THERMAL PROCESSES DURING THE INTERACTION OF OPTICAL RADIATION WITH HETEROGENEOUS LAYERED MEDIA

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We develop a method for the calculation of thermal and thermochemical processes during the interaction of optical radiation pulses of duration $t_p > 10^{-6}$ sec with heterogeneous layered media.

In the theoretical analysis of the interaction of intense optical radiation with matter, of considerable interest is the study of thermal processes [1]. In recent years, a great deal of attention has been given to such processes during the interaction of radiation with heterogeneous layered media where one or several layers contain small particles which absorb and scatter the energy of the radiation. This is due to the necessity of studying the interaction of optical radiation with dielectrics and multilayer optical coverings which contain absorbing inhomogeneities, the interaction of radiation with heterogeneous layered biotissues which contain pigment particles, in the optical recording of information in layered magnetorheological media, etc. [1-4]. In these cases, the energy of the incident radiation is absorbed by the particles and by the medium as a whole, the heated particles exchange heat with the surrounding medium by virtue of the nonlinear heat conduction, the molecules of the medium can experience thermochemical transformations, etc. These processes will be studied in the present work.

We consider the propagation of a beam of optical radiation in the layered medium along the x axis. The radiation has wavelength λ and intensity distribution I over the cross section. The individual layers have different optical and thermophysical properties. The dividing boundaries of the layers $x_i (i = 0, 1, 2, \ldots)$ are perpendicular to the x axis, the thickness of layer i is equal to $\Delta x_i = x_i - x_{i-1}$, and the characteristic thickness of a layer is $\sim$10-100 μm. Let us suppose that the layer with boundaries x_1, x_2 and thickness $x_2 - x_1$ contains a collection of spherical or spheroidal particles with characteristic size $\sim$1 μm which absorb and scatter the energy of the radiation, and the remaining layers are homogeneous. For simplicity we assume that the particles are monodispersed. The propagation of radiation in the layered medium will be described by the quasistationary transport equation

$$\frac{\partial I}{\partial x} + (\alpha_{ai} + \alpha_{si}) I = 0, \tag{1}$$

Here, for the heterogeneous layer which contains the particles, $\alpha_{a2} = N_0 S_s K_a$, $\alpha_{s2} = N_0 S_s K_s$ where $S_s = \pi r_0^2$ for a sphere, and $S_s = \pi r_a^2 r_B \left[r_B^2 \sin^2\left(\frac{\pi}{2} - \alpha\right) + r_a^2 \cos^2\left(\frac{\pi}{2} - \alpha\right)\right]^{-1/2}$ for a spheroid. The equation of thermal balance of a single particle which absorbs the energy of the radiation and emits energy into the surrounding medium by nonlinear heat conduction has the form, in the approximation of uniformly heated particles,

$$V_0 \rho_0 c_0 \frac{\partial T_0}{\partial t} = K_a I S_s - j_T S_0. \tag{2}$$

The thermal conductivity of the medium which surrounds the particles is specified in the form

$$\varkappa_i = \varkappa_{i\infty} (T_m / T_\infty)^a, \tag{3}$$

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where $\varkappa_{i\infty}=\varkappa_i(T_m=T_\infty)$, $a=\text{const}$. Since the characteristic time t_c needed to establish the temperature field around a particle is short relative to the duration of the heating t_p (see below), the heat exchange of the particle with the medium will be considered in the quasi-stationary approximation [5]. The quasistationary heat exchange of a spherical particle with allowance for (3) was studied by Pustovalov and Romanov [5] who, for $a \neq -1$, obtained the following formulas for the temperature distribution around the particle T(r) and j_T

$$T = T_m\left[1+\frac{r_0}{r}\,\{(T_0/T_m)^{a+1}-1\}\right]^{1/(a+1)}, \tag{4}$$

$$j_T = -\varkappa\frac{dT}{dr}\bigg|_{r=r_0} = \frac{\varkappa_{i\infty}T_m^{a+1}}{(a+1)T_\infty^a r_0}\left\{\left(\frac{T_0}{T_m}\right)^{a+1}-1\right\}. \tag{5}$$

