Research and selection of methods for operational control of diffusion coefficient in block products made of capillary-porous materials

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Abstract. For studies of heat and mass transfer in porous media, operational pulse methods are becoming increasingly common. Unlike most known methods, they eliminate the need for destruction of controlled products for preparing special samples of a given configuration. The metrological analysis of the methods providing the possibility of measuring the diffusion coefficient of polar solvents in block products made of porous materials without their destruction is performed. The areas of their preferred use are justified in the context of ensuring acceptable accuracy, in the presence of pronounced anisotropy of properties including.

1. Introduction

The production and operation of products from porous materials (PM) occurs under conditions of significant energy consumption and is associated with the migration of moisture or other substances distributed in the solid phase. Currently, products from PM in the form of blocks and panels are widely used. These products can be attributed to massive bodies in terms of heat and mass transfer processes. Block products from porous materials are regularly used in the energy sector [1], in production and use of promoters, catalysts and nanostructured materials [2, 3], in the field of creating block systems for air purification and regeneration [4], and other prospective lines of production development [5]. For example, in the construction industry, these are bricks, blocks, plates, drywall panels, cement-bonded particleboards, molded heat insulators, etc. Diffusion processes are fundamental in the production and operation of such products. Diffusion of moisture, for example, in porous building products largely determines the indoor microclimate and the effectiveness of heat-shielding properties of enclosing structures [6, 7]. To improve the efficiency of production and operation of block products from porous materials requires knowledge of the diffusion coefficient, which is part of the equations for mathematical models of processes. Technological features for obtaining the class of products under consideration or targeted measures in the manufacture of products in some cases lead to a noticeable anisotropy of PM properties [8]. This leads to the need to use more accurate mathematical models in the development of appropriate methods and the complexity of measurement procedures [9].

At present, the diffusion coefficients in PM block products are measured by methods related to the need to register the time evolution for the spatial distributions of the diffusant in the solid phase of the samples under study, which is quite challenging. In this case, it is necessary either to cut the test

samples into parts and determine the diffusant content by various methods, which introduces distortions in the mass transfer process [10] or to use complex, expensive equipment of significant dimensions and weight [11,12]. In most cases, the product itself is not subject to control. Usually an analogue subjects to such control, which is a research sample specifically made according to the formulation and technology of the product [11] or obtained from the product by mechanical processing. After processing the product completely or partially loses its consumer properties [10]. Therefore, most of the applied methods and measuring devices implementing them are difficult to use directly for operational measurement of the diffusion coefficient in finished products [11]. A significant increase in the performance of measuring equipment for the study of heat and mass transfer processes is achieved by using pulse methods in the presence of flat areas of controlled products with specified sizes [13]. These methods allow one to ensure the safety of products subject to control. They can be attributed to contact non-destructive testing (NDT) methods [14]. Control of block products from PM in terms of measuring the diffusion coefficient can also be performed by similar methods of nondestructive testing.

2. Problem statement

The aim of the study is to perform metrological analysis of nondestructive testing of diffusion coefficient in block products made of porous materials and determine preferences in their use for studies of isotropic and anisotropic porous materials.

To achieve the goal, it is necessary to solve the following tasks:

– to analyze the accuracy of the methods under consideration, to identify the dominants of the resulting measurement error for the desired diffusion coefficient, and to obtain the calculated expressions of their dependence on the influencing factors;

– to assess expediency of using these methods to determine the diffusion coefficient in cases of anisotropy properties presence or absence in block products from porous materials.

3. Theory

When designing pulse methods for non-destructive testing of the diffusion coefficient in block products from porous materials, it is necessary to control the reaction of the object to pulse actions with a dose of a diffusant by changing its concentration in the solid phase at given distances from the point of exposure [15]. Moreover, to ensure acceptable sensitivity of the measuring equipment and the speed of the method, the distances are specified within a few millimeters. Therefore, a high locality of the diffusant concentration control should be ensured, since the error in determining the coordinate δr_0 included in the calculated expressions for the resulting error in determining the diffusion coefficient increases significantly with increasing the thickness of the layer. The concentration change in this layer affects the output characteristic of the sensors used [16]. As such, most of the known electrophysical transducers are difficult to use when implementing NDT methods, since the considered component of the resulting error can reach several tens of percent [16]. The reason is the comparable values of thickness in the informative layers of the material, these layers being adjacent to the sensitive elements of the concentration sensors and distances from the points of pulse action to the sensors. Another reason is a continuously and dynamically changing concentration of the diffusant in these layers [17]. In addition, almost all the measuring equipment used to control the concentration of diffusants distributed in the solid phase requires individual calibration for each new diffusant-porous material system. The exception is for those diffusants employing low-performance weight and chemical methods. The most commonly used methods of layer-by-layer concentration measurement in the study of mass transfer characteristics by cutting products into parts [10] are unacceptable due to the need to ensure non-destructive testing.

