

Economic Assessment of the Hydrogenation of CO₂ to Liquid Fuels and C₂ to C₄ Petrochemical Feedstock

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Dedicated to Professor Schlögl on the occasion of his 60th birthday

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Abstract

The catalytic hydrogenation of carbon dioxide leads to C₂ to C₄ (petrochemical feedstock) and liquid C₅₊ hydrocarbons (fuel). Carbon dioxide is obtained by its removal by absorption from the effluent of coal- or gas-driven power plants while hydrogen can be sustainably generated by electrolysis of water using electricity either from windmills or photo voltaic. Thus, environmentally harmful CO₂ may be converted sustainably to useful products. On the basis of a process flow sheet, the costs for producing the hydrocarbons, i.e. the processing of CO₂, are estimated for different plant sizes. The price of hydrogen contributes significantly to the overall production costs. Depending on plant size and hydrogen price, the costs of the different hydrocarbon fractions range from approx. 1 to 4 €/kg based on the present hydrogen price of (4 €/kg being a conservative estimate) and on its future prediction (2 €/kg). Further price reductions may be achieved by final engineering optimization of the process.

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Keywords

Hydrogenation of CO₂

Hydrocarbon products

Liquid fuel

C₂ - C₄ hydrocarbons

Economic process assessment Effect of hydrogen on price of hydrocarbon products

1. Introduction

The catalytic performance of selected catalysts for the hydrogenation of CO₂ towards both hydrocarbon fuels (C₅-C₁₅) and lower hydrocarbons (C₂-C₄ olefins and paraffins) was recently reported [1a, 1b]. This is definitely a sustainable route for removing large amounts of highly concentrated CO₂ contained in the off-gas of coal- or gas-driven power-plants and

in other miscellaneous sources. Against storage of CO₂ in underground caverns [2a, 2b] which has been repeatedly suggested, the proposed process offers an important advantage: chemical interactions of CO₂ with surrounding underground structures are excluded, which could possibly lead to mechanical instability of the caverns. Advantageously, the hydrocarbon products can be used as fuel and chemical feedstock. It should be mentioned that processing of hydrocarbons from CO₂ solved in sea water has been recently suggested by the US navy [3].

The feasibility of such a process is discussed on the basis of a tentative process scheme, which is to a large extent similar to that of well-known conventional Fischer-Tropsch (FT) technology. The preliminary economic assessment of the process was carried out for pilot- and demonstration-plant scale; subsequently, the processing costs were extrapolated to large-scale operation. The respective product costs are derived from a rough estimate of investment costs and the assumed feed costs for carbon dioxide and hydrogen. The prices of “sustainable” hydrogen from electrolysis (electricity production from photo-voltaic or windmill) are, however, still difficult to assess for the next-decade. Also the prices for CO₂ from coal- or fuel-driven power plants are still uncertain for political reasons but they are not very essential for the overall processing costs. The ranges of such prices for both raw materials are discussed (see section 2.2.2 on cost of feedstock etc). From a chemical point of view the transformation of CO₂ to carbon monoxide via the reversed water-gas-shift reaction is a necessity as an initial step for the FT synthesis.

2. Principles of Technology and its Economics

2.1 Technology

The suggested process is based on the performance of selected catalyst compositions for the hydrogenation of CO₂ to higher hydrocarbons (CO₂-FTS), which had been recently tested [1]. Only iron oxide-based catalysts modified by the oxides of K and Cu or Zn can reach high CO₂ conversion combined with high selectivity to the desired C₅ to C₁₅ and C₂ to C₄ hydrocarbons as well as low methane formation; TiO₂ is the most suitable support for obtaining low methane selectivity. High conversion of CO₂ to hydrocarbons can only be achieved when the Fischer-Tropsch reaction is fast enough to overcome thermodynamic limitation of the first reaction step, that is the reverse water-gas shift reaction. For this purpose, higher reaction temperatures, higher pressure, and an excess of H₂ in the feed are helpful since they increase the reaction rate of the required FT reaction. However, catalysts to be used should also minimize methane formation. Fortunately, some catalyst compositions fulfil this need [1].

