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Optimal Blue Hydrogen Process with CO_2 Capture, Utilisation and Storage

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Hydrogen (H_2) energy has a high potential to become a source of future sustainable fuel to replace fossil fuel. At present, natural gas conversion is the conventional process to produce H_2. However, this process produces carbon dioxide (CO_2) emissions as a by-product. CO_2 capture, utilisation, and storage (CCUS) technologies can be integrated in conventional H_2 plants to address this issue. By doing so, conventional H_2 plants can be retrofitted to produce cleaner H_2 called blue H_2. This work presents a mathematical model to optimise blue H_2 processes integrated with CCUS technologies. The research objectives are to determine optimal and feasible decarbonisation systems for H_2 production and optimal storage technologies for the produced H_2 with minimum cost. The developed optimisation-based model considers different grey H_2 processes, CO_2 capture technologies, CO_2 transportation, utilisation, storage, and H_2 storage. The model factors technology efficiency, costing and overall energy consumption. The developed model is demonstrated with a blue H_2 production case study. The optimised blue H_2 process with CCUS was obtained with the optimisation objective of minimising total annualised cost (TAC).

1. Introduction

Approximately 40 % of the global CO_2 emission is due to the combustion of fossil fuel for electricity generation on residential, commercial, and industrial scales (Abdul Latif et al., 2021). These detrimental effects have alerted policymakers and researchers to decarbonise the energy sector. Hydrogen (H_2) energy is an alternative fuel resource of the future and does not produce CO_2 emissions when utilised. Steam methane reforming (SMR) is the most used technology to convert natural gas to H_2. This H_2 production path is categorised as grey H_2. To achieve net-zero CO_2 emissions, grey H_2 processes need to be retrofitted with carbon capture, utilisation, and storage (CCUS). This turns the previously mentioned grey H_2 to blue H_2 production, where no CO_2 is expected to be emitted into the environment (ATCO, 2022). The implementation of blue H_2 processes can become realisable when optimisation is done on the economic aspect. In literature, research work can be found on H_2 processes and carbon capture systems. For instance, Li et al. (2020) proposed a multi-criterion decision-making model (MCDM) to study the sustainability assessment of grey H_2 production. Santibanez-Gonzalez (2017) used a stochastic MILP model to minimise the carbon capture system's investment and construction cost. Many scholars have done optimisation-based research on the integration of H_2 production processes and carbon capture systems. Cormos et al. (2018) investigated the technical and economic performances of H_2 production from SMR and autothermal reforming integrated with pre-combustion carbon capture. Besides, Roussanaly et al. (2020) compared the H_2 production cost through SMR without and with carbon emissions capture and storage. However, several research gaps are identified. The available carbon capture research discussed above targets the cement industry instead of H_2 production plants. In addition, the blue H_2 research work above considered limited technological options in their analysis. In other words, these works focused only on one grey

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H_2 production route and one carbon capture technology. Such focus may lead to restricted possibilities of achieving higher H_2 production performance as no alternative technologies were considered.

2. Research novelty

To address these research gaps, an optimisation-based mathematical model is employed in this research to enhance blue H_2 performance with various technologies being considered. In addition, optimisation of the blue H_2 process can be investigated from many perspectives, which include total annualised production cost, annual energy consumption and annual CO_2 production/emissions. Therefore, this research focus on the optimisation objective of achieving minimum TAC for the blue H_2 process.

3. Methodology

The first step of this work focused on technology compilation through a literature review. The compiled technologies are then put into a single diagram called a superstructure. Superstructure illustrates all the possible interconnections between technologies considered. For this work, the superstructure developed is shown in Figure 1. In addition, performance parameters for each technology in the superstructure were collected, which include grey H_2 yield, carbon capture efficiency, energy consumption for grey H_2 production, carbon capture and H_2 storage and economic data for each technology.

