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# Why the Energy Transition in the Downstream Industry Pass Through Renewable Hydrogen?

Dr. Marcio Wagner da Silva

## INTRODUCTION AND CONTEXT

Demand for hydrogen raised strongly in the last decades following the necessity of hydrotreatment units installations in refineries to comply with the pressure to reduce the content of contaminants like sulfur and nitrogen in the petroleum derivatives and consequently minimizing the environmental impact caused by fuels burn.

Given the greater offer of natural gas in the last years, the hydrogen generation process through Methane (main natural gas component) reforming reactions has consolidated like the principal route to produce hydrogen and syngas to the production of the most diversified chemical products like ammonia and conversion of methanol to olefins (MTO processes), etc.

Regarding the hydrogen, this compound became a fundamental enabler to the crude oil refining chain. Due to the increasingly

necessity to reduce the environmental impact of the crude oil derivatives, it's practically impossible to produce marketable crude oil derivatives without at least one hydroprocessing step, raising the hydrogen demand as aforementioned. In this sense, even to efforts related to energy transition by the downstream industry, the hydrogen presents a key role.

As aforementioned, hydrogen is a key enabler to the future of the downstream industry and the development of renewable sources of hydrogen is fundamental to the success of the efforts to the energy transition to a lower carbon profile. According to the Wood Mackenzie data the green and blue hydrogen generation routes tends to take-off in the next years, these data are presented in Figure 1.

In the current scenario, the best alternative to refiners is optimize the hydrogen consumption minimizing the operating costs and CO<sub>2</sub> emissions.

## Green hydrogen capacity takes off post-2030 (LHS) and starts to penetrate hard-to-abate sectors

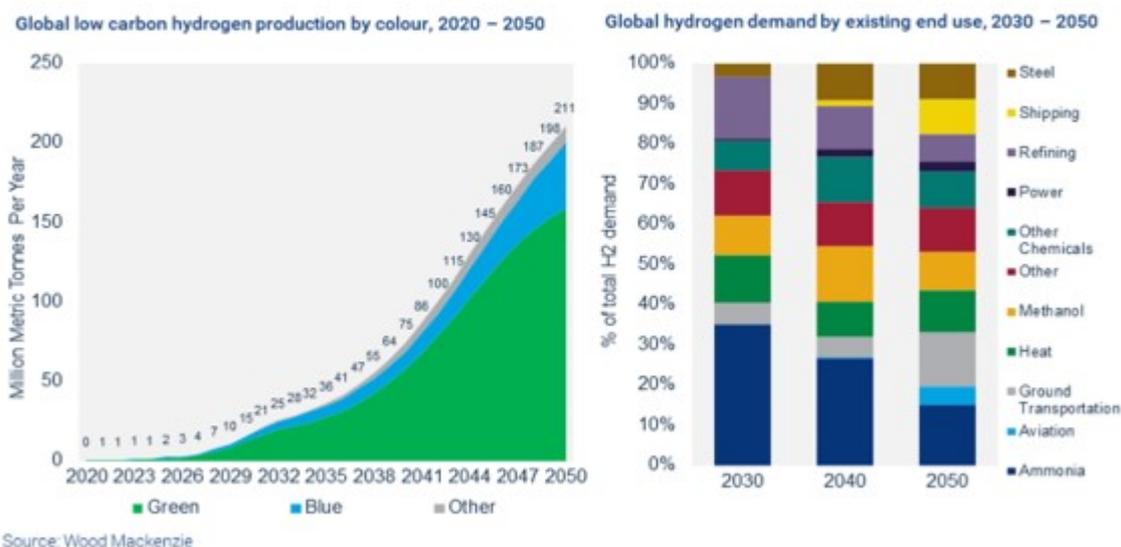
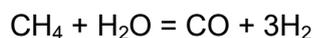


Figure 1 – Evolution of the Cleaner Hydrogen Production Routes (Wood Mackenzie, 2020)

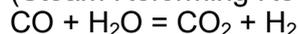
## HYDROGEN AND SYNGAS PRODUCTION ROUTES - INTRODUCTION

Methane steam reforming is currently the most employed technology to produce syngas and hydrogen, moreover, it's presented at the moment as the most economical route to produce hydrogen in large scale.

The methane steam reforming process can be chemically represented like presented below:



(Steam Reforming Reaction - Endothermic)



(Shift Reaction - Exothermic)

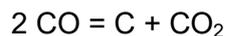
The reforming reaction is endothermic, so is favored by higher temperatures (700 – 850 °C) and the catalyst commonly employed is a catalyst with a high content of nickel (Ni) over alumina (Al<sub>2</sub>O<sub>3</sub>). The reaction equilibrium is favored by lower pressures, however, to avoid the necessity of produced gasses compression the reactions are conducted under moderate pressures (15 – 25 bar).

Shift reaction is slightly exothermic and occurs under mild reaction conditions (200 – 350 °C) over iron oxide catalyst promoted with cobalt and copper.

Figure 2 shows a basic arrangement for process unit dedicated to producing hydrogen by Methane steam reforming.

Once the nickel catalyst is strongly sensitive to contaminants like sulfur that can cause his deactivation, in the process have a treatment step dedicated to removing these contaminants from methane stream.

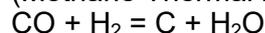
Some undesired reactions can occur during the methane steam reforming process conducting the coke deposition over the catalyst leading to loss of chemical activity or a complete deactivation, as described below:



(Boudouard Reaction)



(Methane Thermal Decomposition)



(CO Reduction)

To minimize the risk of carbon deposits, the process is conducted with higher steam/hydrocarbon ratio (3 – 4 mole of H<sub>2</sub>O per carbon mole), however, the steam/hydrocarbon ratio can't be much high because can lead to excessive dimensions of the process equipment and the lower H<sub>2</sub>O/hydrocarbon ratio can be compensated by temperature rising. Another side effect of the rise of H<sub>2</sub>O/hydrocarbon ratio is the CO reduction that changes the CO/H<sub>2</sub> ratio. Figure 3 presents a basic process flow diagram for a hydrogen generation unit through natural gas steam reforming by Haldor Topsoe Company.