To measure the diffusion coefficient of polar solvents in porous material products, a number of non-destructive testing methods have been developed [18-20]. In these methods, a galvanic transducer (GT), that is a generator-type sensor, is used as a diffusant local concentration sensor [17]. When the electrodes of GT are located on the equipotential surfaces of the controlled products, a high locality of

diffusant concentration control is ensured [17]. These methods are based on the use of point [18, 19] or linear [19,20] pulse actions on the flat surface of the products. The methods can be conditionally divided by type of products from PM, for which they may be suitable. These products can be thin-sheet or block ones.

In [18], the analysis of NDT methods for the control of thin-sheet products from porous materials was performed and recommendations were given for their use in order to ensure acceptable accuracy. Let us perform a metrological analysis of the NDT methods, designed to determine the diffusion coefficient in block products from PM.

Method of point pulse action. When implementing the method, a measuring probe [21] is pressed to the flat surface of the test product, this probe having a hole for temporary placement of a diffusant dose point source. GT electrodes are placed on the surface of the measuring probe facing the test product and at a predetermined distance from the hole for applying the pulse action. By supplying a diffusant pulse in the investigated block product from PM, mass transfer is performed corresponding to the diffusion process in an unbounded ball under the action of a point source. The diffusion coefficient is calculated by the following expression [19]:

$$D = r_0^2 / (6\tau_{max}), \tag{1}$$

where r_0 is the coordinate of reaction control to pulse action; τ_{max} is the time to reach the maximum amplitude response to the action.

Using a GT as a sensor of diffusant concentration [17] allows one to determine the value τ_{max} at the time the electromotive force (EMF) of this transducer reaches its maximum value. This ensures high productivity of research, as the need for calibration of the GT for each PM and diffusant is eliminated.

Method of point pulse action. The method is implemented using a measuring probe pressed against the flat surface of the test product [21]. The probe has a slot for temporary placement of a diffusant dose linear source with constant productivity. GT electrodes are placed on the surface of the measuring probe facing the test product and at a predetermined distance from the hole for applying the pulse action. By supplying a diffusant pulse in the investigated block product from PM, mass transfer is performed corresponding to the diffusion process in an unbounded ball under the action of a point source. The diffusion coefficient is calculated by the following expression [19]:

$$D = r_0^2 / (4\tau_{max}), \tag{2}$$

where r_0 is the distance from the pulse line to the electrodes of GT; τ_{max} is the time at which the maximum is achieved on the curve of the diffusant concentration change.

Let us analyze the methods in terms of ensuring minimum measurement errors. In accordance with the concepts of classical metrology, the resulting measurement error of the desired diffusion coefficient when using both methods has the same form, determined by the form of the calculated relations (1) and (2) [19,20]:

$$\delta D = \sqrt{4(\delta r_0)^2 + (\delta \tau_{max})^2 + (\delta_m)^2},$$

where $\delta r_0 = \Delta r_0 / r_0$ is the relative measurement error of the placement coordinate of the GT electrodes [17]; $\delta \tau_{max} = \Delta \tau_{max} / \tau_{max}$ is the relative error of determination τ_{max}; δ_m is the error of method [18,19].

When GT is in use, a quantity $\pm d / 2r_0$ can be applied to estimate δr_0, where d is the diameter of the galvanic transducer electrodes [17]. The nature of the dependence δr_0 is shown in Figure 1c. At the same distances from the sources to the GT electrodes, the value δr_0 can be considered the same for both methods. Let us also assume that the error of method δ_m takes approximately the same values when we use both methods.

