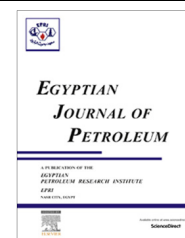




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FULL LENGTH ARTICLE

Automated targeting technique for indirect inter-plant hydrogen integration



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Abstract The increase of hydrogen demand has been one of the focal point recently. Indirect inter-plant hydrogen integration (IPHI) is a new issue for reducing hydrogen consumption and hydrogen discharge, in which hydrogen networks for different plants are integrated indirectly through centralized partitioning regeneration unit where hydrogen is regenerated for further reuse/recycle. This work is an extension of the automated targeting technique that was developed for single hydrogen network and the automated targeting that was developed for inter-plant water integration. The concentration cascade model has been developed to optimize hydrogen networks through partitioning regeneration unit such as pressure swing adsorption (PSA) or membrane separation. Two case studies from literature are studied to illustrate the applicability of the proposed model.

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1. Introduction

Hydrogen management is getting essential in refineries and petrochemical complexes, since refineries face an increasing challenge of meeting the growing demand for clean fuels. Along with the legislation for environment protection higher gasoline and diesel quality specifications have been implemented to reduce pollutants from automotive exhausts. Oil refineries consume hydrogen in large amounts during the past decade to remove sulfur and nitrogen compounds and produce lighter fuel with high quality, since crude oil gets heavier and contains more sulfur and nitrogen. Thus the hydrogen supply in many refineries is becoming a critical problem [1,2].

Hydrogen resources are units that either consume or produce hydrogen. Management of these two resources leads to lower consumption of hydrogen and improves refinery profitability [3,4]. The production sources of hydrogen in refining are catalytic reformers, hydrogen plants, ethylene plants and hydrogen import. Where the most common hydrogen consumption units are hydrocracker and hydrotreaters. The hydrogen demands of hydrogen consumers vary with the changes of operational load mostly and raw material [5–7]. As the demand for hydrogen grows, the management and optimization of hydrogen system in refinery is becoming increasingly important [8]. Development process integration has been proven as a promising approach in maximizing potential resource conservation [9]. The hydrogen network integration optimizes the hydrogen network with all the hydrogen demands and hydrogen supplies. Consequently it can significantly increase the reuse of the process hydrogen and minimize the hydrogen utility consumption. The management of hydrogen network

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Nomenclature

Abbreviations

AF	annualization factor	MINLP	mixed-integer nonlinear programming
A_{MEM}	membrane area, m ²	MP	mega Pascal
AWH	annual working hours	NLP	nonlinear programming
A	fractional interest rate per year	P	maximum pressure of the pipeline in MPa
CCR	continuous catalytic reformer unit	P_n^{feed}	inlet feed pressure to the regenerator
CNHT	cracked naphtha hydrotreater unit	$P_n^{product}$	top product pressure of the regenerator
D	length of the cross-plant pipeline, m	PSA	pressure swing adsorption unit
DHT	diesel hydrotreater unit	SK	sink
$F_{cp,k}$	feed flowrate to the regenerator from network, k	SR	source
$F_{FH,k}^{FIPHI}$	fresh inter-plant hydrogen integration for network, k	TAC	total annual cost measured by US\$
$F_{DH,k}^{FIPHI}$	inter-plant hydrogen discharge flowrate from network, k	UB _{cp}	upper bound of the cross-plant pipeline flowrate
$F_{i,k}^{EXP}$	flowrate exported from source i in network k to the regenerator	US\$	American dollar
$F_{SK,j,k}$	flowrate of sink j for network k	$W_{cost,k}$	unit cost of fresh hydrogen for network, k
$F_{SR,j,k}$	flowrate of source i for network k	$X_{INDIR,k}$	binary variable indicates the existence of a cross-plant pipeline from all sources of network k to the regenerator
$g_{cp,k}$	top product of the regenerator to network k	Y_F	inlet impurity concentration to the regenerator
GPU	cm ³ (STD) * 10 ⁻⁶ /cm ² s cmHg	$y_{H_2in\ feed}$	hydrogen concentration in the feed of the regenerator
HCU	hydrocracker unit	$y_{H_2in\ product}$	hydrogen concentration in the top product of the regenerator
I	is a source in hydrogen networks	Y_i	impurity concentration of source I
IPHI	inter-plant hydrogen integration	Y_p	impurity concentration of the top product of the regenerator
I_{pipe}	capital cost of pipeline, US\$	Y_r	impurity concentration of the bottom product of the regenerator
I_{psa}	capital cost of pressure swing adsorption, US\$	Y_r^{min}	the minimum concentration of all sources in the participating networks when they are regenerated individually
$I_{regenerator}$	capital cost of regenerator, US\$	Y_r^{max}	the maximum concentration of all sources in the participating networks when they are regenerated individually
ISO	isomerization unit	$Z_{INDIR,k}$	binary variable indicate the existence of a cross-plant pipeline from the bottom product of the regenerator to all sinks of network k
J	is a sink in hydrogen network	$\delta_{m,k}$	net material flowrate from level m for network k
JHT	jet fuel hydrotreater unit	$\varepsilon_{m,k}$	residue of impurity load from concentration level m for network k
L	number of years	α_L	hydrogen recovery
LB _{cp}	lower bound of the cross-plant pipeline flowrate		
$L_{cp,k}$	bottom product of the regenerator for network k		
L_{gH_2}	permeability of hydrogen component through membrane		
LP	linear programming		
M	index for concentration levels		
MMSCFd	million standard cubic feet per day		
$N_{INDIR,k}$	binary variable indicates the existence of a cross-plant pipeline from the top product of the regenerator to all sinks of network, k .		

involves two alternatives: reuse and regeneration [10]. For the two alternatives, many researchers focused on the optimization of hydrogen network by graphical or mathematical methods. For graphical optimization, Towler et al. [1] presented a value composite curve method to analyze the hydrogen network. Alves and Towler [2] presented the purity profiles and hydrogen surplus profiles to calculate the minimum fresh hydrogen consumption. Zhao et al. [11] proposed a simple graphical method for determining fresh hydrogen demand at any concentration. El Halwagi et al. [12] presented a rigorous graphical targeting technique to obtain the minimum fresh hydrogen utility. Agrawal and Shenoy [13] proposed a unified conceptual approach for water and hydrogen network. Zhao et al. [14] improved an iterative targeting method considering multiple impurities for hydrogen network.

For mathematical optimization, Liu and Zhang [15] introduced a detailed model of regeneration units by selecting an optimum regeneration unit. Foo et al. [16] presented an algebraic algorithmic method based on concentration intervals. Ng et al. [17,18] introduced the automated targeting technique based on pinch calculation for resource conservation problems for hydrogen reuse and hydrogen regeneration system. Lou et al. [19], presented an improved graphical targeting approach (pinch sliding approach) for water and hydrogen networks with different types of regenerator. The minimum flowrates of utility and regenerator feed are targeted by integrating the triangle rule and the optimal condition theorem. Zhou et al. [20], introduced a systematic modeling methodology for both the economic efficiency and environmental performance of hydrogen network. The economic efficiency is based on the

evaluation of total annual cost while the environmental performance is based on the CO₂ emission of the network. Yang et al. [9], presented an iterative method for hydrogen network design involving regeneration reuse. Liao et al. [21], proposed a new concept, mixing potential to study the disturbance resistance ability of networks. Lou et al. [22], presented a new method for the design of hydrogen networks with multiple contaminants. They minimized the utility consumption by minimizing the thermodynamic irreversibility of the satisfying process. The thermodynamic irreversibility which is minimized is the entropy for each sink. Lou et al. [23], introduced robust optimization as a framework for optimizing hydrogen network. Numbers of scenarios which represent possible future environments are included. Wu et al. [24] presented a mathematical optimization model for hydrogen network considering pure hydrogen load and inlet hydrogen concentration of hydrogen consuming reactors as constraints. Jhaveri et al. [25] developed five mathematical optimization models for optimizing hydrogen network in refinery. Two models are nonlinear program (NLP) networks and the other three are mixed integer nonlinear program (MINLP). Wang et al. [10] proposed a linear programming (LP) model for optimal operation of hydrogen network. Umana et al. [26] provide a framework to assess the interaction between hydroprocessing reactions and hydrogen distribution systems to improve overall network performance. They studied the effect of changes in light hydrocarbon composition in the recycle and purge of hydrogen consuming processes on hydrogen requirements.

