

TECHNOLOGIES FOR CONVERTING CO₂ AND CO₂-RICH NATURAL GAS SOURCES AND THEIR POTENTIAL APPLICATIONS IN VIETNAM

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ABSTRACT:

The greenhouse effect is one of the most concerning issues for humanity, mainly caused by the excessive emission of CO₂ into the environment from industrial production activities. For climate change mitigation, it is significant to capture CO₂ and transform it into chemicals and materials. As a result, various technologies for CO₂ conversion have been developed. Vietnam is one of the countries heavily affected by climate change and the country has great potential for applying CO₂ conversion technologies for resolving emissions by industrial activities or utilizing CO₂-rich natural gas sources.

Keywords: CO₂ emissions, natural gas with high CO₂ content, renewable resources, CCUS.

1. Overview of CO₂ and energy transition trends

For over 150 years since the world began the industrial era, due to increased production activities and energy demand, the concentration of CO₂ has risen by over 50% [1]. According to research from the Earth System Science Data, the concentration of CO₂ in the Earth's atmosphere reached 417.2 ppm at the end of 2022 [2]. In the latest analysis by the International Energy Agency, the amount of CO₂ in 2022 is expected to increase by 300 million tons, bringing the total amount of CO₂ in the atmosphere to 33.8 billion tons [3].

The world is beginning a trend of the energy transition, reducing the proportion of fossil fuel use

and increasing the use of renewable energy such as solar energy, wind energy, wave energy, etc. The latest development is the use of green hydrogen energy to serve the goal of carbon neutrality by 2050. Current trends in energy transition are:

- Change of primary energy sources: coal → oil → natural gas → renewable energy.
- Change of final energy uses: coal → oil → natural gas → electricity → hydro energy.
- Other trends: optimizing and conserving energy sources, recycling waste, using biofuels, CCU and CCUS technology, etc.

The use of CO₂ as a new raw material provides humans with many opportunities to reduce emissions or even diversify the energy supply for

production activities, although this is still facing many difficulties and challenges.

Regarding opportunities, using CO₂ helps to reduce emissions and aims to lower the concentration of CO₂ in the air to an acceptable level. It can be transformed into many high-value products, making it a key link in sustainable development and a renewable resource, promoting the development of green energy technology.

On the other hand, the biggest challenge in utilizing CO₂ is its thermodynamic properties. Carbon dioxide is a relatively inert compound with a low heat of formation of -393.5 kJ/mol and hybridization, making it difficult to undergo chemical reactions under normal conditions.

2. The issue of CO₂ in Vietnam and Vietnam's advantages in utilizing CO₂

According to the World Bank statistics, in 2019, on average, each person in Vietnam emitted 3.5 tons of CO₂ into the atmosphere, equivalent to a total of 336.7 million tons of CO₂ emissions. Recognizing this issue, Vietnam has committed to reducing CO₂ emissions to zero by 2050 at COP26. In Vietnam's NDC in 2022, Vietnam commits to an unconditional reduction of 15.4% in CO₂ emissions and a conditional reduction of 43.5% with international support by 2030 [4].

In the Global Climate Risk Index report in 2019, Vietnam is one of the top 10 countries most affected by climate change. From 1951 to 2000, the average temperature in Vietnam increased by 0.7 degrees Celsius and sea levels rose over 20 cm compared to the year 1965. Therefore, converting CO₂ gas and CO₂-rich natural gas is necessary to benefit the environment.

It can be seen that the development of CO₂ utilization brings various benefits to Vietnam's growth. Firstly, Vietnam has significant reserves of natural gas, with 700 billion cubic meters of proven reserves, but many of the discovered gas fields have high levels of CO₂ ranging from 10 - 60 mol%. CO₂ utilization helps to exploit and use our

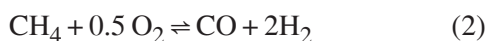
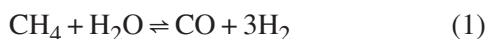
natural resources efficiently. Secondly, Vietnam is a developing industrial country with many factories that emit a large amount of CO₂ into the environment, such as steel mills, cement plants, and thermal power plants. Applying direct CO₂ conversion technologies helps to recover these emissions and convert them into valuable products, as well as encouraging the construction of facilities for CO₂ capture and storage. Last but not least, CO₂ utilization will contribute to Vietnam's green growth, directing us to achieve our Net Zero Emission target by 2050.

