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DETERMINATION OF COEFFICIENT OF THERMAL EXPANSION FOR BINARY COMPOSITE MATERIALS

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UDC 539.32

The coefficient of thermal expansion is determined for two-phase materials on the basis of the percolation model.

1. Structural Model of Nonhomogeneous Material. The analysis of heat conduction and electrical conduction in binary heterogeneous materials with random distribution of equipollent components is usually based on the percolation theory (theory of flow-through) [1, 2]. One of the main applications of this theory is determining the topology of an infinite cluster or, in other words, the distribution of components in such material as their volume concentration changes. It has been established [1, 2] that as the volume concentration v_1 of the first component in a continuous binder changes over the $0 \leqslant v_1 \leqslant v_c$ range (v_c is the percolation threshold), there appear isolated inclusions (insular clusters) of the first component and with $v_c \leq v_1$ bonds develop between them which transforms these insular clusters into infinite ones, this jumpwise transition occurring at the concentration $v_1 = v_c$ and being followed by formation of two equipollent infinite clusters when $v_1 \simeq v_2 \simeq 0.5$ in the material. A further increase of the volume concentration of the first component $v_1 > 0.5$ results in a structural reversal, namely an infinite cluster of the second component will decrease and at the concentration $v_2 = v_c$ be jumpwise transformed into an insular one while an infinite cluster of the first component continues growing.

Using the concepts of the percolation theory and the methods of reduction to an elementary cell, a structural model of a heterogeneous material has been proposed [3, 4], an elementary cell of which is shown here in Fig. 1. The geometrical parameters of such an elementary cell are given in Table 1. On the basis of this model theoretical relations for the effective thermal and electrical conductivities which agree closely with experimental data [3, 4] have then been derived.

Now, using this percolation model, we will determine the coefficient of thermal expansion for a heterogeneous material.

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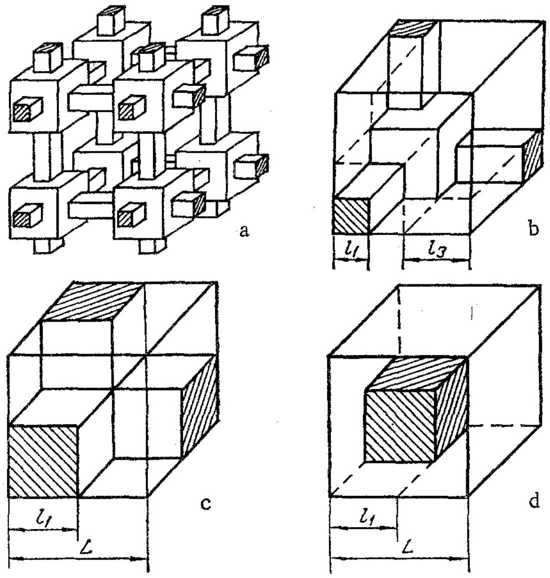

Fig. 1. Percolation model of heterogeneous material: (a) model; (b) elementary cell; (c) $v_1 = v_2 = 0.5$; (d) $v_1 < v_C$.

2. Coefficient of Thermal Expansion. Both upper and lower bounds for the effective coefficient of thermal expansion α_{ll} of heterogeneous materials, based on the principle of minimum potential energy after deformation of a body, have been established [5, 6] by the integral method of sections

$$\alpha'_{ll} \leqslant \alpha_{ll} \leqslant \alpha''_{ll}, \tag{1}$$

where

$$\alpha'_{ll} = \{\alpha^{(2)}_{ll} + \overline{S}_1(x_k)(\alpha^{(1)}_{kk} - \alpha^{(2)}_{kk})\, B^{(1)}_{llkk}(x_k)\}_L, \tag{2}$$

$$\alpha''_{ll} = \{N_{kl}(x_i,\ x_j)\, M_{klmn}(x_i,\ x_j)\}_S\, S_{klnm}, \tag{3}$$

