





# A Multi-objective Optimization Approach for Integrating Isomerization Hydro-processing Unit with Hydrogen Network

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Abstract

# Article Info

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#### Introduction

Due to stringent environmental regulations calling for reduced use of aromatics and olefins in gasoline, naphtha isomerization has been shown to be a simple and cost-effective technology for producing clean gasoline components with high octane number [1]. The isomerization process is one of the hydrogen consumers in the refinery. Besides isomerization, there are other hydrogen consuming processes such as hydrocracking and hydrotreating. On the other hand, there are hydrogen producers such as catalytic reforming [2-7]. Catalytic reforming produces aromatic compounds from the recycling and dehydrogenation of hydrocarbon molecules and is used to increase the octane number of heavy naphtha. At the same time, large amounts of hydrogen are produced as a by-product. If the

Therefore, the disconnection between the hydrogen network and increasing the octane number in the isomerization unit will lead to a shortage of hydrogen and unmet demand for hydrogen. The present work is an integrated approach to the isomerization process in refineries and hydrogen network design. Modeling and optimization of the overall hydrogen network objective to minimize the total consumption of fresh hydrogen and maximize the isomerization octane number using a goal programming multi-objective optimization approach is presented. The model is introduced for two cases, reuse/recycling and hydrogen regeneration networks. The global network model is a multi-objective nonlinear programming model, and is optimized using Lingo optimization software version 14. The results showed that by integrating the isomerization model with the hydrogen reuse/recycling network, the fresh hydrogen saving was 20.871 kmol/h compared to 7.772 kmol/h in the case of neglecting the isomerization model. On the other hand, a significant reduction of 447.898 kmol/h in fresh hydrogen was achieved in the hydrogen regeneration network when including the isomerization model compared to that achieved when neglecting the isomerization model (343.261 kmol/h). In addition, the optimum operating conditions, feed specifications and hydrogen consumption of the isomerization unit that gives the maximum research octane number and the minimum fresh hydrogen were determined for both the reuse/recycling and hydrogen regeneration network cases.

The isomerization unit improves the research octane number of light naphtha with better

environmental impacts. Recent developments in increasing the octane number in the isomerization unit lead to an increase in the demand for hydrogen in the refining.

hydrogen produced by catalytic reforming is insufficient, additional hydrogen can be provided by constructing a hydrogen plant that produces hydrogen gas either by steam reforming or partial oxidation of hydrocarbons. Alternatively, hydrogen could be imported via pipeline. Finally, the off-gases from hydrogen consumers are also resources. They can be reused in other consumers, sent for fuel, or used as feed for hydrogen purifiers to recover hydrogen and increase the hydrogen purity required for the hydro-processing units. The most widely used processes for hydrogen purification are pressure swing adsorption, membrane separation, and cryogenic separation [8-11].

In recent years, refineries have been facing increasing demand for clean fuels. To meet the required end product specifications of crude oil, it has

become necessary to increase the use of hydrogen in hydro-processing units. But hydrogen production capacities are often limited and bottlenecks exist. To reduce these bottlenecks, an optimization should be applied to improve hydrogen consumption in the hydrogen network to achieve better and maximum hydrogen reuse.

There are two methods to optimize the hydrogen network, pinch analysis and mathematical programming optimization techniques [12].

The pinch analysis technique can be graphical or algebraic optimization. For the graphical optimization based on pinch technique, there are different aspects that are optimized for targeting and designing the hydrogen network. El-Halwagi et al. [13] presented a material recycling pinch diagram, Zhao et al. [14] introduced the impurity deficit diagram, and Saw et al. [15] proposed a material surplus composite curve. Other works on algebraic optimization-based pinch technique have been presented. Deng et al. [16] combined the design of a hydrogen network with valuable light hydrocarbons. Huang and Liu [17] optimized hydrogen separators in a hydrogen network. Gai et al. [18] developed a method that combines multiple-level resource pinch analysis with waste hydrogen regeneration to reduce the use of fresh hydrogen and the waste hydrogen discharged.

On the other hand, many studies based on mathematical programming have been reported in the literature. This technique relies on superstructure to formulate hydrogen optimization problems as mathematical models using computer programs [19]. Jagannath et al. [20] proposed an improved superstructure and a mixed integer nonlinear programming (MINLP) model to synthesize a hydrogen network with the minimum annual total cost. Deng et al. [21] developed a superstructure and a mathematical model to synthesize intermediate hydrogen headers in a hydrogen network. Other works have been conducted using mathematical programming to solve complex problems in hydrogen network optimization [22–32].

Previous optimization methods on the hydrogen network were mainly developed assuming constant isomerization performance with constant hydrogen consumption. Few works have addressed the integration between the hydrogen consumption of the hydro-processing unit and the hydrogen network. Mao et al. [33] presented the integration of vacuum gas oil (VGO) hydrocracking reaction and hydrogen network. They used a quantitative equation relating hydrogen consumption, sulphur content in the feed oil of the VGO reactor, and hydrogen entering the hydrocracking reactor. Umana et al. [34] presented a framework for evaluating the interactions between hydro-processing reactions and hydrogen distribution systems. The effect of changes in hydrogen partial pressure, hydrogen oil ratio, operating conditions, and hydrogen consumption on the required sulphur specifications of the hydrodesulphurization process and the overall hydrogen network performance were studied. Furthermore, the production of light hydrocarbons associated with hydrodesulphurization

reactions was incorporated into the hydrogen network model.

Isomerization often does not occur under ideal conditions, resulting in excessive amounts of hydrogen being used and allowing excess hydrogen and associated impurities to be sent either to the flare or for use as fuel. Additionally, the effect of changes in hydrogen consumption, operating conditions, and feed specifications on the required octane number and overall performance of the hydrogen network has not been considered in previous research.

The main objectives of this work were to integrate the isomerization hydro-processing model into reuse/recycling and into hydrogen network regeneration considering the effect of isomerization operating conditions, feed specifications, and hydrogen consumption on product research octane number.