3. Simultaneous CO₂ and CH₄ conversion technologies

3.1. Natural gas with its CO₂ content up to 10 mol% - A feedstock for urea production

Ammonia (NH₃) is one of the chemicals that is widely used in many fields such as industry, agriculture, household products, metal processing, etc. Urea is produced through the reaction between NH₃ and CO₂. The most common application of urea is as a type of nitrogen fertilizer. The synthesis of ammonia and urea is derived from natural gas, which goes through a synthesis gas production phase using various reforming methods, typically steam reforming, as it has the highest H₂/CO ratio suitable for ammonia production. All of the CO₂ generated in the Water Gas Shift (WGS) reaction is used in the synthesis of urea.

The transformation of natural gas with methane as a representative component into urea is described by Equations 1 - 6.



The impact of CO₂ on natural gas

In the production of ammonia, the presence of CO₂ in natural gas is undesirable as it only adds to

the energy cost. In contrast, for the urea synthesis process, if CO₂ is present in the initial natural gas, it will help minimize the production or collection of CO₂ for the urea synthesis stage. For natural gas that has gone through reforming to create a high H₂/CO ratio and thus produces a lot of NH₃, there will be excess NH₃ compared to the amount of CO₂ obtained. Therefore, the presence of CO₂ in natural gas will help balance the amount of NH₃ and CO₂ for the urea production stage.

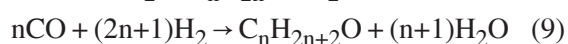
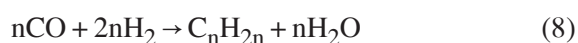
The optimal concentration of CO₂ in natural gas for the ammonia/urea synthesis process depends on the composition of the natural gas (lean or rich). If it is lean natural gas, the optimal CO₂ concentration is around 5-10% [5]. However, if it is rich natural gas, then the amount of carbon is already sufficient, and it may even be in excess, so it would be better if this gas does not contain any CO₂.

Currently, the Ca Mau Fertilizer Plant is operating with 8% CO₂ in the input stream, showing that the conversion efficiency of NH₃ and CO₂ in the high-pressure reaction equipment is quite high, about 63%, and for pure urea products up to over 99%.

The scale of application: commercialization. Currently, many companies provide licenses for these technologies such as Haldor Topsoe (Denmark), KBR (USA), or Casale (Switzerland).

3.2. Fuel production by the Fischer-Tropsch reaction

The Fischer-Tropsch (F-T) reaction is a chemical reaction that allows the conversion of CO and H₂ into liquid hydrocarbons, developed by Franz Fischer and Hans Tropsch in 1925. The conversion of synthesis gas into liquid fuels through the F-T reaction helps create a cleaner burning fuel source with fewer air pollutants such as sulfur or nitrogen. The synthesis process takes place according to the following reactions:



The impact of CO₂ on natural gas

The presence of CO₂ will decrease the H₂/CO ratio after the reforming process, depending on the desired liquid product, the CO₂ content may have a positive or negative impact. Typically, the CO₂ content in natural gas should be below 30% to create hydrocarbons with a high-octane number (ON) suitable for use as fuel. Hydrocarbons produced from a high H₂/CO ratio tend to have lower molecular weights. In addition, CO₂ can act as a diluent to reduce the partial pressure of the input reactants and promote hydrocarbon formation, leading to higher selectivity of liquid fuels and reducing the formation of methane gas. However, a large amount of CO₂ can reduce the overall conversion rate and hydrocarbon yield because CO₂ does not directly participate in the conversion process [6].

