Conference Paper

Determination of Diffusion Coefficient in Hydrogel

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Abstract

Background Oriented Schlieren method was used to obtain diffusion coefficient of glucose in hydrogel in the experiment with radial symmetry.

Keywords: background oriented schlieren, diffusion, hydrogel, holographic sensors, glucose.

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1. Introduction

Hydrogels are polyacrylamide polymers that are long molecular chains with cross-links forming the bulk network. Due to their structure, hydrogels possess a very high degree of absorption. They are able to absorb water in amounts up to five hundred times of their own weight. Due to the similarity of physical properties of hydrogels with the properties of physiological tissues, they are widely used in medicine, in particular, in tissue engineering and as sustained-release of drug delivery system. If the cross-links of the hydrogel chains selectively interact with a given substance, the hydrogel swells or contracts upon contact with the corresponding components of the aqueous solutions. This allows one to design holographic sensors by recording in the hydrogel a thick-layer hologram that reflects only the component resonant to the period of the layers when the white light is directed onto it [1-3]. When the sensor shrinks or swells, the wavelength of the reflected radiation changes, which makes it possible to measure the concentration of that component of the solution to which the crosslinking of the hydrogel matrix is "tuned." When designing a sensor, it is important to select its parameters to ensure the correct operation mode and sufficient accuracy of measuring the wavelength of the reflected light. The sensor is a thick-layer hologram, so it is important to know the penetration rate of the test solution into the depth of the hydrogel matrix.

2. Materials and methods

To determine the diffusion coefficient of glucose in the hydrogel, a model experiment was performed with a hydrogel matrix without glucose sensitive crosslinks. The matrix does not swell or contract, and the penetration rate of glucose is estimated from the change in the refractive index of the hydrogel using Background Oriented Schlieren method [4]. This method does not require precise tuning of optics and is very convenient for studying processes in optically transparent media [5-7]. In the case of slow processes, a conventional consumer camera can be used as the recording equipment [8].

The essence of the method is as follows. A transparent optically heterogeneous object, in this case a cuvette with a hydrogel, is installed between the camera and a flat screen with random filling with black and white spots similar to the laser speckle structure. In the process of changing optical inhomogeneities, the screen shots are taken through the cuvette. The shooting frequency depends on the speed of the changes, in this case the photos were taken about once every 10 minutes. The scale of the "artificial speckles" of the screen was chosen so that the size of one information element on the camera's matrix was 2.5 - 3 pixels. This size is optimal from the point of view of the signal-to-noise ratio [9]. The deviation of the beam caused by its passage through the cuvette leads to a shift of the image of the corresponding area of the random pattern by a certain distance Δx in the plane of the camera sensor. Assuming that the deviation angles of the rays forming the image from the optical axis are small, one can calculate the gradient of the refractive index of the contents of a flat cuvette from the displacement Δx

$$\frac{dn}{dx} \approx \Delta x \frac{(L-f)}{lfd}, \quad (1)$$

where L is the distance from the random screen to the camera, l is the distance from the screen to the cuvette, f is the focal length, and d is the thickness of the cuvette. The distribution of the displacement Δx along the plane of the cuvette was determined by the correlation scanning of pairs of photographs, the reference one made through the cuvette filled with water only, and the other one made at a certain time with the hydrogel. The scheme of photographic recording is shown in Fig. 1. The hydrogel was placed in the cuvette 5 cm high, the thickness d = 2.5 mm. The distance from the screen to the cuvette was l = 28 cm, and the distance from the screen to the camera L = 53 cm. The camera Canon SX700 was used, and the focal length when shooting was 15 mm.

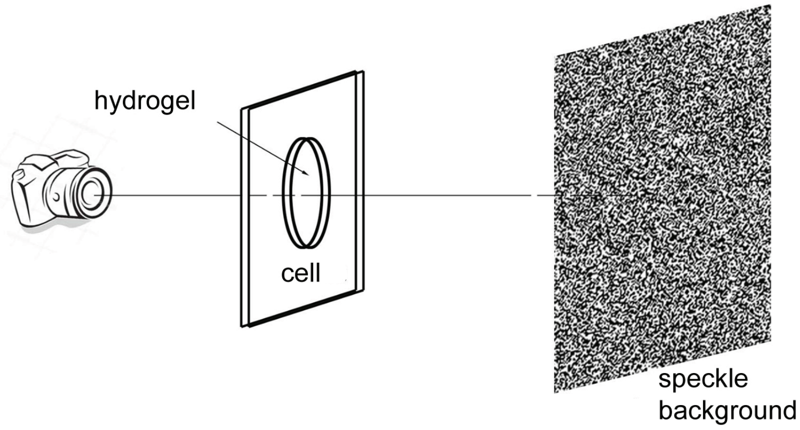

Figure 1: Schematic diagram of diffusion process shooting.

