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G.A. Mun[1*], I. Moldakhan[2], A.M. Serikbay[1], D. Kaldybekov[1], I.E. Suleimenov[3], Kinam Park[4]

[1]Al-Farabi Kazakh National University, Almaty, Kazakhstan
[2]Almaty University of Power Engineering and Telecommunications, Almaty, Kazakhstan
[3]Institute of Information and Computational Technologies, Almaty, Kazakhstan
[4]Purdue University, West Lafayette, USA
[*]e-mail: mungrig@yandex.ru

Hydrophilic interpolymer associates – the key to solving the problem of pre-biological evolution

Abstract. A new approach to the analysis of the processes that led to the appearance of Life on Earth was proposed. The approach is based on the idea of the evolution of the Earth's shells as a system whole, which corresponds to the interpretation of the concept of "complex system" based on the analogy with neural networks. This analogy allows us to show that there are evolutionary mechanisms for which the primary one is the evolution of an analog of a neural network that is complementary to a specific complex system. The transformation of the system in this case takes place at the level of restructuring its structure, which at the next stage of evolution creates prerequisites for the "selection" of elements that correspond to the new state of the system as a whole. Experimental evidence of the validity of this approach for the interpretation of the origin of Life has been obtained based on the analysis of the formation of a comparative new class of macromolecular systems – hydrophilic interpolymer associates. It was shown that a solution of two interacting polymers, in which hydrophilic interpolymer associates are formed, is a direct analog of a neural network in a defined range of conditions. This object is capable of evolving as a whole, which proves the possibility of implementing an alternative mechanism of evolution considered in the framework of Darwinian theories.

Key words: complex system, polymers, interpolymer complexes, hydrophilic interpolymer associates, origin of life, neural network analog, evolution.

Introduction

Currently, active research is continuing to uncover the mystery of the origin of Life on our planet [1-3]. The authors of the vast majority of works carried out in this direction, either way, are guided by an approach that dating back to the theory of the origin of Darwinian's species [1-3].

However, there is every reason to believe that attempts to interpret the mechanisms of evolution that precede biological evolution, based on theories that go back to Darwin's point of view, cannot lead to success. This follows, among other things, from philosophical (methodological) judgments [4, 5]. Darwinian theories have faced and will continue to face insurmountable difficulties in trying to establish the mechanisms of evolution that led to the emergence of Life on Earth for fundamental reasons.

Darwin's theory the origin of species successfully describes the evolution of existing biological species, but is unable to adequately answer the question of where Life came from as a system whole. The last clarification (the systemic integrity of the phenomenon of Life) is extremely important. It is on this basis that even purely philosophical methods can show that mutational mechanisms are not able to give an adequate picture of the evolution that preceded the biological one.

Namely, Life exists in the form of ecosystems, therefore mutation mechanisms cannot be consistent even from the point of view of General philosophical concepts: they can explain the transformation of

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ecosystems, but they cannot explain the instantaneous emergence of an ecosystem with a rather complex and well-developed structure. Thus, the appearance of Life can only be the result of a certain qualitative leap (the emergence of a new quality), otherwise called aromorphosis, the prerequisites for this point of view are set out in well-known review papers [6, 7].

In [4, 5] it was shown that it is possible to propose a mechanism of evolution alternative to those that follow Darwinian theories. The corresponding theory is based on fundamentally new ideas about complex systems.

In the cited works, a hypothesis was put forward that a system of arbitrary nature should be interpreted as complex, when there is a complementary analog of a neural network. This means that for any complex system (a system that meets the category of complex in the philosophical sense), first of all, certain processes of information processing must be characterized that qualitatively different from the processes of information processing, characteristic to its individual components. The system (in the philosophical sense of this term) is something that qualitatively different from a simple set of its constituent elements – this idea is characteristic of the entire system approach, which goes back to the works of L. von Bertalanffy.

The proposed interpretation of the concept of "complex system" based on the analogy with neural networks, as well as the resulting philosophical understanding of the category of complex, indeed allow to propose a mechanism of evolution that is fundamentally different from Darwin's.

Indeed, a complex system can evolve even when the characteristics of its individual elements remain unchanged. In fact, this means that in this system, it is not the elements themselves that change, but the architecture of the relationships between them. The analogy with a neural network is important in that way because the rearrangement of links between system elements can be interpreted in the language of information theory as the evolution of an analog of a neural network that is complementary to the complex system under consideration.