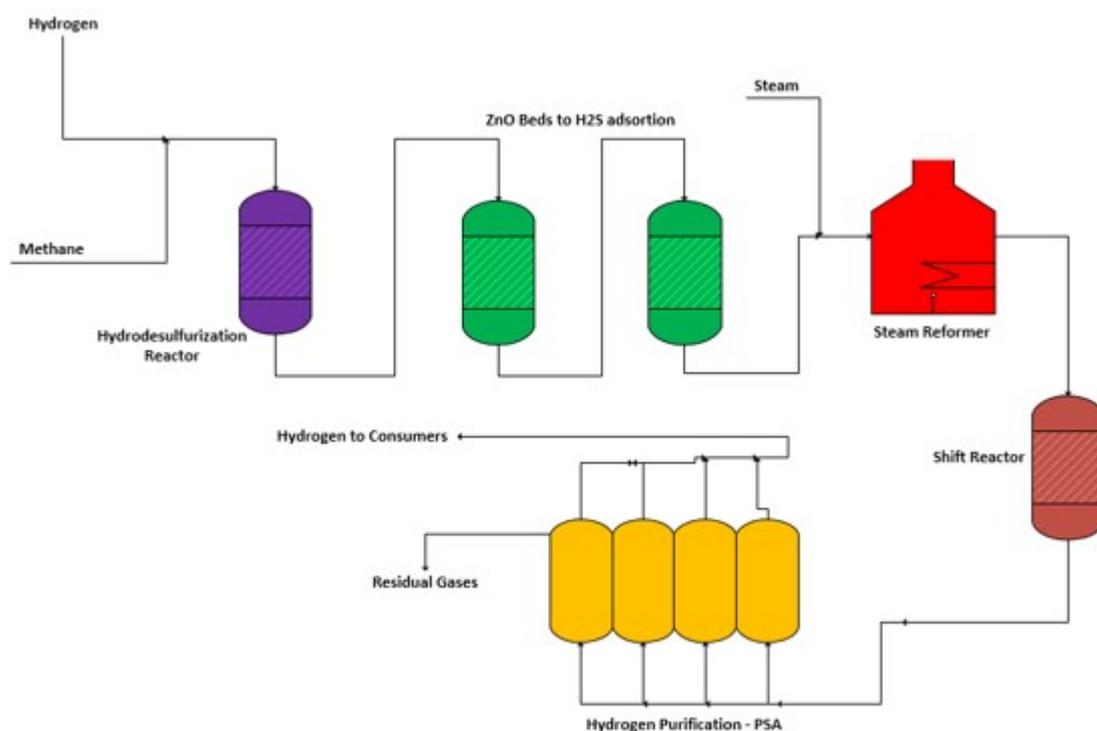


Figure 2- Basic Process Flow Scheme for Methane Steam Reforming Process

Another much-studied technology aiming to the hydrogen production is called methane dry reforming, the principal chemical reaction of the dry reforming process is presented below:

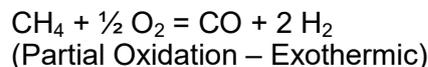


Methane dry reforming reaction is endothermic and conducted under high temperatures (higher than 700 °C) over a nickel-based catalyst. The dry reforming production route is attractive from the environment point of view because can minimize the water consumption and the main reagent is a combustion sub-product that is partially responsible for the greenhouse effect. Another point in favor of dry reforming technology is that the syngas from this process had the ratio  $\text{H}_2/\text{CO} = 1$ , this characteristic is ideal for producing oxygenated compounds as acetic acid and dimethyl ether.

Meanwhile, the main challenge to be overcome to the development of the dry reforming technology is the strongly tendency to coke deposition over the catalysts. Due to the non-existence of water in the process and the low H/C ratio in the process feed, the coke formation over the catalysts applied in the dry reforming process is much severe.

Several researchers had studied ways to develop more resistant catalysts against deactivation by carbon deposition to provide practical application for methane dry reforming technology. Another point to be considered in the dry reforming process development is the necessity of  $\text{CO}_2$  purification that can contribute in a negative form to the economic viability of the process when compared with the steam reforming process.

Methane Partial Catalytic Oxidation Process shows attractive once produces syngas with  $\text{H}_2/\text{CO} = 2$ , that's ideal for Fischer-Tropsch synthesis. The main chemical reaction in the methane partial catalytic oxidation process is presented below:



This technology applies nickel based catalysts, however, some research was developed using platinum, palladium, rhodium and ruthenium as active metals, despite the higher cost of these metals when compared with the nickel.

The presence of oxygen in the process reduces the carbon deposit formation over the catalyst, but the use of pure oxygen is not economically competitive because needs the installation of cryogenic separation unit to separate the oxygen from air, on the other hand, using air in the process would increase the equipment dimensions due to the  $\text{N}_2$  presence. Another problem associated with methane partial oxidation is related to the safety owing to the process feed is a mixture of  $\text{CH}_4$  and  $\text{O}_2$  that can present explosion risk under some process conditions.

Another promising technology to produce syngas and hydrogen from methane is called Autothermal Reforming Process. This technology is target of several studies and is basically a combination of partial oxidation and steam reforming processes, the principal reactions involved in this process are showed below:

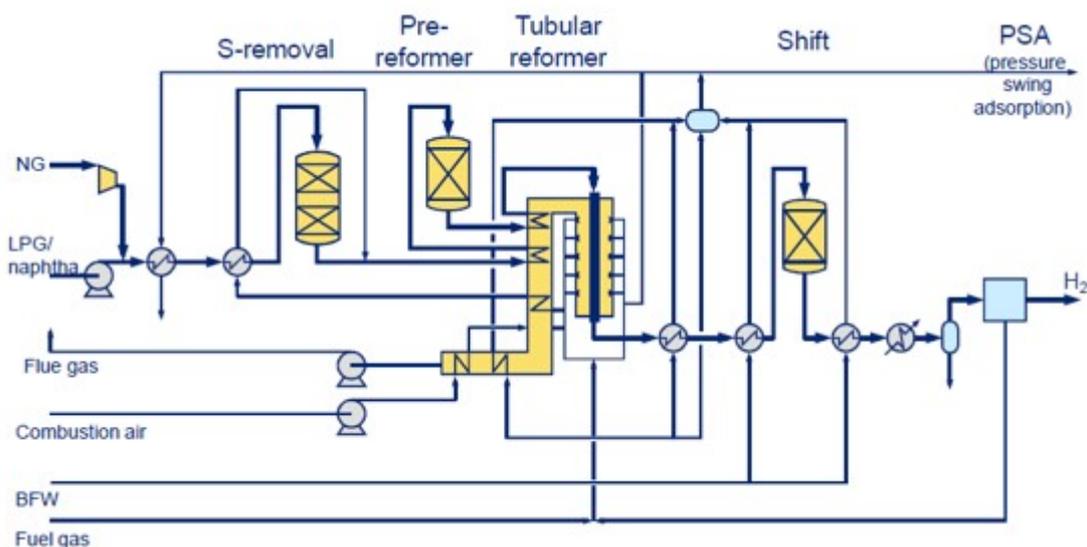
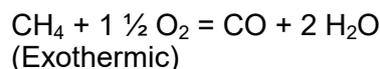


Figure 3 – Hydrogen Generation Process by Haldor Topsoe Company (PEIRETTI, 2013)

$\text{CH}_4 + \text{H}_2\text{O} = \text{CO} + 3 \text{H}_2$   
(Steam Reforming Reaction – Endothermic)

$\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$   
(Shift Reaction – Exothermic)

The process is called autothermal because involves endothermic and exothermic reactions and, in theory, the heat produced in the partial oxidation reaction is consumed in the reforming reaction.