The object was a tablet of dry polyacrylamide hydrogel 0.4 mm thick, 4.5 mm in diameter. The thickness of the sample was selected so that when the hydrogel was completely swollen, it rose up to the walls of the cuvette. In this case, the glucose solution penetrates into the hydrogel only through the lateral (cylindrical) surface. Samples of greater thickness when swollen are destroyed due to strong internal stresses. At the first stage of the experiment it is necessary to obtain the equilibrium state of the hydrogel disc in the distilled water. The process of swelling of the dry hydrogel takes about a day. The chart of the hydrogel diameter growth is shown in Fig. 2.

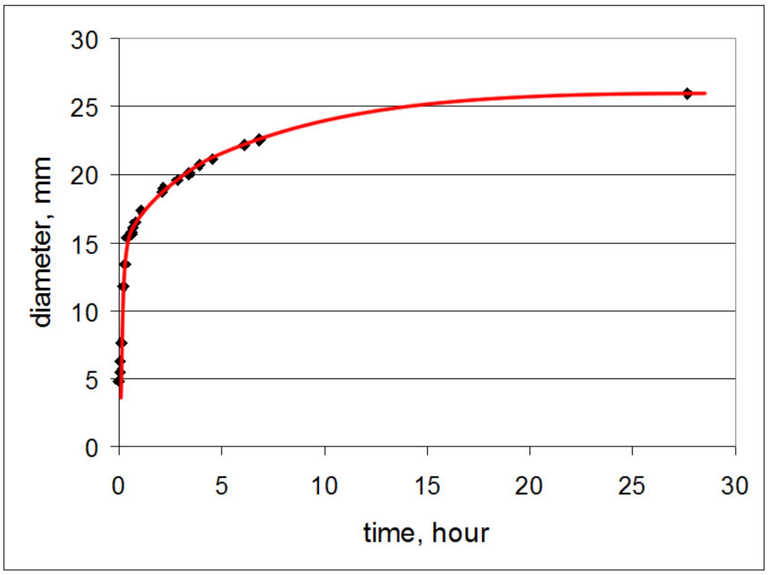

Figure 2: Dependence of the diameter of the hydrogel sample on time when swelling in distilled water.

During the first 20 minutes after immersion in the water, the hydrogel swells about three times. The sample is highly optically heterogeneous, and its internal structure can not be investigated by a Background Oriented Schlieren method (Fig. 3a). Further, the optical inhomogeneities are slightly smoothed out, and they can be measured. Figure 3b shows the results of the correlation measurements of the internal structure of the hydrogel during swelling. Maps of the refractive index derivative along the horizontal coordinate are given. The vertical component is given as an illustration for one of the moments (Figure 3c), its structure and time behavior are the same as those of the horizontal component. It can be seen that the optical density variations, sharp at first, are smoothed over time. More, the growth of the sample occurs unevenly. Structural features are most pronounced during the second hour of swelling. Further, the compressed areas between growth petals disappear one by one, and in the equilibrium state the structure disappears completely. The residual value (in the equilibrium state of the hydrogel) of the derivative of the refractive index is ~ $4*10^{-5}$ 1/mm, which, when integrated, gives the difference in the refractive index of the hydrogel saturated with water and water ~ $5*10^{-5}$.

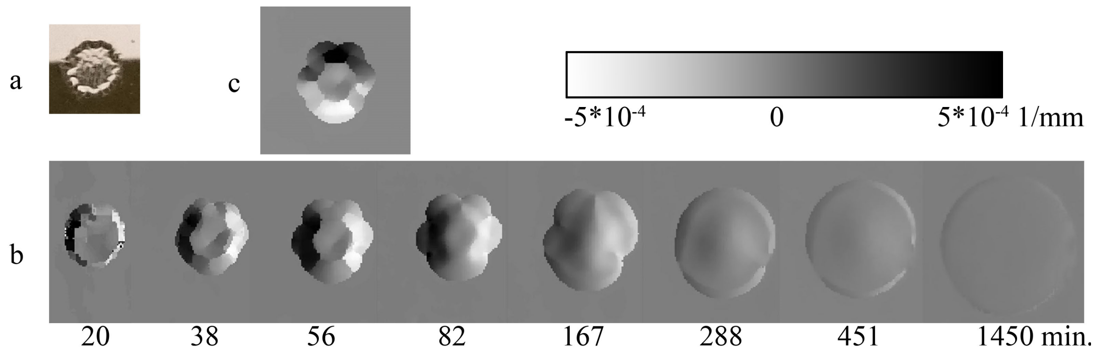

Figure 3: The process of hydrogel swelling. a) Photo of the sample 15 minutes after the swelling starts. b) Maps of the derivative of the refractive index along the horizontal coordinate for some moments of time. c) The vertical component of the refractive index derivative for t = 56 min.

