Non-destructive testing method for determining the solvent diffusion coefficient in the porous materials products

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Non-destructive testing method for determining the solvent diffusion coefficient in the porous materials products

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Abstract. Ensuring non-destructive testing of products in industry is an urgent task. Most of the modern methods for determining the diffusion coefficient in porous materials have been developed for bodies of a given configuration and size. This leads to the need for finished products destruction to make experimental samples from them. The purpose of this study is the development of a dynamic method that allows operatively determine the diffusion coefficient in finished products from porous materials without destroying them. The method is designed to investigate the solvents diffusion coefficient in building constructions from materials having a porous structure: brick, concrete and aerated concrete, gypsum, cement, gypsum or silicate solutions, gas silicate blocks, heat insulators, etc. A mathematical model of the method is constructed. The influence of the design and measuring device operating parameters on the method accuracy is studied. The application results of the developed method for structural porous products are presented.

1. Introduction
Most studies of the diffusion in the products made from porous building materials is focused on the moisture diffusion coefficient determination. In the applied measurement devices unidirectional isothermal mass transfer is usually used in specially prepared samples for the stationary or non-stationary measurement processes [1 - 3]. The methods are characterized by substantial costs and time for experiment preparation. Applied instruments are also cumbersome and have significant cost. For the determination of spatial moisture distributions inside the specimens, made from building materials, traditional weight methods with the cutting of the test specimen into pieces are often used. Now there are modern methods of nondestructive testing: nuclear magnetic resonance [4], game scope [2], neutron radiography, computed tomography and x-rays [3]. However, the methods are not able to carry out non-destructive testing of finished products as it is necessary to have samples of a given configuration, and the measurement process is characterized by a significant duration, which does not allow to obtain a rapid information of the products quality. A significant increase in efficiency control may be obtained through the use of dynamic and pulse methods. However, the known pulse techniques (methods instantaneous source mass) are also associated with the need to manufacture samples [5].

2. Formulation of the problem
The aim is to develop a new impulse method for measuring the diffusion coefficients of solvents distributed in the solid phase of porous materials (PM). The method should ensure the possibility to investigate finished products without destroying them and significantly reduce the time spent on monitoring. To achieve the goal it is necessary to solve the following tasks:
- develop a physical and mathematical model of the method that provides a fast flowing mass transfer process in the controlled product and get without the product destruction the necessary measurement;
- to develop and investigate a mathematical model of errors in measuring the diffusion coefficient;
- to investigate the influence of the measuring device design parameters and the regime variables for the accuracy measurement process of determining the diffusion coefficient, give recommendations on rational parameters of measuring devices;
- to test experimentally the working capacity of the developed method on products for construction purposes.

3. Theory

The research results are based on the analytical theory of heat and mass transfer, mathematical physics, mathematical modeling, the classical metrology theory and mathematical statistics. The main idea of the developed method is to use the impulse action on a PM product in the form of a solvent dose. Then, the response to this effect is analyzed as a the solvent concentration change in the PM solid phase. From the point of the mathematical mass transfer process description, the sample is viewed as an unbounded body of the canonical form - the ball. For this purpose, the checked sample must have a flat surface of specified dimensions. This allows one-sided access to the product and replacement the measuring device on this part of the surface. In such case non-destructive control of the diffusion coefficient of solvents is possible. Figure 1 represents the physical model of the method.

![Physical model of the method](image)

When implementing the method, product is used with a uniform initial distribution of the solvent. At the beginning of the experiment, a pulse solvent dose $Q$ is applied within a «spot» of diameter $s$ on the flat sample surface. After the pulse is applied, the flat product surface is waterproofed. With the help of electrodes, located on a concentric circle with radius $r_0$ in respect to the point of impulse action on the sample, the electromotive force (EMF) change $E(r_0, \tau)$ of the electrochemical converter (ECC) is measured in time[6]. The process of spreading a solvent in a solid PM product is described by the following boundary-value mass transfer problem in an unbounded medium:

$$
\frac{\partial C(r, \tau)}{\partial \tau} = \frac{D}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C(r, \tau)}{\partial r} \right) + \frac{Q}{\rho_0} \delta(r, \tau),
$$