Let us perform the analysis $\delta\tau_{\max}$. The studies show that dependence of $\delta\tau_{\max}$ on influencing factors is complex. Figure 1 shows the results of numerical experiments for the method with a linear source for determining the dependence of the maximum concentration values (a), the relative error of their measurement $\delta U_{\max} = \Delta U / U_{\max}$ (b), the components δr_0 and $\delta\tau_{\max}$ (c) of the resulting error δD on the distance r_0, where ΔU is the absolute error of concentration measurement. The calculation results are given for three values of the pulse dose W in the diffusion of ethanol in a cement-fiber plate. The maximum concentration values are presented in the relative form $U_{\max} / U_{\mathrm{p}}$, where U_{p} is the equilibrium concentration of the diffusant with its own saturated vapor in the solid phase of the PM under study at a given temperature. The curves $\delta U_{\max} = \Delta U / U_{\max}$ were obtained under the assumption that ΔU was constant for a relative measurement error of the EMF transducer $\delta E = 0.5\%$ in the rational range of the GT static characteristics [19]. The diameter of the GT electrodes in the calculations δr_0 was taken equal to 0.2 mm.
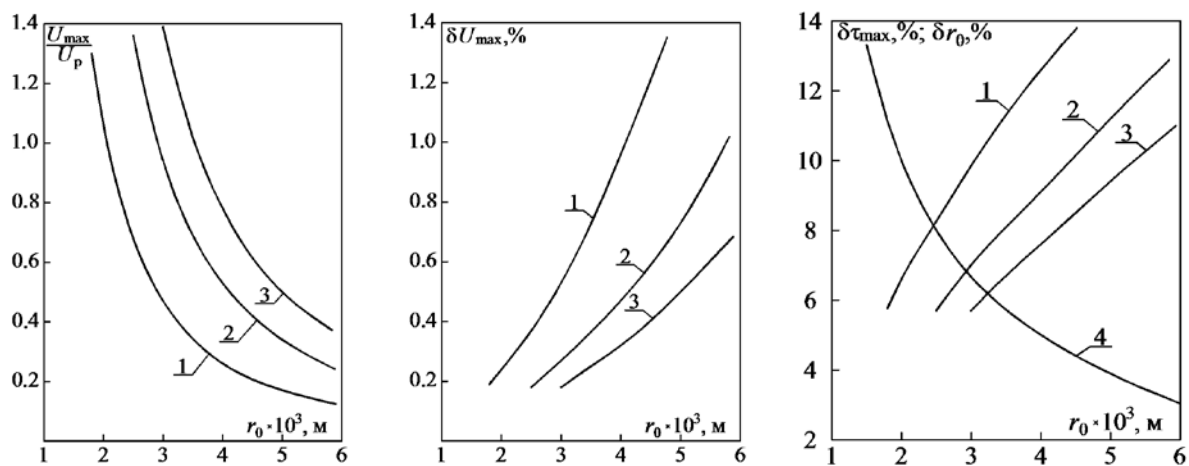
Figure 1. Influence of the distance from the pulse source to the transducer electrodes on the technological and metrological parameters of the method at different source power $W (\times 10^3 \, \mathrm{kg/m})$: 1 – 3.0; 2 – 6.0; 3 – 9.0.

Similar studies were performed for the method with a point source [22]. An analysis of the curves in [22] and in Figure 1 indicates that when the electrodes are located at a distance of $r_0 \geq 4\,\mathrm{mm}$, the dominant of the resulting measurement error of the desired diffusion coefficient when using any of the methods under consideration is $\delta\tau_{\max}$. The remaining components of the resulting error of the two methods, when the same size of the GT electrodes and the distances r_0 from their locations to the sources are used, have approximately the same values [15,19,20,22].

We obtain calculated expressions of the dependence $\delta\tau_{\max}$ on the measurement error δ of the diffusant local concentration for each of the considered methods. Let $U_{\max}$ be the maximum concentration value reached at a time $\tau = \tau_{\max}$ at the GT electrodes locations. Figure 2 illustrates the time variation in concentration in the form of a curve typical for both methods [18]:
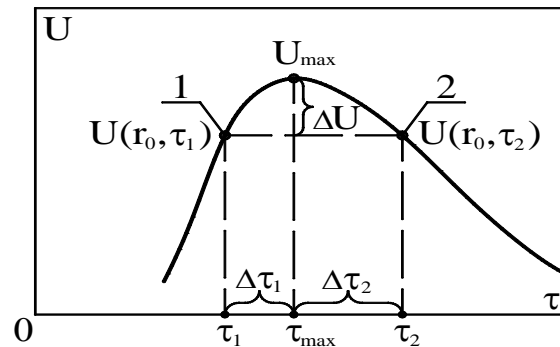
Figure 2. Time variation of the diffusant concentration.

Due to the presence of an error ΔU in measuring the concentration, the maximum concentration can be erroneously recorded at the time instants τ_1 (point 1) or τ_2 (point 2), respectively, before and after the actual value of U_{max} maximum with an error $\Delta\tau_1$ or $\Delta\tau_2$ occurs (Figure 2) [18].

