The results obtained can be used for predicting the effective thermal conductivity of polydispersed (two-component and multicomponent) granular systems and for developing programs for the experimental investigation, monitoring, analysis, and generalization of the measurement results.

NOTATION

P, porosity of the packing; d_I, d_{II}, d_{III}, average particle diameters; r_I, r_{II}, r_{III}, average particle radii; λ_1, λ_2, λ_3, λ_p, thermal conductivities of the grains and the component in the pores; r_c, r_{c_1}, r_{c_2}, radii of "cuffs"; $\lambda_{eff.III}$, $\lambda_{eff.II}$, $\lambda_{eff.I}$, effective thermal conductivities of intermediate fractions and of the entire system; $\lambda_{sk.II}$, thermal conductivity of the skeleton; y_c, y_{c_1}, y_{c_2}, y_1, y_2, y_3, y_4, relative radii of element with averaged parameters; V_g, V_p, volumes of the grains and the pores; h, Φ, height of the microroughness of the grains and heat flux spreading function (determined by formulas from [2]).

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CALCULATION OF THE THERMAL CONDUCTIVITY OF HETEROGENEOUS MATERIALS WITH DISORDERED STRUCTURE

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UDC 536.21

Structural models for determining the effective transport coefficients for two-component heterogeneous materials are analyzed and the results are compared with experimental data.

The problem of calculating the thermal conductivity of heterogeneous materials with the help of the theory of generalized conductivity is still an important problem, in spite of the progress made [1, 2]. The development of new technologies for obtaining heterogeneous materials greatly increases the range of possible structures, whose neglect can lead to large errors in determining conductivity.

It should be noted that sometimes this structural dependence of the conductivity is not used correctly: secondary properties are chosen as a foundation for the new model and for the computed characteristics [3]. The classical starting information for calculating the characteristics determining the conductivity includes the volume fractions (m_1, m_2) and coefficients of thermal conductivity of the components (λ_1, λ_2). We will examine heterogeneous materials consisting of two solid components and we will estimate the accuracy that we can expect for the computational results. If it is assumed that the coefficients of thermal conductivity of the components are known to within ~10% and the volume fractions to within 5%, then a calculation using the equation for a structure with cubic isolated inclusions [2]

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 41, No. 4, pp. 607-616, October, 1981. Original article submitted July 30, 1980.

0022-0841/81/1404-1069$07.50

$$\lambda = \lambda_2 \left[\left(1 - \sqrt[3]{m_1} + \frac{1}{\nu - 1} \right)^{-1} m_1 + 1 \right], \qquad (1)$$

where $\nu = \lambda_1/\lambda_2$ and the index 1 corresponds to the inclusion component, leads to a mean square relative error ~13%. It stems only from the errors in the initial data and does not take into account the correspondence between the model and the real structure. The validity of the computed characteristic is checked by comparing it to experimental data, which are also uncertain to an extent. Thus, a deviation of ±15-20% of the computed results from the experimental results can be assumed to be completely permissible. Within the error range, the computed characteristics must be checked by comparing them with the statistically representative quantity of experimental data, analyzing first the distribution law for the deviations.

The structures of combined (sintered, cast, porous, and nonporous) materials having the properties of a mechanical mixture, prepared from quasiisomeric particles (particle sizes with respect to coordinates differing by not more than a factor of 2-3), are divided into two large groups: with isolated inclusions of one of the components (matrix structure) and with interpenetrating components [1, 2, 4]. The fact that uniformity in the types of structures indicated over a wide range of variation of volume fractions (concentrations) of components, up to limiting cases, can be achieved by certain technological methods has not been excluded. In this respect, most of the computed characteristics are applicable without restrictions over a range of variation in the volume fractions from 0 to 1. However, if special techniques for controlling the structure in the technology of preparing the materials are not provided, then it is difficult to attain uniformity of a single type of structure over the entire range of variation of volume concentration. For low concentration, a matrix structure is most probable, while for high concentration, an interpenetrating structure is most probable.

