Kinetics model of convective drying

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Abstract. The article considers various approaches to modeling the drying of finely dispersed products: based on the classical theory of diffusion, from the standpoint of the thermodynamics of an irreversible process. An analysis of the scientific and technical literature on the problem revealed not high accuracy of the convective drying models of finely dispersed products developed on the existing theoretical basis. It is shown that the improvement of drying modeling can be based on the kinetics theory of chemical reactions. In this case, drying is considered as a quasi-topochemical heterogeneous reaction. At the same time, a combined approach to drying modeling is proposed: the phenomenological equation of chemical kinetics, which describes the speed of the process, is supplemented by the experimentally established relationship between the temperature and moisture content of the material. Based on the proposed approach, mathematical models of the kinetics of convective drying of finely dispersed products for periods of constant and decreasing speed have been developed, which allow to establish a clear type of the influence of the temperature regime on the kinetics of the process and reduce the amount of experimental research. A mathematical model of the temperature curve of drying for a period of decreasing speed is developed. The relationship between the speed of the first and second drying periods is established. The reliability of the obtained mathematical models is confirmed by experimental verification of the kinetic equation obtained for a period of constant velocity.

1. Introduction
The creation of innovative drying technologies and efficient drying devices is impossible without further development of the theoretical foundations of this process. Existing methods of calculation and design of drying equipment do not provide sufficient accuracy in forecasting the actual process flow. Therefore, the development of innovative drying methods involves a long period of experimental research and subsequent stages of testing and finalizing the prototype. This causes high development cost and decrease of production development rates. In this regard, there is an urgent need to develop theoretical models of drying and engineering methods for calculating drying processes that provide a significant acceleration in new equipment development. In our opinion, the development of the drying theory can be based on another theoretical foundation. At the same time, the disadvantages of known approaches can be compensated by the advantages of new ones. In particular, drying can be represented as a thermal heterogeneous physical and chemical process and, therefore, the laws of chemical kinetics can be applied to the calculation of this process velocity.

In connection with the above, the overall goal of this work is to develop theoretical provisions for convective drying of dispersed food products based on the laws of chemical kinetics, the use of which will reduce the amount of experimental research, costs and time for the development of innovative technology and drying techniques.
2. Analysis of the completed researches of the issue, setting the goal and objectives of the research

The scientific basis of mathematical modelling of any process is the physical laws and physical and chemical relations that this process follows, as well as general methods of research and calculation of processes and devices for food and chemical industries.

The most important issue in the theory of drying is the kinetics of the process, knowledge of which allows you to determine one of its main characteristics - the duration of drying. According to established concepts [1,2,3,4,5,6], based on the analysis of a large volume of experimental data, the process of convective drying includes two stages: period of constant velocity (the first period), when

\[
\frac{dU}{d\tau} = N_1 = \text{const}, \quad U_{i} \geq U \geq U_{k}, \quad (1)
\]

and the period of velocity ramp down (the second period), when

\[
\frac{dU}{d\tau} = N_2(U), \quad U_{k} > U \geq U_{i}, \quad (2)
\]

where \(U_i\), \(U_k\), \(U_{sp}\), \(U\)—correspondingly, initial, critical, final and current moisture content in the product; \(\tau\)—process duration.

Th. Sherwood and Lewis proposed using the apparatus of the classical diffusion theory based on Fick's law as a theoretical basis for modelling the kinetics of convective drying. However, the differential equation of mass conductivity obtained with this approach can only be used in approximate solutions under isothermal drying conditions [6]. In non-isothermal drying conditions, a temperature gradient occurs along the thickness of the product, causing an additional flow of moisture. Therefore, it is necessary to consider the phenomena of mutual heat and mass transfer. This is most successful from the point of view of the thermodynamics of an irreversible process. In the thermodynamic approach, the final energy actions of the driving forces are determined, and the molecular structure of the substance and the mechanism of the process are not considered. Applying the equations of non-equilibrium thermodynamics to the drying modelling, A.V. Lykov and his colleagues developed the theory of moisture and heat transfer, which is now a foundation for research and mathematical modelling of process [1]. In this case, wet materials are considered as capillary-porous colloidal systems, the moisture transfer in which is determined by the coefficients of moisture content and temperature. The intensity of any flow of energy or mass is determined by phenomenological laws as the product of the kinetic coefficient to the corresponding thermodynamic force [1]. Such approach to drying requires a separate consideration of determination of the driving force of the process [7, 8], which has so far been determined by a priori postulating a simplified model of the drying process.