The quasistationary heat exchange of a spheroidal particle with allowance for (3) was studied by Pustovalov and Bobuchenko [6] who obtained, in the one-dimensional approximation T = T(ξ) with respect to ξ, for $a \neq -1$,

$$T = T_m\left[1+\frac{\ln\operatorname{th}\xi/2}{\ln\operatorname{th}\xi_0/2}\left\{\left(\frac{T_0}{T_m}\right)^{a+1}-1\right\}\right]^{\frac{1}{a+1}}, \tag{6}$$

$$j_T = -\frac{\varkappa_{i\infty}T_m^{a+1}\arcsin(1/\operatorname{ch}\xi_0)\{(T_0/T_m)^{a+1}-1\}}{(a+1)cT_\infty^a\ln\operatorname{th}(\xi_0/2)\operatorname{sh}\xi_0}. \tag{7}$$

The two-dimensional nonstationary heat-conduction equation for the entire volume of the layered medium with allowance for energy dissipation has the form

$$\rho_c c_c\frac{\partial T_m}{\partial t} = \frac{1}{R}\frac{\partial}{\partial R}\left(R\varkappa(T_m)\frac{\partial T_m}{\partial R}\right)+\frac{\partial}{\partial x}\left(\varkappa(T_m)\frac{\partial T_m}{\partial x}\right)+Q. \tag{8}$$

The initial and boundary conditions for (1)-(8) are as follows:

$$I(x_0) = I_0\exp(-R^2/R_c^2),\quad 0\leqslant t\leqslant t_p;\quad I=0,\quad t>t_p;$$
$$T_0(x,\ R,\ t=0)=T_\infty;\quad T_m(x,\ R,\ t=0)=T_\infty;\quad T_m(R=R_{max})=T_\infty;$$
$$T_m(x=x_0)=T_\infty;\quad T_m(x=x_{max})=T_\infty;\quad \frac{\partial T_m}{\partial R}\bigg|_{R=0}=0. \tag{9}$$

The distribution of radiation intensity across the section of the beam was assumed to be Gaussian. The radius R_{max} and length $x_{max}-x_0$ of the computational volume were chosen such that the unperturbed initial conditions are satisfied on its surface in the time interval under consideration. The heat sources in (8) are specified in the form

$$i=1,\ 3,\ 4,\ 5,\ \ldots,\ Q=\alpha_{ai}I;\quad i=2,\ Q=N_0S_0j_T. \tag{10}$$

The kinetic equation for the thermochemical transformation (TCT) of the molecules will be taken from [7]:

$$\frac{\partial f}{\partial t} = -\frac{kT}{h}\exp\left(-\frac{\Delta H - T\Delta S}{R_g T}\right)f \tag{11}$$

with the initial condition f(t = 0, x, R) = 1.0. Condition (8) is written in the approximation of quasicontinuous medium which indictes that the temperature is locally balanced in a physically infinitesimally small volume which contains many particles whose dimensions are smaller than the characteristic dimensions of the problem: $N_0^{-1/3} < \Delta x_i$, R_c, $\sqrt{t_p\chi_i}$. The use of (10) indicates that the particles are assumed to be pointlike heat sources with identical