In what follows, we analyze a model and computed characteristics which take into account the rearrangement of a structure with isolated inclusions into an interpenetrating structure with the volume concentration of the component varying over a wide range. For a concentration m_1 less than 0.125, the structure is viewed as a matrix structure, where cubic inclusions of component 1, uniformly distributed within component 2, are isolated from one another, and the conductivity is calculated using (1). Further increase in concentration leads to a change in the type of structure. Connecting bridges with a square transverse cross section appear between isolated inclusions and the structure is transformed into an interpenetrating one (Fig. 1). As the concentration increases, the transverse cross section of the bridges increases and for $m_1 = 0.5$, the structure is a spatial orthogonal lattice with cubic symmetry [5]. The threshold volume concentration (0.125) at which the structure begins to rearrange was chosen taking into account the data in [6-8].

The theory of percolation, based on mathematical modeling using a computer, as well as certain experimental data [8-11], indicate that threshold concentration occurs at $m_1 = 0.15 \pm 0.03$.

In principle, if there is the required justification, the threshold concentration for the appearance of an interpenetrating structure can be related to high or low values of m_1 [9]. Comparison of experimental data with computational results using the model in [11] indicates the change in the magnitude of the threshold concentration as a function of the ratios of the component coefficients of conductivity, which is explained by the specific properties of the structure.

We choose, as the unit cell, 1/8 of the unit cube according to symmetry conditions. The cube is separated by adiabatic surfaces $1-2-3-4$, $1'-2'-3'-4'$, $1''-2''-3''-4''$, and $1'''-2'''-3'''-4'''$ into parts which form 9 branches connected in parallel with 14 thermal resistances (Fig. 2) [7].

The conductivity of the cell λ, inversely proportional to the total thermal resistance, is defined as:

$$\lambda = \lambda_2 \left\{ 0.75 + A^2 \left[\frac{\nu - 1}{\nu - A(\nu - 1)} \right] + \frac{\nu}{\nu + 1} [0.5 + A^2(\nu - 1)] \right\}, \qquad (2)$$

where $\nu = \lambda_1/\lambda_2$.

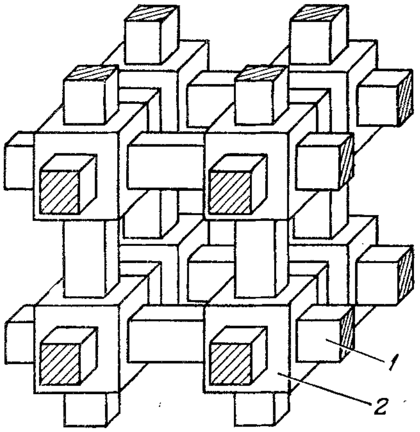

Fig. 1

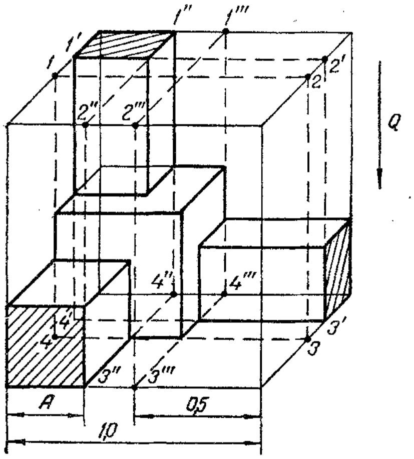

Fig. 2

Fig. 1. Structural model of a heterogeneous material with m > 0.125; 1) connecting bridge; 2) core.

Fig. 2. Unit cell: $1-2-3-4$; $1'-2'-3'-4'$; $1''-2''-3''-4''$; $1'''-2'''-3'''-4'''$ are auxiliary adiabatic surfaces; A is a dimensionless structural parameter; the arrow indicates the direction of heat flow Q.

The dimensionless structural parameter A, which is numerically equal to the length of an edge of the square transverse cross section of a connecting bridge, is expressed in terms of the volume concentration:

$$A = \sqrt{2(m_1 - 0.125)/3}, \tag{3}$$

where $0.125 \leqslant m_1 \leqslant 0.5$ and $0 \leqslant A \leqslant 0.5$. For $m_1 = 0.125$, Eq. (2) is modified and it can be represented as (1). If $m_1 = 0.5$, then $A = 0.5$ and the sides of the connecting bridges attain the sizes of the core, which corresponds to Dul'nev's structural model [5]. The conductivities of models calculated according to [5] and (2) coincide.

Thus, the structural model and expression (2) relate two basic types of structures (matrix and interpenetrating) as limiting cases.

The use of Eq. (2) under conditions when the conductivity of the component in the pore space is $\lambda_1 = 0$, $\nu = 0$ is determined by the maximum porosity 0.875.