Based on the above, a combined production of liquid fuels (mainly C₅ to C₁₅ hydrocarbons) and lower hydrocarbons, in particular C₂ to C₄ hydrocarbons are obtained which may be used as feedstock in chemical industry. Methane being a side product serves mainly for heat generation required within the process. It is obvious, that such a process is a means of reducing CO₂ from fossil-fuel-driven power plants that would be otherwise released into the atmosphere; moreover, the liquid fuel fraction could be more easily stored, e.g. in underground caverns, than the gaseous CO₂ itself. Moreover, such process may substitute or supplement part of present crude-oil supply. The economics may be improved if the expected CO₂ tax applied in the EU and other industrial countries becomes world-wide effective; (this is presently an open question due to political reasons). In any case, cheap hydrogen sources are needed. Such hydrogen can be generated by water electrolysis using electricity from photo-voltaic or windmill-based power generation.

The economics of the process are affected by the price of carbon dioxide and hydrogen which depend in particular on technological but also political boundary conditions. The process economics are also influenced by the costs of capture and separation of CO₂ from the effluent of the power plants, and finally, by the cost of “green” hydrogen generation from sustainable sources based on the electrochemical conversion of water. Depending on the cost for CO₂ and H₂, the suggested technology “CO₂ → fuel plus petrochemical feedstock” may become economically feasible in the long run; this is dealt with in the subsequent sections.

As an introduction to the topic, a simplified block diagram for the process of CO₂ hydrogenation to hydrocarbons is shown in Figure 1. A more detailed scheme of the process for investment cost estimation is presented in Figure 2 (see further below).

Insert Figure 1

The block diagram includes the reactor and the respective apparatuses for unit-operations leading to the final products, which comprise C₅₊ hydrocarbons and C₂ to C₄ hydrocarbons consisting mainly of olefins, besides CH₄. In particular, the following operations are required:

- Mixing of fresh reactants with re-cycled feed of non-converted hydrogen and CO₂
- Compression and pre-heating of the feed of reactants
- Chemical conversion of the feed gas
- Cooling down of the effluent/product streams

- Water separation
- Absorption of non-reacted CO₂, its de-sorption and recycling to the reactor inlet
- Flashing of light components and gases (CO, methane, hydrogen)
- Purification of final hydrocarbon products.

The feed composition at the reactor inlet is a crucial process parameter. By recycling part of non-converted hydrogen and CO₂, the feed composition is modified and the ratio of CO₂/H₂ is changed. Thus, it is very important to control the feed composition after recycle addition. Furthermore, CO₂ will be separated from gaseous products by absorption. Removal of CO₂ from the product is not quantitative; only 95 % can be withdrawn and recycled. On the other hand, the absorption from gaseous products is mostly very selective to CO₂ and no other components are recycled. However, due to desorption of CO₂ from the absorbent at lower pressure of usually 5-10 bar, the recycle should be added to the feed before its compression.

2.2 Economics

The economics of the process, i.e. the processing costs, depend on the required plant investment, the costs for the feed, i.e. CO₂ and H₂, as well as for cost of the required catalyst and general expenditures. On the basis of the proposed process scheme tentative cost estimates for necessary plant investment and the main running expenses are derived. On all this information including specific reaction conditions, first, detailed investment costs were estimated for a pilot-plant (4.580 kg of C₅ to C₁₅₊ hydrocarbons <fuel> and 6.000 kg of C₂ to C₄ hydrocarbons per day) and for a demonstration-plant of tenfold capacity. The results are presented in the subsequent section 2.2.1. In addition, by extrapolation of the investment costs of the demonstration plant also investment-cost estimates of medium-sized and large-scale plants (114.000, 229.000, and 458.000 kg p. d. of fuel and the respective amount of C₂ to C₄ as indicated above) were derived as outlined in the subsequent section. The results lead to indications about the process economics and their further improvement. Although the above procedure is subject to larger inaccuracies the approximate total processing costs towards the C₅₊ fraction (fuel) and towards the C₂ to C₄ fraction (petrochemical feedstock) are accessible taking the prices for CO₂ and H₂ into account.