We emphasize that this paper uses these representations. The evolution of an analog of the neural network that is complementary to a complex system of arbitrary nature suggests that a complex system could become a system for processing information even if it is composed of extremely simple elements. It is appropriate to emphasize that the existing artificial neural networks are also built from very simple elements that perform extremely simple functions, but when combined into a single system, they allow to implement a new quality, new information processing systems.

If a complex system experiences a quantitative and qualitative transition associated with the emergence of a qualitatively new information processing system, then it could be converted into a filter that creates preferential conditions for the existence or appearance (reproduction) of those elements that correspond to its qualitatively new state.

In other words, from the proposed point of view, the primary one is not the evolution of elements that are driven by mutations, but the evolution of the system as a whole; it can cause the very qualitative leap that, in particular, could lead to the emergence of Life as a system whole, i.e. already in the form of an ecosystem.

In other words, according to this point of view, Life on our planet arose due to a very specific complex system was able to process information in a very specific way.

The relationship between the philosophical interpretation of the category of information reflected in [8] and the proposed interpretation of prebiological evolution in this paper is as follows.

The simplest form of life, namely a virus, can be considered as a macromolecular object that performs a single function – the preservation of its genetic code, "its information". The virus contains a single informational macromolecule, and all the rest perform auxiliary functions. This example clearly shows that the origin of life can and should be interpreted purely from the position of information theory.

Further, as was shown in [9] based on the concept of dialectical positivism [8], information systems have a well-defined hierarchy.

What is called a message is an information object of the simplest type. At the next level are information systems, which are some rules for processing information. The simplest example of a more complex information system is a set of operating rules with binary numbers (addition, multiplication, etc.) or any other algorithm. At the highest level of the hierarchy of information objects is human intelligence, understood as a purely informational object (this object cannot be identified with the brain or processes occurring in it, just as you cannot identify a punched card and information written on it).

We emphasize that this work is not about identifying certain protocells, but about identifying the conditions when these protocells could have occurred. Returning to the idea of the existence of Life in the form of ecosystems, this means that the entire system as a whole is rebuilt and all the attributes of Life are created simultaneously in it. This, in particular, excludes the criticism that was addressed to Darwinian theories, and which was based on the extremely low probability of spontaneous occurrence of such molecules as DNA. From this point of view, this question is not worth it at all. It is not about the probabilities of random processes; it is about the evolution of the system as a whole.

Thus, to solve the problem of the origin of Life on our planet, it is necessary to show how such physical and chemical systems as solutions of hydrophilic macromolecules can be transformed into information processing systems. The choice of these systems as the base object is obvious: the main information macromolecules (DNA and RNA) are hydrophilic polymers.

In the present paper, we assume that the first step in this direction already has all the prerequisites. Namely, it is shown that there are quite certain conditions under which a solution containing two types of hydrophilic macromolecules interacting with each other is transformed into a non-trivial information processing system.

This transformation becomes possible due to the fact that hydrophilic interpolymer associates (HIA) could be formed in systems of this type [10, 11], and their formation can be accompanied by phase transitions of a stadial character. Experimental proof of the existence of staged phase transitions for the same system was given in [12] by the example of reactions between polyvinylcaprolactam (PVCL) and polyacrylic acid (PAA).

In this paper, the same system is experimentally studied and the conditions when phase transitions become stadial are analyzed. We emphasize that the multiplicity of phase transitions in such systems is quite easy to interpret from the point of view of the physical chemistry of polymers. Namely, in [10-12], it was shown that hydrophilic interpolymer associates are product that is intermediate by its properties with respect to both classical interpolymer complexes, true solutions, and cross-linked hydrophilic networks (Figure 1).

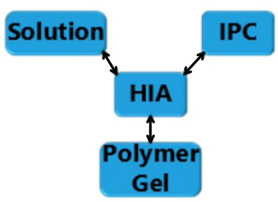

Figure 1 – An intermediate position of the HIA among the known macromolecular objects

The hydrophilic interpolymer associate (HIA), as shown in [11, 12], is an analog of a cross-linked polymer network and HIA is a dynamic system. In other words, the hydrogen bonds that stabilize it are disrupted constantly and appeared again.

Experimental

Materials

Polyacrylic acid (PAA) with molecular weights of 2×10^3; 2.5×10^5; 4.5×10^5; 7.5×10^5 and 1.079×10^6 Da was purchased from Sigma-Aldrich (St. Louis, MO, USA). Polyvinylcaprolactam (PVCL) was synthesized according to a report using free-radical polymerization technique [19].