# Methodology

This work aims to integrate the isomerization process model with the overall hydrogen network. Section 1 in this work is the general formulation of the goal programming multi-objective optimization model. Section 2 introduces the integration of the isomerization model with the hydrogen reuse/recycling network. This section is divided into four parts, the first part is the isomerization model based on the relationship between hydrogen consumption in the isomerization reactor and the research octane number of isomerate, the second part is the formulation of the hydrogen reuse/recycling network as described in the previous work, the third part is the integration of the isomerization model with the hydrogen reuse/recycling network when the isomerization model equations are used as new constraints for the objective function of the minimum fresh hydrogen, and the fourth part is the multi-objective optimization model for integrating the isomerization model with the hydrogen reuse/recycling network. Section 3 introduces the integration of the isomerization model with the hydrogen regeneration network. This section is divided into three parts, the first part is the formulation of the hydrogen regeneration network, the second part is the integration of the isomerization model with the hydrogen regeneration network when the isomerization model equations are used as new constraints for the objective function of the minimum fresh hydrogen and regeneration costs, and the third part is the multi-objective optimization model for integrating the isomerization model with the hydrogen regeneration network. For Sections 2 and 3, a case study is solved to illustrate the applicability of the approach.

#### Multi-objective optimization formulation using Goal programming technique

There are two ways to solve multi-objective optimization models; Mathematical programming methods and algorithms of approximation to the Pareto set. Scalarization and non-scalarization methods are a classification of mathematical programming methods. The scalarization methods require reformulating a single-objective model related to a multi-objective model through a scalar function [35, 36]. Goal programming, weighted sum method, and epsilon constraint method are the most widely used methods of mathematical programming [37-42].

In this research, the goal programming method was used for multi-objective optimization. The general formulation for goal programming multi-objective optimization is given in Equation (1) and subjects to constraints as shown in Equations (2) and (3) [37]. In this way the model seeks to minimize the overall deviation of targets

$$\min_{\mathbf{x},\delta^+,\delta^-} \sum_{i=1}^{p} (\delta_i^+ + \delta_i^-)$$
(1)

$$f_i(x) + \delta_i^+ - \delta_i^- = ai \quad \forall i = 1, ..., p$$
(2)

$$\delta_i^+, \delta_i^- \ge 0 \quad \forall i = 1, \dots, p \tag{3}$$

Where  $f_i(x)$  is the individual objective function and its number starts from i to P,  $\delta_i^+, \delta_i^-$  are the deviations of the objective function from the optimum goals (ai).

# Integration of isomerization model with reuse/recycling hydrogen network

The integration methodology applied to this work opens opportunities to exploit varying degrees of freedom that can improve the performance of the overall integrated optimization framework. This work presents the mathematical formulation and optimization of a hydrogen network for reuse/recycling and regeneration system under variable operating conditions, feed specifications, and hydrogen consumption constraints for an isomerization hydro-processing unit to meet the maximum product research octane number and minimum fresh hydrogen for the overall hydrogen network.

#### Isomerization unit model

The hydrogen requirement for the isomerization unit giving minimum fresh hydrogen consumption in optimizing the overall hydrogen network for reuse /recycling and regeneration may not be accurate when the maximum octane number required for isomerization is ignored.

This work aims to provide a mathematical optimization for evaluating the interactions between the isomerization hydro-processing process and the hydrogen distribution network to improve the overall performance of the hydrogen network. It also shows the effects of changes in the specifications of the light naphtha feed stream in the inlet feed to the isomerization reactor and changes in operating conditions such as temperature, liquid hourly space velocity, and hydrogen consumption of the isomerization unit on the inlet hydrogen flowrate required for the isomerization reactor (sink) and the hydrogen flowrate at the outlet (source) and thus the total hydrogen requirement of the hydrogen network.

Figure 1 shows the current non-recyclable isomerization process as presented by Shehata et al. [43]. The light liquid naphtha feed stream is mixed with a hydrogen-rich gas, heated and fed to the isomerization reactor. The reactor operates at the required hydrogen flowrate depending on the maximum octane number targets. Hydrogen is consumed by saturating benzene and naphthene to paraffin and then to iso-paraffin. The liquid flow from the reactor is cooled and directed to a highpressure flash separator. The gas released from the separator is purged into the fuel system.

Note that the existing isomerization unit does not have a recirculation loop as in other hydro-processing process units in the refinery such as hydrotreatment and hydrocracking. Off-gas from hydro-processes is either reused as external recirculation in the same unit or purged into the fuel system.



Figure 1 An extended isomerization model showing different interactions in the hydrogen network model

There are three main locations: the reactor inlet (Giso), the isomerization reactor, and the reactor outlet (W<sub>iso</sub>). The interconnection between these variables and their intermediate currents in the hydrogen network is achieved using the behavior of the system. Hence, hydrogen consumer models sufficiently detailed to capture important operating features of the system are required in the early stage of a refinery hydrogen network [20]. The methodology developed in this work proposes a general representation of the isomerization process model embedded in the hydrogen network superstructure to produce an integrated process and network configuration. There are three main challenges in this methodology: obtaining an isomerization model that can predict the optimal operational conditions of the process (reactor temperature, and liquid hourly space velocity), feed specifications (naphthenes and benzene wt.%), and hydrogen consumption giving maximum research octane number and minimum fresh hydrogen of the overall hydrogen network in refinery. The model of the isomerization process presented in Figure 1 was presented by Shehata et al. [43]. They presented the relationship between the product's research octane number (RON) and other variables; The weight percentage of naphthene (naph) and benzene (BZ) in the naphtha feed to the isomerization unit, temperature (T), liquid hourly space velocity (LHSV), and hydrogen consumption (HC) in the isomerization reactor as illustrated in Equation (4).

