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Research of heat transfer processes in hydrogels by holographic interferometry and gradient thermometry

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The study of natural convection in structured optically transparent materials using pure and combined agarose-gelatin gels was carried out by optical holography. The article presents data on visualization of the occurrence and development of convective flows in such gels with non-stationary conductive heating from below. The similarities and differences of the conditions of heat transfer and the occurrence of convection in structured materials and droplet liquids are analyzed. For the first time experimentally obtained data on the effect of two interpenetrating and interacting structured media on the transition from conductive to convective heat transfer.

Keywords: natural convection in gels, optical holography, hydrogels, three-dimensional bioprinting.

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Today practical application of soft matter is of widespread interest, including mobile structured media, such as gels. Gels are usually understood as heterogeneous systems whose dispersed phase is bound by intermolecular links, and the dispersed phase filling the available space is a droplet liquid. Gels can be of different physical and chemical nature. The number of substances forming gels is large and varied: e.g. gelatin, hyaluronic acid, agarose, alginates, etc. [1,2]. Practical applications of gels are increasingly expanding [3–5] due to the fact that they belong to the so-called „smart materials" because of their unique rheological, thermophysical and physical-chemical properties. In recent years, gels are of special interest also due to the fact that they are considered to be the main working material for bioprinting — one of the promising areas of regenerative medicine development [6].

For practical implementation of additive technologies in the formation of bioreactors, gels are extremely convenient, since they allow multiple phase transitions from liquid to gel and back [7]. Gels are capable of creating artificial microchannels that intensify nutrients and oxygen supply into their space, as well as remove metabolic products in deep cell cultivation [8,9]. All of this imposes additional strict limitations on the methods for investigation and diagnosis of such systems, since gel-like materials require noninvasive methods of investigation, optical ones being the most preferable. It should be noted that gels as such are not stationary media, since they alter their structure with time. They possess properties of syneresis and thixotropy due to their relaxation processes, and anisotropy occurring during gel formation.

Depending on the source components, the method of production, the planned application and the physical-chemical properties of various gel materials, there are a large number of experimental methods to research their characteristics. These include, for example, differential scanning calorimetry [10], infrared spectroscopy [11], and broadband dielectric spectroscopy [12] and scanning electron microscopy [13] methods. These methods, as well as a number of others, enable research of the properties of a wide range of both organic and synthetic gel materials. However, the study of the thermal-physical properties of gels, especially taking into account phase transformations and availability of modifying components, such as cell cultures, requires the use of optical noninvasive methods [14–16].

The aim of the present paper is to implement an experimental complex for non-contact diagnostics of temperature fields and heat flows under non-stationary heating of hydrogel materials, allowing to determine their thermal-physical characteristics.

To visualize heat transfer dynamics in gel materials during the melting phase transition, the optical method of holographic interferometry with registration of interference fringes in real time was applied. This method is based on dependence between medium refractive index and its properties, composition, and temperature. The used experimental setup, the scheme of which is shown in Fig. 1, includes a helium-neon laser of 20 mW power with radiation wavelength $\lambda = 0.63\,\mu$m, an optical system consisting of reflecting and semi-transparent mirrors, an attenuator, space filters, lenses and a photoplate. According to the method, first a hologram was obtained on the photoplate. Then an experiment was done to obtain video frames (interference patterns) of the processes under study using a video camera focused on the working area. The camera was set up so that the image of the object and the interference fringes were observed simultaneously. The hologram was registered, and the photoplate was chemically