Molecular weight characteristics and polydispersity of the PVCL were determined using a gel permeation chromatography (GPC Aligent 1100 series RI detector).

Methods

The pH of polymer solution mixtures was gradually decreased by adding 0.1M HCl dropwise and the optical density (D) of mixtures was determined using a Shimadzu UV-2401PC UV-Vis spectrophotometer (Shimadzu Corp., Kyoto, Japan) with a Shimadzu CPS-240A thermoelectrically temperature controlled cell positioner at 400 nm wavelength and at 25 °C.

Results

The dependence of the optical density (turbidity) of polymer mixtures on pH, containing equimolar amounts of PVCL and PAA of different molecular weights, are presented in Figures 2-4. The turbidimetric titration curves are shown in Figure 7 for the case of PAA with Mw 4.5×10^5 Da.

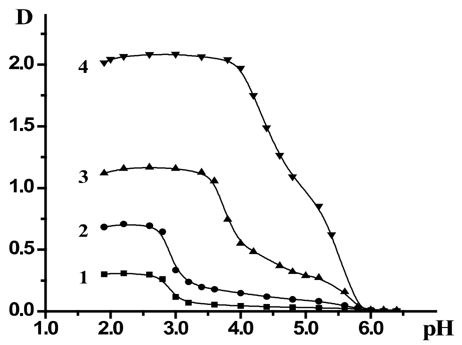

Figure 2 – The dependence of the optical density of polymer solution mixtures of PVCL and PAA on pH; Mw (PAA) = 2.5×10^5 Da, $C_{polymers}$ = 0.005 (1); 0.01 (2); 0.02 (3) and 0.05 mol/L (4)

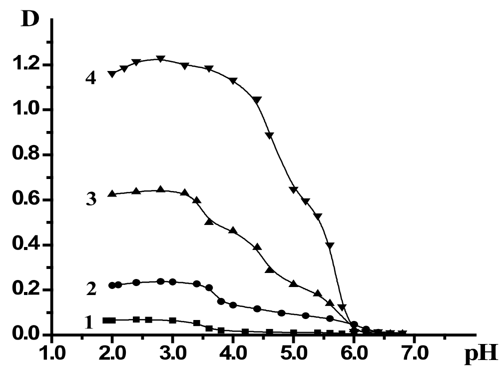

Figure 3 – The dependence of the optical density of polymer solution mixtures of PVCL and PAA on pH; Mw (PAA) = 4.5 × 10^5 Da, $C_{polymers}$ = 0.0025 (1); 0.005 (2); 0.01 (3) and 0.02 mol/L (4)

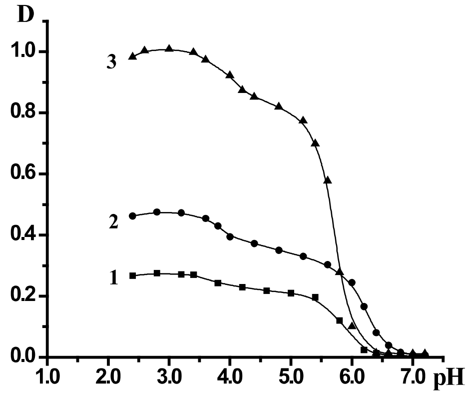

Figure 4 – The dependence of the optical density of polymer solution mixtures of PVCL and PAA on pH; Mw (PAA) = 7.5 × 10^5 Da, $C_{polymers}$ = 0.0025 (1); 0.005 (2) and 0.01 mol/L (3)

Discussion

PVCL belongs to a class of thermo-responsive polymers which possess a lower critical solution temperature (LCST) due to hydrophobic interactions in solutions [13, 14]. Complexes formed by such polymers are usually also sensitive to variations in other parameters that could shift the hydrophobic-hydrophilic balance, in particular, to variations in pH. This is confirmed by the data displayed in Figures 2-4.

In particular, it is clear that there is a critical pH value that limits the pH range, in which there is a sharp increase in the optical density of the solution and this is usually associated with the formation of a complex [15, 16].

A more detailed analysis of the depicted curves reveals important features of another product of interpolymer reactions between the macromolecules – hydrophilic interpolymer associate (HIA). The mechanism of formation of HIA, in accordance with the arguments reported in [10-12], is as follows.

