Numerical modelling and experimental study of liquid evaporation during gel formation

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Abstract. Gels are promising materials in biotechnology and medicine as a medium for storing cells for bioprinting applications. Gel is a two-phase system consisting of solid medium and liquid phase. Understanding of a gel structure evolution and gel aging during liquid evaporation is a crucial step in developing new additive bioprinting technologies. A numerical and experimental study of liquid evaporation was performed. In experimental study an evaporation process of an agarose gel layer located on Petri dish was observed and mass difference was detected using electronic scales. Numerical model was based on a smoothed particle hydrodynamics method. Gel in a model was represented as a solid-liquid system and liquid evaporation was modelled due to capillary forces and heat transfer. Comparison of experimental data and numerical results demonstrated that model can adequately represent evaporation process in agarose gel.

1. Introduction
Development of additive technologies, including areas such as medicine and biotechnology requires an understanding of the dynamics of the processes of formation and aging of gels with the aim of developing optimal technological processes for bioprinting technologies [1]. The formation of gels occurs due to the complex hydrodynamic and thermo-physical processes occurring in the internal micro-structure of the gel. The processes of mass transfer in the gel involve the interaction of multiple phases on small linear scales [2]. In order to determine the dynamics of the process of liquid evaporation during gel formation, experimental and numerical study of measuring the rate of evaporation of the liquid phase in the course of cooling of the sample gel was performed.

Currently gels are considered a high-potential structure-forming material for formation of artificial tissues from immobilized cells using the method of additive manufacturing. Due to their rheological properties, gels refer to an intermediate state between liquid and solid. The common definition of gels is a dispersive system with liquid dispersing medium, and the dispersion phase makes up a spatial structured mesh due to intermolecular interaction in the contact areas [3]. Gels are capable of displaying both elasticity and plasticity.

Dynamics of gas liquid flow inside irregular channels and gels are not studied enough, both experimentally and numerically. Gel is a specific two-phase system. One phase represents a relatively sparse spatial network of polymer molecules which are bonded at the intersections of intermolecular
bonds. The second phase is a liquid. In the study of additive processes of application of gels in relation
to problems of bioprinting, we found the effect of microchannels formation between the layers of gel,
filled with gas and fluid.

In the case of the presence of microorganisms within the gel, a carbon dioxide is produced within
the gel due to activity of microorganisms. For the normal functioning of microorganisms in the gel it is
required a withdrawal of produced products. In this case, the rate of drainage is influenced by the
intensity of absorption and the speed of ascent of gas bubbles, which leads to the necessity of
conducting experimental and theoretical studies of the process of mass transfer of gas bubbles in the
gel. The relevance of such studies is also due to the active development of technologies of three-
dimensional printing of living tissues and bodies [1].

2. Numerical model
Model of a gas phase moving inside gel was developed based on the smoothed particle hydrodynamics
method. The method is based on representing the fluid as a set of particles, which are located at some
distance from each other, the distance called smoothing [4]. The influence of each particle on the
properties of nearby particles is calculated on the basis of its density and the distance from the particle
for which parameters. Evaporation rate was calculated by procedure proposed in [5]. First, vapour
pressure is calculated by applying ideal gas equation, then saturation pressure with a given surface
temperature can be calculated using Magnus formula, finally evaporation rate is computed by Smith
formula [5].

For the calculation we introduce the notion of kernel functions. As a kernel function, typically uses
a Gaussian function. In the case of motion of a gas bubble of carbon dioxide in the channel with
obstacles (figure 1). For verification of the developed model was used for calculation of test problem
on simulation of freely rising gas bubble in a pure liquid at various angles of inclination of the tube for
the purpose of comparison with previously obtained experimental data.

Mathematical model of a liquid flow was based on smoothed particle hydrodynamics method. Method
is based on representing liquid as an ensemble of smoothed particles [4]. Influence of each particle on neighborhood particles is calculated based on their density and distance to other particles (1).

\[
A(r) = \int A(r') W(r - r', h) dr'.
\]  

(1)

In discrete representation the value of an arbitrary point is presented in the following form (2):

\[
A(r) = \sum m_i A_i \frac{\Delta}{\rho_i} W(|r - r^i|, h).
\]  

(2)

For pressure (3) and velocity (4) calculation the following difference scheme is applied:

\[
p_i = -\sum m_j \frac{p_j - p_i}{\rho_j} W(|r_i - r_j|, h),
\]  

(3)

\[
v_i = \mu \sum m_j \frac{v_j - v_i}{\rho_j} \nabla W(|r_i - r_j|, h).
\]  

(4)

Evaporation condition is defined as follows (5)

\[
ev = \max \left( \frac{\partial m_s}{\partial \tau} \right)
\]  

(5)
\[ \frac{\partial m_s}{\partial \tau} = S \left( a + b \cdot \|v\| \right) (p_{\text{sat}} - p_v), \]

where \( p_{\text{sat}} \) is the saturation vapor pressure, \( p_v \) is the vapor pressure, \( S \) is the surface of liquid-air contact, \( v \) is the velocity, \( a, b \) are arbitrary parameters [5].

