

Kinetics of the process of obtaining calcium cyanamide from lime, carbon dioxide and ammonia

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Annotation

Technical solutions have been developed to create a technology for obtaining calcium cyanamide from lime, carbonate anhydride and industrial ammonia. A technological scheme for obtaining nitrogen fertilizer and an effective defoliant, calcium cyanamide, has been developed and optimal technological parameters of the process have been determined. An experimental batch of calcium cyanamide has been produced at an experimental installation. The experiments investigated the effect of changes in the initial composition of one of the gas components of a chemical reaction with the relative constancy of the other component. Experimental studies were carried out by the excess method, in which an excess of ammonia was used in one series of experiments, and an excess of carbon dioxide was used in another

Keywords. Carbon dioxide, ammonia, expansion gas, calcium cyanamide, waste gases.

In kinetic studies, first of all, the order of the chemical reaction between calcium oxide, ammonia and carbon dioxide was determined.

The experiments investigated the effect of changes in the initial composition of one of the gas components of a chemical reaction with the relative constancy of the other component. Experimental studies were carried out by the excess method, in which an excess of ammonia was used in one series of experiments, and an excess of carbon dioxide was used in another. The excess was high enough that the concentration of the excess component from experience to experience practically remained constant. The concentration of the other component, by which the reaction order was determined, changed very significantly from experience to experience – several times.

According to the results of experiments conducted at different initial concentrations of the gas component and the duration of obtaining calcium cyanamide, a number of curves in coordinates from the percentage of nitrogen in the product were obtained - C_{N_2} and time $-T$.

Further, to determine the reaction order, the initial velocity method was applied, according to which the $C_{N_2}-\tau$ curves were graphically differentiated to determine the process speed (DC_{N_2}/d) at any time from the start of the reaction.

In the process speed – time coordinates, curves were plotted in accordance with the speed values (dC_{N_2}/dT). The constructed curves were extrapolated and then the speed of the process was determined in the initial period ($T=0$).

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After that, a curve was plotted in the coordinates of the logarithm of the initial velocity – the logarithm of the initial concentration of the tested component. The constructed curve had the character of a straight line, the tangent of the angle of inclination of which to the abscissa axis showed the reaction order for the studied component, i.e. the particular reaction order for carbon dioxide or ammonia.

During the experiments, the duration of the synthesis of calcium cyanamide varied from 25 to 105 minutes in increments of 15 minutes, the ratio NH_3 : CO_2 ranged from 12:1 to 1:12. The charge was prepared on the basis of natural limestone of the Jamansai deposit.

The temperature of 800°C was kept constant, which turned out to be optimal in previous experiments, the volume velocity of the initial gas mixture was 6000^{-1} . According to classical concepts, the reaction order can be 0, 1, 2, and 3, as well as fractional. In this regard, the experimental data obtained were checked using known equations and diagrams that showed their inconsistency. Therefore, from a theoretical point of view, the reaction is not three-molecular, apparently, it has an order associated with a complex reaction mechanism for the formation of calcium cyanamide.

