The characterization of Ni-Au supported alloy as catalytically active system in partial oxidation of methane.

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Introduction

The monometallic Ni/support catalysts are usually used in processes of partial oxidation of CH_4 (POM), but unfortunately they are susceptible on carbon deactivation during long term reaction. The gold doped nickel supported catalysts are highly active, selective and resistant for carbon deactivation during CH_4 reforming processes. Up to now the characterization of nickel-gold catalysts is not clearly describe as well as gold influence on catalytic performance of Ni catalysts. The aim of this work is the catalyst characterization and finding the correlation between physicochemical properties and catalytic performance of 5%Ni and 5%Ni-2%Au/support ($CrAl_3O_6$, Al_2O_3).

Materials and Methods

The alumina and chromium alumina binary supports were precipitated with ammonia using aqueous solution of aluminum and chromium nitrates $Al(NO_3)_3$ and $Cr(NO_3)_3$. Nickel and gold metal phases were impregnated using $Ni(NO_3)_2{\cdot}6H_2O$ and $HAuCl_4$ compounds. After drying the catalysts were calcined 4h in air at 400 °C. The physicochemical properties were studied by TPR, XRD, TOF-SIMS, SEM-EDS, XPS measurements and tested in partial oxidation of methane.

Results and Discussion

The strong metal oxide support interaction SMOSI: $CrAl_3O_6$ $NiCrO_3$ and $NiAl_2O_4$ were determined by XRD analysis for 5%Ni-2%Au/$CrAl_3O_6$. The generation of spinel like compounds is supposed as a origin of well dispersed Ni species which are responsible for high activity of catalysts in POM. The 5%Ni and 5%Ni-2%Au/$CrAl_3O_6$ catalysts achieved 100% of CH_4 conversion at 900°C, whereas 5%Ni and 5%Ni-2%Au/Al_2O_3 only 20 and 30% of CH_4 conversion, respectively (Table 1).

The gold doped Ni catalysts show the decrease of carbon deactivation in comparison to monometallic nickel catalyst from 24 to 7% and from 8 to 1% of carbon for catalysts supported on Al_2O_3 and $CrAl_3O_6$, respectively (Table 1).This finding is attributed to Ni sites dilution and blocking of highly reactive the exposed edge and kink sites on the surface of Ni particles in the Ni-Au alloy particles.

Table 1.Catalyst activity and resistance against carbon deactivation.

Catalysts	CH_4 conversion [%]		Carbon deposition after 24 h POM
	at 800°C	at 900°C	at 900°
5%Ni-2%Au/$CrAl_3O_6$	98	100	<1
5%Ni/$CrAl_3O_6$	99	100	8
5%Ni-2%Au/Al_2O_3	18	30	7
5%Ni/Al_2O_3	18	20	24

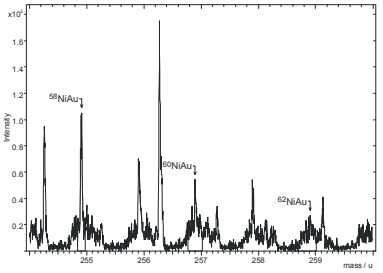

Fig. 1 TOF-SIMS (+) spectra – combined $NiAu^+$ ions distribution for 5%Ni-2%Au/$CrAl_3O_6$.

Significance

The Ni-Au alloying seems to be protectable phenomenon for carbon formation and resultant deactivation during partial oxidation of methane to synthesis gas.