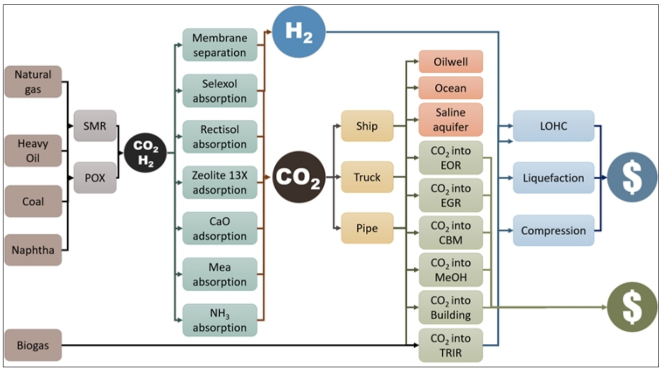

Figure 1: Superstructure of blue H_2 process with CCUS

After collecting useful performance data for the relevant technologies, the next step was to formulate mathematical equations based on the developed superstructure. These equations include relationships like material mass flowrates, the energy consumption of the process, annualised capital, and the operating cost of the process. Figure 2 illustrates the general principle used to formulate equations for this work. It serves as a guiding framework for how the equations were developed for this work. As shown, three main sections are identified throughout the process, namely feed (*f*), technology (*g*), and product (*h*). For instance, in grey H_2 production, feed (*f*) refers to natural gas, heavy oil, coal, and naphtha fed into various technologies. The considered technologies (*g*) are steam methane reforming (SMR) and natural gas partial oxidation (POX). Then, the product formed from the technology (*h*) is the syngas containing mainly H_2 and CO_2.

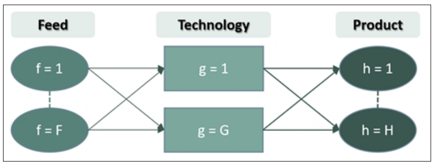

Figure 2: Generic superstructure

Figure 2 starts with feed $f \varepsilon F$. The available mass flowrate of feed is denoted as F_f while the actual mass flowrate of feed flow taken by particular technology is denoted as F_{fg}, as shown in Eq(1). The inequality sign is used in Eq(1) to provide flexibility to the model to decide how much feed is necessary (F_{fg}) based on the available feed input (F_f). Next, the product mass flowrate for particular technology (F_{gh}) can be determined based on the technology efficiency (X_{fgh}), as shown in Eq(2).

$$\mathrm{F_f} \geq \sum_{g=1}^{G} F_{fg} \tag{1}$$

$$F_{gh} = \sum_{f=1}^{F} F_{fg}\mathrm{X_{fgh}} \qquad \forall g \forall h \tag{2}$$

The energy consumption of the overall process is expressed as Eq(3). The energy consumption of each technology (E_g) is obtained by multiplying the product mass flowrate (F_{gh}) with the energy factor of each technology (W_g). Energy factors are typically expressed as the energy required per unit flow of product.

$$E_g = \sum_{h=1}^{H} F_{gh}\mathrm{W_g} \qquad \forall g \tag{3}$$

Furthermore, the annualised capital and operating cost of the process are defined in Eq(4) and Eq(5), respectively. The costs of the process can be determined based on either the inflow of raw material (F_{fg}) or the total product produced from the technology (F_{gh}). The cost factors compiled include the capital cost per unit input, capital cost per unit output, operating cost per unit input and operating cost per unit output as expressed by $\mathrm{C_g^{input}}$,$\mathrm{C_g^{product}}$,$\mathrm{O_g^{input}}$ and $\mathrm{O_g^{product}}$. The objective of minimising TAC is shown in Eq(6) where H_2 sale and CO_2 sale represent the expected profit gained from selling H_2 and captured CO_2

$$BC_g = \left(\sum_{f=1}^{F} F_{fg}\mathrm{C_g^{input}} + \sum_{h=1}^{H} F_{gh}\mathrm{C_g^{product}} \right) \times capital\ recovery\ factor(r) \qquad \forall g \tag{4}$$

$$BO_g = \sum_{f=1}^{F} F_{fg}\mathrm{O_g^{input}} + \sum_{h=1}^{H} F_{gh}\mathrm{O_g^{product}} \qquad \forall g \tag{5}$$

$$Min\ TAC = \sum_{g=1}^{G} BC_g + \sum_{g=1}^{G} BO_g - H_2\ sale - CO_2\ sale \tag{6}$$

As mentioned previously, the generic equations above serve as a guiding framework to model the interactions in Figure 1. The following section discusses a case study where the developed equations are coded in LINGO software to optimise the process shown in Figure 1.

4. Case study background

This case study focuses on recommending the optimal blue H_2 process with minimum TAC. A linear model was developed to perform process optimisation using the information below. The U.S. Energy Information Administration (2021) claims that Malaysia holds approximately 1.18 trillion cubic metre of proved natural gas reserves as of January 2020. The information on the available natural gas supply in Malaysia provides a sound way to estimate the feed flow of fossil fuel in this case study. The feed flow estimated is 10Mt/year for each raw material feed flow involved in Figure 1. Other general information is listed in Table 1.