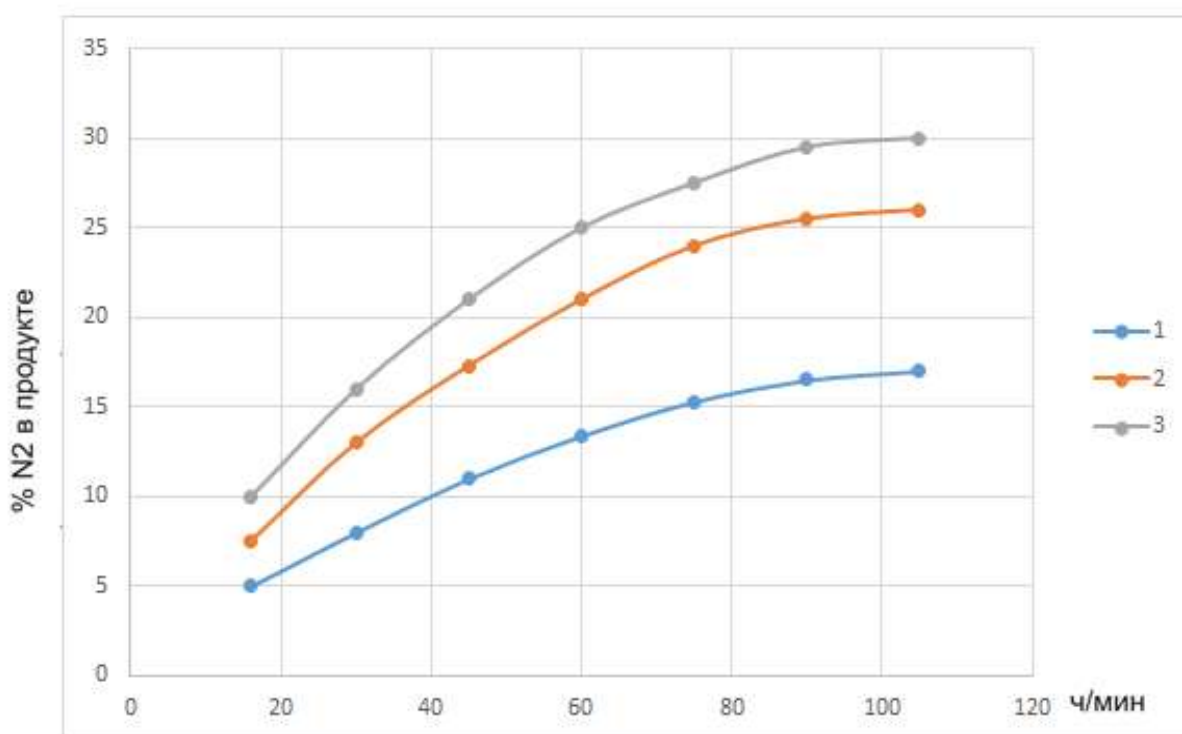
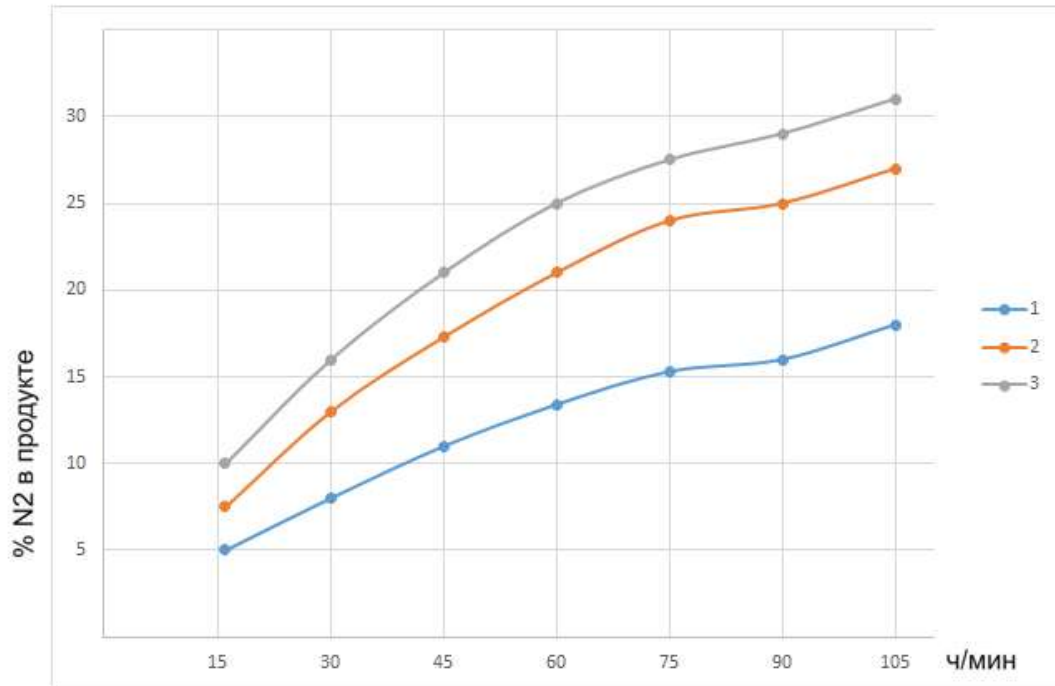


Fig. 1 Dependence of the nitrogen content in the product on the duration of the process and the concentration of ammonia in the gas mixture

The first series of experiments to determine the order of the carbon dioxide reaction was carried out at the ratio CO_2 : NH_3 = 3:1, 5:1, 1:12.

