

Production Entropy and Negentropy

Mailybi Aldabergenov

«Modern Chemistry» LLP, Almaty, Kazakhstan

Email address:

aldabergenov_m@mail.ru

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Abstract: Chemical reactions in an open system are accompanied by direct and reverse transformations of components. In this case, irreversible processes are characterized by production of entropy of both direct and reverse transformations. Negentropy is production of the entropy of the reverse transformation. According to the second law of thermodynamics, production of entropy is a positive value. The difference between production of entropy of direct and reverse transformations (negentropy) is called the useful production of entropy. Negentropy is a barrier to the growth of the useful production of entropy of the system, which not all reactions can overcome. The relationship between the useful production of entropy and negentropy determines the path of evolution of the system at the bifurcation point. Based on the "The Gibbs function normalized to the total number of electrons" and on the example of triangulation of the $\text{CaO-SiO}_2\text{-H}_2\text{O}$ system, entropy production, negentropy, entropy flux, total entropy change, affinity, thermodynamic force, transformation temperature, rate constant at the interaction of Ca(OH)_2 and $\text{Ca}_3\text{Si}_2\text{O}_6(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ are calculated. In chemical reactions, production of entropy and negentropy exist in parallel.

Keywords: Nonequilibrium Thermodynamics, Entropy Production, Negentropy

1. Introduction

Irreversible processes are the driving force that creates order in the system. In chemical reactions far from equilibrium, ordered structures arise under the action of nonlinear interactions at the bifurcation point and lead to production of entropy. Non-equilibrium situations according to I. Prigogine [1] lead to space-time structures - dissipative structures. Production of entropy and its exchange with the environment can lead to the evolution of the system, passing into the mode of self-organization [2].

With regard to living organisms, E. Schrödinger [3] introduced the concept of "negative entropy" to explain the compensation of entropy production, L. Brillouin, justifying the process of obtaining information, called it negentropy [4].

Currently, negentropy is used mainly as the amount of information equal to the difference between the initial (before receiving the message) and final (after receiving the message) entropy, and as the reciprocal quantify of entropy, expressing the degree of order of material objects.

The second law of thermodynamics represents the change in the entropy of an open system in the form of two

components [1]:

1. production of entropy due to irreversible processes;
2. exchange of entropy with the environment.

Evolutionary processes in chemical reactions can be described on the basis of transformation of the phase composition of the system compounds as causal relationships between phases. At the point of bifurcation, the system loses its stability and passes into a nonequilibrium state, in which it is possible to acquire a new quality of evolution of the dynamic system. There take place a restructuring of the nature of the movement and the structure of the system. When a system moves far from equilibrium to a bifurcation point, the chemical potential and affinity of the system play a significant role. At the point of bifurcation, the system begins to produce entropy caused by irreversible processes, the passage of the reaction is characterized by a certain rate constant. It is these factors that determine which evolutionary branch the system will follow after passing through the bifurcation point. When chemical reagents interact in a real system, it is possible to determine all the evolutionary paths of the system. After all, evolutionary processes between two points of bifurcation obey deterministic laws [1].

Transformation of the components of a chemical reaction

can only occur spontaneously from a phase with a high potential to a phase with a low potential. In chemical reactions, various phases of substances exchange matter and energy, this being the basis of irreversible processes. Irreversible processes generate entropy production.

During chemical reactions, old bonds are broken and new ones are formed. In a phenomenological description of the nature of a chemical bond, one can proceed from the fact that electrostatic interactions during the formation of a molecule uniformly reduce the energy of all its electrons. This is directly reflected in the energies of the core electronic levels, and the so-called exchange energy includes the interference energy and the energy of mutual penetration [5], each of which contains competing kinetic and potential contributions. Therefore, when interpreting the phenomenon of chemical bonding, it is not enough to take into account only the valence electrons least bound to the nucleus - the contribution of the electronic system as a whole is important. The distribution of the electron density and the energy of the atomic core levels determine many properties of molecules, including their electronically excited states. We have proposed a new concept "The Gibbs function normalized to the total number of electrons" ($\Delta\bar{G}_{298}^0$) [6], the physical meaning of which is to define the chemical bond as a collective effect of electron-nuclear interaction. It is calculated by dividing the value of the Gibbs energy of the formation of a compound by the total number of electrons in a given compound and by the Avogadro number, i.e. it determines the density of the formation energy of the compound per one electron.

$$\Delta\bar{G}_{298}^0 = \frac{\Delta_f G_{298}^0}{n_e N_A}$$

where n_e is the total number of electrons in a given compound;

N_A is Avogadro's number;

Density usually refers to some volume. In this definition of the "The Gibbs function normalized to the total number of electrons", density refers not to volume, but to the number of electrons, i.e. characterizes accumulation of the Gibbs energy density on molecular bonds. During formation of bonds, the "The Gibbs function normalized to the total number of electrons" responds adequately to all changes in the structure of the compound, makes it possible to judge the reactivity of the compound, its stability, determine the value of the chemical potential, calculate the degree of conversion of components in the course of a chemical reaction, triangulate multicomponent systems and determine mechanisms and the rate of chemical reactions.