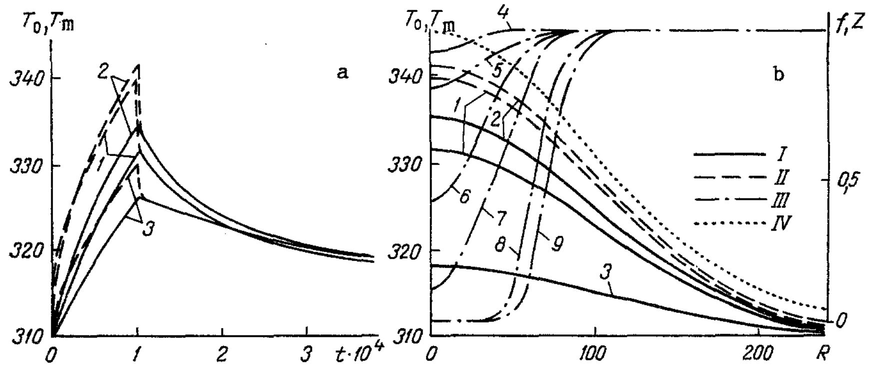

Fig. 1. a) Dependence of the temperature of the medium T_m (I) and particles T_0 (II) in points R = 0; $x = x_1$ (1); x_1 + 2 μm (2); x_2 (3) on t. b) The distributions T_m (I), T_0 (II) and the degree of thermochemical transformations f (III) along R for the medium immediately at the surface of the particles (8, 9) and far from the particles (4, 5, 6, 7) for 10^{-4} (1, 2, 4, 6, 8, 9), $6 \cdot 10^{-4}$ (3, 5, 7, 8, 9) sec, and the cross sections $x = x_1$ (1, 3, 4, 5, 8) x_1 + 2 μm (2, 3, 6, 7, 9). The distribution of the normal intensity of the radiation Z along R for $x = x_1$ for $0 \leq t \leq t_p$ (IV) (b). T_0 and T_c are in deg K, t is in sec, and R in μm.

temperature in a physically small volume. The volume occupied by the particles themselves and by the heat aureolas around the particles (see formulas (4), (6)) is assumed to be much smaller than the volume of the surrounding medium.

A numerical method for the solution of the system of equations (1)-(11) was developed and realized. This method is based on the calculation of (1), (2) and (11) by an explicit Runge–Kutta difference scheme of the second order of accuracy. Equation (8) is solved by a conservative implicit difference scheme of the second order of accuracy with respect to the spatial variable, and of the first order of accuracy with respect to the time variable [8]. This formulation of the problem and the numerical method for its solution can be used for the analysis of various problems which involve the interaction of intense optical radiation with heterogeneous layered media. By way of example, we consider the interaction of optical radiation with λ = 0.514 μm with layered biotissues containing a layer with pigment particles (grains) which absorb the energy of the radiation. The optical and geometrical characteristics of the individual layers of the biotissue are taken from [9], for example, $x_2 - x_1 = 5$ μm, $x_3 - x_2 = 20$, $x_4 - x_3 = 80$ μm. The thermophysical characteristics are taken from [10]. The following values of the parameters were used: radius of the spherical pigment grain are taken from [11].

The energy of the incident radiation in the pigment layer is selectively absorbed by the pigment particles; the absorption in the surrounding medium is considerably weaker. During the irradiation, therefore, the temperature of the particles exceeds the temperature of the surrounding medium. Energy is transferred to the entire volume by heat exchange of the particles with the surroundings. The characteristic time of the heat exchange for a particle with radius $r_0 \sim 1$ μm can be estimated using the formula $t_c \sim r_0^2/4\chi_c \sim 1.4 \cdot 10^{-6}$ sec, where $\chi_c = 1.73 \cdot 10^{-7}$ m²/sec is the thermal conductivity of the medium [10]. Consequently, the ratio between the duration of the radiation pulse t_p and the characteristic time of the heat exchange of a particle t_c determines the character of the process. During the interaction of the radiation with the heterogeneous medium for $t_p < t_c$, the particles are heated considerably in excess of the medium and there is no appreciable heat exchange between the particles and the medium during the pulse [3]. For $t_p > t_c$, the interaction of radiation with the heterogeneous medium takes place under a developed heat exchange of the heated particles with the surrounding medium and under a general temperature increase. This case is studied in the present work. The pigment particles have high optical and thermal stability (heating to ∼620°K does not lead to any changes [12]) and the TCT processes are considered for the surrounding medium.