Table 1: General information for case study

Parameter	Value	Unit
H_2 production rate (Zapantis, 2021)	1.3	kt/d
Annual operating period	351	d
Annual H_2 production rate	456.3	kt
Plant lifespan	30	y
Interest rate (Eria, 2020)	3%	-
Capital recovery factor (r)	0.51	-

In addition, some assumptions have been made throughout the case study as follows:

- 100% blue H_2 sale with consistent price in all cases
- 100% CO_2 storage and/or utilisation
- 99% purification stage in grey H_2 production

- No capacity limitation set for all technologies

In this case study, the minimum H_2 purity requirement is 40 % based on its use in the power generation sector (Thyer et al., 2009) who investigated the flame stability of H_2/CO_2 mixture with different concentration ratios and the results revealed that stable combustion is achieved starting at an H_2/CO_2 concentration ratio of 40:60. In other words, H_2 with a purity of 40 % can be safely combusted to generate energy for power generation purposes. Therefore, the lowest H_2 purity requirement to be sold to the power generation sector is set at 40% in this case study to ensure a safe power generation process using H_2. Furthermore, the performance data of technologies considered are listed in Table 2, Table 3 and Table 4.

Table 2: Performance data for grey H_2 processes, carbon capture technologies and H_2 storage

Performance data	Process efficiency (%)	Energy consumption (kWh/y)	Capital cost ($/t flow/y)	Operating cost ($/t flow)
Grey H_2 processes				
SMR	84	8.24	8.45	729
POX	90	56.2	30.6	1,632
TRIR	99	72.7	1,020,000	589
Carbon capture technologies				
Membrane separation	85	62.6	653,000	54
Selexol absorption	90	488	602,000	26.66
Rectisol absorption	85	159		26.6
MEA absorption	89	11,708	855,000	70
NH_3 absorption	95	3,186		55
CaO looping	85	486	1,068,000	40
Zeolite 13X adsorption	70	1,900		30
H_2 storage				
Liquid H_2 organic carrier	-	16 /t H_2	-	0.0678
Liquefaction	-	12,500 /t H_2	-	8
Compression	-	4,500 /t H_2	-	7.55

Table 3: Performance data for carbon transportation and storage

	CO_2 transportation			CO_2 storage		
	Pipe	Ships	Truck	Oilwell	Ocean	Saline
Operating cost ($/t CO_2)	3	1,640	0.025	0.65	14	15.2

Table 4: Performance data for CO_2 utilisation

CO_2 utilisation	Enhanced oil recovery (EOR)	Enhance gas recovery (EGR)	Coalbed methane (CBM)	MeOH industry	Building industry
Sale price ($/t CO_2)	20	10	15	300	467

5. Result and discussion

5.1 Optimal production route

The optimisation objective studied using the developed model was minimising the TAC of blue H_2 production. No constraint was defined for the target purity of H_2 in the model to provide flexibility on the technology selection. Instead, the obtained H_2 purity from the model will be analysed later to confirm that the purity is above the minimum requirement of 40 %. The results are shown in Figure 3.

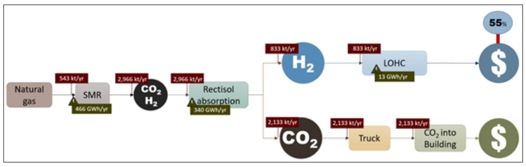

Figure 3: Preferred production route to achieve minimum TAC

Figure 3 presents the selected production path for blue H_2 production to meet the objective function of minimising production annualised cost without constraint being set on H_2 purity. Note that in grey H_2 production, only the mass flowrate of natural gas is presented, while other raw materials such as steam are not illustrated in Figure 3. This is because natural gas is the non-renewable element identified among the raw materials in SMR which will eventually contribute the carbon emissions. The H_2 yield from SMR is dependent on the feed flow of natural gas and thus only natural gas flow is presented. The results show that SMR is preferred over partial oxidation (POX) to produce grey H_2. This is explained by the lower annualised capital expenditure (CAPEX), and operating expenditure (OPEX) observed in SMR as compared to POX. The other aspects, such as H_2 yield, and energy consumption, are factored in the optimisation model however do not play a significant role in this case as the objective function only targets the annualised cost. Next, the produced grey H_2 is sent to CO_2 capture treatment by Rectisol absorption. Prior to carbon capture section, the syngas produced from SMR is subjected to purification stage with assumed efficiency of 99% to obtain a product stream with major components of CO_2 and H_2 gas only, as mentioned under Section 4. Rectisol absorption is the most favoured CO_2 capture technology due to the lowest annualised CAPEX and OPEX. Plus, the CO_2 capture efficiency of Rectisol absorption is reported to be 85 %. This will yield the blue H_2 with a purity of 55 % with the remaining impurities being the uncaptured CO_2. As discussed in the previous section, the minimum required purity of H_2 to be sold to the power generation sector is 40 %. Therefore, the blue H_2 produced (55 %) fulfils the purity requirement and is allowed to be used for power generation purposes. Next, the captured CO_2 is transported by truck as this transportation route is the most economical option as compared to pipeline and ship. Moreover, the captured CO_2 can either be stored or utilised. Since the storing of captured CO_2 will add up to the overall expenditure of the process, CO_2 utilisation appears to be an ideal option. Among all utilisation opportunities, the construction industry has the highest CO_2 sale value and thus became the most advantageous route to minimise the overall production cost of blue H_2. Lastly, the produced blue H_2 is recommended to be stored by a liquid H_2 organic carrier (LOHC), which has the lowest storing cost as compared to liquefaction and compression. In this case study, H_2 purity has no effect on the performance of H_2 storage. The total annualised production cost obtained is 88,953 M USD with a total annual energy consumption of 800GWh. The detailed cost breakdown is listed in Table 5.

