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Near Ambient Pressure Photoelectron Spectroscopy Studies of CO Oxidation on Spinel Co3O4 Surfaces: Electronic Structure and Mechanistic Aspects of Wet and Dry CO Oxidation

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Abstract : The CO oxidation is a primary reaction in heterogeneous catalysis due to its potential to overcome the air pollution caused by various reasons. Indeed, in the study of sustainable catalysis, the role played by water is very important. The present work is focused on studying the effect of moisture on the sustainability of Co3O4 NR catalyst for CO oxidation reaction at ambient temperature. The catalytic activity, electronic structure and the mechanistic aspects of spinel Co3O4 nanorod surfaces have been explored in dry and wet atmosphere by near-ambient pressure photoelectron spectroscopic techniques (NAP-PES) with conventional x-ray (Al kα) and ultraviolet sources (He-I).Comparative NAPPES studies have been employed to understand the elucidation of the catalytic reaction pathway and the evolution of various surface species. The presence of water with CO+O2 plummet the catalytic activity due to the change in electronic nature from predominantly oxidic (without water in the feed) to few intermediates covered Co3O4 surface. However, ≥ 375 K Co3O4 surface recovers and regain oxidation activity, at least partially, even in the presence of water. Above mentioned observations are fully supported by the changes observed in the work function of Co3O4 in the presence of wet (H2O+CO+O2) compared to dry (CO+O2) conditions. Various type of surface species, such as CO(ads), carbonate, formate, are found to be on the catalyst surface depending on the reaction conditions. Under dry condition, CO couples with labile O atoms to form CO2, however under wet conditions it also interacts with surface OH groups results in the formation carbonate and formate intermediate. The carbonate acts at reaction inhibitor at room temperature, however proves as active intermediate at temperature 375 K or above. On the other hand, formate has proved to be reaction spectator due to its high stability. The intrinsic role of these species to suppress the oxidation has been demonstrated through a possible reaction mechanism under different reaction conditions.

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