2.2.1 Investment costs for a pilot and a demonstration plant

The investment costs being the backbone of the subsequent economic considerations have been estimated for a 4.580 and 45.800 kg of fuel per day in addition to the petrochemical feedstock. The costs are based on a detailed process scheme comprising the different main

apparatuses and process streams as presented in Figure 2 along with Tables 1 and 2 by taking into account the overall mass and enthalpy balances of the various process streams. The total investment costs are composed of the sum of costs for major and auxiliary equipment (ME and AE).

Insert Figure 2 and thereafter Tables 1 and 2

2.2.1.1 Major equipment costs

The costs of ME were estimated by using open literature sources, knowledge from industry and various databanks, comparable with the earlier ICARUS computer program [4-6]. For the sake of correctness it should be mentioned that the scale-up extrapolation of the individual types of apparatuses depends for different plant capacities on the range of apparatus sizes. For the above mentioned plant capacities, i.e. 4.580 and 45.800 kg of C₅₊ hydrocarbons per day, the data of ME costs are listed in Table 3; (the data for investment costs for the higher-capacities plants were obtained by extrapolation of these data, see section 3.2)..

Insert Table 3

2.2.1.2 Total investment costs consisting of major and auxiliary equipment

The total investment costs including the costs for auxiliary equipment were calculated on the basis of the ME costs by using Guthrie's module method [7], often used in chemical-engineering process design [8]. For the present case, a module factor of 3.5 for estimating total investment costs has been chosen as usually done for only partly proven new technologies. The total investment costs include ME and AE as well as all other costs required for commissioning of the plant, comprising measurement and control processes variables, and engineering services, pipelines and electricity supply, etc. The total investment costs for the two capacities as mentioned in section 2.2.1.1 are also reported in Table 3. More precision of the final total investment cost estimation of the CO₂-based FT synthesis process plant can be obtained by a more detailed engineering approach for the proposed plant. This was, however, not the intention of this preliminary study. Only a preliminary estimate of the various investment costs was looked for. It should only offer a simplified comparison of the production costs for hydrocarbons as obtained by this tentative CO₂-based FT process with

present-day technology for producing C_2 to C_{15+} hydrocarbons from carbon dioxide; for this purpose the plant sizes were extrapolated to higher capacities. – A special paragraph 3.2 is devoted to the extrapolation of total investment costs (main and auxiliary equipment) to large-scale plants in order to assess the effect of scale-up on processing costs

2.2.2 Costs of feedstock, catalysts and fixed costs

For the subsequent estimation of the production costs of hydrocarbons (C_2 to C_4 and C_{5+}) from CO_2 and H_2 , the prices of feedstock, catalysts, and utilities, which were ascribed to fixed costs, are required.

2.2.2.1 Carbon dioxide

Industrial sources of CO_2 are coal- or natural gas-fired power plants for electricity production. Severe environmental damage is ascribed to CO_2 ; (carbon dioxide from automobiles is not considered since it cannot economically be converted as suggested in this paper due to its low concentration in the ambient air). In the long-range future it is intended to introduce mandatory taxes (certificates) for CO_2 emitted from power plants in all industrial states; these taxes are presently only enforced in the EU. Tax certificates for CO_2 are presently available for 4 to 6 € per metric tonne at the European Energy Exchange in Leipzig (February to April 2013) although a price of ca. 20 €/t CO_2 is scheduled for the future. For a production cost estimate the mandatory CO_2 tax should be at least partly deducted from the future price of CO_2 used as raw material in the FT synthesis process. That is to say, the price for separation and purification of CO_2 emitted from power plants for the proposed subsequent catalytic Fischer-Tropsch (FT) process plants ought to be reduced by at least 10 € per metric tonne CO_2 for the respective FT synthesis presented in this paper.

According to the state of technology, post-combustion carbon-dioxide capture from coal- or gas-fired power stations is separated from the off-gas by liquid chemical absorption using commercially available absorbents. Several alternative cost estimates exist which differ significantly.

