



# Effect of active thermal insulation on methane and carbon dioxide concentrations in the effluent of a catalytic partial oxidation reactor for natural gas conversion to synthesis gas

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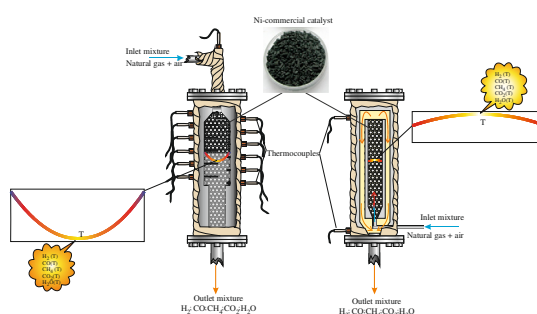
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## HIGHLIGHTS

- Local thermodynamic equilibrium is established in CPOX pilot scale reactors.
- CPOX products composition is controlled by heat transfer processes in catalyst beds.
- Active thermal insulation has a positive effect on conversion.
- Preheating up to 700 °C allows for reduction in CO<sub>2</sub> content to 0.4 vol.%.
- Catalyst deterioration is observed at bottom–top gas filtration through reactor.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Two reactor designs with either passive or active thermal insulation were tested to produce synthesis gas from natural gas using catalytic partial oxidation on a system that exceeded laboratory scale. Preheating the working mixture significantly improves the conversion parameters. Preheating levels are limited by two factors: mixture ignition in the heater and overheating at the catalyst bed inlet area. The use of hot conversion products to provide active thermal insulation significantly improves the quality of the conversion. Because the same amount of identical catalyst was used in both reactors, the observed effect supports the hypothesis that the product composition depends primarily on the heat transfer process.

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## 1. Introduction

Efforts to more efficiently use natural gas reserves have raised interest in synthesis gas (syngas) production, which is currently used for a broad number of applications [1] including modern

gas-to-product technologies [2] and fuel cells [3]. Steam reforming is frequently used for syngas and hydrogen production; however, this process is not ideal in terms of time or efficiency. The catalytic partial oxidation (CPOX) process is an alternative to steam reforming [4]. CPOX is more energy efficient [5]. Comparative thermodynamic analyses have confirmed that partial oxidation process has advantage over steam reforming process for syngas production [6]. Highly endothermic steam reforming processes require

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external heating [7] with intensive heat delivery. Steam reforming process requires 2.8 times higher energy supply than partial oxidation process to convert one mole of methane to synthesis gas at 1000 °C. In the face of rising natural gas prices, the partial oxidation process is becoming more economically attractive, especially with small and medium production capacities [6,8].

Numerous studies have described and analyzed CPOX of hydrocarbons. These studies primarily focused on methane, which is the principal constituent of natural gas. Recent reviews have covered the major findings in the field and refer to key publications on theoretical and experimental CPOX processes, catalysts, and reaction kinetics [4,9]. An overwhelming majority of these publications involved the investigation of fundamental CPOX processes and were aimed at clarifying the catalysis kinetics, developing effective catalyst compositions (active agent/promoter/carrier), studying catalyst degradation mechanisms, and increasing catalyst service life.

Typically, kinetic methane CPOX studies are investigated experimentally with a laboratory-scale reactor [10–16]. It has been established that, noble and transition metals are feasible CPOX catalysts, meaning that in laboratory conditions, the thermodynamic equilibrium composition is reached for 10–50 ms. These characteristic residence times mean that the chemical reaction rate is “very high”.

At the same time, active research on laboratory scale operating in kinetic mode are conducted where the selectivities of the various components of the produced synthesis gas depend on choice of residence time [17]. Such effect is particularly observed in the membrane reactors employed to remove hydrogen efficiently from the reaction volume [18]. High flow rates are not required to exclusively study scientific aspects of catalytic processes. In laboratory studies typical flow rates are in the range of 50–300 sccm [18]. Implementation of scientific concepts developed using laboratory to scale up processes to industrial plants usually causes a number of problems that do not occur and are not discussed in case of small scale installations.

Despite some advantages of reactors operating in kinetic modes all industrial partial oxidation plants are exploited at equilibrium conditions. The design of an industrial-scale plant requires optimization of the heat and mass transfer processes rather than identifying effective catalysts. The most feasible catalyst is primarily chosen based on its cost and durability at approximately 1000 °C.

