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Plasma Catalyzed Non-Oxidative Coupling Conversion of Methane to Hydrogen and Ethane

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Abstract

Under standard environmental conditions, methane coupling and plasma pyrolysis were carried out. With varying H_2/CH_4 ratios and N_2 flow rates, methane was coupled through gliding arc plasma with conversion ranging from 25% to 40% and high acetylene selectivity above 90%. Plasma emission spectra showed that the IH/IH ratio was lowest at an H_2/CH_4 ratio of 3, and that it increased as N_2 feed rate increased from 0 L/min to 2 L/min, varying similarly to acetylene selectivity. With increasing Pd loading and Ag/Pd ratio, metallic Pd species (335.4 eV) were transferred to electron rich Pd species (334.8 eV and 334.1 eV), which resulted in a red shift of the linear adsorbed CO stretching frequency from 2060 cm^1 to 2068 cm^1 to 2055 cm^1-2015 cm^1. This was shown by X-ray photoelectron spectra for reduced catalysts. With 86.5% selectivity of ethane and 36.2% conversion of methane, superior plasma catalytic performance above 0.1 Pd 0.5 Ag/Al_2O_3 catalysts has been attained.

***Keywords:** Methane plasma catalysis; Hydrogen and ethene production; Gliding arc; Pd based catalyst; Plasma*

Introduction

Direct conversion of methane, the second largest contributor to historical global warming, has steadily gained attention due to the urgency of the stated "carbon neutral" strategy. The hydrogen economy is incredibly appealing from a societal and technical standpoint as a potential remedy for the energy crisis and its effects on the environment. Methane has the highest C:H ratio (4:1) and can be used as a green hydrogen source to generate high value compounds with no carbon emissions and realize clean energy conversion. As a result, methane has tremendous potential value as a source of clean fossil energy or as a raw material, and it may be economically brought to use. Methane Steam Reforming (MSR), Methane Dry gas Reforming (MDR), methane Oxidative Coupling reaction (OCM) and methane Non-Oxidative Coupling reaction (NOCM) are the primary steps in the process of producing hydrogen from methane. The manufacture of light olefins like ethylene, propylene and benzene important building blocks for a variety of commodities can be accomplished by the methane coupling reaction. Due to the potential for economic gain, studies on coupling reactions have drawn a lot of attention for many years. However, its industrial application is constrained by a high working temperature and poor reaction efficiency. Non-thermal plasma can initiate the methane coupling reaction at lower temperatures, according to recent research on unconventional technologies for plasma conversion of methane.

Description

Plasma conversion has advantages over conventional processes in terms of safety, environmental friendliness and adaptability, which can help it overcome the drawbacks of the thermal reaction process. Explosion risk during reactant mixing is eliminated and high temperature tolerance is improved by the anaerobic reaction procedure. Atomic usage can achieve 100% because of no CO_2 output. Plasma reactors with high throughput that are smaller in size have the potential to be compact, distributed hydrogen production units that are incorporated into hydrogen energy systems. Different plasma forms, such as thermal arc Dielectric Barrier Discharge (DBD), microwave plasma Gliding Arc Discharge (GAD), etc. which have various mechanisms of methane dissociation, can be produced with various plasma generator structures and discharge settings. Despite the fact that thermal arc discharge technology is still in its infancy, temperatures beyond 3500 K tend to produce carbon black, hydrogen and a negligible amount of acetylene. Dielectric barrier discharge can evenly spread the arc, but due to the dielectric's weakening of the electric field and low electron density, ethane is the principal product. By varying the microwave power, the microwave plasma may flexibly alter the product. The product can be transformed from ethane to acetylene by increasing the methane input energy.

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However, the development of microwave plasma is still hampered by energy consumption and power supply design. In hydrogen atmospheres, gliding arc plasma, heated plasma, mostly produces acetylene. The straightforward reactor design offers more room for reducing the energy required for methane conversion, making it appropriate for expanding industries. To efficiently convert methane into ethylene at the moment, a catalytic procedure must be used on the plasma products before they can be further transformed. Because the process conditions are more favourable than those of ethane reforming, acetylene hydrogenation is preferred over ethylene synthesis. Pd has a strong hydrogenation activity at low temperatures, making it an ideal active metal for acetylene hydrogenation catalysts. However, acetylene can be easily hydrogenated into ethane using a monometallic catalyst. Acetylene hydrogenation's product distribution can be adjusted by the effects of space geometry and the electrical environment. Therefore, it is necessary to add a second metal, such as Ag or Zn to Pd catalysts in order to enhance their electrical characteristics and reduce the size of their Pd particle constituents. PdAg and PdCu catalysts are two potential bimetallic Pd-based catalysts that have garnered a lot of interest. K modified Pd and PdAg/zeolite catalysts for the selective hydrogenation of acetylene were investigated by Huang and colleagues. It was determined that the addition of Ag to Pd/K^{+}-zeolite catalyst produced a high degree of ethylene selectivity and minimal hydrogenation activity. In comparison to all hydrogenation to ethane over monometallic Pd catalysts, Datye, et al., conducted acetylene hydrogenation tests with 99.3% conversion and 50% selectivity to ethylene over $PdAg/SiO_2$ catalysts. Including Khan $PdAg/Al_2O_3$ displayed similar catalytic activity to previously reported PdAg catalysts when the adsorption and co-adsorption of acetylene and ethylene on alumina supported Pd-Ag catalysts were studied. However, Panpraot's investigation of nanocrystalline titania as supports for Pd and Pd-Ag catalysts led to low activity and selectivity for the hydrogenation of acetylene to ethylene over $PdAg/TiO_2$ catalyst. For the hydrogenation of acetylene over Pd and Pd Cu catalysts, Leviness, et al., discovered that copper addition to palladium lowered the catalytic activity and enhanced its stability. PdCu catalysts greatly increased selectivity in the semi hydrogenation of internal and terminal alkynes, according to Markov and colleagues. Yang, et al., produced solid palladium-copper alloy based PdCu/C catalysts to increase the ethylene selectivity while maintaining high activity. To validate the high performance from the modification of Ag and Cu on Pd, some studies using DFT calculations have simulated the composition of PdAg and Pd Cu alloy as well as the adsorption process of acetylene and hydrogen on the alloy surface. When Huang investigated the catalytic behaviour of Pd with a Group IB bimetallic catalyst, he discovered that the selectivity of the PdAg catalyst was superior to that of the PdCu catalyst. According to Kim, et al., when promoters were introduced impregnation, Ag promoted catalysts had higher ethylene selectivity and lower activity than Cu promoted ones. Our research indicates that the industrial method for the purification of ethylene is very different from the conversion of methane through plasma with an H_2 atmosphere, which can result in high temperature pyrolysis gas comprising roughly 70%-90% hydrogen, 3%-8% acetylene and 5%-15% methane. However, activity and selectivity for the hydrogenation of trace acetylene at low temperature may not be comparable to catalytic performance over conventional PdAg catalyst at high temperature and hydrogen rich environment

Conclusion

In this study, we built a new PdAg catalyst for acetylene hydrogenation of plasma and designed an effective plasma catalytic method for the conversion of methane to ethylene. The reaction mechanism of active species from methane and hydrogen during gliding arc plasma discharge has been explained in conjunction with experimental findings and emission spectrum. The electron characteristics of the Pd metal surface were studied using X-ray Photoelectron Spectroscopy (XPS) and Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) of CO adsorption. It has been determined how to balance the prevention of excessive hydrogenation with the high activity of acetylene hydrogenation.