A further means to enhance hydrogen recovery is via inter-plant hydrogen integration (IPHI) between hydrogen networks of different neighbor plants. Such technique can be used to achieve greater hydrogen saving than that technique when hydrogen conservation is implemented in individual plant separately. However in IPHI each participating plant seeks to maximize its own benefits [9,28].

Two types of inter-plant hydrogen integration schemes exist, direct and indirect integrations. In the direct inter-plant hydrogen integration, hydrogen sources and sinks from different networks are integrated directly via cross-plant pipelines [28]. Hence a hydrogen source in one network may be fed to another network as a new source. On the other hand in the indirect inter-plant hydrogen integration, hydrogen sources and sinks from different networks are integrated indirectly via a centralized hydrogen partitioning regeneration unit such as pressure swing adsorption (PSA) or membrane separation. The first targeting method in inter plant water system is addressed by Liao et al. [27], they presented the design of flexible multiple plant water networks. The design is based on the combination of pinch insight with mathematical programming. Chew et al. [28], analyzed the direct and indirect inter-plant water integration using mathematical optimization techniques. Chew and Foo [9], presented an automated targeting for direct and indirect inter plant water integration.

All the previous works for inter-plant integration are direct integration and indirect integration for single pass water regeneration system with one inlet stream and one outlet stream. The inter-plant integration with partitioning regeneration system with one inlet stream and two outlet streams as used in inter-plant hydrogen integration are not covered by the literature. In addition, the capital cost of the regenerator which is the function of the regenerator capacity (its feed flowrate)

has not been considered in targeting the minimum flowrate of the fresh utility in inter-plant integration.

In this paper, the automated targeting technique which was developed by Ng et al. [18] for single hydrogen network and the automated targeting technique that was developed for inter-plant water integration by Chew and Foo [9] are extended into inter-plant hydrogen integration.

In IPHI sources/sinks of different hydrogen networks are integrated via a centralized partitioning regeneration unit as pressure swing adsorption (PSA) or membrane separation. The hydrogen sources for any participating hydrogen network can either be integrated with the hydrogen sinks in the same hydrogen network or sent to the regeneration unit or sent to the discharge. The partitioning regenerator is used to improve hydrogen source quality sent from the participating networks giving two product streams, top product stream, and bottom product stream. The top and the bottom product regenerated streams of the regenerator can be either reused/recycled in the participating hydrogen networks or sent to the discharge.

Note that there are no direct connections between the sources and the sinks of the participating hydrogen networks. A mixed integer nonlinear program model (MINLP) is formulated in LINGO version 12. The model is solved using a PC with 1.7 GHz Intel Core i5-4210U CPU Processor.

2. Problem description

The inter-plant hydrogen integration problem that will be considered in this work can be **stated** as follows.

There is a set of hydrogen networks k of the fixed flow rate type problem. Each hydrogen network consists of a set of process sources $\{i = 1, 2, \dots, N_{\text{sources}}\}$ and a set of process sinks $\{j = 1, 2, \dots, N_{\text{sinks}}\}$. Each source i , has a flow rate of $F_{\text{SR},i}$, and a composition of single contaminant, y_i and each sink j , has a flow rate of $F_{\text{SK},j}$, and a maximum composition of single contaminant, Z_j . There are fresh inter-plant hydrogen sources ($F_{\text{FH},k}^{\text{IPHI}}$) that can be purchased to supplement additional hydrogen requirement of the process sink that is not satisfied by the process sources. There is a centralized partitioning regenerator (PSA or membrane unit) between the hydrogen networks for regenerating the sources from each network.

The indirect inter-plant hydrogen integration among these hydrogen networks is optimized to achieve the minimum total annual cost.

3. Automated targeting for partitioning regeneration system

The first automated targeting technique was presented by El Halwagi and Manousiathakis [29] for the synthesis of mass exchange network. For in-plant reuse/recycle integration system, Ng et al. [30] extended the automated targeting to resource conservation network (RCN) which is related to the water cascade analysis developed by Manan et al. [31]. Ng et al. [32] extended the automated targeting to the property based resource conservation network and also, to single-impurity resource conservation networks. For in-plant regeneration system, Ng et al. [18] extended the automated targeting to resource conservation networks for single-pass and partitioning waste regeneration systems. For inter-plant integration system, Chew and Foo [9] extended the automated targeting to

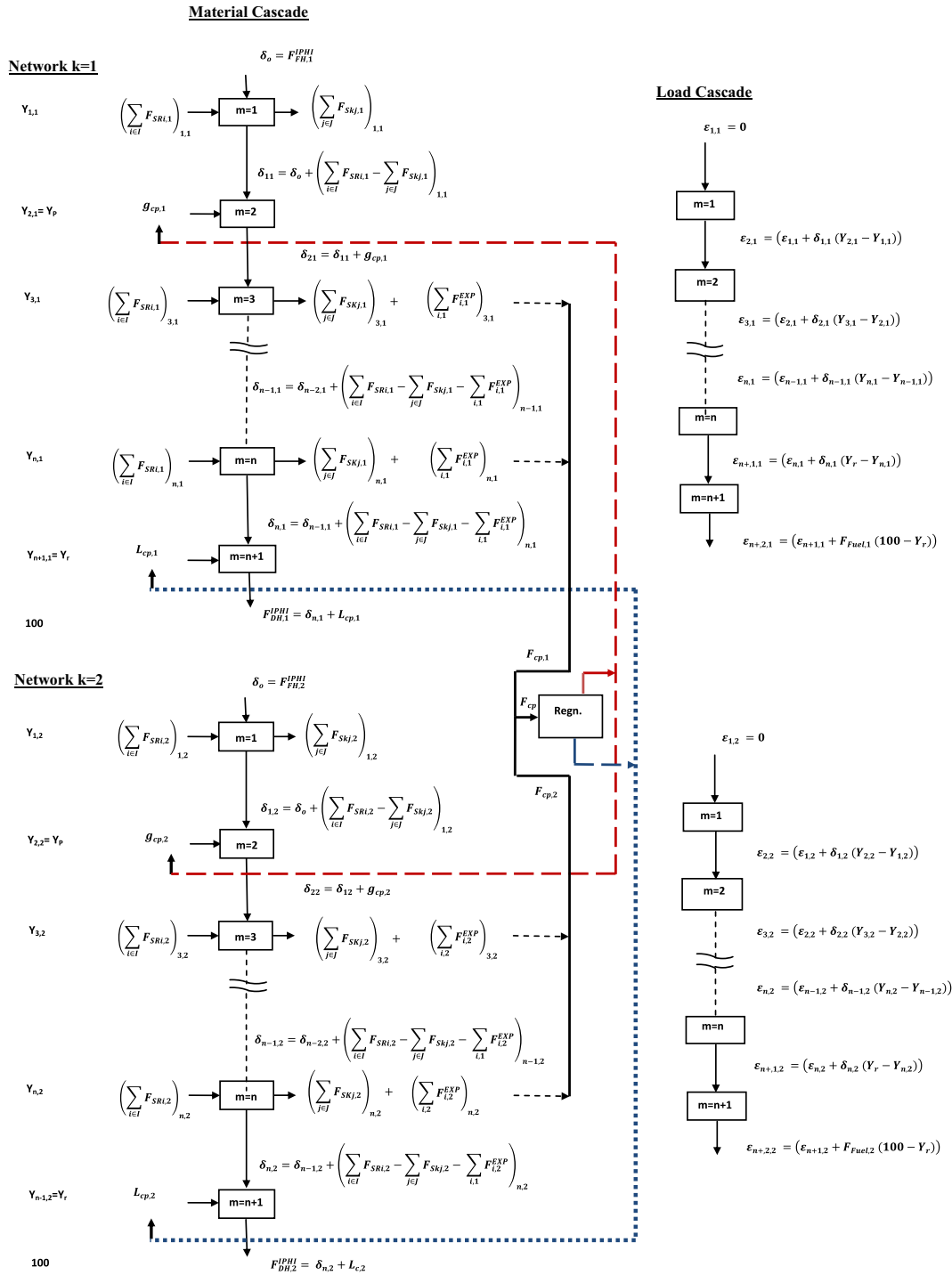


Figure 1 Impurity concentration cascade diagram for indirect inter-plant hydrogen integration.

inter-plant water integration system and inter-plant property-based integration system for single-pass regeneration system. However, this previous work did not consider the inter-plant hydrogen integration with partitioning regeneration system. In this work, the automated targeting is extended to inter-plant hydrogen integration system since, the indirect inter-plant hydrogen integration is based on the use of partitioning regenerator as a regenerator unit. The first step in the automated targeting technique for IPHI is the construction of the

hydrogen conservation cascade diagram as presented in Fig. 1. In the cascade diagram, a total of n sink/source concentrations (Y_m) for each hydrogen network k are arranged in an ascending order, starting from the lowest ($m = 1$) to the highest ($m = n$). Another concentration level is added for fresh hydrogen source, if it does not exist among the process sinks and sources. When the fresh hydrogen source is pure, zero concentration level is added as a first level and if the fresh hydrogen is impure, it will be located at its respective concentration

level as any source. Also, another two concentration levels for top (Y_p) and bottom product impurity concentration (Y_r) of the regenerator are added. A final concentration level (100%) is added to calculate the residue impurity load [9,18].