Autothermal reforming can produce syngas with  $\text{H}_2/\text{CO}$  ratio close to 2, adjusting the reactants proportion in the process feed. Normally the catalyst used in the process is nickel-based catalyst over alumina, however, coke deposition over the catalyst represents a challenge that needs to be overcome in the methane autothermal reforming too.

Like aforementioned, several technologies have been studied and developed, but at the moment methane steam reforming process still is the most economical route to produce hydrogen in large scale.

### **RENEWABLE HYDROGEN GENERATION ROUTES – FUNDAMENTAL ENABLER TO THE ENERGY TRANSITION**

In the current scenario, is increasingly high the pressure from the society to energy transition efforts aiming to reduce the fossil fuels participation in the global energetic matrix. As aforementioned, the hydroprocessing technologies achieve a fundamental role in any refining hardware, but this fact has a side effect due to the high  $\text{CO}_2$  emissions during the natural gas steam reforming process to produce hydrogen.

Some refiners are adopting the co-processing of renewable materials in the crude oil refineries aiming to produce high quality and cleaner transportation fuels. Despite the advantages of environmental footprint reduction of the refining industry operations, renewables processing presents some technological challenges to refiners.

The renewable streams have a great number of unsaturations and oxygen in his molecules which lead to high heat release rates and high hydrogen consumption, this fact leads to the necessity of higher capacity of heat removal from hydrotreating reactors aiming to avoid damage to the catalysts. The main chemical reactions associated with the renewable streams hydrotreating process can be

represented as below:

$\text{R-CH=CH}_2 + \text{H}_2 \rightarrow \text{R-CH}_2\text{-CH}_3$   
(Olefins Saturation)

$\text{R-OH} + \text{H}_2 \rightarrow \text{R-H} + \text{H}_2\text{O}$   
(Hydrodeoxygenation)

Where R represents a hydrocarbon.

These characteristics lead to the necessity of higher hydrogen production capacity by the refiners as well as quenching systems of hydrotreating reactors more robust or, in some cases, the reduction of processing capacity to absorb the renewable streams. In this point it's important to consider a viability analysis related to the use of renewables in the crude oil refineries once the higher necessity of hydrogen generation implies in higher  $\text{CO}_2$  emissions through the natural gas reforming process that is the most applied process to produce hydrogen in commercial scale.

$\text{CH}_4 + \text{H}_2\text{O} = \text{CO} + 3\text{H}_2$   
(Steam Reforming Reaction - Endothermic)

$\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$   
(Shift Reaction - Exothermic)

This fact leads some technology licensors to dedicate his efforts to look for alternative routes for hydrogen production in large scale in a more sustainable manner. Some alternatives pointed can offer promising advantages:

- Natural Gas Steam Reforming with Carbon Capture – The carbon capture technology and cost can be limiting factor among refiners;
- Natural Gas Steam Reforming applying biogas – The main difficult in this alternative is a reliable source of biogas as well as their cost.;
- Reverse water gas shift reaction ( $\text{CO}_2 = \text{H}_2 + \text{CO}$ ) – One of the most attractive technologies, mainly to produce renewable syngas;
- Electrolysis – The technology is one of the more promising to the near future.

Figure 4 presents a processing scheme to produce hydrocarbons applying renewable hydrogen, based in the Roland Berger Company concept.

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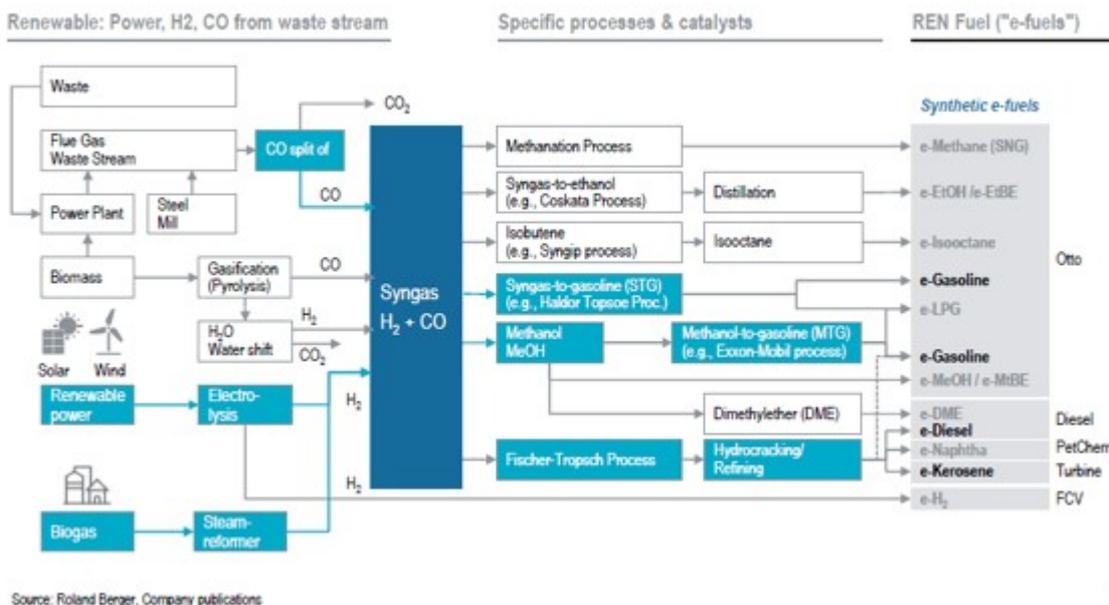


Figure 4 – Hydrocarbons Production Routes Applying Renewable Hydrogen (Roland Berger Company, 2020).