The system of equations (3), (4), (5) are solved jointly for each particle over the period of time of the numerical experiment.

3. Experiment

3.1. Experimental setup

In the experimental study an agarose gel with concentration of 0.6 and 1% was used. At the beginning of the experiment, a prepared sample of the gel at a temperature of 60 °C was applied to the surface of the Petri dish. Then Petri dish was mounted on a heat insulating plate and installed at the scale. Further thermopair was injected into the gel. Before starting the experiment a timer was set to measure time of experiment. Differences in values presented by thermopair measurements and electronic scales were recorded in the table with the corresponding time value according to the timer. The experimental setup is shown in figure 1.

![Figure 1. Experimental setup: 1 – layer of agarose gel, 2 – thermopair, 3 – Petri dish, 4 - heat-insulating plate, 5 – electronic scale, 6 – vapour flux, 7 – Multimeter Fluke 117](image)

Thermopair was connected to the multimeter Fluke 117 which supports temperature measurements. Thermopair could be located in arbitrary area of a Petri dish. After series of experiments it was found that temperature distribution among the plate can be considered homogenous and hence thermopair location can be arbitrary. However thermopair is not recommended to locate near the dish wall in order to avoid boundary effects. The thickness of the gel layer was measured by moving thermopair while considering the bottom of the plate as a reference point. The moment when thermopair reached the surface of the gel was fixed by drop in a temperature when thermopair leaved the gel since there is a difference between temperature in gel and atmosphere.

The estimated model of the process of evaporation is based on the idea of considering gel as a system of capillaries. The motion of the liquid within the gel is associated with the movement of the liquid phase in microchannels of complex configuration, where capillary force provides a supply of fluid from the inner layers of the gel to the surface during evaporation [6]. The density of capillaries
per unit surface area was determined using qualitative analysis of photographs of the microstructure of the gel obtained by electron microscopy [7].

3.2. Experimental study

Liquid evaporation from gel was observed for 2 hours and 40 minutes. This time interval allows to capture the characteristic of stage of formation and aging of the gel, including the initial stage of structure formation and polymerization. Time step was set to 5 minutes. The data obtained is the dependence of change of weight of gel over the time (figure 2). Data is presented in normalized form where normalized time is current time in experiment divided by end time of an experiment. In this case 1 corresponds to a fully formed gel. Relative mass is mass divided by initial mass of a gal filled with liquid.

Figure 2. Dynamics of gel evaporation with dependence from time for different gel concentrations

In the first 20 minutes of the experiment there is a linear dependence of change of weight of gel over the time. Dependence may be associated with the fact that in this period liquid evaporation occurs from the surface of the gel, with the supply of fluid from the deep layers of the gel by the action of the capillary forces. In the interval from 20 minutes to 2 hours, dependency becomes nonlinear, which may occur due to the action of additional factors on the evaporation process such as the process of shrinkage of the gel. After 2 hours stabilization of the evaporation process was observed. The cause of stabilization can be explained by the fact that the remaining liquid is not enough for the action of capillary forces to supply the liquid to the surface of the gel, which leads to the fact that the evaporation process continues within the pores, where the total surface evaporation is less in comparison with the gel surface.

4. Discussion

Temperature measurements in time allowed to identify the mode of establishing a stationary temperature, in which case mathematical model was applied (figure 3).
Figure 3. Dynamics of gel evaporation with dependence from time for different gel concentrations

For this mode a numerical calculation of the dynamics of evaporation of the liquid was performed and a comparative analysis with experimental results for agarose gel concentrations of 0.6% and 1% was made. The variation of the mass of the gel is of a linear nature in the area of stationary temperature. Evaporation rate for a given concentrations does not vary significantly. The similarity of the nature of evaporation for these concentrations may be related to the fact that the difference in concentration is insufficient for the occurrence of changes in the internal structure of the gel.

5. Conclusions

Three stages of evaporation of the gel were identified. These stages are: linear which corresponds to the gel formation process, nonlinear, associated with the action of capillary forces and shrinkage of the gel, and the stage of polymerization characterized by plateau. The comparison of results of calculation and experiment shows the effectiveness of the model for the initial stage of gel formation. The character of dependencies of evaporation of the gel over the time at the stage of gel formation demonstrates a linear character, the dynamics of evaporation agarose gel with a concentration of 0.6% differs from the concentration of 1% gel insignificantly, which allows to make a conclusion about the similarity of the internal structure of the gel at these concentrations.

Acknowledgments

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