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## THE SOL-GEL TRANSITION IN HYDROGELS AS THE FIRST-ORDER PHASE TRANSITION

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*Thermodynamic features of the sol-gel transition have been considered. It is found that the sol-gel transition in hydrogels is the first-order phase transition. Using the classical theory of phase transformations, the kinetics of sol-gel transition in various regimes is analyzed. It is shown that, under certain conditions, the sol-gel transition can acquire features characteristic of the percolation transition.*

*Keywords:* sol-gel transition, hydrogel, first-order phase transition, percolation.

### 1. Introduction

Among polymers and biopolymers that are used in many production areas cellulose derivatives are the most applicable today [1]. Cellulose is the most widespread natural polymer. Its structure is represented by thread-like fragments of d-glucopyranose [2]. Cellulose is a hydrophilic material [3, 4]. However, it is insoluble in water and most organic solvents [5]. It occurs due to the presence of strong intramolecular and intermolecular hydrogen bonds between the individual chains. To increase its solubility, cellulose undergoes a chemical modification, the esterification of hydroxyl groups in the cellulose chain. This enables the production of a large class of cellulose derivatives with unique properties [6].

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The studies of hydrogels created on the basis of water-soluble cellulose derivatives have gained rapid development in recent decades owing to the extensive application of these materials in industrial technologies and biomedicine [7–11]. An important role in those applications has information about the factors that affect the gel formation, namely, information about the mechanism of the sol-gel transition. This circumstance proves the challenging character of the problem considered in this article.

Below, we will use the terminology adopted in work [12].

- The term “gel” has two meanings. It is used to refer to a network (a framework) formed by polymer chains. It is also used as the name of a structure in a polymer solution, where the chains form a framework. Below, when using the term “gel”, we mean exactly the second variant.

- The term “sol” is the name of a structure of a polymer solution where the chains do not form a framework.

- The term “sol-gel transition” is used as the name of a structural transition in a polymer solution accompanied by the formation of a framework.

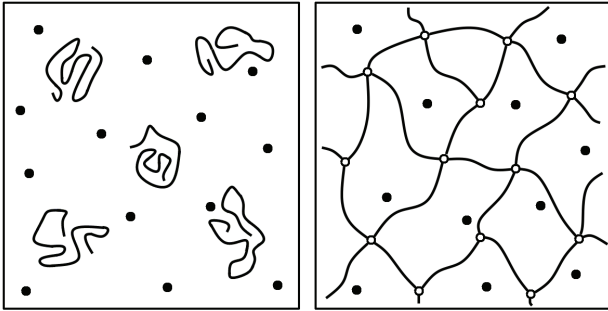


Fig. 1. Structure of polymer solution: sol (a), gel (b)

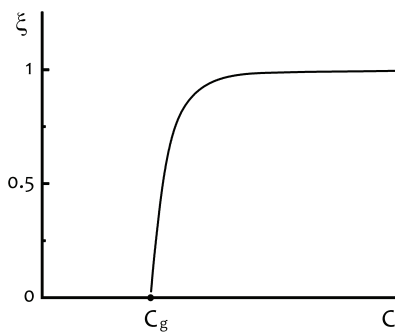


Fig. 2. Dependence of the relative gel volume  $\xi$  on the solution concentration  $C$ , provided that the sol-gel transition is a percolation one

The sol and gel structures are schematically depicted in Fig. 1. In this figure, the polymer chains are drawn as solid curves, the solvent molecules are shown as solid circles, and the network nodes (spatial regions where the chains are connected with one another) are exhibited as hollow circles.

Nowadays, it is generally accepted to describe the gelation process in the framework of the approach based on the percolation theory [13–15]. Most of the results of this theory were obtained with the help of computer simulations and using Monte Carlo algorithms [16–18].

The application of the percolation model [12, 13, 19] makes it possible to obtain the dependence of the relative gel volume  $\xi$  on the solution concentration  $C$ . Figure 2 illustrates the general behavior of this dependence. As one can see from Fig. 2, according to the percolation theory, the gel is formed at solution concentrations corresponding to the inequality

$$C \geq C_g, \tag{1}$$

where  $C_g$  is the so-called percolation threshold, i.e., the concentration at which the gel forms an infinite cluster.