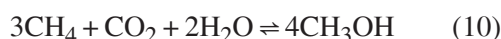
The scale of application: commercialization. The commercialization of the Fischer-Tropsch process to produce liquid fuels from natural gas has been applied for a long time in many countries such as Qatar, South Africa, and Malaysia.

3.3. Natural gas with its CO₂ content of 10 - 30 mol% - A feedstock for methanol and dimethyl ether (DME) production

Fossil fuels and natural gas are currently the main sources of energy supply. However, they are gradually being depleted and their uses emit large amounts of GHG. Methanol is considered a new source of energy with an octane rating of up to 113 and a specific gravity equal to 1/2 compared to gasoline [7]. Methanol is also an important raw material for synthesizing acetic acid, olefins, MTBE, and fuel additives, ... which are raw materials for more than 30% [8] of the chemical industry's processes. On the other hand, methanol can be dehydrated into dimethyl ether (DME) which has been promoted as an alternative fuel for LPG and diesel.

Methanol can be produced from various renewable feedstocks such as biomass, CO₂,

biomethane, ... The most common method to produce methanol is through the production of synthesis gas (indirect method), the appropriate H_2/CO ratio for methanol synthesis is 2, so the optimal method to create syngas with this ratio is bi-reforming while if using steam reforming, the ratio must be adjusted by reducing the hydrogen content. If natural gas has a CO_2 content of 10-30%, it will be suitable for the production of methanol and dimethyl ether (DME). CO_2 is needed for the methanol synthesis process and occurs according to the total reaction:



Stoichiometrically, it can be seen that a mixture of CH_4 and CO_2 with its molar ratio of 3 (i.e., the gas contains 25 mol% of CO_2) is the right feedstock for methanol production.

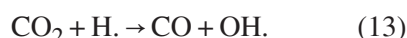
The impact of CO_2 on natural gas

The optimal CO_2 content for the methanol production process is about 25-30%, thereby reducing the cost of pre-treating CO_2 and contributing to increasing the efficiency of the process. The presence of CO_2 increases the carbon ratio in the reforming process, making it easier to form coke, causing loss of activity, and cost for catalyst regeneration or replacement. CO_2 also incurs additional operating costs due to the corrosion of pipes due to the formation of carbonic acid during transportation.

The scale of application: commercialization. Now, the Air Liquide Group has put into operation a methanol production plant from natural gas with a CO_2 content of 31% on Hainan Island, China since 2006 [9]. In 2014, Haldor Topsoe demonstrated a pilot plant to perform a bi-reforming of CH_4 - CO_2 mixture without significant coke formation in Brazil, this is a reforming method that is eligible for methanol production [10].

Recently, a new method has just been patented in the US for the synthesis of DME through only one stage. John E. Stauffer, from the University of Greenwich, proposed a direct synthesis reaction of

methanol from CH_4 and CO_2 without going through the syngas synthesis process, eliminating many stages in the previous process. In addition, the reaction between CH_4 and CO_2 takes place according to the free radical reaction and does not require the presence of a catalyst with a reaction temperature above $1,000^\circ C$ [11].

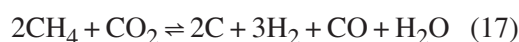
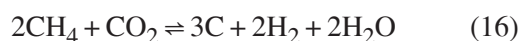


The problem that needs to be solved is the above one-stage process is still being researched in the laboratory because the methanol conversion efficiency has only reached about 5%, so it is necessary to improve the process to increase the efficiency to 10% before it can be commercialized [10].

3.4. Natural gas with its CO_2 content of 25 - 50 mol% - A feedstock for dry reforming and carbon nanotubes (CNTs) production

CNTs as well as other nanomaterials show excellent physical properties such as high tensile strength, large surface area, ultra-lightweight, good electrical and thermal conductivity, etc., leading to high potential for use in semiconductor, electronics, energy battery, or drug delivery fields. CNTs have a good load-bearing capacity (10-100 times higher than the best steel) per unit mass [12].

