

NEW PROCESS DEVELOPMENT OF NATURAL GAS CONVERSION TECHNOLOGY TO LIQUID FUELS VIA OCM REACTION.

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Abstract

The conversion of methane via an OCM (Oxidative Coupling of Methane) to transportable liquid fuel has been investigated in order to utilize remote natural gas effectively. A conceptual view of this new process was developed for gasoline production based on reviews of other conventional processes and sensitivity analyses. The process developed, "ORIGINAL", is characterized by simplification of the total process and application of an advanced fluidized bed reactor to the OCM reaction. Full economic comparisons between the "ORIGINAL" and conventional OCM technologies were carried out. The results showed that the "ORIGINAL" process is substantially more economical when compared with existing technologies under the same conditions.

Introduction

Many natural gas fields have been discovered recently in Southeast Asia, which has become one of the main areas for oil and gas exploration. In many cases, however, the developments of the discovered gas fields have been obstructed by the demand for large investments in the transportation facilities (e.g., gas pipeline) and other related infrastructure.

Some liquefaction methods of natural gas, which include liquefying natural gas at an extremely low temperature (LNG), and converting natural gas into liquid fuel through the synthetic gas, have already been put into industrial use. These methods, however, also require large investments in the facilities and are expensive to operate. Consequently, even the existing methods remain a significant obstacle in the development of natural gas fields, in particular marginal fields.

Thus, to exploit natural gas resources more widely, a new method is required which converts natural gas into liquid fuel more efficiently and economically than the existing methods. With this in mind, the purpose of this study is to research and to develop a new process which converts methane, the main component of natural gas, into liquid fuel (e.g., gasoline), rather directly than through synthetic gas.

Scope of work

First of all, we devoted our time largely to reviewing the current development trends overseas, and conducted a feasibility study. Among the various direct reaction processes in which methane is converted into a highly reactive intermediate product, the oxidative coupling of methane (OCM) reaction process, in which ethylene is the intermediate product, was chosen from the standpoint of economic efficiency and feasibility in practical use. Furthermore we conducted a sensitivity analysis on the existing OCM processes which we thought promising, to evaluate the impact of various process parameters on the economy of the process itself. A "conceptual view" of the newly integrated "ORIGINAL" process was developed on the basis of these findings and the process was evaluated from the standpoint of economy.

Description of the conventional technology

A sensitivity analysis was carried out on the conventional methane conversion technology[2] [3]. This liquid fuel synthesizing process is based on the OCM. Conceptual block flow diagrams of this process, which have been modified under our consideration to simplify the process, is shown in Figure 1.

This process called Co-feed mode is characterized by point that methane and oxygen are co-fed to the oxidative coupling reactor, therefore air separation unit is required. Pyrolysis of C2 + hydrocarbon is undergone at the upper side of the reactor which oxygen is relatively free. The pyrolysis approach leads to the removal of heat from the OCM reaction and the production of more olefins. Coupling products after separation of unconverted methane and byproducts are fed with oligomerization reactor. Higher hydrocarbon produced are refined to gasoline in a distillation unit. Unconverted methane, carbon oxide and hydrogen mixed with natural gas are fed to a decarbonator to remove carbon dioxide. In the following methanation reactor, carbon oxides are converted to methane by the hydrogen produced in the pyrolysis reactor in order to utilize natural gas effectively.

Sensitivity analysis of existing processes

In discovering factors relative to product costs, we conducted sensitivity tests on the parameters listed below regarding conventional technology. When each factor was analyzed to study the causative effect on economics, other parameters took respective basic values underlined to ignore an effect of themselves.

- (1) Plant scale (natural gas volume): 10,50,100 (* 10 thousand Nm³/D)
- (2) Methane conversion ratio : 10,15,20 (%)
- (3) C2+ selectivity : 70,80,90 (%)
- (4) Reaction pressure : 1,4,10 (Kg/cm²G)
- (5) GHSV : 5000,10000,20000 (hr-1)
- (6) C5+ yield rate : 60,70,80 (%)
- (7) Natural gas price : 0.5,1.0,2.0 (\$/MMBTU)

The impact of variable factors on the gasoline production cost is shown in Figure 2. Within the scope of the present evaluation, the reaction results of the oxidative coupling process had a much more significant impact than those of the polymerization process. In general, several sensitivity studies indicated that C2+ selectivity was more important economically to the OCM reaction than methane conversion ratio. This sensitivity analysis showed that the reaction results for the methane conversion ratio were found to have been subject to a much more significant impact within the various factors involved in the oxidative coupling reaction under these conditions. Among the factors other than the reaction result, plant size was the significant factor which had the most impact on gasoline production cost.

We also carried out the sensitivity analysis with respect to conventional Redox-mode process [1] and got the same tendency with Co-feed mode.

