THE INFLUENCE OF ETHANE ADDITION ON OPERATING CONDITIONS AND ACETYLENE PRODUCTION BY THE NON-CATALYTIC PARTIAL OXIDATION OF METHANE

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Summary

The non-catalytic partial oxidation of methane under fuel-rich conditions was studied in a nozzle type reactor. The influences of preheat temperature, oxygen ratio and ethane addition were studied. The axial species concentration profiles were measured with a mass spectrometer using a sampling probe that could be moved along the length of the reactor. Increasing the preheat temperature was favorable for acetylene production. For a methane feed of 5 L/min, the maximum acetylene concentration (0.0567) was at 140 mm distance from the nozzle and oxygen ratio of 0.58~0.60. When 5% ethane was added, the maximum acetylene concentration (0.0587) was at 130 mm and oxygen ratio 0.60~0.62.

Keywords

Non-catalytic partial oxidation; Methane and ethane; Acetylene; Species concentrations;

Introduction

Non-catalytic partial oxidation is a proven efficient process to utilize methane and other low hydrocarbons. Besides methane, other low carbon number hydrocarbons also exist in low but significant concentrations in coal bed gas, coke oven gas, refinery gas and some natural gas. It is useful to use these to produce chemicals. The design and control of thermal partial oxidation is difficult because of the very high temperature, fast mixing, intensive turbulence, and very rapid reaction and quenching.(1) Although C_2 and higher hydrocarbons are often <10% in natural gas and other feed gases, their concentrations significantly affect the optimum operating conditions.(2, 3) In this work, the influence of C_2 and higher hydrocarbons was studied with mixtures of methane and ethane to quantify the influences on the operating conditions.

2. Experimental

The experiments were carried out in a nozzle type reactor. The experimental apparatus is shown in Fig. 1. The nozzle was 5 mm in diameter. The quartz reactor chamber was 10 mm i.d. and 200 mm in length. Pure methane, ethane and oxygen from cylinders were used as reactants. Gas flow rates were controlled by mass flow controllers. The feed gases were first heated in tubular heaters and then mixed in the mixing chamber. The flame was stabilized by a secondary oxygen flow at the sides of the nozzle. Ignition was by electrode discharge. Species concentration axial profiles were measured with a mass spectrometer using a movable sampling probe placed inside the reactor, which was designed to avoid further reaction in the sampling line. The tip of the sampling probe was 100 μm i.d. and 5 mm long, and the transfer line was 1 mm i.d. The low pressure (less than 100 Pa) and decreased temperature in the sampling probe made further reactions in it very slow.

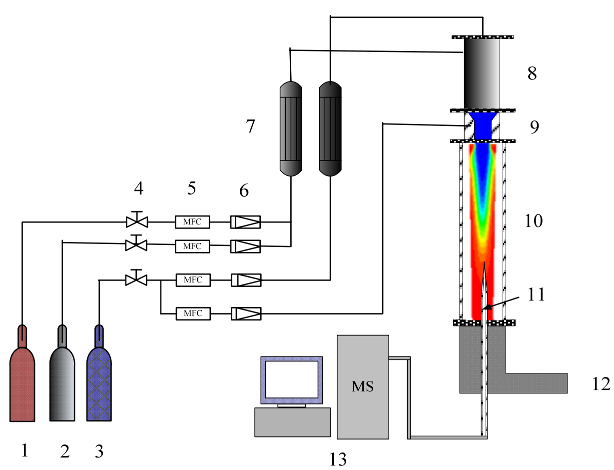

Fig. 1. Schematic of the experimental apparatus
1-oxygen cylinder, 2-methane cylinder, 3-ethane cylinder, 4-stop valves, 5-mass flow meters, 6-check valves, 7-tubular heaters, 8-mixing chamber, 9-nozzle, 10-reactor, 11–sampling probe, 12-gas tank, 13-mass spectrometer.

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3. Results and Discussion

The main operating parameters that influence the partial oxidation are preheat temperature, oxygen ratio, and reaction time. C_2 and higher hydrocarbon concentrations also have significant influences. The oxygen ratio was defined as the molar ratio between oxygen and methane for pure methane system, and between oxygen and the mixture of methane and ethane when ethane was added.