Carbon nanotubes (CNTs) are synthesized through the decomposition of methane (mainly by the CVD - chemical vapor deposition method). The process of forming carbon nanotubes from CO_2 -rich natural gas is as below:



It has also been discovered that the formation of CNTs during coking in the dry reforming reaction opens up a greater opportunity to take advantage of

the disadvantage of forming a lot of coke in DRM into an advantage and brings this technology closer to the industrial scale.

The impact of CO₂ on natural gas

The presence of CO₂ in the raw material helps to lower the reaction temperature and improve product quality. CO₂ participates in many reaction chains such as dry reforming, Boudouard oxidation-reduction reaction (between CO and CO₂), or RWGS reaction generating steam, etc., helping to eliminate defects arising during CNTs synthesis even when significantly narrowing the diameter of carbon nanotubes.

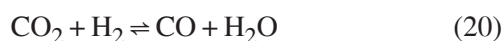
The scale of application: The conversion of natural gas with a CO₂ content above 25% into CNTs is still in the research stage in the laboratory. Currently, the production of CNTs from powder catalysts must go through the product separation process because CNTs formed are mixed with catalysts and this mixture needs to be refined and filtered to obtain pure CNTs, increasing production time, chemical costs, reducing product yield, causing defects that reduce the quality of CNTs while investment costs, operating and maintenance costs of refining plants are very large, possibly accounting for up to 90% of the product cost.

4. Direct CO₂ conversion technologies

4.1. Production of methanol through the CO₂ hydrogenation process

Currently, many countries around the world have started researching and developing these technologies such as China, Japan, Australia, and European countries. For example, in Reykjavik, Iceland, a methanol production plant was completed in 2012, producing about 4,000 tons of methanol per year from CO₂ and H₂, treating about 5,500 tons of CO₂. This plant uses about 10% of CO₂ emissions from electricity production, H₂ used also comes from electrolysis. CO₂ and H₂ are converted into methanol thanks to the CZA catalyst system at conditions of 250°C and 5-10 MPa [8].

The reaction of converting CO₂ into methanol and other side reactions such as:



The presence of by-products such as CO and H₂O from the RWGS reaction is a current issue with this reaction. Water can decrease the catalysts activity and alter the reactions direction. As a result, it is important to concentrate on enhancing catalyst efficiency and eliminating by-products like H₂O.

The biggest problem that makes this process not yet widely developed and operated on a large scale is due to the inertness of CO₂. To obtain a high-selectivity MeOH recovery process with good conversion, it is necessary to operate at high pressure and medium temperature.