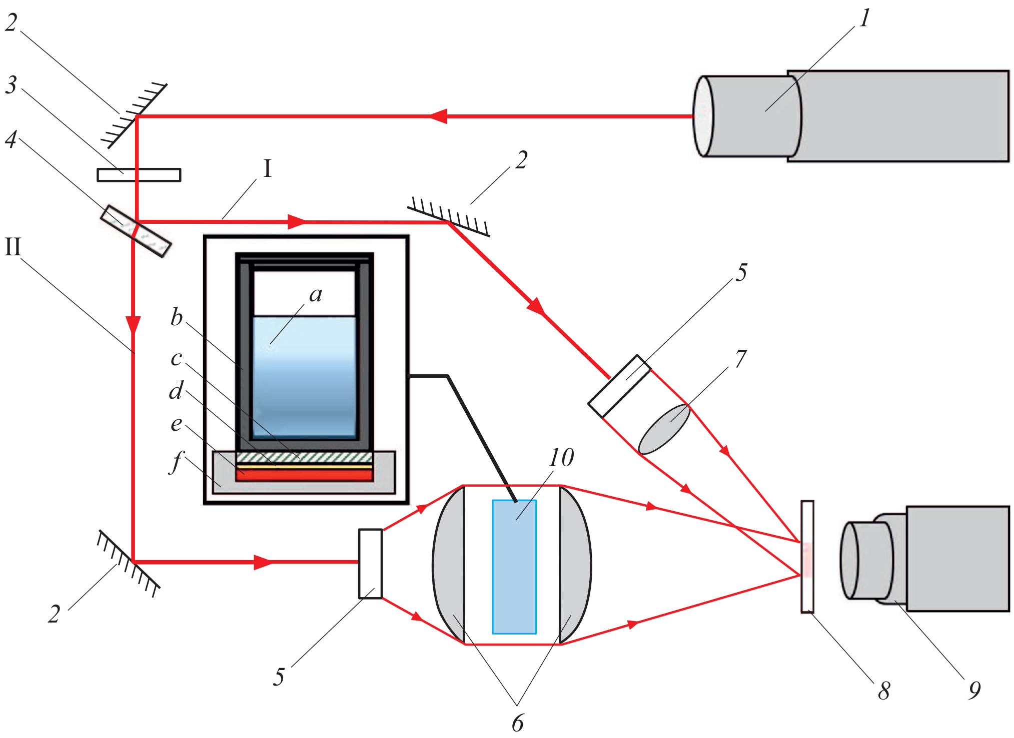

Figure 1. Experimental setup diagram. *1* — helium-neon laser, *2* — reflecting mirrors, *3* — attenuator, *4* — semitransparent mirror, *5* — space filters, *6* and *7* — lenses, *8* – photoplate, *9* — video camera, *10* — working area (*a* — hydrogel, *b* — optical cuvette, *c*— gradient sensor of heat flow, *d* — electrical insulation, *e* — electrical heater, *f* — heat insulation). I — non-object-bearing beam, II — object-bearing beam.

treated in a fixed position. The most important feature of the experiments in this study was the use of elements *3*, *5*, *7* and a high-resolution photographic plate PFG-01, designed to produce holograms with high resolving power. A beam attenuator *3* was used to regulate the contrast of interference fringes in process of making a video with the camera during the experiments. To improve the quality of the object-bearing and non-object-bearing beams, collimator systems comprising a combination of optical elements *5*, *6* and *5*, *7* were used.

Studies of heat exchange processes in the gel samples were carried out in a specially designed working area. The working area is an optically transparent cuvette with size of 5×10 mm and height of 15 mm, which was filled with the studied medium. The initial temperature of the test samples in the experiments was monitored using thermocouples connected to an electronic thermometer. A gradient heat flow sensor and an electrical heater were mounted in the bottom of the cuvette to supply heat with the regulated thermal load application on a direct current source. The heating power varied between 1.5 and 4.5 W. Use of a gradient heat flow sensor combined with the heating system made it possible to both measure and control the surface heat flow density brought to the bottom part of the working area, i. e. the intensity of heating.

Dynamics of non-stationary heating of samples was recorded by means of an installed universal video camera Fast Video-500M and was displayed on the computer screen. Video was recorded directly on the matrix of the video camera with dimensions of 30×20 mm. The resolution of the camera with a video recording rate of 1000 fps was 816×620 pixels. The distance from the matrix to the photoplate was 325 mm, the camera angle was 32°.

Studies of heat exchange processes were performed on various samples of hydrogel materials synthesized on the basis of agarose and gelatin. They were obtained by mixing dry agarose and gelatin powders of different concentrations with distilled water followed by heating over a pan of simmering water until the components were completely dissolved, and a homogeneous solution was obtained. The prepared solution was poured into an experimental cuvette, then the samples were cooled in an air thermostat to the initial temperature of the experiment $T = 20^\circ$C. The agarose content of the samples varied between 0.1–0.6 mass%, and the gelatin content — between 4.0–8.0 mass%. Samples with higher content of a gel-forming agent for the optical scheme (photoplate) used proved to be optically too dense for quality imaging.