The conductivities of porous materials, for which $\nu = 0$, are calculated after simplifying (2):

$$0.125 \leqslant m_1 \leqslant 0.5, \ \lambda = \lambda_2(0.75 - A), \tag{4}$$

$$0.5 \leqslant m_2 \leqslant 0.875, \ \lambda = \lambda_2 A^{*2}. \tag{5}$$

In (5), the parameter A^* is determined with the help of the volume concentration of the conducting component 2:

$$A^* = \sqrt{2(m_2 - 0.125)/3}. \tag{6}$$

Equations (1) and (2) are valid for limiting transitions with $\nu = 1$ and with $m_1 = 0$ or $m_1 = 1$.

It is well known that, based on one structural model, several computed characteristics, differing by the method of linearizing the heat flow paths in the model volume, can be proposed. The most widely used methods involve separating the unit cell by adiabatic or isothermal and adiabatic surfaces [1, 12-14]. The computed data under the worst conditions ($\nu = 0$) can differ by 30-40%. Comparison of the computed characteristics with the numerical solutions [1, 12, 13] permits choosing the most accurate equation.

It is shown in [1] that the arithmetic mean value of the conductivities, found with different methods for linearizing the heat flow paths, agrees with the numerical solution. It should be noted that the adequacy of the structural model in this case is not subject to discussion and a more accurate equation gives results close to the numerical calculation exclusively for a specific model.

TABLE 1. Distribution of Experimental Data on the Volume Content of Components

Volume content interval	0–0,1	0,1–0,2	0,2–0,3	0,3–0,4	0,4–0,5
No, of data points	68	98	103	101	46

TABLE 2. Comparison of Computed and Experimental Data

Positions in Figs. 3, 4	Ref. for eq. used	No. of data points	$\bar{R}$, %	s, %
a	[15]	399	–30,9	24,0
b	[5]	416	+ 5,2	18,5
c	[5], [15]	415	–13,5	22,0
d	[40]	404	–26,0	24,0
e	[41]	415	–16,5	27,6
f	[30]	374	–18,1	20,0
g	[42]	416	–12,0	18,3
h	(1), (2)	416	+ 0,4	17,6

The data for the numerical calculation for the model in Fig. 2 were obtained for condition $\nu = 0$ using Zaidel's method with the unit cell separated into 4096 nodes. The error in the solution, found using Runge's method, does not exceed 5%. The results of the numerical solution exceed the data calculated using (2) by 25%.

Probably, this difference stems from the schematic nature of the model proposed for the structure, which is expressed in the cubic shape of particles and in the uniformity of the transverse cross section of the bridges over their length. The urge to obtain a simple computational relation for determining the conductivity of the mixture explains the choice of geometry for the model of the structure and the adiabatic method for separating the unit cell.

We compared the computed characteristics for determining the effective conductivity, proposed for sintered materials (dense compositions, porous metals, etc.), with experimental data, which contained 416 experimental points for the thermal and electrical conductivities of 378 substances. Most of the materials were porous and their porosity does not exceed 0.5 by volume fraction [1, 7, 15-39].

The distribution of experimental points over the volume content of the components is shown in Table 1. The nonuniformity of their distribution over the intervals is explained by the fact that for low concentrations (<0.1) most computed characteristics for matrix structures lead to close results, while for high concentrations (0.4-0.5), especially for porous materials, a number of technological problems in preparing the materials appear. If we take into account the fact that before the action of external conditions (pressing, sintering, etc.), the particles in a free-fill state form a substance with a porosity of 0.4-0.5, while during pressing the porosity decreases, then the reason for the sharp drop in the amount of experimental data in the literature on materials with porosity exceeding 0.4 becomes clear.

For dense materials, the quantity ν falls in the interval 0.05-20, while for porous materials, $\nu = 0$.

After analyzing specific conditions for the experiment, experimental data which were obtained with conductive heat transfer were chosen. The equations obtained by Frey [15], by Dul'nev [5], the mean-square value using the equations in [15] and [5], the equations obtained by Bruggeman [40], Odelevskii for a random mixture [41], Skorokhod [30], by Zarichnyak and Novikov [42], as well as (1) and (2), were used to compute λ_{comp}. In the latter case, for concentrations of one of the components less than 0.125, Eq. (1) is used, while for high concentrations (2) is used.

The choice of the computational equations stems from the necessity of comparing structural models with different characteristics, in order to investigate the possibility of using them over a wide range of variation of component concentrations.