1) MEA (Mono Ethanol Amine) as a solvent has been used by Dave et al. [3]. CO_2 is being separated by thermal treatment from the chemical absorbent. The cost of CO_2 avoidance in the effluent amounts to ca. 69 AUS\$ per metric tonne; a reduction of cost could be achieved with a solvent having a 50 % higher absorption capacity; the authors mention ca. 48 AUS\$ per metric tonne CO_2 (1 AUS\$ amounts to approximately 0.69 €).

2) The Siemens company reports [9] a best case scenario for capture, compression, transportation and intermediate storage of CO_2 :

Capture of CO_2 : 25 to 60 € per metric ton CO_2 ,

Compression, transportation, storage: 10 to 30 € per metric tonne CO₂.

That is to say, the cost for CO₂ avoidance should amount to approximately 35 to 90 € per metric tonne CO₂.

3) The International Energy Agency reported in 2010 values for “cost of CO₂ avoided” (in US \$/ metric ton CO₂) from nine organizations collected for the years 2005 to 2009: 62, 69, 60, 87, 128, 75, 76, 75, 90 US\$ per metric tonne CO₂. This amounts to an average of 80 US\$ per metric ton CO₂ [10].

The data of the three cases 1) to 3) on a € basis (August 2013), which were obtained for avoidance of CO₂ emission in the atmosphere, and which should correspond to the feed price of CO₂ are summarized in Table 4.

Insert Table 4

These data are in close agreement with those recently reported by Wilson et al. [11] and presented in Table 5.

Insert Table 5

On the basis of the above data, a preliminary price of 45 to 49 €, i.e., an average of 47 € per metric ton of CO₂ is most probably an acceptable choice. If the European tax for CO₂ is considered, this price might be reduced by at least ca. 10 € (see further above). That is to say, the CO₂ price should amount to ca. 40 € per metric tonne.

2.2.2.2 Hydrogen

The prices for required “green” hydrogen gained by electrolysis of water using electricity from wind, photo voltaic, geothermal and hydro-power, or ocean-tides power (the latter taking advantage of the differences between high and low tides in coastal areas), are similarly subject to economic uncertainties as for CO₂. For production costs only electric power from sources based on photovoltaic and/or wind are considered since they appear most advanced for the present purpose.

The price of hydrogen from natural gas (produced via steam reforming) may serve for comparison with photovoltaic- or windmill-based technologies to assess their suitability as compared to conventional procedures. E.g., the price of hydrogen based on steam-

reforming of natural gas amounts to 4 to 5 US\$/ kg of H₂ as pointed out below which could be used as a reference.

Electricity from windmills

The range of hydrogen prices covers a wide span. Bockris and Veziroglu [12] reported for 1 kilogram of hydrogen including a tax of 1.25 US\$ the following prices:

- hydrogen from wind (via electrolysis) is estimated to 8 to 10 US\$,
- for comparison, hydrogen from nuclear power (via electrolysis) would cost 7.50 to 9.5 US\$

Troncoso et al. [15] reported the implementation of a water-electrolysis plant in combination with a wind-power plant for the generation of “green” hydrogen. For different wind fluid-dynamic conditions (wind curtailment) a range of prices varying from 20 to 30 US\$ (ca. 20 €) per kilogram of hydrogen was stated, which appears extremely high as compared to the above data.

A significantly lower price for producing hydrogen by electrolysis was reported by Thompson J. R. et al. [13-14]; it amounted to 3.1 US \$/ kg of hydrogen.

Electricity from photo-voltaic

Price data on photovoltaic-driven aqueous alkaline electrolysis of water for hydrogen production at different conditions were communicated by Licht et al. [16]; four alternative process conditions resulted into US\$ 5.74, 4.96, 3.01, and 2.61 per kg hydrogen.

Similar results have been reported in a very recent NREL-report (National Renewable Energy Laboratory) updated in June 04, 2013). The hydrogen cost targets for the year 2015 are US\$ 3.10 for central hydrogen plants and 3.70 for distributed hydrogen plants [17].