and S_{klmn} is the compliance tensor. The tensors

$$B_{llkk}(x_k),\ N_{kl}(x_i,\ x_j),\ M_{klnm}(x_i,\ x_j) \tag{4}$$

are determined from the equalities

$$\{\sigma^{(i)}_{ij}(\mathbf{r})\}_{S_i} = B^{(i)}_{ijkl}(x_k)\,\{\sigma_{kl}(\mathbf{r})\}_S, \qquad \{\sigma_{ij}(\mathbf{r})\}_{L_i} = B^{(i)}_{ijkl}(x_i,\ x_j)\,\{\sigma_{kl}(\mathbf{r})\}_L,$$
$$\{\sigma_{kl}(\mathbf{r})\}_L = M_{klnm}(x_i,\ x_j)\,\{\varepsilon_{nm}(\mathbf{r})\}_L, \qquad \{\sigma_{ij}(\mathbf{r})\,\alpha_{ij}(\mathbf{r})\}_L = N_{kl}(x_i,\ x_j)\,\{\sigma_{kl}(\mathbf{r})\}_L, \tag{5}$$

which yield

$$M_{klnm}(x_i,\ x_j) = \overline{L}_1(x_i,\ x_j)\, C^{(1)}_{klij} B^{(1)}_{ijnm}(x_i,\ x_j) + \overline{L}_2(x_i,\ x_j)\, C^{(2)}_{klij} B^{(2)}_{ijmn}(x_i x_j), \tag{6}$$
$$N_{kl}(x_i,\ x_j) = \overline{L}_1(x_i,\ x_j)\, \alpha^{(1)}_{ij} B^{(1)}_{ijkl}(x_i,\ x_j) + \overline{L}_2(x_i,\ x_j)\, \alpha^{(2)}_{ij} B^{(2)}_{ijmn}(x_i,\ x_j).$$

Here the brackets {...} and <...> denote averaging with respect to variables x_1, x_2, x_3:

$$\{\ldots\}_L = \frac{1}{L}\int\limits_0^L (\ldots)\, dx_k;\ \{\ldots\}_S = \frac{1}{S}\iint\limits_{(S)} (\ldots)\, dx_i dx_j, \tag{7}$$
$$\langle \ldots \rangle = \{\{\ldots\}_L\}_S = \{\{\ldots\}_S\}_L = \frac{1}{V}\iiint\limits_V (\ldots)\, dx_i dx_j dx_k.$$

Here the lower bound α'_{ll} has been determined on the assumption that

$$\frac{\partial}{\partial x_k}\{\sigma_{kl}(\mathbf{r})\}_S = 0, \tag{8}$$

and the upper bound α''_{ll} has been determined on the assumption that

$$\frac{\partial}{\partial x_l}\{\varepsilon_{nm}(\mathbf{r})\}_L = 0. \tag{9}$$

The superscripts in the tensor notation and the subscripts in the scalar notation indicate to which component of the material a given quantity refers.

TABLE 1. Geometrical Parameters of Percolation Model*

Range of v_1 variation	$\overline{S}_1^{(M)}$	$\overline{S}_2$	$\overline{S}_3$	$\overline{S}_4$	$\overline{l}_1$	$\overline{l}_2$
$0 \leqslant v_1 \leqslant v_c$	0	$v_1^{2/3}$	0	$1-v_1^{2/3}$	0	$v_1^{1/3}$
$v_c < v_1 \leqslant 0,5$	$\frac{1}{3}\left(\frac{v_1-v_c}{1-v_c^{1/3}}\right)$	$v_c^{2/3}-\overline{S}_1$	$2(S_1^{(M)})^{1/2} \times (1-v_c^{2/3})$	$1-v_c^{2/3}-\overline{S}_3$	$(S_1^{(M)})^{1/2}$	$v_c^{1/3}$
$0,5 < v_1 \leqslant 1-v_c$	$1-v_c^{2/3}-\overline{S}_3$	$v_c^{2/3}-\overline{S}_4$	$2S_4^{1/2} \times (1-v_c^{2/3})$	$\frac{1}{3}\left(\frac{v_2-v_c}{1-v_c^{1/3}}\right)$	$1-\overline{S}_4^{1/2}$	$1-v_c^{1/3}$
$1-v_c < v_1 \leqslant 1$	$1-v_2^{2/3}$	$v_2^{2/3}$	0	0	0	$1-v_2^{1/3}$