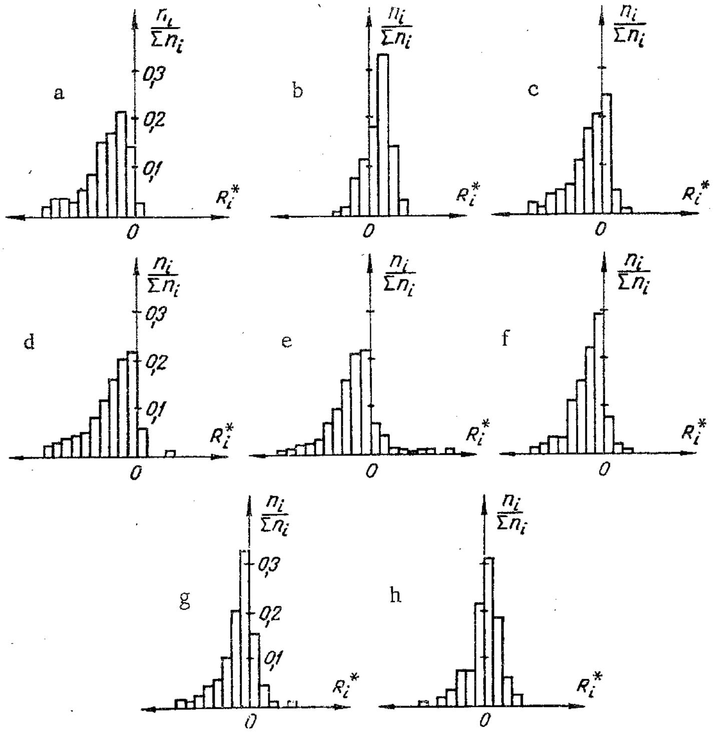

Fig. 3. Histograms of deviations between experimental and computed data: a) [15]; b) [5]; c) [5, 15]; d) [40]; e) [41]; f) [30]; g) [42]; h) according to (1) and (2); $n_i/\Sigma n_i$ is the relative fraction of data in the interval R_i^*.

The equations from [5, 15] and the arithmetic mean value of the results obtained with them concern models of the interpenetrating type. The dependences from [40] and (1) were obtained for matrix structures. The effective properties are calculated according to [30, 41, 42] for random mixtures.

The use of the model for sintered materials [43] is complicated due to the absence of the necessary starting information in works where experimental data on the conductivity of materials are presented.

Table 2 compares the computed and experimental data. Some of the changes in the number of experimental points for different equations are related to the fact that experimental points differing from computed values by more than ±100% were rejected as rough errors. In analyzing the data in [30], only porous materials were considered.

If it is assumed that the normal distribution is valid for deviations of the form $R = (\lambda_{exp}-\lambda_{comp})/\lambda_{exp}$ (in percent) and the symmetry properties are taken into account, then the arithmetic mean of the deviation must equal zero. The shift in the arithmetic mean indicates the systematic error that affects the computed data. Table 2 also shows the mean-square deviation (s) from the arithmetic mean value $\bar{R}$ with a confidence probability of 0.67.

Most of the equations examined lead to results that are too high compared to experimental data and, in addition, the fact that $\bar{R}$ differs from zero cannot be explained by the random nature of the deviations. Equations (1) and (2), for which the shift in the arithmetic mean of the deviations can be assumed to be random with a confidence of 0.95, are exceptions.

The mean-square deviations (s) of the computed data using the equations differ less and agree with previously presented estimates of the deviation of experimental and computed results. Analyzing the mean-square deviations, it should be noted that the disagreement between [5, 42] and (1), (2) with a confidence of 0.95 is accidental (checked according to R. Fisher's criterion), while the disagreement between (1) and (2) and other equations is not accidental.

Figure 3 shows histograms of the deviations of experimental data from the computed data.

