SETTING TARGETS FOR WATER AND HYDROGEN NETWORKS USING CASCADE ANALYSIS

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ABSTRACT

This paper describes the cascade analysis technique as a new method to establish the utility targets for water and hydrogen networks. Cascade analysis is a numerical alternative to the graphical targeting technique known as the surplus diagram. The cascade analysis is to the surplus diagram in water and hydrogen integration as problem table analysis is to the grand composite curves in heat integration. By eliminating the tedious iterative steps of the surplus diagram, the cascade analysis can quickly yield accurate utility targets and pinch point locations for water or hydrogen network, thereby offering a key complimentary role to the surplus diagram in the design and retrofit of a process (water or hydrogen) network. In water pinch analysis, this numerical tool is known as *water cascade analysis* (WCA) while in hydrogen pinch analysis, it is known as *hydrogen cascade analysis* (HCA).

INTRODUCTION

Mass integration is a holistic methodology that provides a fundamental understanding of the global mass flow and allocation within a process (El-Halwagi, 1997). This concept was initiated in the late 1980s when El-Halwagi and Manousiouthakis (1989) extended the concept of heat recovery pinch into mass transfer processes, in particular, for the optimal synthesis of mass exchange network (MEN). The MEN synthesis concept was then extended to a much wider range of problems. These problems include the automated synthesis procedure for MEN (El-Halwagi and Manousiouthakis, 1990a), simultaneous synthesis of mass exchange and regeneration networks (El-Halwagi and Manousiouthakis, 1990b); synthesis of reactive MEN (El-Halwagi and Srinivas, 1992; Srinivas and El-Halwagi, 1994a); synthesis of combined heat and reactive MEN (Srinivas and El-Halwagi, 1994b); synthesis of waste-interception networks (El-Halwagi, Hamad and Garrison, 1996); heat induced separation networks (Dunn, Srinivas and El-Halwagi, 1995; Dye, Berry and Ng, 1995; Richburg and El-Halwagi, 1995; El-Halwagi, Srinivas and Dunn, 1995).

Besides, two of the special cases of MEN synthesis that receive a lot of attention from both academia and industrial are the *water recovery network*, which is often called the "water pinch analysis" (Wang and Smith, 1994; 1995; Dhole et al., 1996; Olesen and Polley, 1997; Sorin and Bédard, 1999; Polley and Polley, 2000; Bagajewicz, 2000; Dunn and Wenzel, 2001a, 2001b; Xiao and Seider, 2001; Hallale, 2002; Tan, Manan and Foo, 2002; Foo *et al.*, 2003) as well as the *hydrogen distribution network*, or the "hydrogen pinch analysis" (Towler *et al.*, 1996; Hallale and Liu, 2001; Alves and Towler, 2002; Hallale, Moore and Vauk, 2002). The main driving force behind these activities is that, apart from the stringent environmental regulation, the fresh utility (fresh water and hydrogen supply) cost and waste treatment cost have both rose significantly in these recent years. The process plants are now taking more serious measurement towards the minimisation of utility (fresh water and hydrogen use) consumption. This corresponds to a reduced waste generation as a mean to reduce production cost and to ensure a sustainable business activities growth.

This paper will firstly present the *Water Cascade Analysis* (WCA) as a new technique to establish the minimum water and wastewater targets for water recovery network synthesis. The WCA is a numerical alternative to the graphical water targeting technique known as the water surplus diagram (Hallale, 2002). By eliminating the tedious iterative steps of the water surplus diagram, the WCA can quickly yield accurate water targets and pinch point locations for a *maximum water recovery* (MWR) network, thereby offering a key complimentary role to the water surplus diagram in the design and retrofit of a water recovery network.

The concept of cascade analysis is next extended into another special case of mass integration, i.e. the hydrogen distribution network synthesis. With appropriate modification, the concept of cascade analysis is used to determine the minimum hydrogen consumption (minimum utility) for a refinery hydrogen network. This new tabular tool is called the Hydrogen Cascade Analysis (HCA). HCA yields accurate hydrogen target and pinch point(s) locations for a hydrogen network by eliminating the tedious iterative steps of the hydrogen surplus diagram.

CASCADE ANALYSIS FOR WATER RECOVERY NETWORK SYNTHESIS

The current drive towards environmental sustainability and the rising costs of fresh water and effluent treatment have encouraged the process industry to find new ways to reduce freshwater consumption and wastewater generation. Concurrently, the development of systematic techniques for water reduction, reuse and recycling within a process plant has seen extensive progress. The advent of Water Pinch Analysis (WPA) as a tool for the design of optimal water recovery network has been one of the most significant advances in the area of water minimization over the last ten years. The WPA technique as proposed by Wang and Smith (1994) generally considers the potential of using fresh or recycle water as a lean stream to absorb certain contaminants from various process operations, provided there exist a driving force for mass transfer. Maximising water reuse and recycling can minimise freshwater consumption and wastewater generation.

Types of water-using operations

Water-using operations in a process plant can generally be classified into the mass transfer-based and the non-mass transfer-based operations. A mass transfer-based water-using operation is characterised by the preferential transfer of species from a rich stream to water, which is being utilised as a mass separating agent (MSA). A typical example of such operation is the cleaning of a process vessel using fresh or recycle water. During cleaning, water is fed into the vessel (as a demand) while wastewater is generated (as a source) as shown in **Figure 1a**. Another example of the mass transfer-based water-using operation is the absorption process where water is the MSA used to remove contaminants such as H_2S and SO_2 from a sour gas stream (see **Figure 1b**).

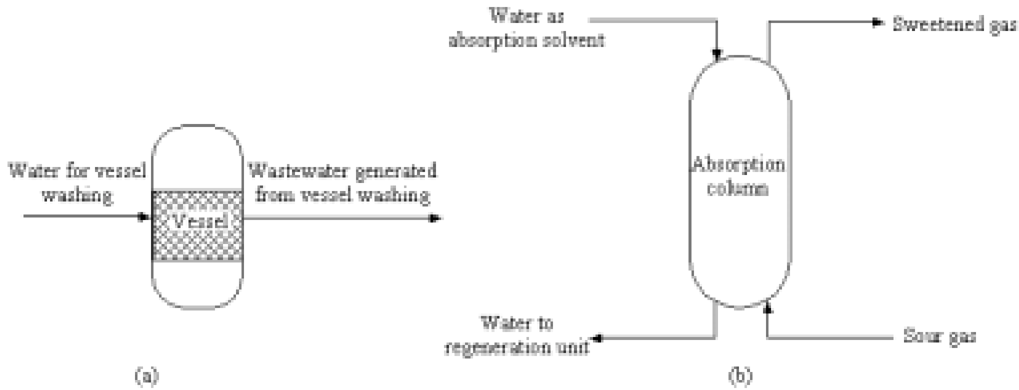

Figure 1. Mass transfer-based water-using operations: (a) Vessel washing; (b) Sour gas absorption where water demand and water source exist.