Using the method of holographic interferometry described above, video images of interference fringes characterizing changes in the temperature field over time caused by heating hydrogel samples of different composition at different values of the heat flow supplied were obtained for

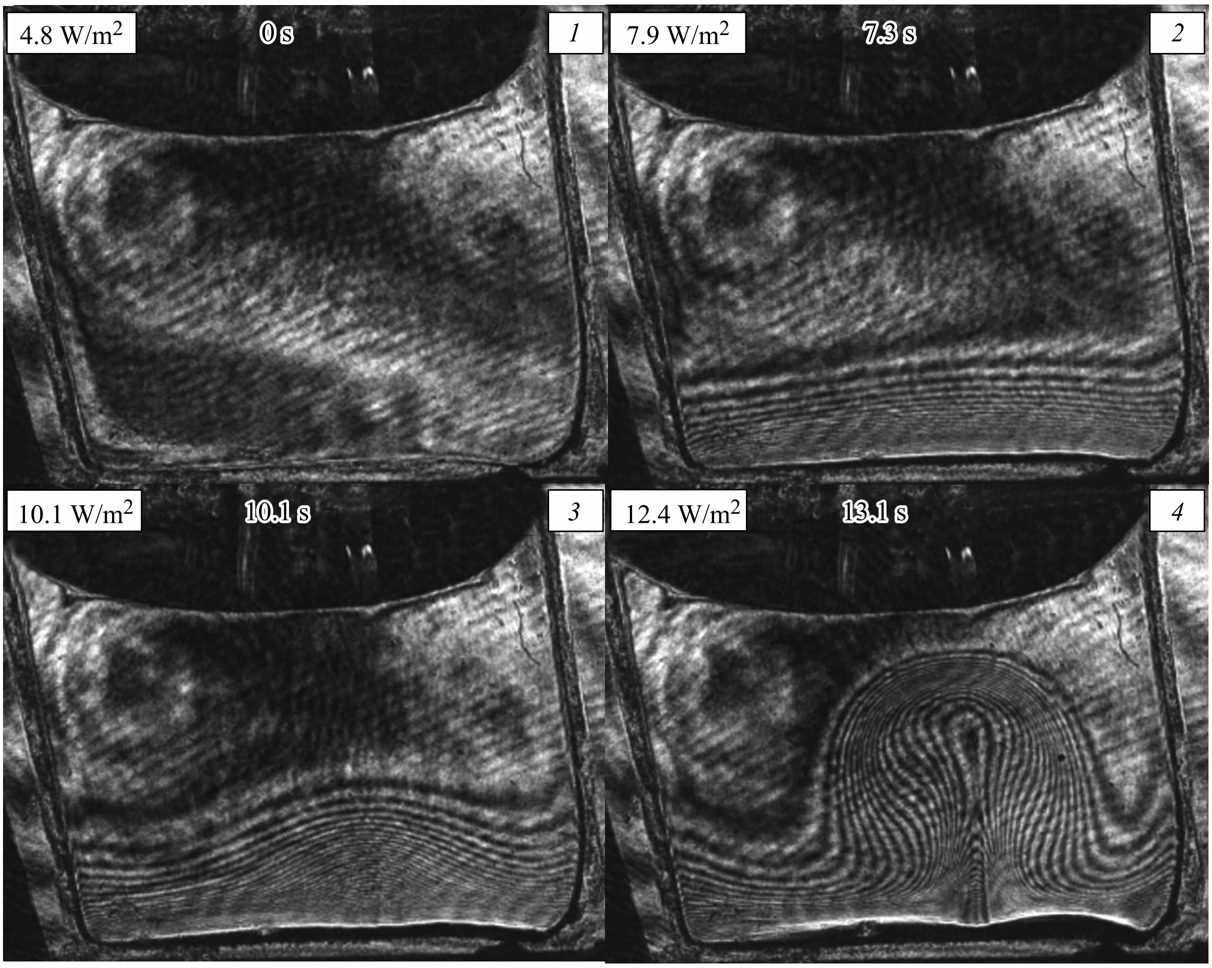

Figure 2. Interference patterns of free convection origination and development in a cuvette with gelatin (4.0 mass%) gel at $N = 4.5\,\mathrm{W}$. Explanation in the text.

the first time. Thus, experimental data on the origination and dynamics of convective flows in the cuvette with pure gelatin gel (4.0 mass%) at a constant heat load $N = 4.5\,\mathrm{W}$ are shown in Fig. 2.

