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# On the design, development and operation of an energy efficient $CO_2$ removal for the oxidative coupling of methane in a miniplant scale

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ABSTRACT

The catalytic oxidative coupling of methane to higher hydrocarbons (OCM) as an alternative route to ethylene or liquid fuels obtained much attention. Besides the intensive research and novel developments in the catalyst field are a number of investigations conducted with reaction and separation engineering aspects at the TU Berlin. The aim of this research is the demonstration of the technical feasibility of the OCM process, including product recovery in a miniplant scale. In this article the gas treatment of the reaction product gas is discussed as a key part of the production chain from raw material to the product. In the OCM process the $CO_2$ is treated as an unwanted by-product or waste. Nevertheless, the energy and the costs requirements for the whole OCM process are crucial for industrial applicability and must be kept as low as possible to operate economically. Therefore different separation methods are examined and several process alternatives are investigated. In this article the energy saving of a hybrid separation process based on a membrane-absorption process for the $CO_2$ capture of the OCM process is presented and proved experimentally in miniplant scale. In a case study of the oxidative coupling of methane (OCM) process the development of an integrated gas treatment process for $CO_2$ capture in the miniplant scale is presented and experimental results are discussed.



## 1. Introduction

Oxidative coupling of methane to ethylene provides with natural- or biogas in combination with other alternative processes [1,2] a new feedstock for the chemical industry and is a promising alternative for the petrochemistry [1]. Due to the yield limitation and high separation costs for by-products, the process is not applied in the industry yet, although several process alternatives were proposed in the literature [2]. In a novel approach of concurrent engineering of the whole process, including the downstream processes are investigated simultaneously in a miniplant scale, to study novel reactor concepts, [3] as well as alternative separation processes [4]. Inevitably, the $CO_2$ has to be removed from the product chain in an early process stage to prevent condensation and freezing in the further product separation unit, which is a cryogenic distillation in the state of the art process. While research for energy optimal $CO_2$ capture processes focus in the last years on power plants [5,6], natural gas treatment and other industrial processes [7], these knowledge has to be applied and transferred in an early process design stage.

In this article is the focus on the separation process for 90% $CO_2$ removal under the particular OCM conditions of up to 25 mol% $CO_2$ in the raw gas at a feed pressure of 32 bar. Beside other possible separation principles the chemical absorption process with 30 wt% Monoethanolamine was figured out as a as state of the art process and therefore as the OCM base case $CO_2$ removal process [8]. After extensive preliminary investigations of [9] and [10], a flexible miniplant system was designed, constructed and put into operation. The design of an economical downstream gas refining process is a project goal, besides improving of the catalysts, due to the moderate ethylene yield for currently available OCM catalysts of 30%. This can be achieved only through an integrated downstream concept that is based on energetically and economical enhanced processes.

To investigate the entire process concept concurrently, the process is divided into three units (Fig. 1): the reaction unit, the gas scrubbing unit and the product separation unit.

Based on a holistic view and on the approach of simultaneous process development, the design tasks for each unit and stream purities are defined considering their interactions among each other. Based on the process schematic of the flow diagram in Fig. 1,

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