In summary, the hydrogen prices reported per kilogram range from about 3 to a maximum of 30 US\$; most of these prices are, however, below 10 US\$. Thus, a conservative price of about 4 to 5 US\$ (equivalent to about 3.8 €) may appear appropriate for the cost estimation put forward in section 3 of the present paper.

Looking at the future price development even a price of 2 € per kg appears possible.

2.2.2.3 Catalysts

Suitable catalyst compositions for the CO₂-based FT synthesis have been reported [1]. An

optimized composition was implemented into the process scheme. According to our earlier work the oxides of $11.3\text{La}14.1\text{Cu}71\text{Fe}3.9\text{K}$ were applied.

Since no recent market price for FT catalysts was accessible, the present estimate is based on an earlier report issued by BMFT in which a catalyst price of 750 DM (being equal to ca. 375 €) per metric tonne was suggested for the year 1974 [18]. Assuming an average inflation rate of 3 % p.a. since 1974, a price of 1.223 €/metric tonne appears appropriate for the years 2013/14.

2.2.2.4 Fixed costs for energy and utility

The feed is preheated in two steps: (1) reaction heat from the reactor is used and (2) combustion energy of methane in the methane-fired heat exchanger is utilized. The price for methane combustion is neglected since there is a source of sufficient methane available within the process. Cooling of the reactor effluent is realized by cooling water, pumped in a loop and assuming a spray unit to cool down the water after heat exchange. The costs for the spray unit are included in the total investment costs.

As outlined further above, the process includes a reactor and common process engineering operations, in particular unit operations (including compression of gases). Moreover, pumps are required for product separation besides preheating and cooling of process streams in the heat exchange loops. The price for electricity from wind energy was set to 0.0354 € per 1 kWh [19], [20]. Furthermore, the costs for operating personal (lab and maintenance) were integrated in the fixed cost calculation. Roughly, 500.000 €/year have been assumed for 3-shift working performance.

All the above costs are ascribed to “fixed costs for energy and utilities” [19]; the costs for catalysts are, however, independently dealt with in the following cost compilation since they could be clearly specified.

3. Processing cost estimation of differently-sized plants

Plants of different sizes (capacities) were conceptually designed on the basis of the flow-sheet presented in Figure 2. The product-stream compositions at the reactor outlet were obtained from earlier experiments [1]. The estimation of processing costs was targeted on total investment costs for the main as well as auxiliary equipment and the processing costs of the pilot plant as a whole as well as of the demonstration plant and the larger-sized plants.

The various plants were assumed to be operated within a frame of conditions to earlier CO₂-based FT plants and experiments recently carried out for the CO₂-based synthesis.

Frame of conditions for plant design and operation

- Source of hydrogen from water electrolysis; at a later stage the required electricity supply should be provided from wind energy or photo voltaic;
- Reaction conditions of CO₂-based FT-Synthesis: 300-350 °C,
- Ratio of CO₂/H₂: 1/3,
- Total pressure 10 to 15 bar
- Reactor: Catalyst development and catalytic experiments have been carried out in fixed-bed reactors.
- It is anticipated that the CO₂-based FT synthesis is carried out in a 2-phase (gas/solid catalyst) multi-tubular
- Fixed-bed reactor with product separation and recycle of non-converted reactants CO₂ and H₂. At a later stage, the use of a slurry reactor may be also considered.

3.1 Estimation of processing costs for a pilot-plant and demonstration plant

The processing costs were based on the total investment cost (as outlined in Table 3) and the direct as well indirect costs of the pilot and demo-plant the total production costs for both plant capacities (4.580 and 45.800 kg hydrocarbons per day) were derived as presented in Table 6; the costs for gaseous C₂ to C₄ hydrocarbons and of liquid fuel (C₅₊) are listed in this table. Moreover, for the sake of simplification, the table contains also data for the larger-scale plants which are discussed in the subsequent paragraph 3.2.

