

Setting the Minimum Utility Gas Flowrate Targets Using Cascade Analysis Technique

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This work describes the gas cascade analysis (GCA) as an extension to cascade analysis technique for targeting the minimum flowrates for water and property-based networks. The GCA technique enables quick and accurate identification of the minimum flowrate targets, pinch-point location(s), and resource allocation targets for a utility gas network. Multiple-pinch problems and appropriate selection of gas purification techniques were systematically assessed via the GCA. The GCA technique was successfully used to determine the minimum flowrate targets for nitrogen, oxygen, and hydrogen utility gas networks prior to detailed design.

Introduction

Process integration emerged as an effective energy-saving tool during the world energy crisis of the 70s.^{1–6} Since then, it has evolved into a generic methodology for resource conservation and pollution prevention, covering the optimal synthesis of mass-exchange network,^{7–16} maximum water-recovery network,^{17–31} hydrogen networks,^{32–41} and property-based network.^{42–49}

The typical process integration technique for resource conservation comprises two steps, i.e., setting the minimum resource targets followed by design of a resource-conservation network to achieve the minimum resource targets. Generic targeting techniques applicable for various types of resource-conservation networks include graphical composite curves^{7,17,22,23,26,34,47} and numerical targeting techniques.^{8–10,24–28,30,49} Once the resource targets are established, the resource-conservation network can be designed to achieve the minimum targets using various network design tools.^{7,10,17,21,22,26,31} Recent numerical targeting methods such as cascade analysis technique have, thus far, been proven applicable to water and property-based networks.^{24–25,27–29,49} This paper describes the gas cascade analysis (GCA) technique as a numerical tool to establish the minimum flowrate targets for a utility gas network. GCA is an extension of the cascade analysis technique for targeting water and property-based networks. The problem definition for utility gas network synthesis is given as follows:

Given a set of utility gas-using processes, it is desired to synthesize a network of stream interconnections among the processes so that the overall consumption of fresh utility gas is minimized.

Despite many similarities, there exist two key differences between synthesis problems of water and property-based networks with that of a utility gas network. First, while pure freshwater feed (zero contaminant load) has always been assumed to be available for synthesis of water network, it is quite common to encounter impure fresh feeds for utility gas networks. Also, there are cases where the fresh feeds may

contain higher levels of impurities compared to the effluent gas streams or gas sources. This rarely happens in a water network.

Second, a water network may consist of a mix of mass-transfer-based and nonmass-transfer-based processes. For mass-transfer operations, water is used to remove impurities from a set of rich process streams. Note that any flowrate that can remove impurities from a rich process stream is acceptable. Hence, the flowrate for water feeds (i.e., water demands) may not be fixed. However, this is not the case for utility gas networks. To maintain steady-state operation (e.g., to maintain the system pressure in a gas-phase hydrotreating reactor), it is necessary to maintain a uniform inlet mass flowrate and gas concentration for a gas-consuming process. This means that each gas sink requires a fixed amount of gas flowrate and inlet concentration.

This paper describes the GCA as a tool to set the minimum flowrate targets for utility gas networks. The technique proposed in this work will address the two previously mentioned key limitations associated with the current state-of-the-art resource targeting techniques.^{24–25,27–29,49} Four industrial case studies involving the reuse/recycle of nitrogen, oxygen, and hydrogen gases will demonstrate the extension of cascade analysis technique to the setting up of the minimum utility targets for a gas network.

Example 1—Nitrogen Integration

Figure 1 shows the process flow diagram of a magnetic-tape manufacturing process.¹⁰ Coating ingredients are dissolved in an organic solvent and form a slurry mixture. The slurry is then suspended with resin binders and special additives. Next, the coating slurry is deposited on a base film. Nitrogen gas is used to induce evaporation rate of solvent that is proper for deposition. A small amount of the solvent is decomposed into other organic species in the coating chamber. The decomposed organics are then separated from the coating chamber exhaust gas using a membrane unit. The retentate stream consisting mainly of nitrogen and laden with 1.9 wt/wt % organic solvent leaves the membrane unit at a flowrate of 3.0 kg/s.

The coated film undergoes a drying process where nitrogen gas is used to evaporate the remaining solvent. The dryer exhaust gas has a flowrate of 5.5 kg/s and a solvent concentration of

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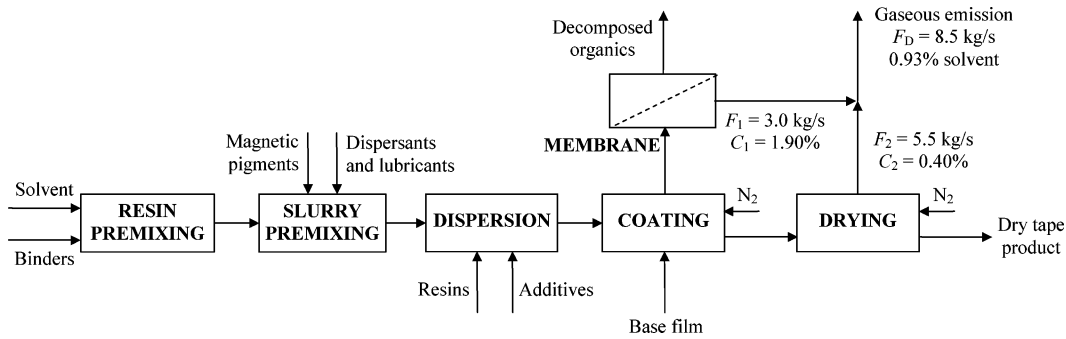


Figure 1. Process flow diagram of a magnetic tape manufacturing process in Example 1.¹⁰

Table 1. Limiting Data for Example 1 (Nitrogen Integration)¹⁰

gas sinks, SK _j		flowrate	concentration
<i>j</i>	stream	<i>F_j</i> (kg/s)	<i>C_j</i> (wt/wt %)
1	drying	5.5	0.1
2	coating	3.0	0.2

gas sources, SR _i		flowrate	concentration
<i>i</i>	stream	<i>F_i</i> (kg/s)	<i>C_i</i> (wt/wt %)
1	membrane retentate	3.0	1.9
2	drying	5.5	0.4
fresh supply		to be determined	0

0.4 wt/wt %. The two exhaust gas streams are mixed and disposed of. To reduce nitrogen consumption, it is proposed to reuse/recycle the solvent-laden exhaust gas to the coating and drying processes, subject to the following constraints:

i. Coating

- $3.0 \leq$ flowrate of gaseous feed (kg/s) ≤ 3.2
- $0.0 \leq$ wt % of solvent ≤ 0.2

ii. Drying

- $5.5 \leq$ flowrate of gaseous feed (kg/s) ≤ 6.0
- $0.0 \leq$ wt % of solvent ≤ 0.1

The outlet concentrations for the coating and drying processes are assumed to be independent of the feed gas concentration. The constraints listed above require the gaseous flowrate and its solvent concentration in the units to fall within the given range. El-Halwagi¹⁰ and Foo et al.²⁹ specify two key strategies to maximize reuse/recycle. First, the maximum permissible inlet concentration should be assigned as the *limiting concentration*, while the minimum feed flowrate should be assigned as the *limiting* flowrate for each process sink. Second, the sources should be segregated into individual streams, since mixing tends to degrade the source streams' qualities. For this example, the solvent-laden exhaust gas should be segregated into membrane retentate and dryer exhaust gas streams. Fresh makeup nitrogen

is solvent-free and, hence, has a limiting concentration of 0 wt %. The limiting data for the gas sinks and sources are listed in Table 1.