As aforementioned, hydrogen is a key enabler to the future of the downstream industry and the development of renewable sources of hydrogen is fundamental to the success of the efforts to the energy transition to a lower carbon profile.

Recently, the literature is classifying the hydrogen production routes in four classes as follow (Based on IEA data from 2019):

1 – Brown Route – Hydrogen production from coal gasification without carbon abatement system (CCS). This route presents the higher emission rate of greenhouse gases (19 t CO<sub>2</sub>/t H<sub>2</sub>) and an average production cost of US\$ 1,2 to 2,1 per kg H<sub>2</sub>;

2 – Gray Route – This is the conventional hydrogen production route adopted by the most part of the refiners, which applies steam reforming of natural gas without CCS. This route still presents high emission of greenhouse gases (11 t CO<sub>2</sub>/t H<sub>2</sub>) and an average production cost of US\$ 1,0 to 2,1 per kg H<sub>2</sub>;

3 – Blue Route – This route encompasses the conventional steam reforming of natural gas with CO<sub>2</sub> abatement system. In this case, the CO<sub>2</sub> emissions are drastically reduced (0,2 t CO<sub>2</sub>/t H<sub>2</sub>), but the average production cost reaches US\$ 1,5 to 2,9 per kg H<sub>2</sub>;

4 – Green Route – As presented above, the green route is based on electrolysis through renewable electricity. In this case it's possible to reach zero CO<sub>2</sub> emissions, but the average

production cost is still considered high (US\$ 3,0 to 7,5 per kg H<sub>2</sub>).

The technology development and scale-up gains tends to reduce the production costs of cleaner routes over the next years. Currently, the best alternative to refiners is to optimize the hydrogen consumption to keep under control the operating costs as well as, control the emissions of greenhouse gases.

Nowadays, as presented in Figure 5, the crude oil refining industry is the main hydrogen consumer followed by the ammonia production.

Still based on data from Figure 6, 71 % of the hydrogen produced by dedicated processes is from natural gas steam reforming and 27 % from coal gasification, both routes present high emissions of greenhouse gases (mainly CO<sub>2</sub>). According to the reference, the difference (close to 75 Mt of hydrogen) is related to the generation where the hydrogen is produced as a by-product like naphtha catalytic reforming or propane steam cracking as example. Crossing the data from Figure 6, it's clear the relevance of the necessity of the energy transition efforts in the downstream sector to the success of the global transition to a low carbon and hydrogen economy.

#### THE GAS TO LIQUIDS TECHNOLOGIES – SYNGAS PRODUCTION

One of the most promising and well-developed technologies to produce liquid hydrocarbons from natural gas currently is the

## Today's production of hydrogen is via carbon-intensive processes, with use of hydrogen concentrated in the refining, ammonia, and methanol sectors

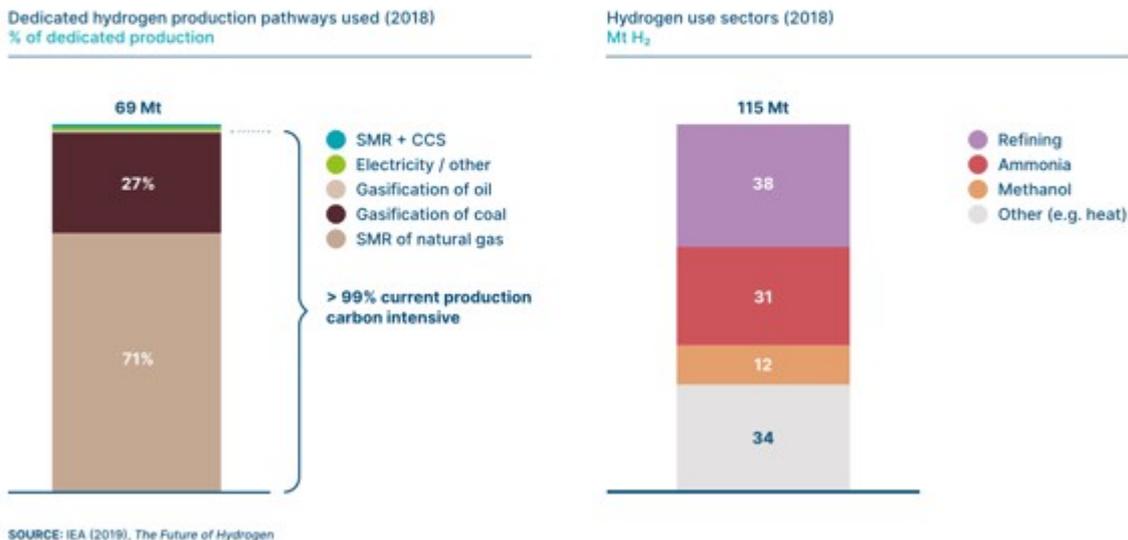


Figure 5 – Main Production Routes and Hydrogen Consumers (ETC Global Hydrogen Report, 2021)

conversion of syngas ( $\text{CO} + \text{H}_2$ ) in longer-chain hydrocarbons such as gasoline and other liquid fuel products, known as Gas to Liquids Technologies (GTL). The liquid hydrocarbons production can be carried out by direct syngas conversion, in Fischer-Tropsch synthesis reactions or through methanol production as intermediate product (Methanol to Olefins technologies).

Shown process in Figure 6 is based in the syngas gas generation from steam reforming of natural gas, this is the most common route, however, there are process variations applying syngas production through coal, biomass, or petroleum coke gasification route.

The process starts with syngas generation and, as aforementioned, the produced hydrocarbon chain extension is controlled in the Fischer-Tropsch synthesis step through the  $\text{CO}/\text{H}_2$  ratio in the syngas fed to the FT reactors (beyond temperature and reaction pressure), following the produced hydrocarbons are separated and sent to refining steps as isomerization, hydrotreating, hydrocracking, catalytic reforming, etc. According to application of the produced derivative (Gasoline, Diesel, Lubricant, etc.).