The second step in the concentration cascade diagram is the building of the material cascade. It is performed across all concentration intervals for each hydrogen network. At each concentration level m , the flowrate balance takes into account the sinks and the sources flowrates and the flowrates to and from the partitioning regeneration unit. The inter-plant flowrate that is sent from each source i to the regenerator is called export flowrate $F_{i,k}^{\text{EXP}}$, and is put as a sink at its source concentration level on the right of the material cascade. On the other hand, the inter-plant flowrates that are received from the regenerator unit to each network k are called the import flowrates, $g_{\text{cp},k}$, $L_{\text{cp},k}$, and they are added as sources at their concentration levels (Y_p , Y_r) on the left of the material cascade. Note that, Y_p and Y_r impurity concentration levels are the top and the bottom product concentrations of the partitioning regeneration unit. As described in Eq. (1), the net material flowrate from level m for network k ($\delta_{m,k}$), is the summation of the net material flowrate cascaded from the earlier concentration level ($\delta_{m-1,k}$) with the net flowrates at concentration level m ($\sum_{i \in I} F_{\text{SR},i,k} - \sum_{j \in J} F_{\text{SK},j,k} - \sum_{i,k} F_{i,k}^{\text{EXP}}$). The fresh inter-plant hydrogen integration for each network $F_{\text{FH},k}^{\text{IPHI}}$ is added at the first concentration level ($\delta_o = F_{\text{FH},k}^{\text{IPHI}}$) when the fresh hydrogen is pure and if the fresh hydrogen is impure, it will be located at its concentration level as any source in the individual network. The inter-plant hydrogen discharge $F_{\text{DH},k}^{\text{IPHI}}$, is located at the last level of the material cascade in each network as shown in Fig. 1.

$$\delta_{m,k} = \delta_{m-1,k} + \left(\sum_{i \in I} F_{\text{SR},i,k} - \sum_{j \in J} F_{\text{SK},j,k} - \sum_{i,k} F_{i,k}^{\text{EXP}} \right)_m$$

$$k = 1, 2, 3, \dots, K_{\text{networks}} \quad (1)$$

The third step in the concentration cascade diagram is the building of the load cascade. The residual contaminant load at each concentration level m ($\varepsilon_{m,k}$) for each network k as described in Eq. (2), is composed of two parts. The first part is the residual contaminant load from the earlier level ($\varepsilon_{m-1,k}$) and the second part is the contaminant impurity load since, the contaminant impurity load is the net material flowrate at level $m-1$ ($\delta_{m-1,k}$) and the difference between the concentration levels Y_{m-1} and Y_m [9,28].

$$\varepsilon_{m,k} = \varepsilon_{m-1,k} + \delta_{m-1,k} (Y_m - Y_{m-1}) \quad \nabla k \in K, \quad \forall m \in M_{\text{levels}} \quad (2)$$

Note that, the net material flowrate (δ_m) may take a positive or negative sign. The positive value is achieved when the material flowrate is from the lower concentration level to the higher concentration level and the negative value is achieved when the material flowrate is from the higher to the lower concentration level [9,18].

Note also that, the residual contaminant ($\varepsilon_{m,k}$) must take positive value so, Eq. (3) is taken into consideration. Of the advantages of the automated targeting technique is that it combined the pinch analysis technique and the mathematical technique [30] since, the pinch point is determined at the residual contaminant load ($\varepsilon_{m,k}$) equal to zero [9].

$$\varepsilon_{m,k} \geq 0, \quad \forall k \in K, \quad \forall m \in M \quad (3)$$

The objective function considered in this work is the minimization of a total annual cost (TAC) that includes operation cost and annualized capital costs. The operation cost depends on the fresh hydrogen cost while the capital cost is the regenerator and the pipes costs as described in Eq. (4).

$$\text{TAC} = \left[\sum_{k \in K} F_{\text{FH},k}^{\text{IPHI}} \cdot W_{\text{cost},k} \right] \text{AWH} + [(I_{\text{regenerator}} + I_{\text{pipe}})] \text{AF} \quad (4)$$

where $F_{\text{FH},k}^{\text{IPHI}}$ and $W_{\text{cost},k}$ represent the indirect inter-plant hydrogen fresh flow rate to each network and the unit cost of each fresh hydrogen, respectively. The fresh hydrogen in each network is placed at its impurity concentration in the cascade diagram. AWH is the annual working hours per year.

The pressure swing adsorption (PSA) and the membrane separation are commonly used as regenerators in the regeneration of hydrogen networks [18]. The capital cost of the PSA regenerator (I_{psa}) can be calculated using Eq. (5) [33].

$$I_{\text{psa}} = [503,800 + (347,400 F_{\text{cp}})] \quad (5)$$

where F_{cp} is the inlet flowrate of the regenerator and measured in MMSCFd. When membrane separation is used as a regenerator, the capital cost of the regenerator (I_{MEM}) can be calculated using Eqs. (6) and (7) [15,33].

$$A_{\text{MEM}} = \frac{F_{\text{cp}}}{\text{Lg}_{\text{H}_2} \cdot [P_n^{\text{feed}} \cdot y_{\text{H}_2 \text{ in feed}} - P_n^{\text{product}} \cdot y_{\text{H}_2 \text{ in product}}]} \quad (6)$$

$$I_{\text{MEM}} = [50,000 + 400 \cdot A_{\text{MEM}}] \quad (7)$$

where A_{MEM} is the membrane area, Lg_{H_2} represents the permeability of hydrogen and measured in GPU ($\text{cm}^3(\text{STD}) * 10^{-6} / \text{cm}^2 \text{ s cmHg}$), P_n^{feed} and P_n^{product} are the inlet pressure and the top product pressure of the regenerator, and $y_{\text{H}_2 \text{ in feed}}$ and $y_{\text{H}_2 \text{ in product}}$ are the hydrogen purity in the inlet feed and the top product of the regenerator respectively.

The pipelines from the sources of the networks to the regenerator and from the regenerator to sinks of the networks can be calculated using Eq. (8) [15].

$$I_{\text{pipe}} = D \left[\left(420.74 X_{\text{INDIR}}(k) + 1484.76 \frac{0.02352 F_{\text{cp},k}}{P} \right) + \left(420.74 N_{\text{INDIR}}(k) + 1484.76 \frac{0.02352 g_{\text{cp},k}}{P} \right) + \left(420.74 Z_{\text{INDIR}}(k) + 1484.76 \frac{0.02352 N_{\text{cp},k}}{P} \right) + \left(420.74 M_{\text{INDIR}}(k) + 1484.76 \frac{0.02352 F_{\text{DH},k}^{\text{IPHI}}}{P} \right) \right] \quad (8)$$

where $F_{\text{cp},k}$ is the summation of all export flowrates from sources of network k to the regenerator as described in Eq. (9), $g_{\text{cp},k}$ is the top flowrate of the regenerator sent to network k , and $N_{\text{cp},k}$ is the bottom flowrate of the regenerator ($L_{\text{cp},k}$) sent to network k as described in Eq. (10). $F_{\text{DH},k}^{\text{IPHI}}$ is the discharge hydrogen flowrate that is sent from network k to the fuel system. P is the max pressure of the pipelines. D is the length of the cross-plant pipeline. Note that, the pipelines cost within each individual network are assumed neglected, because their cost are very small when compared to the other pipelines

Table 1 Hydrogen processes data for case study 1.