ORIGINAL process implication

A conceptual view of the basic original overall process "ORIGINAL" was developed for gasoline production on the basis of reviews of the conventional processes, sensitivity analysis and reaction results likely to be obtained in the oxidative coupling reaction and polymerization reaction. A general outline of the designed process can be seen on flowsheet (Figure 3).

This process incorporated the OCM and pyrolysis reaction is similar to the Co-feed mode OCM process. It also includes facilities for oligomerization, product separation and methanation. The following two points characterize the new process. In general, with conventional technology wholly recycled gas including unconverted methane is fed to the decarbonation unit to remove CO₂ and treated recycled gas enters the methanator with new additional CO₂ which corresponds with H₂ and CO in order to enhance the carbon utilization efficiency.

Firstly, the flowsheet shows recycled unconverted methane gas, hydrogen, CO₂ and CO are split in two and are fed to a decarbonator and a methanator respectively. Regarding reduction of scale of the decarbonation unit, it is effective to feed directly part of the recycled gas, including

CO₂ corresponding to methanation with the methanator, not through the decarbonator. The integrated process with the conceptual view is economically competitive compared with existing methods because of the reduced plant cost.

Secondly, the question which we must consider is what types of reactor to apply to the OCM reaction. The OCM reaction is very exothermic and operated in a relatively narrow temperature range, placing high demands on heat removal and temperature control. In general, application of the fixed bed for OCM reactions has several disadvantages, difficulty selecting cooling agents and a complicated reactor structure. As for the bubbling fluidized bed reactor, which has a high heat transfer coefficient, scaling-up is known to be an important problem.

This research activity has also led to the identification of new natural gas conversion concepts using a circulating fluidized bed design with a riser reactor. This reactor is used to combine the methane oxidative coupling step with the pyrolysis of ethane and higher alkane components present in natural gas, to provide an efficient method for total conversion of natural gas to liquid fuel. The circulating fluidized bed is schematically illustrated in Fig. 4. The OCM is carried out in the riser reactor as FCC technology and an important feature of the process is pyrolytic conversion within a bubbling fluidized bed, which is the disengaging section located at the top of the riser zone, using heat generated by the OCM reaction via catalyst particles. For the circulating fluidized bed, it is possible to recycle the catalyst continuously, and to replace it if deactivation occurs. The temperature inside both reaction phases can be easily controlled and scaling-up to a commercial size is relatively simple.

Economic evaluations

The concept of the ORIGINAL process was then evaluated from the standpoint of process efficiency. From the sensitivity analysis, we understood that the methane conversion ratio is more important than C₂₊ selectivity under our condition. Therefore we investigated process efficiency and plant costs in a high methane conversion region. A correlation between methane conversion and C₂₊ selectivity, which is derived from extrapolation of catalytic performances of the best catalysts known in literatures at present[4], used in our evaluations is shown in Table 1.

Under these conditions we calculated the investment cost on plant construction regarding ORIGINAL as shown in Figure 5 and found CASE-2 to be the more economical condition. Furthermore the economic evaluations of ORIGINAL and conventional technologies in the region of high methane conversion (25~30%) are summarized in Table 2. The results showed that ORIGINAL is more economically feasible compared with existing technologies.

Conclusion

We have analyzed the relationship and causative effects of several factors on plant and product costs to existing methods. It was noted that the rate of methane conversion was economically significant compared to C₂₊ selectivity under 30% methane conversion. On the basis of these results we have designed a new process with conceptualized the circulating fluidized bed reactor to OCM reaction and carried out the economical evaluations. The result showed that the correlation between about 30% of methane conversion and 77% of C₂₊ selectivity was the optimum condition for reducing plant costs for our process. Furthermore, this ORIGINAL process has shown to be more efficient and economically feasible than conventional technologies under the same conditions.

References

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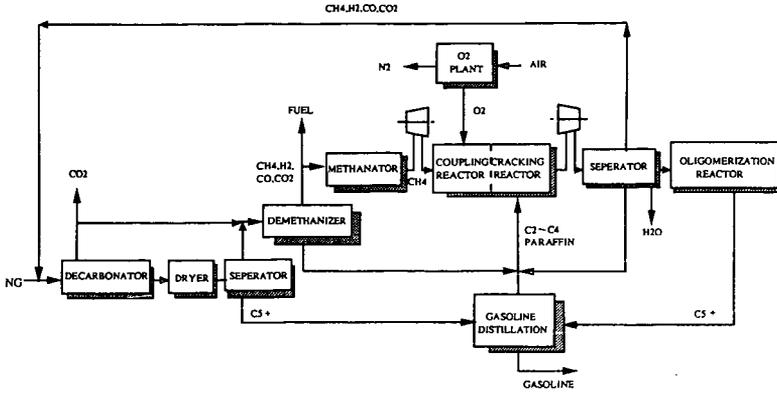