The dimensions of polyacid and non-ionic polymer coils differ greatly, even when the medium is not very acidic. This is due to the well-known polyelectrolyte effect, which occurs until the ionization of the polyacid is suppressed caused by the low-molecular component. Therefore, for the formation of the classical interpolymer complex (IPC) in an alkaline or slightly acidic region, it is necessary that the energy needed to stabilize its bonds exceeds a certain critical value.

This value is determined by the work that must be done either against the forces of polyelectrolyte stretching of the coil formed by the ionic polymer or, on the contrary, against the forces of elastic compression of the coil formed by the non-ionic component. It is obvious that to form the classical IPC, a sufficiently large number of hydrogen bonds must be formed between two macromolecules. This could only be occurred when the coils they form are of comparable size. Consequently, in the acidic environment, the formation of IPC stabilized by hydrogen bonds is facilitated, since the size of the coil formed by the ionic coil is reduced due to the suppression of polyelectrolyte swelling. This conclusion is directly confirmed by the experimental data presented above (Figures 1-4) and by the theory developed in [16].

In contrast, in a weakly acidic or alkaline environment, the size of coils formed by ionic and non-ionic macromolecules are not comparable. In this case, a significant number of bonds could be formed only when a single coil of non-ionic polymer interacts with several polyacid molecules instantly (or vice versa). Therefore, in this case, the polyacid molecules (due to the fact that the coil they form is relatively larger in size) serve as a cross-linking agent for a non-ionic polymer. Since the energy of formation of hydrogen bonds is relatively low (these bonds are not strong enough), then a loose network is formed in the solution, the bonds of which are in dynamic equilibrium that disrupt constantly and appear again. This dynamic network, according to the reports in [10-12], is interpreted as HIA. HIA is a product of an interpolymer reaction, intermediate both with respect to the classical IPC and to a true solution containing non-interacting macromolecules, as well as with respect to a hydrogel with physical cross-linking nodes.

A schematic illustration of consecutive transitions from a solution containing non-interacting macromolecules to the classical IPC through HIA is depicted on Figure 5.

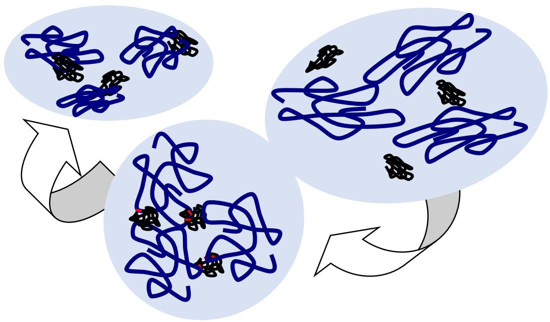

Figure 5 – A schematic illustration of sequential transitions in a system that allows the formation of HIA: non-interacting macromolecules–HIA–IPC

A comparison of the curves on Figures 2-4 demonstrates that their shape also changes markedly as the molecular weight of PAA increases. A detailed analysis of these curves allows to conclude that there are at least a number of intermediates in PVCL and PAA solution mixture. Moreover, as will be clear from the following, the transitions between such forms are of a stadial nature, which, in turn, allows to consider the environment in which the HIA is formed as a system with distributed memory.

In [10, 12], the following approximation was proposed for the dependence of the optical density of a polymer solution undergoing a phase transition.

$$D = \frac{D_0}{1 + \exp\left(\left(pH - pH_0\right)/\tau_0\right)} \qquad (1)$$

where: D_0 – an extrapolation extremum of the optical density (i.e. the value that would be realized if the reaction between the polymers took place completely, but the reaction product did not precipitate); pH_0 – a parameter that determines the boundary of the area of existence of the complex by pH (this parameter is directly related to the critical pH value determined by the standard method, but differs slightly from it in numerical value); τ_0 – a parameter that determines the slope of the curve describing the transition from a solution of non-interacting polymers to a complex.

The type of curve (1) could be obtained, including, based on semi-phenomenological theory [12], parameters D_0, pH_0 and τ_0 are calculated based on the experimental data. However, it is seen that the type of this dependence coincides with the logistic curve which is often used in the theory of neural networks [17]. In other words, this curve could be used for approximate description of any type of phase transition, regardless of whether it corresponds to the phenomenological model [12] or not.

Curve (1) satisfies a first-order differential equation

$$\frac{dD}{dpH} = \frac{D^2}{D_0 \tau_0} - \frac{D}{\tau_0} \qquad (2)$$

which could be obtained by directly differentiating (1) by a variable pH and expressing exponential factors through the function D in the equation (1) [19].