The non-mass transfer-based water-using operation covers functions of water other than as a mass separating agent. A typical example includes water being fed as a raw material, or being withdrawn as a product or a by-product in a chemical reaction (see **Figure 2**). The non-mass transfer-based operation also covers cases where water is being utilised as heating or cooling media. For such operations, usually, only water demands or water sources exist. Note that, for the non-mass transfer-based water-using operations, the water flowrate is more important than the amount of contaminant accumulated. Though the conventional water network studies have focused on the mass transfer-based model (Takama *et al.*, 1980; Wang and Smith, 1994), recent studies have shown that the non-mass transfer-based water-using operations are also important to consider (Dhole *et al.*, 1996; Hallale, 2002).

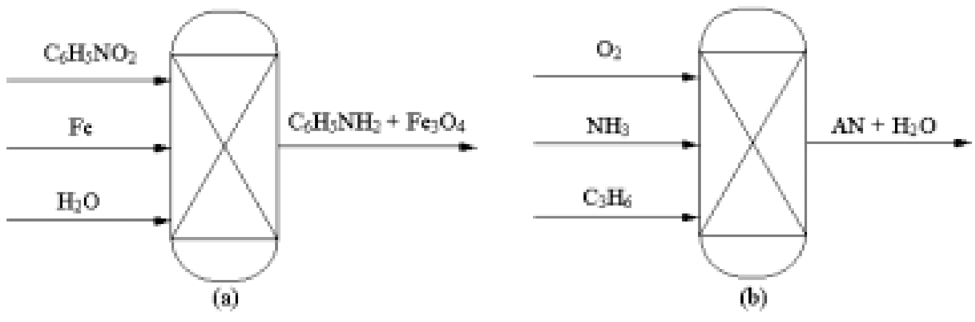

Figure 2. Non-mass transfer-based water-using operations: (a) a reactor that consumes water in aniline production; (b) a reactor that produces water as a byproduct in acrylonitrile (AN) production

Previous work on water targeting

In targeting the minimum utility requirements and in locating the pinch points, the graphical technique such as the composite curves and the numerical technique such as the problem table have both been used in the heat (Linnhoff *et al.*, 1982), mass (El-Halwagi, 1997) and water recovery problems (Mann and Liu, 1999) that are based on pinch analysis. Why then are both techniques usually used together even though they apparently yield the same information? The answer lies in the complimentary roles they play in pinch analysis. The graphical tool like the composite curves is vital in terms of providing an understanding of the overall heat and mass transfer potentials in a process. On the other hand, the numerical targeting tools like the problem table in heat integration (Linnhoff *et al.*, 1982) or the composition interval table (CIT) in mass integration (El-Halwagi and Manousiouthakis, 1989) are advantageous from the point of view of accuracy and speed, and therefore, are more amenable to computer programming. Note that the majority of researchers have extended the use of composite curves and problem table analysis established for heat recovery based on pinch analysis to the mass recovery, and later, to the water recovery problems.

Wang and Smith (1994) introduced the plot of composition versus contaminant mass load, or the water composite curves, for which they termed as the limiting water profile, for graphical water targeting. They also made use of the composition interval table from mass integration to pinpoint the pinch location and generate the exact minimum water targets prior to network design. The limiting water profile represents a major stride in establishing the baseline water requirement and wastewater generation for a process. However, its applicability is only limited to mass transfer-based operations. Water as cooling and heating media in cooling towers and boilers, and as a reactant may not be appropriately represented as mass transfer operations. To overcome this limitation, Dhole *et al.* (1996) introduced the water source and demand composite curves. They also suggested process changes like mixing and bypassing to further reduce the fresh water consumption. However, Polley and Polley (2000) later pointed out that, unless the correct stream mixing system was identified, the apparent targets generated by Dhole's technique could be substantially higher than the true minimum fresh water and wastewater targets.

Sorin and Bédard (1999) developed the Evolutionary Table to numerically determine the fresh water and wastewater targets. They pointed out that the targeting technique introduced by Dhole *et al.* (1996) could result in a number of "local" pinch points, which might not necessarily be the actual or the "global" pinch points. However, Hallale (2002) recently showed that, when more than one global pinch points occurred in water-using processes, the Evolutionary Table failed to locate them correctly.

Hallale (2002) presented an alternative graphical method called the water surplus diagram (**Figure 3**) to target the minimum fresh water and wastewater. The method, which was adapted from the hydrogen pinch analysis (Alves and Towler, 2002), had a similar representation to the water source and demand composite curves proposed by Dhole *et al.* (1996), thereby was not limited to the mass transfer-based operations. The new representation by Hallale (2002) could handle all mixing possibilities and yet resulted in the true pinch point and reuse target.

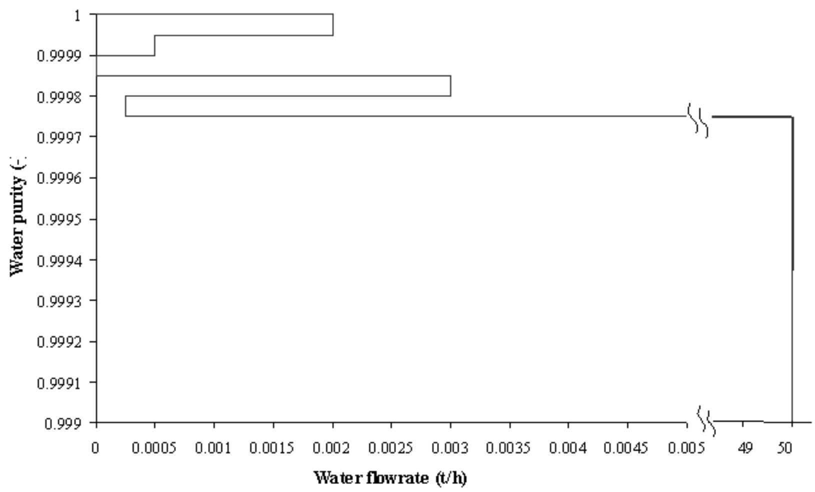

Figure 3. Water surplus diagram by Hallale (2002)

However, the water surplus diagram has the same drawbacks like the composite curves. It is tedious and time consuming to draw as it involves trial an error to find the pinch points and water targets. Besides, it has limitations in terms of generating highly accurate targets due to its graphical nature. The tedious iterative procedure to construct the water surplus diagram is shown in **Figure 4**. In order to eliminate the trial an error steps and compliment the graphical method, there is a need for a numerical equivalent of the water surplus diagram similar to the composition interval table in mass integration. This is the subject of this paper.

