Stable Au-Ag alloy nanoparticles encapsulated in single crystal multi-hollow zeolites: application to CO removal at low temperature studied by operando IR spectroscopy

R. Atwi, A. Tuel, M. Maffre, L. Burel, F.C. Meunier*
University of Lyon - CNRS, IRCELYON, UMR 5256, Villeurbanne, France

The preparation of well-defined and stable alloy nanoparticle remains a challenge. We have shown that it was possible to prepare well-defined alloy nanoparticles encapsulated in single crystal hollow zeolite nanoboxes that were stable at 500°C in the presence of steam. The overall metal loading in these single hollow nanoboxes had yet to be kept low if the preparation of small particles was sought. We have recently reported a novel method to prepare well-dispersed nickel nanoparticles at high loading (i.e. 6 wt.%) using multi-hollow single crystal zeolites.

The present contribution reports for the first time the preparation of Au and Au-Ag nanoparticles dispersed in multi-hollow silicalite-1 single crystals, derived from the method reported earlier. Highly dispersed Au (not shown) and Au-Ag (Fig. 1, left) nanoparticles with a diameter around 3 nm were obtained, even after calcination at 550°C. The CO oxidation activity and the species present at the surface of the catalyst were evaluated using a diffuse reflectance FT-IR spectroscopic (DRIFTS) cell described elsewhere. The Au and Au-Ag materials exhibited a maximum of activity (Fig. 1, Middle), as expected. Surprisingly, the Au-only sample was as active as the Au-Ag sample at lower temperatures, possibly due to a large fraction of low coordination sites that are able to adsorb and activate CO. This was confirmed by the operando DRIFTS data (Fig. 1, Right), which showed a significantly higher band at 2134 cm⁻¹, corresponding to CO adsorbed on step and corner sites. The proportion of these sites was lower on the Au-Ag sample, but the alloy exhibited partially oxidised Ag sites (bands at 2189 and 2172 cm⁻¹) that could too adsorb CO. The activity of an Ag-only sample was significantly lower over this temperature range (Fig. 1, Middle), stressing again the synergy existing between Au and Ag for CO oxidation.

This work shows that it is possible to prepare highly dispersed and stable Au and Au-Ag nanoparticles in multi-hollow zeolites that exhibit interesting activity for CO oxidation at room temperature.

Figure 1. (Left) TEM picture of the 2 wt.%Au+0.2wt.%Ag@silicalite-1 sample calcined at 550°C. (Middle) CO conversion as a function of temperature. Feed: 2.8% CO + 40% O₂ in He, total flow rate 50 mL min⁻¹, 10 mg of catalyst. (Right) Operando DRIFTS spectra at 100°C corresponding to the catalytic data reported the previous figure.

References

Corresponding author email: fcm@ircelyon.univ-lyon1.fr