Gas Cascade Analysis (GCA) Technique

The first step in conducting a GCA is to locate the various gas sinks and sources at their respective concentration levels. As shown in Table 2, the concentration levels (C_k) are arranged in an ascending order and the flowrate of each gas sink (F_j) and source (F_i) is located individually at its respective concentration level k in columns 3 and 4. Column 5 represents the net flowrate ($\sum_i F_i - \sum_j F_j$) between gas sources and sinks at each concentration level k ; positive values indicate a surplus, negative values indicate a deficit.

The next GCA step is to cascade the net flowrate surplus/deficit down the concentration levels to yield the cumulative surplus/deficit flowrate (F_C) shown in column 6 of Table 2 and, ultimately, the overall net surplus/deficit for the network. A zero fresh nitrogen flowrate is assumed at this stage to facilitate the search for the minimum utility gas flowrate. The cascade principle is conceptually explained in Figure 2. In Figure 2a, 50 kg/s of waste gas is available as a gas source at an impurity concentration of 5%. On the other hand, a gas sink exists at 100 kg/s and can accept up to an impurity concentration of 10%. Without considering reuse, 50 kg/s of waste gas would have to be purged, while 100 kg/s of fresh gas would have to be supplied to satisfy the sink. However, as shown in Figure 2b, by making use of 50 kg/s of the gas source at $C = 5\%$ to satisfy the gas sink that can tolerate a higher impurity of 10%, it is possible to avoid purging the gas source directly as emission. Doing so reduces not only the waste gas generation but also the fresh gas intake, in both cases, by 50 kg/s.

The two final steps in getting the minimum utility targets involve setting up the cumulative impurity load cascade (cum Δm , column 8), and finally calculating the flowrate of fresh gas needed at each concentration level, or the *interval fresh*

Table 2. Infeasible Gas Cascade Table for Example 1 (Nitrogen Integration)

level, <i>k</i>	concentration <i>C_k</i> (wt/wt %)	$\sum_j F_j$ (kg/s)	$\sum_i F_i$ (kg/s)	$\sum_i F_i - \sum_j F_j$ (kg/s)	<i>F_C</i> (kg/s)	Δm (kg/s)	cum Δm (kg/s)	<i>F_{F,k}</i> (kg/s)
					0			
1	0				0			
2	0.1	5.5		-5.5	-5.5	-0.006	-0.006	-2.750
3	0.2	3.0		-3.0	-8.5	-0.017	-0.023	-5.625
4	0.4		5.5	5.5	-3	-0.045	-0.068	-3.553
5	1.9		3.0	3.0	0	0.000	-0.068	-0.068
6	100							

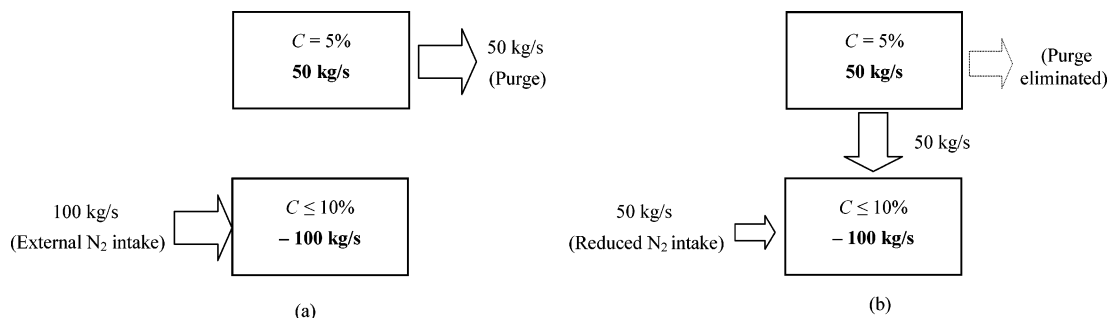


Figure 2. Concept of gas cascade: (a) without nitrogen cascading; (b) with nitrogen cascading.

Table 3. Feasible GCT for Example 1 (Nitrogen Integration)

level, k	concentration C_k (wt/wt %)	$\sum_j F_j$ (kg/s)	$\sum_i F_i$ (kg/s)	$\sum_i F_i - \sum_j F_j$ (kg/s)	F_C (kg/s)	Δm (kg/s)	cum Δm (kg/s)
					$F_F = 5.625$		
1	0				5.625	0.006	
2	0.1	5.5		-5.5	0.125	0.000	0.006
3	0.2	3.0		-3.0	-2.875	-0.006	0.006
4	0.4		5.5	5.5	2.625	0.039	0 (pinch) 0.039
5	1.9		3.0	3.0	$F_D = 5.625$	5.518	
6	100						5.558

gas flowrate, ($F_{F,k}$). The largest negative $F_{F,k}$, which actually represents the minimum fresh gas target that cascaded down the cumulative gas flowrate column (F_C), yields the minimum discharge flowrate.

Note that the impurity load (Δm) shown in Column 7 of Table 2 is obtained from the product of cumulative gas flowrate (F_C) and the concentration difference across two concentration levels (ΔC). Cascading the impurity load down the concentration levels of column 8 yields the cumulative load (cum Δm). Note that the (cum Δm) is essentially the numerical equivalent of the surplus diagram.³⁴ Table 2 is an infeasible gas cascade table (GCT) since the (cum Δm) value is negative. This means that there is insufficient utility gas at the respective concentration level. The amount of interval fresh gas flowrate ($F_{F,k}$) is obtained by dividing cum Δm by the difference between the concentration at level k (C_k) and the fresh feed concentration (C_F), i.e.,

$$F_{F,k} = \frac{\text{cum } \Delta m}{C_k - C_F} \quad (1)$$

The absolute value of the largest negative $F_{F,k}$, i.e., 5.625 kg/s, is essentially the minimum fresh gas flowrate target (F_F). This amount is cascaded down the F_C column of the feasible GCT in Table 3, to produce the minimum nitrogen discharge flowrates (F_D) of 5.625 kg/s. The minimum fresh feed and discharge flowrates identified by the GCA correspond to a reduction of 34% from the original feed and discharge flowrates (sum of the individual process flowrates in Table 1). The pinch concentration for the gas network exists at 0.4%, where there is zero cum Δm . From the limiting data in Table 1, emission from the drying process (SR₂) was identified as the *pinch-causing source* for this example. To achieve the minimum gas targets, 2.875 kg/s of SR₂, which exists between intervals $k = 3$ and $k = 4$, should be allocated to the region above the pinch, while 2.625 kg/s of SR₂ between $k = 4$ and 5 should be allocated below the pinch. The exact pinch-causing stream distribution to meet the minimum gas targets is termed as the *resource*

allocation targets.^{25,28,49} In the next section, a case study involving oxygen reuse/recycle will be presented to illustrate the network-design technique to achieve the various network targets established.