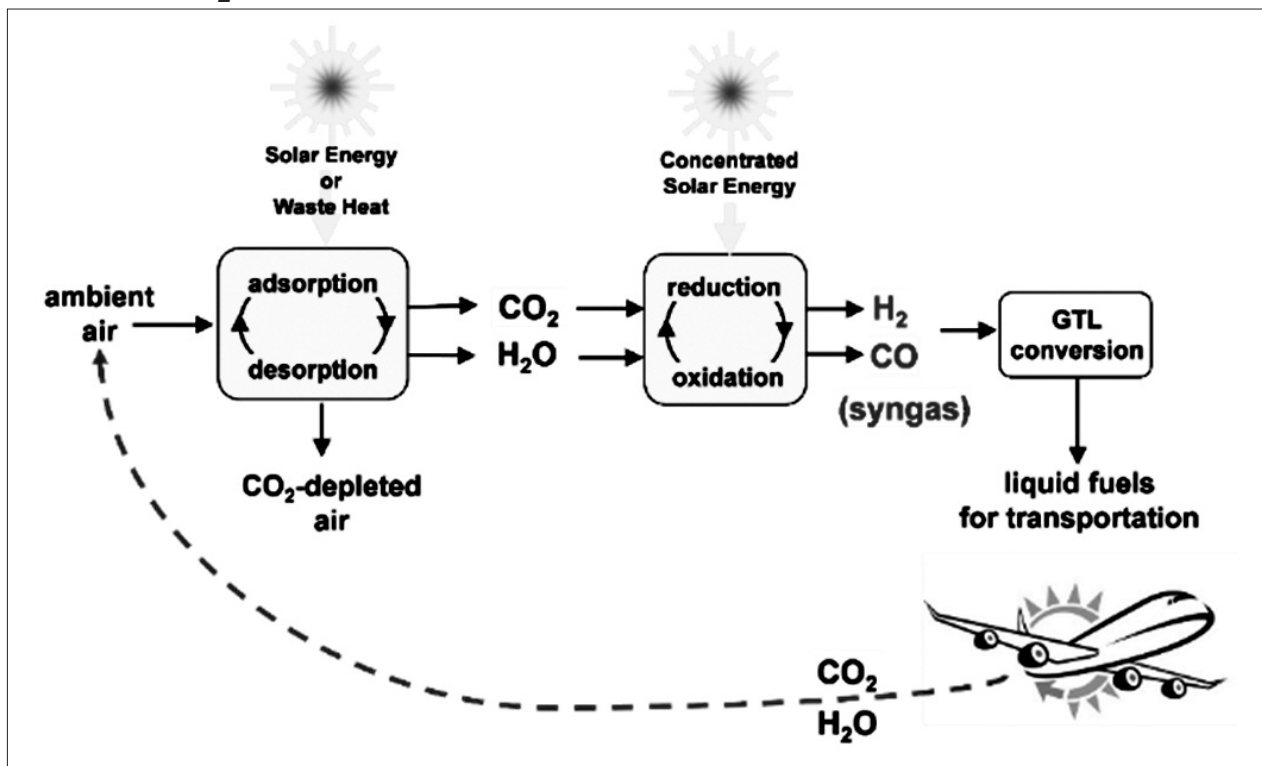
4.2. Conversion of CO₂ into fuel

A method to convert CO₂ into fuel has been developed by ETH Zurich (Switzerland) with the technology of producing liquid fuel by converting CO₂ and H₂O in exhaust gas using solar energy as the energy source. The goal of the project is to help reduce at least 50% of CO₂ during fuel production and use [13].

This process consists of two main steps. First, CO₂ and H₂O will be captured from the air through the absorption process. This mixture will be transformed into synthesis gas through solar radiation energy at a temperature of about 1,500°C in the presence of a CeO₂ catalyst, and synthesis gas is formed through the decomposition process of CO₂ and H₂O [13]. Then, the synthesis gas will be converted into liquid fuels such as diesel or ketone through the Fischer-Tropsch reaction. The catalyst used in this Sun-to-Liquid process is CeO₂, which is chosen because of its good thermal stability, making it suitable for use in this reaction without being degraded.

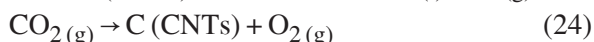
4.3. Producing Carbon nanotubes by electrolysis reaction

The method for producing carbon nanotubes (CNTs) directly from CO₂ is called C2CNT and

Figure 1. The CO₂ conversion process of ETH Zurich (13)

involves the electrolysis of molten salts. This process, developed by scientists at George Washington University, allows for the conversion of CO₂ from fossil fuel combustion, such as natural gas, into carbon and oxygen. The oxygen is then sent back to the power plant to increase the efficiency of the combustion reaction, forming a closed loop to optimize energy and reduce CO₂ emissions.

The process of electrolysis of molten CO₂ salt into C and O₂ uses Li₂CO₃ as the carbonate electrolyte, a nickel anode, and a Galvanize steel cathode. The representation is as follows:



We can see that all CO₂ gas will be converted into CNTs, and based on their molecular weight, to produce 1 ton of CNTs, we will consume 3.7 tons of CO₂, bringing significant benefits to the environment. This process is expected to be

integrated into an energy plant in Alberta, Canada to convert around 5 tons of CO₂ per day into CNTs instead of releasing them into the environment as it is now [14].

