January, 1970] 291

SHORT COMMUNICATIONS

Oxidation of Propylene Adsorbed on Oxide Catalysts

Yutaka Kubokawa, Takehiko Ono, Takeshi Tomino and Norio Yano

Department of Applied Chemistry, University of Osaka Prefecture, Sakai, Osaka

(Received October 15, 1969)

Although the oxidation of olefins on oxide catalysts has been investigated by many workers, the nature of the adsorption complex during the oxidation reaction still remains to be unclarified.1) present work, the interaction of oxygen with propylene adsorbed on various oxides was investigated. On the catalyst2) which had already adsorbed propylene it was found that the amount of oxygen adsorbed increased markedly compared to that on the cleaned catalyst. In the case of Co₃O₄, the increase in the amount of oxygen adsorbed was proportional to the amount of propylene previously adsorbed as seen in Fig. 1. On ZnO the amount of oxygen adsorbed on propylene-covered surface was very small at 0°C. After the measurement at 0°C the temperature of the catalyst was raised in oxygen. It was found that an appreciable adsorption of oxygen occurred above 80°C. The amount of oxygen adsorbed at 80°C after contact for 3 hr was also proportional to the amount of propylene adsorbed.

After the oxygen adsorption experiment, the oxygen remaining in the gas phase was removed

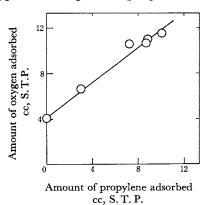


Fig. 1. Enhancement of oxygen adsorption on Co₃O₄ (8.55 g; 100 m²).

by Töpler pump and desorption experiments were carried out. It was found that the desorbtion of oxygen as well as of propylene decreased markedly compared to the desorption in the case where only oxygen or propylene had been adsorbed. This suggests that enhancement of the oxygen adsorption results from the interaction of oxygen with propylene adsorbed with the formation of an adsorption complex.

After formation of the propylene-oxygen complex, the temperature of the catalyst was raised in steps in oxygen. It was found that the complex was oxidized to carbon dioxide and water. The result obtained with ZnO is shown in Fig. 2. It is seen that the amount of oxygen reacted at each stage passes through two maxima, one corresponding to the uptake at 80°C with the formation of complex, and the other corresponding to the reaction at 350°C with the formation of carbon dioxide. This suggests that the oxidation of propylene adsorbed on ZnO proceeds in the two consecutive steps, i. e., the complex formation with a low activation energy, and the oxidation of the complex to carbon dioxide and water with a high activation energy. It was found that such a two step mechanism could be applied in the catalytic oxidation of propylene on oxides as well as the oxidation of propylene adsorbed. Details will be published in a forthcoming paper.

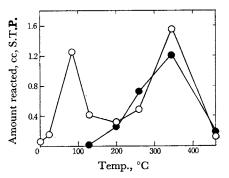


Fig. 2. Interaction of oxygen with propylene adsorbed on ZnO (5 g; 15.3 m²), —, O₂ reacted; —, CO₂ formed.

¹⁾ H. H. Voge and C. R. Adams, "Advances in Catalysis," Vol. 17, Academic Press, N.Y. (1967), p. 151.

²⁾ Prior to the experiment the catalyst was evacuated at 450°C. The time of evacuation was 24 hr on Co₃O₄ and 10 hr on ZnO.