Isomerate RON

= -78.81947 - (5.7610 \* naph) + (19.16095 \* BZ)- (1.7566 \* Temp) - (11.92181 \* LHSV) + (6.82721 \* HC) + (0.21942 \* naph \* BZ) + (0.032018 \* naph \* Temp) + (0.19833 \* naph \* LHSV) - (0.21345 \* BZ \* HC) + (0.21577 \* Temp \* LHSV) - (0.050521 \* Temp \* HC) - (0.25395 \* LHSV \* HC) - (0.25974 \* BZ<sup>2</sup>) + (0.019167 \* Temp<sup>2</sup>) + (0.009074 \* HC<sup>2</sup>) (4)

Note that Equation (4) is presented based on operational data collected from an existing isomerization unit. It is applicable according to the range of operating conditions, feed specifications, hydrogen consumption, and research octane number of the isomerization process. Where the range of naphthene and benzene in light naphtha feed is 5.0-34 wt% and 0.31-3.03 wt% respectively, the reactor temperature range is  $156-170^{\circ}$ C, the LHSV range is 0.82-2.81 h<sup>-1</sup>, the hydrogen consumption range is 102.6-124 kmol/h, and the research octane number range is 86-88.6. This equation should not be used to determine the relative influence of each factor because the coefficients are scaled to accommodate units of each factor and the intercept is not at the center of the design space.

In this work, Equation (4) was included in the optimization and integration of the hydrogen network of the plant containing this isomerization unit in the case of reuse/recycling and in regeneration. The effect of changing the operating parameters (Section A, B and C in Figure 1) on the flowrates of fresh hydrogen required for the isomerization unit (Section D), hydrogen entering the

isomerization reactor (Section E) and hydrogen discharge (Section F) will be studied.

Besides Equation (4) included in the integration of the isomerization process with the hydrogen network as a constraint, the overall mass and components balances around the reactor were taken into account. The hydrogen inlet streams to the reactor ( $G_{iso}$ ) (isomerization sink) and hydrogen outlet ( $w_{iso}$ ) (isomerization purge or isomerization source) are set as unknown variables. Their flowrates are based on determining the hydrogen consumption determined from Equation (4).

As presented by Umana et al. [34], and Jia et al. [35], the hydrogen material balance of the hydro-processing reactor (Equation (5)) is based on the hydrogen consumption (HC) in the reactor determined from Equation (4).

$$w_{iso} (1 - y_{iso}) = G_{iso} (1 - z_{iso}) - HC$$

i, j for isomerization unit (5)

Where the purge hydrogen outlet of the reactor  $(w_{iso} (1 - y_{iso}))$  results in the inlet hydrogen flowrate to the reactor  $(G_{iso} (1 - z_{iso}))$  minus the hydrogen consumption in the reactor (HC).  $z_{iso}$  and  $y_{iso}$  are the concentrations of impurities entering and exiting the isomerization reactor, respectively. The hydrogen consumption (HC) in Equations (4) and (5) is the amount of hydrogen required for the isomerization reactions plus the hydrocarbons are produced from any hydroprocessing unit and refer to any gaseous or semi-gaseous molecule with a molecular weight less than or equal to that of pentane (C1-C5). The range of the operational parameters in Equation (4) are set as a constraints in the isomerization model as presented in Equations (6)-(11).

$naph^{L} \le naph \le naph^{U} $	6)
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 $BZ^{L} \le BZ \le BZ^{U} \tag{7}$ 

 $Temp^{L} \le Temp \le Temp^{U}$ (8)

$$LHSV^{L} \le LHSV \le LHSV^{U}$$
(9)

$$\mathrm{HC}^{\mathrm{L}} \le \mathrm{HC} \le \mathrm{HC}^{\mathrm{U}} \tag{10}$$

 $RON^{L} \le RON \le RON^{U} \tag{11}$ 

Each parameter ranged from the lowest value in its range to the highest value in its range. naph<sup>L</sup>, BZ<sup>L</sup>, Temp<sup>L</sup>, LHSV<sup>L</sup>, HC<sup>L</sup>, and RON<sup>L</sup> are the lower value of naphthene, benzene, temperature, liquid hourly space velocity, hydrogen consumption in the isomerization reactor, and the research octane number in their rang, respectively, while the naph<sup>U</sup>, BZ<sup>U</sup>, Temp<sup>U</sup>, LHSV<sup>U</sup>, HC<sup>U</sup>, and RON<sup>U</sup> are the upper values of the operating parameters and the research octane number in their range, respectively.

#### Reuse/recycling hydrogen network

The hydrogen network in refinery is distributed to sources and sinks as described in Figure 2. Hence, source streams are the outlet hydrogen streams from the hydro-processes units as hydrotreating, hydrocracking, and isomerization units. Sink streams are the hydrogen demand of the previous units and are added to the reactor of each unit. There is fresh hydrogen can be supplied to hydrogen sink streams when hydrogen sent to sinks is not satisfied by hydrogen sources. The fresh hydrogen sources may be from catalytic reforming or hydrogen plant. Each source  $W_i$  may be sent to hydrogen sink  $(w_{i,j})$  or to fuel system (fuel<sub>i</sub>) as described in Equation (12) [1, 9].

$$W_{i} = \sum_{j=1}^{N_{sinks}} w_{i,j} + \text{ fuel}_{i} \qquad \forall i = 1, ..., N_{sources} (12)$$



Figure 2 Source/ Sink allocation [1]

Each sink j takes its demand of hydrogen  $(G_j)$  from source i  $(w_{i,j})$  or from fresh hydrogen  $(Fresh_j)$  as shown in Equation (13).

$$G_{j} = \sum_{i=1}^{N_{sources}} w_{i,j} + Fresh_{j} \quad \forall j = 1, ..., N_{sinks}$$
(13)

The impurity material balance for sink j is described in Equation (14):

$$G_{j}z_{j} = \sum_{i=1}^{N_{sources}} w_{i,j} y_{i} + Fresh_{j}x_{f}$$

$$\forall j = 1, ..., N_{sinks+1}$$
(14)

where  $x_{f_i} z_j$  are the impurity concentrations of the fresh hydrogen and sink j, respectively, The minimum  $(z_i^{min})$  and maximum  $(z_i^{max})$  of impurity

concentration of sink stream are used as constraints as shown in Equation (15).

$$z_j^{\min} \le z_j \le z_j^{\max} \tag{15}$$

The total fresh hydrogen of the hydrogen network is the sum of all fresh hydrogen streams supplied to the hydrogen sinks as shown in equation (16).

Fresh hydrogen = 
$$\sum_{j=1}^{N_{sinks}} Fresh_j$$
 (16)

For non-negativity, Equations (17) and (18) are taken into consideration.

