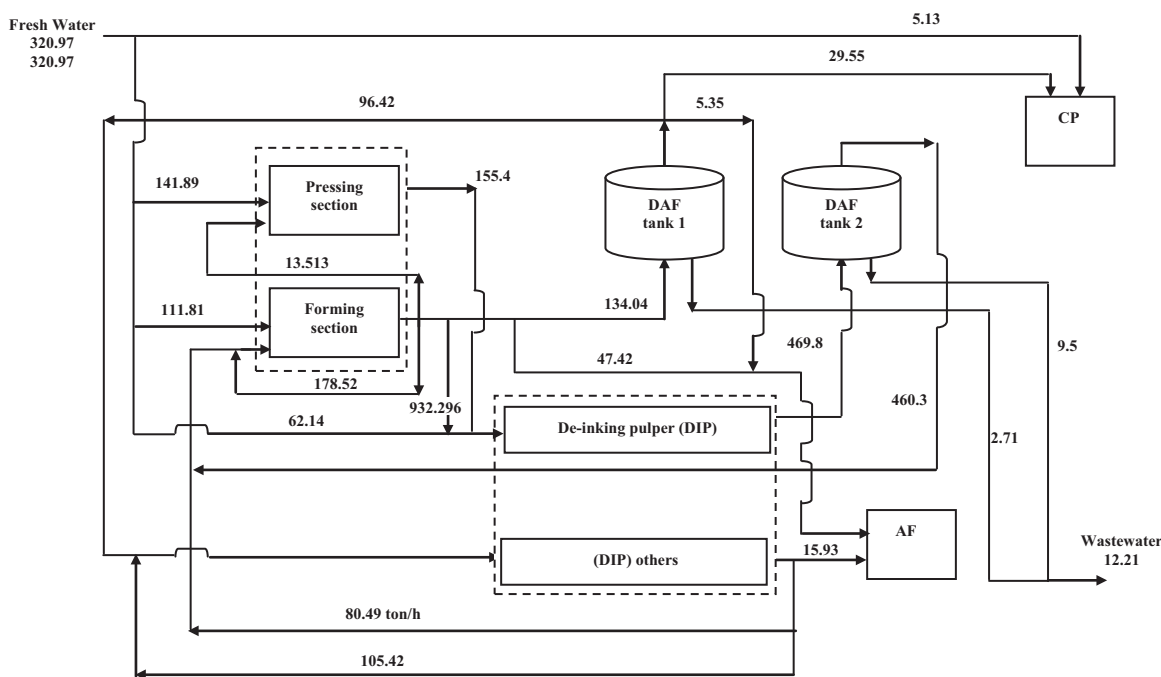


Figure 9 Optimized Water network flowsheet design for case study 3 (flowrate in t/h).



## 5. Conclusion

In this work, a linear optimization model is represented to solve waste resource minimization problem by using partitioning regeneration system. The two product streams of the regenerator can be reused/recycled with the sink streams in the network. This model can be applied on water and hydrogen networks. This method indicates the number of partitioning regenerators required for the regeneration based on the number of required regenerated sources. The model is applied onto the partitioning regeneration systems with fixed outlet concentration and systems with fixed removal ratios.

Several case studies from the literature have been solved with the proposed model and the results showed that;

- (1) All results agree with the previous results in the literature.
- (2) The results proved that the minimum fresh resource and minimum discharge flowrate are the same in the two cases, regeneration in individual regenerators without mixing the regenerated sources as described by the proposed method and in the case of using one regenerator and mixing the regenerated sources as described by Tan et al. work [3].

Future work will focus on the extension of the method to multi-contaminant resource problems.

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