	Hydrogen sources	Flow rates (MMSCFd)	Impurity (vol.%)	Hydrogen sinks (demands)	Flow rates (MMSCFd)	Impurity (vol.%)
Network 1	Outlet unit A (SR1)	350	9.0	Inlet unit A (SK1)	400	7.2
	Outlet unit B (SR2)	500	15.0	Inlet unit B (SK2)	600	12.4
	Outlet unit C (SR3)	223	25.0	Inlet unit C (SK3)	240	22.3
	Outlet unit D (SR4)	248	30.0	Inlet unit D (SK4)	270	24.6
Fresh hydrogen			To be determined			1.0
Network 2	CCR (SR5)	14.463	17.0	HCU(SK5)	152.26	8.689
	HCU (SR6)	75.635	18.28	NHT (SK6)	34.285	22.178
	NHT (SR7)	32.95	23.43	DHT (SK7)	181.01	24.792
	DHT (SR8)	168.723	28.62	CNHT (SK8)	40.008	30.304
	CNHT (SR9)	35.543	33.52			
Fresh hydrogen			To be determined			1.0

cost [28]. Binary variable $X_{\text{INDIR}}(k)$ indicates the existence of a cross-plant pipeline from all sources of network k to the regenerator [15,33]. $N_{\text{INDIR}}(k)$, and $Z_{\text{INDIR}}(k)$ indicate the existence of a cross-plant pipeline from the top and the bottom product of the regenerator to all sinks of each network. $M_{\text{INDIR}}(k)$ indicates the existence of a cross-plant pipeline from network k to the fuel discharge.

$$F_{\text{cp},k} = \sum_{i \in I} F_{i,k}^{\text{EXP}} \quad \forall k \in K \quad (9)$$

$$N_{\text{cp},k} = L_{\text{cp},k} - F_{\text{DH},k}^{\text{PHI}} \quad (10)$$

To annualize the capital cost an annualization factor (AF) is added as described in Eq. (11) [15].

$$\text{AF} = \frac{a \cdot (1 + a)^L}{(1 + a)^L - 1} \quad (11)$$

where a , and L are a fractional interest rate per year and the number of years, respectively. Eq. (12) is the overall material balance on the regenerator, and it states that the sum of all export hydrogen flowrates from sources of the participating

networks $\left(\sum_{k \in K} \sum_{i \in I} F_{i,k}^{\text{EXP}}\right)$ to the regenerator must equal the sum of all hydrogen flowrates from the top product $\left(\sum_{k \in K} g_{\text{cp},k}\right)$ and all hydrogen flowrates from the bottom product $\left(\sum_{k \in K} L_{\text{cp},k}\right)$ of the regenerator.

$$\sum_{k \in K} \sum_{i \in I} F_{i,k}^{\text{EXP}} = \sum_{k \in K} g_{\text{cp},k} + \sum_{k \in K} L_{\text{cp},k} \quad (12)$$

As presented in Fig. 1 all export flowrates from the participating hydrogen networks $\left(\sum_{k \in K} F_{\text{cp},k}\right)$ are mixed in a single export cross-plant pipeline (F_{cp}) before sending them to the regenerator as described in Eq. (13), on the other hand, hydrogen flowrate from the top product of the regenerator (g_{cp}) is distributed to the hydrogen networks through cross-plant pipelines $\left(\sum_{k \in K} g_{\text{cp},k}\right)$. Also, the hydrogen flowrate from the bottom product of the regenerator (L_{cp}) is distributed to the hydrogen networks through cross-plant pipelines $\left(\sum_{k \in K} L_{\text{cp},k}\right)$ as described in Eqs. (14) and (15).

$$F_{\text{cp}} = \sum_{k \in K} F_{\text{cp},k} \quad (13)$$

Table 2 Individual regeneration results of the two networks for case study 1.

Regeneration results	Network 1	Network 2	Total
<i>Base case</i>			
Fresh hydrogen (MMSCFd)	278.13	109.6	387.73
Hydrogen discharge (MMSCFd)	9.13	29.351	118.481
<i>Regeneration by PSA</i>			
Fresh hydrogen (MMSCFd)	208.238	88.956	297.194
hydrogen discharge (MMSCFd)	19.238	8.707	27.945
Regenerated flowrate (MMSCFd)	52.084	21.708	73.792
<i>Regeneration by membrane</i>			
Fresh hydrogen (MMSCFd)	206.478	88.254	294.732
hydrogen discharge (MMSCFd)	17.478	8.005	25.483
Regenerated flowrate (MMSCFd)	54.376	22.705	77.081

Table 3 Indirect inter-plant hydrogen integration results for case study 1.

Cases	PSA	Membrane
Fresh hydrogen flow rate (MMSCFd) for the two networks	296.6393	294.4314
Hydrogen discharge flow rate (MMSCFd) for the two networks	27.3903	25.1824
Inlet feed flow rate (MMSCFd) to the regenerator	73.7800	77.0067
Inlet feed concentration (vol.%) to the regenerator	31.0337	30.9904
Bottom concentration (vol.%) of the regenerator	83.425	90.651
Top product of the regenerator (MMSCFd)	46.3897	51.8243
Bottom product of the regenerator (MMSCFd)	27.3903	25.1824
No. of cross-plant pipelines (N)	4.0	4.0
Capital cost of the regenerator (MU\$)	26.1349	0.6066
Capital cost of the pipelines (MU\$)	1.486	1.514
Total capital cost (MU\$)	27.6209	2.1206

$$g_{cp} = \sum_{k \in K} g_{cp,k} \tag{14}$$

The overall impurity balance on the regenerator is given by Eq. (16).

$$L_{cp} = \sum_{k \in K} L_{cp,k} \tag{15}$$

$$\sum_{k \in K} \sum_{i \in I} F_{i,k}^{EXP} Y_i = \sum_{k \in K} g_{cp,k} Y_p + \sum_{k \in K} L_{cp,k} Y_r \tag{16}$$

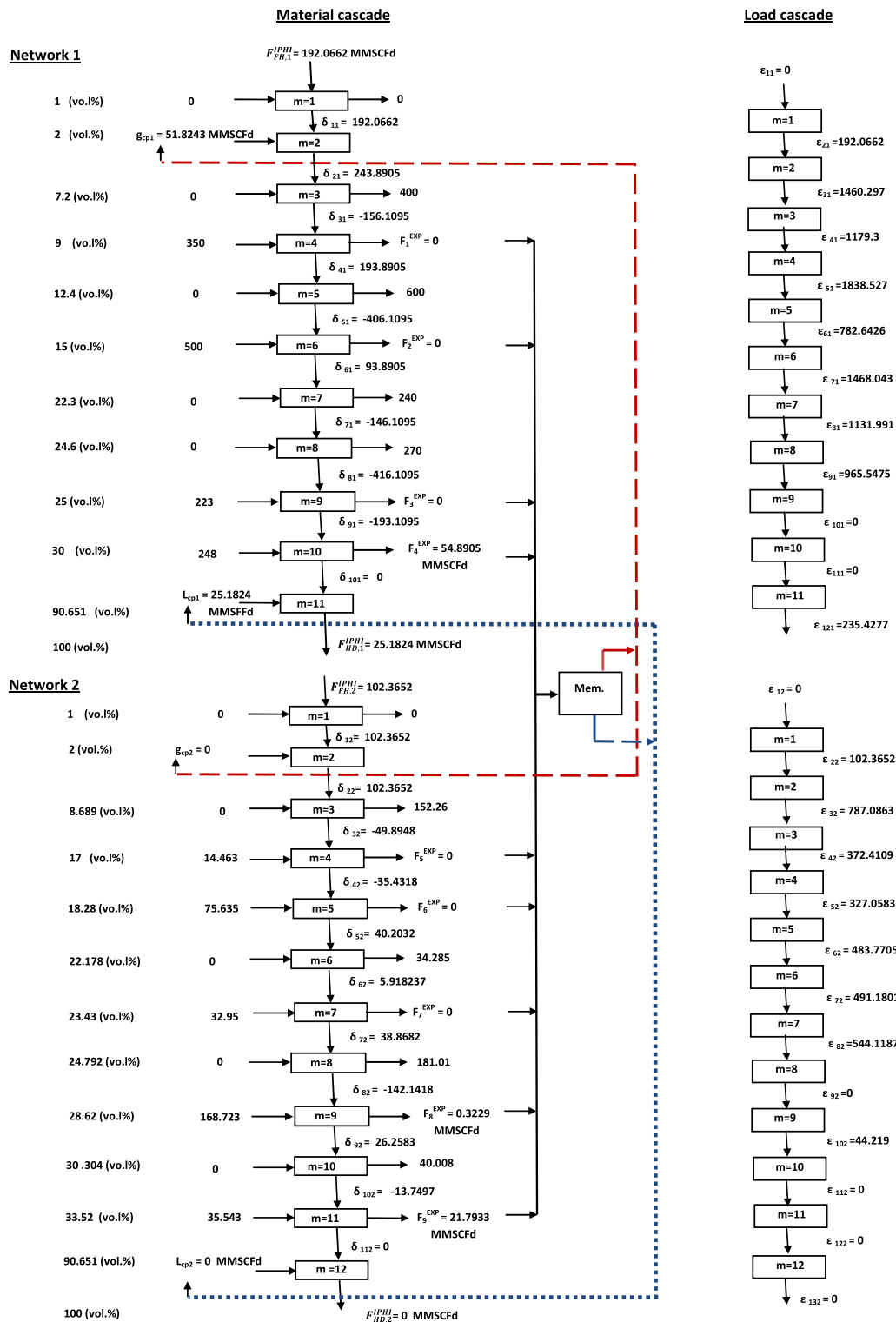


Figure 2 Impurity concentration cascade diagram for network 1 and network 2 with the indirect inter-plant hydrogen integration for case study 1 using membrane.