Equation (2) could be interpreted as the equation of the phase portrait of *D(pH)* curves, which is assumed as the dependence of the derivative of some function on the values of this function. Such phase portraits were used, in particular, in [18] to analyze the swelling kinetics of hydrogels in the presence of copper ions.

If the phase portrait of the curve under consideration obeys equation (2), i.e. it allows parabola approximation, then this curve itself will be described by the equation (1) [18].

Figures 6-8 display phase portraits of the experimental curves illustrated in Figures 2-4, respectively.

The depicted phase portraits were obtained using numerical differentiation with an approximate equation:

$$\left.\frac{dy}{dx}\right|_n = \frac{1}{10\Delta x}\left(2y_{n+2} + y_{n+1} - y_{n-1} - 2y_{n-2}\right) \qquad (3)$$

The Figures show that the phase portraits of these curves allow (with acceptable accuracy) approximations by a set of parabola fragments.

It can be seen that the phase portraits of individual curves shown in Figures 2-4 are indeed described with satisfactory accuracy by parabolic dependence. This, in particular, indicates the applicability of equation (1) to describe a change in the transparency of the solution during the phase transition (at least when such a transition is the only one).

However, there are curves whose phase portraits differ significantly from the parabolic one, too. Nevertheless, each of the presented phase portraits could be divided into separate sections, each of which is approximated by parabolic dependencies with satisfactory accuracy.

Based on this, it can be concluded that there are several different phase transitions take place in the system under consideration, i.e. the transition from a solution containing non-interacting macromolecules to IPC has a stadial nature. In particular, the phase portraits displayed on Figures 6-7 clearly show that the transition under consideration occurs in two stages.

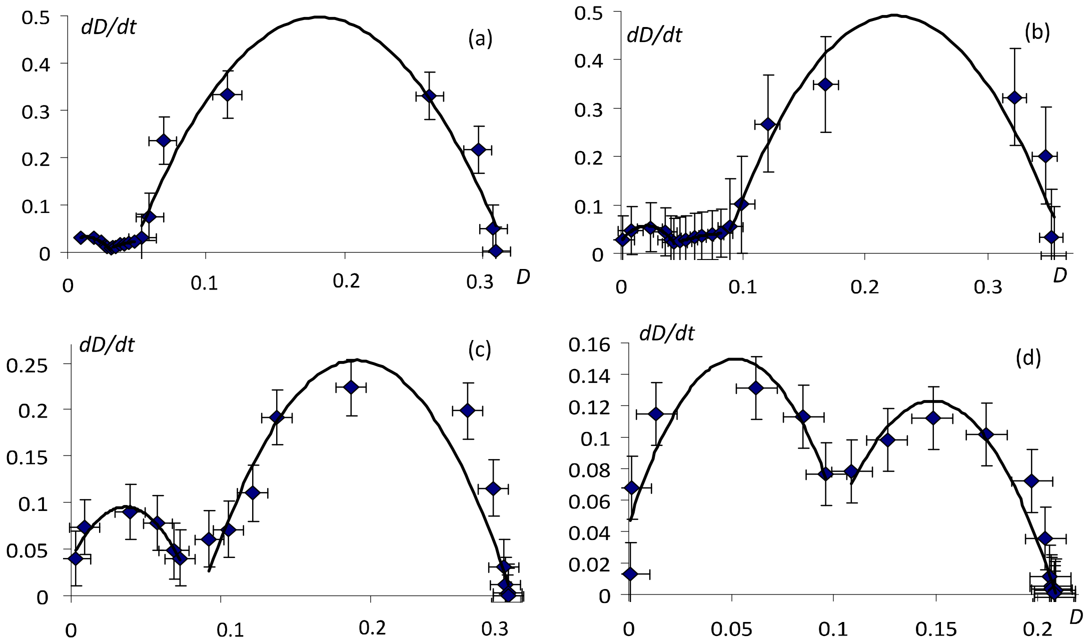

Figure 6 – Phase portraits of curves 1 (a), 2 (b), 3 (c) and 4 (d) shown in Figure 2

The two-stage phase transition fully corresponds to the scheme of Figure 5, specifically, the presence of an intermediate product of the interpolymer reaction – a hydrophilic interpolymer associate.

However, it can also be specified the case when the number of individual stages reaches three, which indicates the existence of not single, but two intermediate products. As will be clear from the following, even a system in which the phase transition occurs in two stages already possesses the properties with its inherent distributed memory (more specifically, such a system can become an analog of a neural network consisting of two neurons).