The second series of experiments to determine the order of the ammonia reaction was carried out at the ratios of carbon dioxide to ammonia 1:3, 12:1, 1:9.

According to the results of experiments (Figs.1 and 2), it can be concluded that with the same duration of the process, ammonia accelerates the formation of calcium cyanamide to a greater extent than carbon dioxide.



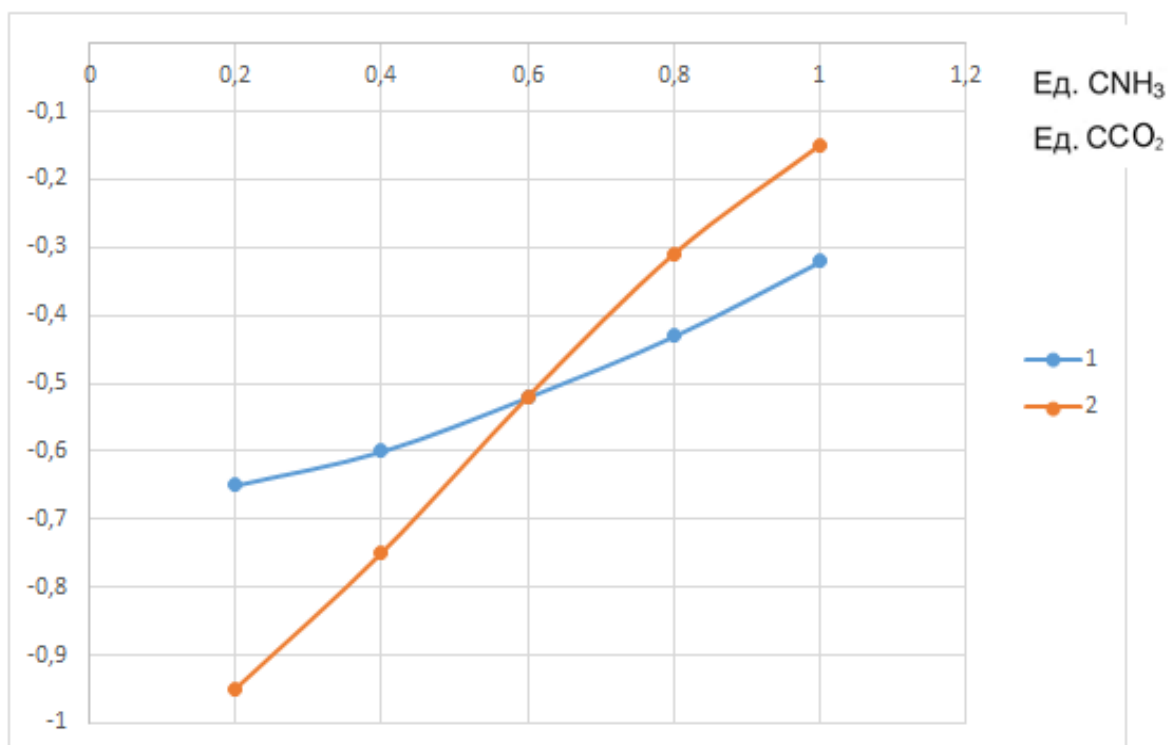
1-4%, 2-7%, 3-10%

Fig. 2 Dependence of the nitrogen content in the product on the duration of the process and the concentration of carbon dioxide in the gas mixture

Definition of particular orders (Fig.3) shows that the order of the formation reaction for carbon dioxide is 0.416, and for ammonia – 0.71205.

Since the indices of the reaction orders are fractional, this indicates that the process of chemical interaction of calcium oxide with ammonia and carbon dioxide is complex and its mechanism cannot be expressed by a simple stoichiometric equation.

Based on certain reaction orders, it can be assumed that the heterogeneous process of formation of calcium cyanamide from lime, ammonia and carbon dioxide is a complex process involving a number of successive stages. In this regard, the overall speed will be determined by the speed of the slowest stage.