Table 4 Saving in the fresh hydrogen and hydrogen discharge for case study 1.

Cases	Fresh hydrogen (MMSCFd)	Hydrogen discharge (MMSCFd)
Base case	387.73	118.481
<i>For PSA</i>		
Inter-plant integration	296.6393	27.3903
Saving	91.091	91.091
Saving percentage %	23.49	76.88
<i>For membrane</i>		
Inter-plant integration	294.4314	25.1824
saving	93.299	93.299
Saving percentage %	24.06	78.75

where Y_i is the impurity concentration of each source i sent to the partitioning regenerator. Also, Eqs. (17) and (18) are added as a material and impurity balance on the regenerator.

$$F_{cp} = g_{cp} + L_{cp} \quad (17)$$

$$F_{cp} Y_F = g_{cp} Y_P + L_{cp} Y_r \quad (18)$$

where Y_F represents the impurity concentration of the feed to the regenerator. The upper bound of the export flowrate sent from any source i in network k ($F_{i,k}^{EXP}$) to the regenerator is taken as its source flowrate ($F_{SRi,k}$) as described in Eq. (19).

$$0 \leq F_{i,k}^{EXP} \leq F_{SRi,k} \quad \forall i \in I, \quad \forall k \in K \quad (19)$$

The upper (UB_{cp}) and lower bounds (LB_{cp}) of the cross-plant flowrates from each network to the regenerator ($F_{cp,k}$), from top product of the regenerator to each hydrogen network ($g_{cp,k}$), from bottom product of the regenerator to each network ($N_{cp,k}$), and from each network to the fuel discharge

($F_{DH,k}^{IPHI}$) are used as constraints as described in Eqs. (20)–(23). The existence of the cross-plant pipelines are indicated by the binary variables $X_{INDIR,k}$, $N_{INDIR,k}$, $Z_{INDIR,k}$ and $M_{INDIR,k}$. Note that, the total number of cross-plant pipelines are limited to N in Eq. (24). Note also that the binary variables in Eqs. (20)–(23) produced a MINLP formulation.

$$LB_{cp} X_{INDIR,k} \leq F_{cp,k} \leq UB_{cp} X_{INDIR,k} \quad (20)$$

$$LB_{cp} N_{INDIR,k} \leq g_{cp,k} \leq UB_{cp} N_{INDIR,k} \quad (21)$$

$$LB_{cp} Z_{INDIR,k} \leq N_{cp,k} \leq UB_{cp} Z_{INDIR,k} \quad (22)$$

$$LB_{cp} M_{INDIR,k} \leq F_{DH,k}^{IPHI} \leq UB_{cp} M_{INDIR,k} \quad (23)$$

$$\sum_{k \in K} X_{INDIR,k} + \sum_{k \in K} N_{INDIR,k} + \sum_{k \in K} Z_{INDIR,k} + \sum_{k \in K} M_{INDIR,k} \leq N \quad (24)$$

For non-negativity of fresh hydrogen Eq. (25) is added to the model.

$$F_{FH,k}^{FIPHI} \geq 0 \quad (25)$$

The impurity concentration of the top product of the regenerator (Y_p) is a fixed characteristic of the regeneration system in hydrogen network [18], but the impurity concentration of the bottom product (Y_r) is unknown and set to be between a minimum ($Y_{r,i}^{\min}$) and a maximum ($Y_{r,i}^{\max}$) value as presented in Eq. (26).

$$Y_{r,i}^{\min} \leq Y_r \leq Y_{r,i}^{\max} \quad (26)$$

The minimum and the maximum value of the impurity concentration of the bottom product of the regenerator are the lowest and the highest values of the impurity concentrations ($Y_{r,i}$) of all sources of the participating networks when they are regenerated individually without inter-plant hydrogen integration as described in Eq. (27).

Table 5 Indirect inter-plant hydrogen integration costs for case study 1.

Cases	Operation cost (MUS\$/yr)	Capital cost (MUS\$/yr)	Total annual cost (MUS\$/yr)
(Base case)	258.487	—	258.487
<i>For PSA</i>			
Inter-plant integration	197.759	27.6209*0.231	204.1399
Operating cost saving	60.728		
Saving cost percentage %	23.49		
PSA cost (MUS\$)	26.1349		
Pipe cost (MUS\$)	1.486		
Capital cost (MUS\$)	27.6209		
Payback period (months)	5.5		
<i>For Membrane</i>			
Inter-plant integration	196.288	0.4899	196.7778
Operating cost saving	62.199		
Saving cost percentage %	24.06		
Membrane cost (MUS\$)	0.6066		
Pipe cost (MUS\$)	1.514		
Capital cost (MUS\$)	2.1206		
Payback period (day)	13		

$$Y_{r,i} = \frac{\left[Y_i - \frac{(100-Y_i)}{(100-Y_p)} \cdot \alpha_L \cdot Y_p \right]}{\left[1 - \frac{(100-Y_i)}{(100-Y_p)} \cdot \alpha_L \right]} \quad (27)$$

where Y_i and Y_p are the impurity concentrations of each source i and the impurity concentration of the top product of the regenerator. α_L is the hydrogen recovery [18].

4. Case studies

Two case studies are solved to illustrate the indirect inter-plant hydrogen integration model by conservation cascade diagram. The first case study illustrates the indirect inter-plant hydrogen integration when the two networks have the same fresh

hydrogen impurity source. The second case study illustrates the application of the proposed model when the two networks have different fresh hydrogen impurity source.

4.1. Case study 1

This case under investigation involves two hydrogen networks with the same fresh hydrogen impurity. The first network is taken from Liu [33] and the other network is taken from Jia [34]. The limiting hydrogen data for the two networks are tabulated in Table 1. The fresh hydrogen and the hydrogen discharge for network 1 are 278.13 MMSCFd (1% impurity), and 89.13 MMSCFd (18.1% impurity), respectively. The fresh

