METHANE DRY REFORMING WITH CO_2: A STUDY ON SURFACE CARBON SPECIES

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Introduction

Methane dry reforming with CO_2 has been studied for a long time. One of the major problems is the deactivation and the type of carbon formed on the surface. In the present work 8 wt% $NiMgAl_2O_4$ sample was prepared using the method described in ref. 1. The aim of the present work is to elucidate the structure of the various surface carbons leading to the deactivation of the catalyst. Second, we wish to study the role of gold interacting with nickel to prevent excess carbon deposit formation.

Materials and Methods

The catalyst was prepared by incipient wetness method using $Ni(NO_3)_3$ and $HAuCl_4$. The reaction was carried out with a mixture of 29% CO_2 and 71% CH_4 in a plug flow reactor with temperature programmed mode. The components were measured by QMS and/or GC and the conversion was calculated. The retained carbon was recovered by temperature programmed oxidation (TPO)- The catalyst samples were characterized by X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and transmission electron microscopy (TEM)

Results and Discussion

In Fig. 1 a typical curve is presented indicating the temperature programmed reaction. The reaction starts at 500°C and is completed at 700°C. It was established that in sequential reactions the catalyst was somewhat deactivated due to graphite like carbon deposit measured by XRD and TEM (Fig. 2 shows the TEM image). Discrepancy found by XPS (Fig. 3)can be explained by the different types of carbon, i.e. after the first reaction most of the carbon deposited can be recovered by TPO and it is carbide (XPS), whereas after several subsequent reactions the carbon on the surface is graphitized forming carbon nanotubes (TEM).In the gold promoted catalyst the amount of graphite (nanotubes) is significantly reduced.

Significance

The methane containing over 30% CO_2 can be converted to syngas to reduce the amount of CO_2

References

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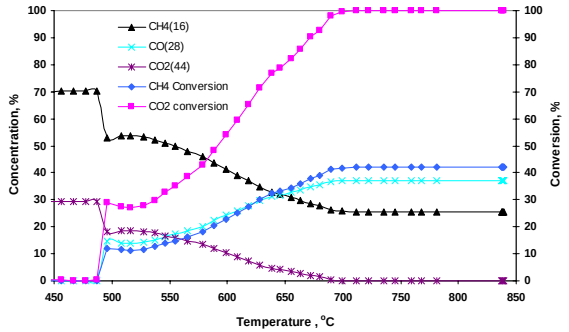

Fig. 1 Temperature programmed reaction of CH_4-CO_2 mixture

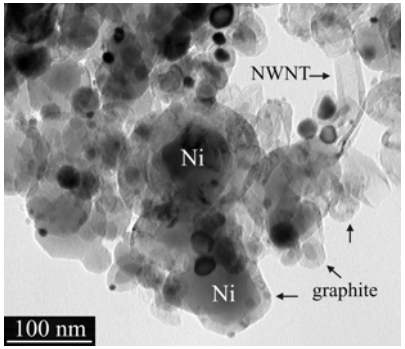

Fig 2 TEM image

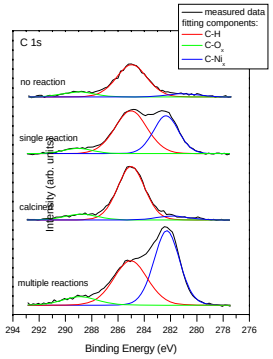

Fig. 3 XPS data