Propylene epoxidation with H\textsubscript{2} and O\textsubscript{2} over Au supported on ZrO\textsubscript{2} with different crystal phase

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ZrO\textsubscript{2} with different crystal phases were used as carriers of Au/ZrO\textsubscript{2} catalysts for the direct vapor-phase epoxidation of propylene with H\textsubscript{2} and O\textsubscript{2}. Despite their catalytic performances were inferior to those of Au catalysts supported on Ti-containing, however, an interesting feature was that propylene epoxidation over Au/ZrO\textsubscript{2} appreciably depended on calcination temperature of support, that is, the phase of ZrO\textsubscript{2}. Au/t-ZrO\textsubscript{2} (t: tetragonal) benefits high propylene conversion, whereas only tiny amounts of PO was obtained. On the contrary, PO selectivity can reach 100\% for Au/m-ZrO\textsubscript{2} (m: monoclinic) at reaction temperature 100\°C.

The effect of pre-calcination temperature of ZrO\textsubscript{2} on the structural and surface properties of the supports and Au/ZrO\textsubscript{2} catalysts was investigated by XRD, N\textsubscript{2}-physisorption, Raman, HRTEM, and XPS characterizations. Correlating to the catalytic results, it is found that the catalytic performance of Au/ZrO\textsubscript{2} catalysts strongly depends on the concentration of oxygen vacancy and monoclinic structure in the lattice (see Figure 1). XPS results disclose that electrons can be transferred from the supported Au to ZrO\textsubscript{2} and the interaction between Au and ZrO\textsubscript{2} is strongest for Au/ZrO\textsubscript{2}-400, which is responsible for the high propylene conversion. Moreover, theoretical calculation showed that the monoclinic crystal lattice is assumed to be a crucial structural fact for PO selectivity.

Figure 1. TEM images of as-synthesized Au/ZrO\textsubscript{2} catalysts: (a) Au/ZrO\textsubscript{2}-400, (b) Au/ZrO\textsubscript{2}-650, (c) Au/ZrO\textsubscript{2}-800, and (d) Au/ZrO\textsubscript{2}-1000.

References

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