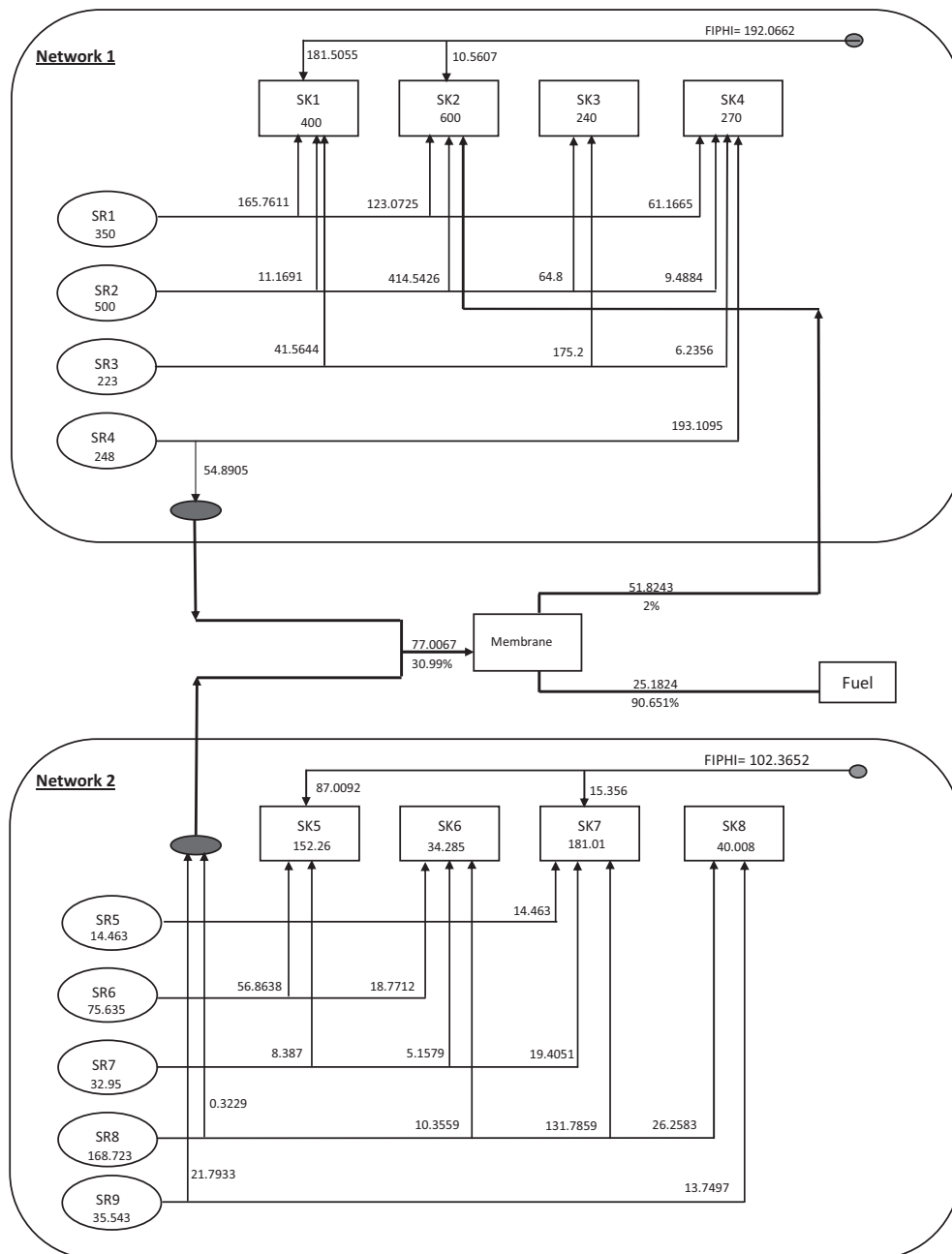


Figure 3 Optimized indirect inter-plant hydrogen integration design for case study 1 (flowrate is given in MMSCFd).

Table 6 Hydrogen processes data for case study 2.

	Hydrogen sources	Flow rates (MMSCFd)	Impurity (vol.%)	Hydrogen sinks (demands)	Flow rates (MMSCFd)	Impurity (vol.%)
Network 1	CCR (SR1)	14.463	17.0	HCU (SK1)	152.26	8.689
	HCU (SR2)	75.635	18.28	NHT (SK2)	34.285	22.178
	NHT (SR3)	32.95	23.43	DHT (SK3)	181.01	24.792
	DHT (SR4)	168.723	28.62	CNHT (SK4)	40.008	30.304
	CNHT (SR5)	35.543	33.52			
Fresh hydrogen			To be determined			1.0
Network 2	CCR (SR6)	23.5	25.0	HC (SK5)	124.48	19.7
	CNHT (SR7)	40.22	25.0	CNHT (SK6)	44.96	22.89
	HC (SR8)	96.99	25.0	DHT (SK7)	12.87	24.75
	DHT (SR9)	10.17	30.0	ISO (SK8)	0.04	25.0
	JHT (SR10)	7.92	35.0	JHT (SK9)	12.25	27.94
	NHT (SR11)	10.14	40.0	NHT (SK10)	15.67	31.181
Fresh hydrogen			To be determined			8.0

hydrogen and hydrogen discharge for the second network are 109.6 MMSCFd (1% impurity), and 29.351 MMSCFd (25.45% impurity), respectively as described in the base case without any hydrogen integration. It is required to synthesize indirect inter-plant hydrogen integration for the two networks. The cost of fresh hydrogen is taken as US\$ 2000/MMSCF [15,33]. 8000 h/yr is taken as the operating hours per year. The lower and maximum bounds of the cross-plant pipelines flowrate are taken as 5 MMSCFd and 500 MMSCFd, respectively.

The regeneration for the two hydrogen networks is achieved through a pressure-swing adsorption (PSA) or a membrane. The PSA product impurity Y_P is specified as 0.1% and with recovery $\alpha_L = 90\%$. For the membrane, the product impurity Y_P is specified as 2% and with recovery $\alpha_L = 95\%$ [18,35]. The hydrogen permeability of the membrane is set as 500 GPU that is equal to 125 times of the impurity permeability. The inlet and product pressure of the membrane are assumed to be 11.61 and 3.24 MPa, respectively. The capital cost is annualized for five years, with 5% interest rate. The piping distances from the regenerator to the sinks and the sources are assumed to be 500 m at high pressure of 4 MPa [33].

LINGO optimization software version 12 is used to solve the model presented by Ng et al. [18] for individual regeneration. Table 2 illustrates the minimum fresh hydrogen and hydrogen discharge flowrates for each network in the base case and when each network is regenerated individually.

Note that, the capital cost of the regenerator was not covered in the previous automated targeting presented for individual regeneration of hydrogen network [18]. In this work the total annual cost of the inter-plant integration is calculated.

The minimum and maximum impurity concentrations of the bottom product of the regenerator (Y_r) are calculated to be 49.495 vol.%, and 83.425 vol.%, respectively in case of using the PSA regenerator using Eq. (27). For indirect inter-plant hydrogen integration, solving the objective function in Eq. (4) subjected to the constraints Eqs. (1)–(3), (5), and (8)–(26) for PSA yields the minimum fresh hydrogen and hydrogen discharge for the two networks of 296.6393 and 27.3903

MMSCFd, respectively as described in Table 3 (column b). On the other hand when the membrane separation is used as a regenerator the minimum and maximum impurity concentrations of the bottom product of the regenerator (Y_r) are calculated to be 61.394 vol.%, and 90.651 vol.%, respectively. The proposed model is resolved. The constraints in this case are Eqs. (1)–(3), and (6)–(26). The optimal solution as shown in Table 3 (column c) shows that, the fresh hydrogen and hydrogen discharge for the two networks are 294.4314 and 25.1825 MMSCFd, respectively.

It is noted also from Table 3 that the total capital cost in case of using the membrane as a regenerator (US\$ 2.1206 million) is very small when compared with the capital cost of the PSA (US\$ 27.6209 million). Fig. 2 is the cascade indirect inter-plant diagram for networks 1 and 2 using the membrane as a regenerator. As shown in this figure, 54.8905 MMSCFd from source 4 in network 1 and 0.3229 MMSCFd from source 8 and 21.7933 MMSCFd from source 9 in network 2 are sent as export flowrates to the regenerator. Also, the import flowrates that are sent from the top product of the regenerator to the two networks are 51.8243 MMSCFd for network 1 and zero for network 2. On the other hand, the import flowrates that are sent from the bottom product of the regenerator to the networks are 25.1824 MMSCFd for network 1 and zero for network 2. All the bottom product of the regenerator that is sent to the first network is discharged as a fuel. The pinch point for each network was determined at residual contaminant load equal to zero.

As illustrated in Table 4 the saving in total fresh hydrogen flowrate for the two networks is 91.091 MMSCFd for the PSA regenerator with a saving percentage of 23.49% and 93.299 MMSCFd for the membrane regenerator with a saving percentage of 24.06%. Also, the saving amount of 91.091 MMSCFd is achieved for the hydrogen discharge for the two networks with saving percentage of 76.88% when PSA is used as a regenerator and 93.299 MMSCFd with saving percentage of 78.75% is achieved for the two networks when membrane separation is used as a regenerator.