After the hydrogel had reached equilibrium state, the water in the cuvette was replaced with a glucose solution (0.25 M / L), and this concentration was maintained outside the sample throughout the experiment. The solution contacted the hydrogel only along its cylindrical surface. The object was photographed for further correlation processing about two to three times per hour during several hours.

3. Results

By correlation processing of the pictures, maps of the derivative of the hydrogel refractive index were obtained for a set of time moments. Some of them, with the indication of the time passed from the start of diffusion process, are shown in Fig. 4. Similar to Fig. 3, the vertical derivative of the refractive index for one of the moments is given for illustration.

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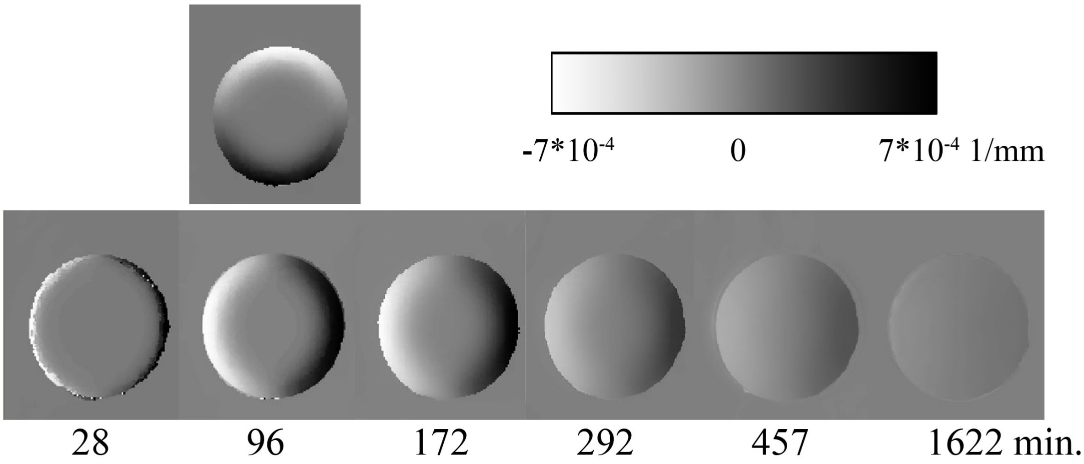

Figure 4: Maps of the derivative of the refractive index for the process of diffusion of glucose.

It can be seen that the picture has a radial symmetry. During the first three hours the sample has a decreasing central region with the constant refractive index. The graphs of the dependence of the derivative of the refractive index on the radius are shown in Fig. 5.

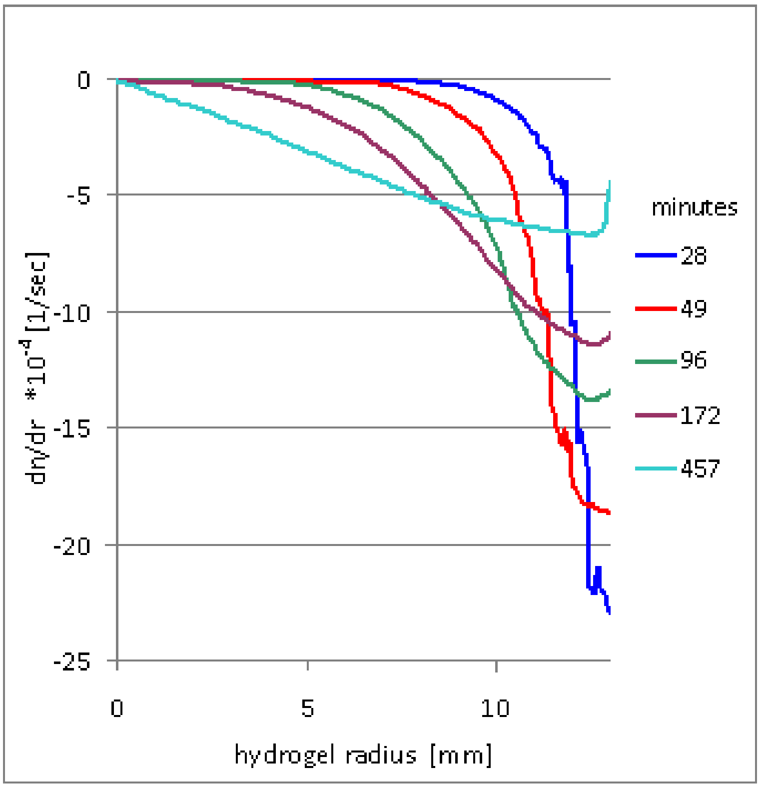

Figure 5: Dependence of the derivative of the hydrogel refractive index on the radius in the process glucose diffusion into the water-saturated hydrogel.