Table 5: Cost breakdown for optimised process

Production path	Selected technology	Annualised cost		
		CAPEX	OPEX	Profit
Grey Hydrogen	SMR	\$ 197,000	\$ 361 M	-
CO_2 capture	Rectisol absorption	\$ 91,000 M	\$ 57 M	-
CO_2 transportation	Truck	-	\$ 53,000	-
CO_2 utilisation (sale)	Building material	-	-	\$ 955 M
H_2 storage	LOHC	-	\$ 56,000	-
H_2 sale	-	-	-	\$ 1,580 M
Total:		\$ 88,953 M		

4.2 Effect of CCUS on CO_2 emission reduction

Verification steps are conducted to benchmark the result obtained. According to the EU Commission (2022), the definition of low-carbon H_2 (blue H_2) is H_2 that is produced from non-renewable sources and meets a GHG emissions reduction threshold of 70 % as compared to grey H_2 (fossil-based H_2). Thus, a CO_2 emissions reduction analysis is carried out to study the validity of blue H_2 in each case. The analysis began with obtaining the total CO_2 production from the blue H_2 process. This was done by summing up the CO_2 production from grey H_2 as a by-product and carbon emission due to energy consumption concerning three major sections which include grey H_2 production, carbon capture and H_2 storage. On the other hand, the introduction of CO_2 emissions from CO_2 transportation and utilisation are not factored in this model. According to the U.S. Energy Information Administration (2022), the CO_2 emission factor is around 385 t of CO_2 per GWh of energy consumed. Thus, the CO_2 produced due to energy consumption can be determined by multiplying the total energy consumption of the process with the reported CO_2 emission factor. In this case study, CO_2 production from grey H_2 will be sent for CO_2 capture treatment while no further treatment is available for the CO_2 production from energy consumption. Since the implementation of CCUS is the key element to retrofit the grey H_2 into blue H_2 process, the CO_2 emission reduction is calculated using Eq(7). The results are listed in Table 6.

$$CO_2\ emission\ reduction = \frac{Annual\ CO_2\ capture}{Total\ annual\ CO_2\ emission} \times 100\ \% \qquad (7)$$

Table 6: CO_2 emission reduction result

	From grey H_2 production	From energy consumption	Total CO_2 emissions	CO_2 capture rate	CO_2 emissions reduction
CO_2 flow (kt/y)	2,510	308	2,818	2,133	76 %

The outcome in Table 6 concludes that implementation of CCUS has effectively reduced primary greenhouse gas (CO_2) emission to a good level (>70 %). Also, this achievement has direct effect on relieving the pressure and stress of global warming caused by the massive growth of CO_2 emissions in H_2 generation sector.

6.0 Conclusion

In conclusion, this research covers the implementation of CCUS into grey H_2 production to produce blue H_2. An optimisation-based mathematical model was developed to determine the ideal blue H_2 process with minimum cost, and energy consumption. The case study has shown that a minimum annualised cost of 88,953 M USD can be achieved with H_2 purity of 55 %. There are limitations in this work that serve as motivation for future work. Firstly, carbon capture was not included for carbon emission due to energy consumption. Besides, the emissions generated from CO_2 transportation and utilisation was not considered for this work. In future work, the proposed model can be improved by performing CO_2 capture on the CO_2 emissions due to energy consumption, transportation and utilisation. Moreover, detailed information concerning the products' market value and demand could be determined accordingly to the interested country so that the process TAC can be resolved more precisely. Lastly, technology limitations such as maximum capacity can be factored in to analyse the system's flexibility.

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