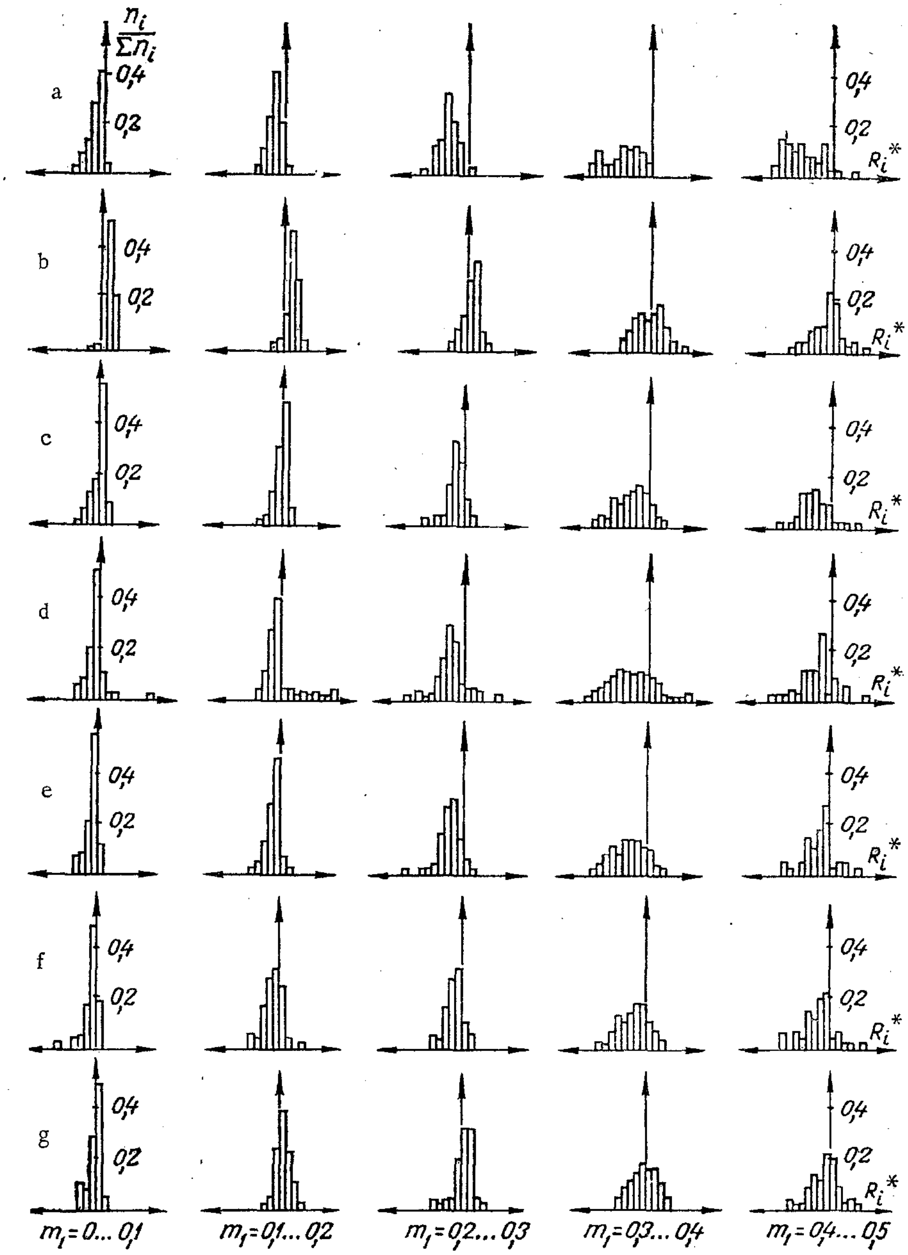

Fig. 4. Histograms of the deviations between experimental and computed data, calculated for different intervals of volume concentration of components m_1 (m_1 = 0...0.1; 0.1...0.2; 0.2...0.3; 0.3...0.4; 0.4...0.5). The histograms are constructed as in Fig. 3.

For most equations, the computed data exceed the corresponding experimental results, which leads to a large shift in the arithmetic mean deviation toward negative values. Therefore, the equations in [5, 30, 40, 41] cannot be recommended for calculating conductivities over a wide range of variation of the volume content of components, since they do not take into account the corresponding rearrangement of the structure.

Let us analyze the distribution law for the deviations in different volume content intervals. For this purpose, we will distribute the experimental data over five groups with the following intervals for variation of the volume content: 0-0.1; 0.1-0.2; 0.2-0.3; 0.3-0.4; and 0.4-0.5.

Figure 4 shows histograms of deviations for different computational equations.

As expected, in the volume content interval for a single component from 0 to 0.2, all equations give the smallest mean square deviation of computed data from experimental data, but the arithmetic mean is shifted from zero for [5, 15, 41]. For large concentrations, the mean-square deviation increases, especially for [1, 15], and there is a noticeable shift in the arithmetic mean value of the deviations for [1, 15, 30, 41, 42].