To calculate the diffusion coefficient of glucose in the hydrogel, we consider the diffusion equation in the case of radial symmetry

$$D\Delta n\left(\vec{r},t\right) = \frac{\partial n\left(\vec{r},t\right)}{\partial t} \tag{2}$$

with the boundary conditions

$$\frac{\partial n\left(0,t\right)}{\partial r} = 0 n\left(\vec{r},t\right)\Big|_{|\vec{r}|=a+\xi} = 0 \tag{3}$$

and the initial distribution

$$n\left(\vec{r},0\right) = \theta\left(a-r\right), \tag{4}$$

where a is the radius of the disk, and $\xi << a$. One can solve it with the method of separation of variables. Then, the radial solution is looked for in the form

$$n\left(r,t\right) = n_0 \sum_k A_k e^{-D\mu_k^2 t/(a+\xi)^2} J_0\left(\frac{\mu_k}{a+\xi} r\right), \tag{5}$$

where μ_k - are solutions to the equation

$$J_0\left(\mu_k\right) = 0. \tag{6}$$

The constants A_k are

$$A_k = \frac{2}{J_1^2\left(\mu_k\right)} \int_0^{\frac{a}{a+\xi}} J_0\left(\mu_k r\right) r dr \tag{7}$$

where J_0 и J_1 are Bessel functions. In the experiment, the refractive index derivatives are measured, so we compare the numerically calculated derivatives of expression (5) with allowance for (6) and (7). For different values of the parameter *Dt* in the exponential index (or, which is the same, for different time values at a constant diffusion coefficient), the derivatives of the refractive index along the radius look as shown in Fig. 6. The calculations were carried out taking into account the first twenty zeros of the Bessel function. Since the time for each experimental curve is fixed, the diffusion coefficient *D* remains the only fitting parameter, except the scaling factors. The fit was performed over the region $0 < r < 0.9\ a$ to avoid the area at the disk boundary. In this area, with a large change of refractive index gradient on small scale, the correlation calculations may provide the data not reliable enough.

As a result, for various times, the diffusion coefficient for a given glucose concentration was calculated (see Table 1). The value $D \sim 5{*}10^{-6}$ cm^2/s is obtained.

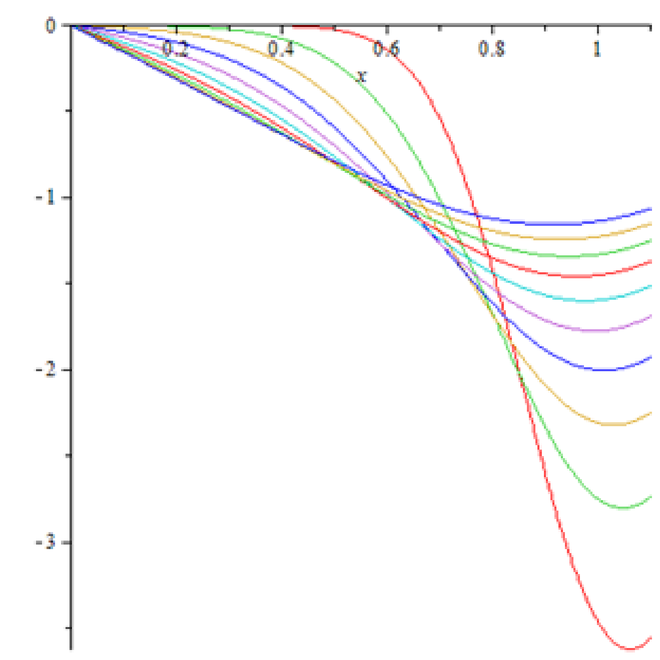

Figure 6: The result of an approximate calculation of the derivative of the refractive index.

Table 1: Values of the diffusion coefficient.

t, minutes	D, cm^2/sec
148	4,9482 E -06
172	4,99467 E -06
292	5,01598 E -06
457	4,93071 E -06
1622	5,20962 E-06
Mean value	5,02 E-06

4. Conclusion

The solution of the diffusion equation with radial symmetry gives the dependence of the derivative of the refractive index on the radius for different times from the diffusion start. By adjusting the parameters, the diffusion coefficient was calculated for a given glucose concentration. The value $D \sim 5*10^{-6}$ cm^2/s is obtained which practically coincides with the diffusion coefficient obtained for diffusion of glucose solution and water in the experiment carried out by the method described in [10]. Thus, this type of hydrogel has practically no effect on the diffusion rate.

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