The obtained video frames demonstrate different modes of non-stationary heat transfer in the gelatin gel over time. In Fig. 2 video frame *1* corresponds to the state of the studied sample at the initial moment of time before heating. It contains no interference fringes of thermal origin (corresponding to isotherms) appearing during heating.

From the time of heating and in the next time interval, interference fringes parallel to the heating surface appear. For these moments of time, there is a classical non-stationary thermal conductivity mode (video frame *2* in Fig. 2). The characteristic time when parallel isotherms curve with respect to the heating surface is identified with the time of convection start when interpreting the experimental data. As can be seen from Fig. 2 (video frame *3*), deformation of interference fringes occurs, i. e. the heat transfer mode changes from conductive to convective one, and, accordingly, non-stationary free convection begins to develop in the sample. Given that gels — are structured disperse systems with a 3D polymer frame formed by the dispersed phase with certain mechanical strength, the presence of curved isotherms is associated with gel melting, when it is transferred to the melt (solution) state.

Further heating of the gel in the cuvette forms a symmetric thermal comparable to the size of the cuvette (video frame *4* in Fig. 2). In the center is a drop of melt heating the adjacent gel layers. Such heating reduces mechanical strength of the surrounding gel and allows the droplet to rise upwards, overcoming its resistance. Behind the droplet, you can clearly see a concurrent layer of liquid. Since the viscosity of gelatin gel melt is high, the convective motion of the thermal remains laminar, at least under thermal loads investigated in the paper.

Fig. 3 shows the results of experiments when recording a video of convection onset in both pure agarose and gelatin gels and in a mixed gel derived from them. The experiments were performed at reduced heater power $N = 3.5\,\mathrm{W}$, because of risk of overheating and damage to the gradient heat flow sensor due to poor heat removal from the heater in dense gels.

