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LASER PHOTOACOUSTICS METHOD FOR DETERMINATION OF THE COEFFICIENTS OF THERMAL CONDUCTIVITY AND THERMAL DIFFUSIVITY OF MATERIALS

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In the article are given results of studies of laser photoacoustic for determination of the thermophysical properties of materials by the microphone detection circuits. It has been shown that the photoacoustic method for determining the thermophysical properties of materials, complementing other existing methods. The characteristic features of the application of the photoacoustic method based on the Rosencweig-Gersho fundamental theory are considered. The mathematical model of the problem for determining the thermal diffusivity of solid-state materials in one-dimensional and three-layer photoacoustic cell is developed. The proposed photoacoustic method can be successfully applied to the study of materials that are complex in structure, such as structurally inhomogeneous, multilayer, composite, powdery, nanomaterials, etc.

Keywords: photoacoustic method, thermal properties, thermal diffusivity and conductivity of materials.

Introduction

The developments of the modern fundamental science and innovative technologies, as a rule, contribute to the emergence of new physical and chemical research method sand to various materials with predetermined properties. In particular, the fundamentals study of the structure of substances at the atomic and quantum levels, laid down in the beginning of the last century, led in the second half to the discovery of optical quantum generators in the optical range: masers in the IR (1954) and lasers (1961) - in the visible radiation region, the authors of which were awarded the Nobel Prize in Physics. Thus, the discovery of lasers, in turn, contributed to the emergence of a number of completely new, research methods of substances, including laser photoacoustic (PA) spectroscopy.

Thermophysical methods for studying the properties of materials, in addition to studies of generally accepted standard thermophysical coefficients (thermal conductivity, heat capacity, thermal expansion, thermal diffusivity, etc.), as well as temperature changes and other related coefficients: optical, mechanical, structural, and many others, are human activities (sciences, technology and production).As noted, despite some progress and the development of theoretical methods for determining the thermophysical properties (TP) of materials (analytical, numerical, computer simulations, etc.), experimental methods still remain the real source for determining and obtaining information [1-2].The correct choice of the method for determining the TP of materials depends on a number of factors that must be considered in their research: on the possibility of the method itself, the physical states and characteristics of the object of study, on the conditions of the experiments set (measurement accuracy, temperature range, source selection, etc.It is known that at present, there are numerous standard equipment and instruments for determining TP materials from different manufacturers.

It may be noted, for example, the German company Netzsch, which offers a sufficient set of thermophysical instruments and equipment, including those based on the laser flash method (LFA) in a wide temperature range. However, despite their high quality indicators, there are a number of necessary limitations that should be carried out when conducting experimental work. Characteristic features of the considered laser PA methods are: high sensitivity and resolution, versatility, non-contact, high information content, speed, wide areas (practical) applications, sufficient simplicity and inexpensive, available experimental equipment. Due to these qualities, PA methods are successfully applied in almost all areas of scientific research, both fundamental and practical [3-5]. In the framework of this work, we will consider the possibilities of the laser FA method with an indirect (microphone) registration scheme in determining the thermal diffusivity of materials.

1.Mathematical model of the problem.

The theoretical fundamentals of laser photoacoustic methods for determining the thermal characteristics of materials are currently rather well studied, as a rule, at moderate power densities of optical radiation [4-6] and far from the phase transition point.

The main regularities between the values of the PA signal and the desired thermal parameters of the studied substances, with an indirect registration scheme, are described in the framework of the Rosencweig-Gersho (RG) linear theory [8]. Basically, precisely in the linear mode, the results of PA experiments in determining the TPS of various materials and substances are the most preferred and coincide with the literature for known materials [7-11].We present an analysis of the theory of the laser PA method for determining the thermal diffusivity of solid-state materials, following the condition of [8], i.e. for one-dimensional and three-layer PA chamber consisting of: a transparent gas (air), a sample with an absorption coefficient β and a substrate (Fig.1). The amplitude modulated laser radiation with intensity $I = I_0\left(1+\cos\omega t\right)/2$ and modulation frequency ω falls on the surface of the sample under study. Denote by lg, ls and lb the thickness of the gas layers, the samples and the substrate, respectively.

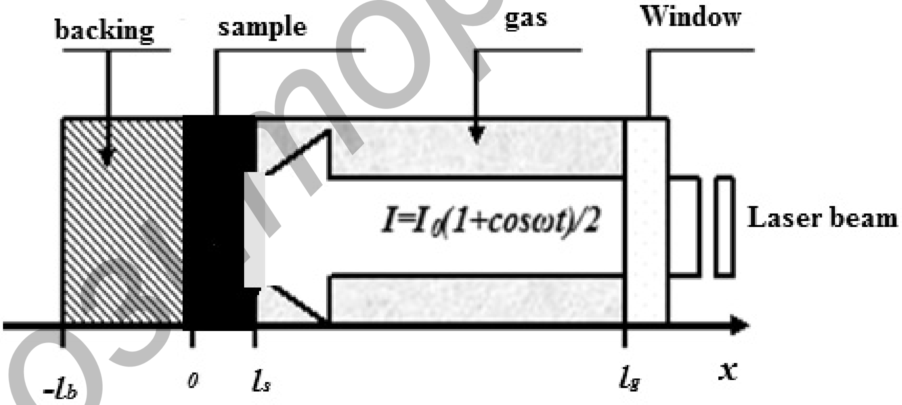

Fig.1. One-dimensional and three-layer PA cell.