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*$\overline{S}_1 = \overline{S} + (\overline{S}_1^{(M)} - \overline{S})g(z)$; $\overline{S} = \left(\frac{v_1-v_c}{1-v_c}\right)^{1,6}$; $g(z) = 5.53z - 8.3z^2 + 3.23z^3 + 0.54z^4$; $z = K_2/K_1$, $K_2 < K_1$.

When the components are homogeneous and isotropic, then expressions (2) and (3) yield

$$\alpha' = \left\{\alpha_1\overline{S}_1(x_k) + \alpha_2\overline{S}_2(x_k) + 2\frac{\overline{S}_1(x_k)\overline{S}_2(x_k)(\alpha_2-\alpha_1)(a_1-a_2)}{a_1\overline{S}_1(x_k)+a_2\overline{S}_2(x_k)}\right\}_L, \tag{10}$$

$$\alpha'' = \{\delta(x_i, x_j)\,\eta(x_i, x_j)\}_L E^{-1}, \tag{11}$$

$$\delta(x_i, x_j) = \alpha_1\overline{L}_1(x_i, x_j) + \alpha_2\overline{L}_2(x_i, x_j) + 2\frac{\overline{L}_1(x_i, x_j)\overline{L}_2(x_i, x_j)(\alpha_1-\alpha_2)(c_1-c_2)a_1a_2}{a_1\overline{L}_1(x_i, x_j)+a_2\overline{L}_2(x_i, x_j)}, \tag{12}$$

$$\eta(x_i, x_j) = \left[\frac{\overline{L}_1(x_i, x_j)}{E_1} + \frac{\overline{L}_2(x_i, x_j)}{E_2} - 2\frac{(c_1-c_2)^2\overline{L}_1(x_i, x_j)\overline{L}_2(x_i, x_j)a_1a_2}{a_1\overline{L}_1(x_i, x_j)+a_2\overline{L}_2(x_i, x_j)}\right]^{-1}, \tag{13}$$

$$a_i = \frac{18K_i\mu_i}{3K_i+4\mu_i},\quad c_i = \frac{3K_i-2\mu_i}{18K_i\mu_i};\quad E = \frac{9K\mu}{3K+\mu},$$

where $L_i(x_i, x_j)$ is the length of a segment parallel to the Ox_k axis in the i-th component, $S_i(x_k)$ is the area of the intersection of a representative volume V by a plane perpendicular to the Ox_k axis and occupied by the i-th component (i = 1, 2), $\overline{L}_i(x_i, x_j) = L_i(x_i, x_j)/(L_1(x_i, x_j)+L_2(x_i, x_j))$, $\overline{S}_i(x_k) = S_i(x_i, x_j)/(S_1(x_k) + S_2(x_k))$.

In subsequent calculations, besides geometrically modeling the structure of a heterogeneous medium, we will also use the method of step-by-step quasihomogenization [5, 6]. The gist of this method is to do the following: first extracting a representative volume V of the heterogeneous material, then partitioning this volume V into regions readily integratable and determining their effective properties, and then, with each region regarded as a quasihomogeneous one, determining the effective properties of the entire representative volume V.