Some side reactions can occur during the hydrocarbons production process, leading to coke deposition on the catalyst, causing

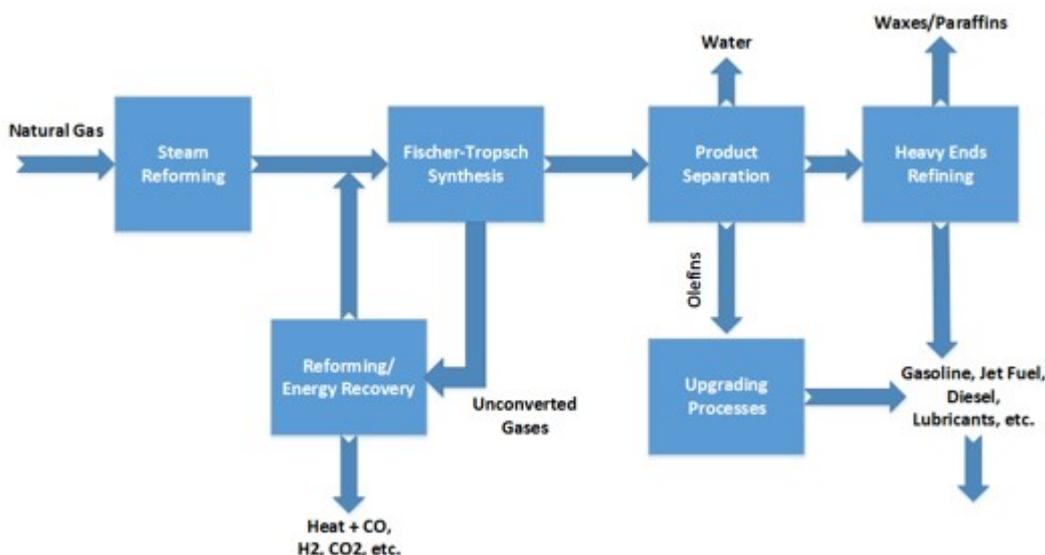
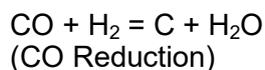


Figure 6 – Block Diagram to a Typical Fischer-Tropsch GTL Process Plant.

his deactivation according to following chemical reactions:



The type of reactor applied in the FT synthesis step have strong influence on the yield and quality of the obtained products, the campaign time of the process units also depends on the type of reactor. Fixed bed reactors are widely employed to FT synthesis, however, show a reduced campaign time due to the low resistance to catalyst deactivation phenomenon. Modern process units apply fluidized bed or slurry phase reactors that present a higher resistance to coke deposition on the catalyst and better heat distribution, leading to higher campaign periods.

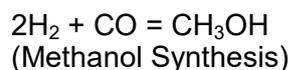
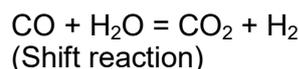
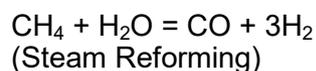
Most recently is observed a reduction trend in the demand by transportation fuels and some refiners are looking for change his production focus from transportation fuels to petrochemicals. The gas to liquids can be applied in synergy with conventional refining processes to improve the yield of petrochemicals in the refining hardware through the production of high quality naphtha that can be applied to FCC or steam cracking units to produce light olefins, ensuring higher added value to the processed crudes and gas as well as participation in a growing market.

Another attractive alternative and synergy opportunities to refiners is the production of ammonia that are the base of any fertilizer.

Despite the flat demand over the last years, is expected a growing market in the next years due to the increasingly demand by food at global level. As presented in Figure 7, is also expected a growing demand of Methanol in the next years, this intermediate can be used to produce high demand products like formaldehyde that is applied to produce plastics and coatings, allowing great added value to the crude oil and natural producing chain.

### METHANOL TO OLEFINS/GASOLINE TECHNOLOGIES – LIQUID HYDROCARBONS FROM METHANOL

Another alternative route to produce liquid hydrocarbons from syngas is the non-catalytic conversion of the natural gas to methanol followed by the polymerization to produce alkenes. Methanol is produced from natural gas according to the following chemical reactions:



In the sequence, the methanol is dehydrated to produce Dimethyl-Ether which is posteriorly dehydrated to produce hydrocarbons, as shown in the sequence:

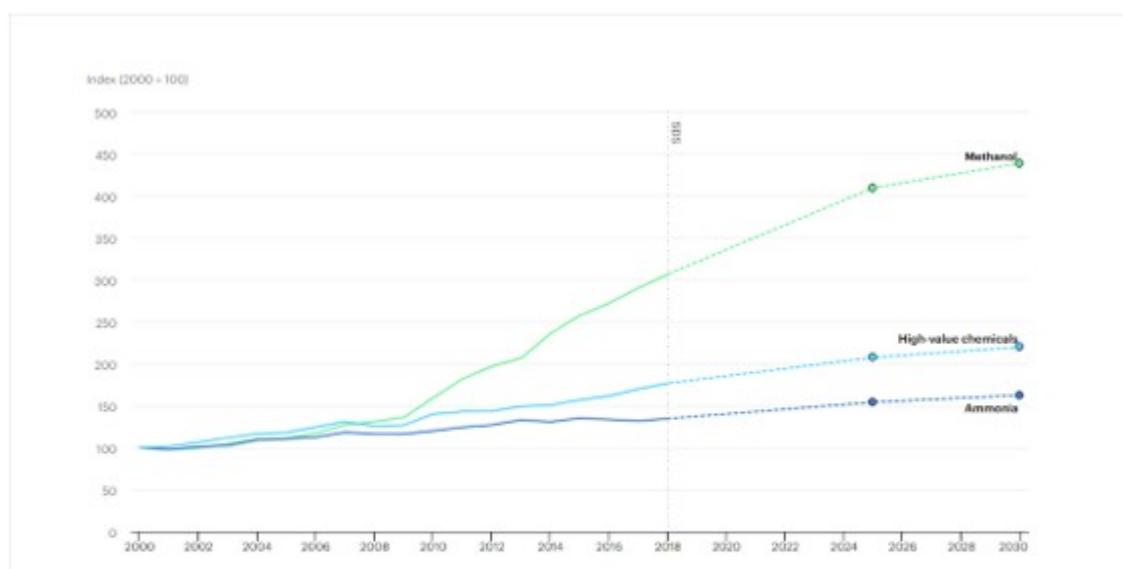
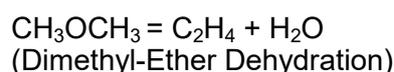
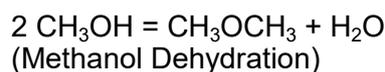


Figure 7 – Primary Chemicals Production Forecast (IEA, 2020)

The methanol conversion to olefins into hydrocarbons is called Methanol to Olefins (MTO) or Methanol to Gasoline (MTG) technologies. Figure 8 presents a typical unit dedicated to produce methanol from natural gas through the two-step reforming process.

reforming step that offer great scale economy when compared with traditional production processes (One step and two-step reforming processes). Figure 9 presents a basic process flow diagram for the ATR™ process, developed by Haldor Topsoe Company.

