Enhancement of Hydrogen Adsorption on an Acetone-covered Nickel Catalyst

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In order to obtain information on the nature of the adsorption complex during the hydrogenation of acetone on metal catalysts, the interaction of hydrogen with acetone adsorbed on a silica-supported nickel catalyst (15 wt % Ni) was investigated. On the catalyst which had already adsorbed acetone it was found that the amount of hydrogen adsorbed was increased markedly compared to the adsorption on the cleaned catalyst. Such an enhanced adsorption of hydrogen was an activated process, its rate being increased with increasing temperature. The activation energy of adsorption was determined as 10-13 kcal/mol in the temperature range 0-20°C. Adsorption equilibrium was almost attained after several hours at 20°C. The increase in the amount of hydrogen adsorbed at equilibrium $q_{\rm H_2}$ was proportional to the amount of acetone previously adsorbed q_A

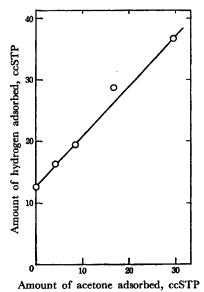


Fig. 1. Adsorption of hydrogen on the acetone-covered surface.

The amount of catalyst and the equilibrium pressure were 6.67 g and 20 mmHg, respectively.

as seen in Fig. 1. The ratio of $q_{\rm H2}$ to $q_{\rm A}$ was 0.83 at 20°C.¹⁾ After the adsorption of hydrogen, the hydrogen remaining in the gas phase was removed by Töpler pump at 0°C. The temperature of the catalyst was raised up in stages, the desorbed gas being collected. It was found that by evacuation at 0°C 25 per cent of the hydrogen adsorbed was removed while after evacuation at 50°C only 20 per cent of the hydrogen adsorbed remained on the surface. On the other hand, the amounts of acetone and isopropyl alcohol appearing in the gas phase during such experiments were very small even at 50°C.²⁾ Such behavior suggests that hydrogen is held relatively weakly on the acetone-covered nickel surface.

The infrared spectra of the adsorbed layer were investigated by means of an apparatus similar to that used by Peri.³⁾ When hydrogen was admitted to the acetone-covered surface, no new bands due to hydrogen adsorption were detected, and in addition, little or no change in the band at 1706 cm⁻¹ due to the C-O stretching vibration, was observed. The spectra obtained were completely different from those with the surface which had adsorbed isopropyl alcohol.

The results described above suggest the formation of an acetone-hydrogen complex on the nickel catalyst when hydrogen is adsorbed at room temperature in the presence of acetone. In the temperature range above 80°C at which formation of isopropyl alcohol takes place considerably, the situation may be somewhat different. It seems necessary, however, to take into consideration the formation of such an acetone-hydrogen complex when we work out the mechanism of the hydrogenation of acetone.

¹⁾ Such an increase in the hydrogen adsorption caused by the preadsorbed acetone was also found for a non-supported nickel catalyst.

²⁾ The desorbed products were analyzed gas chromatographically by using a 2 m polyethylene glycol column at 65°C.

³⁾ J. B. Peri, Discussions Faraday Soc., 41, 121 (1966).