It should be emphasized that the method of phase portraits used to plot Figures 6-8 was originally developed in macromolecular chemistry to identify the stadial nature of the processes under consideration. Thus, it could be considered as experimentally proven that there are conditions under which the phase transition from the true solution of non-interacting macromolecules to the classical interpolymer complex may undergo through several different stages, some of which may not be determined in experiments, since the phase portrait method has a limited resolution, which depends both on the precision of optical measurements and on the possible superposition of one stage on another (the corresponding effect is also seen in above phase portraits).

It should also be emphasized that the nature of these stages, generally speaking, is not primarily determined by the nature of the interaction between macromolecules of the types under consideration.

Indeed, in fact, it is about a well-defined labile network structure, which could be considered as a complex system (in the philosophical meaning of the term).

As reported in [19-23], the behavior of complex systems, in which the structure of bonds can be transformed, is very non-trivial. It is emphasized that in the cited papers, extremely abstract network structures composed of formal elements, the only property of which is the formation of links, have been studied. Despite the fact that the elements in such grids are extremely simple, it was found that such objects are also characterized by pronounced phase transitions [22, 23]. Moreover, proofs of the existence of phase transitions in abstract grids, whose elements are endowed with the only property – the ability to form links with other elements of the system – can be obtained by direct calculation [24] without the use of numerical modeling methods used in such works as in [22, 23].

The results of work carried out in the study of complex network structures of various nature depict that there are conditions when in systems of this kind there are grids occur that differ from each other in structure, and differences in the nature of the structure could be associated with the occurrence of local inhomogeneities.

This conclusion could be used to justify the possibility of converting a hydrophilic interpolymer associate into a non-trivial information processing system.

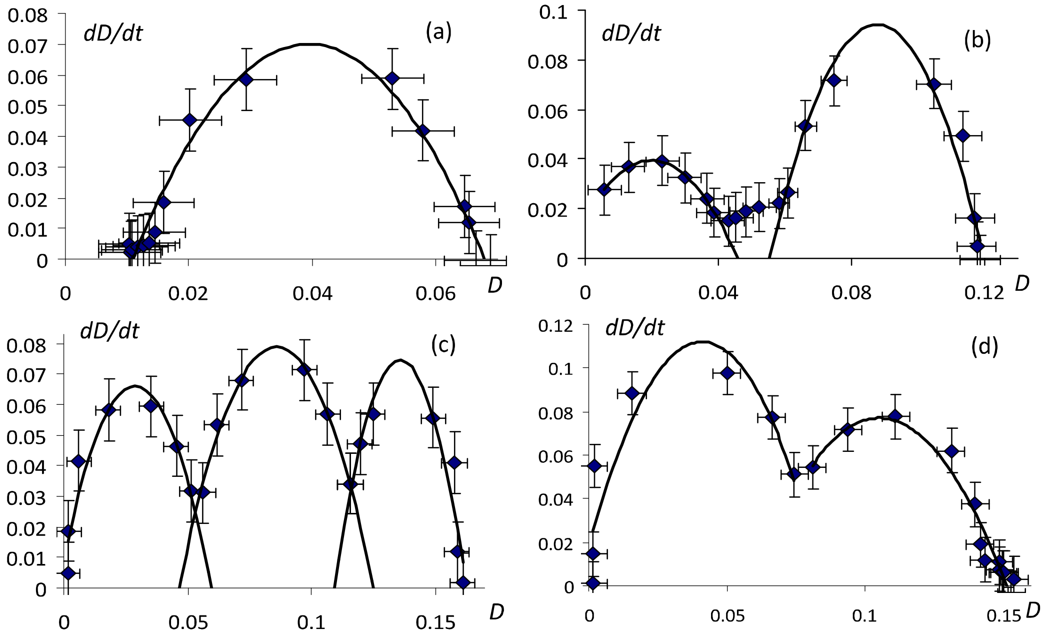

Figure 7 – Phase portraits of curves 1 (a), 2 (b), 3 (c) and 4 (d) shown in Figure 3