$$
\tau > 0, \; 0 \leq r < \infty, \; C(\tau,0) = C_0; \; \frac{\partial C(0, \tau)}{\partial r} = 0; \; C(\infty, \tau) = C_0,
$$

where $C(r_0, \tau)$ - the solvent concentration on the sphere surface of radius $r_0$ with respect to the solvent dose impulse delivery point at time $\tau$; $D$ - the solvent diffusion coefficient; $\delta(r, \tau)$ - Dirac delta function; $\rho_0$ - the density of the absolutely dry test material; $Q$ - the amount of liquid phase supplied from the dispenser to the flat sample surface; $C_0$ - initial concentration of solvent in the material at the time $\tau = 0$.

The diffusion coefficient can be found from the formula [7]:

$$
D = \frac{Q}{\rho_0} \int_0^\infty r^2 \frac{\partial C(r, \tau)}{\partial \tau} \, dr.
$$
where $\tau_{\text{max}}$ is the time corresponding to the maximum on the $C(r_0, \tau)$ curve of the concentration change at a distance $r_0$ from the source.

Due to the monotonicity of static ECC characteristic [8], the moments of reaching the maximum concentration $C(r_0, \tau)$ and the converter maximum EMF $E(r_0, \tau)$ coincide, it is possible to realize the measurement procedure without preliminary calibration of the ECC. The experience with the application of electrochemical converters [8, 9] shows that it is expedient to use the range of the static characteristic near the value $0.5C_e$, characterized by high sensitivity and stable signal. Here $C_e$ is the diffusing substance concentration in the PM solid phase at equilibrium with saturated solvent vapors at a given temperature. Therefore, in carrying out the experiment it is expedient to obtain the value $C_{\text{max}}$ in the vicinity of the given value:

$$C_{\text{max}} \approx 0.5C_e$$  \hspace{1cm} (2)

The values of the ECC location coordinate $r_0$ and the solvent dose $Q$ supplied at the beginning of the experiment active part exert a significant influence on the $C_{\text{max}}$ value. In this case, the solvent concentration change in the zone of source action is described by the function:

$$C(r, \tau) - C_0 = Q \left[ 8 \rho_0 \left( \frac{\pi D \tau}{r_0} \right)^{3/2} \exp \left( \frac{r_0^2}{4D\tau} \right) \right].$$  \hspace{1cm} (3)

From (3), taking (1) into account, assuming for simplicity $C_0 = 0$, we can obtain the dependence of the attained maximum $C_{\text{max}}$ at $\tau = \tau_{\text{max}}$ from the solvent dose $Q$:

$$C_{\text{max}}(r_0, \tau_{\text{max}}) = Q \left[ 8 \rho_0 \left( \frac{\pi}{6} \right)^{3/2} \left( \frac{1}{\sqrt{e}} \right) \frac{r_0^3}{r_0^3} \right].$$  \hspace{1cm} (4)

We will carry out a study of the proposed method as the design measuring device parameters and the technological conditions of the measurement process affect on the diffusion coefficient accuracy. The main design parameters are: the minimum permissible flat section dimensions and the product thickness in the area of action of the pulsed source; allowable «spot» size $s$ of substance dosing; the coordinate $r_0$ of the ECC electrodes location relative to the location of the pulse application. The main technological variables are the parameters of the applied mass pulse: the duration $t$ and the power $Q$.

The basis for finding the design measuring device parameters is the determination of coordinate $r_0$ for monitoring the EMF change after pulse application.

The mathematical model of the relative error root-mean-square estimate in determining the sought diffusion coefficient has the form [8, 10]:

$$\delta D = \sqrt{4(\delta r_0)^2 + (\delta \tau_{\text{max}})^2 + (\delta h)^2},$$  \hspace{1cm} (5)

where $\delta r_0 = \Delta r_0 / r_0$ and $\delta \tau_{\text{max}} = \Delta \tau_{\text{max}} / \tau_{\text{max}}$ are, respectively, the relative error in determining the coordinate of the calculated cross-section and the time to reach the maximum on the curve of the concentration change in the calculated section; $\delta h$ is the total value of the methodical error due to the incomplete correspondence of the mathematical model used to real physical processes.

