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STUDY OF THE DEPENDENCE OF THE CATALYTIC ACTIVITY AND THE ENERGY OF THE METAL-HYDROGEN BOND

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The catalytic activity of various metals and alloys in reactions involving hydrogen can be associated with various volumetric and surface properties of catalysts (1,3). According to Balandin's multiplet theory, an important role in catalysis is played by the presence of a geometric (structural) correspondence between the reacting group of atoms in the substrate molecule and the location of the catalyst atoms on its surface. An equally important role is played by the principle of energy correspondence, which consists in the presence of a certain range of catalyst—substrate binding energies, at which the catalyst exhibits maximum activity. To solve the issues related to the thermodynamic determination of the catalyst of the highest activity, attempts have been made to establish a relationship between the specific activity of the catalysts and the heats of formation of the proposed intermediate compounds or the heats of certain intermediate stages through which the process may proceed. Catalytic reactions involving hydrogen are carried out through the stages of formation of a bond between the surface of the catalyst and hydrogen, it is quite justified, in our opinion, to search for a correlation of the catalytic activity of metals with such an energy characteristic of their surface as the strength of the metal—hydrogen adsorption bond. Of course, from the point of view of the multiplet theory, it would be more justified to choose as a correlation parameter (Fig.1) the value of the adsorption potential of the catalyst q, that is, the sum of the energies of a group of reactions and catalysts, for which one or another of the selected parameters plays a dominant role in determining the catalytic properties.

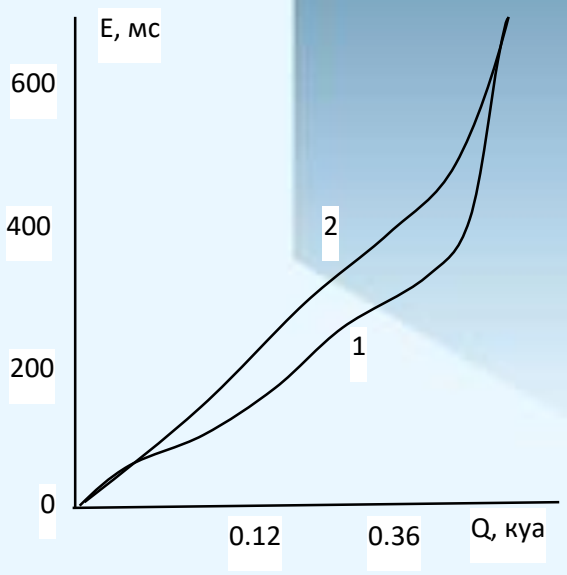

Fig.1. Charging curves of the Pt/Pt electrode in sulfate solutions of OD N IPSO; +KOH; 30°; current density 5-10~5 a/cm2 at pH 1.2 (curve 1), 13.0 (curve 2).

There is some correlation between the atomic radii of the metal (Pd, Rh, and their alloys) and the mechanism of the exchange reaction between hydrogen and fatty acids. Using electrochemical methods for studying catalysts, it was found that the activity of catalysts during the hydrogenation of vegetable oils depends on the binding energy of the adsorbed hydrogen and the unsaturated compound with the contact surface. With an increase in the concentration of alkali in the reaction medium, the binding energy of hydrogen to the surface of the catalyst increases, and the rate of hydrogen renewal and its activation on the surface of the catalyst decreases. It is confirmed that the medium affects only the amount of adsorbed hydrogen and does not affect the amount of dissolved hydrogen. Using modern research methods, using the example of hydrogenation of cottonseed oil, we tried to find out the effect of pH on the binding energy of the reaction components with the surface of the catalyst. We have removed the charging curves of rhodium catalysts. As can be seen from Figure 1, with increasing pH, the extent of the hydrogen region and the potential for hydrogen desorption increase, which indicates an increase in the degree of coating of the rhodium surface with hydrogen and an increase in the strength of the Rh–H bond. The hydrogen region on the charge curve for rhodium is an isotherm of hydrogen adsorption. Isotherms were removed at several temperatures to determine the heat of

adsorption. However, the determination of this value is also possible by one isotherm. Such a calculation based on charging curves is quite acceptable for determining the topography of hydrogen adsorption on metals of the rhodium group. The determination of the heat of hydrogen adsorption makes it possible to calculate the binding energy of the hydrogen catalyst. Since the heat of hydrogen adsorption decreases with increasing degree of surface filling, the Rh-H binding energies calculated by us also change with surface filling. For example, with an increase in the degree of filling from 0 to 1 at pH 1.0, the Rh—H binding energy varies from 65.8 to 57.8 k/cal • mol-1. Measuring the potential of the catalyst during hydrogenation makes it possible to judge the ratio of concentrations of reacting substances on the surface of the catalyst and approximate the degree of filling it with hydrogen, and therefore the binding energy of the hydrogen involved in the reaction. Since when fatty acids are applied to the degassed surface of rhodium, their deep decomposition occurs, measuring the adsorption of the organic component, and therefore determining the heat of adsorption in this way is not reliable enough. Using the kinetic method of calculating the binding energies of reacting molecules with the surface of the catalyst, it was possible to find the value of the binding energy of rhodium with carbon in double bonds. The values of the Rh-H binding energies used for this calculation were calculated by us in each case for the degree of filling of the rhodium surface corresponding to the potential of the working catalyst.

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