

PLASMA-CATALYTIC NON-OXIDATIVE METHANE COUPLING BY GLIDING DISCHARGES ENHANCED BY Pd AND Pt CATALYSTS

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Abstract. Plasma-catalytic methane conversion was studied under gliding discharge (GD) conditions using a mobile (spouted) bed of Pt/Al₂O₃ and Pd/Al₂O₃ catalysts. C₂ hydrocarbons and hydrogen were the main products with some amounts of non-volatile substances (mainly soot). In a homogeneous system and with alumina-ceramic particles, mainly acetylene was produced from the CH₄+H₂ mixture with none or a minor share of other C₂ hydrocarbons. The product composition changed in the presence of Pt/Al₂O₃ and Pd/Al₂O₃ catalysts, with enlarged contents of ethylene and ethane and a much lower share of acetylene.

1. INTRODUCTION

The growing utilization of natural gas (NG) for chemical syntheses encourages researchers to undertake studies on simpler and more economical processes for the conversion of simple hydrocarbons into useful products, unsaturated and aromatic hydrocarbons, as well as oxygenates, e.g. methanol, formaldehyde. Nowadays, the main drawback of high-temperature NG processing is the high energy demand. Therefore, novel methods of low temperature NG conversion (partial oxidation, reforming, oxidative and non-oxidative coupling) are searched in last years, including those using low-temperature non-equilibrium plasmas for chemical reaction initiation^[1,2]. Due to the active species generation in plasma with electrons of high energy (>1 eV), a number of chemical reactions may be effectively conducted at moderate bulk gas temperatures. The reactions of methane and other simple hydrocarbons were investigated under conditions of different kinds of discharges including spark, corona and dielectric barrier discharges often used in pulsed-plasma system^[3-14]. Furthermore microwave and RF discharges were used for methane conversion into acetylene^[15,16]. The performance of different systems using both thermal and non-thermal plasmas for hydrocarbon conversions to hydrogen has recently been discussed^[17,18]. An efficient method for methane conversion into synthesis gas in non-equilibrium plasma, has been developed using gliding discharges (GD)^[19-25]. On the other hand, it was found that unsaturated hydrocarbons (mainly acetylene) and some oxygenates, e.g. methanol, may also be obtained in this kind of discharge. The selectivity of methane conversion to acetylene was usually significant, however, the formation of carbon (soot) could not be avoided^[26].

In order to improve the efficiency and selectivity of those processes, the use of hybrid systems were investigated by combining the plasma activation of reactants with the action of a solid catalyst stimulating the generation of the desired product. A number of studies was performed with the use of the combined systems for converting simple hydrocarbons by oxidative or non-oxidative coupling to obtain olefins, acetylene, aromatics, oxygenates etc.^[27-39]. The efficiency of plasma-catalytic system based on gliding discharges was the object of our studies over the last years^[1,2]. Taking into account a number of hitherto performed experiments with plasma-catalytic systems, it is possible to conclude that their efficiency depends not only on the kind of discharge and the catalyst's activity but also on the reaction volume configuration. In many cases, low efficiency of these processes results from an excessive time elapsing between the active species generation in the plasma and their contact with the catalyst. Thus, an essential question had to be answered: is there any catalyst which can effectively be