An alternative technology developed by the Haldor Topsoe Company to produce methanol from natural gas is the Autothermal reforming process, called ATR™ that offers improvements related to the reforming furnace. A significant advantage of the ATR™ process is the lower required ratio Steam/Carbon in the

The most known processes dedicated to converting methanol in hydrocarbons are the processes MTG™ developed by ExxonMobil Company and the MTO-Hydro™ process, developed by UOP Company. Figure 10 presents the process flow diagram for the MTG™ process by ExxonMobil Company.

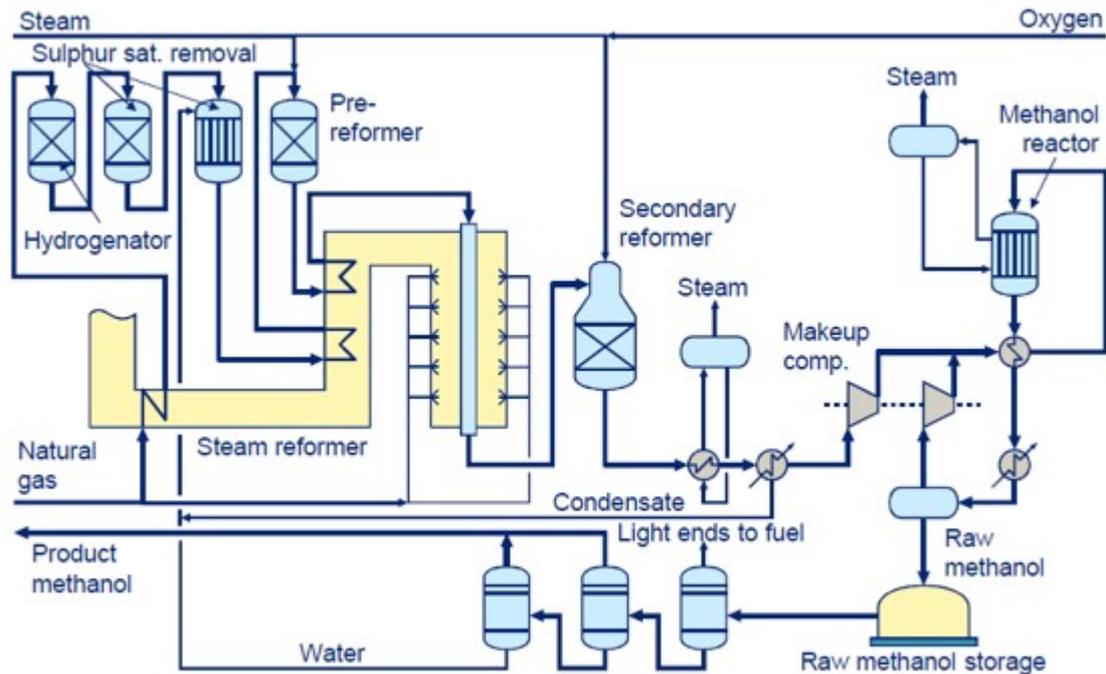


Figure 8 – Methanol Production Process from Natural Gas through two-reforming Process (PEIRETTI, 2013)

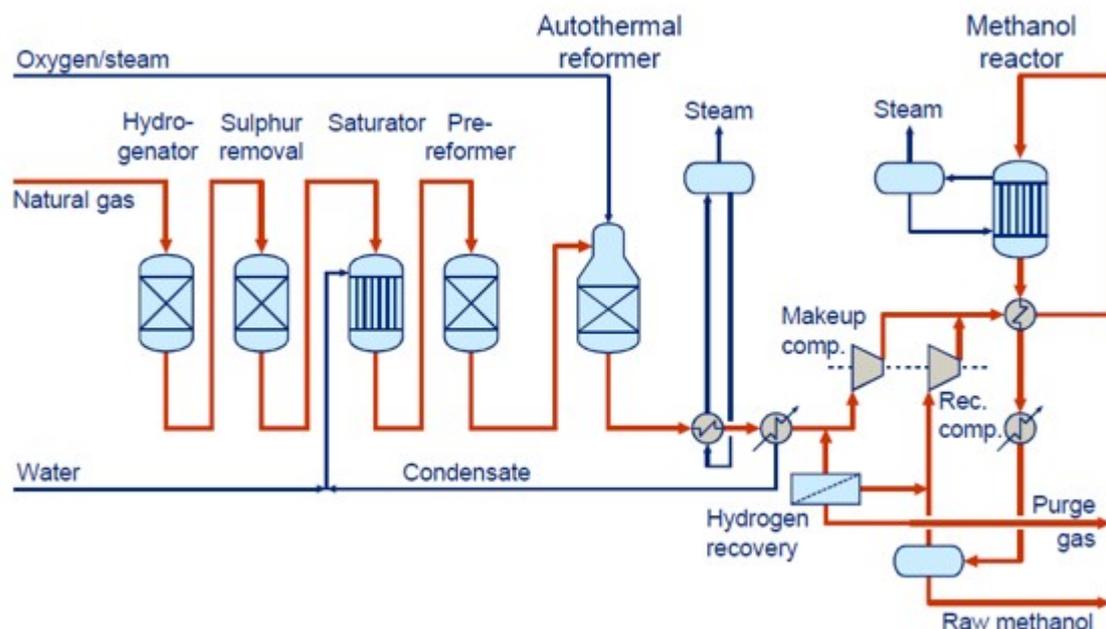


Figure 9 – Autothermal (ATR™) Process for Methanol Production from Natural Gas by Haldor Topsoe Company (PEIRETTI, 2013)