The system, by A.V. Lykov, of nonlinear differential equations of heat and mass transfer, which describes the non-stationary process of heat and mass transfer in wet bodies under any conditions was widely recognized and is widely known. However, as the author himself rightly points out, for calculating the velocity of the second drying period, the solution of this system of equations “can be used as the first approximation” [1].

This circumstance leads to the fact that, along with the general theory of transport in the drying process, attempts are made to build simpler, private models, as well as to study the possibilities of building models on another theoretical basis [4, 6].

An empirical approach based on generalization of experimental data using theory of similarity, probability, and mathematical statistics is widely used for modeling purposes.[2,5,8,9]. The experiment data is processed in the form of relation equations between generalized variables (criteria and similarity simplexes).

However, due to variability of numerical values of the criteria and coefficients in the drying process, the models and methods of calculation based on them are limited and approximate [6]. In addition, the experimental modeling method causes large material and time expenses, since it requires production of a prototype and conducting experiments.
Therefore, for specific processes, they tend to use simpler descriptions that reflect only the most significant types of transfer, for example, the thermal conductivity equation [1, 6]. These equations include equivalent transfer coefficients that are easier to estimate and measure. But such models have significant limitations.

Engineering practice has widely used methods for modeling drying kinetics based on approximating kinetic curves in the form of: \( N_2(U) = f(U) \). Thus, G.K. Filonenko [10] based on experimental data analysis, proposed a model of kinetics for the second drying period in the form of an empirical dependence:

\[
N_2(U) = N_1 \cdot (U - U_\rho)^n /[A + B \cdot (U - U_\rho)^m], \tag{3}
\]

where \( m, A, B \) – constants determined from experience; \( m \) – characterizes type of the moisture relation with material and does not depend on other factors.

The analysis of experimental models of drying kinetics makes it possible to emphasize the experimental fact revealed by many researchers: the velocity of the second drying period can be represented as the product of the velocity of the first period (the maximum speed) to a certain function \( \psi(U) \) [10]. This means that for any drying methods, the mathematical model of kinetics of the second period can be represented by equality:

\[
N_2(U) = N_1 \cdot \psi(U). \tag{4}
\]

The simplest generalizations of experimental studies of heat and mass transfer are obtained in the form of simple empirical power dependencies of the form [11]:

\[
y = AX_1^n \cdot X_2^n \cdots X_k^n, \tag{5}
\]

where \( X_1, X_2, \ldots X_k \) – drying parameters, \( Y \)–characteristic (for example, velocity) of the process.

The main drawback of all empirical models is that they are obtained as a result of generalization of experimental data obtained for specific products dried on experimental prototypes. Drying conditions on prototypes do not fully correspond to real conditions in industrial devices, which causes insufficient accuracy of the obtained models.

To achieve this goal, it is essential to solve the following objectives:

1. Develop general form of mathematical model of convective drying based on the laws of chemical kinetics.
2. Determine the form of mathematical model for the period of constant drying velocity.
3. Determine the form of mathematical model for the period of ramping down drying velocity.