For small concentrations, we can recommend Eq. (1) and [1, 30, 40, 41, and 42], while for high concentrations, (2) and [5]. The latter is understandable also because for equal volume concentrations 0.5 both equations give results that agree exactly.

In collecting the experimental data, materials with porosity exceeding 0.5 ($\approx$ 30 data points), prepared from quasiisomeric particles, are also grouped together.

The results of the calculation using the equations systematically exceed the experimental data. Their disagreement exceeds 100% for 14 materials in calculations according to [15, 40], for 10 materials for calculations according to (2) and [5], and for two according to [11]. The best agreement is observed with the use of the equations in [42] (a single deviation by more than 100%), but the shift in the arithmetic mean value of the deviations is 11.7% with a mean-square deviation of 43.8%. The well-known difficulties in preparing highly porous bodies give rise to the use of diverse technological methods, which undoubtedly affect the structure and, as a result, the conductivity of the material.

Thus, in order to determine the conductivity of nonporous heterogeneous materials with a disordered structure over a wide range of variation of the volume content of components with $\nu > 0.01$, we can recommend Eqs. (1) and (2). For porous bodies ($\nu = 0$), the equations indicated provide the required accuracy only for porosities $\leqslant 0.5$.

Calculation of the conductivities of highly porous materials (porosity >0.5) requires further investigation and equations must be developed that take into account the specific conditions of the technology used to prepare them and their structure.

NOTATION

m_1 and m_2, volume concentrations of components 1 and 2; λ_1 and λ_2, coefficients of thermal conductivity of components 1 and 2; λ, effective thermal conductivity of a heterogeneous material; $\nu = \lambda_1/\lambda_2$, ratio of the coefficients of thermal conductivity of components 1 and 2; A and A*, dimensionless parameters of the structure; R, deviation of the computed value of the thermal conductivity from the experimental value, in %; $\bar{R}=\frac{1}{n}\Sigma R$, arithmetic mean value of the deviation; n, overall number of experimental points compared; $s=\sqrt{\Sigma(R-\bar{R})^2/n}$, mean-square deviation around $\bar{R}$; R_i, where i = 0...10%; 0... −10%; 10...20%; −10... −20%, etc., are the deviation intervals; n_i, number of data points falling into the deviation interval R_i; $n_i/\Sigma n_i$, relative fraction of data points with deviation R_i.

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EXPERIMENTAL STUDY OF DENSITY AND VISCOSITY OF FOUR-COMPONENT LIQUID SYSTEMS

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UDC 547.571:532.14.001.5.532.1-133

Results are offered from an experimental study of the P—V—T functions and viscosity of multicomponent liquid systems, for which an equation of state is developed.

The basic task of this experimental study was an examination of density and viscosity of two- and multicomponent liquid systems, consisting of alcohols and aldehydes of normal structures and their isocompounds. These systems in the liquid phase are widely used in technological processes.

Analysis of experimental methods of density study [1-4] demonstrated that for multicomponent liquid systems the most suitable approach is that of hydrostatic weighing [5, 6].

Having determined the compositions of the specimens and the range of state parameters at which the experiments were to be performed, we prepared the apparatus, with consideration of the polarity of the individual components and the polarity of the solution itself, which can significantly affect normal operation of the tracking system sensor if no precausions are taken.

Initially the densities of the following binary solutions were studied: n-butyl alcohol (80%)-isobutyl alcohol (20%); n-butyric aldehyde (80%)-isobutyric aldehyde (20%) [7, 8]. Subsequently, three systems of these two mixtures were prepared: 1) 10% first, 90% second; 2) 60% first, 40% second; 3) 90% first, 10% second.

The alcohols and aldehydes used to prepare the solutions were purified by the technique of [9]. Special care was taken to eliminate water from the alcohols. Purity of the alcohols and aldehydes was 99.96 and 99.97% by weight. The pure aldehydes and alcohols were maintained in the dark, and the multicomponent liquid systems were prepared immediately before the experiments.

Density measurements were made over the range 285-500°K at pressures of 0.1-50 MPa, with temperature determined by a platinum resistance thermometer. The results obtained are presented in Table 1.

Analysis of the experimental results permitted establishment of a generalized equation of state for each of the three systems in the form

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0022-0841/81/4104-1077$07.50