5. Evaluation of the potential application of advanced technologies for CO₂ and CO₂-rich natural gas conversion in Vietnam

The application of direct CO₂ conversion technologies or simultaneous CO₂ and methane conversion technologies depends on the supply source, market demand for the products, level of technological development, as well as the cost and feasibility of the project.

5.1. Direct production of methanol from CO₂

Similar to what has been presented, the direct production of methanol from CO₂ through hydrogenation reaction is considered one of the good solutions for reducing greenhouse gas emissions for our country as well as bringing in large revenue from valuable products. Currently, the main source of CO₂ emissions in our country

comes from industries such as steel production or construction as well as fuel combustion at power plants, etc.

The steel industry in Vietnam is one of the developing industries with many large-scale factories. The emission intensity of the Vietnamese steel industry is also higher than the world average due to the use of many fossil fuels [15]. Therefore, the ability to recover and convert CO₂ obtained from steel mills plays an important role in our countrys emission reduction goals.

With the consumption market for methanol in 2030 being 640 KTA, assuming that the conversion of CO₂ into methanol can meet 30% of the market, which is 192 KTA, the number of CO₂ emissions reduced is 264 KTA. Assuming that Vietnams steel industry also contributes 7% of total CO₂ emissions, the average CO₂ emissions of our steel industry is 336.7 million tons of CO₂ x 7% = 23,569 KTA. So, converting CO₂ into methanol handles 1.12% of CO₂ emissions from the steel industry.

Another aspect to consider is the CO₂ emissions from burning fuel. For example, in Urea plants CO₂ emissions come from burning fuel to supply energy for reaction processes. Almost all of the CO₂ in the initial natural gas and the CO₂ generated from the WGS reaction are used for urea synthesis, so there is not much CO₂ emitted into the environment during production.

The Ca Mau Fertilizer Plant sources natural gas from the pre-treated PM3 gas, which contains approximately 8% CO₂ with a flow rate of around 510 million m³ per year, and the permeate gas contains 50% CO₂ with a flow rate of around 3,000 m³/h. The majority of CO₂ emissions come from 40% of natural gas used as fuel for the reforming reactor, boiler, and power turbine.

For the Phu My Fertilizer Plant, the natural gas source is from the Southeastern region containing 2-3% CO₂ with an input flow rate of about 630 million m³/year. The majority of CO₂ emissions come from 30% of feedstock used as fuel.

Through calculations of the combustion reaction providing energy to the reaction furnace with each percentage of the feedstock used, plus the amount of CO₂ in the natural gas, the result shows that the CO₂ emission amount of the Ca Mau Fertilizer Plant is 365 KTA, while that of the Phu My Fertilizer Plant is 400 KTA.

The CO₂ emissions from the combustion activities at these two factories can be treated by recovering, storing, and using them to directly convert into methanol by hydrogenation. The source of hydrogen used is green hydrogen, this type of energy is gradually being paid attention to and developed in Vietnam, so this demand can be met from abundant renewable energy sources such as wind power and solar power in the Central Coastal provinces and the Mekong Delta. Evidence is that recently in Tra Vinh province, the groundbreaking ceremony for the first green hydrogen plant in Vietnam was held [16].

Based on the reaction equation, the amount of green H₂ needed for CO₂ conversion in the urea field is estimated to be about 104 kilotons per year. With the projected cost of producing green hydrogen in Vietnam in 2030 at about 2 USD/kg [17], the cost of the required hydrogen would be approximately 208,636 USD per year.

5.2. Producing nanocarbon tubes from the Ca Voi Xanh gas field

The Ca Voi Xanh gas field was discovered in 2011 off the coast of Central Vietnam about 100 km away. It is the largest gas field discovered in our country to date, with a reserve of about 150 billion m³ of natural gas. However, the CH₄ content in this gas field is only about 58%, while the rest are mostly other impurities such as CO₂, N₂, and H₂S, especially CO₂ accounts for about 30%.