We obtain the calculated expressions for the relative values $I\zeta = \Delta\tau_1 \cdot (\tau_{max})^{-1}$ and $\omega = \Delta\tau_2 \cdot (\tau_{max})^{-1}$ of the errors in determining the point in time τ_{max} in the method that implements a point pulsed action, where, $\Delta\tau_1 = \tau_{max} - \tau_1$ and $\Delta\tau_2 = \tau_2 - \tau_{max}$. Over time, after applying a pulse, the diffusant concentration at a distance r_0 from the source changes as follows [19]:

$$U(r_0,\tau) = \frac{W}{8\rho_0} \cdot (\pi D\tau)^{-\frac{3}{2}} \exp\left(-\frac{r_0^2}{4D\tau}\right), \tag{3}$$

where is the density of the absolutely dry porous material, kg / m^3; W is power of a pulse source, kg.

At a point in time τ_1, the concentration reaches the value:

$$U(r_0,\tau_1) = \frac{W}{8\rho_0} \cdot (\pi D\tau_1)^{-\frac{3}{2}} \exp\left(-\frac{3\tau_{max}}{2\tau_1}\right), \tag{4}$$

On the other hand, it is connected by the relation (Figure 2):

$$U(r_0,\tau_1) = U_{max}(1-\delta). \tag{5}$$

The exact value of the maximum concentration at a time τ_{max} taking the relationship (1) into account is:

$$U(r_0,\tau_{max}) = \frac{W}{8\rho_0} \cdot (\pi D\tau_{max})^{-\frac{3}{2}} \exp(-1,5). \tag{6}$$

Equating the right parts of equations (4), (5) and using (6) we have the expression:

$$1-\delta = \left(\frac{\tau_{max}}{\tau_1}\right)^{\frac{3}{2}} \exp\left(\frac{3}{2}\left(1-\frac{\tau_{max}}{\tau_1}\right)\right),$$

from which we obtain the dependence of the error ζ of the erroneous registration for the maximum concentration at a time τ_1 (Figure 2):

$$\left((1-\zeta)^{-1} \exp\left(\frac{\zeta}{\zeta-1}\right)\right)^{\frac{3}{2}} = 1-\delta, \tag{7}$$

At a point in time τ_2, the concentration reaches the value:

$$U(r_0,\tau_2) = \frac{W}{8\rho_0} \cdot (\pi D\tau_2)^{-3/2} \exp\left(-\frac{3\tau_{max}}{2\tau_2}\right). \tag{8}$$

On the other hand, it is connected by the relation (Figure 2):

$$U(r_0,\tau_2) = U_{max}(1-\delta) \tag{9}$$

Transforming (6), (8), (9) similarly to the above, we obtain the equation:

$$1-\delta = \left(\frac{\tau_{max}}{\tau_2}\right)^{3/2} \exp\left(\frac{3}{2}\left(1-\frac{\tau_{max}}{\tau_2}\right)\right),$$

from which, a dependence on the δ of error ω of erroneous registration for the maximum concentration at a time τ_2 is found (Figure 2):

$$\left((1+\omega)^{-1}\exp\left(\frac{\omega}{1+\omega}\right)\right)^{3/2} = 1-\delta. \tag{10}$$

Let us analyze the method of linear pulse action and compare the results of the accuracy assessment with the analysis results of the previous method. We obtain the calculated expressions for the relative values $y = \Delta\tau_1 \cdot (\tau_{max})^{-1}$ and $z = \Delta\tau_2 \cdot (\tau_{max})^{-1}$ of the error in determining the instant of time τ_{max} in a method of a linear pulse action.

Over time, after applying the pulse, the concentration of the diffusant at a distance r_0 from the source changes as follows [20]:

$$U(r_0,\tau) = \frac{Q}{4\rho_0} \cdot (\pi D\tau)^{-1} \exp\left(-\frac{r_0^2}{4D\tau}\right) \tag{11}$$

where Q is the power of the "linear" source r=0r=0, calculated as the ratio of the diffusant amount to the length of the pulse action line L, kg / m.

Thus, in terms of the mathematical description the mass transfer in a block product under the linear pulse action proves to be similar to the mass transfer in a thin-sheet product when applying a point pulse action [18]. Therefore, as equations for the relative values $y = \Delta\tau_1 \cdot (\tau_{max})^{-1}$ and $z = \Delta\tau_2 \cdot (\tau_{max})^{-1}$ of the errors in determining the point in time τ_{max} we can use the relations obtained earlier for the method of thin products control that implements a point pulse action [18]:

$$(1-y)^{-1}\exp\left(\frac{y}{y-1}\right) = 1-\delta. \tag{12}$$

$$(1+z)^{-1}\exp\left(\frac{z}{1+z}\right) = 1-\delta. \tag{13}$$

We note that the linear pulse action method allows one to study the diffusion coefficient in products not only from isotropic PMs, but also from anisotropic ones [20].