We calculated the thermal processes in heterogeneous layered media for the threshold energy density $E_{0t} = 1.88\cdot10^3$ J/m², $t_p = 1\cdot10^{-4}$ sec, $R_c = 140$ μm taken from experiment [13]. Immediately after the beginning of the irradiation, the particles are heated, and they exchange heat with the surrounding medium (see Fig. 1a). The excess heating of the particles with respect to the medium $\Delta T_0 = T_0 - T_m$ reaches a maximum value ∿8-10 K and depends on the local value of the intensity, size and optical characteristics of the particles, etc. The particles at $x = x_1$ begin to heat up more rapidly than particles for $x > x_1$ since, for $x > x_1$, the particles are acted upon by radiation with lower intensity because of the attenuation by the medium. However, during the formation of a characteristic temperature profile (see Fig. 2) resulting from the simultaneous heating by radiation and cooling by heat conduction, the maxima of the particle temperature and of the temperature of the medium move towards the interior of the layer $x_2 - x_1$. After the radiation is switched off, particles transfer energy to the medium during time $\sim 5\cdot10^{-6}$ sec; their temperature T_0 becomes equal to T_m. The thermal relaxation time of a particle $5\cdot10^{-6}$ sec agrees with the estimate of the characteristic time of heat exchange t_c.

Figure 1b shows the distributions of T_0, T_m, f, $Z = I(x, R)/I_0$ along R in the planes $x = x_1$, $x_1 + 2$ μm for several moments of time. In the cases under consideration, the characteristic radius of the beam R_c exceeds by an appreciable factor the thicknesses of the absorbing layer $x_2 - x_1$. Therefore, during the heat exchange, there is a considerable heating of the regions adjacent to the layer $x_2 - x_1$ along the longitudinal x axis and, to a lesser degree, heating of the medium in the radial direction. With increasing R (decrease of I), the temperature of the particles and the temperature of the medium decrease, and their difference also decreases. The region of continuous TCT broadens considerably in the radial direction when the pulse is terminated and the heated region cools down. The rate of thermochemical reactions (11) depends considerably on the temperature and has a threshold character. The heating (or lowering of temperature) below ∿330°K leads to the absence (or a sharp slowing down) of the reactions during the above time intervals. The presence of a considerable temperature difference between the particles and the medium during the pulse can lead to a sharp difference of reaction rates directly at the surface of the particle (with temperature $T = T_0$) and in the medium (with $T = T_m$). As a result, a selective interaction regime can be realized when the thermochemical reactions proceed in the immediate vicinity of the surface of the heated particles and are absent in the remaining volume of the medium [3], i.e., the TCT microregions are localized near the particles. In the present case, for an interaction with threshold energy [13], one observes a vast region in which a selective interaction is realized, together with a TCT microregion of characteristic size ∿30-50 μm. Figure 1b shows the boundary of the region in which TCT proceed directly at the surface of the particles. In the space between the boundaries of the regions with volume and surface TCT, the microregions broaden until they completely join at the boundary of a TCT macroregion. We note that, for pulses with $t_p = 10^{-5}$-10^{-3} sec and below-threshold energy density $E_0 = (0.5\text{-}0.8)E_{0t}$, a selective interaction with particles is realized, leading to TCT which involves the molecules of the medium directly adjacent to the surface of the particles, without the formation of a continuous transformation macroregion.

Figure 2 shows the distributions of T_0 within the layer $x_2 - x_1$, T_m, f and Z along x for R = 0 for several moments of time. During the pulse, the energy of the applied radiation is adsorbed by the layers $x_2 - x_1$, $x_4 - x_3$, and it is practically not absorbed in layers $x_1 - x_0$, $x_3 - x_2$. This leads to a considerable heating of the particles and medium in the layer $x_2 - x_1$ and to a weaker heating in the layer $x_4 - x_3$ (because of the attenuation of the radiation). As a result, during a two-dimensional nonstationary heat exchange between the heated layers and the surrounding medium, a characteristic temperature distribution is formed, with a subsequent cooling after the termination of the radiation pulse.