It is assumed that the heat released in the sample is transferred to the near-surface layer of gas in the chamber and the substrate, only due to a thermal conductivity, i.e. temperature and heat fluxes at all boundaries are continuous. This layer of gas, thickness is $2\pi\mu_g$, where $\mu_g = \sqrt{2a_g/\omega}$ - the thermal diffusion length, periodically expanding causes an acoustic pressure oscillation in the PA cell. The viscosity and finiteness of the speed of sound in a gaseous medium are neglected, i.e. are met a conditions $l_g << \lambda_a$. Under such assumptions, the main task of the PA effect is reduced to determining the periodic component of the temperature field at the sample – substrate interface. It is may be found from the solution of the system of differential heat conduction equations for a gas, a sample and substrate:

$$\frac{\partial^2 T_g}{\partial x^2} = \frac{1}{a_g}\frac{\partial T_g}{\partial t}, \qquad 0 \le x \le l_g, \tag{1}$$

$$\frac{\partial^2 T_s}{\partial x^2} = \frac{1}{a_s}\frac{\partial T_s}{\partial t} - A[1+\exp(i\omega t)]\exp(\beta x)\ , \ -l_s \le x \le 0 \tag{2}$$

$$\frac{\partial^2 T_b}{\partial x^2} = \frac{1}{a_b}\frac{\partial T_b}{\partial t}, \qquad (-l_s + l_b) \le x \le -l_s \tag{3}$$

Here, $T_i(x,t)$- temperature oscillation, regarding ambient temperature T_0, for the corresponding layers, k_i, $a_i = \kappa_i / C_{pi}$ - heat conductivity and thermal diffusivity coefficients, $C_{Pi} = (\rho c_p)_i$ - specific heat per unit volume, (indices $i = s, b, g$ denote the sample, substrate and the gas filling the PA cell, respectively), $A = \beta I_0 \eta / 2\kappa_s$, η- the efficiency, which the absorbed light is converted into heat through nonradiative transitions.

The necessary boundary conditions for solving the system of equations (1) - (3), follow from the requirement of continuity of temperature and heat fluxes on the substrate – sample and sample – gas boundaries, as well as from the condition of equality of the cell wall temperature with the ambient temperature T_0:

$$\begin{gathered} T_g(0,l_g) = T_b(0,-l-l_b) = 0, \quad T_g(0,t) = T_s(0,t), T_b(-l,t) = T_s(-l,t) \\ k_g \frac{\partial T_g}{\partial x}(0,t) = k_s \frac{\partial T_s}{\partial x}(0,t), \quad k_b \frac{\partial T_b}{\partial x}(-l,t) = k_s \frac{\partial T_s}{\partial x}(-l,t) \end{gathered} \tag{4}$$

Thus, the systems of equation (1) - (3) together with the boundary conditions (4), represent the general thermal part of the mathematical model of the problem [8].To the solve acoustic part of the problem, it is assumed that the generation of acoustic (sound) waves occurs adiabatic (the test sample inside the PA cell is sealed) and the pressure increment can be determined from the adiabatic equation of an ideal gas:

$$\delta P / P_0 = \gamma \delta V / V_0 \quad \text{or} \quad PV^{\gamma} = const, \tag{5}$$

where, $\gamma = C_p / C_V$ - the ratio of the heat capacities of the gas.

The above mentioned condition is $l_g << \lambda_a$, allows to consider the propagation of acoustic waves in the PA cell as a voluminous, and use the ratio $\delta V / V_0 \approx \delta x(t) / l_g$, where are the movements $\delta x(t)$ can be defined as: $\delta x(t) = 2\pi\mu_g \theta(t) / T_0$. For the solving acoustic part of the problem, need to determine the average value of $T_g(x,\omega)$ by thickness $2\pi\mu_g$, determined from the system of equations (1) - (4) for the thermal part of the problem. Therefore, for the acoustic part we have:

$$\delta P_g(t) = \frac{P_0 \gamma 2\pi\mu_g \theta_g(t)}{l_g T_0}. \tag{6}$$

The systems of equation (1) - (4) together with (6) represent a mathematical model of the problem.