$\operatorname{fresh}_{j} \geq 0$	(17)
$w_{ii} \ge 0$	(18)

The objective function applied is the minimization of fresh hydrogen requirement of hydrogen network as illustrated in Equation (19).

$$\min = Fresh hydrogen \tag{19}$$

This model is nonlinear programming and is solved by commercial LINGO optimization software V.14.

To integrate the isomerization model with the hydrogen reuse/recycling network, the isomerization model is included in the hydrogen reuse/recycling network model.

#### Integration isomerization model with reuse/recycling hydrogen network

In this section, the integration of the isomerization model with a hydrogen reuse/recycling network is presented. Isomerization model equations were used as new constraints for the objective function Equation (19). The solution of Equation (19) subjected to the constraints of Equations (4)-(11) and (12)-(18) is nonlinear and can be solved by the commercial LINGO optimization software V.14.

Note that Equations (4) and (5) are used as constraints for the minimum fresh hydrogen Equation (19). Also note, when solving this model through Lingo optimization software, the obtained RON may or may not be the maximum value of RON.

To obtain maximum RON and minimum fresh hydrogen in the same model, a multi-objective optimization model is required.

# Goal programming multiobjective optimization model for integration of isomerization model with the reuse/recycling hydrogen network

Maximizing the research octane number of isomerate (max = Isomerate RON) and minimizing fresh hydrogen for the hydrogen reuse/recycling network (min = fresh hydrogen) containing this isomerization process is the objective function of the multi-objective optimization model. In this work, the single objective functions were reformulated with respect to the multi-objective function model (goal programming optimization) and the resulting formulas are as presented in Equations (20)-(26) for the reuse/recycling hydrogen network. In this multi-objective model, there are two objective functions, the first objective function is to minimize fresh hydrogen and the second objective function is to maximize RON. Solving this multi-objective Equation (20) subjected to the constraints equations (21)-(26) and (4)-(18) yields the maximum research octane number, minimum hydrogen consumption in the isomerization process and thus minimum amount of fresh hydrogen for the overall hydrogen network.

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min
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 $= \delta_{\text{fresh hydrogen}}^{+} + \delta_{\text{fresh hydrogen}}^{-} + \delta_{\text{Isomerate RON}}^{+} + \delta_{\text{Isomerate RON}}^{-}$ (20)

fresh hydrogen – 
$$\delta_{\text{fresh hydrogen}}^+ + \delta_{\text{fresh hydrogen}}^-$$
  
=  $a_{\text{fresh hydrogen}}$  (21)

Isomerate RON + 
$$\delta^{+}_{Isomerate RON} - \delta^{-}_{Isomerate RON}$$
  
=  $a_{Isomerate RON}$  (22)

a<sub>fresh hydrogen</sub>

$$= \frac{\text{Isomerate RON} - \text{Isomerate RON}^{\min}}{\text{Isomerate RON}^{\max} - \text{Isomerate RON}^{\min}}$$
(24)

$$\delta^+_{\text{fresh hydrogen}}$$
,  $\delta^-_{\text{fresh hydrogen}} \ge 0$  (25)

$$\delta_{\text{Isomerate RON}}^+$$
,  $\delta_{\text{Isomerate RON}}^- \ge 0$  (26)

The goal required to achieve in multi-objective model is the minimum value of fresh hydrogen and the maximum value of the isomerate RON. Fresh hydrogen<sup>min</sup> in Equation (23) is obtained by solving the objective function Equation (19) subjected to constraints Equations (4)-(18). Fresh hydrogen<sup>max</sup> is maximum fresh hydrogen before optimization [36-39]. Isomerate RON<sup>max</sup> in Equation (24) is the maximum isomerate RON. In this work, maximum isomerate RON is taken as the upper value of RON (88.6) in its operating range as collected from field data of Equation (4) as reported by Shehata et al. [43]. Isomerate RON<sup>min</sup> is the minimum isomerate RON and it is taken as the lowest value of RON in its operating range (86).

# **Case study**

The existing hydrogen network of a refinery in Egypt including the isomerization process, shown in Figure 1, and its operating data are given in Equation (4) is presented here to illustrate the applicability of the proposed approach presented to integrate the isomerization hydro-processing unit with the overall hydrogen network. As shown in Table 1, the hydrogen network consists of two hydrogen producers namely the H<sub>2</sub> plant and the catalytic reforming unit (CRU), and two hydrotreating units namely the naphtha hydrotreater (NHT) and the diesel hydrotreater (DHT), isomerization unit (ISO), and hydrocracking unit (HCU). Currently fresh hydrogen is available to this network at a rate of 2265.71 kmol/h with an impurity concentration of 0.01%. There is a pressure swing adsorption (PSA) that is used to regenerate a certain amount of CRU flowrate at 9.78% impurity concentration to produce a regeneration stream with 0.1% impurity concentration which is sent to the hydrocracking unit and a residual stream with 45.43% impurity concentration. The outlets of the units mentioned in this study are used as sources and the inlets of these units are used as sinks as shown in Table 1.

# Integration of isomerization model with hydrogen regeneration network

#### **Regeneration of hydrogen network**

As presented in Figure 3, the hydrogen source streams may be regenerated in a regenerator as pressure

swing adsorption (PSA), membrane, and cryogenic separation system for more recovery of hydrogen. The regenerator of hydrogen network is partitioning regenerator having top and bottom products [3, 11, 26].

Table 1 Hydrogen sources and sinks for Case Study

Stream sinks SKj	Hydrogen sinks	Flowrates (Kmol/h)	Impurity conc. (Kmol %)
1	Inlet HCU	3276.9	0.1
2	Inlet PSA	1287.8	9.78
3	Inlet NHT	135.56	9.78
4	Inlet DHT	2873.48	15.77
5	Inlet Isomerization	241.3 (To be determine)	9.78
Stream	Hydrogen	Flowrates	Impurity
sources	sources	(Kmol/h)	conc.
SRi			(Kmol %)
1	Outlet 1 PSA	1011.19	0.1
2	CRU	2141.2	9.78
3	Outlet DHT	2396.94	16.96
4	Outlet HCU	515.15	26.52
5	Outlet 2 PSA	276.61	45.43
6	Outlet NHT	97.9	53.16
7	Outlet Isomerization	122.6 (To be determine)	58.22
	Fresh supply (H <sub>2</sub> Plant)	2265.71 (To be determine)	0.01

The top product has high hydrogen purity and the bottom product has low hydrogen purity. Each hydrogen source (Q<sub>i</sub>) may be integrated with sinks in hydrogen network ( $w_{i,j}$ ), sent to the regenerator ( $w_{regen,i}$ ), or sent to fuel system as presented in Equation (27).

$$Q_{i} = \sum_{j=1}^{N_{sinks}} w_{i,j} + w_{regen,i} + Fuel_{i}$$
  
for i = 1,2,3 ..., N<sub>sources</sub> (27)

Where  $\sum_{j=1}^{N_{sinks}} w_{i,j}$  is the summation of all flowrates

from source i to sinks j.