It was shown in [8] that when ECC electrodes are located on the equipotential samples surfaces with respect to a pulsed source of solvent dose, a value $\pm a/r_0$ can be used as an estimate $\delta r_0$, where $a$ is the ECC electrodes radius. The value $\delta \tau_{\text{max}}$ essentially depends on the nature of the change in the concentration $C(r_0, \tau)$ and the error in its determination. In figure 2, 3 it is shown as an example, the calculated dependences in the relative $C_{\text{max}}/C_e$ concentrations change and the error $\delta C_{\text{max}} = \Delta C/C_{\text{max}}$ on $r_0$ obtained in the study of the diffusion coefficient of ethanol in a dry state gas silicate plate (530 kg/m$^3$) for different dose values of the solvent pulse $Q$. The dependences of figure 3 are obtained on the basis of (4), assuming the AC constancy with the relative error $\delta E$ (2) of measuring the EMF ($\delta E = 0.5\%$ in the chosen rational ethanol concentration range). Studies show that for a given material, for example, with an impulse of $8.0 \times 10^4$ kg, the measuring device must be equipped with a sensor located at a distance $r_0 \leq 2.5$ mm.
Figure 2. The influence of the sensor location on the maximum concentrations reached at different source powers $Q$ (kg):
1 - $8.0 \times 10^{-6}$; 2 - $3.5 \times 10^{-5}$; 3 - $7.0 \times 10^{-5}$.

Figure 3. The influence of the sensor location on the error in their measurements at different source powers $Q$ (kg):
1 - $8.0 \times 10^{-6}$; 2 - $3.5 \times 10^{-5}$; 3 - $7.0 \times 10^{-5}$.

At $r_0 > 3$ mm, the maxima on the change curves $C(r_0,\tau)$ and, respectively $E(r_0,\tau)$, will be inadequate. This will lead to a significant increase in the relative error $\delta C_{\text{max}}$ and $\delta E_{\text{max}}$, respectively, the concentration and EMF of the ECC and, consequently, the relative error in determining the time to reach the maximum on the curve of the concentration change in the calculated section. At a pulse of $7.0 \times 10^{-5}$ kg, the sensors information with coordinates $r_0<4$ mm does not allow to calculate the diffusion coefficient at all. The value of $C_{\text{max}}$ exceeds $C_s$ and the ECC static characteristic is on the saturation plateau [8].

Figure 4 presents the results of numerical experiments when we determine the dependence $\delta r_{\text{max}}$ of the ethanol diffusion coefficient in a gas silicate plate on $r_0$ obtained in the study for the same values of solvent doses. In the same place, the error curve is shown for the ECC electrode diameters to 0.2 mm.

Figure 4. The influence of the sensor position on the error $\delta r_0$ (4) and $\delta r_{\text{max}}$ at different source powers $Q$ (kg):
1 - $8.0 \times 10^{-6}$; 2 - $3.5 \times 10^{-5}$; 3 - $7.0 \times 10^{-5}$. 
The analysis of the curves (figures 2, 4) shows that if the condition (2) is satisfied when setting the experiment with different source power, then the error $\delta \tau_{\text{max}}$ has approximately the same values. At the same time, on the graphs of the dependence $\delta \tau_{\text{max}}$ on $r_0$ (figure 4), it is necessary to take the sections, corresponding to the range of the best ECC sensitivity near the value $0.5 C_e$ (figure 2). Therefore, the distance choice $r_0$, according (5) and $\delta_m = \text{const}$, is achieved by a compromise solution based on the error $\delta \tau_{\text{1}}$ and duration of the experiment. As the analysis result (figure 4), it was decided to place the main electrodes of the electrochemical converter at a distance $r_0 = 4$ mm from the point of impulse action and additional electrodes at a distance of 3 and 5 mm. Removing the converter from the source more than 5 mm causes an increase in the duration of the experiment, in the dimensions of the products flat sections and in the overall dimensions of the measuring device with a slight increase in the control accuracy. Using (4), and taking into account the calculated constants, it is possible to calculate in advance the required impulse action solvent dose $Q$ which provides the fulfillment of condition (2):

$$Q \approx 3.41 \rho_0 C_e r_0^3.$$  

(6)