Example 2—Oxygen Integration

Some preliminary work on oxygen integration has focused on the optimum reuse of oxygen in a wastewater treatment plant.^{50–52} The following case study is an attempt to explore the interactions and possible site-wide integration between process and utility sections for maximum oxygen recovery.

Figure 3 shows a process plant that consumes a large amount of high-purity oxygen in its oxidation processes. A relatively small portion of the oxygen is also fed to enhance the combustion system as well as for the aerobic section of the wastewater treatment plant. The sources of oxygen emitted from the oxygen-consuming processes are still of good quality and should, therefore, be reused/recycled to reduce the fresh oxygen

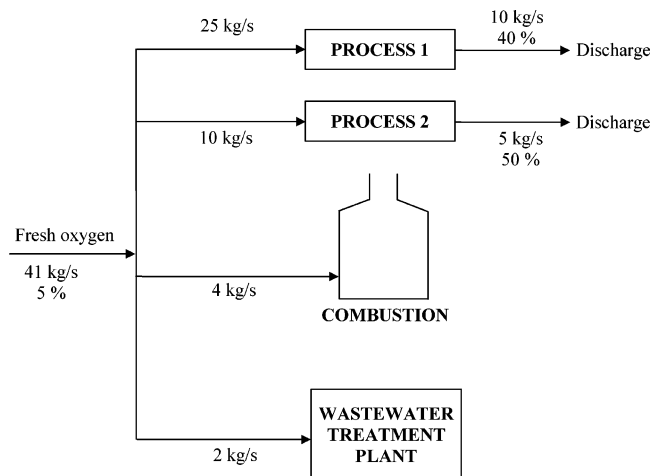


Figure 3. Process flow diagram for oxygen integration in Example 2.

Table 4. Limiting Data for Example 2 (Oxygen Integration)

gas sinks, SK _j		flowrate	concentration
<i>j</i>	stream	<i>F_j</i> (kg/s)	<i>C_j</i> (wt/wt %)
1	process 1	25	10
2	process 2	10	10
3	combustion	4	65
4	wastewater treatment	2	65

gas sources, SR _i		flowrate	concentration
<i>i</i>	stream	<i>F_i</i> (kg/s)	<i>C_i</i> (wt/wt %)
1	process 1	10	40
2	process 2	5	50
fresh supply		to be determined	5

Table 5. GCT for Example 2 (Oxygen Integration)

level, <i>k</i>	concentration <i>C_k</i> (wt/wt %)	$\sum_j F_j$ (kg/s)	$\sum_i F_i$ (kg/s)	$\sum_i F_i - \sum_j F_j$ (kg/s)	<i>F_C</i> (kg/s)	Δm (kg/s)	cum Δm (kg/s)
<i>F_F</i> = 30							
1	5			0			
2	10	35		-35	30	1.50	1.50
3	40		10	10	-5	-1.50	0.00
4	50		5	5	5	0.50 (pinch)	0.50
5	65	6		-6	10	1.50	2.00
6	100			0	<i>F_D</i> = 4	1.40	3.40

intake. Limiting data for all oxygen sinks and sources are given in Table 4. Note that, since GCA is able to handle impure fresh feeds, the 5% impurity in fresh oxygen supply should not pose any problems in getting accurate minimum gas flowrate targets. The GCT for this case study (Table 5) gives fresh oxygen supply (*F_F*) and discharge (*F_D*) targets at 30 kg/s and 4 kg/s, respectively. This corresponds to a fresh oxygen reduction of 27% and a discharge gas reduction of 73%, as compared to the base-case system shown in Figure 3.

Network Design

This section briefly reviews the design technique for a utility gas network.³⁵ Note that alternative techniques for water-network design^{10,26,31} may also be applied in combination with the flowrate targets and pinch points established using the GCA technique.

The pinch concentration identified in the targeting stage plays an important role in designing a network for maximum gas reuse/recycle. The pinch divides a network into two design regions, i.e., the regions above the pinch and those below the pinch, for problems involving a single pinch. It is essential to

observe the pinch division in order to achieve the minimum flowrates established during the targeting stage. In the region above the pinch, cumulative impurity load surplus is in mass balance with load deficit. On the other hand, there is always excess impurity load in the region below the pinch. The region above the pinch is, therefore, controlling the overall gas flowrate balance for the network and, hence, is the most constrained part of the network.

Following the above observation, in designing a gas network, a gas source above the pinch (including the external gas supply) must not be fed to a gas sink below the pinch. Violating this rule will result in a higher fresh gas flowrate. This rule also applies during mixing of different gas sources belonging to the different pinch regions. The pinch-causing stream(s) that exist at the pinch concentration is, however, an exception to this rule. Recall that part of the pinch-causing stream(s) belongs to the region above the pinch, while the remaining belongs to the region below the pinch.

To maintain steady-state operation of the gas-consuming processes, we need to maintain a uniform mass flowrate and concentration at the inlet stream of the gas-consuming process. The following guidelines can be used to design the network:^{10,35}

(a) For sinks

$$\text{Flowrate: } \sum_i F_{SR_i, SK_j} = F_{SK_j} \quad (2)$$

where *F_{ij}* is the flowrate between source *SR_i* and sink *SK_j*.

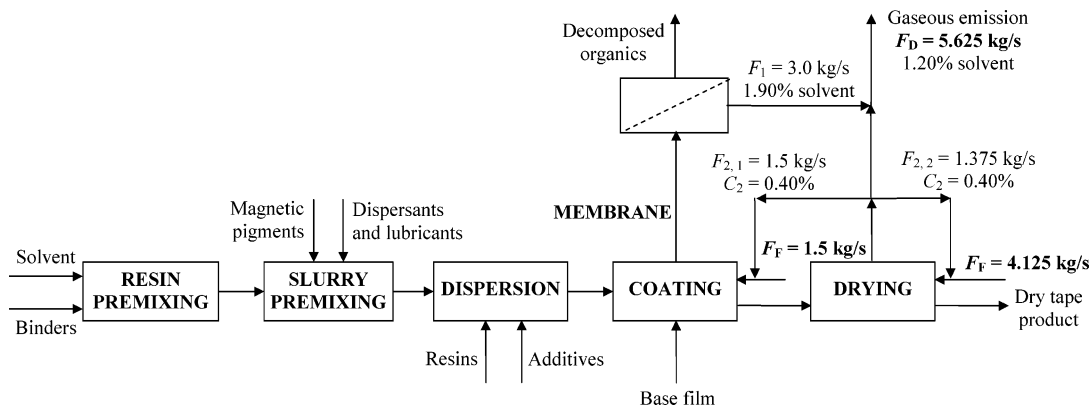
$$\text{Gas content: } \sum_i F_{SR_i, SK_j} C_{SR_i} = F_{SK_j} C_{SK_j} \quad (3)$$

(b) For sources

$$\text{Flowrate: } \sum_i F_{SR_i, SK_j} = F_{SR_i} \quad (4)$$

Equations 2 and 3 are constraints/limits representing the flowrate and the gas content required by a sink. Equation 4 states that the flowrate of source *i* to be fed to sink *j* is limited by the availability of sink *j*. Any gas from a source that is not fed to a sink will leave as a discharge stream.