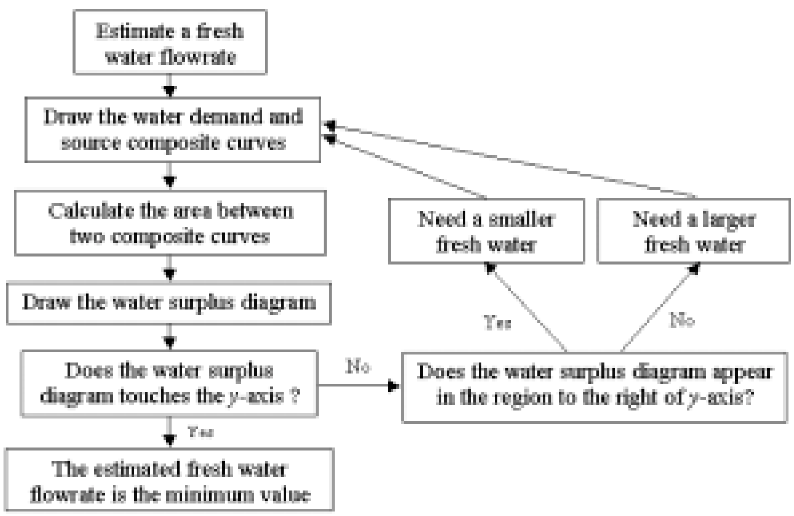

Figure 4. The tedious iterative steps of constructing the water surplus diagram

This work presents the *water cascade analysis* (WCA), a new numerical technique to establish the minimum water and wastewater targets in a water recovery network. The WCA eliminates any tedious iterative step to quickly yield the exact utility targets and the pinch location(s). As in the case of the water surplus diagram, the WCA is not limited to mass transfer-based operations and is therefore applicable to a wide range of water using operations. A case study on water minimisation involving acrylonitrile production from El-Halwagi (1997) is used to illustrate the procedure for water and wastewater targeting using the WCA.

Acrylonitrile case study

Acrylonitrile (AN) is produced via the vapour-phase ammoxidation of propylene that takes place in a fluidised-bed reactor at 450ºC and 2 atm, according to Equation 1.

$$C_3H_6 + NH_3 + 1.5O_2 \xrightarrow{catalyst} C_3H_3N + 3H_2O \qquad \textbf{(1)}$$

This is a single-pass reaction with almost complete conversion of propylene. Products from the reactor is cooled and partially condensed. The reactor off-gas is sent to a scrubber that uses fresh water as the scrubbing agent. The bottom product from the scrubber is separated into the aqueous layer and an organic layer in a decanter. The organic layer is later fractionated in a slightly vacuumed distillation column that is induced by a steam-jet ejector. **Figure 5** shows the process flow diagram for AN production along with the pertinent material balance data.

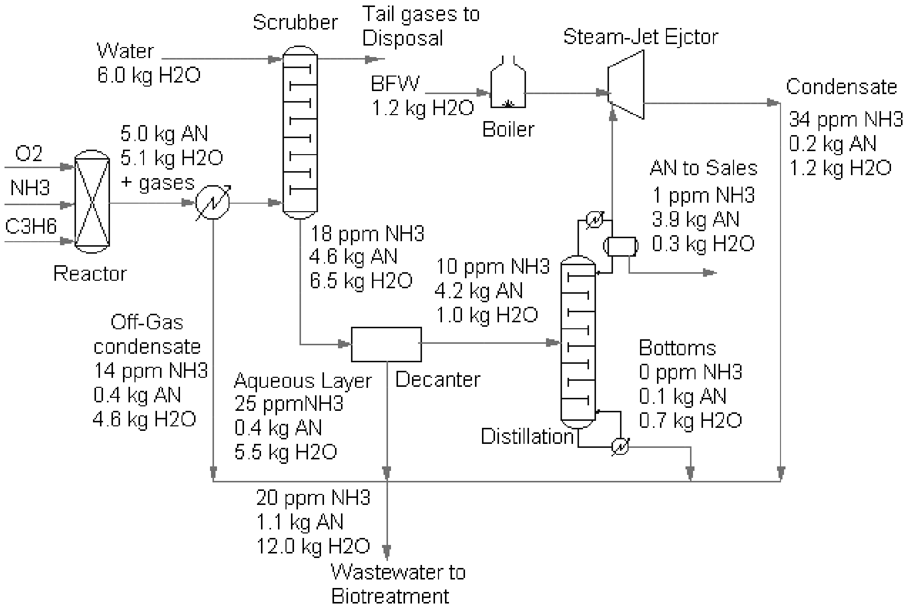

Figure 5. The flowsheet for AN production

There are two water demands for this process - the boiler feed water (BFW) and the water feed stream to the scrubber. There are four water sources that include the off-gas condensate, the aqueous layer from the decanter, the distillation column bottoms product and the condensate from the steam-jet ejector. Ammonia (NH_3) is the main contaminant in this process. Here, the water sources are regarded as wastewater and sent to a bio-treatment facility operated at full capacity.

One way to debottleneck the overall process is via water reuse and recycling. However, any proposed solution must comply with the flowrate and concentration constraints imposed on the water demands and sources, as listed below:

i. Scrubber

- $5.8 \leq$ flowrate of wash feed (kg/s) ≤ 6.2
- $0.0 \leq NH_3$ content of wash feed ≤ 10.0

ii. Boiler feed water

- NH_3 content = 0.0 ppm
- AN content = 0.0 ppm

iii. Decanter

- 10.6 ≤ feed flowrate (kg/s) ≤ 11.1

iv. Distillation column

- 5.2 ≤ feed flowrate (kg/s) ≤ 5.7
- 0.0 ≤ NH_3 content of feed (ppm) ≤ 30.0
- 80.0 ≤ AN content of feed (wt%) ≤ 100.0

The first step in establishing the minimum water target is to identify the limiting water data for the process, subject to the constraints listed above. Note that, of the four listed constraints, only the first two (i.e. scrubber and boiler feed water) that involve the streams selected for water reuse analysis, are considered. The first constraint requires the flowrate and the concentration of NH_3 in the scrubber wash feed to be bounded within the given range. Hence, in order to maximise water reuse, one should maximise the NH_3 concentration while keeping the flowrate of this water source to a minimum. The second constraint means that only pure water should be used as the boiler feed water (BFW). The limiting data for the water demands and sources are summarised in **Table 1**. Note from **Figure 5** and **Table 1** that none of these operations can be modelled as a mass transfer process.