Insert Table 6

A careful analysis of the data leads to interesting insights in the economics of the CO₂-based FT process:

- + The cost of hydrogen being one of the two feedstock components needed for the process exceeds that of carbon dioxide significantly; the cost of CO₂ amounts to only 7.5 % of the total cost for feed; i.e., the processing costs are essentially determined by the hydrogen price. This is illustrated in comparing the results on the processing costs presented in Tables 6 and 7)
- + The production costs depend on the scale of the two plants. With increasing capacity (from 4.580 → 45.800 kg liquid hydrocarbons p. d. plus the respective amount of C₂ to C₄ hydrocarbons) the processing cost of the hydrogenation of carbon dioxide decreases per mass of products (gas and fuel) significantly for the higher capacity of the demo-plant. Hence, the cost estimation was extended to even higher capacities assuming similar

effects. From the latter results (see paragraph 3.2), it is obvious that similar but only less significant effects on the cost structure were observed.

3.2 Extrapolation of processing costs to processing plants of higher-capacities

The experimental conditions of the plants of higher capacities were set the same as for the pilot and demonstration plants (see preceding section).

Against the above results the plant extension to higher liquid hydrocarbons (fuel) capacities was increased by extrapolation from

45.800 kg p. d → 114.500 → 229.000 → 458.000 kg p. d.

The total investment costs I_x for the three plants were obtained by extrapolation from the total investment $I_{45.800 \text{ kg p. d}}$ for the base-case capacity of 45.800 kg fuel p. d. to higher capacities C_x

$$I_x = I_{45.800 \text{ kg p. d.}} \cdot (C_x / C_{45.800 \text{ kg p. d.}})^{0.7} \quad (\text{equ.1})$$

The direct costs were only adjusted to a minor degree. The resulting investment costs and the respective processing costs are presented in Table 6. The results indicate that the costs for total hydrocarbons decrease only slightly with increasing plant capacity. That is to say, there is only a minor scale-up effect above capacities of ca. 100.000 kg of fuel p. d.

As already indicated further above (section 2.2.2.2), processing costs can be significantly reduced for lower hydrogen prices. The results of processing costs in Table 6 are based on a hydrogen price of 3,8 € per kg hydrogen. It appears, however, possible that the hydrogen price may be reduced to about 2 € per kg (see above). For this situation the processing costs and the respective product prices were tentatively assessed. The results presented in Table 7 clearly show that the product prices are significantly reduced; they approach present-day prices of liquid fuel and petrochemical feedstock based on crude oil.

Insert Table 7

4. Conclusions

The tentative rough economic assessment of a process for the hydrogenation of carbon

dioxide to the total of C₂ to C₄ and liquid-fuel C₅₊ hydrocarbons results presently into significantly higher hydrocarbon costs as compared to processing of crude oil to such products. Further optimization of the chemical-engineering unit operations would presumably result in different (minimized) investment costs changing the outcome of this assessment slightly. However, the main potential for decreasing the production costs significantly would be a decrease of the hydrogen price as outlined above

Thus, from the present point of view, hydrogenation of carbon dioxide to hydrocarbons indicates an economic potential for its future elimination from the off gas of coal or fuel-driven power plants, where it prevails in high concentrations; by chemical conversion of carbon dioxide into useful hydrocarbon products a significant contribution to reducing carbon dioxide in the atmosphere will be possible if the described technology comes to bear. The product composition could be, in principle, adjusted by changing the catalyst.

For more accurate cost figures, a more detailed engineering approach is certainly required. Moreover, a pilot plant as described in this paper would be of high value to validate the present results*). Altogether, it is assumed that the pathway of converting environmentally harmful carbon dioxide to valuable hydrocarbons will significantly remove CO₂ from the atmosphere.

*) As a location for a pilot and/or demonstration plant the Ruhr area (close to Oberhausen, Germany, Europe) may be considered since the required infrastructure for a small-scale pilot plant as well for the larger-scale demonstration plant should be presumably partly available; due the available coal- and gas-driven power plants. Thus, in this area, also sufficient carbon dioxide could be delivered as feedstock for a modified Fischer-Tropsch synthesis.

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