Investigation of Natural Gas Adsorption For the Recovery of Zeolite Waste

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ABSTRACT

The purpose of this work is to study the utilization of zeolite waste for use in the adsorption treatment of natural gases and their influence on adsorption-desorption characteristics in natural gas desulfurization processes for the purpose of their secondary use in desulfurization processes. A method has been developed to restore the original properties of zeolites based on the oxidation of air pollution products with oxygen at moderate temperatures. The quality of the recovered zeolites at the pilot plant was controlled by laboratory analysis of the degree of recovery and pilot tests of the adsorption – desorption characteristics of the recovered zeolite samples at the GPU "Shurtangaz".

Keywords: adsorption, zeolite waste, desulfurization, natural gas, desorption characteristics

Chemical processing of natural gases makes it possible to obtain a wide range of valuable products: motor fuels, polymer materials, etc. The various aggressive impurities included in the composition of natural and associated petroleum gases - acid gases (hydrogen sulfide and CO_2), water vapor, etc. lead to negative consequences for the processing of gases. The results of comprehensive studies of the mechanism of formation of contaminants in porous structural zeolites in the adsorption – desorption cycles of gas processing have established that only their transport pores are contaminated.

Purification of hydrocarbon gases from acidic components and inert gases, as well as water vapor, which complicate the processing processes, is carried out using: adsorption; absorption; catalytic methods; membrane technology. The most optimal way to dry and purify natural gas from H_2S, CO_2 and NO_2 is adsorption purification.

Gas purification with synthetic zeolites is widely used and has a number of advantages: pronounced selectivity of adsorption of polar molecules; high adsorption capacity at room temperatures and low partial pressures of the sorbed component; proximity of the diameters of the entrance windows in the zeolite cavity to the size of the molecules, which allows selective adsorption, low specific consumption of adsorbent, high environmental performance [1].

The conducted studies made it possible to develop a methodology for determining the structure of transport pores, which made it possible to identify changes in the dynamic characteristics of zeolites from the narrowing of the capillary size as a result of contamination.

Based on these considerations, a method was developed to restore the original properties of zeolites, based on the oxidation of air pollution products with oxygen at moderate temperatures.

Based on the above and critically analyzing possible options for the recovery of zeolites, a pilot plant with a capacity of 1.2 tons /day was developed and built. The principal advantages of this installation is that it operates in a continuous mode, i.e. the zeolite layer is in motion from top to bottom, and hot air is supplied to the recovery column.

The proposed recovery process is a stationary process and, thanks to simple thermodynamic calculations, the mode is easily regulated and the degree of recovery of zeolite almost reaches 100% due to the movement of the zeolite layer. Figure 1 shows the developed plant for the recovery of zeolite waste.

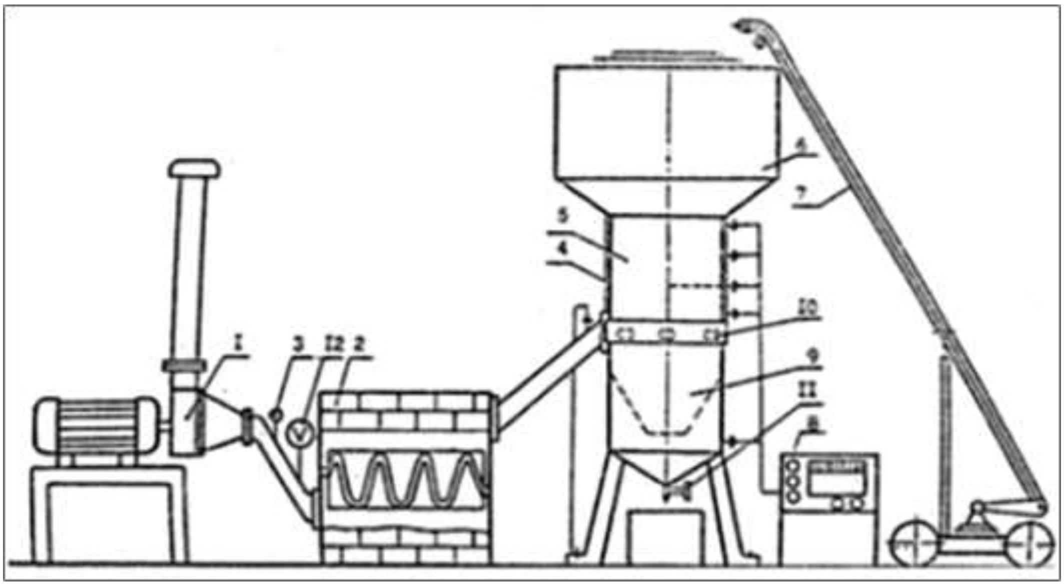

Fig.1 Zeolite waste recovery plant

The installation (Fig.1.) consists of a recovery column (4) with a diameter of 0.5 m and a height of 2 m, which consists of two parts; the upper (5) serves to restore the zeolite, where hot air is supplied, the lower (9) serves to cool the restored zeolite.