The rationale for using the remaining design parameters and regime variables of the diffusion coefficient measurement process was obtained by analyzing the methodical error $\delta_m$. They are chosen from the condition of reaching negligibly small values $\delta_m$ in comparison with the remaining resulting error $\delta D$ components according to [8, 11]. The minimum dimensions $d_e$ of the flat section diameter for controlled massive PM product and a measuring probe are obtained from the condition: $d_e > 20 r_0$.

The product thickness $h$ in the area of action of the source must be at least 10 $r_0$. Dimensions of the "spot" diameter s, within which the solvent dose is applied to the test material surface, should not exceed $0.05 \times r_0$. The pulse duration $t$ of the mass must not exceed $0.05 \times \tau_{\text{max}}$. Based on the maximum location value (5 mm) of the ECC electrodes we determined the minimum permissible diameter of the products flat piece (100 mm) on which the measuring device is placed. The minimum sizes of controlled massive products in the area of the point source action are also determined. The dimensions of the products must exceed the hemisphere dimensions of a radius 50 mm centered at the point of pulse application. This allowed us to take the upper estimate of 5% for the total methodical error $\delta_m$. Taking this into account, the root-mean-square estimate of the relative error $\delta D$ in determining the unknown diffusion coefficient according to (5) and taking into account the data in figure 2 for distances 3, 4 and 5 mm up to a point source, respectively, is 12; 11 and 10% (for $\delta E = 0.5\%$ and the condition for using the optimal doses, calculated by equation (6) for each $r_0$).

4. Experimental results

To carry out experimental studies, we used the information-measuring system developed by the "National Instruments" technology [12] with a modernized measuring device [8]. Before carrying out the active experiment, the required value of the pulse mass was preliminarily determined by formula (6). In figure 5, as an example, the results of measuring the diffusion coefficient of ethanol in gas-silicate dry slabs 50 mm thick with a $530 \text{ kg} / \text{m}^3$ density are presented which obtained using the proposed method. The total number of studies i for each PM-solute system was at least 20 runs, the ethanol pulse value for this example was $3.2 \times 10^{-5} \text{ kg}$, $r_0 = 4$ mm. Experimental studies have shown that the mathematical expectation of the ethanol diffusion coefficient in the tested gas-silicate plate with a confidence probability $\alpha = 0.95$ is $5.4 \times 10^{-9} \text{ m}^2/\text{s}$. The random error of the diffusion coefficient determining is less than 9% the duration of the experiment does not exceed 12 minutes.

5. Discussion of the results

This method allows us to measure quickly the diffusion coefficient of polar solvents in porous materials products without destroying them. Products must have at least one flat surface with a diameter more than 100 mm. A significant increase in control performance is ensured by:

- excluding the need of making samples for research;
- using more dynamic mass transfer process under impulse action;
- elimination the need for preliminary calibration of the electrochemical converter used for each
new material and diffusing solvent (tens of hours).

The efficiency of this method was checked by comparing the obtained data with the results of
determining the diffusion coefficients of moisture and ethanol in samples from massive porous
materials by the zonal method [13] and the sorption method [14]. The discrepancy in the numerical
values of the coefficients did not exceed 8 ... 10%.

6. Conclusion

The dynamic method has been developed that makes it possible to significantly improve the
efficiency of controlling the polar solvents diffusion coefficients in finished articles from porous
materials without destroying them. The method can be used for the operational diffusion coefficient
control in portable devices for finished construction products made from porous materials. As a
studies result, the minimum permissible dimensions of the product flat surface on which a measuring
portable device can be placed are determined. The rational value of the electrochemical converter
electrodes location coordinate relative to the point of impulse action application is determined from
the permissible accuracy and duration of the measurement process. The equation is obtained for
calculating the required solvent dose for impulse action. The statistical processing the obtained
experimental data and its comparison with the known results indicate the operability of the proposed
nondestructive testing method.

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