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Numerical study of the process of calcinations small-grained limestone in a fluidized bed of inert granular material.

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ABSTRACT

The results of mathematical modeling of the process of production of highly effective sorbent by the method of calcination of small-grained limestone particles in a fluidized bed of inert granular material are given. Dynamic characteristics of the main parameters of the obtained sorbent - total porosity and specific surface in the working range of changes in the technological parameters of the calcination process and the variation of the initial porosity and the pore diameter of the raw limestone - are constructed. The obtained results allow to determine at the design stage the initial data for the hardware and technological design of the process of calcination of small-grained limestone.

***KEYWORDS**: limestone, calcium oxide, sorbent, specific surface, porosity, fluidized bed.*

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I. INTRODUCTION

The use of disperse materials on a carbonate basis (limestone, dolomite, carbonate-containing bentonite clay) are widely used as sorbents in various industries: for the capture of harmful gas compounds (SO_2, HCI, HF) [1]; the absorption of petroleum products from the aqueous medium [2], etc. To obtain the sorbent, the raw material must be activated by thermal, chemical or other means [3-4]. One of the sorbents used is highly reactive lime: the product of burning carbonaceous rock with high porosity and specific surface area. Obtaining highly reactive lime with the necessary sorption properties in traditional apparatuses with coarse-grained material calcination is due to the complexity of ensuring uniform distribution of pores in grain volume. Until the required degree of transformation in the center of the particle in the surface layers of CaO formed, there is a sintering of pores with a significant loss of adsorption capacity of the sorbent. In order to obtain a highly reactive sorbent, it was suggested that the process of burning small-grained limestone in a fluidized-bed apparatus of inert granular material [5] was proposed, which allows to provide the desired temperature and time of the residence of small-grained particles in the reaction zone, to form a product with desired physic-chemical properties.

II. LITERATURE REVIEW AND PROBLEM STATEMENT

The process of burning limestones under different conditions was carried out by a number of authors in works [6-7]. Investigation of the properties of various limestone after calcining by different methods shows a large difference in the values of their specific surface from 6.6 to 104 m^2/g. This testifies to the fact that in samples of a sorbent with a low specific surface there was a sintering of pores due to excessive time of staying in the high temperature zone. The limestone calcination process, even for relatively small fractions <100 microns, depends to a great extent on the size of the particles, the heat and mass transfer, the kinetics of the reaction and the temperature of the firing. Thus, in order to obtain highly reactive lime, it is necessary to burn the small-grained material within a time sufficient to achieve the full conversion of the raw material, at such a temperature of the external gas phase, which provides the maximum calcination rate, but excludes the possibility of sintering of pores in the outer layer of the sorbent particles.

The physical process of calcination of small-grained limestone particles can be represented as an idealized quasi-homogeneous model [6], based on the notion that the decomposition reaction of $CaCO_3$ (1) passes through the entire volume of a particle with a velocity depending on temperature and CO_2 concentration in pores whose values change in radius:

$CaCO_3 \leftrightarrow CaO + CO_2\uparrow.$ (1)

The quality of the sorbent also depends to a large extent on the dispersed composition of the raw material, the initial porosity, pore size etc. For the time of the residence of small-grained particles in the reaction volume, the mass of which varies during the processing, the aerodynamics of the fluidized bed have a dominant influence.

III. THE AIM AND OBJECTIVES OF RESEARCH

The purpose of the study is to carry out detailed numerical experiments on the analysis of the burning of small-grained limestone particles in order to determine the nature of the dependence of the velocity of the process of calcination and, accordingly, changes in porosity and specific surface and the degree of conversion of limestone from the factors characterizing the particle-the particle radius, initial porosity and the diameter of the channels, and the conditions of the process - the temperature and composition of the external gas phase. Knowledge of the dynamics of the changes of the indicated indicators of the calcination process for each type of limestone is necessary for the choice of structural and technological parameters of the installation, in which the temperature and gas dynamic conditions in the fluidized bed will guarantee a time of contact of the particle with the surrounding environment, which is necessary to achieve the desired degree of development of porosity and specific surface in the absence of baking contact surface.

IV. NUMERICAL EXPERIMENT

4.1 Staging A Numerical Experiment

Obviously, achieving the necessary qualitative characteristics of the sorbent (porosity structure, specific surface), depending on their many interconnected parameters, is a complex optimization problem, the solution of which is experimentally problematic. In this connection, a mathematical model was constructed [7], an appropriate mathematical framework was developed, on the basis of which a detailed numerical study was performed on the process of small-grained limestone calcination in a fluidized bed of inert granular in a wide range of changes in technological parameters and characteristics of the raw material. As a result, a rather complete picture of the process of activation of limestone is obtained.

