Production of synthesis gas from biogas on composite materials prepared by combustion synthesis method

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Introduction

Self-propagating high-temperature synthesis (SHS) method is used worldwide [1,2] for the low-cost production of engineering and functional materials such as advanced ceramics, intermetallics, catalysts and magnetic materials. The method exploits self-sustaining solid-flame combustion reactions for the internal development of very high temperatures over very short periods. It therefore offers many advantages over traditional methods such as much lower energy costs, ease of manufacture and capability for producing materials with unique properties and characteristics. The basic principles of SHS can be summarized as follows: rapid auto wave combustion-like self-sustaining reactions yielding resultant products of desired composition and structure, complete or partial elimination of external energy supply by the utilization of the internal heat released in the chemical reactions, control of the process rate, temperature, degree of conversion, composition and structure of products by variation in the rates of heat release and transfer [2,3]. It was found that prepared SHS materials are new class of catalysts and carriers.

For over the last few years, conversion of carbon dioxide, in particular by its reaction with methane to form carbon monoxide and hydrogen (commonly known as dry reforming of methane), has gained a lot of attention. It is known that nanosized metallic and polyoxide catalysts are developed for catalytic partial oxidation (CPO) of CH_4 into synthesis-gas during last decade too [4]. The aim of work was investigation the reaction of CH_4 with CO_2 (dry reforming) and O_2 (partial oxidation) catalyzed by Co-Mg-O, which were prepared by SHS method and traditional incipient wetness impregnation to demonstrate the benefits of SHS method.

Materials and Methods

The solution combustion synthesis catalysts on the base of $Co(NO_3)_2$-$Mg(NO_3)_2$-H_3BO_3 with glycine / urine were prepared from initial nitrates, boric acid and glycine / urine. The resulting materials were characterized by X-ray diffraction (XRD) using $CuKa_1$ radiation and Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray Spectroscopy (EDX). Further analyses were carried out using TEM and the particle size distributions were estimated by XRD peak broadening analysis. Surface area of the samples was determined by BET method.

Results and Discussion

It was found, that reducer (urine or glycine) play significant role in formation of spinel compositions due to different volume and speed of gas formation during SCS and as consequence different temperature of combustion. SEM analysis determine the various phases of spinel formation, it helps to clarify the doping mechanism by cations in spinel lattice. It was found that $CoMg_3O_4$ spinel formation is due to magnesium cations substitution of cobalt cations. Mg^{2+} ions are replaced by ions of Co^{2+}, as a consequence $CoMg_3O_4$ lattice parameters increase, since the ionic radius of Mg^{2+} (0.66 Å) less than Co^{2+} (0.72 Å). CoB_2O_4 formation probably occurs during the replacement of Co^{2+} (ionic radius 0.72 Å) ions B^{3+} (ionic radius 0.23 Å), which reduces the spinel lattice parameters. It has been shown that in the case of addition of urea in the catalytic composition the concentration of cobalt cations in the spinel lattice is higher. In this case, the Co_3MgO_4 is formed, and in the case of glycine $CoMg_3O_4$ is formed basically. It was found that the initial mixture of catalyst 60% Co $(NO_3)_2$ + 40% Mg $(NO_3)_2$ + 22% glycine + 37% H_3BO_3, containing glycine has a higher activity than the catalyst containing urea.

Significance

When comparing the activity of catalysts on the base of $Co(NO_3)_2$-$Mg(NO_3)_2$-H_3BO_3 with glycine / urine prepared by SHS and impregnation method has been shown that the impregnation method concedes to SHS method by activity whereby a higher feed conversion and H_2 yield were obtained.

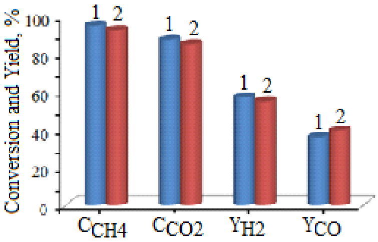

Figure 1. Effect of the method of preparing the catalyst on the conversion of methane and carbon dioxide as well yield of hydrogen and carbon monoxide. 1 - SHS method, 2 - impregnation method, GHSV = 860 h^{-1}.

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