At the same time, the percolation theory does not allow the sol-gel transition to be described at length. The matter is that the researched transition, like any other structural transformation, is affected not only by the concentration  $C$  but also by the temperature  $T$  and other external parameters (for example, the pressure  $p$ ). Furthermore, gelation is a process that evolves in the time  $t$ . Thus, the quantity  $\xi$  must be a function of many variables,

$$\xi = \xi(p, T, C, t) \tag{2}$$

rather than a function of only one variable,  $C$ , as is predicted by the percolation theory. Taking into account that this is dependence (2) that holds in reality, we propose another approach to describe gel formation, which differs from that adopted in the literature. The fundamentals of this approach were laid in the previous work [20], where a continuum model of the phase transition of the first kind was developed. The model is based on the ideas of the classical theory of phase transformations [21]. A general formula was obtained that relates the relative volume of the initial phase to the temperature that changes linearly with time.

The presented work is a continuation of work [20]. Its novelty consists in that we show that the sol and gel-structures of a solution are different thermodynamic phases, and accordingly the sol-gel transition is a phase transition of the first kind. This result allows the classical theory [21] to be used while describing gelation and analyzing the kinetics of the sol-gel transition in various regimes.

## 2. Thermodynamic Features of the Sol-Gel Transition

From the topological viewpoint, the configuration formed by gel chains is connected, whereas the configuration formed by sol chains is disconnected. Therefore, it is impossible to transform either of those configurations into the other using a continuous transformation. This fact means that the thermodynamic characteristics of the polymer solution – namely, the first derivatives of the thermodynamic potential – do not change continuously at the sol-gel transition, and the indicated derivatives have a

break. Such a behavior, as is known, means that the sol-gel transition is the first-order phase transition.

In the sol, as in any liquid, there are regions with the short-range order. Let  $\ell'$  denote the size of such regions. As compared to the sol, the gel is a more ordered phase, which is associated with the formation of nodes. Indeed, for a link in the sol, the number of its nearest neighbors along the chain equals two. At the same time, the corresponding number for a given node in the gel equals three, four, or more. This circumstance leads to the appearance of short-range regions in the gel with the size  $\ell'' \gg \ell'$ .

Below, when describing the kinetics of the sol-gel transition, we use the spatial scale with the scale  $\Delta\ell$  corresponding to the condition

$$\ell' \ll \Delta\ell \ll \ell''. \quad (3)$$

This choice of the spatial scale allows us to consider the sol as a completely disordered system.

Let  $\xi$  denote the relative volume occupied by the gel. Before the transition, when the system consists exclusively of the sol,  $\xi = 0$ . After the transition,  $\xi = 1$ . The quantity  $\xi$  plays, in this case, the role of the order parameter, and the gel is considered as a completely ordered system. Therefore, as it should be for phases with the transition of the first kind between them, the values of the order parameter in the sol and the gel differ by a finite magnitude, which is equal to unity in this case.

Let  $p$  denote the pressure, and let  $T$  denote the temperature of the system. By definition, the equilibrium thermodynamic potentials of the sol,  $\Phi_s$ , and the gel,  $\Phi_g$ , have to be functions of the variables  $p$ ,  $T$ , and  $C$ :

$$\Phi_s = \Phi_s(p, T, C), \quad (4)$$

$$\Phi_g = \Phi_g(p, T, C). \quad (5)$$

Consider the case where the sol-gel transition occurs at  $p = \text{const}$ . In work [22], a phase diagram for the “polymer–solvent” system, which corresponds to this case, was proposed. Figure 3 demonstrates a fragment of this diagram. In the  $T - C$  diagram, the region confined by the curve OABH corresponds to the sol, and the region confined by the curve DEF corresponds to the gel. In other words, function (4) is defined in the former domain, and function (5) in the latter one. Both phases – the sol and the gel – coexist in the region HBED.