The numerical experiment is carried out in the range of operating parameters of the calcination process: the temperature of the external gas phase t = 900-1200 °C, the diameter of the particle d = 80-200 microns, the initial porosity of the limestone ε_0 = 0.03-0.48, pore diameter d_p = 3.84 - 17 nm, diffusion coefficient of CO_2 in pores D_{por} = 0.0002 m^2/s, CO_2 diffusion coefficient between the gas stream and the surface of the limestone particles D = 0.002 m^2/s with.

4.2 The Results Of The Numerical Experiment

Fig. 2-6 shows the dynamic characteristics of the basic parameters of the calcination process, on which the rate of conversion of a limestone particle into a high-calcium oxide (sorbent) and sorption property of the obtained CaO depends. The speed of the calcination process is determined by the particle temperature and the concentration of carbon dioxide in the limestone pores, which are distributed over the particle radius. Changing these parameters in time and in coordinates r = 0 (center of particle) and r = 1 (particle surface) is presented in Fig. 2-3 (since the diameter of the particle is small and its heating is very rapid, the temperature profiles on the surface and in the center practically coincide) . The speed of the particle conversion process is of paramount importance because of the limitation of the residence time in the reaction zone, which is determined by the hydrodynamics of the fluidized bed. In Fig. 4-6, the dynamic characteristics of the parameters that actually characterize the dynamics of the calcination process and the quality of the sorbent obtained, namely, the degree of conversion of the output product x (i.e. the degree of use of the raw material), the change in porosity ε and the specific surface of the particle S, m^2/g. Moreover, the specific surface represented as the total for the whole particle, is converted into a unit of its mass.

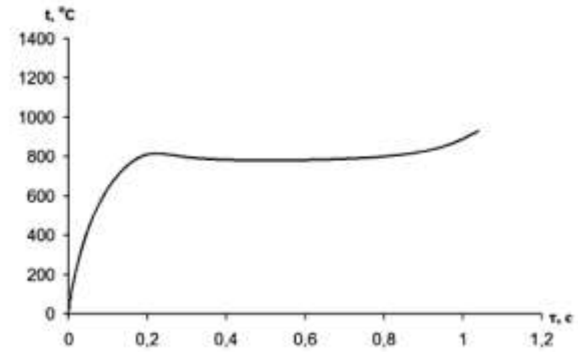

Fig. 2. Dependence of the temperature within the particle on the particle residence time

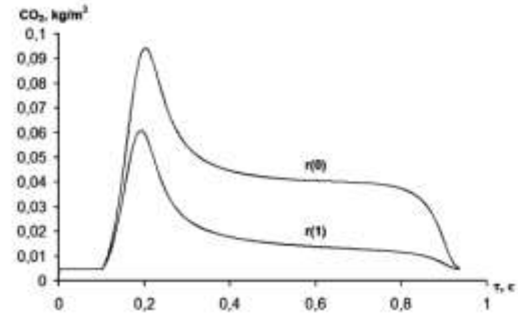

Fig. 3. Dependence of the concentration CO_2 by decomposition of the particle on the particle residence time

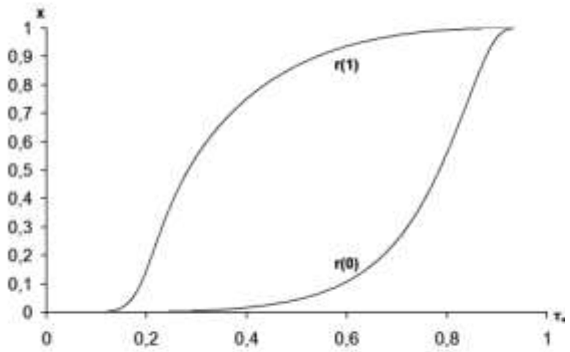

Fig. 4. Dependence of the extent of calcination on the particle residence time

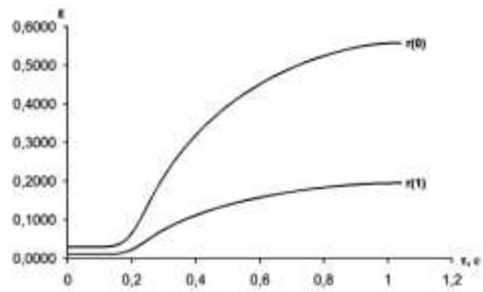

Fig. 5. Dependence of the porosity on the particle residence time

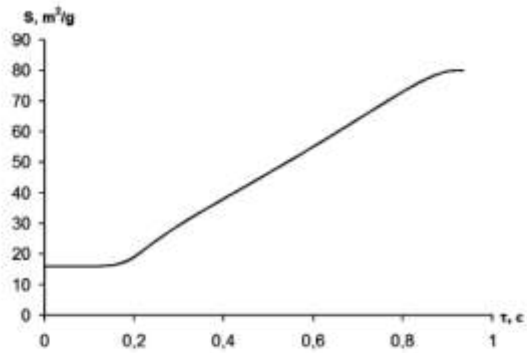

Fig. 6. Dependence of the specific surface area on the particle residence time

4.3 Parametric Sensitivity Of The Process Of Calcinations

The main purpose of the computational experiment is to determine the nature of the influence of the quality of the raw material (dispersion, porosity, pore diameter, thermophysical characteristics, which depend on the natural origin of the limestone), and the variables of the parameters of the calcination process (temperature, residence time of the particle in the reaction zone, determined by hydrodynamics of the fluidized bed) the disered product (porosity and specific surface of the sorbent), the degree of use of raw materials and process performance.