Above the column there is a hopper (6) with a diameter of 1.0 m and a height of 1.0 m. where zeolite waste is continuously fed by a conveyor (7). The air is supplied by a high-pressure fan (1) to the electric furnace (2), where it is heated to a temperature of 4500C and evenly enters the middle part of the column evenly through the distribution device (10). The air supply speed is regulated by a valve (3), and the speed of movement of the zeolite is regulated by a valve (11) located at the bottom of the recovery column. In addition, the unit is equipped with thermocouples (T) at six points with a recording potentiometer (8).

The main design parameters are the air supply speed and the movement of the zeolite through the column. Calculation of the process parameters is performed:n, given that the mode is stationary, the desired parameters relate to a unit of time. Based on experimental data obtained from previous studies on the operation of a pilot recovery plant and, being set by the speed of movement of the zeolite layer in the recovery column, i.e. by the capacity of the plant for manufactured products, the amount of heat flow and air flow were determined, which are equal to: heat flow Q = 14.85 kW, hourly consumption Vvod.=108

m^3/h. Based on these calculated data, a high-pressure fan with a capacity of at least 108 m^3/h and an electric furnace with a capacity of 14.85kW were selected for installation. The quality of the recovered zeolites at the pilot plant was controlled by laboratory analysis of the degree of recovery and pilot tests of the adsorption – desorption characteristics of the recovered zeolite samples at the GPU "Shurtangaz".

It is known that the main characteristic of adsorbents is their absorption capacity, i.e. the degree of gas purification during adsorption. The absorption capacity of the spent zeolites was determined by removing the adsorption isotherms on a device, the principle of operation of which is based on measuring the change in gas pressure during adsorption at a constant volume.

Determination of the degree of reduction was reduced to comparative estimates of H_2S adsorption isotherms on spent, recovered and fresh imported zeolite on a laboratory device (Fig.2).

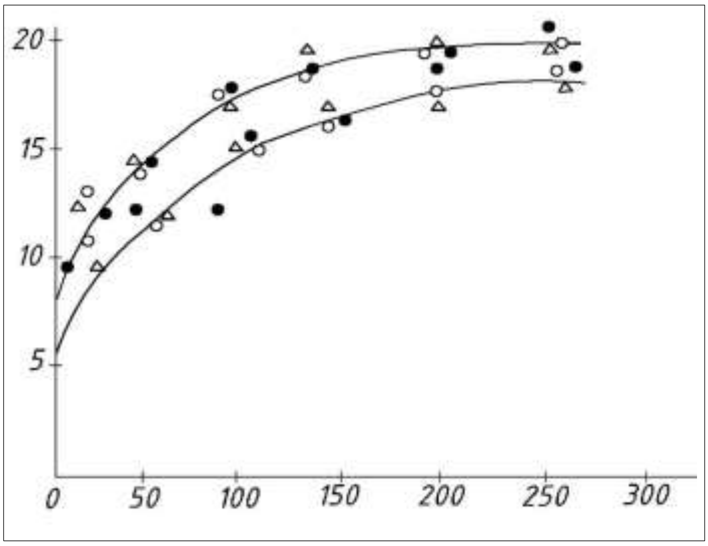

Fig.2. Isotherms of H2 adsorption on zeolites

– untreated zeoli○, – on reconstituted ze●lite,
– on fresh tsiolite △

Figure 2 shows the isotherms of H_2S adsorption on spent and fresh zeolites, from which it can be seen that the service life of zeolites at desulfurization plants almost does not affect the adsorption isotherms. This indicates that during multiple adsorption-desorption cycles of gas purification, only the transport pores of zeolites are polluted, and the adsorption cavities are practically not exposed to contamination.

To study the dynamic characteristics of the recovered zeolites, tests were carried out in the GPU "Shurtangaz" on a pilot installation. The following table 1 shows the test results.

Table 1.

Type of adsorbent	Dynamic capacity after recovery	% recovery
Used zeolite "Bitterfeld"	0,78	
Restored zeolite "Bitterfeld"	1,38	98,5
Fresh imported zeolite "Bitterfeld"	1,4	
Spent CaA zeolite (5A)	0,58	
Reconstituted CaA zeolite	1,18	98,3
Fresh imported CaA zeolite	1,2	

During long-term operation of zeolites in the adsorption–desorption cycles of desulfurization plants, they are contaminated with oxidation-reduction reaction products contained in crude gas (hydrocarbons, hydrogen sulfide, carbon dioxide and water vapor), which leads to a sharp narrowing of transport pores, resulting in a decrease in its dynamic activity.

As a result of studying the kinetics of hydrogen sulfide adsorption on spent and recovered zeolites and using the mathematical apparatus of the theory of mass transfer, a method for determining the structure of transport pores was developed and a change in the dynamic characteristics of zeolites from the narrowing of the capillary size as a result of contamination was revealed.

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