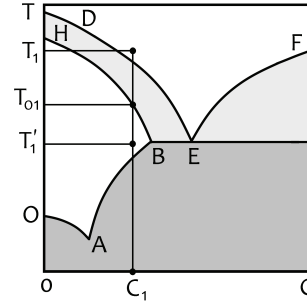


Fig. 3. Fragment of the phase diagram of the “polymer–solvent” system

Every value  $\xi < 1$  characterizes a certain non-equilibrium state of the solution. According to the definition [23, 24], the thermodynamic potential  $\Phi$  of such a state is a function of the variables  $p$ ,  $T$ ,  $C$ , and  $\xi$ :

$$\Phi = \Phi(p, T, C, \xi). \quad (6)$$

The thermodynamic potential is the thermodynamic function that characterizes the equilibrium state. It becomes possible to ascribe a certain value of the thermodynamic potential to a non-equilibrium state if this state is relatively stable; this is a metastable state, i.e., a state of incomplete equilibrium.

Similar considerations are applicable to the temperature because the temperature is determined for an equilibrium state. Hence, when the matter concerns the temperature of the system (in our case, the polymer solution) in a non-equilibrium state, it is obvious that this state must be a state of incomplete equilibrium.

Let us seek an answer to the following question: what is the state of incomplete equilibrium at the molecular structural level? When trying to answer it, we use a molecular model that corresponds to the adiabatic approximation of quantum mechanics [25]. In the framework of this model, solvent molecules and the links of the polymer chain are considered as structureless particles (point-like force centers). The model assumes the existence of equilibrium in the electronic subsystem at any time moment. This statement defines the time scale in which the indicated model “works”. This is the scale  $t'$  with the increment  $\Delta t'$  satisfying the condition

$$\Delta t' \approx \tau', \quad (7)$$

where  $\tau'$  is the time required for the electronic subsystem to reach a stationary state (the relaxation time).

Let  $N$  denote the number of particles in the polymer solution, and  $X \equiv \{x_j\}$  be a  $3N$ -dimensional vector whose components are the Cartesian coordinates  $x_j$  of the particles. As a result of thermal motion, the vector  $X$  randomly changes in time. In the indicated model, as is known, there are two types of thermal motion of particles: oscillatory and diffusive. Accordingly, the vector  $X$  can be presented in the form

$$X = X_0 + U, \tag{8}$$

where the summand  $X_0$  is determined by the diffusive motion, and the summand  $U$  by the oscillatory one.

It is also known [21] that the diffusion is a sequence of elementary acts of particle motion. The mechanism of every act consists in that a particle (or a group of particles, i.e., a kinetic unit) overcomes an energy barrier with the help of thermal fluctuations and moves to a certain distance, which is called the elementary displacement. The average duration of an elementary act  $\tau$  is determined by the formula

$$\tau = \tau_0 \exp\left(-\frac{\Delta F}{k_B T}\right), \tag{9}$$

where  $k_B$  is the Boltzmann constant,  $T$  is the temperature,  $\Delta F$  is the free energy of activation, and  $\tau_0$  is a characteristic time whose order of magnitude is considered to be equal to the period of thermal fluctuations.

It is obvious that, during the time interval  $\tau$ , the vector  $X_0$  remains unchanged. It is reasonable to describe the behavior of this vector in time by considering this vector as a function

$$X_0 = X_0(t) \tag{10}$$

and using the time scale  $t$  with the increment  $\Delta t$  that satisfies the condition

$$\Delta t \approx \tau. \tag{11}$$

Formula (9) was derived under the condition

$$\frac{\Delta F}{k_B T} \gg 1, \tag{12}$$

whence the inequality

$$\tau \gg \tau_0 \tag{13}$$

follows. This inequality means that a significant number of particle vibrations occur during the time interval  $\tau$ . Therefore, remaining unchanged within the time interval  $\tau$ , the vector  $X_0$  actually determines the equilibrium positions of the particles around which the vibrational displacement  $U$  takes place.