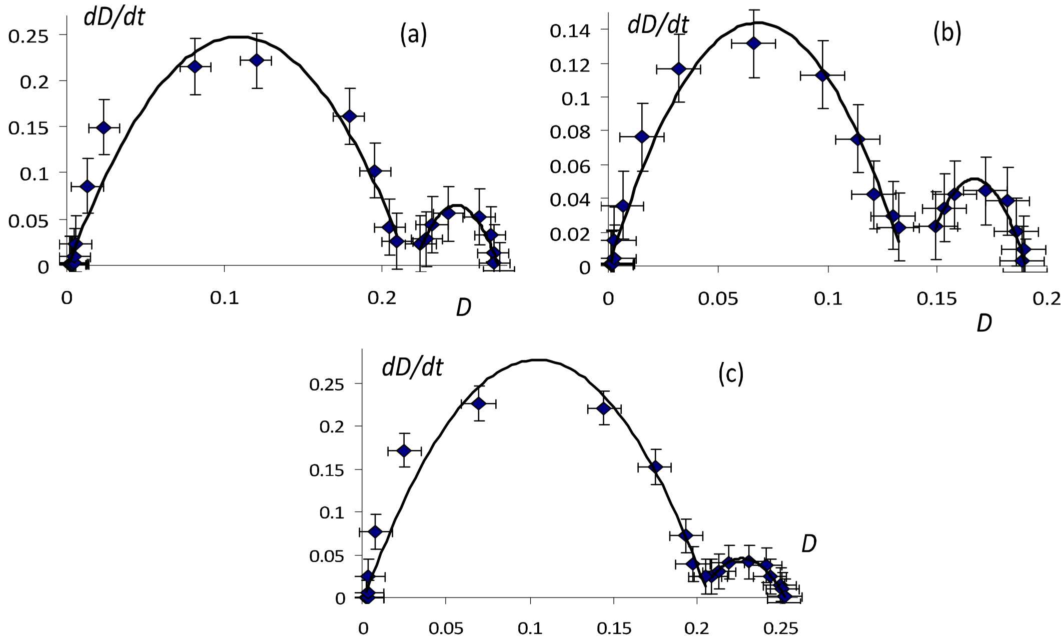

Figure 8 – Phase portraits of curves 1 (a), 2 (b) and 3 (c) shown in Figure 4

Transition from a dynamic network, i.e. from the hydrophilic interpolymer associate to the true interpolymer complex could be considered as a limiting case of "thickening of the grid" in some of its local regions. In this case, the density of bonds between two macromolecules at a local point reaches a maximum, and the links between these two macromolecules with others are terminated.

In other words, the true interpolymer complex can be considered as a limiting example of a hydrophilic associate in some parts of which the degree of heterogeneity of the dynamic network has reached its maximum value (maximum bond density between two separate macromolecules upon all other bonds are broken). This understanding, as well as the results of the works cited above, suggests that in fact intermediate cases can be implemented in such a system, i.e. situations when the distribution of the density of bonds along the dynamic grid is uneven. Apparently, the transitions of this kind that demonstrate the experimental results described above.

Thus, it becomes clear that at least some phase transitions in the system under consideration are not related to a change in the nature of the chemistry of bonds between macromolecules, but to changes in the structure of the network itself. In other words, it is about the observed manifestations of changes in the network structure at local scales, but exceeding the size of individual macromolecular coil by their characteristic dimensions.

The existence of staged phase transitions, which, as shown by the experiments described above, can be very complex (the number of stages could be up to three and, possibly, more) allows to state that such a system itself can have distributed memory.

Provided that the phase transition has a stadial nature, then the solution containing a hydrophilic interpolymer associate can be considered as a neural network (Figure 9). In accordance with the scheme shown in Figure 9, the solution is conditionally divided into regions, in each of which a phase transition occurs, and which corresponds to one of the stages discussed above. This stage corresponds to the transition from a hydrophilic interpolymer associate of the simplest type to the hydrophilic interpolymer associate, in which there are pronounced heterogeneities of the distribution of bond density.

As it was shown above, this stage is expressed for some conditions and can be interpreted as a transition from more homogeneous HIA to less homogeneous HIA, i.e., to the one in which a kind of nuclei of the interpolymer complex are formed, and more correctly, regions are formed in which the density of bonds between molecules of two interacting varieties is locally increased.

It is emphasized once again that, in accordance with the most general ideas about the nature of structural transitions in complex networks, any phase transition that does not change the nature of the interacting elements themselves can only be associated with structural transformations in the network itself. Thus, there is a transition associated with a local increase in the density of bonds and, accordingly, the density of swollen coils. Each of these sections can conditionally be considered as a neuron, and the output state of the neuron meets a logical zero is placed under the distribution corresponding to a more uniform grid, i.e. HIA of first type mentioned above, and the state corresponding to a logical unit is denser, i.e. HIA of second type.