Since determining the coefficient of thermal expansion for a subregion of volume V involves the elastic properties, which must be known, we will use here the following relations for the bulk modulus K and the shear modulus μ of any subregion

$$K' = \left\{\left(\left\{\frac{n}{K}\right\}_L - 2\frac{\{d\}_L\{P\}_L}{\{KP\}_L}\right)^{-1}\right\}_S, \tag{14}$$

$$\mu' = \left\{\left(\left\{\frac{1}{\mu}\right\}_L\right)^{-1}\right\}_S, \tag{15}$$

$$K'' = \left[\left\{\left(\frac{\{P\}_S}{\{KP\}_S}\right)\right\}_L\right]^{-1}, \tag{16}$$

$$\mu'' = \left[\left\{\left(\frac{1}{\{\mu\}_S}\right)\right\}_L\right]^{-1}, \tag{17}$$

where $P = 6m/(3 + 4m)$, $d = (3 - 2m)/(3 + 4m)$, $n = 9/(3 + 4m)$, $m = \mu/K$, with K', μ' obtained on the basis of assumption (8) and K'', μ'' obtained on the basis of assumption (9).

The braces $\{\ldots\}_L$ and $\{\ldots\}_S$ in expressions (14)-(17) are, according to definitions (7), put in the form

$$\{f\}_L = f_1\overline{L}_1(x_i, x_j) + f_2\overline{L}_2(x_i, x_j),\quad \{f\}_S = f_1\overline{S}_1(x_k) + f_2\overline{S}_2(x_k).$$

TABLE 2. Moduli of Elasticity of WC–Co Alloy*

Volume concn., %	Young's modulus, (kgf/mm^2)·10^{-4}		Shear modulus, (kgf/mm^2)·10^{-4}		Bulk modulus, (kgf/mm^2)·10^{-4}	
	calc.	expt.	calc.	expt.	calc.	expt.
5,3	6,76	6,73	2,83	2,81	3,696	3,75
10,0	6,4	6,419	2,672	2,665	3,548	3,62
16,4	5,90	5,92	2,53	2,447	3,41	3,5
20,8	5,67	5,64	—	—	—	—
25,0	5,34	5,327	2,178	2,184	3,247	3,17
30,5	5,04	5,013	2,044	2,026	3,170	3,14; 3,18
45,0	4,28	4,27	—	—	—	—

*Experimental data taken from [8].

TABLE 3. Young's Modulus of W–S_iO_4 Composite

Volume concn. of tungsten, %	Young's modulus, (kgf/mm^2)·10^{-4}		
	calc. according to relations (25)-(34)	calc. according to relations in [10, 11]	expt. [9]
0	0,805	0,805	0,805
10	0,895	0,920	0,909
20	1,040	1,060	1,055
30	1,226	1,210	1,180
40	1,489	1,441	1,375
50	1,728	1,692	1,599
100	3,550	3,550	3,550

For determining the effective properties we will use the combined method with sections of an elementary cell [7], which yields results close to

$$\alpha = \frac{1}{2}(\alpha' + \alpha''), \quad K = \frac{1}{2}(K' + K''), \quad \mu = \frac{1}{2}(\mu' + \mu''). \tag{18}$$

Upon partitioning an elementary cell into subregions as shown in Fig. 1b, we define the coefficient of thermal expansion as

$$\alpha = \frac{\delta_4 b_4 (\bar{S}_1 + \bar{S}_4) + \delta_3 b_3 (\bar{S}_2 + \bar{S}_3)}{b_4 (\bar{S}_1 + \bar{S}_4) + b_3 (\bar{S}_2 + \bar{S}_3)}, \tag{19}$$

where (20)

$$\delta_1 = \alpha_1 \bar{l}_2 + \alpha_2 (1 - \bar{l}_2) + 2 \frac{(c_1 - c_2)(\alpha_1 - \alpha_2) \bar{l}_2 (1 - \bar{l}_2) a_1 a_2}{a_1 \bar{l}_2 + a_2 (1 - \bar{l}_2)};$$