Table 1. Limiting water data for AN production

Water demands, D_j		**Flowrate**	**Concentration**
j	Stream	F_j (kg/s)	C_j (ppm)
1	BFW	1.2	0
2	Scrubber	5.8	10

Water sources, S_i		**Flowrate**	**Concentration**
i	Stream	F_i (kg/s)	C_i (ppm)
1	Distillation bottoms	0.8	0
2	Off-gas condensate	5	14
3	Aqueous layer	5.9	25
4	Ejector condensate	1.4	34

El-Halwagi (1997) proposed a targeting technique for the limiting data in **Table 1**. However, his simplified technique only considers the water flowrate balance and ignores the driving force for water reuse. **Figure 6** is a conceptual representation of the targeting technique proposed by El-Halwagi (1997). Clearly, without considering the thermodynamic constraint (the concentration driving force), one could easily overlook the true minimum target. Based on this simplified technique, El-Halwagi (1997) reported that no fresh water was needed for this operation while the wastewater flowrate was targeted at 4.8 kg/s. As will be shown later in this paper, the target predicted by El-Halwagi (1997) was only valid after implementing a process change, i.e. by substituting the steam-jet ejector with a vacuum pump. Note that there was no mention of the minimum water target for the base case process, i.e. before regeneration and process changes.

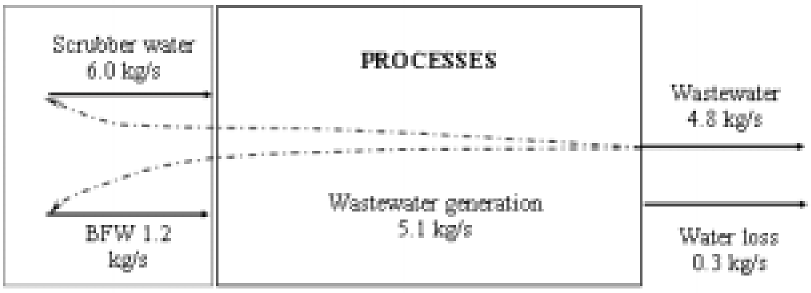

Figure 6 Targeting model by El-Halwagi (1997)

In the same work, El-Halwagi (1997) also proposed a systematic method to design the water reuse network for the process by means of the source-sink mapping diagram. However, without the knowledge of the *true minimum* target ahead of design, there will always be questions as to whether further improvements on the network are still possible Clearly, it is essential to have a good targeting tool to determine the true minimum target. As a guideline, a good tool should satisfy three basic requirements as follows:

i. it should be capable of handling mass transfer or non mass transfer-based water operations.
ii. it should consider the flowrate and the concentration driving force for water reuse.
iii. it should be non-iterative, therefore, can quickly yield the exact targets.

In the next section, we demonstrate the use of WCA as a new tool for water targeting that fulfils all the basic requirements outlined.

THE WATER CASCADE ANALYSIS TECHNIQUE

The main objective of the Water Cascade Analysis (WCA) is to establish the minimum water targets, i.e. the overall fresh water requirement and wastewater generation for a process after looking at the possibility of using the available water sources within a process to meet its water demands. To achieve this objective, one has to establish the net water flowrate as well as the water surplus and deficit at the different water purity levels within the process under study. The *interval water balance table* has been introduced for this purpose. The AN production case study described in the previous section is used to illustrate the WCA water targeting technique is presented next.

The first step in the WCA is to set up the *interval water balance table* (**Table 2**) to determine the net water source or water demand at each purity level. The first column of **Table 2** contains the contaminant concentration levels (C) arranged in ascending order. Each concentration level is expressed in terms of the water purity (P) in the second column. With the concentration of pure water set at one million ppm, the fraction of pure water in a contaminated stream, or the *water purity*, can be expressed as (Hallale, 2002):

$$\text{Purity, } P = \frac{1000000 - C}{1000000} \tag{2}$$

where:

C = contaminant concentration in ppm.

Table 2. The interval water balance table for AN production case study

Column no.	1	2	3	4	5	6	7
Interval, n	Concentration C_n (ppm)	Purity, P_n	ΔP	$\Sigma F_{D,j}$ (kg/s)	$\Sigma F_{S,i}$ (kg/s)	$\Sigma F_{D,j} + \Sigma F_{S,i}$ (kg/s)	Net water source / demand
1	0	1.000000		-1.2	0.8	-0.4	Demand
			0.000010				
2	10	0.999990		-5.8		-5.8	Demand
			0.000002				
3	14	0.999986			5.0	5.0	Source
			0.000013				
4	25	0.999975			5.9	5.9	Source
			0.000009				
5	34	0.999966			1.4	1.4	Source
			0.999966				
6	1000000	0					

The number of purity intervals (n) equals the number of water demands (N_D) and the number of water sources (N_S) minus any duplicate purity (N_{DP}):

$$n = N_D + N_S - N_{DP} \tag{3}$$

Column 3 of **Table 2** contains the water purity difference (ΔP), calculated as follows:

$$\Delta P = P_n - P_{n+1} \tag{4}$$

Columns 4 and 5 contain the flowrates for the water demands ($\sum_j F_{\mathrm{D},j}$) and water sources ($\sum_i F_{\mathrm{S},i}$) at their corresponding purity levels. The flowrate of water demand is fixed as negative, and the water source positive. These flowrates are summed up at each purity level to give the *net interval* water flowrate, ($\sum_j F_{\mathrm{D},j} + \sum_i F_{\mathrm{S},i}$, column 6); (+) representing *net water source*, (-) *net water demand* (column 7).

The next key step in the WCA is to establish the fresh water and waste water targets for the process. In doing so, it is important to consider *both* the water flowrate balance and the concentration driving force (water purity) so that the *true minimum* water targets can be obtained. The water flowrate balance involves using the water cascade concept to get the *cumulative* net water source/demand for a process (F_{C}). A conceptual illustration of how water cascading can minimise fresh water needs and wastewater generation is represented by **Figure 7**. By making use of 100 kg/s of the net water source at the purity level of 0.999900 (100 ppm) to satisfy the water demand of 50 kg/s at the purity level of 0.999800 (200 ppm), it is possible to avoid sending part of the net water source directly to effluent. Doing so not only reduces the wastewater generation but also the fresh water consumption, in both cases, by 50%.

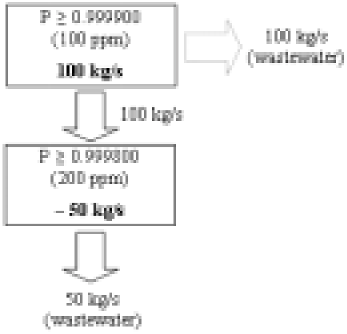

Figure 7. The principle of water cascading

For the water cascade diagram in **Figure 8(a)**, a fresh water flowrate (F_{FW}) of 0 kg/s is assumed. Here, the net water demand of 0.4 kg/s at the first purity level is cascaded to the second purity level to meet another water demand of 5.8 kg/s, giving a cumulative net of –6.20 kg/s (demand). This cumulative demand meets only net water sources down the next three purity levels to yield a cumulative water source, or wastewater flowrate (F_{WW}), of 6.10 kg/s at the lowest purity level of the water cascade diagram.

