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Model of continuous production of fine silicon carbide

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Abstract. A detailed analysis of heat and mass transfer processes and chemical transformations in a high-temperature gas flow with solid particles is a very difficult task. Using the phenomenological approach, the authors succeeded in obtaining a closed system of equations, the solution of which allows us to determine the main parameters of the stationary process for the production of silicon carbide. It is assumed that the implementation of two technological conditions: maintaining the required temperature level of the system "fluidizing gas - solid particles" in the production process and the continuous removal of both the final product - silicon carbide, and small unreacted particles. Variant calculations were carried out with the determination of the parameters of the continuous process for the production of fine silicon carbide, in particular, the optimal ratio of the initial sizes of carbon-containing particles and SiO_2 particles and the maximum possible yield of the final product. The qualitative agreement of the calculation results with the data obtained in experiments with periodic loading of reacting components is shown.

1. Introduction

The work is devoted to the creation of a general model, which describe the production of fine silicon carbide in a boiling (fluidized) high-temperature layer of reactive particles in an installation for continuous production of the final product.

1.1. Basic provisions of the model

The flow of heated gas in the annular space moves upwards. It has axial and tangential velocity components.

A constant electric (thermal) power is supplied with the boiling (fluidized) bed. Maintaining the temperature level in the layer is carried out by means of radiation-convective heat exchange of the flow with hot electrodes and as a result of direct heat release in electrically conductive particles.

When stochastic movement in the reaction space, SiO_2 particles become liquid and in the presence of carbon-containing material particles evaporate to form SiO and CO.

The particles of the carbon-containing component react with gaseous SiO to form silicon carbide.

Basic chemical reactions [1]

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$$C + SiO_2 = SiO + CO \tag{1}$$

and

$$2C + SiO = SiC + CO \tag{2}$$

Formation SiC occurs on the surface of a porous reactive carbon-containing particle with a decreasing effective contact area with time in the reaction volume.

1.2. Key Assumptions

The temperatures of the gas and the solid carbon-containing carbide-forming particles are equal to each other. The temperature of the liquid phase SiO_2 is equal to the melting point of silicon oxide. Within the reaction zone, these temperatures do not change. The process of carbide formation is generally regarded as quasistationary. Particles of carbon-containing material can be divided into 2 groups (corresponding indices «c_1» and «c_2»): «c_1» - the first group of particles - accumulators of the final product (SiC) according to reaction (2); «c_2» - the second group of particles that, together with the particles SiO_2 around them, form a source of gaseous generation SiO by reaction (1) - section I of the installation.

All particles at any stage of the process are spherical

Any of the particles in the reaction zone is involved in any process that results in a change in its mass.

Particles of the solid fraction are not removed with the gas stream that provides fluidization.

2. Key notes

R, r - initial particle radii; $\bar{r}_i$ - the calculated radii of the particles in the reaction zone; M - molar mass; τ - time; T - temperature; m - weight; μ - mass flow; ρ - density; σ - surface tension; $\tilde{R}$ - universal gas constant; q - heat flux density; α_C - the share of carbon in the particle; k_D - the rate of chemical reaction; C - concentration; E - activation energy; D - diffusion coefficient; V - gas flow rate; $\tilde{n}$ - number of particles; n - particle feed rate; F - is the cross-sectional area; H - is the height of the fluidized bed.

3. Basic equations of carbide formation process

3.1. Evaporation of the liquid phase (droplets) of SiO_2.

We take advantage of the result on the evaporation of dispersed particles obtained in [2] and take into account the factor limiting the evaporation process. This factor is the necessary heat influx from the outside to maintain the melting point with intensive cooling of the particle due to evaporation. We obtain a differential equation to determine the current radius r_{SiO2} of the liquid particle SiO_2during the heat supply, which limits the evaporation process.