3. Presentation of the research materials

Developing a mathematical model for convective drying of finely dispersed food products, the following process features were taken into account. The modern development of the theory and technology of drying is characterized by a transition from drying product in a layer to development of dryers with an active hydrodynamic mode, which provides a high intensity of heat and mass transfer. In this process, due to the small size and relatively high thermal conductivity of a product particles, heat transfer often occurs under external conditions, i.e. it is determined by the rate of heat supply to a material; and the process of mass transfer is performed under the internal conditions [1, 6], since the coefficients of moisture diffusion in food products are very small. The analysis of numerous experimental studies [1, 6] showed that during the first drying period, when the product humidity is high, the process is limited by the heat transfer rate at the interface, and during the second drying period by the heat and moisture transfer rate inside a particle. Since the use of active hydrodynamic modes allows the use of a relatively high temperature of the drying agent, evaporation occurs at a certain constant depth (\( \delta = \text{const} \)) [1, 6]. The distribution curves of the material temperature and relative humidity remain constant. From the point of view of chemical kinetics, the evaporation zone can be considered as a kinetic area, and for small particle sizes (\( d \leq 2\delta \)), it can be assumed that the entire drying process takes place in the kinetic area.
Consider drying as a heterogeneous physical and chemical process and apply the laws of the kinetics of chemical reactions to calculate velocity of the process. From the point of view of the goal and objectives of this study, the issue of mathematical modeling of drying kinetics is similar to mathematical modeling of the kinetics of topochemical reactions. These include the processes of burning mineral raw materials (dolomite, magnesite, calcite, etc.), dehydration of crystallohydrates (sulphates of copper, zinc, and others) [12]. Experimental study of the thermal decomposition reaction usually begins with determining the type of curve that expresses the change of the decomposed substance fraction $\alpha$ over time $\tau$ [12], similar to how the type of curve for drying a wet substance is determined [3, 5]. The value $\alpha$ in the kinetics of chemical reactions is also called the degree of transformation of the substance and is determined by the equation:

$$\alpha = \frac{\Delta m_{i}}{\Delta m}, \quad (6)$$

where $\Delta m_{i}$, $\Delta m$ – current and final values of the substance amount converted during the process.

Analyzing the thermal decomposition curves, the attention was drawn to the analogy between these curves of some substances and the drying curves. For example, during the dehydration of crystallohydrates, there are periods of constant and decreasing (slope period) process velocity both on the thermal decomposition (separation) curves and on the drying curves [13]. Since topochemical reactions follow the usual chemical laws, this analogy indirectly indicates the possibility of formal application of the laws of chemical reaction kinetics to the description of drying kinetics.

The dependence of the reaction rate on the concentration of reacting substances expresses the law of active masses. It is based on the assumption that the value of the reaction rate depends on the number of collisions of reacting molecules per unit of time with a frequency proportional to the product of the concentrations of reacting substances in degrees whose indicators correspond to stoichiometric coefficients [12]. Theories of the kinetics of chemical reactions explain the influence of various factors on the reaction rate (primarily the influence of temperature) and the calculation of the velocity constant. One of the first of such theories is the Arrhenius theory of active collisions [12]. According to Arrhenius, every reaction proceeds through an intermediate stage associated with the formation of a special modification-active molecules that possess increased energy. The mathematical expression of the dependence of the reaction rate constant on temperature (Arrhenius equation) has the form [12]:

$$k = k_{0} \cdot \exp\left(-\frac{q}{RT}\right), \quad (7)$$

where $k$– the rate constant of the chemical reaction; $k_{0}$ – a pre-exponential multiplier that depends on the physical and chemical properties of the reaction system; $q$-the activation heat, which characterizes the thermal effect of the transition of molecules from the normal to the active state. In the practice of kinetic research, the activation energy $E$ is usually considered instead of the activation heat $q$, since for most reactions the difference between $E$ and $q$ does not exceed 2%.

Equation (7) contains the temperature of the material as an unknown. Thus, a combined approach to modeling drying is proposed: the phenomenological equation describing the velocity of the process is supplemented by an experimentally established relationship between the temperature and the moisture content of the material.