The feed to the regeneration unit (w<sub>regen</sub>) is the summation of all flowrates sent from hydrogen sources  $(\sum_{i=1}^{N_{sources}} wregen_i)$  to the regenerator as described in Equation (28).

$$w_{\text{regen}} = \sum_{i=1}^{N_{\text{sources}}} \text{wregen}_i$$
 (28)

Material and impurity balance on the regenerator is illustrated in Equations (29) and (30).

$$\sum_{i=1}^{N_{sources}} wregen_i = wregen_p + wregen_r$$
(29)

$$\sum_{i=1}^{N_{sources}} wregen_i y_i = wregen_p y_p + wregen_r y_r \quad (30) \qquad G_j = \sum_{i=1}^{N_{sources}} w_{i,j} + wregen_{pj} + wregen_{rj} + Fresh_j$$
for j = 1,2,3, ..., N<sub>sinks</sub> (35)



Figure 3 Structural representation for hydrogen regeneration network

Where  $wregen_p$  and  $wregen_r$  are the top and bottom products of the partitioning regenerator, respectively;  $y_i$  is the impurity concentration of source i;  $y_p$  and  $y_r$  are respectively the impurity concentration of the top, and the bottom flowrates of the regenerator.

For the regenerator, the product can be determined according to the hydrogen recovery ( $\alpha_1$ ) as described in Equation (31) [3, 11]:

$$w_{\text{regen}} \alpha_{l} (1 - y_{f}) = \text{wregen}_{p} (1 - Y_{p})$$
(31)

Where  $y_f$  is the impurity concentration of inlet feed to the regenerator;  $(1 - y_f)$  is the hydrogen purity concentration of the inlet feed to the regenerator;  $(1 - y_p)$  is the hydrogen purity concentration of the top product stream of the regenerator.

The top and the bottom products of the regenerator are considered as a sources streams and can be sent to sink j ( $wregen_{pj}$ ,  $wregen_{rj}$ ) or sent to the fuel system ( $wasteregen_p$ ,  $wasteregen_r$ ) as described in Equation (32) and (33).

$$wregen_{p} = \sum_{j=1}^{N_{sinks}} wregen_{pj} + wasteregen_{p} \quad \text{for } j$$
$$= 1,2,3 \dots, N_{sinks} \quad (32)$$

wregen<sub>r</sub> = 
$$\sum_{j=1}^{r}$$
 wregen<sub>rj</sub> + wasteregen<sub>r</sub> for j

$$= 1,2,3..., N_{sinks}$$
 (33)

waste = 
$$\sum_{i=1}^{N_{sources}} waste_i + wsteregen_p$$
  
+ wasteregen\_r (34)

Material and impurity balance on sink j is given in Equations (35) and (36):

$$G_{j}z_{j} = \sum_{i=1}^{N_{sources}} w_{i,j} y_{i} + wregen_{pj}y_{p} + wregen_{rj}y_{r} + Fresh_{j}x_{j}$$
(36)

where  $\sum_{i=1}^{N_{sources}} w_{i,j}$  is the summation of all hydrogen flowrates sent from sources to sink j; wregen<sub>pj</sub> is the hydrogen flowrate sent from top product of the regenerator to sink j; wregen<sub>rj</sub> is the hydrogen flowrate sent from bottom product of the regenerator to sink j; Fresh<sub>j</sub> is the fresh hydrogen flowrate sent from fresh source to sink j; x<sub>j</sub> is the fresh hydrogen impurity concentration. The fresh hydrogen of the hydrogen streams  $(\sum_{i=1}^{N_{sinks}} fresh_j)$  sent to sinks as presented in Equation (37).

Fresh hydrogen = 
$$\sum_{j=1}^{N_{sinks}} \text{fresh}_j$$
 (37)

The lower and the upper bound of the bottom product of the regenerator is described in Equation (38):

$$y_r^{\rm L} \le y_r \le y_r^{\rm U} \tag{38}$$

Where  $y_r^L$  is lower concentration of bottom stream of all sources when they regenerated without mixing;  $y_r^U$ is the upper concentration of bottom stream of all sources when they regenerated without mixing. To get the lower and the upper concentration of  $y_r$  it is required to determine  $y_r$  for each source stream with impurity concentration  $y_i$  according to Equation (39) [11].

$$y_{r} = \frac{\left[y_{i} - \left(\frac{100 - y_{i}}{100 - y_{p}}\right)\alpha_{l} y_{p}\right]}{\left[1 - \left(\frac{100 - y_{i}}{100 - y_{p}}\right)\alpha_{l}\right]}$$
(39)

Where  $\alpha_l$  is the hydrogen recovery.

The objective function of the hydrogen regeneration network takes into account the fresh hydrogen and the regeneration costs as described in Equation (40) [11]:

min = [Fresh hydrogen \* 
$$W_{cost}$$
  
+  $w_{regen} \cdot R_{cost}$ ] AWH (40)

Where  $W_{cost}$  is the unit cost of fresh hydrogen supplied to all sinks;  $R_{cost}$  is the unit cost of the regenerated flowrate; AWH is the annual working hours.

The hydrogen regeneration network model presented above is non-linear and can be solved by Lingo optimization program version 14.

#### Integration isomerization model with hydrogen regeneration network

To integrate the isomerization model with the hydrogen regeneration network, the isomerization model was included in the hydrogen regeneration network model. Isomerization model equations were used as new constraints for the objective function Equation (40). The solution of Equation (40) subjected to the constraints of Equations (4)-(11) and (27)-(38) is nonlinear and can be solved by the commercial LINGO optimization software V.14.