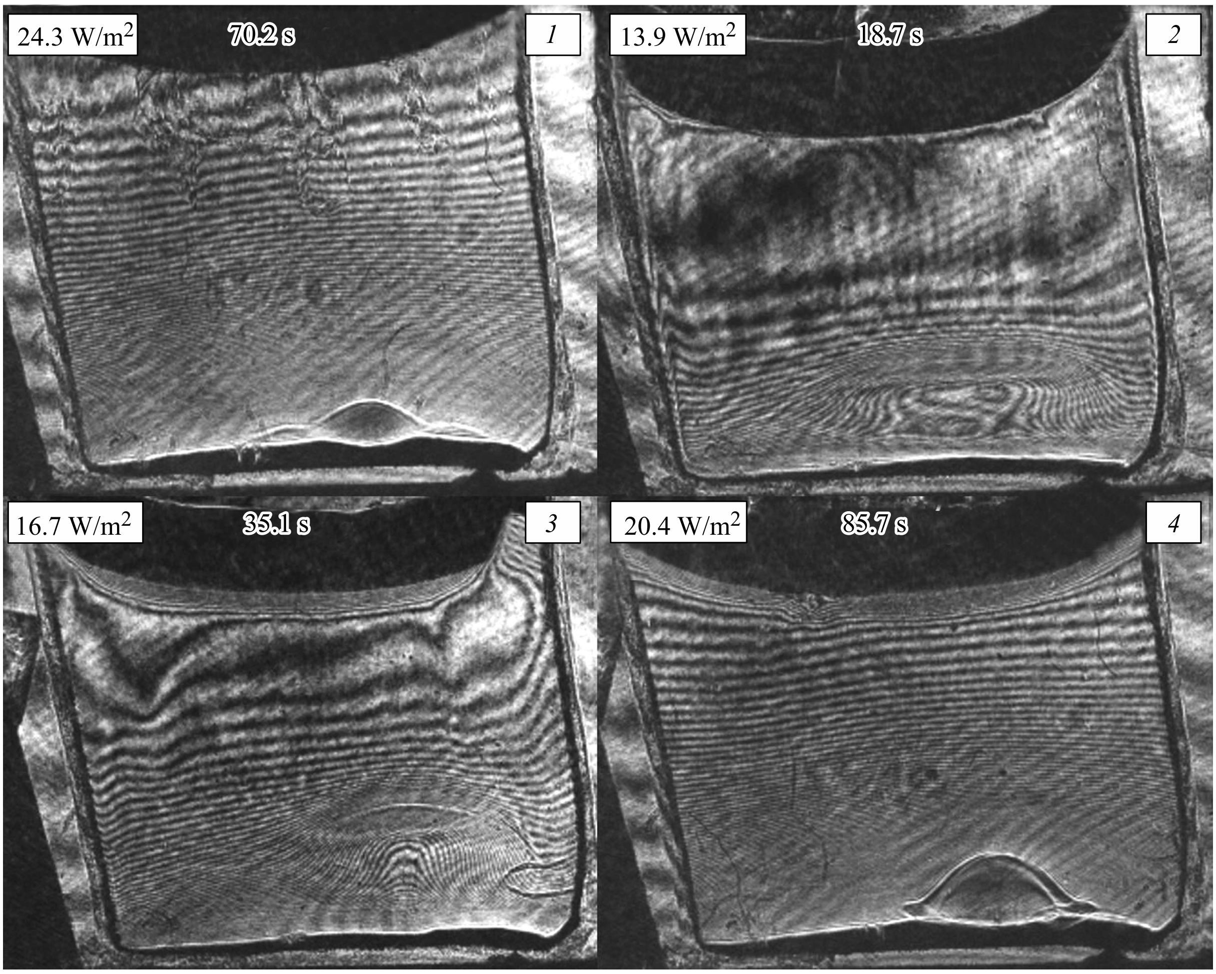

Figure 3. Interference patterns at the onset of convection for various hydrogel samples at heating power $N = 3.5\,\mathrm{W}$. *1* — agarose gel (0.4 mass%), *2* — gelatin gel (4.0 mass%), *3* — gelatin gel (8.0 mass%), *4* — mixed gel of agarose and gelatin (0.1 and 4.0 mass%, accordingly).

It can be seen (Fig. 3) that of the gel samples presented, gelatin (4.0 mass%) has the minimum time for convection to occur (video frame *2*). Video frame *3* in Fig. 3 shows a situation similar to that shown in frame *2*, but the time for convection start is noticeably longer here, since the gel density is increased by a factor of 2. In the case of pure agarose gel with a minimum concentration (0.4 mass%) (video frame *1* in Fig. 3) the convection start time is significantly longer than that for gelatin gels with 4.0 and 8.0 mass%. It follows from Fig. 3 (video frame *4*) that the heating conditions and natural convection in the mixed gel differ significantly from those in the pure gels being its components (the formation time and thermal shape). This indicates fundamental changes in the rheology of the mixture material related to changes in the intermolecular links in the disperse phase, and not only to differences in the thermal properties of mixed and single-component gels. When selecting the composition of the mixed sample, it was found that this was observed even at a negligible concentration of the modifying agarose component.

To conclude, we would like to note that the experimental studies conducted demonstrate the possibility of applying the methods of holographic interferometry in combination with gradient thermometry to study the processes of heat and mass transfer in complex and non-stationary structured media. Their development will enable non-contact (by the width and speed of interference fringe propagation) determination, by solving the inverse problem of thermal conductivity, the thermal-physical characteristics of gels depending on their temperature. Moreover, it is principally possible to visualize the concentration fields of nutrients, oxygen and carbon dioxide (by measuring the local density) during deep cultivation of microbioobjects in gels of different composition by holographic interferometry method.

New experimental data obtained on the occurrence of natural convective flows in structured hydrogel materials show their significant difference from similar flows in droplet liquids. Based on the example of gelatin gel, the characteristic stages of the heat exchange process, when the mode of non-stationary heat conductivity is replaced by heat transfer under conditions of free convection, have been determined. Since one of the current trends in the creation of bioprinting materials is the use of mixed gels, the proposed holographic method can be used to correctly select the thermal modes of bioprinter printing devices. Their calculation is impossible without determining the thermal properties of working media such as hydrogel systems.

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Conflict of interest

The authors declare that they have no conflict of interest.

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