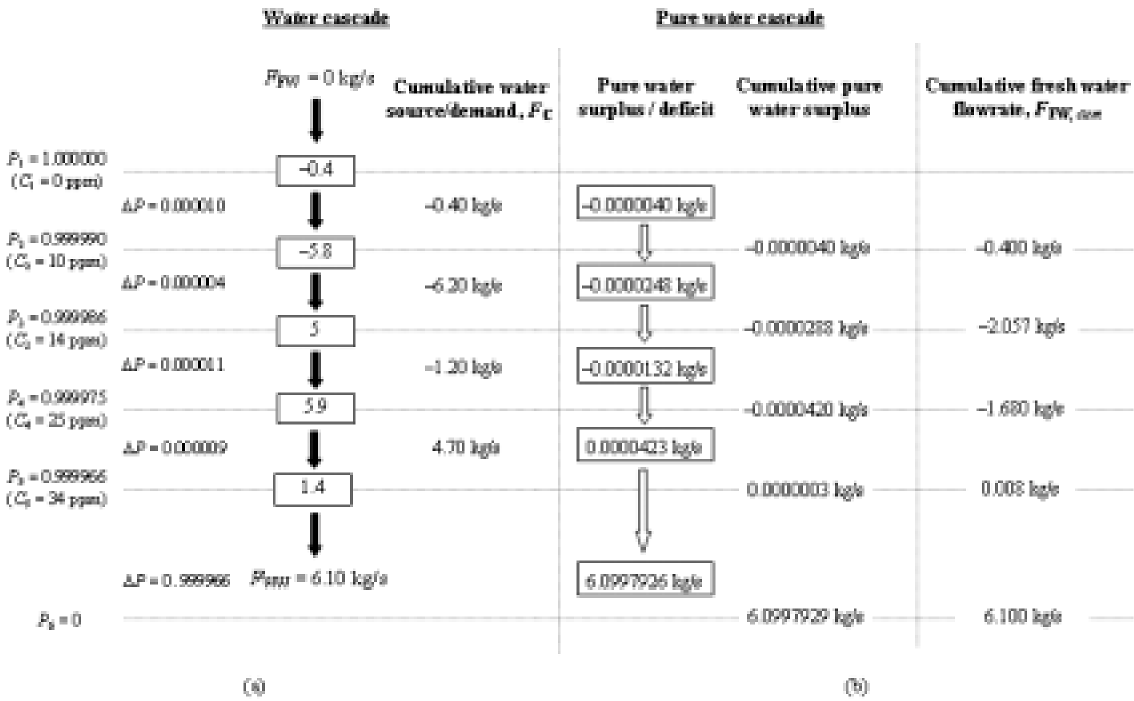

Figure 8. (a) Water cascade diagram with an assumed fresh water flowrate of 0 kg/s; (b)Pure water cascade is used to check the feasibility of the water cascade

The *cumulative* net water source/demand for the process (F_C) at each purity interval forms the net interval water cascade diagram. The water cascade diagram is similar to the *interval heat balance table* for the problem table algorithm in heat integration (Linnhoff *et al.*, 1982) and the table of exchangeable loads for mass exchange cascade diagram in mass integration (El-Halwagi, 1997).

The water cascade diagram depicting the preliminary water balance (i.e., with $F_{FW} = 0$ kg/s) is essential as a basis to generate a feasible water cascade, and ultimately, the true minimum water targets. Note again that, in addition to considering the water flowrate balance, the true minimum targets can only be realised by also taking into account the pure water surplus or deficit, which is a product of the cumulative net water source/demand (F_C) and the purity difference (ΔP) across two purity levels (**Figure 8b**). A pure water surplus (+) means that water is available with purity higher than what is required in this region. On the other hand, a pure water deficit (-) means that water of higher purity than those available is required (Hallale, 2002). Cascading the pure water surplus/deficit down the purity intervals yields the pure water cascade that represents the cumulative amount of pure water surplus/deficit (**Figure 8b).** The cumulative pure water surplus/deficit at each purity level is a numerical representation of the water surplus diagram introduced by Hallale (2002) (**Figure 3**).

Notice that the first three purity levels (i.e. P_1, P_2 and P_3) of the pure water cascade in **Figure 8(b)** consist of cumulative pure water deficits. The deficits on the pure water cascade, which correspond to the negative region of Hallale's water surplus diagram (**Figure 9**) (Hallale, 2002), indicate that the pure water cascade is "infeasible". These deficits mean that there is insufficient fresh water in the network and are the result of assuming zero fresh water flowrate (F_{FW}) during water cascading. Thus, additional fresh water should be supplied to remove all pure water deficits and yield a feasible pure water cascade.

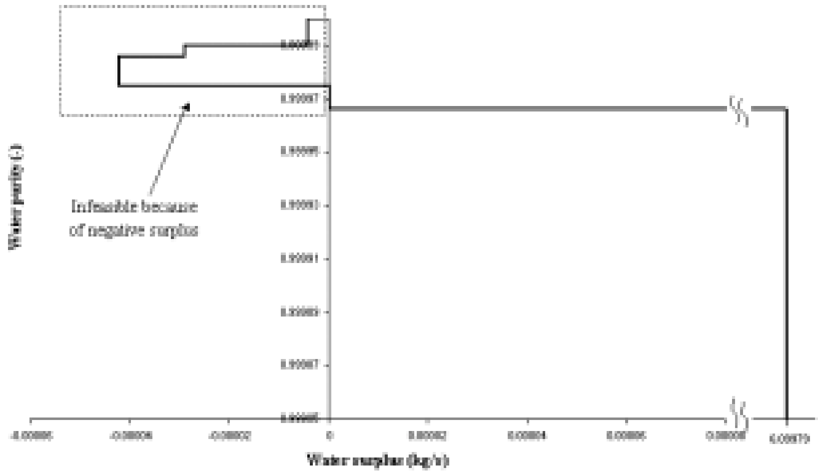

Figure 9 A negative water surplus in the water surplus diagram indicate an infeasible water network

Figure 8(b) shows that the *cumulative fresh water flowrate* ($F_{\mathrm{FW},cum}$) for each purity level is obtained by dividing the cumulative pure water surplus/deficit by the purity difference between the fresh water utility (P_{F}) and purity level of interest, as follows,

$$F_{\mathrm{FW},\,cum} = \frac{\text{cumulative pure water surplus/deficit}}{P_F - P_n} \quad (5)$$

A negative $F_{\mathrm{FW},cum}$ means that there is insufficient fresh water whereas a positive $F_{\mathrm{FW},cum}$ means that there is excess fresh water at the given purity level. In order to ensure that there is sufficient fresh water at all points in the network, a fresh water flowrate (F_{FW}) of exactly the same magnitude as the absolute value of the largest negative $F_{\mathrm{FW},cum}$ should be supplied at the highest purity level of a *feasible water* *cascade* (**Figure 10**). $F_{FW,cum}$ of –2.057 kg/s found at the third purity level (P_3) of the cumulative fresh water cascade in **Figure 8(b)** is the largest negative $F_{FW,cum}$. This quantity of fresh water is added at the highest purity level of the feasible water cascade in **Figure 10**. Note that a feasible water cascade is the one that results in positive, or at least, zero cumulative pure water surplus value in the pure water cascade. The feasible water cascade yields the *true minimum* fresh water and wastewater flowrate targets of 2.057 kg/s and 8.157 kg/s respectively for the AN case study.