Note that Equations (4)-(11) are used as constraints for the objective function Equation (40). Also note, when solving this model through Lingo optimization software, the obtained RON may or may not be the maximum value of RON.

To obtain minimum fresh hydrogen, minimum regenerated flowrate, and maximum RON, in the same model, a multi-objective optimization model is required.

#### Goal programming multi-objective optimization model for integration of isomerization model with the hydrogen regeneration network

Minimizing fresh hydrogen (min = fresh hydrogen), minimizing the regeneration flowrate (min =  $w_{regen}$  ), and maximizing the research octane number are the objective functions of the multi-objective goal programming optimization model when the isomerization model is integrated with the hydrogen regeneration network. In this work the individual objective functions are reformulated in terms of the goal programing multiobjective optimization model and the resulting formulas are presented in Equations (41)-(50). In this multiobjective model, we have three objective functions, the first objective function is to minimize fresh hydrogen for the hydrogen network, the second objective function is to minimize the regenerating flowrate, and the third objective function is to maximize RON. The isomerization unit is integrated into the hydrogen regeneration network of the plant containing this isomerization unit.

#### min

$$= \delta_{\text{fresh hydrogen}}^{+} + \delta_{\text{fresh hydrogen}}^{-} + \delta_{\text{wregen}}^{+} + \delta_{\text{wregen}}^{-} + \delta_{\text{Isomerate RON}}^{+}$$
(41)

(Fresh hydrogen  $* W_{cost}$ ) -  $\delta^+_{fresh hydrogen}$ 

 $+ \delta_{\text{fresh hydrogen}}^{-} = a_{\text{fresh hydrogen}}$ (42)

$$(w_{regen} \cdot R_{cost}) - \delta^+_{wregen} + \delta^-_{wregen} = a_{wregen}$$
 (43)

Isomerate RON +  $\delta^+_{Isomerate RON} - \delta^-_{Isomerate RON}$ =  $a_{Isomerate RON}$  (44)

a<sub>fresh hydrogen</sub>

$$= \frac{\text{fresh hydrogen} - \text{fresh hydrogen}^{\min}}{\text{fresh hydrogen}^{\max} - \text{fresh hydrogen}^{\min}}$$
(45)

awregen

$$=\frac{w_{\text{regen}} - w_{\text{regen}}}{w_{\text{regen}} - w_{\text{regen}} \min}$$
(46)

a<sub>Isomerate RON</sub>

$$= \frac{\text{Isomerate RON} - \text{Isomerate RON}^{\text{Infin}}}{\text{Isomerate RON}^{\text{max}} - \text{Isomerate RON}^{\text{min}}}$$
(47)

 $\delta^+_{\text{fresh hydrogen}}, \delta^-_{\text{fresh hydrogen}} \ge 0$  (48)

$$\delta_{\text{wregen}}^+, \delta_{\text{wregen}}^- \ge 0$$
 (49)

 $\delta^+_{\text{Isomerate RON}}, \delta^-_{\text{Isomerate RON}} \ge 0$  (50)

The minimum value of fresh hydrogen, the minimum value of regenerating flowrate, and the maximum value of isomerate RON are the target of the goal programming multi-objective function. Fresh hydrogen<sup>min</sup> , and  $w_{regen}$ <sup>min</sup>, in Equation (45) and (46), respectively, are obtained by solving the objective function Equation (40) subjected to the constraints of Equations (4)-(11) and (27)-(38). Fresh hydrogen<sup>max</sup> is the maximum fresh hydrogen before optimization.  $w_{regen} \stackrel{max}{\longrightarrow}$  in Equation (46) is the maximum sources flowrates that will be regenerated and is taken as the sum of all sources flowrates assuming that maximum regeneration is obtained when all sources streams are regenerated. Isomerate  $\ensuremath{\mathsf{RON}^{\max}}$  in Equation (47) is the maximum isomerate RON. In this work, maximum isomerate RON is taken as the upper value of RON (88.6) in its operating range as collected from field data of Equation (4) as reported by Shehata et al. [43]. Isomerate RON<sup>min</sup> is the minimum isomerate RON and it is taken as the lowest value of RON in its operating range (86).

The same case study presented above was resolved in case of hydrogen regeneration network. A PSA regenerator was used for regeneration with the specified product with 0.1% impurity concentration ( $\gamma_p$ ) and 90% hydrogen recovery ( $\alpha_i$ ). The unit price of fresh hydrogen and the unit cost of regenerated flowrate were assumed to be 1 and 0.6 USD/ kmol, respectively [3, 11].

#### **Results and Discussion**

Solving the presented case study by the reuse/recycling method described in this paper without incorporating the isomerization process model into the hydrogen network (solving Equation (19) subject to the constraints of Equations (12)-(18)), it is found that fresh hydrogen decreased from 2265.71 to 2257.938 kmol/h (Table (2, column 2)). Therefore the decrease in fresh hydrogen is 7.772 kmol/h. On the other hand, when the isomerization model is included with the hydrogen reuse/recycling network (solving the objective function equation (19) subject to the constraints of equation (4)-(11) and (12)-(18)) the fresh hydrogen significantly decreased from 2265.71 to 2244.839 kmol/h. The saving in fresh hydrogen is 20.871 kmol/h when hydrogen consumption and the research octane number equation

for isomerization unit are included in the optimization process. The optimal research octane number for the isomerization unit that gives the minimum fresh hydrogen flowrate is determined at its lowest value of 86. Note that in this step, the isomerization model equations are used as constraints for the objective function of the minimum fresh hydrogen for the hydrogen network. The optimal operating conditions for the isomerization process that yield a minimum amount of fresh hydrogen are illustrated in Table (2, column 3).

The advantage of using the optimized isomerization model in the hydrogen network is the increased savings in fresh hydrogen of 20.871 kmol/h. Another advantage of the proposed model is that the isomerization process sink (reactor inlet) is reduced from 241.26 to 113.722 kmol/h. The discharge stream of the isomerization process (purge stream) was also reduced from 122.6 kmol/h to zero. In the previous optimization of the hydrogen network without considering the isomerization process model, and stabilizing the hydrogen consumption, approximately velocity, naphthene and benzene content). The optimal research octane number is determined to be at its lowest value of 86.0 at the optimal operating conditions of 102.6 kmol/h HC, 156 °C Temp, 2.81 LHSV, 13.96 wt. % Naphthene and 0.31 wt. % benzene.