Table 5 shows that, US\$ 60.728 million per year operation cost saving with saving percentage equal to 23.49% was

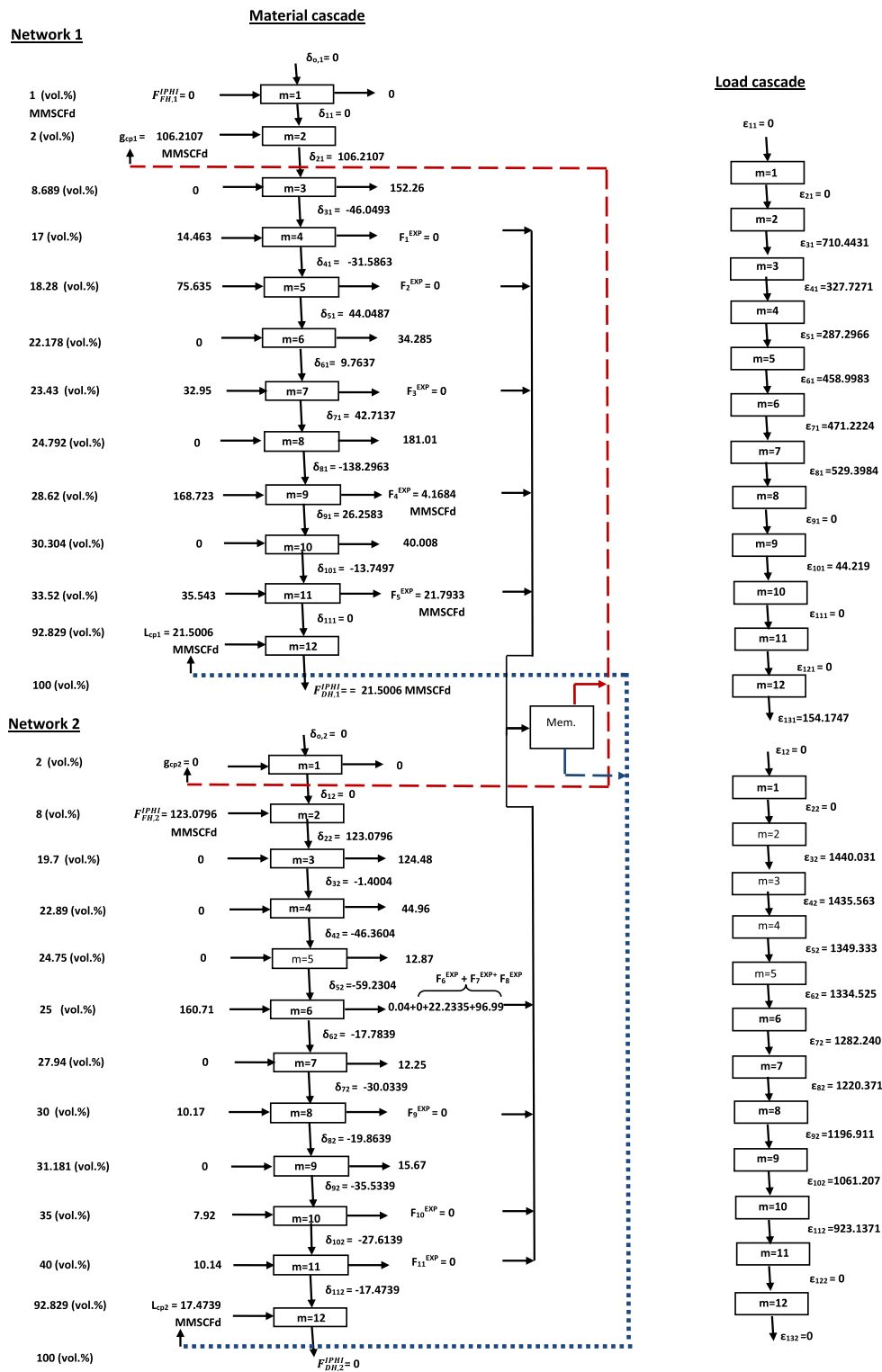


Figure 4 Impurity concentration cascade diagram for network 1 and network 2 with indirect inter-plant hydrogen integration for case study 2 using membrane.

achieved in the case of using the PSA as a regenerator. But in the case of using the membrane separation as a regenerator the saving in operation cost was US\$ 62.199 million per year with 24.06% saving percentage. Table 5 shows also that the total annual cost is calculated to be US\$ 196.7778 million per year

in case of using the membrane separation and this is lower than its value in the case of using the PSA as a regenerator (US\$ 204.1399 million per year). The increase in the total annual cost in the case of the PSA is due to the high capital cost of the PSA.

Table 7 Individual regeneration results of the two networks for case study 2.

Regeneration results	Network 1	Network 2	Total
<i>Base case</i>			
Fresh hydrogen (MMSCFd)	109.6	45	154.6
Hydrogen discharge (MMSCFd)	29.351	34.24	63.591
<i>Regeneration by PSA</i>			
Fresh hydrogen (MMSCFd)	88.95558	28.30452	117.2601
Hydrogen discharge (MMSCFd)	8.706576	6.974516	15.68109
Regenerated flowrate (MMSCFd)	21.70777	19.21649	40.92426
<i>Regeneration by membrane</i>			
Fresh hydrogen (MMSCFd)	88.25378	27.79098	116.04476
Hydrogen discharge (MMSCFd)	8.004781	6.460979	14.46576
Regenerated flowrate (MMSCFd)	22.70538	19.89399	42.59937

Table 8 Indirect-Inter plant hydrogen integration results for case study 2.

Cases	PSA	Membrane
Fresh hydrogen flow rate (MMSCFd)	0.0 + 124.6832	0.0 + 123.0796
Hydrogen discharge flow rate (MMSCFd)	23.1043	21.5006
Inlet feed flow rate (MMSCFd) to the regenerator	142.2406	145.1852
Inlet feed concentration (vol.%) to the regenerator (Y_F)	26.16	26.38
Residue concentration (vol.%) of the regenerator	86.941	92.829
Top product of the regenerator (MMSCFd)	99.56120	106.2107
Bottom product of the regenerator (MMSCFd)	42.6794	38.9745
No. of cross-plant pipelines (N)	6	6
Capital cost of the regenerator (MUSS)	49.9182	0.9948
Capital cost of the pipelines (MUSS)	2.2937	2.3194
Total capital cost (MUSS)	52.2119	3.3132

Table 9 Saving in the fresh hydrogen and hydrogen discharge for case study 2.

Cases	Fresh hydrogen (MMSCFd)	Hydrogen discharge (MMSCFd)
<i>Base case</i>		
	109.6 + 45.0	63.591
	154.6	
<i>For PSA</i>		
Inter-plant integration	0.0 + 124.6832	23.1043
Saving	29.917	40.487
Saving percentage %	19.35	63.67
<i>For Membrane</i>		
Inter-plant integration	0.0 + 123.0796	21.5006
saving	31.520	42.090
Saving percentage %	20.39	66.19

As a result of the above, the payback period which has been calculated as the ratio of the capital investment and the saving in the operation cost was very small (13 days) in case of the membrane separation when compared with its value in case of PSA (5.5 months). The corresponding indirect inter-plant hydrogen integration design for the two networks is shown in Fig. 3 with a total of four cross-plant pipelines.

Table 10 Indirect inter-plant hydrogen integration costs for case study 2.

Cases	Operation cost (MUSS/yr)	Capital cost (MUSS/yr)	Total annual cost (MUSS/yr)
<i>Base case</i>			
	94.067	—	94.067
<i>For PSA</i>			
Inter-plant integration	58.1856	12.0609	70.2465
Operating cost saving	35.8814		
Saving cost percentage %	38.14		
PSA cost (MUSS)	49.9182		
Pipe cost (MUSS)	2.2937		
Capital cost (MUSS)	52.2119		
Payback period (Year)	1.46		
<i>For Membrane</i>			
Inter-plant integration	57.4372	0.7655	58.2027
Operating cost saving	36.6298		
Saving cost percentage %	38.94		
Membrane cost (MUSS)	0.9948		
Pipe cost (MUSS)	2.3194		
Capital cost (MUSS)	3.314		
Payback period (day)	33		

4.2. Case study 2

This case study is represented to illustrate the indirect inter-plant hydrogen integration when the fresh hydrogen of the participating networks has different fresh hydrogen impurity concentration. It involves two hydrogen networks. The first network is taken from Jia [34] and the other one from Liu [33]. The limiting hydrogen data for the two networks are tabulated in Table 6. The fresh hydrogen and the fuel discharge for network 1 are as described in case study 1. The fresh hydrogen and fuel discharge for the second network are 45.0 MMSCFd (8 vol.% impurity), and 34.24 MMSCFd (30.39 vol.% impurity), respectively as described in the base case without any hydrogen integration. It is required to synthesize indirect inter-plant hydrogen integration for the two networks. The unit cost of fresh hydrogen for the first network is taken as US\$ 2000/MMSCF [33] (fresh hydrogen 1 vol.% impurity)

and for the second network it is assumed to be US\$ 1400/MMSCF (fresh hydrogen 8 vol.% impurity). The lower bound of the cross-plant flowrate is taken as 0.04 MMSCFd and the upper bound of the cross-plant flowrate is taken as 500 MMSCFd. The properties of the regeneration units are taken as the previous case study.