$$\delta_2 = \alpha_1 \bar{l}_1 + \alpha_2 (1 - l_1) + \frac{(c_1 - c_2)(\alpha_1 - \alpha_2) \bar{l}_1 (1 - \bar{l}_1) a_1 a_2}{a_1 \bar{l}_1 + a_2 (1 - \bar{l}_1)}; \tag{21}$$

$$\delta_3 = \frac{\delta_1 b_1 \bar{S}_2 + \delta_2 b_2 \bar{S}_3}{b_1 \bar{S}_2 + b_2 \bar{S}_3}; \tag{22}$$

$$\delta_4 = \frac{\alpha_1 a_1 \bar{S}_1 + \alpha_2 a_2 \bar{S}_2}{a_1 \bar{S}_1 + a_2 \bar{S}_2}; \quad a_i = \frac{18 K_i \mu_i}{3K_i + 4\mu_i}, \ i = 1, \ 2; \tag{23}$$

$$b_i = \frac{18 \varkappa_i \gamma_i}{3\varkappa_i + 4\gamma_i}; \quad P_i = \frac{6m_i}{3 + 4m_i}; \quad m_i = \frac{\mu_i}{K_i}; \quad c_i = \frac{3K_i - 2\mu_i}{18 K_i \mu_i}; \tag{24}$$

$$h_i = \frac{6m_i'}{3 + 4m_i'}; \quad m_i' = \frac{\gamma_i}{\varkappa_i}; \quad d_i = \frac{3 - 2m_i}{3 + 4m_i}; \quad n_i = \frac{9}{3 + 4m_i}; \tag{25}$$

$$\varkappa_1 = \left\{ \frac{n_1}{K_1} \bar{l}_2 + \frac{n_2}{K_2} (1 - \bar{l}_2) - 2 \frac{[d_1 \bar{l}_2 + d_2 (1 - \bar{l}_2)][P_1 \bar{l}_2 + P_2 (1 - \bar{l}_2)]}{K_1 P_1 l_2 + K_2 P_2 (1 - \bar{l}_2)} \right\}^{-1}; \tag{26}$$

$$\varkappa_2 = \left\{ \frac{n_1}{K_1} \bar{l}_1 + \frac{n_2}{K_2} (1 - \bar{l}_1) - 2 \frac{[d_1 \bar{l}_1 + d_2 (1 - \bar{l}_1)][P_1 \bar{l}_1 + P_2 (1 - \bar{l}_1)]}{K_1 P_1 \bar{l}_1 + K_2 P_2 (1 - \bar{l}_1)} \right\}^{-1}; \tag{27}$$

$$\varkappa_3 = \frac{\varkappa_1 h_1 \bar{S}_2 + \varkappa_2 h_2 \bar{S}_3}{h_1 \bar{S}_2 + h_2 \bar{S}_3}; \tag{28}$$

$$\varkappa_4 = \frac{K_1 P_1 \bar{S}_1 + K_2 P_2 \bar{S}_4}{P_1 \bar{S}_1 + P_2 \bar{S}_4}; \qquad (29)$$

$$\gamma_1 = \left(\frac{\bar{l}_2}{\mu_1} + \frac{(1-\bar{l}_2)}{\mu_2}\right)^{-1}; \; \gamma_2 = \left(\frac{\bar{l}_1}{\mu_1} + \frac{1-\bar{l}_1}{\mu_2}\right)^{-1}; \qquad (30)$$

$$\gamma_3 = \gamma_1 \frac{\bar{S}_2}{\bar{S}_2 + \bar{S}_3} + \gamma_2 \frac{\bar{S}_3}{\bar{S}_2 + \bar{S}_3}; \qquad (31)$$

$$\gamma_4 = \mu_1 \frac{\bar{S}_1}{\bar{S}_1 + \bar{S}_4} + \mu_2 \frac{\bar{S}_4}{\bar{S}_1 + \bar{S}_4}. \qquad (32)$$