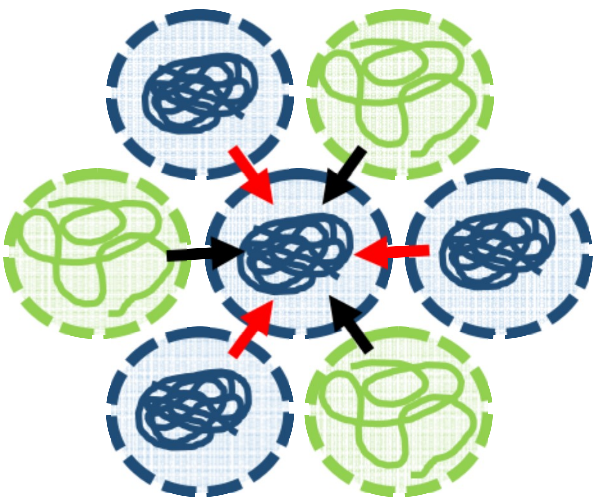

Figure 9 – A solution containing HIA as an analog of a neural network

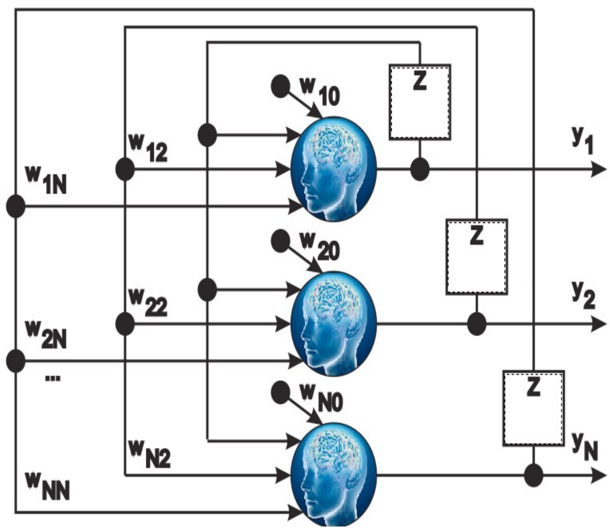

Figure 10 – Diagram of the Hopfield neuroprocessor

Obviously, there is a well-defined impact of neighboring regions on each other in the grid structure. Conditionally, such effect is shown by arrows on Figure 10. Therefore, this structure could be likened to a Hopfield neural network, in which all elements are covered by a feedback system (Figure 10). It is the fact that this system could be considered by analogy with the Hopfield neural network that proves that information can actually be written into a system of this type, that is, this system has distributed memory.

Conclusion

Thus, a solution of two interacting polymers in which hydrophilic interpolymer associates are formed is a direct analog of a neural network in a defined range of conditions. This object is able to evolve as a whole, which proves the possibility of implementing an alternative mechanism to the evolution considered in the framework of Darwinian theories.

The results obtained in this work could also be considered as a direct experimental confirmation of the concept of dialectical positivism. Specifically, it is about the interpretation of evolutionary processes in terms of understanding the information as a philosophical category, a paired category of matter.

From this point of view, the evolution of complex systems is complementary to the evolution of information objects. More precisely, qualitative-quantitative transitions that link different structural levels of the hierarchy of matter are complementary to structural levels that correspond to qualitative-quantitative transitions in the world of information entities.

We emphasize that it is about the evolution of the system as a whole. Even being composed of the same elements, it could experience transitions from quantity to quality, each of which can raise the level of information complexity, i.e. the emergence of the system of the new entities capable of processing the information qualitatively in different way than the components of the system elements.

As a result, the combination of such qualitative and quantitative transitions may well lead to a new state of the system, as a result of which it is converted into something qualitatively, that is different at the physical level. In other words, it turns into a kind of "filter" capable selecting the elements that are most relevant to its new state.

It is obvious that this kind of representation returns, including to the concept of Gaia J. Lovelock. More precisely, the results obtained allow to look at the problem of the origin of life from the point of view of J. Lovelock.

The primary is not the evolution of individual elements that later became components of the biosphere, the entire shell of the Earth evolved as a whole. Of course, a detailed consideration of these issues is beyond the scope of this work, however the results obtained in it already allow us to say that systems based on hydrophilic macromolecules can indeed evolve as a system whole and a well-defined structural memory occurs in them in a natural manner.

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