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INVESTIGATION OF PALLADIUM CATALYST ON ALUMINUM OXIDE IN THE PROCESS OF ACETYLENE HYDROGENATION IN ETHANE-ETHYLENE FRACTION

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ABSTRACT

The order of acetylene hydrogenation on a palladium catalyst for acetylene is one, while the order for hydrogen is zero; the order of ethylene hydrogenation for hydrogen is one, while the order for ethylene is zero. At temperatures of 50-70 ° C, the reaction proceeds in the kinetic region. The service life of the catalyst, its selectivity, the course of side processes, and its mechanical strength all influence the features of the corundum carrier.

Keywords: Industrial catalyst, acetylene hydrogenation, ethane-ethylene fraction, hydrogen conversion, acetylene conversion, pilot plant.

INTRODUCTION

Pyrolysis is still the most common way to make lower olefins like ethylene and propylene. Pyrolysis facilities currently have capacity of 113 million tons per year for ethylene (100 percent of global production) and 38.6 million tons per year for propylene (just over 65 percent of global production). The following is the current global structure of pyrolysis raw materials: 27.6% (wt.) ethane, 14.0 percent (wt.) liquefied gases, 53.1 percent (wt.) straight-run gasoline (naphtha), 5.3 percent (wt.) hydrotreated kerosene and oil fractions (wt.) The separation of the hydrocarbon flow into fractions: methane-hydrogen, ethane - ethylene, propane-propylene, etc. occurs during cooling at temperatures from 110 to - 130 °With and pressure from 0.5 to 5.0 MPa.

Catalytic hydrogenation is used to separate the ethane-ethylene fraction from the acetylene contaminant. The reaction of catalytic hydrogenation of alkynes is used to purify the ethane-ethylene fraction (EEF) of pyrogas from the mixing of acetylene hydrocarbons. Metal or oxide catalysts are used to catalyze the hydrogenation reaction. A contact containing 0.04 wt was utilized as a catalyst to evaluate the kinetics of the acetylene hydrogenation process. % palladium on - aluminum oxide, 9 m2/g specific surface area. Tests of the finished catalyst were

carried out at temperatures of 40 - 60 0C, a pressure of 2.3-2.5 MPa and a molar ratio of hydrogen and acetylene 1.6-1.9. The selectivity of acetylene hydrogenation on this catalyst was 90%.

EXPERIMENTAL PART

Testing the primary features of a new type of catalyst under conditions as close to real-world industrial conditions as possible is an important step in its development and implementation. Such an approach is required to anticipate the stability of its operational features, such as activity and selectivity, as well as service life, which is especially important when using a costly metal like palladium.

The mission was completed by measuring the prototypes' activity and selectivity values and comparing them to industry catalysts in four different reactors: laboratory, pilot, experimental, and industrial. On a real industrial gas mixture - ethane-ethylene fraction of the composition: ethylene, approximately, 55 percent, ethane, 43 percent, acetylene, 0.3 percent, hydrogen, 0.9 percent, the rest - methane, propylene, propane, carbon monoxide, etc. - comparative tests of prototypes and industrial catalyst G-58I were carried out on laboratory, pilot, and pilot plants. The acetylene content in the contact gas should not exceed 10 ppm.We conducted comparative tests of 32 prototypes and the G-58I industrial catalyst: the minimum temperatures of the acetylene hydrogenation process at which it is completely eliminated (up to 1 ppm), hydrogen conversion and selectivity corresponding to these temperatures, the formation of green oil as a result of side reactions of ethylene and acetylene oligomerization at acid centers the inner surface of the carrier.

"Ethanolamine" prototypes No. 12, 13, 15 were treated with an aqueous solution of sodium sulfide.

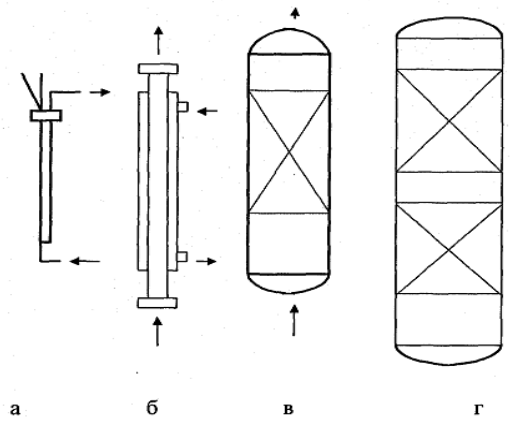

Fig.1. Large-scale test levels.

A-laboratory reactor, b-pilot reactor, experimental reactor,d-industrial reactor.

Table 1.Results of comparative tests of prototypes and industrial catalyst G-58 I in the process of acetylene hydrogenation in EEF [C2H2]starting=0.3 vol. %, [H2]starting = 0.9 vol. %

№№ for examples	[Pd], %	Min.full cleaning temperature EEF, 0C	$[C_2H_2]$ conc, no more than ppm	$[H_2]$ conc, %	Note
G-58I	1	65	1	0.37	green oil
1	0.1	68	1	0.20	green oil
2	0.1	70	1	0.18	green oil
3	0.05	75	1	0.31	green oil
4	0.2	65	1	0.10	green oil
5	0.05	70	1	0.31	green oil
6	0.05	70	1	0.37	
7	0.05	72	1	0.39	
8	0.2	65	1	0.44	
9	0.1	68	1	0.44	
10	0.3	65	1	0.40	
11	0.3	65	1	0.38	
12	0.1	70	1	0.46	
13	0.1	70	1	0.41	

The optimal palladium content in the catalyst according to the results of comparative tests was 0.2%, above this value, the catalyst is characterized by high activity to the detriment of selectivity, below its catalyst is characterized by instability of operation during prolonged tests.