According to inequality (13), the time scale with the increment  $\Delta t$  cannot be used when the vibrations are dealt with. In this case, as a result of the obvious inequality

$$\tau' \ll \tau_0, \tag{14}$$

a time scale with the increment  $\Delta t'$  has to be applied and, accordingly,  $U$ , has to be represented in the functional form

$$U = U(t'). \tag{15}$$

It is known [26] that the temperature concept is valid, if there is a Maxwellian distribution of particles over their velocities in the system. Let  $\tau_M$  denote the time needed for this distribution to be reached. According to work [27], in the case of liquids,  $\tau_M$  is the time required for the equilibrium with respect to the vibrational degrees of freedom to be reached, i.e., the time during which the random function  $U(t')$  becomes almost stationary.

According to formula (9), an elementary act of diffusion can take place only if a certain temperature value is established in the system. In other words, for this act to be realized, the following inequality must hold:

$$\tau_M \ll \tau. \tag{16}$$

It gives an answer to the question posed above concerning the state of incomplete equilibrium that arises in the course of the sol-gel transition process. According to formula (16), the state of incomplete equilibrium is an equilibrium state with respect to the variable  $U$ , i.e., the equilibrium state of the vibrational subsystem. Accordingly, the solution temperature at the sol-transition process is a temperature that characterizes the mentioned state of the vibrational subsystem, and the nonequilibrium thermodynamic potential  $\Phi$  is the thermodynamic potential of the indicated state.

Every equilibrium state of the vibrational subsystem corresponds to a certain vector  $X_0$ . In turn, the

latter corresponds to a “specific” value of the parameter  $\xi$ . Therefore, we have the following expression for the potential  $\Phi$ :

$$\Phi = E(p, T, \xi) + F(p, T, \xi) + pV(p, T, \xi), \quad (17)$$

where  $E$  is the energy of particles at their equilibrium positions,  $V$  is the system volume, and  $F$  is the free vibrational energy determined by the well-known formula [28]

$$F = k_B T \sum_{j=1}^{3N} \ln \left( 2 \sinh \frac{\hbar \omega_j}{2k_B T} \right), \quad (18)$$

where  $\hbar$  is Planck’s constant, and  $\omega_j$  is the  $j$ th eigenfrequency of vibrations in the system.

### 3. Kinetics of the Sol-Gel Transition at the Constant Solution Temperature and Concentration

The assertion that the sol and the gel are different thermodynamic phases allows the classical theory of phase transitions of the first kind to be used when describing the kinetics of the sol-gel transition [21]. In this theory, phases A and B with the same chemical composition are considered. Let the letter A denote the sol, and the letter B the gel. The requirement of the same chemical composition of both phases means that the polymer concentrations in them must be identical. Let this concentration be equal to  $C_1$  (see Fig. 3). At this concentration, as one can from Fig. 3, the phase transition A  $\rightarrow$  B occurs at temperatures exceeding  $T_{01}$  (for example, at the temperature  $T_1$ ). In work [21], the case where the temperature remains constant during the phase transition was considered. Obviously, in this case, the concentration always remains equal to  $C_1$ .

According to work [21], the mechanism of the A  $\rightarrow$  B phase transition is as follows. As a result of thermal fluctuations, nuclei of phase B are formed in phase A. Their formation leads to a change in the thermodynamic potential of the solution by a certain value  $\Delta\Phi$ . In work [21], these nuclei were assumed spherical, which led to the formula

$$\Delta\Phi = -(\phi' - \phi'') \frac{4\pi R^3}{3} + \sigma 4\pi R^2, \quad (19)$$

where  $R$  is the nucleus radius;  $\sigma$  is the surface tension at the boundary between phases A and B;  $\phi'$  and  $\phi''$

are the densities of the thermodynamic potentials of phases A and B, respectively, and their difference is determined by the equality

$$\phi' - \phi'' = \lambda \frac{T_1 - T_{01}}{T_{01}}, \quad (20)$$

where  $\lambda$  is the heat of the A  $\rightarrow$  B phase transition per unit volume.