The gas network for Examples 1 and 2 designed using the above formulation is shown in Figures 4 and 5, respectively. The flowrate targets as well as the gas allocation targets for the pinch-causing stream identified during the targeting stage are realized here. Note that, during the design stage, the gas network should be designed independently for the individual pinch regions. This is to avoid the gas sources from being fed to the

**Figure 4.** Process flow diagram of magnetic tape manufacturing process (Example 1) with exhaust gas reuse and recycling.

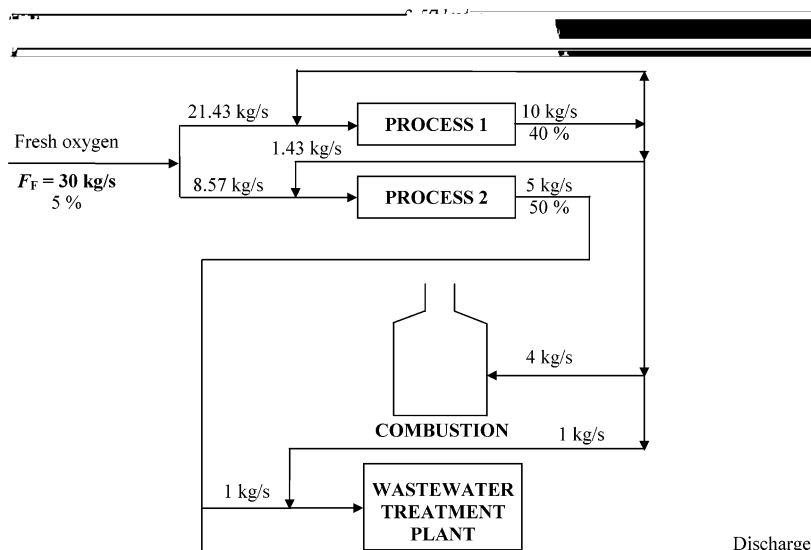


Figure 5. Oxygen reuse/recycle network for Example 2.

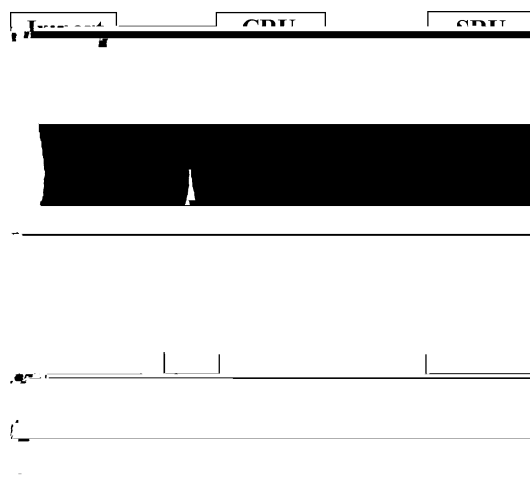


Figure 6. Refinery hydrogen network in Example 3: numbers represent the total gas flowrate in mol/s and impurity concentration (mol %).³⁴

sinks in different design regions as well as any inappropriate mixing of gas sources between regions, particularly in a multiple-pinch problem. Note also that many network design options may result as the number of gas sinks and sources increase. A designer can always impose other constraints on network design, such as forbidden or forced connections, due to safety, operation, or geographical reasons.

Example 3—Hydrogen Integration

Since hydrogen is a valuable utility gas in crude oil refineries and petrochemical plants, hydrogen reuse and recycle has been the subject of extensive research.^{32–41} Among the most promising graphical tools to set the minimum hydrogen targets are the hydrogen surplus diagram³⁴ and the material reuse/recycle pinch diagram.²³

Figure 6 shows a hydrogen network case study from Alves and Towler,³⁴ where the existing fresh hydrogen consumption is reported at 277.2 mol/s. There are four hydrogen-consuming processes in this case study, which consist of hydrocracker unit (HCU), naphtha hydrotreater (NHT), cracked naphtha hydrotreater (CNHT), and diesel hydrotreater (DHT). There are

Table 6. Limiting Data for Example 3 (Hydrogen Integration)^{34,23}

gas sinks, SK _j		flowrate	concentration
<i>j</i>	stream	<i>F_j</i> (mol/s)	<i>C_j</i> (mol %)
1	HCU	2495.0	19.39
2	NHT	180.2	21.15
3	CNHT	720.7	24.86
4	DHT	554.4	22.43
gas sources, SR _i		flowrate	concentration
<i>i</i>	stream	<i>F_i</i> (mol/s)	<i>C_i</i> (mol %)
1	HCU	1801.9	25.00
2	NHT	138.6	25.00
3	CNHT	457.4	30.00
4	DHT	346.5	27.00
5	SRU	623.8	7.00
6	CRU	415.8	20.00
fresh supply		to be determined	5.00

two hydrogen-producing facilities in this network, i.e., catalytic-reforming unit (CRU) and steam-reforming unit (SRU). These are the internal hydrogen sources for the network, and their use is to be maximized before considering the purchase of hydrogen. A fresh hydrogen supply having 5% impurity is available for this case study. Limiting data for all hydrogen sources and sinks are shown in Table 6.³⁴ Note that the limiting data for a hydrogen network cannot be directly extracted from its process flow diagram such as that in Figure 6; the detailed procedure to extract the limiting data is presented elsewhere.³⁴

The results of GCA are shown in Table 7. Table 7 shows the minimum fresh hydrogen target (*F_F*) and discharge flowrate (*F_D*) at 268.82 mol/s and 102.52 mol/s, respectively. These values agree with that reported by the graphical targeting tools.^{23,34} The GCT identifies the CNHT as the pinch-causing stream, from which 354.88 mol/s must be sent to the region above the pinch and 102.52 mol/s must be sent to the region below the pinch.

To further reduce fresh hydrogen intake, hydrogen sources can be partially treated for reuse and recycle in the hydrogen sinks. The next section demonstrates how GCA can be used to appropriately place a hydrogen purifier in the context of an overall process.