3.1 Influence of preheat temperature

In partial oxidation, acetylene is produced by the pyrolysis of methane at a temperature determined by the heat released from methane oxidation. An increased preheat temperature can increase the reaction temperature and is favorable for acetylene production because pyrolysis is endothermic. However, the preheat temperature also has to be limited to prevent premature ignition in the mixing chamber and the pyrolysis of methane in the heater.[(4)]

3.2 Influence of oxygen ratio

In partial oxidation, the methane reactions can be divided into two types: the exothermic oxidation reactions to increase the temperature, and the endothermic pyrolysis reaction to produce acetylene and H_2.[(4,5)] For maximum acetylene yield, there is an optimum oxygen ratio to balance the amounts used to produce heat and used in pyrolysis. The influence of oxygen ratio from 0.52 to 0.60 on the concentrations at different axial distances from the nozzle is shown in Fig. 2. The maximum acetylene concentration (0.567) was at 140 mm and oxygen ratio of 0.58~0.60. When oxygen ratio was above 0.60, acetylene concentration decreased with an increase in oxygen ratio.

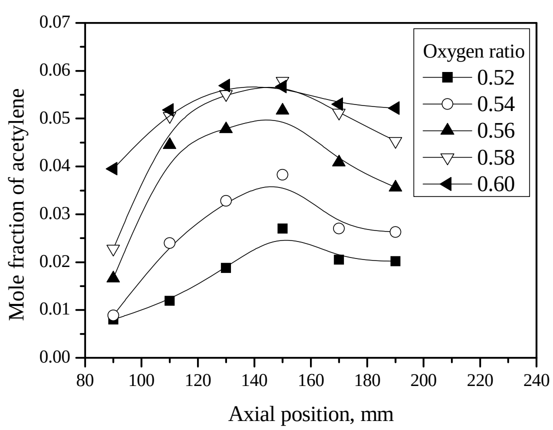

Fig. 2. Axial concentration profiles of acetylene with pure methane as reactant (Q_{CH4}=5NL/min, T_{mix}=320^{o}C)

3.3 Influence of ethane addition

Ethane reacts more easily than methane [(3)] and its C/H ratio is higher than methane. Both these aspects make it better for the production of acetylene. As shown in Fig. 3, with added ethane, the maximum acetylene was obtained at the axial position of 130 mm and oxygen ratio of 0.60~0.62. Compared with methane, a larger concentration of acetylene (0.587) was obtained at a shorter axial position and with a higher oxygen ratio.

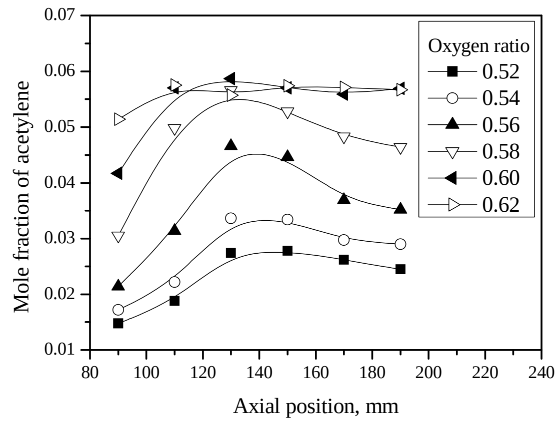

Fig. 3. Axial concentration profiles of acetylene with 95%methane+5%ethane as reactant ($Q_{CH4+C2H6}$=5NL/min, T_{mix}=320^{o}C)

4. Conclusions

In the non-catalytic partial oxygen of methane, a higher preheat temperature is favorable for acetylene production. With a methane feed of 5 L/min, the maximum concentration of acetylene (0.0567) was obtained at the axial position of 140 mm and oxygen ratio of 0.58~0.60. When 5% ethane was added to use a mixture feed, the maximum acetylene concentration (0.0587) was obtained at the axial position of 130 mm and oxygen ratio of 0.60~0.62.

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