As one can see from formula (19), at the radius value

$$R^* = \frac{2\sigma T_{01}}{\lambda(T_1 - T_{01})}, \quad (21)$$

the function  $\Delta\Phi(R)$  has a maximum equal to

$$\Delta\Phi^* = \frac{16\pi}{3} \frac{\sigma^3 T_{01}^3}{\lambda^2 (T_1 - T_{01})^2}. \quad (22)$$

According to the thermodynamics of irreversible processes [23], if  $R < R^*$ , the size of the nucleus should decrease spontaneously, and if  $R > R^*$ , it should grow spontaneously, where  $R^*$  is the critical nucleus radius.

According to work [21], the kinetics of the phase transition of the first kind is governed by two processes: the fluctuation-driven formation of critical nuclei and their subsequent growth. Let  $v$  denote the linear growth rate of the nucleus, and  $n$  denote the number of nuclei of critical size that are formed in a unit volume per unit time. The quantity  $n$  is determined by the formula [21]

$$n = n_0 \exp \left\{ -\frac{\Delta\Phi^*}{k_B T} \right\}, \quad (23)$$

where  $n_0$  is a temperature-independent constant.

For further transformations, let us apply the theoretical model of the kinetics of the phase transition of the first kind that was proposed in work [20]. This model is based on the assumption that the probability of the formation of a nucleus in a certain volume  $\Delta V$  within the time interval  $\Delta t$  is proportional to the product  $\Delta V \Delta t$ . Then, the dependence of the relative amount  $\xi$  of the formed phase B on the time  $t$  at a constant solution temperature and a constant solution concentration should be determined by the formula

$$\xi = 1 - \exp \left\{ -\frac{\pi n v^3 t^4}{3} \right\}. \quad (24)$$

#### 4. Kinetics of the Sol-Gel Transition at the Varying Solution Temperature and the Constant Solution Concentration

The hypothesis that the sol-gel transition is a phase transition of the first kind was tested in works [20, 29]. In work [20], using the approach adopted in works [21, 28], the kinetics of the phase transition under the condition of the linear temperature growth,  $T = T_{01} + \mu t$ , was considered. The dependence of the nucleus volume  $V'$  on the time  $t$  was written in the form

$$V'(t) = at^\alpha, \quad (25)$$

where  $a$  and  $\alpha$  are certain parameters.

For the temperature dependence of  $\xi$ , the following formula was obtained:

$$\xi = 1 - \exp\left\{-D(T - T_{01})^{3(1+\alpha)} \exp\left[-\frac{M}{(T - T_{01})^2}\right]\right\}, \quad (26)$$

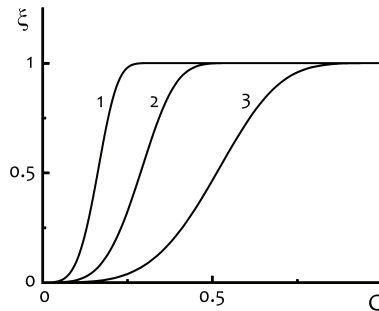
where the notations

$$D = 2an_0e^{-\alpha} \sqrt{1 - 2^{-1/\alpha}} \left[\frac{\alpha}{2\mu M}\right]^{1+\alpha}, \quad (27)$$

$$M = \frac{16\pi\sigma^3T_{01}}{3k_B\lambda^2} \quad (28)$$

were used.

Basing on experimental results [29], it was found that the dependence  $\xi(T)$  for the sol-gel transition in hydrogels prepared on the basis of hydroxypropyl cellulose is described by formula (26). This fact confirms the thesis that the sol-gel transition is a phase transition of the first kind.



**Fig. 4.** Dependences of the relative gel volume  $\xi$  on the solution concentration  $C$ , provided that the sol-gel transition is a phase transition of the first kind, for various values of the parameter  $\beta = 10^3$  (1),  $10^2$  (2), and 10 (3)

#### 5. Kinetics of the Sol-Gel Transition at the Constant Solution Temperature and the Varying Solution Concentration

Let the solution temperature be  $T_1$ , and the solution concentration changes from 0 to  $C_1$  during the sol-gel transition (see Fig. 3). For the time dependence of concentration, let us introduce the notation

$$C = C(t). \quad (29)$$

The rate of nucleus formation per unit volume,  $n$ , and the rate of linear growth of a nucleus,  $v$ , depend on the solution concentration, so they must also be some functions of the time

$$n(t) = n(C(t)), \quad v(t) = v(C(t)). \quad (30)$$

Following the model from work [20], we write down

$$\xi(t) = 1 - \exp\left\{-\frac{4\pi}{3} \int_0^t n(p) [v(p)p]^3 dp\right\}. \quad (31)$$