As noted above, the fluid dynamics of the fluidized bed, the temperature and residence time of fine particles in the reaction zone are the determining factors of influence on the final parameters of the limestone sorbent, since the excessive presence of a limestone particle after the endothermic reaction of decomposition of limestone in the high temperature zone leads to the sintering of the particle, while the porosity and specific surface area decreases and, consequently, the reactivity of the sorbent is lost. On the other hand, the inadequate residence time of the small-grained particle in the reaction zone leads to insufficient transformation of limestone, the removal of raw material, and the degradation of the quality of the final product. A series of numerical experiments was carried out to determine the influence of the particle temperature and its dimensions on the main indicators of the quality of the sorbent (porosity, specific surface) at the variation of these parameters in a possible range of their changes, the results of which are presented in Fig. 9-12.

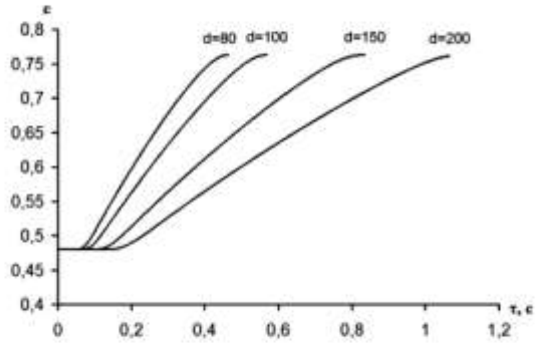

Fig. 9. Dependence of the porosity on the different particle size as a function of the particle residence time, t = 1000 °C

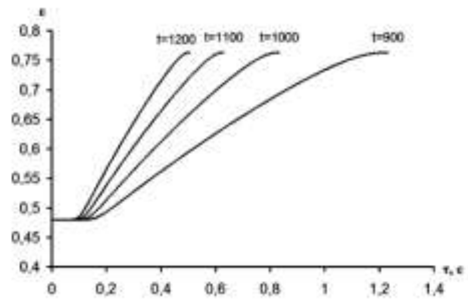

Fig. 10. Dependence of the porosity on the different temperatures as a function of the particle residence time, d = 100 μm

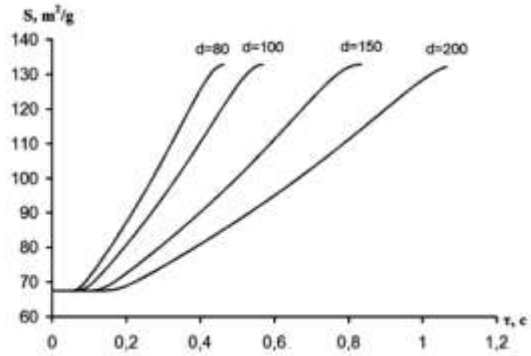

Fig. 11. Dependence of the specific surface area on the different particle size as a function of the particle residence time, t = 1000 °C

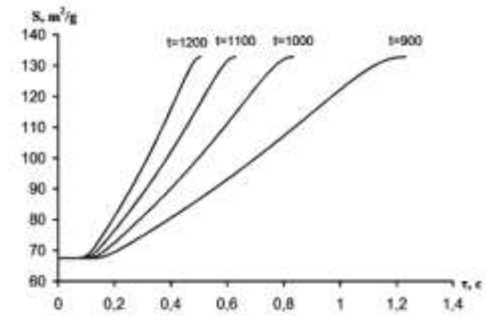

Fig. 12. Dependence of the specific surface area on the different temperatures as a function of the particle residence time, d = 100 μm

In particular, the obtained dependences of Fig. 9-12 allow us to determine the required residence time of particles of different diameters in the reaction volume to ensure the full degree of their transformation (x = 1), depending on the temperature of the particle (Fig. 13-14).