3. Analysis of the thermal coefficients

As the analysis shows, the expression for, the dependence of the parameters of the PA signal included in (6) on the optical, thermal, and geometric parameters of the studied media, as well as the substrate and the buffer gas, is rather complex and ambiguous for the practical application, i.e. analysis of the results of PA experiments. Therefore, for the physically clearerand practical application [8], it is expedient to allocate 6 special cases, for the two most practical important cases:

A) for optically transparent samples, where the inequality is true $l_s < \mu_\beta$, where $\mu_\beta = \beta^{-1}$ the optical absorption length (that is, when the laser radiation is not completely absorbed by the sample and its significant part passes through the medium);

B) for optically opaque samples takes place $l_s > \mu_\beta$, when the sample completely absorbs laser radiation at a distance μ_β significantly smaller than its thickness l_s. The relations between the quantities both μ_g and μ_β on the one hand, μ_β and l_s on the other, make it possible to obtain the six particular cases for A) and B) cases.

In the RG- theory [8], a more detailed analysis is given for each case, the type of dependence of the parameters of the PA signal on the thermal, optical and geothermic characteristics of all the layers of the PA camera and the source of (laser) radiation is determined. Now we are consider some typical special cases of the RG-theory in determining the thermal characteristics of materials

3.1 Optically transparent and translucent samples $l_s < \mu_\beta$

For this case were identified the thermal conductivity and thermal diffusivity coefficients for a number of transparent and translucent nanocomposite polymeric materials [12].In particular, when the substrate is considered optically thick, i.e. $r_b >> 1$ and the thermal coupling, between the layers of the PA cell, is taken to be: $b_{gs} << 1$ and $b_{bs} << 1$, for the complex value of the PA signal, we have:

$$\left|\delta P_g\right| \approx \frac{P_0 \gamma \eta_b I_0}{\sqrt{2} l_g T_0} \frac{\mu_g}{k_b \sigma_b} \left(\frac{4 b_{bs}}{\exp(\sigma_s L_s) - \exp(-\sigma_s l_s)} \right). \qquad (7)$$

Where, for amplitude and phase of the signal we get:

$$\left|\delta P_g\right| \approx A \frac{\mu_g b_{bs}}{k_b \sigma_b} \exp(-\sigma_s l_s), \qquad (8)$$

and $\varphi = -l_s \sqrt{\frac{\omega}{2 a_s}} - \frac{\pi}{2}$. (9)

Here, $A = \frac{P_0 \gamma \eta_b I_0}{\sqrt{2} l_g T_0}$. It can be seen that in this case the phase and amplitudes of the PA signal have frequency dependencies as $\sqrt{\omega}$, ω^{-1}, respectively.

3.2 Analysis of a particular case for optical opaque and thermally thick samples $(\mu_s < l_s < \mu_\beta)$

This corresponds to case 5.2 (b), according to [8]. The characteristic amplitudes of the PA signal and its frequency dependences are as follows:

$$\left|\delta P_g\right| \approx \frac{P_0 \gamma I_0}{4 l_g T_0} \frac{\mu_g \mu_s r^2}{k_s R^2}, \qquad (10)$$

and the frequency is: ω^{-1}. Here r and R, the radii of the laser beam and the PA cell, respectively.

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Obviously, with FA experiments the value of $\left|\delta P_g\right|$ on the left part of the (10) determined by microphone (i.e. experimentally) and the required thermal characteristics (conductivity, diffusivity) are determined from the corresponding dependences of the quantities in the right-hand side. In particular, determining the coefficient of thermal diffusivity as, can be determined other thermophysical coefficients, as well as, the thermal diffusion length $\mu_i = \sqrt{2a_i/\omega}$ depending on the frequency of modulation of laser radiation or the amount of thermal activity (effusivity): $A = \sqrt{\rho C_p k}$ of the materials. For example, when performing calculations for the coefficient of thermal conductivity of quartz powder (Si –powder), with the values of the parameters [13]:

$$\gamma = 1.402\,; P_0 = 9.9\times10^5\,Pa\,; I_0 = 47.2\,mW\ / cm^2; \mu_g = 2.5\times10^{-2}\,cm\,;$$

$$l = 1.1cm\,; T_0 = 293\,K\,; r = 0.27\,cm\,; R = 0.5cm\ \text{жс}\ \delta P = 0.034\,Pa$$

was received: $\kappa_s = 0.99\,J/s\times m\times K$.

Conclusion

Thus, we can conclude that, the PA method for studying the thermal properties of materials has a number of features that substantially differ from other methods. The analysis shows that, despite the numerous methods for determining the thermophysical properties of materials, the PA method with a microphone detection, due to a number of its advantages, can also be successfully used in determining the thermal characteristic of various materials, especially where it is difficult to determine them by other methods. By changing the modulation frequency of the laser radiation, one can determine the length of thermal diffusion and, thus, determine the coefficient of thermal diffusivity of samples to different depths. This allows us to determine the thermophysical parameters, such "inconvenient" for other methods, samples such as multilayer, coated, composite, nanomaterials and others.

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