In the layer $x_2 - x_1$, a region of volume TCT is formed as well as a wider region of transformations which take place directly at the surface of the particles. The maximum excess heating of the medium which takes place in point $x_1 + 2$ μm and which was calculated for the experiments [13] for $R_c = 140$ μm is, on average, equal to 25-30°K and depends weakly on t_p in the interval 10^{-3}-10^{-4} sec.

Figure 3a shows the heating isotherms for the medium $\delta T_m = T_m - T_\infty$ and particles $\delta T_0 = T_0 - T_\infty$ and the lines of a constant degree of TCT for the molecules of the medium f = 0.5 for several moments of time. During the heat exchange and cooling for $t > t_p$, the TCT macroregion with $f < 0.5$ broadens considerably. At the same time, the region of transformations in the immediate vicinity of the surface of the grains for $t > t_p$ conserves a constant form since,

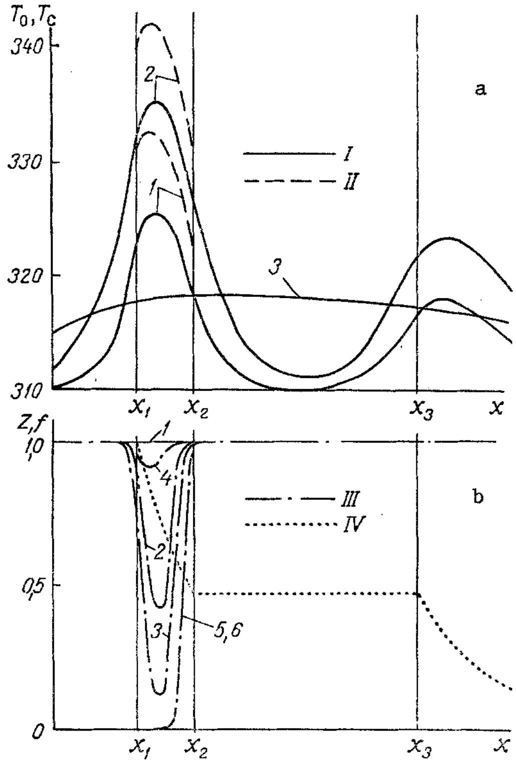

Fig. 2. a) Distributions of T_m(I) and T_0(II) along x for R = 0 for t = $0.5\cdot10^{-4}$ sec (1); $1\cdot10^{-4}$ (2); $6\cdot10^{-4}$ (3). b) The distribution of f(III) for the medium near the surface of the particle (4, 5, 6) and far from it (1, 2, 3) for t = $0.5\cdot10^{-4}$ sec (1, 4); $1\cdot10^{-4}$ (2, 5); $6\cdot10^{-4}$ (3, 6). The distribution of Z along x for R = 0 for $0 \le t \le t_p$ (IV) (b). (The temperatures T_0 and T_m are in °K).