In Vietnam, as presented, it is expected that our countrys demand for CNTs consumption in 2030 will be about 3,700 KTA and need to use 13.5 million m³ of natural gas. Taking advantage of the high CO₂ content in natural gas to produce high-quality nanocarbon tubes will bring a large source

Table 1. The potential market for some products using CNTs (KTA) (19)

Products	2020	2025	2030	2035
Methanol	251	296	296	296
Poly Ethylene (PE)	1,557	1,812	2,022	2,196
Poly Propylene (PP)	1,286	1,532	1,762	1,958
Potential for producing PE and PP from methanol*	143	167	189	208
*Assuming to meet 10% of the PP and PS market.				

of revenue for our country such as exporting or serving the domestic market. The cost of one gram of CNTs ranges from 0.1 - 600 USD/gram depending on the type and quality of the product [18]. Therefore, with the production of CNTs from the Ca Voi Xanh gas field, we can obtain a very large cost from selling this product.

With the CO₂ gas content in the Ca Voi Xanh gas field being about 30%, we can reduce emissions by about 30% x 13.5 million m³ = 4.05 million m³ of CO₂ gas corresponding to about 7.57 kilotons of CO₂ per year.

In Vietnam, the production of CNTs is at the laboratory, small scale, and has been synthesized at the Research and Development Center for Petroleum Processing (Vietnam Petroleum Institute), Faculty of Materials Technology (Ho Chi Minh City University of Technology), etc.

5.3. Production of methanol and petrochemical products

Methanol is in the development plan of the chemical industry in Vietnam. It is expected that by 2030, our country will invest in the production of methanol and some products from this chemical to meet domestic demand and export, limiting the export of crude materials. The development of petrochemical products from methanol through MTO or MTP technology has

also been studied to create a foundation for the development of a petrochemical chain in Vietnam. The demand for methanol and PP, PS plastics is presented in Table 1.

In response to the demand for methanol as well as PP and PE in 2030, it is expected that the methanol consumption market will be worth about 674,000 tons per year and we can process a large amount of CO₂ in natural gas. For the production of methanol by the bi-reforming method, it is expected to reduce emissions by 242 KTA compared to steam reforming, while dry reforming technology will reduce 924 KTA of CO₂.

6. Conclusions

To fight climate change, we need to reduce greenhouse gas emissions now. One solution is to capture, store and use CO₂ (CCUS) as a renewable material for industry. In Vietnam, these technologies are not fully developed, CO₂ conversion technology and CO₂-rich natural gas have different levels of maturity and need to be addressed to be widely commercialized, according to the orientation and goal of reducing CO₂ emissions in our country as well as the increasing demand for products in the domestic and regional markets, the application of these technologies is completely feasible and has great practical significance ■

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CÔNG NGHỆ CHUYỂN HÓA CO₂ VÀ KHÍ THIÊN NHIÊN GIÀU CO₂ - TIỀM NĂNG ỨNG DỤNG TẠI VIỆT NAM

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TÓM TẮT:

Hiệu ứng nhà kính đang là một trong những vấn đề được quan tâm hàng đầu, nguyên nhân là do việc phát thải quá mức khí nhà kính, đặc biệt là CO₂ vào khí quyển do các hoạt động sản xuất công nghiệp của con người. Để giảm thiểu tác động của biến đổi khí hậu, việc thu hồi và chuyển hóa CO₂ thành những sản phẩm có giá trị được xem là một phương pháp quan trọng và mang lại hiệu quả cao. Do đó ngày nay, rất nhiều nghiên cứu về các công nghệ chuyển hóa CO₂ đã và đang được phát triển. Việt Nam là một trong những quốc gia đang bị ảnh hưởng rất nặng nề bởi biến đổi khí hậu, tuy nhiên chúng ta cũng có nhiều tiềm năng trong việc áp dụng những công nghệ chuyển hóa CO₂ từ các hoạt động công nghiệp hay tối ưu hoá nguồn khí thiên nhiên giàu CO₂ có sẵn tại Việt Nam.

Từ khóa: phát thải CO₂, khí thiên nhiên giàu CO₂, nguyên liệu tái tạo, CCUS.

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