Study of catalytic properties of microwave-induced plasma in CO₂ utilization reactions

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Abstract: Two microwave-induced plasma reactors are being investigated. As first approach, a small scale plasma reactor (100 - 300 ml/min) so-called Surfatron is studied to gain insight into gas/plasma phase reactions. A parametric study of various key gas phase reactions is being performed. Furthermore, an up-scaled plasma reactor (100 l/min) will be utilized to understand the needs towards developing an industrial scale plasma reactor for chemical synthesis. Primary results from the small scale plasma reactor are shown.

Keywords: microwave-induced plasma reactor, CO₂ utilization, chemical synthesis

1. Introduction

In the context of developing novel alternative energy technologies, plasma processing applications are investigated. The main objective is to study the catalytic properties of microwave-induced plasma so that it can be used as an alternative for heterogeneous catalysts, thus avoiding its economic and operational problems, i.e., catalyst deactivation, expensive manufacturing processes. In particular, plasma sources can provide both energy (high temperature) and highly reactive species (electrons, ions and radicals) simultaneously. Hence, aimed reactions can selectively and efficiently be activated [1]. Carbon dioxide (CO₂) utilization reactions are considered as CO₂ is the main contributor (by 60%) to the total greenhouse gas emissions. It can be utilized as carbon source for production of valuable chemicals such as methanol, DME and transportation fuels. Therefore, relevant reactions include: reverse-water-gas-shift reaction (RWGS) and dry reforming of methane. Other reactions might be investigated as well, steam reforming or partial oxidation of methane and N₂ hydrogenation for ammonia production.

2. Experimental description

A solid-state microwave generator (MiniFlow 200S, Fig. 1a) in combination with an electromagnetic surface wave launcher (Surfatron, Fig. 1b) is used to ignite and sustain plasma. The microwave generator is operated at 2.45 GHz with a maximum power of 200 W. As for analytical techniques, mass spectroscopy is employed to quantify product composition whereas optical emission spectroscopy enables the characterization of plasma parameters (electron temperature and density) as well as gas temperature which are relevant for the performance of plasma-assisted reactions.

3. Modeling work

A two-dimensional axisymmetric model is presented. A multiphysics approach is taken towards developing the model, which includes gas flow, electromagnetic wave propagation, electron/ion transport properties, heavy species transport properties and heat transfer. Up to date, the model is based on argon plasma. However, other chemistries are being studied such as CO₂ plasma, H₂ plasma, CH₄ plasma and also gas mixture plasma. A schematic representation of the computational domain is shown in Fig. 2. In order to reduce complexity of the model, an antenna parallel to the axes of symmetry is considered. By comparing the EM field distribution of a 3-D geometry (perpendicular antenna) with the 2-D axisymmetric approach, it is proven that this assumption does not affect such distribution [3]. Key plasma parameters as well as the temperature of heavy species can be predicted by the model. Among the plasma parameters, electron number density (1/m³) is presented in Fig. 3. A maximum value of 4.4 x 10²⁰ m⁻³ is reached in front of the excitation surface in which a maximum electric field strength (V/m) is also observed. The results are in good agreement with previous microwave plasma modeling work [3-5].
4. Results and discussion

The RWGS reaction is successfully performed in a non-catalytic microwave-induced plasma reactor. CO₂ conversion is found in the range of 50 - 75% (red dots, Fig. 4). Several advantages are identified when comparing the non-catalytic plasma process to the thermal counterpart, (1) compactness, it eliminates the need for preheating reactants upstream the reactor; (2) clean process, no by-products formation is noticed as compared to previous studies [2], which in turn decreases the recycle ratio and flow rate of the purge stream; (3) simplicity and low price in terms of equipment; (4) super-equilibrium conversions due to the catalytic properties of plasma (excellent chemical medium); (5) no need for solid catalyst avoiding aforementioned issues; (6) milder operating conditions (non-thermal plasma), the conventional process requires 973 K to reach the same conversions (Fig. 4). Nevertheless, it is observed that the input electrical energy is mainly dissipated into heat and light energy instead of chemical energy. Note that the system is not designed to operate at high energy efficiencies, rather its purpose is to carry out fundamental studies of gas/plasma-phase chemical reactions. Other configurations at larger scale exist though achieving better yields [7], which may be adapted to our present context.

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6. References