To obtain the maximum research octane number and the minimum fresh hydrogen at the same time, the goal programming multi-objective optimization is applied (solving equation (20) subject to the constraints of equations (4)-(11), (12)-(18), and (21)-(26)).

It is found that the operating conditions for the isomerization process change to obtain the minimum fresh hydrogen for the hydrogen network and the maximum RON for the isomerization process as shown in Table (2, column 4). It is observed that to get the minimum fresh hydrogen and the maximum RON in the same objective function, the naphthene and liquid hourly space velocity decreased to 7.83 wt. % and 2.18 h<sup>-1</sup>, respectively. Figure 4 is the optimized hydrogen network obtained when the isomerization model is embedded in the

**Table 2** Optimum hydrogen network conditions with and without embedding isomerization model in reuse/recyclinghydrogen network

	Reuse/recycling				
Item	Without embedding isomerization model	With embedding isomerization model and using isomerization equations as constraints to minimum fresh hydrogen	With embedding isomerization model using multi-objective optimization program		
Optimization formulation	Non- linear programing Objective function Equation (19) subjected to constraints Equations (12)-(18)	Non- linear programing Objective function Equation (19) subjected to constraints Equations (4) – (11) and (12)-(18)	Goal programming multi- objective with integration of isomerization model with reuse/recycling hydrogen network Equation (20) subjected to constraints equations (21) – (26), (4) – (11), and (12)-(18)		
Minimum fresh hydrogen of hydrogen network, kmol/h	2257.938	2244.839	2244.839		
Decrease in fresh hydrogen, kmol/h	7.772	20.871	20.871		
Maximum Isomerate RON	-	86.0	88.6		
Fuel discharge of hydrogen network, kmol/h	1004.488	996.367	996.367		
Inlet hydrogen feed to isomerization reactor, G <sub>iso</sub> , kmol/h	241.3 (constant)	113.722	113.722		
Purge hydrogen stream from isomerization unit, W <sub>iso</sub> , kmol/h	122.6 (constant)	zero	zero		
Optimum isomerization reactor temperature, °C	-	156.0	156.0		
Optimum liquid hourly space velocity, h <sup>-1</sup>	-	2.81	2.18		
Optimum naphthens wt.%	-	13.96	7.83		
Optimum benzene wt.%	-	0.31	0.31		
Optimum hydrogen consumption, kmol/h	-	102.6	102.6		

122.6 kmol/h of hydrogen was spent in the fuel system.

Moreover, the optimal research octane number is determined on the basis of the optimal hydrogen consumption in the reactor subjected to the change in operating conditions (temperature, liquid hourly space hydrogen reuse/recycling network.

Integration the isomerization process with the hydrogen network model opens opportunities to exploit different degrees of freedom available for process optimization. For example, the optimal value for the weight percentage of naphthene and benzene, operating conditions, and hydrogen consumption are obtained from the maximum research octane number required for the isomerization process. The optimal hydrogen consumption for the isomerization process translates into the minimum hydrogen flowrate for the entire hydrogen network.

Regarding the regeneration of the hydrogen network, the same case study was solved by using PSA as a regeneration system. The bottom product impurity concentration of PSA ( $y_r$ ) was calculated for all sources according to Equation (39) and it was found that the lower ( $y_r^{LB}$ ) and the upper ( $y_r^{UB}$ ) bottom product limits are 51.8 and 93.30 respectively.

Solving the hydrogen network by the regeneration method without integration the isomerization process model in the hydrogen network (solving objective function Equation (40) subjected to the constraints of Equations (27)-(38)), it is found that 1922.449 kmol/ h was achieved with saving flowrate of 343.261 kmol/hr and saving percentage of 15.15 % (Table (3, column 2)).

The goal programming multi-objective function was applied to obtain the minimum fresh hydrogen for the hydrogen regeneration network, the minimum regenerating flowrate, and the maximum RON. Equation (41) is solved under the constraints of Equations (4)-(11), (27)-(38), and (42)-(50). The results are shown in Table (3, column 4). It is noted that the maximum research octane number of isomerate, the minimum fresh hydrogen and the minimum regenerating flowrate were obtained simultaneously by solving the multi-objective function. As shown in Table (3, column 4), it is noted that the optimal operating conditions for the isomerization process that give the maximum research octane number, the minimum fresh hydrogen, and the minimum regenerating flowrate differ from their values that give the minimum fresh hydrogen and the minimum regenerating flowrates only (Table (3, columns 3 and 4)). It is found that the naphthene and liquid hourly space velocity decreased to 7.191 wt. % and 2.24 h<sup>-1</sup>, respectively, to achieve the goal programming multi-objective function.



Figure 4 Optimum hydrogen network design for case study using integration of the isomerization model with the hydrogen network. The numbers represent the total gas flowrate in kmol/h

On the other hand, embedding the isomerization model in the hydrogen regeneration network by solving the objective function Equation (40) subjected to the constraints of Equations (4)-(11) and (27)-(38), the fresh hydrogen is considerably decreased to 1817.812 kmol /hr with saving flowrate of 447.898 kmol/h and saving percentage of 19.77 %. The minimum regenerating flowrate was determined to be 625.826 kmol/h. The optimum isomerate research octane number giving minimum fresh hydrogen and minimum regenerating flowrate was determined to be at its minimum value of 86. The isomerization process sink (reactor inlet) is reduced from 241.26 to 113.722 kmol/h. The discharge stream of the isomerization process (purge stream) was also reduced from 122.6 kmol/h to zero. All data of optimization is illustrated in Table (3, column 3).