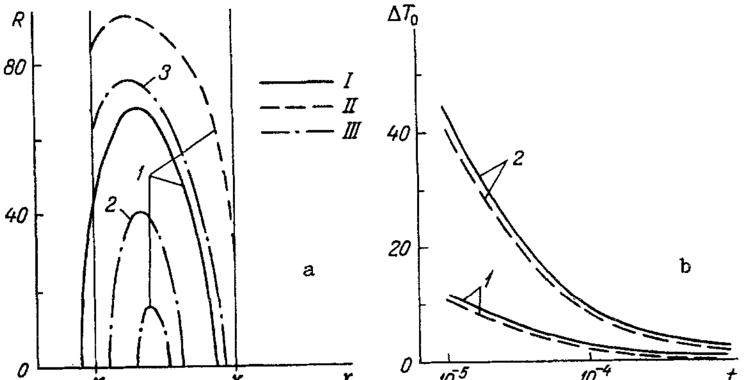

Fig. 3. Heating isotherms of the medium δT_m = 20 K (I) and particles δT_0 = 20 K (II), and the equal level of TCT f = 0.5 (III) at the particles (3) and in the medium (1, 2) for t = $1\cdot10^{-4}$ sec (1, 3); $6\cdot10^{-4}$ (2, 3) (a). b) The dependence of $\Delta T_0 = T_0 - T_m$ on t for spherical particles (I) with r_0 = 0.5 and 1 µm, and for spheroids of the same volume (II) (1, 2), respectively. The quantity R is in µm, t is in sec, and ΔT_0 in °K.

during the characteristic time of thermal relaxation of the grain $t_c \sim 10^{-6}$-10^{-5} sec, no appreciable TCT take place in the present case.

Figure 3b shows the excess heating of the particles ΔT_0 with respect to the medium in the point $x = x_1$, R = 0 calculated for R_c = 140 µm for $t_p = 10^{-3}$, $t\cdot10^{-4}$; $1\cdot10^{-3}$ sec for the conditions of [13]. We took $E_0 = 1 \times 10^3$ J/m^2 for $t_p = 10^{-5}$ sec. Under threshold interaction, the excess heating of the grains ΔT_0 sharply increase with decreasing t_p and reaches ∿45°K for $t_p = 10^{-5}$ sec. For $t_p = 10^{-3}$ sec, the excess heating reaches $\Delta T_0 \lessapprox$ 1-3 K. Consequently, for radiation pulses with $t_p > 10^{-3}$ sec with threshold and below-threshold intensity, the excess heating of the grains does not exceed $\Delta T_0 \lessapprox$ 1-2 K, and the thermal processes can be described in a quasihomogeneous approximation without taking into account the particles. However, for threshold and below-threshold interaction with $t_p < 10^{-3}$ sec and for above-threshold interaction with $t_p > 10^{-3}$ sec, it is necessary to take into account the heterogeneous structure of the absorbing layers. We note that the possibility of using the quasihomogeneous approximation to calculate the thermal processes in heterogeneous layers must be analyzed in each concrete case.

To explain the effect of the size of the particles on their excess heating, we consider ΔT_0 in point $x = x_1$, $R = 0$ calculated for the parameters E_0 and t_p [13], but for $r_0 = 0.5$ µm, $N_0 = 2.86 \cdot 10^{17}$ m^{-3}, $K_a = 0.67$ and the same optical parameters of the heterogeneous layer and same volume filling of the layers by the particles. The qualitative behavior of ΔT_0, T_0, T_m (see Fig. 3b) remains the same. However, the excess heating of the particles ΔT_0 for r_0 = 0.5 µm is considerably lower than for r_0 = 1 µm since the energy absorbed by a particle decreases with decreasing radius. The heating of the medium remains practically the same and, accordingly, the properties and final dimensions of the TCT macroregion remain the same. At the same time, when the excess heating ΔT_0 of the particles is reduced, the size of the zone where the TCT microregions near the particles are formed reduces considerably, and it can lead to its complete absence. Consequently, the particle size can affect considerably the excess heating of the particles with respect to the medium. With increasing size of the particles r_0, ΔT_0 increases and with decreasing r_0, ΔT_0 falls and the properties of the heterogeneous medium approach the properties of the homogeneous media.

