HYDROGEN GENERATION BY PARTIAL OXIDATION OF METHANE IN SUPERADIOBATIC COMBUSTION

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The compact and efficient hydrogen production units for promising hydrogen fuel cell transportation power plants are needed. Among them are hydrocarbon (especially natural gas or methane) reforming units. Currently available methods of natural gas converting include catalytic or non catalytic steam reforming, pyrolysis and partial oxidation.

Recent studies [1-5] demonstrate, that hydrogen can be produced by hydrocarbon partial oxidation in filtration combustion (FC) wave without external heating of reaction zone or using expensive and prone to poisoning catalyst. In this process the temperature needed is gained by "super adiabatic effect" of FC wave. The methane to hydrogen conversion efficiency in such system is strongly dependent on maximal temperature of FC wave, which increases with specific mass flow rate of fuel mixture **g** [2,3].

This paper contains the results of studies made in a more wide range of equivalence ratio γ and **g** to explore optimal methane conversion operating mode in filtration combustion wave. Another goal is to improve the model of numerical simulation of combustion wave using experimental data.

Now we have enough evidence to suspect that well known kinetic patterns of Konnov, GRI, Miller-Bowman, Frenklach, Warnatz are not quite adequate in the case of partial oxidation. One of the probable reasons of such a discrepancy is effect of heterogeneous reactions on a surface of packed bed particles.

The experimental set up consists of porous medium combustion reactor, reactor preheat unit, gas flow rate control equipment, temperature measurement system, gas chromatograph and data acquisition system. The reactor case with 140 mm outer diameter and 6 mm wall thickness is made of stainless steel. The reaction zone is filled with 5-6 mm alumina spheres. The space around packed bed is filled with pressed Kaowool insulation. The porous medium preheat unit as a methane fuelled porous medium burner is provided to start the combustion in reactor.
Air is taken from the high pressure line while methane is taken from the standard cylinder. Flow rates of fuel components are controlled and measured by mass flow controllers. The axial reactor temperature distribution is measured with S-type thermocouples with exposed junction. A PC based data acquisition system was used to process the thermocouple signals. Modified "Chrom 4" gas chromatograph with digital signal processing was used for gas product analysis.

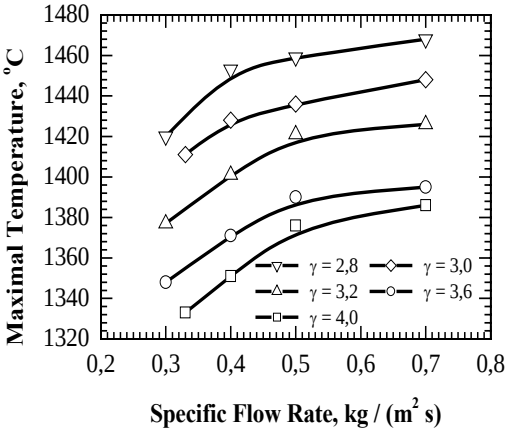

Fig. 1. The effect of equivalence ratio and mass flow rate on maximal temperature

Present study show that efficiency of methane to hydrogen conversion in superadiabatic combustion wave is directly related with maximal reactor temperature.
This temperature is a function of specific flow rate **g** and equivalence ratio γ and is given in Fig.1. For all values of γ a tendency of maximal temperature growth with g is clearly seen, especially for $\mathbf{g} < 0{,}6$. This tendency completely agrees with our previous experimental and numerical simulation results for $\gamma = 4$ [3].

The most sensitive parameters of conversion efficiency are concentration of residual methane in reaction products and methane-to-hydrogen conversion. These parameters are given in Fig.2 and Fig.3 respectively.

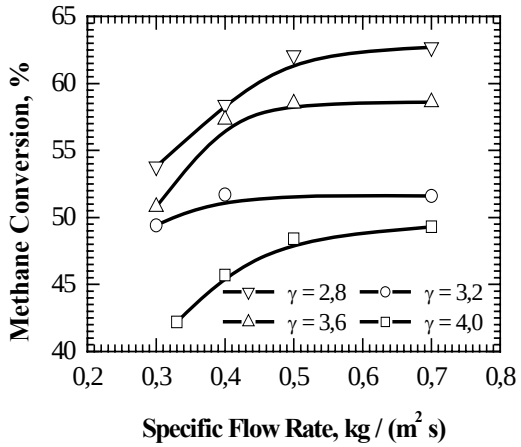

Fig.2. Methane to hydrogen conversion factor

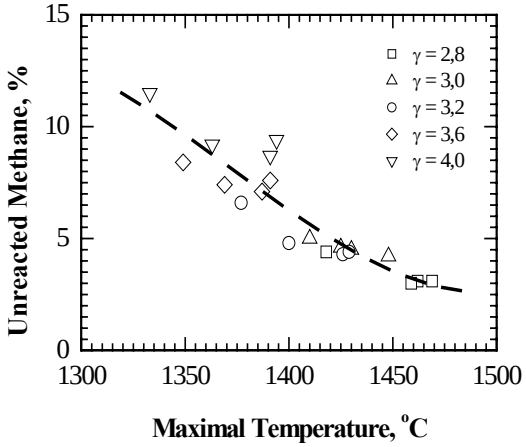

Fig.3. Methane content in reaction products

Partial oxidation of methane can be performed in filtration combustion wave propagating in inert porous medium. The most efficient conversion process seems to be for $\gamma \approx$ 2,8-3. The maximal methane-to-hydrogen conversion efficiency is about 63%. One of the most attractive features of the process is that no soot is produced.

References

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