The advantages of embedded the isomerization model in the hydrogen regeneration network is the increase in the fresh hydrogen saving (447.898 kmol/h), and the decrease in the hydrogen discharge. Another advantage of the presented model for regeneration system is that hydrogen sink to the isomerization process (inlet of the reactor) is decreased from 241.26 to 113.722 k mole/hr. In addition, the source stream (reactor outlet) of isomerization process is decreased to zero so, no hydrogen was routed to the fuel system from the isomerization process. Figure 5 is the optimized hydrogen network obtained when the isomerization model is embedded in the hydrogen regeneration network. 110.8032 kmole/h from catalytic reforming unit (CRU) and 515.0236 kmole/h from outlet of hydrocracking unit (outlet HCU) are sent to the regenerator (new PSA) to produce 430.996 kmol/h as a top product stream with 0.1

% impurity concentration and 194.83 as a bottom product stream of the regenerator with impurity concentration of 75.445 %.

isomerization reactor, the research octane number of isomerate, and the fresh hydrogen flowrate required to optimize the hydrogen network were investigated. Hydrogen network optimization is developed when an

Table 3 Optimum	hydrogen net	vork	conditions	with	and	without	embedding	isomerization	model	in hyd	rogen
	regeneration r	etwo	ork								

	Regeneration				
Item	Without embedding isomerization model	With embedding isomerization model and using isomerization equations as constraints to minimum fresh hydrogen	With embedding isomerization model and using multi- objective optimization program		
Optimization formulation	Non- linear programing objective function Equation (40) subjected to Equations (27)-(38).	Non- linear programing objective function Equation (40) subjected to Equations (4)-(11), and (27)-(38).	Goal programming multi- objective with integration of isomerization model with regeneration hydrogen network Equation (41) subjects to Equations (42)-(50), (4)-(11), and (27)-(38).		
Minimum fresh hydrogen of hydrogen network, kmol/h	1922.449	1817.812	1817.812		
Decrease in fresh hydrogen kmol/h	343.261	447.898	447.898		
Maximum RON	-	86.0	88.6		
Regenerated flowrate, kmol/h	508.521	625.826	625.826		
Fuel discharge of hydrogen network , kmol/h	668.999	569.34	569.34		
Inlet hydrogen feed to isomerization reactor, G <sub>isom</sub> , kmol/h	241.3 (constant)	113.722	113.722		
Purge hydrogen stream from isomerization unit, w <sub>isom</sub> , kmol/h	122.6 (constant)	zero	zero		
Regenerated flowrate, kmol/h	508.521	625.826	625.826		
Regenerator bottom product concentration, yr	78.262	75.445	75.445		
Optimum isomerization reactor temperature, °C	-	156	156.0		
Optimum liquid hourly space velocity, h <sup>-1</sup>	-	2.81	2.24		
Optimum naphthens wt.%	-	13.96	7.191		
Optimum benzene wt.%	-	0.31	0.31		
Optimum hydrogen consumption, kmol/h	-	102.6	102.60		

# Conclusion

To date, studies based on refinery hydrogen management have made the performance of the isomerization process constant in the optimal design of the hydrogen network. In this way, the interactions between variable feed specifications, operating conditions, hydrogen consumption in the isomerization process, the research octane number of the isomerization product, and the fresh hydrogen of the hydrogen network were neglected for the optimization of hydrogen network containing the isomerization unit. The current work takes into account this lack of hydrogen network optimization. The interactions between different operating conditions, feed specifications, hydrogen consumption in the isomerization model is included in the network for reuse/recycling and regeneration using Lingo optimization software version 14. Optimal conditions for the isomerization process that give the maximum research octane number of the product and minimum fresh hydrogen for reuse/recycling and regeneration hydrogen network using goal programming multi-objective optimization approach were determined. The results showed that a reduction of 7.772 kmol/h in fresh hydrogen reuse/recycling network without including the isomerization model, while a saving of 20.871 kmol/h in fresh hydrogen was obtained in the case of including the isomerization model. On the other hand, a significant reduction of 447.898 kmol/h in fresh hydrogen was observed in the hydrogen regeneration network when including the isomerization model compared to that achieved when neglecting the isomerization model (343.261 kmol/h). In addition, the optimum operating conditions of the isomerization unit in the hydrogen reuse/recycling network that gave minimum fresh hydrogen, maximum research octane number were 156 °C, 2.18 h<sup>-1</sup>, 7.83 wt%, 0.31 wt%, 102.6 kmol/h for the isomerization reactor temperature, LHSV, naphthenes weight percentage, benzene weight percentage, and hydrogen consumption in the reactor, respectively, while in the case of the hydrogen regeneration network they were 156 °C, 2.24 h<sup>-1</sup>, 7.191 wt%, 0.31 wt%, 102.6 kmol/h respectively.

W <sub>cost</sub>	Unit cost of fresh hydrogen
Wij	Flowrate sent from source i to sink j
Wregen, p	Top flowrate of regenerator
X <sub>f</sub>	Impurity concentration of fresh hydrogen
<b>Y</b> iso	Impurity concentration of isomerization source
<b>y</b> i	Impurity concentration of source i
Уp	Impurity concentration of regenerator top
	product
<b>y</b> r	Impurity concentration of regenerator bottom
	product
z <sub>iso</sub>	Impurity concentration of isomerization sink
$\alpha_l$	Hydrogen recovery
$\delta_i^-, \delta_i^+$	the deviations of the objective function from the
	optimum goals



Figure 5 Optimum hydrogen regeneration network design for case study 2 using integration of the isomerization model with the hydrogen regeneration network. The numbers represent the total gas flowrate in kmol/h

# Abbreviations

AWH	Annual working hours
Bz	Benzene
CRU	Catalytic reforming unit
DHT	Deseil hydrotreating unit
Fresh <sub>j</sub>	Fresh hydrogen flowrate to sink j
$f_i(x)$	Individual objective function
G <sub>iso</sub>	Isomerization sink flowrate
HC	Hydrogen consumption
HCU	Hydrocracking unit
ISO	Isomerization unit
LHSV	Liquid hourly space velocity
naph	Naphthenes
NHT	Naphtha hydrotreating unit
R <sub>cost</sub>	Unit cost of regenerated flowrate
RON	Research octane number
Temp	Temperature
W <sub>regen,pj</sub>	Flowrate sent from regenerator top product to
	sink j
W <sub>regen,rj</sub>	Flowrate sent from regenerator bottom product
	to sink j
Wiso	Isomerization source flowrate

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