Table 7. GCT for Example 3 (Hydrogen Integration)

level, k	concentration C_k (mol %)	$\Sigma_j F_j$ (mol/s)	$\Sigma_i F_i$ (mol/s)	$\Sigma_i F_i - \Sigma_j F_j$ (mol/s)	F_C (mol/s)	Δm (mol/s)	cum Δm (mol/s)
					$F_F = 268.821$		
1	5.00				268.821	5.3764	
2	7.00		623.8	623.8	892.621	110.5957	5.3764
3	19.39	2495.0		-2 495.0	-1 602.379	-9.7745	115.9722
4	20.00		415.8	415.8	-1 186.579	-13.6457	106.1977
5	21.15	180.2		-180.2	-1 366.779	-17.4948	92.5520
6	22.43	554.4		-554.4	-1 921.179	-46.6846	75.0572
7	24.86	720.7		-720.7	-2 641.879	-3.6986	28.3726
8	25.00		1940.5	1 940.5	-701.379	-14.0276	24.6739
9	27.00		346.5	346.5	-354.879	-10.6464	10.6464
10	30.00		457.4	457.4	$F_D = 102.521$	71.7647	0.0000 (pinch)
11	100.00						71.7647

Table 8. GCT for Example 3 (Regeneration with Membrane)

level, k	concentration C_k (mol %)	$\Sigma_j F_j$ (mol/s)	$\Sigma_i F_i$ (mol/s)	$\Sigma_i F_i - \Sigma_j F_j$ (mol/s)	F_C (mol/s)	Δm (mol/s)	cum Δm (mol/s)
1	2.00		69.6 (permeate)	69.6	69.570	208.71	
2	5.00			$F_F = 196.8$	266.325	532.65	208.71
3	7.00		623.8	623.8	890.125	11 028.65	741.36
4	19.39	2495.0		-2 495.0	-1 604.875	-978.97	11 770.01
5	20.00		415.8	415.8	-1 189.075	-1 367.44	10 791.03
6	21.15	180.2		-180.2	-1 369.275	-1 752.67	9 423.60
7	22.43	554.4		-554.4	-1 923.675	-4 674.53	7 670.92
8	24.86	720.7		-720.7	-2 644.375	-370.21	2 996.39
9	25.00		1940.5	1940.5	-703.875	-1 407.75	2 626.18
10	27.00		346.5	346.5	-357.375	-1 072.13	1 218.43
11	30.00		354.9	354.9	-2.475	-146.31	146.31
12	89.11		33.0 (retentate)	33.0	$F_D = 30.475$	331.84	0.00 (pinch)
13	100.00						331.87

Appropriate Placement of Hydrogen Purifier

Hydrogen purifiers manipulate the properties (e.g., impurity concentration, flowrate, etc.) of hydrogen streams to match the desired properties of hydrogen sinks. Common hydrogen purifiers include pressure swing adsorption (PSA), membrane separation, and cryogenic separation. In contrast to a water regeneration unit that normally produces a single product stream, the hydrogen purification units typically separate single hydrogen feed streams into two product streams, i.e., one with a lower impurity concentration than that of the other. The low-impurity product stream can either be reused in another hydrogen-consuming process or recycled to the same process. A high-impurity byproduct stream is either purged or used as fuel. In the context of an overall network, a hydrogen purifier may be placed above, across, and below the pinch, with purification across the pinch being the best option.³⁷

When dealing with purifier placement, assessment of the impact of changes involves repetitive revisions of the flowrate targets and pinch relocation. Such tasks can be quite cumbersome with the use of graphical tools. The GCA technique, which is very amenable to computer programming has managed to overcome this problem.

Given a gas-separation membrane with a hydrogen recovery of 95%,³⁵ one possible option is to purify the portion of the pinch-causing stream (CNHT) that lies below the pinch at 30%, i.e., 102.52 mol/s to a product stream at 2% impurity. From material-balance calculations, the flowrate of this high-quality permeate stream is 69.57 mol/s. The flowrate and impurity concentration for the retentate stream are 32.95 mol/s and 89.11%, respectively. Adding these two new hydrogen sources at their respective concentration levels yields a new GCT, as shown in Table 8. Note that the permeate stream from membrane

Table 9. GCT for Example 3 (Regeneration with PSA)

level, k	concentration C_k (mol %)	$\sum_j F_j$ (mol/s)	$\sum_i F_i$ (mol/s)	$\sum_i F_i - \sum_j F_j$ (mol/s)	F_C (mol/s)	Δm (mol/s)	cum Δm (mol/s)
1	0.10		64.7 (product)	64.7			
2	5.00			$F_F = 200.0$	64.650	316.785	316.79
3	7.00		623.8	623.8	264.641	529.28	846.07
4	19.39	2495.0		-2 495.0	888.441	11 007.79	11 853.86
5	20.00		415.8	415.8	-1 606.559	-980.00	10 873.86
6	21.15	180.2		-180.2	-1 190.759	-1 369.37	9 504.48
7	22.43	554.4		-554.4	-1 370.959	-1 754.83	7 749.66
8	24.86	720.7		-720.7	-1 925.359	-4 678.62	3 071.04
9	25.00		1940.5	1 940.5	-2 646.059	-370.45	2 700.59
10	27.00		346.5	346.5	-705.559	-1 411.12	1 289.47
11	30.00		354.9	354.9	-359.059	-1 077.18	212.29
12	81.05		37.9 (residue)	37.9	-4.159	-212.29	0.00 (pinch)
13	100.00				$F_D = 33.711$	638.83	638.83

separation at 2% impurity is now much cleaner than the fresh hydrogen supply (5% impurity) and is located at the highest concentration level of the GCT. Hence, the fresh feed and discharge flowrates of the network are now reduced to 196.8 mol/s and 30.475 mol/s, respectively. Note also that the pinch concentration increases to the membrane retentate concentration of 89.11 mol %.

Let us now assess another hydrogen purifier, a PSA unit with a hydrogen-recovery value of 90% and capability to purify the hydrogen stream up to 0.10% impurity.³⁵ From material-balance calculations, one obtains the product and residue flowrates of 64.65 mol/s and 37.87 mol/s, respectively, with the residue-stream impurity concentration at 81.05%. Resetting targets using GCA technique gave a reduced fresh hydrogen flowrates of 200 mol/s, which corresponds to a reduced discharge flowrate of 33.711 mol/s. GCT for this option is shown in Table 9. Note that, in this case, the PSA has also purified the hydrogen source to a higher level relative to the fresh hydrogen supply.

We can now conclude that a gas-separation membrane plays a more important role in reducing the fresh hydrogen target in this hydrogen network, compared to the PSA. This is due to the fact that a membrane with a higher recovery of 95% (as compared to PSA with a recovery of 90%) generates an additional amount of hydrogen from the region of excess hydrogen below the pinch to the region of hydrogen deficit above the pinch. However, one should note that a purifier that gives a higher recovery may not necessarily be better. A purifier that produces a product stream at higher purity (e.g., PSA in this case) may sometimes be a better selection in reducing the overall flowrates of the gas network. We will examine this situation in the multiple-pinch problem that follows.

Example 4—Multiple-Pinch Problems

Table 10 shows the hydrogen sources and sinks data for Example 4. There are six hydrogen sinks and seven hydrogen sources in this network. The fresh hydrogen supply in this case has an impurity concentration of 0.10%. Using GCA, the minimum fresh supply and discharge flowrates are found to be

Table 10. Hydrogen Source and Sink Data for Example 4 (multiple-pinch problem)

gas sinks, SK_j	flowrate, F_j (mol/s)	concentration, C_j (mol %)
1	120.00	0.10
2	27.80	1.40
3	80.00	2.50
4	60.00	2.50
5	100.00	3.00
6	150.00	10.00

gas sources, SR_i	flowrate, F_i (mol/s)	concentration, C_i (mol %)
1	80.00	1.70
2	75.00	15.00
3	28.55	4.00
4	80.00	5.00
5	120.00	10.00
6	40.00	1.70
7	80.00	2.50
fresh hydrogen supply	to be determined	0.10

125.21 mol/s and 90.96 mol/s, respectively (Table 11). This is a typical multiple-pinch problem. Two pinches are observed in the GCT, i.e., at impurity concentrations of 1.70% and 5.0%. Following the terminology in the water network,^{22,25} the pinch concentration at the lower impurity concentration of 1.70% is designated as the *limiting pinch*, i.e., the pinch that influences the overall gas flowrates when any process changes take place.