It is noted that the two networks have different impurity concentration of fresh hydrogen. The first network has fresh hydrogen with 1 vol.% impurity concentration and the other network has fresh hydrogen with 8 vol.% impurity concentration. Each fresh hydrogen is located at its impurity concentration level as shown in Fig. 4. Table 7 summarizes the minimum fresh hydrogen and hydrogen discharge flowrates for the two networks when each network is regenerated individually. For inter-plant integration when the PSA is used as a regenerator the minimum and maximum impurity concentrations of the bottom product of the regenerator (Y_r) are calculated to be

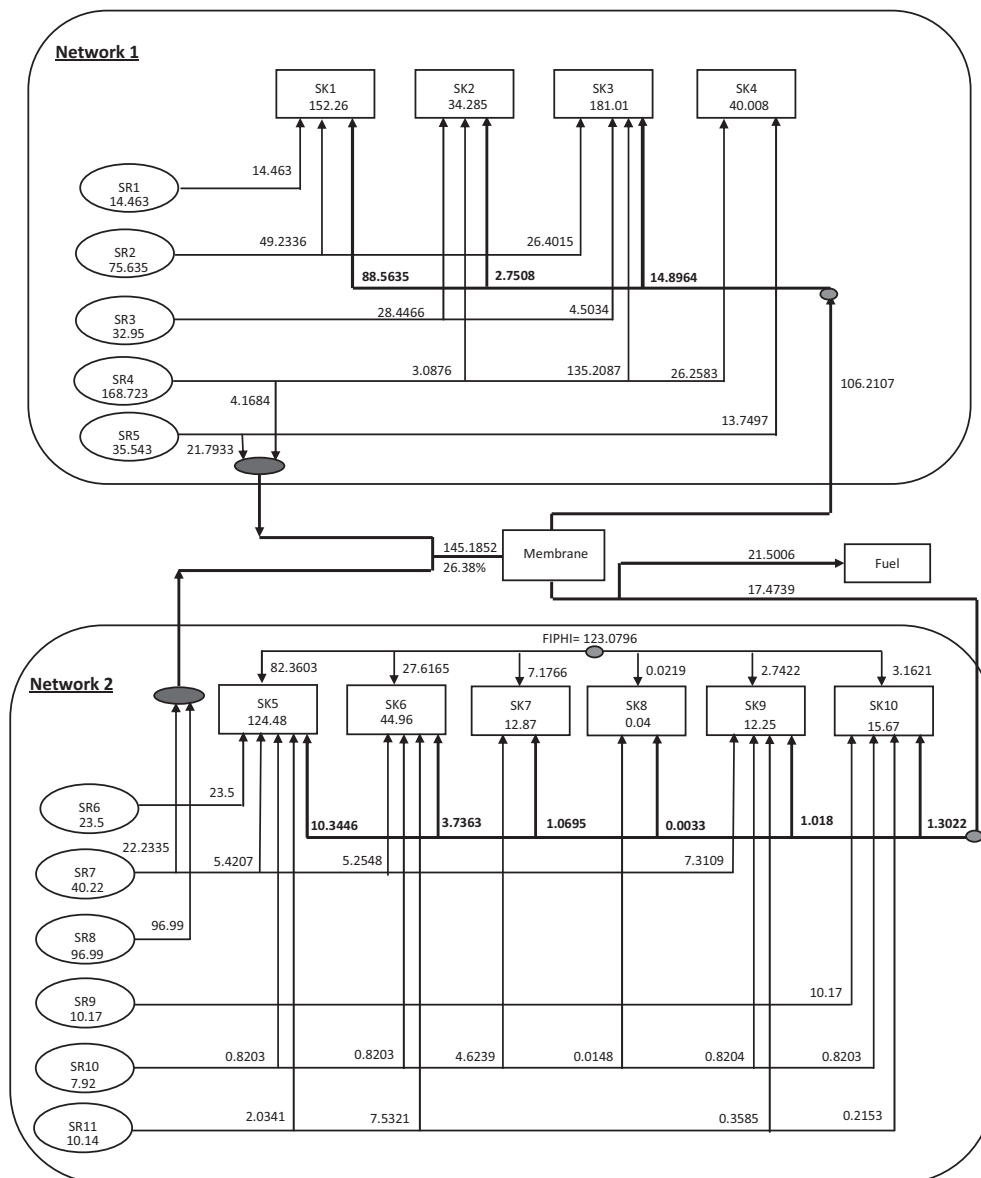


Figure 5 Optimized indirect inter plant hydrogen integration design for case study 2 (flowrate is given in MMSCFd).

67.096 vol.%, and 86.941 vol.%, respectively according to Eq. (27). The MINLP proposed model is solved using the objective function in Eq. (4) to minimize the total annual cost. The results showed that, the minimum fresh hydrogen and hydrogen discharge for the two networks of 124.6832 and 23.1043 MMSCFd, respectively as described in Table 8 (column b) are achieved. On the other hand when the membrane separation is used as a regenerator the minimum and maximum impurity concentrations of the bottom product of the regenerator (Y_r) are calculated to be 78.762 vol.%, and 92.829 vol.%, respectively. The proposed model is resolved. The optimal solution as shown in Table 8 (column c) shows that, the fresh hydrogen and hydrogen discharge for the two networks are 123.0796 and 21.5006 MMSCFd, respectively. The total capital cost in case of using the membrane as a regenerator (US\$ 3.3132 million) is very small when compared with the capital cost of the PSA (US\$ 52.2119 million) as concluded in case study 1. The cascade indirect inter-plant diagram for networks 1 and 2 using the membrane as a regenerator is shown in Fig. 4. It is noted that 4.1684 MMSCFd from source 4 and 21.7933 MMSCFd from source 5 in network 1 and 22.2335 MMSCFd from source 7 and 96.99 MMSCFd from source 8 in network 2 are sent as export flowrates to the regenerator. On the other hand, the import flowrates that are sent from the top product of the regenerator to the two networks are 106.2107 MMSCFd for network 1 and zero for network 2. Also, the import flowrates that are sent from the bottom product of the regenerator to the networks are 21.5006 MMSCFd for network 1 and 17.4739 MMSCFd for network 2. The bottom product of the regenerator that is sent to the first network is discharged as a fuel and the bottom product that is sent to the second network is reused/recycled in the network.

As illustrated in Table 9 the saving in total fresh hydrogen flowrate for the two networks is 29.917 MMSCFd for the PSA regenerator with a saving percentage of 19.35% and 31.52 MMSCFd for the membrane regenerator with a saving percentage of 20.39%. Also, the saving amount of 40.487 MMSCFd is achieved for the hydrogen discharge for the two networks with saving percentage of 63.67% when PSA is used as a regenerator and 42.090 MMSCFd with saving percentage of 66.19% is achieved for the two networks when membrane separation is used as a regenerator.

It is noted that all the inter-plant fresh hydrogen is taken from the second fresh hydrogen of network 2 which has lower price (US\$ 1400/year) than the other fresh hydrogen of network 1 (US\$ 2000/year). So, a high operation cost saving is achieved as illustrated in Table 10. An operation cost saving of US\$ 35.8814 million/year with a saving percentage equal to 38.14% was achieved in case of using the PSA as a regenerator and the saving in operation cost in case of using the membrane separation was US\$ 36.6298 million/year with 38.94% saving percentage. Table 10 shows also that the total annual cost is calculated to be US\$ 70.2465 million per year in case of using the PSA and in case of membrane separation it is US\$ 58.2027 million per year. The previous results lead to payback period of 33 days in case of the membrane separation and 1.46 year for PSA. The corresponding indirect inter-plant hydrogen integration design for the two networks is shown in Fig. 5 with a total of five cross plant pipelines that achieve the targeted total minimum cost.

5. Conclusion

The present study presents the automated targeting for indirect inter-plant hydrogen integration. A proposed model is presented for indirect inter-plant hydrogen integration based on cascade analysis approach in which two types of regenerator units, pressure swing adsorption or membrane are inserted as a part of the overall system. The model is applied to determine the minimum fresh hydrogen consumption and fuel hydrogen discharge for IPHI. In addition, the calculation of total capital cost of the regenerator and the pipelines. Two case studies are studied. In the first case study, the two participating networks have the same fresh hydrogen impurity concentration (1 vol.% impurity concentration) and in the other case study the two networks have different fresh hydrogen impurity concentration (1 vol.% and 8 vol.% impurity concentration). The conclusion and the results which can be withdrawn from the study are that: a significant reduction in the amount of fresh hydrogen required by the process and the hydrogen discharge in IPHI are achieved. Also, a higher reduction was achieved in the operation cost for the participating networks with indirect inter-plant hydrogen integration when compared with the base case in the two case studies, since US\$ 62.199 million per year is achieved for case study 1 and US\$ 36.6298 million per year for case study 2 when the membrane separation is used as a regenerator. All case studies solved showed that the membrane was the optimum regenerator for the participating networks due to its low capital cost comparing with the PSA capital cost.

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