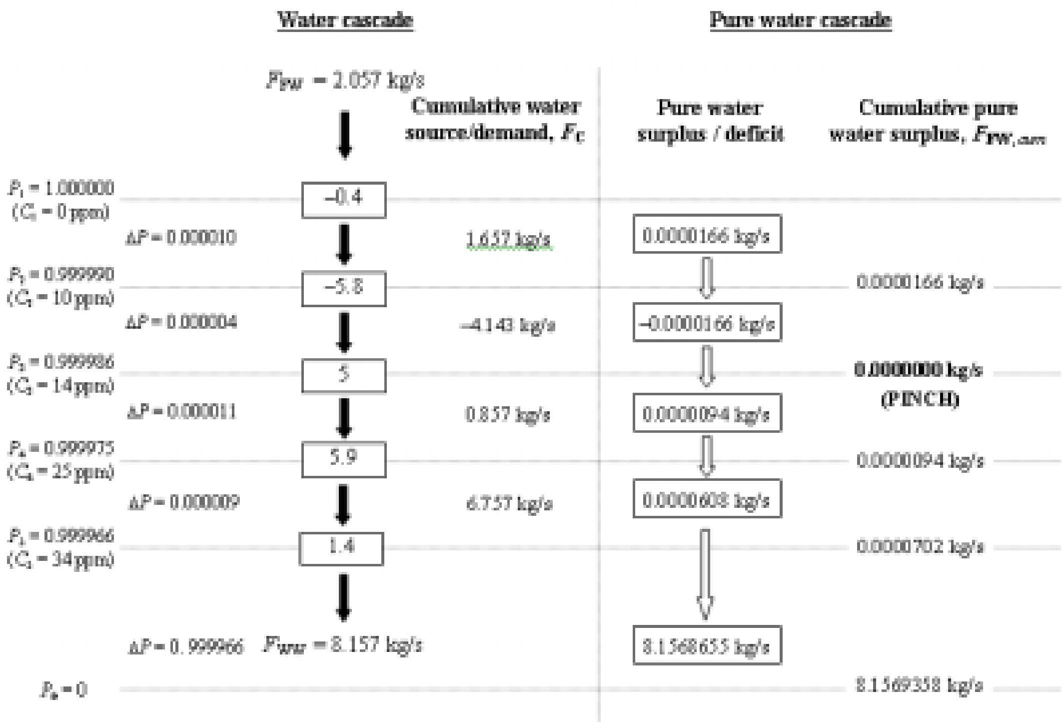

Figure 10. A feasible water cascade for the AN case study

At the third purity level (P = 0.999986) where there is zero cumulative pure water surplus, there exists the pinch for the AN problem. The pinch is the most constrained part of the network that results in maximum water recovery. The detailed network design proposed by El-Halwagi (1997) confirmed the utility targets for this case study. Note that through the WCA, we have obtained the utility targets ahead of design and are able to verify whether the proposed initial design by El-Halwagi (1997) have achieved the objective of maximum water recovery (MWR) for the plant. The water cascade and the pure water surplus cascade diagrams can be integrated with the interval water balance table to form the *water cascade table* (WCT) (**Table 3**).

Table 3 Water Cascade Table (WCT) for AN production case study

Interval n	Concentration C_n (ppm)	Purity, P_n	$\Sigma F_{D,j}$ (kg/s)	$\Sigma F_{S,i}$ (kg/s)	$\Sigma F_{D,j} + \Sigma F_{S,i}$ (kg/s)	F_C, (kg/s)	Pure water surplus (kg/s)	Cumulative pure water surplus (kg/s)
						2.057		
1	0	1.000000	-1.2	0.8	-0.4			
						1.657	0.0000166	
2	10	0.999990	-5.8		-5.8			0.0000166
						-4.143	-0.0000166	
3	14	0.999986		5.0	5.0			0
						0.857	0.0000094	
4	25	0.999975		5.9	5.9			0.0000094
						6.757	0.0000608	
5	34	0.999966		1.4	1.4			0.0000702
						8.157	8.1568655	
6	1000000	0						8.1569358

CASCADE ANALYSIS FOR HYDROGEN DISTRIBUTION NETWORK

The problem definition of a hydrogen distribution network synthesis is given as follows:

Given a set of hydrogen-using processes, it is desired to determine a network of interconnections of hydrogen streams among the hydrogen-using processes so that the overall fresh hydrogen consumption is minimised while the processes receive hydrogen of adequate quality.

Hydrogen is one of the important and expensive utilities in some of the chemical process industries. This includes the crude oil refineries and their associated downstream petrochemical plants. In the crude oil refinery, hydrotreating and hydrocracking are among the main processing steps where large amount of hydrogen is consumed. Other hydrogen-consuming processes in a crude oil refinery include the lubricant plants and isomerisation.

Towler *et al.* [43] initiated the overall analysis of hydrogen distribution network by assessing the cost of hydrogen recovery. A new tool called value composite curve is developed to provide insight into economic tradeoffs that affect the hydrogen management problem in the network. However, as pointed out by Alves and Towler (2002), this approach does not account for the physical constraints that influence the design of the hydrogen network. Alves and Towler (2002) in turn proposed the concept of hydrogen surplus to locate the minimum utility (hydrogen consumption) target in a new hydrogen distribution network. This tool is by far the most promising tool in locating the right minimum utility target for a grassroot process, prior to any commitment of final network design.

The remaining part of the paper aims to present a new tabular technique called the *Hydrogen Cascade Analysis* (HCA), a supplement tool for hydrogen surplus diagram proposed by Alves and Towler (2002). As water surplus diagram, though the concept of hydrogen surplus is powerful in locating the utility target ahead of design, it is tedious and time consuming as it involves trial an error to find the pinch point(s) and utility target via graphical drawings. A minimum of two sets of graphical drawings is required, i.e. the hydrogen source and sink composite curves as well as hydrogen surplus diagram. Often, more than few times of iterative drawing effort are required to finally determine the right target (note that two different graphs are required for each time!).

The tedious iterative procedure to construct the hydrogen surplus diagram is similar to that in Figure 4.