1- Π_{CO_2} =0,416, 2- Π_{NH_3} =0,425

Fig. 3 Dependence of the logarithms of the initial rates of synthesis of calcium cyanamide on the logarithm of the concentration of carbon dioxide and ammonia in the initial gas mixture

We hypothesized that the limiting stage of the process under study is the chemical interaction on the surface of solid lime particles. To verify this, a number of experiments were conducted under optimal conditions for the synthesis of calcium cyanamide from lime, ammonia and carbon dioxide. With the constancy of all other conditions of the experiment, the changes were subjected to: the duration of the process of exposure to the initial gas mixture is from 15 to 120 minutes, as well as temperatures from 700 to 900°C.

Based on experimental data (Fig. 4), graphs are constructed showing the effect of temperature and duration of the process on the nitrogen content in calcium cyanamide. The results of the analysis are quite consistent with experimental data on the effect of temperature on the process of obtaining calcium cyanamide.

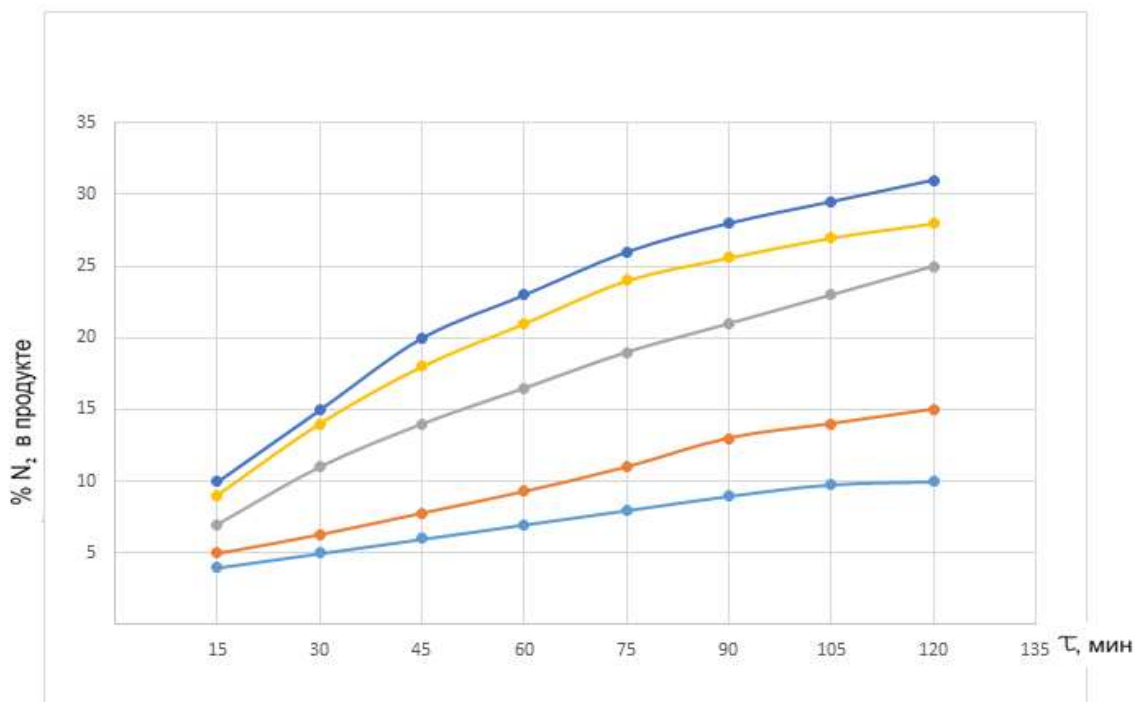


Fig.4 Effect of the duration and temperature of synthesis on the nitrogen content in the product.

The nature of the curves (Fig.4) shows that in the initial period of the synthesis process they differ in steepness, which corresponds to the formation of a product at a high speed. With an increase in the duration of the synthesis process, the curves become flat, tending to a straight line, which reflects the achievement of an equilibrium yield of calcium cyanamide for the corresponding temperatures.

Based on the research results, it can be concluded that the limiting stage of obtaining calcium cyanamide from lime, carbon dioxide and ammonia is the diffusion of the initial gas components through the product layer.

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