4. Experimental results

The dependence of the relative error of the quantity τ_{max} included in equation (1) for the method of the point pulse action is shown in Figure3 in the form of two curves $\zeta(\delta)$ and $\omega(\delta)$.
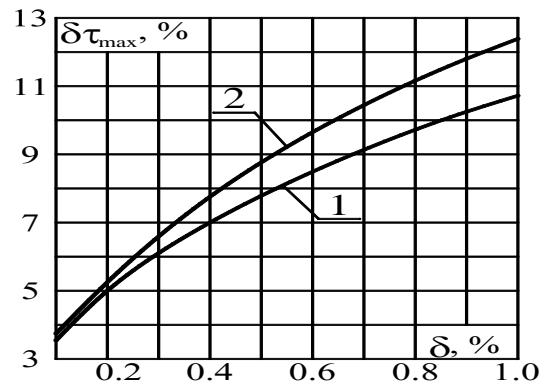
Figure 3. Curves ζ (1), ω (2) for the method with the point pulse action.

They are calculated according to equations (7) and (10) for an erroneously recorded value U_{max} at time instants τ_1 and τ_2, respectively. To construct the resulting curve $\delta\tau_{max}(\delta)$, the average values from $\zeta(\delta)$ and $\omega(\delta)$ can be used. The dependences $y(\delta)$ and $z(\delta)$ calculated according to equations (12) and (13) for the method of a linear pulse action are presented in [18]. For each method, the dependences (7), (10), (12), and (13) allows us to calculate the values $\delta\tau_{max}$ using specific values of equipment inaccuracy for measuring the local concentration of diffusants.

5. Results discussion

The metrological analysis of the presented methods indicates that the change of the relative error in determining the time τ_{max} to reach the maximum concentration U_{max}, depending on the relative error δ in measuring the local concentration of the diffusant for each of the methods considered, occurs in accordance with various laws. In addition, when using each of the considered methods, these dependences are described by different types of equations for negative and positive deviations from the real value τ_{max}. These are equations (7), (10) and (12), (13), respectively. In constructing the generalized dependences of the error $\delta\tau_{max}$ on δ for each of the methods, we propose to use the average values calculated according to equations (7), (10) and (12), (13), assuming that negative and positive deviations from the actual value τ_{max} are equally probable. Analysis of Figure 3 and [18] showed that, other factors being equal, the method of point action on a block product made of porous materials has higher accuracy. For example, with a relative measurement error of the local concentration $\delta = 0.5\%$, the calculated error $\delta\tau_{max}$ of time point measurement is ~ 8.3% and ~ 10.1% for the methods of point and linear effects, respectively, the EMF maximum of the galvanic transducer being observed at this time point (Figure 3 and 2*a* [18] show the average values of the curves).

However, if there is a noticeable anisotropy in the properties of the porous materials used in the products, the point action method is less preferable. Its application is complicated by a significant methodological error δ_m caused by different diffusant propagation speed in different directions of the material, for example, lengthwise and crosswise of fiber arrangement [20]. In this case, it is recommended to use the method with a linear source [20].

Experimental studies of moisture and ethanol diffusion in a number of PM block products confirmed the comparable performance of the presented methods and the reproducibility of the measurement results. The duration of the experiment did not exceed 70 minutes. In the absence of reference materials for measuring the diffusion coefficient, the random component of the error was estimated. When processing the experimental data of measuring the diffusion coefficient, the following random error estimation results were obtained: ~7-8 % for the point source method and ~ 9-11 % for the linear

source method for the control of block products made of isotropic and anisotropic materials, respectively.

6. Conclusion

The metrological characteristics of the non-destructive testing methods of the diffusion coefficient in block products from porous materials are compared, these methods being implemented in measuring devices of various types of mass transfer in conditionally unlimited bodies: a ball and cylinder.

As a result of the analysis, recommendations are formulated on the use of methods for various research objects in terms of ensuring the highest accuracy:

– for block products from isotropic porous materials, a method with a point pulse source of a diffusant dose is recommended;

– for block products made of anisotropic porous materials, a method with a linear pulse source of the diffusant dose is recommended.

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