Hydrogen distribution network case study

Figure 11 shows a hydrogen distribution case study from Alves and Towler (2002). Hydrogen source and sink data for this case study is shown in **Table 4**. There are two hydrogen-producing facilities in this network, i.e. catalytic reforming unit (CRU) and steam reforming unit (SRU). These are the internal hydrogen source for the network and their use are to be maximised before any external hydrogen utility is used. Besides, an import of external hydrogen utility is available at a purity of 95%, with a maximum capacity of 346.5 mol/s. However, all the hydrogen sources have their maximum production capacity in producing hydrogen. The data for various hydrogen sources are also given in **Table 4**. External hydrogen utility consumption reported for this case study prior to the systematic analysis is reported as 277.2 mol/s (Figure 11).

Table 4 Hydrogen source and sink data for case study

Hydrogen sink, D_j		**Flowrate**	**Concentration**
j	Processes	F_{SK} (mol/s)	y_{SK} (mol% H_2)
1	HCU	2495.0	80.61
2	NHT	180.2	78.85
3	CNHT	720.7	75.14
4	DHT	554.4	77.57

Hydrogen sources, S_i		**Flowrate**	**Concentration**
i	Processes	F_{SR} (mol/s)	y_{SR} (mol% H_2)
1	HCU	1801.9	75.00
2	NHT	138.6	75.00
3	CNHT	457.4	70.00
4	DHT	346.5	73.00
5	SRU	623.8 (max)	93.00
6	CRU	415.8 (max)	80.00
7	External source	346.5 (max)	95.00

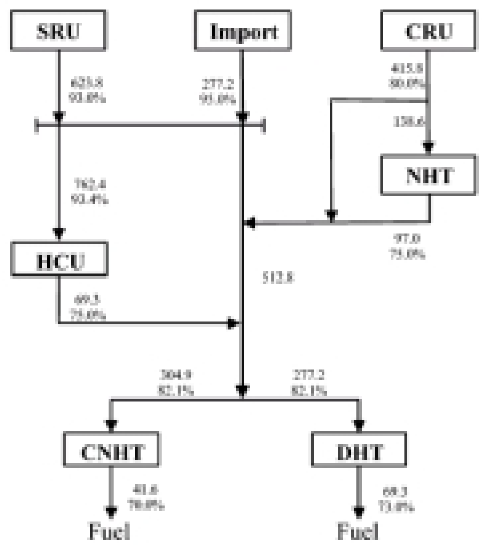

Figure 11 Refinery hydrogen distribution case study in Example 1 [44]. The numbers represent the total gas flowrate in mol/s and hydrogen purity (mol% H_2)

THE HYDROGEN CASCADE ANALYSIS TECHNIQUE

Procedure to carry out the hydrogen cascade analysis (HCA) for a refinery hydrogen network are almost the same with the WCA in water network analysis, as described in the previous section of this paper. One is to construct the *interval hydrogen balance table* as shown in **Table 5**. This table is equivalent to the *interval water balance table* shown in **Table 2**. The only difference between these interval balance tables is that, in hydrogen network analysis, purity (P_n) of the hydrogen stream is expressed directly from the stream data, while water purity is expressed indirectly by the contaminant concentration, via Eq. 2. Hence, an interval hydrogen balance table will have a column less than that in the interval water balance table (**Table 5**).

Table 5 Interval hydrogen balance table for hydrogen network analysis

Column no.	1	2	3	4	5	6
Interval, n	Purity, P_H (mol%)	ΔP	$\Sigma F_{SK,j}$ (mol/s)	$\Sigma F_{SR,i}$ (mol/s)	$\Sigma F_{SK,j} + \Sigma F_{SR,i}$ (mol/s)	Net hydrogen / demand
1	95.00					
		2.00				
2	93.00			623.8	623.8	Source
		12.39				
3	80.61		-2495.0		-2495.0	Demand
		0.61				
4	80.00			415.8	415.8	Source
		1.15				
5	78.85		-180.2		-180.2	Demand
		1.28				
6	77.57		-554.4		-554.4	Demand
		2.43				
7	75.14		-720.7		-720.7	Demand
		0.14				
8	75.00			1940.5	1940.5	Source
		2.00				
9	73.00			346.5	346.5	Source
		3.00				
10	70.00			457.4	457.4	Source
		70.00				
11	0.00					

The rest of the procedure in constructing the HCA is the same with WCA, with the interchange between the variables of water demands and water sources flowrates into the hydrogen sinks and hydrogen sources flowrates.

Following the similar procedure in WCA, one will yield a completed *hydrogen cascade table* (HCT) as shown in **Table 6**. HCA yields the minimum external hydrogen utility target, F_H (equivalent to the fresh water flowrate in WCA) of 268.821 mol/s and a fuel flowrate, F_F (equivalent to the wastewater flowrate in WCA) of 102.521 mol/s for the case study. These values are the same with that originally reported by Alves and Towler (2002) via hydrogen surplus diagram, and hence, proving that HCA is a numerical way of expressing the hydrogen surplus diagram with less tedious work.

Table 6 The hydrogen cascade table (HCT) for case study

Interval, n	Purity, P_n (mol%)	$\Sigma F_{SK,j}$ (mol/s)	$\Sigma F_{SR,i}$ (mol/s)	$\Sigma F_{SK,j} + \Sigma F_{SR,i}$ (mol/s)	F_C, (mol/s)	Pure hydrogen surplus (mol/s)	Cumulative pure hydrogen surplus (mol/s)
					268.821		
1	95.00						
					268.821	5.3764	
2	93.00		623.8	623.8			5.3764
					892.621	110.5957	
3	80.61	-2495.0		-2495.0			115.9722
					-1602.379	-9.7745	
4	80.00		415.8	415.8			106.1977
					-1186.579	-13.6457	
5	78.85	-180.2		-180.2			92.5520
					-1366.779	-17.4948	
6	77.57	-554.4		-554.4			75.0572
					-1921.179	-46.6846	
7	75.14	-720.7		-720.7			28.3726
					-2641.879	-3.6986	
8	75.00		1940.5	1940.5			24.6739
					-701.379	-14.0276	
9	73.00		346.5	346.5			10.6464
					-354.879	-10.6464	
10	70.00		457.4	457.4			**0.0000**
					102.521	71.7647	
11	0.00						71.7647

CONCLUSION

Cascade analysis is a numerical alternative to the graphical targeting technique known as the surplus diagram. Cascade analysis can quickly yield accurate utility targets (water and hydrogen consumption) and pinch point location(s) for a (water or hydrogen) network in a simple way and a precise result. In water pinch analysis, this numerical tool is known as *water cascade analysis* (WCA) while in hydrogen pinch analysis, it is known as *hydrogen cascade analysis* (HCA). All the key features and the systematic nature of the cascade analysis make it easy for the technique to be automated and translated into any computer language for software development. The WCA feature has been recently incorporated in *Heat-MATRIX*, a new software for energy and water reduction developed by the Process Systems Engineering Group, Department of Chemical Engineering, Universiti Teknologi Malaysia (Manan *et al.*, 2003).