Up till now, we have, for simplicity, considered spherical particles. In the general case, the form of the particles which absorb the energy of the radiation can be different, for example, the particles can be spheroidal. The effect of the form of the particles on TCT was studied by a numerical calculation of the interaction of the pulses of radiation with layered media, when the layer $x_2 - x_1$ contains a collection of identical spheroids with r_a = 0.415 µm, r_B = 0.725 and r_a = 0.830, r_B = 1.45 µm. The optical parameters of the heterogeneous layer and the volume filling of the particles remain the same, and the volume of the spheroids is equal to the volume of spherical particles with r_0 = 0.5 and 1 µm, respectively. Figure 3b shows the excess heating of the spheroidal particles ΔT_0 as a function of t_p, calculated for the experiments [13]. For the spheroidal particles, the excess heating is lower by 1-3°K than for spherical particles of the same volume. This is due to an increase of the surface area and to an increase of the heat flux away from the spheroid. Otherwise, the qualitative and quantitative picture of the interaction processes remains practically unchanged.

It is known that a medium which suffers TCT changes its optical properties, in particular, its reflection coefficient for the radiation increases [14]. The time interval after which the optical properties change was determined experimentally in [14] and is equal to 5-500 msec, on average ∿50 msec. In our case, the pulse duration $t_p = 10^{-5}$-10^{-3} sec is considerably less than ∿50 msec, and the change of optical properties of the medium during the pulse of radiation can be neglected.

NOTATION

I_0, intensity on the axis of the beam; R, radial coordinate in the cylindrical coordinate system; α_a, absorption coefficient; α_s, scattering coefficient; N_0, particle concentration; S_s, cross-sectional area of the particle; α, angle between the large axis of the spheroid and the x axis; r_a and r_B, semiaxes of the spheroid; K_a and K_s, efficiency factors for the absorption and scattering of the radiation by a particle; r_0, radius of the spherical particle; V_0, volume of the particle; ρ_0, c_0, density and heat capacity of the particle substance; T_0, temperature of the particle; j_T, energy-flux density carried by thermal conduction away from the surface of the particle; S_0, surface of the particle; T_m, temperature of the medium; T_∞, initial temperature; r, radial coordinate measured from the center of the particle; ξ, spheroidal coordinate; $c = \sqrt{r_B^2 - r_a^2}$, $r_B > r_a$; $\xi = \xi_0$, surface area of the spheroid; ρ_c, c_c, density and heat capacity of the medium; Q, power density of energy dissipation; χ_i, thermal conductivity; R_c, characteristic radius of the beam; f, fraction of molecules of the substance which do not suffer thermochemical transformations; ΔH, ΔS, differences of enthalpies and entropies in the initial and activated state; R_g, universal gas constant; k and h, Boltzmann and Planck constants; E_{0t}, maximum energy density on the axis of the beam for threshold interaction; and Z, normalized intensity of the radiation.

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THE DENSITIES OF MONOHYDRIC SATURATED ALCOHOLS

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Analytic relationships have been derived of the density to the number of carbon atoms for a series of liquid n-alcohols from propan-1-ol to octadecan-1-ol at 263-513°K and atmospheric pressure.

Particular interest attaches to regularity in the properties in homologous series of organic compounds because these can be used to survey and predict properties for largely unexamined members.

The behavior of the density has been examined [1] for n-alcohols containing 1-12 carbon atoms; beginning with propan-1-ol, there is a smooth variation in density at constant temperature and atmospheric pressure within the accuracy of the measurements.

Here we extend the bounds in temperature (263-513°K) and in number of alcohols (C_3–C_{18}). We consider virtually all known alcohols, amongst which the light n-alcohols have been most fully examined and the heavy ones less so.

First, graphs were plotted with ρ and N as coordinates to give isotherms over the range 263-513°K with a step of 10°K for the density as a function of the number of carbon atoms on the basis of [2-8] for the C_3-C_6, C_8, C_{10}, C_{12} alcohols, as well as the accurate measurements of [9] for C_{16} and the densities recommended in [10] for C_{14}, C_{16}, and C_{18}. Then the densities of C_7, C_9, C_{11}, C_{13}, C_{15} and C_{17} alcohols were read from the curves to increase the data volume.

The final processing incorporated all these results and the densities found graphically.

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