The temperature of the material in the constant velocity period can be determined by the temperature of the “wet thermometer” of the drying agent $T_{w}$, and the moisture content in this period varies from the initial $U_{n}$ to the critical $U_{kr}$ value.

Starting from the critical moisture content, the product temperature increases and at the equilibrium moisture content is equal to the temperature of the $T_{c}$ drying agent. At any given time, the material temperature can be determined if the temperature curve is known $T = f(U)$.

Developing general view of mathematical model for convective drying based on the laws of chemical kinetics, we will proceed from the following assumptions.
The transition of moisture from the product to the drying agent is associated with overcoming the energy barrier \( E \), the presence of which is due to the fact that each water molecule is more or less firmly connected to the dry part of the product \( E_{sw} \) and, in addition, the transition to the vapor state is associated with the energy consumption for phase transformation.

Water molecules whose energy is greater than or equal to \( E \) can pass into the vapor state. The portion of such molecules in the wet material follows the Boltzmann distribution law.

The largest cross-cut particle size of the material does not exceed two sizes of the evaporation zone. Under these conditions, one can use Arrhenius’ theory of chemical transformations and express the velocity of the drying process by the equation:

\[
\frac{\partial \alpha}{\partial \tau} = f(\alpha) \cdot A \cdot \exp \left( -\frac{E}{RT} \right),
\]

where \( \alpha \) – degree of substance transformation; \( A \) – coefficient; \( f(\alpha) \) – function of the substance transformation degree; \( R \) – universal gas constant; \( T \) – the absolute temperature of the body; \( E \) – energy of activation. The known determination of the substance transformation degree (6) allows us to establish the relationship between \( \alpha \) and the moisture content of the material:

\[
\alpha = \frac{U_H - U}{U_H - U_p},
\]

where \( U, U_H, U_p \) – respectively, current, initial and equilibrium moisture content of the material.

The function of transformation degree of a substance in the theory of chemical reactions can be represented by various equations. The most universal expression suitable for many chemical reactions, including topochemical ones, is the power law of the form:

\[
f(\alpha) = (1 - \alpha)^n = \left( \frac{U - U_p}{U_H - U_p} \right)^n,
\]

where \( n \) – coefficient, called reaction order.

The intensity of heating the material during drying is usually estimated by the relative (dimensionless) temperature coefficient of drying [10,14]:

\[
B = \frac{\partial T}{\partial T},
\]

where \( \Delta U, \Delta T \) - the fixed values of moisture content and temperature, respectively.

We believe that as the fixed values of temperature and moisture content when calculating the relative temperature coefficient of drying, we should take \( U_{sp} = U_p \) and \( T_c - T_m \).

From the above, it follows that the mathematical model of the convective drying rate in general will be determined by the system of equations:

\[
\begin{align*}
- \frac{\partial U}{\partial \tau} &= (U_H - U_p) \cdot \left( \frac{U - U_p}{U_H - U_p} \right)^n \cdot A \cdot \exp \left( -\frac{E}{RT} \right), \\
- \frac{\partial T}{\partial U} \cdot \frac{\Delta U}{\Delta T} &= f(U),
\end{align*}
\]

that in the first drying period, experimental data on drying various products allow us to conclude that in the first drying period, when the process velocity is constant, the material temperature is equal to the temperature of the wet thermometer \( T_m \), the reaction order can be assumed to be zero. Since, during this period, mainly free or loosely coupled moisture is removed, the binding energy can be neglected and the activation energy of its molecules can be assumed to be equal to the heat of vaporization \( r \). Then, in the period of constant drying rate, the mathematical model of the process will be determined by a system of equations:

\[
\begin{align*}
- \frac{\partial U}{\partial \tau} &= N = \left( U_H - U_p \right) \cdot A \cdot \exp \left( -\frac{r}{RT} \right), \\
- \frac{\partial N}{\partial \tau} &= \left\{ \right. \}
\end{align*}
\]
After taking the logarithm of the drying rate expression (14) we get

\[ \ln N_1 = \ln \left[ \frac{(U_s - U_p)}{A} \right] - \frac{r}{R \cdot T_a}. \]  

(15)

From (15) it follows, that in coordinates \( \ln N_1 - 1/T \) dependence of the velocity of the first drying period on the temperature of drying agent should be linear.