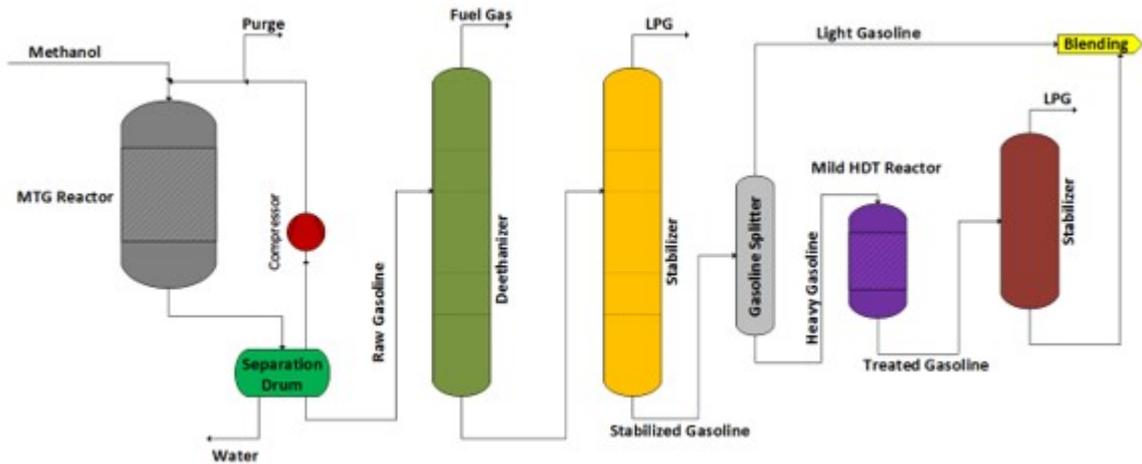


Figure 10 – Process Flow Diagram for MTG™ Technology by ExxonMobil

The MTO technologies presents some advantages in relation to Fischer-Tropsch processes, once show higher selectivity in the hydrocarbon production, furthermore, the obtained products require lower additional processing steps to achieve commercial specifications, another important point in that the installation cost is normally lower to MTO process plants when compared with FT units, once Fischer-Tropsch units are economically viable only in large scale. Regarding the olefins production, the maximization of these derivatives can be especially attractive in the current scenario where there is a trend of reduction in transportation fuels demand followed by the growing market of petrochemicals, creating the necessity of closer integration between refining and petrochemical assets aiming to maximize the added value, share risks and costs, as well as ensure market share in a highly competitive scenario of

the downstream sector. Other great technology developers for methanol production process are Johnson Matthey Company, Linde Company, Chiyoda Corporation, and Jacobs Company.

#### **HYDROGEN NETWORK AND MANAGEMENT ACTIONS IN CRUDE OIL REFINERIES**

As mentioned above, the hydrogen became a fundamental production input to modern crude oil refineries and his adequate management is a key factor to ensure controlled operating costs and competitiveness in the market, as well as allow the production of marketable crude oil derivatives. The hydrogen management actions start with a mass balance involving the hydrogen network that is composed by hydrogen sources, hydrogen purification systems, and the hydrogen consumers as presented in Figure 11.

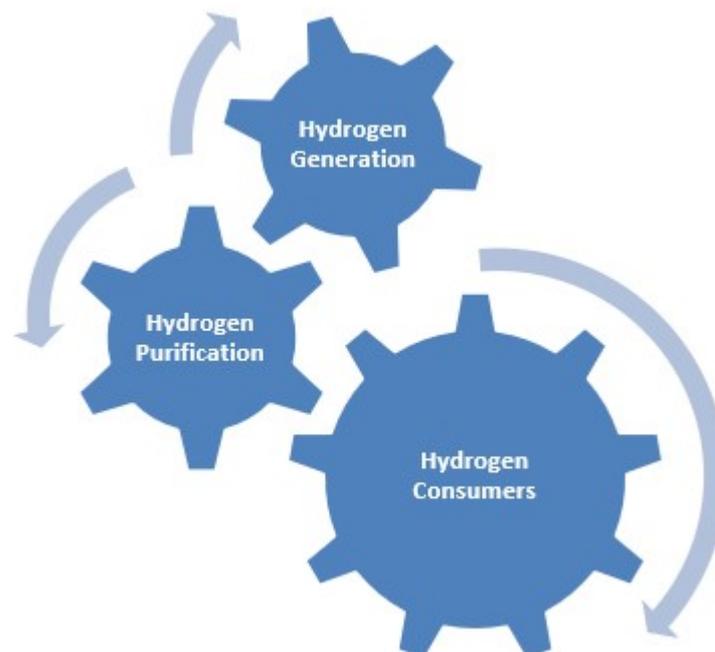


Figure 11 – A Typical Hydrogen Network in a Crude Oil Refinery

The hydrogen generation relies on the refining configuration adopted in the refinery. Normally, refineries that rely on Catalytic Reforming units apply the hydrogen produced in this process unit to compose a relevant part of the hydrogen network becoming an important internal source of hydrogen. As presented above the hydrogen generation route most applied in the refining industry is the steam reforming based on naphtha or natural gas like described by Figure 2.

The hydrogen purifying technologies is another important part of hydrogen network, normally the modern refineries apply Pressure Swing Adsorption (PSA) technologies to purify the hydrogen, reaching purity higher than 99 %. Despite this fact some refiners still use treatments based on amine treatment, as depicted in Figure 12.

Despite the lower capital cost requirement when compared with PSA technologies, the amine treating units produce hydrogen with low purity and this represent great disadvantage, especially to refiners with deep conversion hydroprocessing units. Another hydrogen purifying technologies commercially available are the membrane separations that can reach purity of 98 % and the cryogenic processes that can reach 96 % of purity. The hydrogen purifiers have a key role in the hydrogen management once control the hydrogen recovery in off-gases, one of the main sources of hydrogen losses in the refineries is the burn as fuel gas during poor recovery capacity.

The hydrogen consumers in a crude oil refinery are composed by hydroprocessing units that can be hydrotreating and hydrocracking units, as presented in Figure 13.

