CHARACTERISTICS OF PLASMA BLACKS PREPARED BY PLASMA PYROLYSIS OVER METALS COATED HONEYCOMB CATALYSTS

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Introduction

The development of environmental friendly method for producing hydrogen has attracted due to clean and efficient use of fossil fuel [1]. Thus, the direct disintegration of methane, main component of natural gas, has been interested because the conventional steam reforming process should be a negative effect on global warming through CO_2 emission [2]. Plasma pyrolysis of methane exploits the thermochemical properties of plasma. According to Fulcheri et. al [3], the total enthalpy of methane decomposition at 1600□ is 43.5kcal/mol and the energy, related to carbon mass, varies approximately between 3 and 5 kWh per kg of carbon produced. The direct conversion methane through plasma pyrolysis produces not only hydrogen but also carbon black without CO_2 release. Therefore, practical use of carbon black produced has experimental and theoretical interest in order to meet the economical cost of the plasma pyrolysis process.

In the present study, four kinds of plasma blacks were prepared by plasma pyrolysis system under various metallic catalysts coated on honeycomb, and investigated the catalytic effect on the characteristics of the plasma blacks prepared under plasma pyrolysis condition.

Experimental

The experimental process with a microwave plasma system is presented in Figure1. In order to investigate the plasma catalytic reaction, the reactor was specifically designed. Plasma catalytic reactions were carried out with microwave plasma (2.45GHz, iplas Co.) to generate hydrogen and carbon black from methane using a plasma reactor of quartz tube of 6” O.D. that is connected to the microwave waveguide and resonator. The producing hydrogen is analyzed by a thermal conductivity analyzer (Teledyne, 2000A-EU) and physicochemical properties of plasma blacks are determined in terms of SEM, BET and TGA.

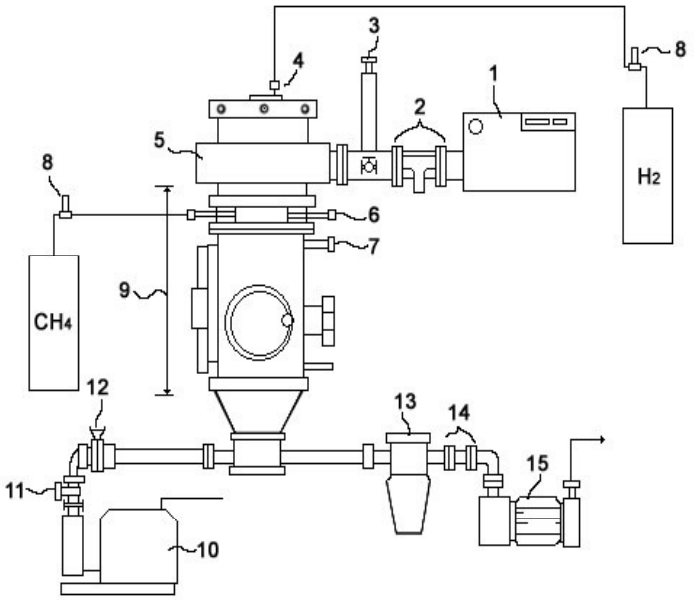

Figure 1. Experimental apparatus for microwave plasma and catalytic reaction system; (1) Magnetron, (2) Waveguide, (3) E–H tuner, (4) Microwave quartz tube, (5) Plasma Generator, (6) Jet inlet nozzle, (7) Cooling line inlet, (8) MFC, (9) Microwave plasma reactor, (10) Vacuum pump, (11,12) Control valve for pressure, (13) Cyclone, (14) Filter, (15) Mechanical diaphragm pump

Results and Discussion

The effect of the CH_4 feed amount on the yields of H_2 and plasma blacks is given in Figure 2. As can be seen, yields of hydrogen and carbon black increases with the amount of CH_4. When an input of 1 mole of methane was introduced, 0.6 mole of carbon and 1.62 mole of hydrogen were produced.

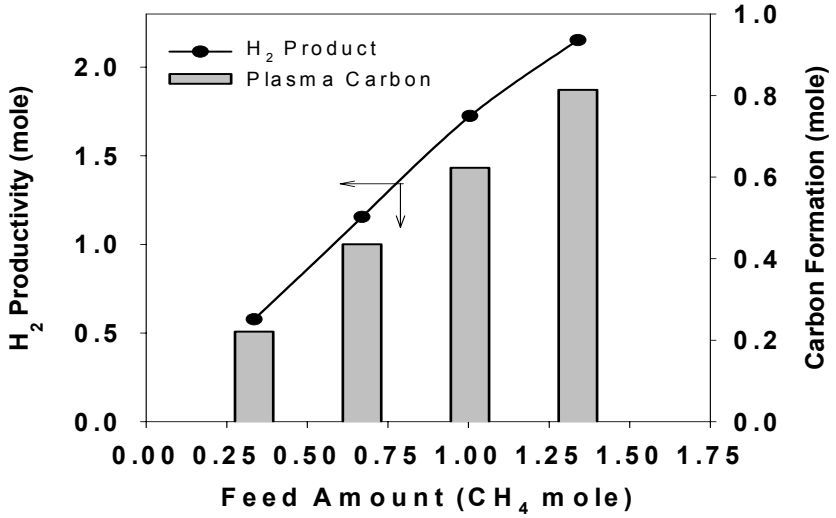

Figure 2. Effect of CH_4 feed on the yield of H_2 and carbon

Physicochemical properties of plasma blacks prepared by plasma pyrolysis over various metals coated honeycomb catalysts are summarized in Table 1. Pt, Pt-Rh, and Pd catalysts were employed as active materials to prepare the plasma blacks. In the experimental range studied, the metallic catalysts reduce the BET surface area, H/C ratio, and electrical resistivity. The data show that dense particle of plasma blacks is produced by the presence of metallic catalysts. The observation indicates that presence of the metallic catalyst reduces the electrical resistivity of plasma blacks due to the decrease in the amount of oxygen functional groups. The highest electrical conductivity of plasma black was observed in the Pt catalyst and then followed by those Pt-Rh, Pd and bare cordierite honeycomb.

Table 1. Physicochemical properties of plasma blacks prepared by plasma pyrolysis over various metals coated honeycomb catalysts.

Plasma black / Properties	Honey comb	Pt	Pt-Rh	Pd
BET surface area (m^2/g)	196.8	130.6	150.4	149.9
H/C ratio	0.0146	0.0095	0.0117	0.0126
Resistivity (Ω・㎝) (at 0.2 g/cm^2 density of plasma black powders)	1933.56	574.72	593.74	1010.69

Conclusions

A microwave plasma couple with catalytic reaction system was employed for production of H_2 and carbon black from natural gas. Pt, Pt-Rh, and Pd catalysts were used as active materials to prepare the plasma blacks. Presence of the catalysts enhanced the decomposition of methane to H_2 and carbon black by activating the reaction of radicals. In the experimental range studied, the metallic catalysts reduced the surface area, surface oxygen content and electrical conductivity of the plasma blacks prepared. It should be also noted that more dense particle of plasma blacks were prepared under existence of metallic catalysts. The highest electrical conductivity of plasma black was observed in the Pt catalyst and then followed by those Pt-Rh, Pd and bare cordierite honeycomb.

References

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