It is to be noted that expressions (25)-(32) yield the effective bulk modulus K and shear modulus μ. Accordingly, we will express the former as

$$K = \frac{\varkappa_3 h_3 (\bar{S}_2 + \bar{S}_3) + \varkappa_4 h_4 (\bar{S}_1 + \bar{S}_4)}{h_3 (\bar{S}_2 + \bar{S}_3) + h_4 (\bar{S}_1 + \bar{S}_4)}, \qquad (33)$$

and the latter as

$$\mu = \gamma_3 (\bar{S}_2 + \bar{S}_3) + \gamma_4 (\bar{S}_1 + \bar{S}_4). \qquad (34)$$

3. Comparison of Theoretical Results with Experimental Data. The graph in Fig. 2 depicts the coefficient of thermal expansion calculated according to relations (19)-(32) with experimental data on W—Co alloys, this comparison indicating a rather close agreement.

A comparison between the moduli of elasticity calculated according to relations (25)-(34) and experimental data on the WC—Co alloy [8] and the W—SiO_2 composite [9] is made in Tables 1 and 2. The maximum error of calculations is here smaller than 8%.

The increasingly stringent precision requirements in determination of thermophysical properties of materials used in power engineering pose a problem of predicting a set of thermophysical properties from a single standpoint.

According to the well-known thermodynamic relation, the difference between specific heat C_p at constant pressure and specific heat C_V at constant volume in homogeneous media (phases) can be defined as [12]

$$C_{pi} - C_{Vi} = 9 K_i \alpha_i^2 V_0 T, \qquad (35)$$

where V_0 is the molar volume and $i = 1, 2$. From the condition for transition in a quasi-homogeneous medium one can conclude that Eq. (35) will be satisfied also for the effective properties

$$C_p - C_V = 9 K \alpha^2 V_0 T. \qquad (36)$$

It also follows from definition (35) that

$$\langle C_p \rangle - \langle C_V \rangle = 9 \langle K \alpha^2 \rangle V_0 T. \qquad (37)$$

The bracket of possible C_p values for a binary material is [13]

$$0 \leqslant \langle C_p \rangle - C_p \leqslant 9 \frac{K_1 K_2 v_1 v_2 (\alpha_1 - \alpha_2)^2}{\langle K \rangle}. \qquad (38)$$

It therefore is possible to predict the values of constant-pressure specific heat C_p and of constant-volume specific heat C_V for a binary material on the basis of relations (35)-(38) with the aid of expressions (19) and (33).

Both thermal and electrical conductivities of binary materials have been determined theoretically on the basis of the percolation model [3, 4], and the results then compared with experimental data. The expressions for the thermal conductivity λ and the shear modulus μ have the same form, moreover, which can be explained by the same form of the equation describing both characteristics

$$q_i = -\lambda_{ij} \nabla T, \; \sigma_i = \mu_{ij} \varepsilon_j. \qquad (39)$$

These expressions, therefore, will yield the coefficient of thermal expansion α as well as the thermal conductivity λ, the specific heats C_p and C_V, and the moduli of elasticity K, μ for binary materials.

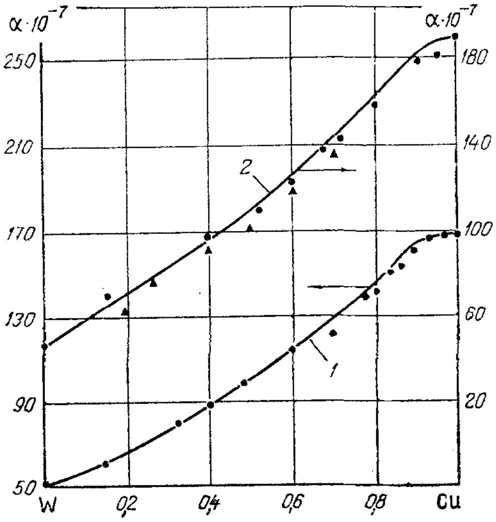

Fig. 2. Coefficient of thermal expansion α (1/°C) of W–Cu alloy: 1) T = 293-373°K; 2) T = 293-773°K; dots represent experimental values [14].