In problems involving multiple pinches, more than two design regions may exist. For Example 4, three distinct design regions exist due to the existence of a limiting pinch at 1.70% purity and a secondary pinch at 5.0%. Utilizing the GCA technique, one can quickly identify the pinch-causing stream(s) and the exact hydrogen allocation for the hydrogen source(s) in each of the design region. This can then be verified using the network-design procedure described previously. One of the possible networks that achieves the various established network targets is shown in Figure 7.

The correct identification of the limiting pinch is crucial in problems involving multiple pinch points and near pinches.

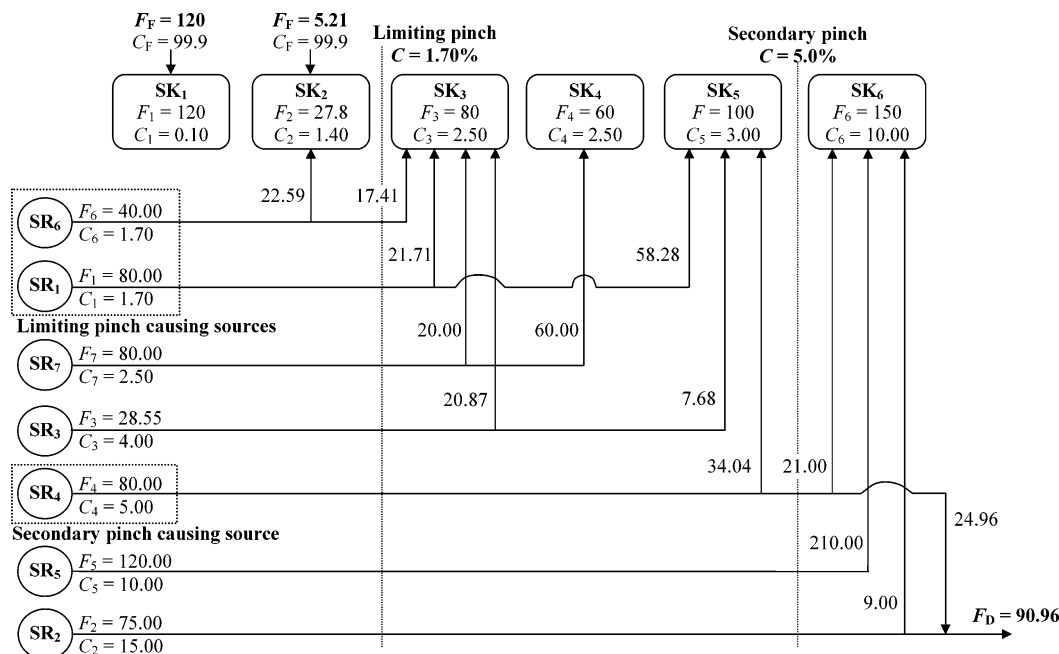


Figure 7. One possible network design for Example 4: flowrate in mol/s while composition in mol % impurity.

Table 11. GCT for Example 4 (Multiple-Pinch Problem)

level <i>k</i>	concentration <i>C_k</i> (mol %)	$\sum_j F_j$ (mol/s)	$\sum_i F_i$ (mol/s)	$\sum_i F_i - \sum_j F_j$ (mol/s)	<i>F_C</i> (mol/s)	Δm (mol/s)	cum Δm (mol/s)
					<i>F_F</i> = 125.21		
1	0.10	120		-120			
2	1.40	27.8		-27.8	5.21	0.068	0.068
3	1.70		120	120	-22.59	-0.068	0.00
4	2.50	140	80	-60	97.41	0.779	(limiting pinch) 0.779
5	3.00	100		-100	37.41	0.187	0.966
6	4.00		28.55	28.55	-62.59	-0.626	0.340
7	5.00		80	80	-34.04	-0.340	0.000
8	10.00	150	120	-30	45.96	2.298	(secondary pinch) 2.298
9	15.00		75	75	15.96	0.798	3.096
10	100				<i>F_D</i> = 90.96	77.318	80.414

Identification of the wrong pinch point will result in missed opportunities during hydrogen-network debottlenecking. This is particularly important for problems involving multiple pinches, where the region above the limiting pinch is the most-constrained part of the network. Debottlenecking strategy will only be beneficial when the hydrogen source flowrate is increased above the limiting pinch. Alternatively, decreasing the need of a hydrogen sink in the region above the limiting pinch will also achieve the debottlenecking objective. This is another area where quick and accurate determination of the true pinch point and flowrate targets is crucial in gas network analysis. Here, once again, GCA has an important role to play.

Given the same hydrogen purifiers in Example 3, we can now determine the appropriate selection of purifier that will reduce the overall hydrogen flowrates of the network. GCT for purifying the hydrogen source using a gas-separation membrane (with a hydrogen recovery of 95% and producing a permeate stream with 2% impurity) is shown in Table 12. Note that, though the secondary pinch purity at 5.0% has been removed,

the minimum flowrate targets for this option are the same as before regeneration (Table 11). In other words, no benefit is obtained by adding a new membrane-separation unit into the network. This is due to the fact that the overall hydrogen flowrate targets are controlled by the most-constrained part of the network, i.e., the region above the limiting pinch. Hence, the purified product stream must achieve an impurity concentration lower than the limiting pinch concentration before the fresh hydrogen requirement can be reduced.

Another hydrogen purifier in Example 3 that was used for evaluation was the PSA unit with a hydrogen recovery of 90%, a resulting residue stream at 52.43% purity, and a product stream containing 0.10% impurity, which was lower than the limiting pinch. Resetting targets using GCA gave a reduced fresh hydrogen target of 54.23 mol/s and a discharge target of 19.98 mol/s (Table 13). Because of the reallocation of hydrogen sources within the whole network, the original limiting and secondary pinches disappeared, while a new pinch emerged at the residue stream concentration of 52.43%. Hence, we can

Table 12. GCT for Example 4 (Regeneration With Membrane)

level, <i>k</i>	concentration, C_k (mol %)	$\Sigma_j F_j$ (mol/s)	$\Sigma_i F_i$ (mol/s)	$\Sigma_i F_i - \Sigma_j F_j$ (mol/s)	F_C (mol/s)	Δm (mol/s)	cum Δm (mol/s)
					$F_F = 125.21$		
1	0.10	120.00		-120			
2	1.40	27.80		-27.8	5.21	0.068	0.068
3	1.70		120.00	120	-22.59	-0.068	0.000
4	2.00		104.69 (permeate)	104.7	97.41	0.292	(pinch) 0.292
5	2.50	140.00	80.00	-60	202.11	1.011	1.303
6	3.00	100.00		-100	142.11	0.711	2.013
7	4.00		28.55	28.5	42.11	0.421	2.434
8	5.00		80.00	80	70.66	0.707	3.141
9	10.00	150.00		-150	150.66	7.533	10.674
10	15.00		75.00	75	0.66	0.033	10.707
11	64.72		15.31 (retentate)	15.3	75.66	37.616	48.323
12	100				$F_D = 90.96$	32.092	80.414