The solution to the equation will look like this:

$$\frac{1}{2}\left(R_{SiO_2}^2 - r_{SiO_2}^2\right) + \left(R_{SiO_2} - r_{SiO_2}\right)\frac{\Delta h_{SiO_2}}{q_{heart}} b = \frac{b}{\rho'_{SiO_2}}\tau, \tag{3}$$

where

$$b = \frac{2M_{SiO_2}\sigma\rho''_{SiO_2}}{\tilde{R}T\rho'_{SiO_2}}\left(\frac{\tilde{R}T}{2\pi M_{SiO_2}}\right)^{1/2}. \tag{4}$$

3.2. Change in the size of the carbide-forming particle

In accordance with the reaction equation (2), for a separate particle of the group « c_1» can be written

$$r_{c1} = R_{c1} - \psi_{c1}\tau, \tag{5}$$

where

$$\psi_{c1} = \frac{2M_c}{\rho_c}\alpha_c k_{D1} C_{SiO}, \tag{6}$$

$$k_{D1} = k_{01} exp\left\{-\frac{E_{c1}}{\tilde{R}T}\right\} f(D_{c1}), \tag{7}$$

$f(D_{c1})$ is a function that takes into account the diffusion of *SiO* through the *SiC* layer.

3.3. Resizing the carbonaceous particle of the group « c_2»
According to the reaction equation (1) and by analogy with equation (5) for the group of particles « c_2» we can write:

$$r_{C_2} = R_{C_2} - \psi_{C_2}\tau \, , \tag{8}$$

where $$\psi_{C_2} = \frac{M_C}{\rho_c}\alpha_C k_{D2} C^H_{SiO} \, , \tag{9}$$

$$k_{D2} = k_{02} exp\left\{-\frac{E_{c2}}{\tilde{R}T_{C_2}}\right\}. \tag{10}$$

3.4. Balance equations for the conservation of mass components of the production of silicon carbide.
Balance for particles SiO_2:

$$\rho_{SiO_2} n_{SiO_2} \frac{1}{3}\left[R^3_{SiO_2} - \left(r^{out}_{SiO_2}\right)^3\right] = \tilde{n}_{SiO_2}\bar{\eta} b \bar{r}_{SiO_2} \, , \tag{11}$$

where $\bar{\eta} = \frac{1}{1+\frac{\Delta h_{SiO_2}}{q_{heart}\bar{r}_{SiO_2}} b}$.

Balance for carbon-containing particles « c_1»

$$\rho_c \, n_{c1} \frac{1}{3}[R^3_{c1} - (r^{out}_{c1})^3] = 2\tilde{n}_{c1} M_c \alpha_c \bar{r}^2_{c1} k_{D1} C_{SiO}. \tag{12}$$

Balance for carbon-containing particles «c_2»:

$$\rho_c \, n_{c2} \frac{1}{3}[R^3_{c2} - (r^{out}_{c2})^3] = \tilde{n}_{SiO_2} (\frac{M_c}{M_{SiO_2}})\bar{\eta} b \bar{r}_{SiO_2} \, . \tag{13}$$

3.5. The flow of the gas mixture at the outlet of the reaction volume
With gases coming out of the reaction volume, gaseous *SiO* is also removed, which has a negative effect on the production efficiency of silicon carbide. Find the minimum required volume flow of exhaust gases. Assuming that at the entrance to the reaction zone the volumetric flow rate of gas (V_{flu}), which provides for the creation of a fluidized bed, corresponds to the minimum velocity — the velocity of the onset of fluidization (u_{mf}). We write the continuity equation for the gas mixture in the form

$$\rho^{out}_g V_{otv} = \rho^{inp}_g V_{flu} + \textstyle\sum_i \Delta\mu_i, \tag{14}$$

Where $\Delta\mu_i = n_i \rho_i \frac{4}{3}\pi R^3_i \left[1\text{-}\left(\frac{r^{out}_i}{R_i}\right)^3\right]$ characterizes the mass loss of solid components - participants in the process per unit time.

We will rewrite equation (14) for the participants of the reactions of the silicon carbide production process taken into account. We get:

$$V_{otv} = \frac{1}{\rho^{out}_g}\left\{\rho^{inp}_g V_{flu} + \Delta\mu_{C_1}\left(1 - \frac{M_{SiC}}{2M_C}\right) + \Delta\mu_{C_2} + \Delta\mu_{SiO_2}\right\} \tag{15}$$

Here: ρ^{inp}_g –- corresponds to the density of the fluidizing gas at the inlet, and ρ^{out}_g - the density of the mixture of gases at the outlet of the reaction volume.