The experimental verification of equation (15) was carried out on the basis of results borrowed from literature sources [3] widely known to scientific community [14]. These data are repeatedly verified, their reliability is in no doubt. Below are the results of the study of convective drying of pressed paper, performed by G.K. Filonenko (figure 1), and the results of the study of conductive drying of a thin layer of cellulose (figure 2), performed by Acad. V.V. Krasnikov [14].

Thus, the results of an experimental study of drying different materials, performed at different times by different authors using different drying methods, prove the validity of the considered approach to mathematical modeling of the drying process.

In the second drying period, bound moisture is removed. The velocity of the process depends not only on the temperature of the product, but also on the moisture concentration (according to the law of active masses). The activation energy is defined as the sum of:

\[ E = r + E_{\text{sw}} = r - R \cdot T \cdot \ln \left[ \varphi(U,T) \right], \]  

(16)

where \( \varphi(U,T) \) – equation of the desorption (sorption) curve for the dried material; \( E_{\text{sw}} \) – the binding energy of moisture with material, which was determined by the well-known equation of Acad. A. Rebinder [10].

For capillary-porous colloidal bodies, the relative temperature rate of drying is a function of the moisture content. Based on a large amount of experimental data on drying various substances, many researchers have concluded that it can be represented by the equation [14]:

\[ B = - \frac{2T}{\partial U} \left( \frac{U_s - U_p}{T_s} \right) = a \cdot (U - U_p), \]  

(17)

where \( a \) and \( m \) – empirical coefficients; \( U_{\text{sp}} \) – critical moisture content.
It is determined [14] that the coefficient \( m \) is a constant value for this material and does not depend on the drying mode. To fulfill the required condition: \( B = 0 \) with \( U = U_p \), represent dependence of the relative temperature factor of drying with the equation:

\[
B = \frac{2\alpha(U_p - U)}{RT} + \frac{(U_p - U)}{m+1} \left[ \frac{1}{U_p - U} \right].
\]  

(18)

Determine temperature of the material by integrating the equation (18):

\[
T = T_c + (T_c - T) \left[ \frac{1}{m+1} \left( \frac{U_p - U}{U_p - U_p} \right)^{m+1} - 1 \right] + \frac{1}{m+1} \left( \frac{U_p - U}{U_p - U_p} \right).
\]  

(19)

The integration constant is found from the condition: at the equilibrium moisture content of the material, its temperature is equal to the temperature of the drying agent \( T_c \). The integration constant can also be determined from the condition: at critical moisture content, the temperature of the material remains equal to the temperature of the wet thermometer. Under these conditions, we get

\[
T = T + (T_c - T) \left[ \frac{1}{m+1} \left( \frac{U_p - U}{U_p - U_p} \right)^{m+1} - 1 \right] + \frac{1}{m+1} \left( \frac{U_p - U}{U_p - U_p} \right).
\]  

(20)

Then in the second drying period the velocity of the process at any moisture content \( U \) can be determined by the equation:

\[
-\frac{\partial U}{\partial T} = N_1(U) = \left( \frac{U - U_p}{U_p - U_p} \right)^{m+1} \left( U - U_p \right) \exp \left( -\frac{r}{RT} \right)
\]  

(21)

The system of equations (20), (21) is a mathematical model of the kinetics of the second drying period of colloidal capillary – porous bodies. Using the dependence (20) and (21), we can prove the previously determined relation (4). Transforming the exponent of the equation (21):

\[
\exp \left( -\frac{r}{RT} \right) = \exp \left( -\frac{r}{RT} \right) \exp \left( \frac{r}{RT} \frac{T - T_c}{T} \right).
\]  