Formula (31) represents the quantity  $\xi$  as a function of the time. To change to the representation of  $\xi$  as a function of the concentration, we introduce the function inverse to function (29):  $t = \varphi(C)$ . We assume that there is a derivative of function (29):  $C'_t = f(t)$ , so that  $dC = f(\varphi(C))dt$ . After replacing the variable,  $p = \varphi(q)$ , in Eq. (31), we obtain

$$\xi(C) = 1 - \exp\left\{-\frac{4\pi}{3} \int_0^C v(q) [w(q)\varphi(q)]^3 \frac{dq}{f(\varphi(q))}\right\}. \quad (32)$$

Let us estimate the behavior of the function  $\xi(C)$ . Since a qualitative analysis is performed, let us simplify expression (32) by assuming that the dependence of the concentration on the time  $t$  is linear, i.e.,

$$C(t) = \eta t, \quad (33)$$

where the rate of concentration growth,  $\eta$ , is a constant, as well as the values of  $n$  and  $v$ :  $n = n_1$ ,  $v = v_1$ . Under such assumptions, formula (32) takes the form

$$\xi(C) = 1 - \exp\{-\beta C^4\}, \quad (34)$$

where

$$\beta = \frac{\pi n_1 v_1^3}{3\eta^4}. \quad (35)$$

In Fig. 4, dependences (34) are shown for various values of  $\beta$ . As one can see, at  $\beta = 10^3$  (curve 1), dependence (34) qualitatively agrees with the dependence presented in Fig. 2. According to formula (35), this coincidence is observed when the inequality

$$\eta^4 \ll n_1 v_1^3 \quad (36)$$

holds, i.e., if the polymer concentration slowly increases in the course of sol-gel transition. This regime can be realized, e.g., by slowly evaporating the solvent [30].

Hence, two seemingly incompatible statements, (i) the sol-gel transition is a percolation transition and (ii) the sol-gel transition is a phase transition of the first kind, do not contradict each other. Really, under condition (36), the dependence  $\xi(C)$  inherent to phase transitions of the first kind takes the form that is inherent to percolation transitions, i.e., the latter is a partial case of the first-order phase transitions.

## 6. Conclusions

The sol and the gel are different thermodynamic phases of polymer solution. This difference is a consequence of their topological nonequivalence. Owing to the latter, the continuity of the first derivatives of the thermodynamic potential becomes broken at the point of the sol-gel transition. This fact means that the sol-gel transition can be classified as a phase transition of the first kind. This circumstance makes it possible to apply the classical theory of phase transformations when describing the features of the sol-gel transition under various conditions, as was done in this article.

This classification does not contradict the generally accepted thesis that the “sol-gel” transition is percolative. Indeed, the sol-gel transition remains to be a phase transition of the first kind, but, in the case where the solution pressure and temperature remain constant during the sol-gel transition, whereas the rate of the solution concentration change is relatively low, also acquires features that are characteristic of the percolation transition. Therefore, the percolation transition is one of the possible forms of the phase transition of the first kind, those forms are different under different conditions.

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### ЗОЛЬ-ГЕЛЬ ПЕРЕХІД У ГІДРОГЕЛЯХ ЯК ФАЗОВИЙ ПЕРЕХІД ПЕРШОГО РОДУ

Розглядаються особливості термодинаміки золь-гель переходу. Встановлено, що золь-гель перехід у гідрогелях є фазовим переходом першого роду. Із використанням класичної теорії фазових перетворень проаналізовано кінетику золь-гель переходу в різних режимах. Показано, що за певних умов золь-гель перехід може набувати рис перколяційного переходу.

*Ключові слова:* золь-гель перехід, гідрогель, фазовий перехід першого роду, перколяція.