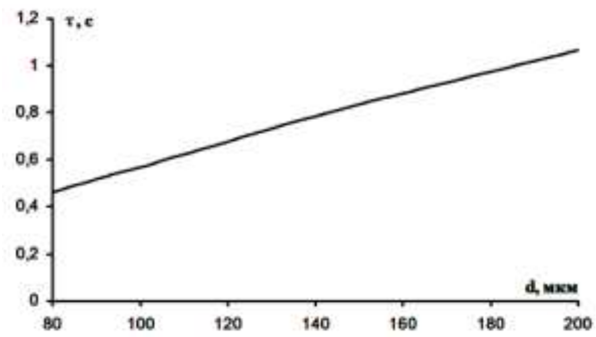

Fig. 13. Dependence of the particle residence time on the different particle size, t = 1000 °C, x = 1

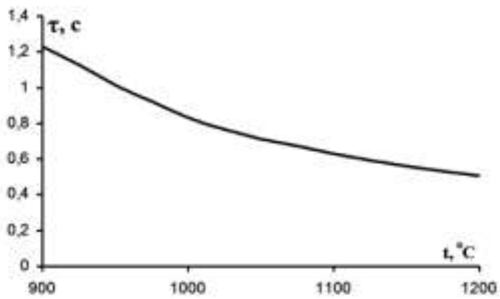

Fig. 14. Dependence of the particle residence time on the different temperatures, d = 100 μm, x = 1

V. DISCUSSION OF THE RESULTS OF A NUMERICAL EXPERIMENT

According to the results of numerical experiments, a detailed course of the calcination process is followed: the heating of the small-grained limestone particles to a temperature of 600 °C occurs within 0.1-0.2 seconds (Fig. 2), which is the beginning of the active process of calcination. Further, during 0.2-0.7 s there is a process of conversion of limestone into quicklime (Fig. 4) with increasing degree of conversion x, a porous structure (Fig. 5) develops with the formation of the corresponding magnitude of the specific surface (Fig. 6).

An important part of understanding the process of calcination is the result of calculating the distribution of carbon dioxide concentration in the particle radius shown in Fig. 3, since the concentration of CO_2 and particle temperature are the determining parameters for the velocity of the endothermic reaction (1).

The main parameters of the quality of the sorbent-final porosity and specific surface, substantially depend on the initial porosity of the source limestone, as seen from Fig. 15,16.

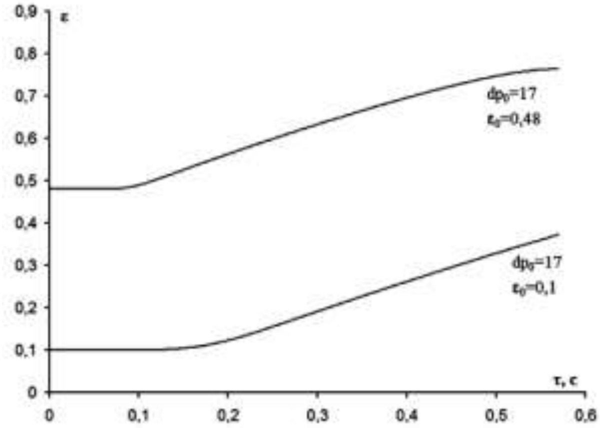

Fig.15. Change of the porosity of particles with different initial porosity.

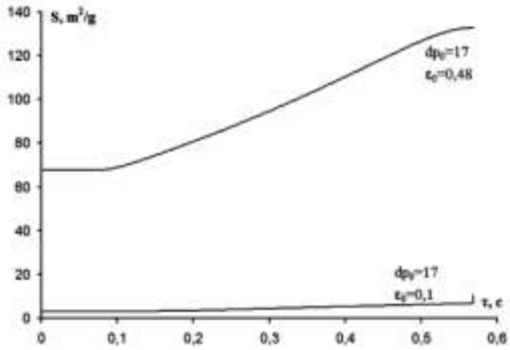

Fig. 16. Change of the specific surface for particles with different initial porosity.

The analysis of the results of computational experiments allows us to formulate the requirements for the raw material and to find optimal solutions of the problem of the choice of technological parameters (temperature, contact time) that provide the basic parameters of the calcination process: the conversion of a limestone particle into lime with the maximum indexes of the degree of conversion of limestone, porosity and specific surface of the sorbent.

The dependencies (Fig. 13-14) allow at the design stage to determine the aerodynamic characteristics of the fluidized bed in which the fine particles of the sorbent are removed from the layer of inert material with the desired qualitative porosity and specific surface properties.

VI. CONCLUSIONS

1. On the basis of the computational experiment, the parametric sensitivity of the burning process of finely divided limestone particles to the physical characteristics of the limestone and the technological parameters of the calcination process was investigated.
2. The results of numerical experiment allow to search for optimal parameters of the process of burning limestone, the determination of which experimentally is problematic or impossible.
3. The computational experiment significantly deepens the understanding of complex physic-chemical interconnected processes in the macropores of the particle, reduces the volume of physical experiments, provides a complete database for determining the optimal design and technological design of the process of calcination with a large-scale transition to a given productivity.

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