Figure 1. Block Flow Diagram of Conventional OCM Process

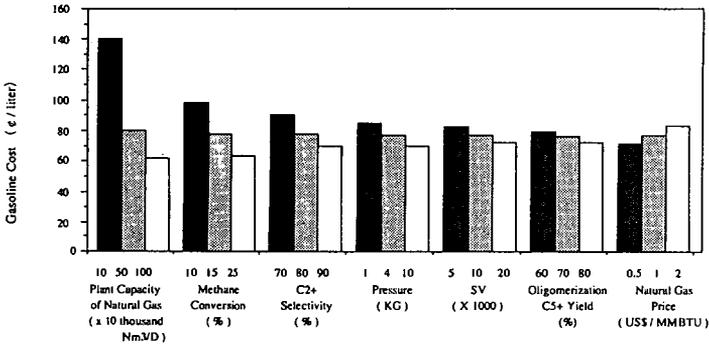


Figure 2. Effects of Various Factors on Gasoline Cost

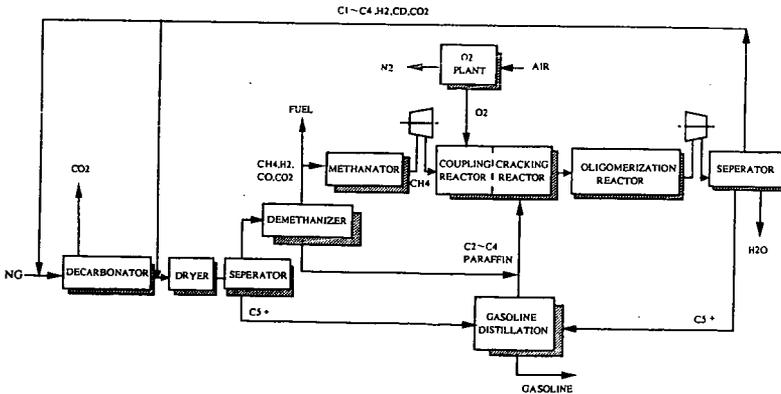


Figure 3. Block Flow Diagram of ORIGINAL Process

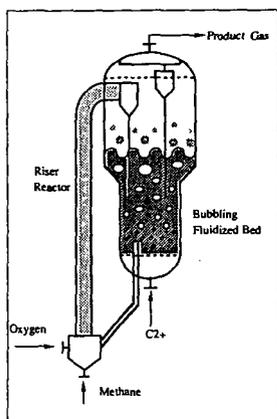


Figure 4. Circulating Fluidized Bed Reactor

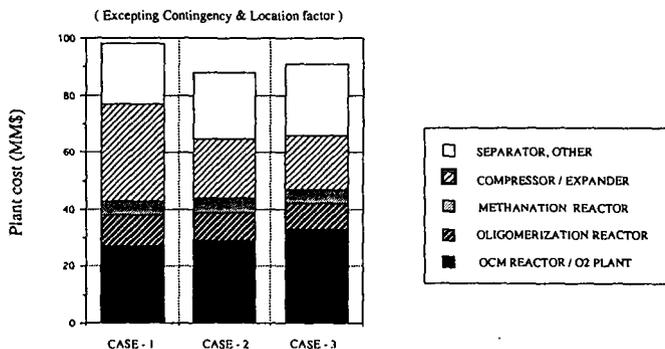


Figure 5. Constituent Proportion of Plant Costs regarding ORIGINAL processes (Plant Capacity : 2,100 BPSD)

Table 1. Estimated Correlation Between Methane Conversion and C₂₊ Selectivity

| | CASE-1 | CASE-2 | CASE-3 |
|---------------------------------|--------|--------|--------|
| METHANE CONVERSION (%) | 20 | 30 | 40 |
| C ₂₊ SELECTIVITY (%) | 84 | 77 | 69 |

Table 2. Comparison of Process Efficiencies and Plant costs (Plant Capacity : 2,100 BPSD)

| | ORIGINAL | OCM (REDOX MODE) | | OCM (COFEED MODE) | |
|---------------------------------|----------|--------------------|------|---------------------|------|
| OCM REACTION CONDITION | | | | | |
| METHANE CONVERSION (%) | 30 | 25 | 30 | 25 | 30 |
| C ₂₊ SELECTIVITY (%) | 77 | 80 | 77 | 80 | 77 |
| PLANT COST (MM\$) | 88 | 103 | 91 | 123 | 111 |
| THERMAL EFFICIENCY (%) | 57.8 | 57.1 | 55.0 | 60.8 | 58.5 |