

THESIS INFORMATION

Title: Investigation of deep oxidation of carbon monoxide and p-xylene on Pt + CuO catalysts using various supports.
Major: Chemical engineering
Code: 62.52.01.03
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Abstract:

CuO-based catalysts modified by Pt supported on γ -Al₂O₃, CeO₂, TiO₂ and γ -Al₂O₃ + CeO₂ for low-temperature oxidation have been prepared. Physico-chemical properties as well as the oxidation activity of catalysts were investigated. The performance of catalysts was assessed for deep oxidation of CO, p-xylene and their mixture in the absence and presence of water vapor. The obtained results emphasized the effects of the composition of catalysts and reactants on the characteristics and activity of Pt + CuO catalysts. Pt and CeO₂ led to weakening the interaction between CuO and carriers and enhancing the dispersion and reducibility of CuO, and CO adsorption, so the activities of Pt + CuO catalysts were increased. The supports determined the specific surface area and the hydrophobic character of catalyst. The optimal composition of all catalysts was proposed. The catalysts containing CeO₂ played the highest activity for CO oxidation since CO was converted completely to CO₂ at 125 °C and space velocity 75,000 h⁻¹. In mixture oxidation, the presence of CO in a mixture with p-xylene exhibited a beneficial effect on the p-xylene conversion, meanwhile the presence of p-xylene showed the inhibition on CO oxidation. The presence of steam in gas mixture had no effect on the conversion of CO, meanwhile it showed the inhibition effect on activity of various catalysts for the deep oxidation of p-xylene. However the deactivation of catalysts by water vapour was reversible.

The kinetics of the deep oxidation of CO, p-xylene and their mixture on three Pt + CuO catalysts were investigated in a gradientless flow-circulating system at low temperature range of 125 – 225 °C. The reactions on catalysts had the same kinetic equations in fractional rational form. These equations imply that the reactions proceed in medium surface coverage with the participation of CO and p-xylene adsorbed molecules and oxygen atoms. The characteristics of carriers and Pt additive only affected the properties, activity and adsorption capacity of catalysts and the activation energy of the reaction. In the oxidation of mixture, a complicated mutual effect associated with the formation of new intermediates takes place, leading to change the kinetics of the reactions.

Based on the results of kinetic investigation in steady state and data of transient responses in unsteady state of reaction, the mechanism of CO oxidation on Pt + CuO catalysts was proposed. Both the lattice and adsorbed atoms O took part in CO oxidation and both Langmuir–Hinshelwood and Mars Van Krevelen models were accepted.

The main contributions of the thesis:

- + Preparing highly effective catalysts based on the mixture of Pt and CuO and carried on hydrophobic supports which expressed the high activity in low-temperature oxidation and the strong inhibition of the presence of water vapor.
- + Emphasizing the interaction of CO and p-xylene being different specificity in their mixture oxidation on Pt + CuO catalysts and clarifying the disadvantage of Pt catalyst in the mixture oxidation – CO inhibition on hydrocarbons excluded on Pt + CuO catalysts.
- + Establishing the kinetic equations of deep oxidation of CO, p-xylene and their mixture on Pt + CuO catalysts and proposing the mechanism of CO oxidation in order to accomplish the theory basis of CO and p-xylene oxidation on catalysts based on the mixture of Pt and CuO using various supports.

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