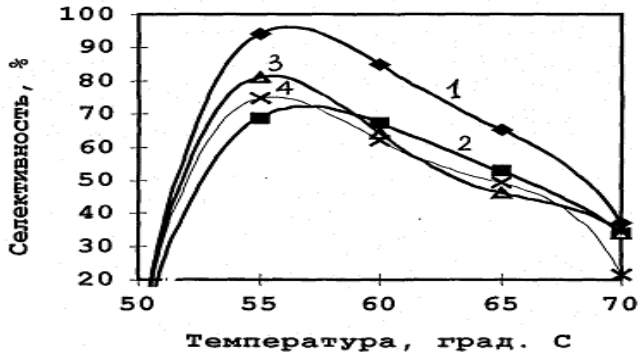

Fig. 2. Dependence of selectivity on temperature. 1 - "ethanolamine" sample No. 8 on carrier A.

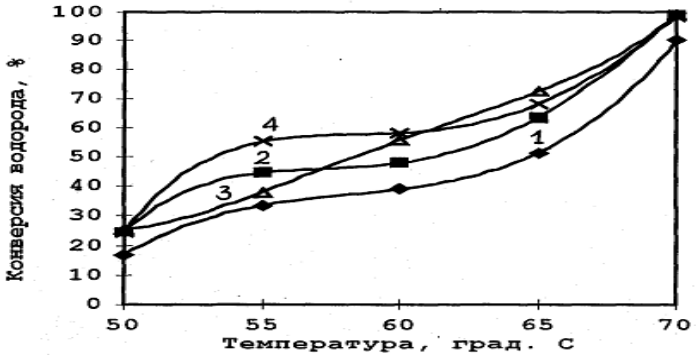

Fig.3. Dependence of hydrogen conversion on temperature.

The results obtained on the experimental samples confirm the presence of two types of active centers on the inner surface of the deposited palladium catalyst: one for acetylene hydrogenation, which is present on the fresh catalyst, and the other for ethylene hydrogenation, whose proportion on the surface increases during operation, i.e. the hydrogenation process selectivity decreases. According to the results of testing prototypes on various brands of corundum carriers containing 0.2 percent palladium and prepared according to various recipes, one was chosen - No. 8, which demonstrated high selectivity, activity, and stability of operation during the hydrogenation of acetylene in the ethane-ethylene fraction. On experimental samples with a palladium content of 0.1 - 0.5%, the hydrogenation rate of acetylene is higher than on the industrial catalyst G-58I.

- Additives of sodium sulfide and sodium formate contribute to a decrease in the rate of acetylene hydrogenation reaction, practically without affecting the rate of ethylene hydrogenation, which explains the decrease in the selectivity of "blackened" prototypes;
- The activity of the experimental samples in the hydrogenation of acetylene prepared by the ethanolamine method is higher than for other samples and the industrial catalyst G-58I;

RESULTS AND DISCUSSION

On a real industrial gas mixture - ethane-ethylene fraction of the composition: ethylene, approximately, 55 percent, ethane, 43 percent, acetylene, 0.3 percent, hydrogen, 0.9 percent, the rest - methane, propylene, propane, carbon monoxide, etc. - comparative tests of prototypes and industrial catalyst G-58I on laboratory, pilot, and pilot plants were carried out. The amount of acetylene in the contact gas should not exceed 10 parts per million. Then, in a laboratory setup, we performed kinetic experiments on the hydrogenation of acetylene and ethylene. For experimental samples and the commercial catalyst G-58I, the temperature dependences of the rate constants of the acetylene and ethylene hydrogenation processes are reported. From the results of comparative tests on a pilot installation and kinetic studies on a laboratory installation, it can be concluded that:

-The hydrogenation reaction of acetylene is of the first order for acetylene, well-left for hydrogen; the hydrogenation reaction of ethylene is of the first order for hydrogen and zero for ethylene;

-The rate of hydrogenation reactions of acetylene and ethylene depends on the palladium content in the catalyst; for experimental samples, it was found that the optimal content is 0.2 - 0.3 wt. %, over 0.5 wt. % it has little effect on the reaction rate;

-Additives of sodium sulfide and sodium formate contribute to a decrease in the rate of acetylene hydrogenation reaction, practically without affecting the rate of ethylene hydrogenation, which explains the decrease in the selectivity of "blackened" prototypes;

 The activity of the experimental samples in the hydrogenation of acetylene prepared by the ethanolamine method is higher than for other samples and the industrial catalyst G-58I;

 The best of the grades of the corundum carrier used for the preparation of prototypes turned out to be a fresh ring with a specific surface area of up to 3.0 m2/g and a bulk density of 790 kg/m3;

 The formation of green oil was not observed on the experimental samples prepared by the ethanolamine method.

CONCLUSION

The order of acetylene hydrogenation on a palladium catalyst is one for acetylene and zero for hydrogen; the order of ethylene hydrogenation is one for hydrogen and zero for ethylene. At temperatures of 50-70 ° C, the reaction takes occur in the kinetic zone. The service life of the catalyst, its selectivity, the course of side processes (oligomerization of acetylene and ethylene with the generation of green oil), and its mechanical strength influence the features of the corundum carrier. The selectivity, total catalytic activity, and catalyst lifespan are all affected by the palladium amount in the tregern hydrogenation catalyst; the ideal palladium content is 0.2-0.3 percent. The addition of alkaline and alkaline earth elements to the catalyst has no discernible effect on the catalyst's characteristics. The addition of sulfur compounds to the catalyst's composition reduces its activity during acetylene hydrogenation while having no effect on its activity during ethylene hydrogenation.

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