Table 13. GCT for Example 4 (Regeneration with PSA)

level, <i>k</i>	concentration C_k (mol %)	$\Sigma_j F_j$ (mol/s)	$\Sigma_i F_i$ (mol/s)	$\Sigma_i F_i - \Sigma_j F_j$ (mol/s)	F_C (mol/s)	Δm (mol/s)	cum Δm (mol/s)
					$F_F = 54.23$		
1	0.10	120.00	97.30 (product)	-22.70			
2	1.40	27.80		-27.80	31.53	0.410	0.410
3	1.70		120.00	120.00	3.73	0.011	0.421
4	2.50	140.00	80.00	-60.00	123.73	0.990	1.411
5	3.00	100.00		-100.00	63.73	0.319	1.730
6	4.00		28.55	28.55	-36.27	-0.363	1.367
7	5.00		80.00	80.00	-7.72	-0.077	1.290
8	10.00	150.00		-150.00	72.28	3.614	4.904
9	15.00		75.00	75.00	-77.72	-3.886	1.018
10	52.43		22.70 (residue)	22.70	-2.72	-1.018	0.000 (pinch)
11	100				$F_D = 19.98$	9.506	9.506

conclude that PSA plays a more important role in reducing the overall hydrogen targets in this network, as compared to a gas-separation membrane. By purifying the hydrogen source in the region of excess hydrogen (below the secondary pinch), PSA supplies an extra amount of hydrogen to the region of hydrogen deficit (above the limiting pinch). Finally, note also that all preliminary screening for appropriate placement of hydrogen purifier is carried out prior to the detailed design of the network.

Conclusion

The cascade analysis tool for targeting the minimum flowrates for water and property-based networks has been extended to utility gas networks. Gas cascade analysis (GCA) enables quick and accurate identification of the minimum flowrate targets, pinch point location(s), and resource allocation targets for a utility gas network. Appropriate selection of gas-purification techniques can be systematically assessed using the GCA.

Problems involving multiple pinches can now be handled more efficiently, more accurately, and with much less effort.

Acknowledgment

The financial support from Ministry of Science, Technology and Environment, Malaysia, through Intensified Research Priority Area (IRPA) research grant and National Science Fellowship (NSF) scholarship is gratefully acknowledged.

Literature Cited

- (1) Hohmann, E. C. Optimum Networks for Heat Exchanger. Ph.D. Thesis, University of Southern California, Los Angeles, CA, 1971.
- (2) Linnhoff, B.; Flower, J. R. Synthesis of Heat Exchanger Networks. *AIChE J.* **1978**, *24* (4), 633-654.
- (3) Umeda, T.; Itoh, J.; Shiroko, K. Heat Exchange System Synthesis. *Chem. Eng. Prog.* **1978**, *73* (7), 70-76.
- (4) Linnhoff, B.; Townsend, D. W.; Boland, D.; Hewitt, G. F.; Thomas, B. E. A.; Guy, A. R.; Marshall, R. H. *A User Guide on Process Integration for the Efficient Use of Energy*; IChemE: Rugby, U.K., 1982.