We take into account [3] that $V_{flu} = u_{mf} \cdot F$, then, for the start of fluidization and the process of expansion of the layer of spherical particles during their random packing

$$V_{flu} = 1{,}667 \frac{u_{mf}}{H} \textstyle\sum_i \frac{m_i}{\rho_i}, \tag{16}$$

Equations (3) - (16) play an important role in determining the roots of the system of algebraic equations describing the physically realizable process for the production of silicon carbide, as well as the choice of loading parameters and the supply of reagents to the reaction volume.

3.6. The yield of the final product

In accordance with reaction (2), an expression can be obtained for determining the performance of a silicon carbide production facility:

$$\mu_{SiC}^{out} = \mu_{C_1} \frac{M_{SiC}}{2M_C}\left[1 - \left(\frac{r_{C_1}^{out}}{R_{C_1}}\right)^3\right] \quad (17)$$

4. Results of calculation of the basic parameters of the process of production of silicon carbide

Transformation of the above system of equations, taking into account the numerous limiting factors reflecting the physical limitations of the roots of the equations recorded for the desired parameters, made it possible to create a working computational program for determining the characteristics of the continuous production of fine silicon carbide in a high-temperature fluidized bed. The results of the calculation of the yield of silicon carbide are shown in figures 1 and 2.

In the calculations $r_{C_1}^{out}$and $r_{C_2}^{out}$asked how dependent on the technological features of a continuous production process.

All other parameters related to the production process as a whole and to individual components - participants of the continuous physical and chemical transformation, are determined by solving the above equations as a system describing a quasistationary process. Missing for the correct solution of the source data are taken from the following scientific publications: the pre-exponential factor and the activation energy for reaction (2) - [4]; reaction rate constant (1) - [5]; the parameters of SiO_2 in the liquid and gaseous state — [6]-[8].

The heat supply to the SiO_2 liquid particle was estimated according to the recommendations of [9] (based on radiation heat transfer) and [10] (based on convective heat transfer).

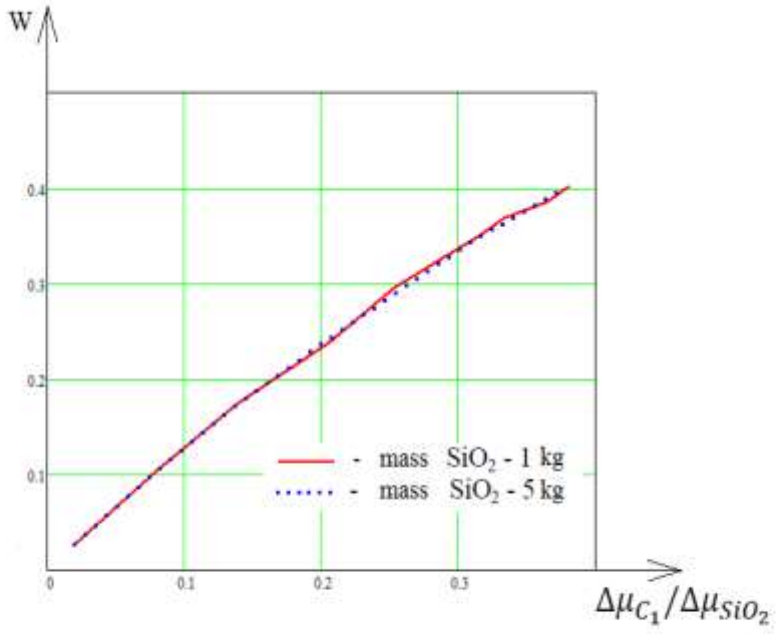

Figure 1. Graph of the relative mass yield of *SiC* as a function of the relative $\Delta\mu_{C_1}/\Delta\mu_{SiO_2}$ with a different mass of SiO_2 in the reaction volume and a constant radius $R_{SiO_2 0}$ = 6.4·10^{-5} m of SiO_2 particles.

