

# Peculiarity of the gold-additive interaction in the Au-Me-zeolite (Me=Cu, Fe, Ni) catalyst for CO oxidation studied by $^{129}\text{Xe}$ nuclear magnetic resonance

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## Introduction

Recently Au supported on metal oxides was shown to be very active in many reactions important for chemical industry such as: CO oxidation, NO reduction, hydrocarbon oxidation, hydrogenation etc. at low temperatures [1]. That opens new horizons for catalytic processes of energy saving and dangerous gases removal. As well nanogold is perspective composite in medicine as an antibacterial mean [2].

Actually different methods of stabilization and the properties of dispersed gold are intensively studied to provide their better application. One of the most interesting supports for gold catalysts are zeolites due to their ability to stabilize special states of the metal inside the bulk frame. It is well established that zeolites exhibit different physicochemical properties when they have different pore structure or Si/Al ratio.

$^{129}\text{Xe}$  NMR of xenon adsorbed in zeolites is an informative method to study species inside zeolitic porous. Xenon is an inert gas with atom size 0.43 nm, smaller than the diameter of zeolite pores. Xenon enters into the zeolite pores and works as a probe molecule with high sensitivity towards the species located in pores. The success of xenon as probe molecule in porous media resides in its very high polarizability.

In order to provide a look on gold species formed in zeolites  $\text{Xe } ^{129}\text{NMR}$  study along with other physico-chemical methods was performed.

## Materials and Methods

Zeolites HY,  $\text{NH}_4$ -mordenite with  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of 14.6 and 20 respectively were supplied by TOSOH Corporation (Japan) and Zeolite International (USA) and used as starting materials.

Au/zeolite samples (2 wt.% of Au) were prepared by ion-exchange of the  $\text{Au}(\text{NH}_3)_4(\text{NO}_3)_3$  complex with the zeolites in solution. Zeolites NaY,  $\text{NH}_4$ -mordenite with  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of 5.6 and 20 respectively were supplied by Zeolite International (USA) and used as starting materials.

Au/zeolite samples were prepared by ion-exchange of the  $\text{Au}(\text{NH}_3)_4(\text{NO}_3)_3$  complex with the zeolites in solution. Previously to xenon loading, samples were dehydrated by gradual

heating up to 573 K in vacuum ( $10^{-4}$  Torr).  $^{129}\text{Xe}$  NMR spectra were recorded between 200 and 298 K in a Bruker DMX-500 instrument Two samples Au/Y were studied, Au-Y(2.5) and Au-Y(0.5) containing 2.5 and 0.5 wt.

## Results and Discussion

In  $^{129}\text{Xe}$  NMR spectra variation in the chemical shift of xenon adsorbed in pores of zeolite after gold loading compared with pure zeolite was observed. It is clear, that dispersed gold goes inside zeolite channels. Au-Fe-NaY and Au-Ni-NaY showed significantly higher chemical shift of Xe adsorbed then for AuY and Fe-NaY, Ni-NaY, Cu-NaY and Au-Cu-NaY. Plots of chemical shift as a function of the amount of xenon adsorbed revealed the introduction of the gold species inside the Y-zeolite cages. In particular the Au-Fe-NaY and Au-Ni-NaY catalysts showed the higher activity of this catalyst series for CO-oxidation. For Au-Cu-HMordenite the side pockets were blocked with the copper or gold particles that were followed by lowest activity in CO oxidation reaction of all the catalyst series.

Microscopic analysis showed, that particles Ni and Fe additives are distributed over all the support, while Cu was placed as isolated

Combination of the catalytic experiments in CO oxidation reaction with study of catalyst structure by TPR, UV-Visible, XPS revealed, that  $\text{Au}^{3+}$  species are not active, clusters showed the activity at low temperature ( $25^\circ\text{C}$ - $250^\circ\text{C}$ ) and nanoparticles at  $400^\circ\text{C}$  and higher.

## Significance

At present, our work focus on the influence of additives such as nickel and copper in the stabilization of the gold particles into two very different zeolitic cavities, the first one the supercages of the faujasite Y zeolite and the second one the main channels of the mordenite zeolite.

## References

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