To scale up the methane CPOX process for syngas production from laboratory scale to industrial scale, a number of engineering problems have to be solved, as the heat and mass transfer coefficients vary substantially. The temperature and concentration distributions within the reactor are formed by heat and mass transfer processes interactions. These phenomena cannot be studied properly using a laboratory-scale reactor. A few publications have been devoted to elaborating on industrial-scale reactor concepts [19,20]. The purpose of this article is to demonstrate that when developing industrial scale CPOX reactors, the greatest impact is obtained from heat transfer rather than from catalyst “efficiency”.

This study expands on the previously published work [21], and testing of the two partial oxidation reactor designs is described. The methane-air mixture CPOX experiments described previously [21] were conducted with a non-preheated inlet mixture with a wide range of CH<sub>4</sub>/air equivalence ratios. The low process temperatures [21] were due to the lack of an external energy supply. These regimes were characterized by minimal heat loss in contrast to hotter regimes, but even in these conditions, the product composition appears to be worse than with adiabatic equilibrium.

The four different catalysts investigated showed no significant differences among their degrees of approximation to the equilibrium composition. This fact led to the conclusion that the deviation in product composition with adiabatic equilibrium is associated with heat loss rather than the lack of residence time.

In the case of an inlet mixture with a C:O ratio of ~1, the obtained CPOX product composition at the reactor outlet was approximately 28% H<sub>2</sub>, 13% CO, 7% CH<sub>4</sub>, and 4% CO<sub>2</sub> (volume percents correspond to the drained mixture), whereas the adiabatic calculation gives 32.8% H<sub>2</sub>, 15.2% CO, 5.5% CH<sub>4</sub>, and 2.6% CO<sub>2</sub>. A simple model assuming local thermodynamic equilibrium showed that the observed deviations could be explained by a non-homogeneous radial temperature profile in the catalyst bed that formed due to heat loss. The calculations predicted a temperature difference of approximately 20 °C between the reactor axis and the cylindrical catalyst surface.

These calculations were based on the assumption that equilibrium was achieved, meaning that “sufficient” residence time was achieved for the working mixture in the catalyst bed. Although the amount of catalyst used and the flow rates of the working mixture correspond to the CPOX residence times described by Korchnak et al. [22], the issues regarding the “adequacy” of the catalyst sample residence time tested by Al-Musa et al. [21] and the actual value of the radial temperature difference remain disputable. The residence time required to reach equilibrium is a key characteristic of the catalyst in CPOX applications. This characteristic depends on the temperature of the fed gas [23], the catalyst grain size, the effective surface area, and the flow conditions. A theoretical study on the effect of heat transfer and particle size on CH<sub>4</sub> partial oxidation was published previously [24]. Periodic temperature perturbations were studied, but the effects of filtration field behavior changes were not considered. In the presence of radial temperature gradients, clearly identifying the impact of residence time on the degree of the approach to equilibrium based on the composition of the outlet mixture is difficult, because the temperature profile affects the chemical process rate and the flow field in the catalyst bed. Studies of mass transfer intensity (i.e., the necessary residence time) should be carried out in a reactor operating at nearly isothermal conditions.

Our aim was to improve the composition of CPOX products using preheating of the inlet mixture and by implementing a novel reactor design to decrease heat loss and therefore lower CH<sub>4</sub> and CO<sub>2</sub> content.

## 2. Materials and methods

### 2.1. Experimental setup of passive thermal insulation

The reactor dimensions and performance have been described in detail previously [21] and are shown in Fig. 1. In the present study, the potential for increasing the conversion depth by preheating the reaction mixture in this reactor was investigated, and the temperature field in the catalytic bed was estimated. For these experiments, 12 thermocouples were installed to record the temperatures. Thermocouples 1, 3, 4, 5, 7, 9, and 11 were located along the reactor axis, whereas thermocouples 2, 6, 8, 10, and 12 were displaced from the reactor axis towards the edge of the thermal insulation (Fig. 1). Ni/γ-Al<sub>2</sub>O<sub>3</sub> catalyst produced by Liaoning Haitai Sci-Tech Development Co., Ltd., China, was used. This granular catalyst with 16 wt% Ni content has cylindrical shape with dimensions, 3.5 mm in length and 1.0 mm in diameter. BET surface area is 91.5 m<sup>2</sup>/g. The volume of the catalytic bed used in experiments is about 327 cm<sup>3</sup>.

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