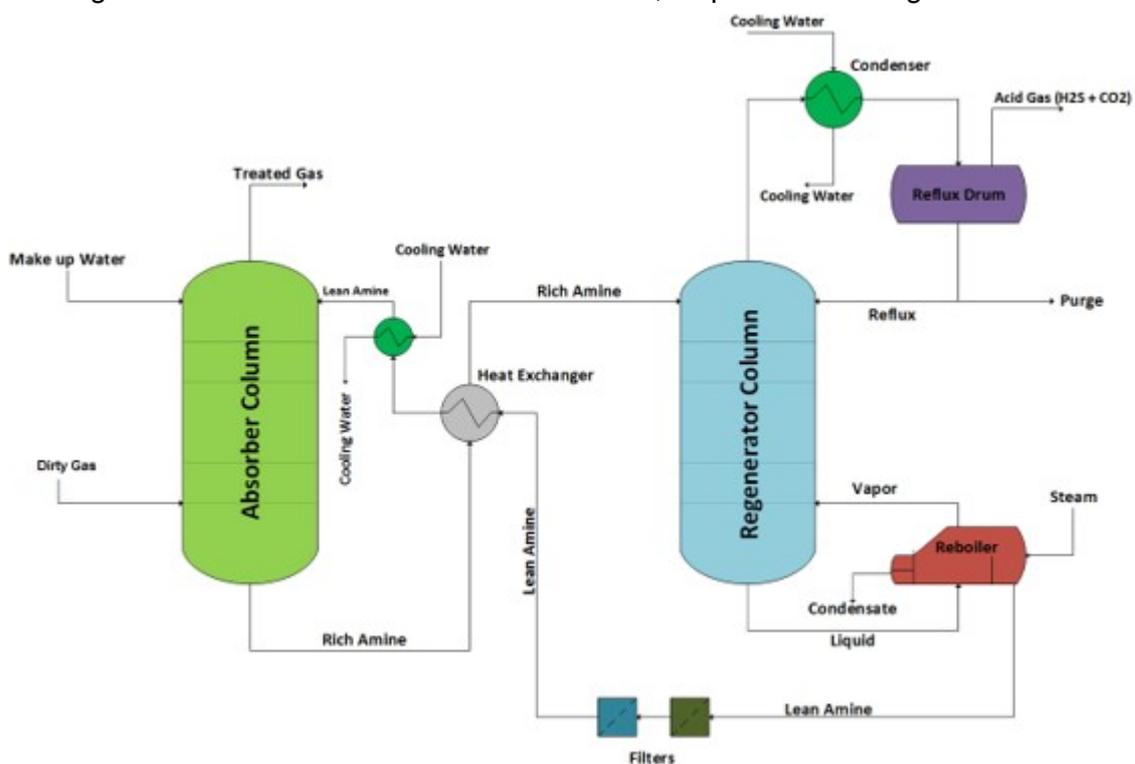


Figure 12 – Typical Amine Treating Unit

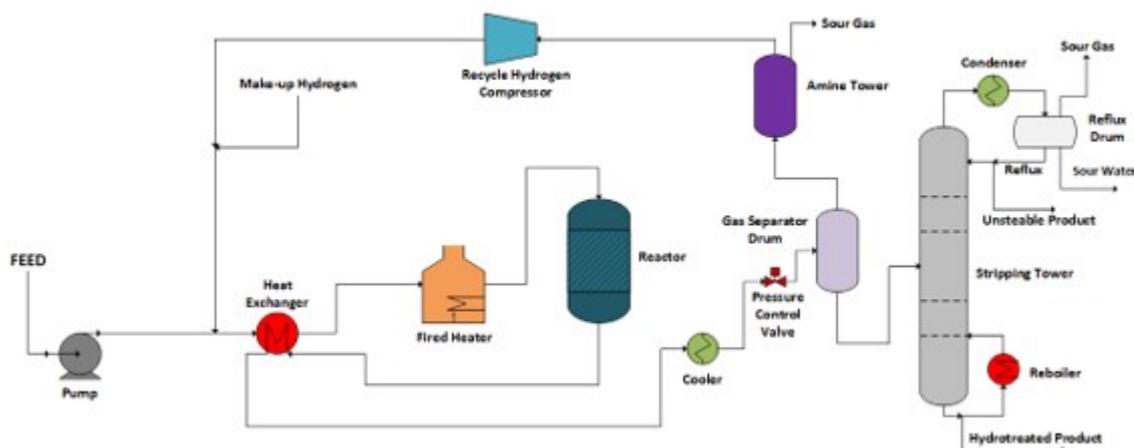


Figure 13 – Typical Low Severity Hydrotreating Unit

According to the refining configuration, the hydrogen consumers in a crude oil refinery can vary, an example is the refineries that rely on isomerization units to increase the production of high-quality gasoline.

The high cost of hydrogen generation as well as the great amount of CO<sub>2</sub> (Greenhouse gas) produced is the main driving force to an adequate hydrogen management in the refining hardware. Process integration technologies like pinch method and mathematical modeling are being applied to reach a most rational use of hydrogen in the refining hardware.

The reliability of hydrogen purification systems as well as the optimization of hydroprocessing units is fundamental to avoid the burn of hydrogen in the fuel gas ring or flare that can raise the operating costs and reduce the refining margins of the refiners. Another key point is the availability of control and instrumentation systems to allow the flow measurement and adequate accuracy of mass balances and actions to define optimization actions and mathematical modeling.

## CONCLUSION

The hydroprocessing technologies became fundamental to the downstream industry both to produce high quality and cleaner derivatives or to prepare feedstocks to the processing units like residue fluid catalytic cracking and this dependence raised, even more after the start of IMO 2020 that requires a deep treatment of bottom barrel streams aiming to comply with the new quality requirements of the marine fuel oil (Bunker). In this sense, the hydrogen generation units achieve strategic character to refiners and the efficient and reliable operation of these units needs to be a priority to refiners.

Nowadays, the cleaner routes of hydrogen production still present higher costs of the traditional routes, but the pressure from the society tends to create stricter regulations forcing the refiners to adopt these routes, especially the blue route in short term, this scenario call the refiners to a more efficient and optimized use of hydrogen in the refining and petrochemical assets. Under this environment, the petrochemical integration necessity of the refining hardware tends to be reinforced once, normally, the petrochemical plants present a surplus of hydrogen which can be offered to integrated refiners with relatively low cost and without new contributions of CO<sub>2</sub> emissions.

Beyond the current status, it's important to understand that the energy transition is no

longer a choice matter to the players of the downstream industry, but a reality, and the efforts to find cleaner sources of hydrogen needs to be supported to refiners aiming to minimize the environmental impact of crude oil processing chain at the same time to ensure the production of high quality and added value derivatives.

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## AUTHOR



Dr. Marcio Wagner da Silva is Process Engineer and Stockpiling Manager focusing on Crude Oil Refining Industry based in São José dos Campos, Brazil. Bachelor's in chemical engineering from University of Maringa (UEM), Brazil and PhD. in Chemical Engineering from University of Campinas (UNICAMP), Brazil. Has extensive experience in research, design and construction to oil and gas industry including developing and coordinating projects to operational improvements and debottlenecking to bottom barrel units, moreover Dr. Marcio Wagner have MBA in Project Management from Federal University of Rio de Janeiro (UFRJ), in Digital Transformation at PUC/RS, and is certified in Business from Getulio Vargas Foundation (FGV).



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