NOTATION

C	=	contaminant concentration, ppm
F	=	flowrate of water streams (kg/s or t/hr) or hydrogen streams (mol/s)
n	=	number of purity intervals
N	=	number of water / hydrogen demands or sources
P	=	purity
y	=	hydrogen purity, mol %
Δ	=	difference
$\sum$	=	summation

Subscripts

C	=	cumulative
D	=	water demands
DP	=	duplicate purities
est	=	estimated fresh water
F	=	hydrogen for fuel
FW	=	fresh water feed
H	=	fresh hydrogen feed
i	=	sources
j	=	demands
S	=	water sources
SK	=	hydrogen sink
SR	=	hydrogen source
WW	=	wastewater

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REFERENCES

Alves, J. J. and Towler, G. P. (2002). "Analysis of Refinery Hydrogen Distribution Systems", Ind. Eng. Chem. Res., 41, 5759-5769.

Bagajewicz, M., 2000, A Review of Recent Design Procedures for Water Networks in Refineries and Process Plants. *Computers Chem. Engng.*, 24, 2093–2113.

Dhole, V. R., Ramchandani, N., Tainsh, R. A. and Wasilewski, M. (1996). "Make your process water pay for itself". Chemical Engineering 103, 100–103.

Dunn, R. F., Srinivas, B. K. and El-Halwagi, M. M. (1995). Optimal Design of Heat-induce Separation Networks for VOC Recovery. *AIChE Symposium Series* 90 (303), 74-85.

Dunn, R. and Wenzel, H. (2001a). Process Integration Design Method for Water Conservation and Wastewater Reduction in Industry. Part 1: Design for Single Contaminants. *Clean Prod. Processes* 3, 307 – 318.

Dunn, R. and Wenzel, H. (2001b). Process Integration Design Method for Water Conservation and Wastewater Reduction in Industry. Part 2: Design for Multiple Contaminants. *Clean Prod. Processes* 3, 319 – 329.

Dye, S. R., Berry, D. A. and Ng, K. M. (1995). Synthesis of Crystallisation-based Separation Scheme. *AIChE Symposium Series*, 91(304), 238-241.

El-Halwagi, M. M. and Manousiouthakis, V. (1989). Synthesis of Mass Exchange Networks. *AIChE Journal* 35(8), 1233 – 1244.

El-Halwagi, M. M. and Manousiouthakis, V. (1990a). Automatic Synthesis of Mass Exchange Networks with Single Component Targets. *Chem. Eng. Sci.* 45 (9), 2813 – 2831.

El-Halwagi, M. M. and Manousiouthakis, V. (1990b). Simultaneous Synthesis of Mass Exchange and Regeneration Networks. *AIChE Journal* 36(8), 1209 – 1219.

El-Halwagi, M. M. and Srinivas, B. K. (1992) Synthesis of Reactive Mass Exchange Networks. *Chem. Eng. Sci.* 47 (8), 2113 – 2119.

El-Halwagi, M. M., Srinivas, B. K. and Dunn, R. F., (1995). Synthesis of Optimal Heat-induce Separation Networks. *Chem. Eng. Sci.* 50 (1), 81–97.

El-Halwagi, M. M., Hamad, A. A. and Garrison, G. W. (1996). Synthesis of Waste Interception and Allocation Networks. *AIChE Journal*, 42 (11), 3087-3101.

El-Halwagi, M. M. (1997). "*Pollution prevention through process integration: systematic design tools*". Academic Press, San Diego.

Foo, C. Y., Manan, Z. A., Yunus, R. M. and Aziz, R. A., (2003). Maximising Water Recovery through Water Pinch Technology – The Use of Water Cascade Table, *Environment 2003*, Malaysia.

Hallale, N. (2002). "A new graphical targeting method for water minimisation". Adv. Env. Res., 6 (3), 377–390.

Hallale, N., and Liu, F. (2001). "Refinery Hydrogen Management for Clean Fuels Production", Adv. Env. Res., 6, 81-98.

Hallale, N., Moore, I. and Vauk, D. (2002). "Hydrogen: Liability or Asset?" Chem. Eng. Prog., September, 66-75.

Linnhoff, B., Townsend, D. W., Boland, D., Hewitt, G. F., Thomas, B. E. A., Guy, A. R. and Marshall, R. H. (1982). "*A user guide on process integration for the efficient use of energy*". IChemE, Rugby, UK.

Manan, Z. A., Ooi, B. L., Lim, F. Y. and Foo, C. Y. (2003). "Heat-MATRIX - A Computer Software For The Reduction Of Energy And Water In Process Plants", 31st Int. Exhibition of Invention, New Techniques and Products of Geneva, Switzerland.

Mann, J. G. and Liu, Y. A. (1999). "*Industrial water reuse and wastewater minimisation*". New York: McGraw Hill.

Olesen, S. G. and Polley, G. T. (1997). A Simple Methodology For The Design Of Water Networks Handling Single Contaminants. *Trans. IChemE*, Part A, 75, 420–426.

Polley, G. T. and Polley, H. L. (2000). "Design better water networks". Chem. Eng. Prog., 96 (2): 47–52.

Richburg, A. and El-Halwagi, M. M. (1995). A Graphical Approach to the Optimum Design of Heat-induced Separation Networks for VOC Recovery. *AIChE Symposium Series*, 91(304), 256-259.

Sorin, M. and Bédard, S. (1999). "The global pinch point in water reuse networks". Trans. IChemE, Part B, 77, 305–308.

Srinivas, B. K. and El-Halwagi, M. M. (1994a). Synthesis of Reactive Mass Exchange Networks with General Non-linear Equilibrium Functions. *AIChE Journal* 40 (3), 463-472.

Srinivas, B. K. and El-Halwagi, M. M. (1994b). Synthesis of Combined Heat and Reactive Mass Exchange Networks. *Chem. Eng. Sci.* 49 (13), 2059 – 2074.

Takama, N., Kuriyama, T., Shiroko, K. and Umeda, T. (1980) "Optimal water allocation in a petroleum refinery", Computers Chem. Eng., 4, 251–258.

Tan, Y. L., Manan, Z. A. and Foo, C. Y. (2002). Water Minimisation by Pinch Technology – Water Cascade Table for Minimum Water and Wastewater Targeting". *9th Asian Pacific Confederation of Chemical Engineering (APCChE 2002) Congress*, New Zealand.

Towler, G. P., Mann, R., Serriere, A. J-L., Gabaude, C. M. D. (1996) "Refinery Hydrogen Management: Cost Analysis of Chemically Integrated Facilities', Ind. Eng. Chem. Res., 35(7), 2378-2388.

Wang, Y. P. and Smith, R. (1994). "Wastewater minimisation". Chem. Eng. Sci., 49, 981–1006.

Xiao, F. and Seider, W. D. (2001). New Structure and Design Methodology for Water Networks. *Ind. Eng. Chem. Res.*, 40, 6140–6146.