Figure 1 reflects a very interesting calculation result - independence of the relative yield of the final product $W = \frac{\mu_{SiC}^{out}}{\sum_i \Delta\mu_i}$ from the mass of an important component of the SiO_2 production process at fixed values of the initial dimensions of carbon-containing components: R_{C_1}, R_{C_2}. The latter indicates the possibility of modeling large-scale industrial production in the laboratory.

Currently, there are options for experimental plants, where they receive fine silicon carbide in the process with periodic loading of the reacting components. One of such installations (reactor ETKS) is described [1], [11]. It is important to compare the recommendations formulated on the basis of experimental studies [11], at the choice of the optimal ratio of the sizes of carbon-containing particles and SiO_2 particles with the results of calculations. In our notation, recommendations are defined as (R_C / R_{SiO_2}) opt = 1.5 - 1.7.

Figure 2 demonstrates the qualitative correspondence of the maximum relative rate of generation of the final product to the optimum ratio of particle sizes of the c_1 and SiO_2 fractions. Indeed, since the graph is constructed for $R_{SiO_2 0}$ = 6.4·10^{-5} m, and in the calculations it was assumed R_{C_1} = R_{C_2} = 2·10^{-4}, then (R_C / R_{SiO_2}) opt corresponds (R_{SiO_2} / $R_{SiO_2 0}$) opt = 1.84 - 2.08.

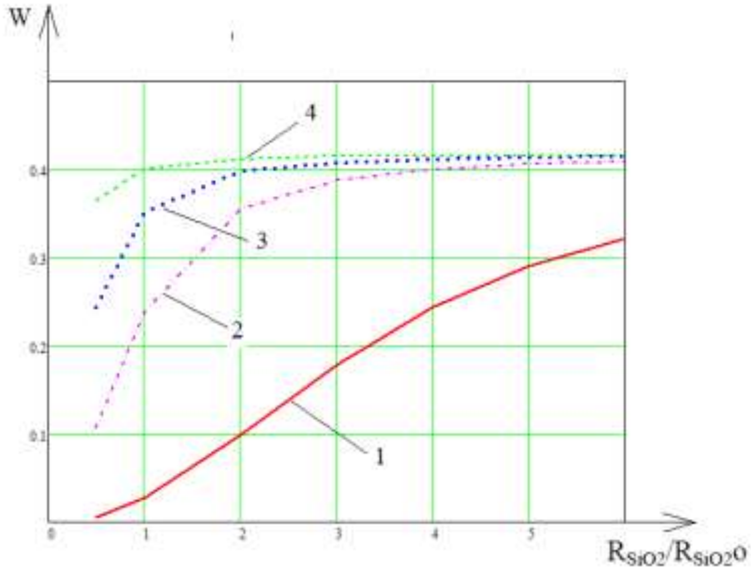

Figure 2. Graph of the relative mass yield of *SiC* on the relative radius of SiO_2 particles $R_{SiO2}/R_{SiO2}o$ with mass of SiO_2 1 kg and different masses of carbon m_c in the reaction zone; 1 - m_c =0.054 kg; 2 - m_c =0.831 kg; 3 - m_c =3.062 kg ; 4 - m_c =13.884 kg.

In [1], an analysis of the elemental and phase composition of the charge samples obtained in the E-TXS is presented. In particular, it was found that the sample of the charge after being in the reaction zone of the ETX reactor for 600 minutes and the variable temperature of the reactor (heating to 1800 ºC) contained 41.3% *SiC* (cubic). Figure 2 shows that the *SiC* yield in a continuous process cannot exceed 40.8%, which can be noted as a qualitative fit between the calculation and the experiment.

5. Conclusion

On the basis of a modern understanding of the physical processes under investigation, a general mathematical model has been created for calculating the main parameters of the stationary continuous production of fine silicon carbide in a boiling (fluidized) high-temperature layer. Calculations showed the consistency of the obtained data on the parameters of different components - participants in the process of production of silicon carbide. The model has been tested at a qualitative level.

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