- (5) Smith, R. *Chemical Process Design*. McGraw-Hill: New York, 1995.
- (6) Shenoy, U. V. *Heat Exchanger Network Synthesis: Process Optimization by Energy and Resource Analysis*; Gulf Publishing Co.: Houston, TX, 1995.
- (7) El-Halwagi, M. M.; Manousiouthakis, V. Synthesis of Mass Exchange Networks. *AIChE J.* **1989**, *35* (8), 1233–1244.
- (8) El-Halwagi, M. M.; Manousiouthakis, V. Automatic Synthesis of Mass Exchange Networks with Single Component Targets. *Chem. Eng. Sci.* **1990**, *45* (9), 2813–2831.
- (9) El-Halwagi, M. M.; Manousiouthakis, V. Simultaneous Synthesis of Mass Exchange and Regeneration Networks. *AIChE J.* **1990**, *36* (8), 1209–1219.
- (10) El-Halwagi, M. M. *Pollution Prevention through Process Integration: Systematic Design Tools*; Academic Press: San Diego, CA, 1997.
- (11) El-Halwagi, M. M. Pollution Prevention through Process Integration. *J. Clean Prod. Process.* **1998**, *1*, 5–19.
- (12) El-Halwagi, M. M.; Spriggs, H. D. Solve Design Puzzles with Mass Integration. *Chem. Eng. Prog.* **1998**, *94*, 25–44.
- (13) Hallale, N.; Fraser, D. M. Capital and Total Cost Targets for Mass Exchange Networks. Part 1 and 2. *Comput. Chem. Eng.* **2000**, *23*, 1661–1699.
- (14) Hallale, N.; Fraser, D. M. Supertargeting for Mass Exchange Networks. Part 1 and 2. *Trans. Inst. Chem. Eng., Part A* **2000**, *78*, 202–216.
- (15) Foo, C. Y.; Manan, Z. A.; Yunus, R. M.; Aziz, R. A. Synthesis of mass exchange network for batch processes. Part I: Utility targeting. *Chem. Eng. Sci.* **2004**, *59* (5), 1009–1026.
- (16) Foo, C. Y.; Manan, Z. A.; Yunus, R. M.; Aziz, R. A. Synthesis of Mass Exchange Network for Batch Processes. Part II: Minimum Units Target and Batch Network Design. *Chem. Eng. Sci.* **2005**, *60* (5), 1349–1362.
- (17) Wang, Y. P.; Smith, R. Wastewater Minimisation. *Chem. Eng. Sci.* **1994**, *49*, 981–1006.
- (18) Dhole, V. R.; Ramchandani, N.; Tainsh, R. A.; Wasilewski, M. Make Your Process Water Pay for Itself. *Chem. Eng.* **1996**, *103* (1), 100–103.
- (19) Sorin, M.; Bédard, S. The Global Pinch Point in Water Reuse Networks. *Trans. Inst. Chem. Eng., Part B* **1999**, *77*, 305–308.
- (20) Polley, G. T.; Polley, H. L. Design Better Water Networks. *Chem. Eng. Prog.* **2000**, *96* (2), 47–52.
- (21) Feng, X.; Seider, W. D. New Structure and Design Method for Water Networks. *Ind. Eng. Chem. Res.* **2001**, *40*, 6140–6146.
- (22) Hallale, N. A New Graphical Targeting Method for Water Minimisation. *Adv. Environ. Res.* **2002**, *6* (3), 377–390.
- (23) El-Halwagi, M. M.; Gabriel, F.; Harell, D. Rigorous Graphical Targeting for Resource Conservation via Material Recycle/Reuse Networks. *Ind. Eng. Chem. Res.* **2003**, *42*, 4319–4328.
- (24) Manan, Z. A.; Foo, C. Y. Setting Targets for Water and Hydrogen Networks Using Cascade Analysis. Presented at AIChE Annual Meeting, San Francisco, CA, 2003.
- (25) Manan, Z. A.; Tan, Y. L.; Foo, D. C. Y. Targeting the Minimum Water Flowrate Using Water Cascade Analysis Technique. *AIChE J.* **2004**, *50* (12), 3169–3183.
- (26) Prakash, R.; Shenoy, U. V. Targeting and Design of Water Networks for Fixed Flowrate and Fixed Contaminant Load Operations. *Chem. Eng. Sci.* **2005**, *60* (1), 255–268.
- (27) Foo, D. C. Y.; Manan, Z. A.; Tan, Y. L. Synthesis of Maximum Water Recovery Network for Batch Process Systems. *J. Clean. Prod.* **2005**, *13* (15), 1381–1394.
- (28) Manan, Z. A.; Foo, D. C. Y.; Tan, Y. L. Use Cascade Analysis Technique to Set Baseline Targets and Optimise Water Network. *Chem. Eng. Prog.* **2006**, in press.
- (29) Foo, D. C. Y.; Manan, Z. A.; El-Halwagi, M. M. Correct Identification of Limiting Water Data for Water Network Synthesis. *Clean Technol. Environ. Policy* **2006**, *8* (2), 96–104.
- (30) Almutlaq, A. M.; Kazantzi, V.; El-Halwagi, M. M. An algebraic approach to targeting waste discharge and impure fresh usage via material recycle/reuse networks. *Clean Technol. Environ. Policy* **2005**, *7*, 294–305.
- (31) Aly, S.; Abeer, S.; Awad, M. A New Systematic Approach for Water Network Design. *Clean Technol. Environ. Policy* **2005**, *7*, 154–161.
- (32) Towler, G. P.; Mann, R.; Serriere, A. J.-L.; Gabaude, C. M. D. Refinery Hydrogen Management: Cost Analysis of Chemically Integrated Facilities. *Ind. Eng. Chem. Res.* **1996**, *35* (7), 2378–2388.
- (33) Linnhoff, B.; Tainsh, R.; Wasilewski, M. Hydrogen Network Management. Presented at The European Refining Technology Conference, Paris, 1999.
- (34) Alves, J. J.; Towler, G. P. Analysis of Refinery Hydrogen Distribution Systems. *Ind. Eng. Chem. Res.* **2002**, *41*, 5759–5769.
- (35) Hallale, N.; Liu, F. Refinery Hydrogen Management for Clean Fuels Production. *Adv. Environ. Res.* **2001**, *6*, 81–98.
- (36) Liu, F.; Hallale, N. Retrofit of Refinery Hydrogen Systems. In *Proceedings of European Symposium of Computer Aided Process Engineering 11 (ESCAPE 11)*; Gani, R., Jørgensen, S. B., Eds.; Elsevier: New York, 2001; pp 445–450.
- (37) Hallale, N.; Moore, I.; Vauk, D. Hydrogen: Liability or Asset? *Chem. Eng. Prog.* **2002**, *98* (9), 66–75.
- (38) Bealing, C.; Hutton, D. Hydrogen Pinch Analysis. *Chem. Eng.* **2002**, *109* (5), 56–61.
- (39) Bealing, C.; Hutton, D. Optimal Hydrogen Output. *Hydrocarbon Eng.* **2002**, *March*, 1–4.
- (40) Ricci, G.; Bealing, C. Using an Integrated Approach Conserves Energy and Hydrogen. *Hydrocarbon Process.* **2003**, *December*, 76–81.
- (41) Li, Z. Pinch Analysis of Hydrogen System in Refineries. In *Proceeding of Process System Engineering (PSE 2003)*; Chen, B., Westerbergh, A. W., Eds.; Elsevier: New York, 2003; pp 952–955.
- (42) Shelley, M. D.; El-Halwagi, M. M. Componentless Design of Recovery and Allocation Systems: A Functionality-Based Clustering Approach. *Comput. Chem. Eng.* **2000**, *24*, 2081–2091.
- (43) El-Halwagi, M. M.; Glasgow I. M.; Eden, M. R.; Qin, X. Property Integration: Componentless Design Techniques and Visualization Tools. *AIChE J.* **2004**, *50* (8), 1854–1869.
- (44) Qin, X.; Gabriel, F.; Harell, D.; El-Halwagi, M. M. Algebraic Techniques for Property Integration via Componentless Design. *Ind. Eng. Chem. Res.* **2004**, *43*, 3792–3798.
- (45) Kazantzi, V.; Harell, D.; Gabriel, F.; Qin, X.; El-Halwagi, M. M. Property-based Integration for Sustainable Development. In *Proceedings of European Symposium on Computer-Aided Process Engineering 14 (ESCAPE 14)*; Barbosa-Povoa, A., Matos, H., Eds.; Elsevier: New York, 2004; pp 1069–1074.
- (46) Kazantzi, V.; Qin, X.; Gabriel, F.; Harell, D.; El-Halwagi, M. M. Process Modification through Visualization and Inclusion Techniques for Property Based Integration. In *Proceedings of the Sixth Foundations of Computer Aided Design (FOCADP)*; Floudas, C. A., Agrawal, R., Eds.; CACHE Corp: Austin, TX, 2004; pp 279–282.
- (47) Kazantzi, V.; El-Halwagi, M. M. Targeting Material Reuse via Property Integration. *Chem. Eng. Prog.* **2005**, *101* (8), 28–37.
- (48) Kazantzi, V.; Qin, X.; Foo, D. C. Y.; El-Halwagi, M. M. Pollution Prevention through Property-Based Design. Presented in AIChE Annual Meeting, Cincinnati, OH, 2005.
- (49) Foo, D. C. Y.; Kazantzi, V.; El-Halwagi, M. M.; Manan, Z. A. Surplus Diagram and Cascade Analysis Technique for Targeting Property-Based Material Reuse Network. *Chem. Eng. Sci.* **2006**, *61* (8), 2626–2642.
- (50) Zhelev, T. K.; Ntlhakana, J. L. Energy-Environment Closed-Loop through Oxygen Pinch. *Comput. Chem. Eng.* **1999**, *23*, S79–S83.
- (51) Zhelev, T. K.; Bhaw, N. Combined Water–Oxygen Pinch Analysis for Better Wastewater Treatment Management. *Waste Manage.* **2000**, *20*, 665–670.
- (52) Zhelev, T. K. Wastewater Treatment Management Using Combined Water–oxygen-Thermal Pinch Analysis. In *Proceedings of European Symposium of Computer Aided Process Engineering 12 (ESCAPE 12)*; Grievink, J., van Schijndel, J., Eds.; Elsevier: New York, 2002; pp 391–396.

Received for review November 28, 2005
 Revised manuscript received May 7, 2006
 Accepted May 24, 2006