(22)

Then the equation of the drying velocity of the second period after simple transformations will be in the form of:

\[
N_1(U) = N_1 \left[ \left( \frac{U - U_p}{U_p - U_p} \right)^{m+1} \left( U - U_p \right) \exp \left( -\frac{r}{RT} \right) \right].
\]  

(23)

The exponent of the equation (23) is converted taking into account the equation (20):

\[
\frac{r}{RT} = \frac{T - T_c}{T} = \frac{1}{m+1} \left( \frac{1}{U_p - U_p} \right)^{m+1} - 1 + \frac{1}{m+1} \frac{\alpha_v}{K}.
\]  

(24)

Indicate:

\[
\alpha_v = \frac{U_p - U}{U_p - U_p}; \quad 1 - \alpha_c = \frac{U_p - U_p}{U_p - U_p}; \quad \frac{T_c}{T} - \frac{U_p}{U_p} = K
\]  

(25)

Expression (24)’ we’ll put in as:

\[
\frac{r}{RT} = \frac{T - T_c}{T} = \frac{1}{m+1} \left( \frac{1}{U_p - U_p} \right)^{m+1} - 1 + \frac{1}{m+1} \frac{\alpha_v}{K}.
\]  

(26)

Indicate:
\[ Z(U) = \frac{1}{m} \left[ (1 - \alpha_U)^{n+1} - 1 \right] + \frac{m+1}{m} \alpha_U. \]  

(27)

Then the expression for the drying velocity of the second period has the form:

\[ N_c(U) = N_1 \left( \frac{U - U_{cr}}{U_{cr} - U_i} \right) \cdot \phi(U, T) \cdot \exp \left( \frac{r}{RT} \cdot \frac{Z(U)}{K + Z(U)} \right). \]  

(28)

Equation (28) confirms the previously established model (4), which proves the validity of the considered approach to modeling convective drying of finely dispersed products.

4. Conclusions

Existing mathematical models of convective drying of fine-dispersed products do not predict the actual course of the process with sufficient accuracy, which causes a high cost of development and a decrease in the rate of production development.

Modeling of convective drying of fine products can be based on the laws of chemical kinetics. Applying two laws of chemical kinetics - the law of active masses and the Arrhenius law - as a theoretical basis for modeling convective drying is not enough. A combined approach to model drying is required: the Arrhenius equation describing the velocity of the process should be supplemented by an experimentally established relationship between the temperature and the moisture content of the material. On this basis, the equations for calculating the drying rate in periods of constant and decreasing velocity are obtained.

The validity of the considered approach to modeling convective drying of fine products is confirmed by experimental verification of the kinetic equation obtained for the period of constant velocity, and the relationship between the obtained model for the period of decreasing velocity and experimental models of this period.

5. Theoretical significance of the research and prospects for further scientific researches in this direction

Many mathematical models developed on the existing theoretical basis represent a system of differential equations that require a preliminary solution in engineering calculations. However, there are often significant mathematical difficulties. The approach proposed in this work allows us to obtain computational equations that, after determining all the empirical coefficients, can be directly applied in engineering calculations of drying processes.

The obtained model of drying with constant speed period allows us to establish a clear type of influence of the temperature mode on the kinetics of the process. As a parameter of the temperature mode, the absolute temperature of the drying agent should be taken from the wet thermometer but not from the dry one, as was previously accepted. It is determined that during the period of constant drying velocity, the logarithm of the drying velocity has a linear dependence on the inverse value of the absolute temperature of the product.

The fulfilled research has scientifically proved the possibility of applying the laws of chemical kinetics for the purposes of modeling convective drying. However, for effective practical application of the obtained models, further research is needed related to the development of methods for experimental determination of the values to activate energy of moisture molecules in the drying process and other empirical coefficients of mathematical models.

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