In conclusion, it will be noted that the model of a binary material (Fig. 1) under consideration here can evolve into the Odelevskii model with isolated inclusions [14] (Fig. 1d) or into the Frey–Dul'nev model with interpenetrating components [15] (Fig. 1c). For describing the properties of a heterogeneous material with isolated inclusions or with interpenetrating components one, therefore, can use expressions (19)-(34) with the geometrical parameters defined either as

$$\overline{S}=\overline{S}_1=\overline{S}_1^{(M)}=\overline{S}_3=0,\ l_1=0,\ \overline{S}_2=v_1^{2/3},$$
$$\overline{S}_4=1-v_1^{2/3},\ \overline{l}_2=v_1^{1/3};\ 0\leqslant v_1\leqslant 1,$$

in the Odelevskii model [15] or as

$$S_1=S_1^{(M)}=C^2,\ C=0.5+A\cos\left(\frac{2\pi-\varphi}{3}\right).$$

in the Frey–Dul'nev model [16]. When $0\leqslant v_2\leqslant 0.5$, then $A = -1$ and $\varphi = \cos^{-1}(1 - 2v_2)$. When $0.5\leqslant v_2\leqslant 1$, then $A = 1$ and $\varphi = \cos^{-1}(2v_2 - 1)$. Here $\overline{S}_2 = 0$, $\overline{S}_3 = 2C(1 - C)$ and $\overline{S}_4 = (1 - C)^2$, where $C = \overline{l}_1 = \overline{l}_2$.

NOTATION

v_i, volume concentration of the i-th component; v_c, percolation threshold; α, coefficient of thermal expansion; σ_{ij}, stress tensor; ε_{ij}, strain tensor; K, bulk modulus; μ, shear modulus; S_{ijkl}, compliance tensor; C_{ijkl}, elasticity tensor; $L_i(x_i, x_j)$, length of a segment through the i-th component perpendicular to both axes Ox_i and Ox_j; and $S_i(x_k)$, area of a section through volume V perpendicular to the Ox_k axis and occupied by the i-th component.

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METHOD OF DETERMINING THE PHASE VARIABLES OF THE SOLID PHASE IN DISPERSE FLOWS

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A method was developed for contactless measurement of the kinematic characteristics of the solid phase in disperse flows. The proposed method was substantiated empirically and the error was determined.

Although knowledge of the coordinates and velocity fields of both phases in multiphase systems fully determines the intensity of the target processes in heat and mass transfer, until now they have been little studied due to the lack of experimental methods of investigating them.

This article attempts to develop a method of determining the empirical probability density function for the coordinates and absolute velocity of a test particle in a disperse flow — in particular, in apparatuses with a monodisperse fluidized bed. To do this, we need an empirical method of determining the phase variables of the test particle.

The familiar method in [1, 2] for measuring the coordinates of an isotope-labeled particle has several advantages over other methods [3, 4]: 1) it allows for continuous recording of the position of the particle in the fluidized bed; 2) it permits measurements to be made at any point in the apparatus; 3) the transducers are located outside the apparatus and do not disturb the natural character of flow of the phases; 4) a test particle labeled with the Co^{60} isotope is representative in the sense that, for practical purposes, it is the same in size and weight as the other particles in the monodisperse bed.

However, the method does have several shortcomings, making it impossible to evaluate the particle concentration field in the phase space; instability of the electronic equipment, since the signal is analyzed in analog form; oscillograms based on calibration curves are analyzed manually; it is necessary to differentiate the experimentally obtained coordinates in order to obtain estimates of the absolute velocity of the tagged particle [5].

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