2. Methods of Research

The bifurcation points in the CaO-SiO₂-H₂O system were determined based on the triangulation of the system [7, 8]. Triangulation of the CaO-SiO₂-H₂O system (Figure 1) and the method for studying the processes of transformation of components are fully described in [8-13]. Thermodynamic parameters of the compounds: "The Gibbs function normalized to the total number of electrons", "Average electron heat capacity", "Average electron entropy" for the initial compounds in the solid state for the examples under consideration are given in Table 1. "Average electron entropy" and "Average electron heat capacity" are calculated similarly to "The Gibbs function normalized to the total number of electrons".

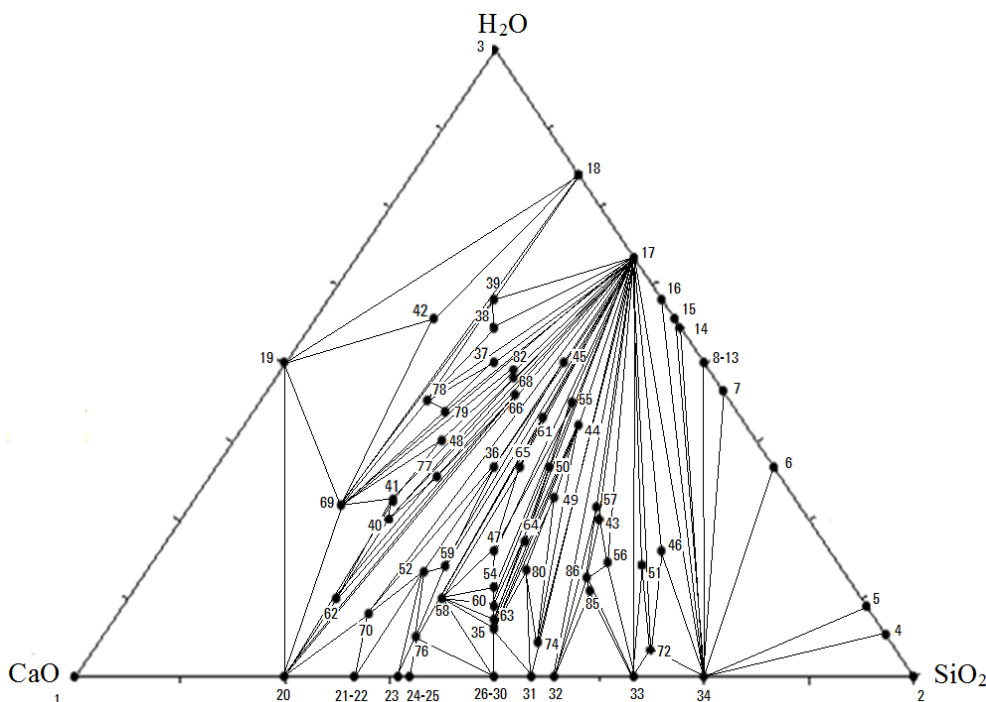


Figure 1. Triangulation of the CaO-SiO₂-H₂O system.

Table 1. Values of The Gibbs function normalized to the total number of electrons ($\Delta\bar{G}_{298}^0$), "Average electron heat capacity" (\bar{C}_p), "Average electron entropy" (\bar{S}) for the initial components in the solid state.

N* of the compound	Compounds	$-\Delta\bar{G}_{298}^0$, eV	\bar{C}_p , eV/K	\bar{S} , eV/K
19	$\text{Ca}(\text{OH})_2$	0.245	$0.239 \cdot 10^{-4}$	$0.228 \cdot 10^{-4}$
69	$\text{Ca}_6\text{Si}_2\text{O}_7(\text{OH})_6$	0.250	$0.181 \cdot 10^{-4}$	$0.187 \cdot 10^{-4}$
42	$\text{Ca}_2\text{SiO}_4 \cdot 4\text{H}_2\text{O}$	0.252	$0.230 \cdot 10^{-4}$	$0.255 \cdot 10^{-4}$
48	$\text{Ca}_3\text{Si}_2\text{O}_6(\text{OH})_2 \cdot 2\text{H}_2\text{O}$	0.262	$0.195 \cdot 10^{-4}$	$0.211 \cdot 10^{-4}$
18	H_8SiO_6	0.311	$0.331 \cdot 10^{-4}$	$0.600 \cdot 10^{-4}$
39	$\text{CaSiO}_3 \cdot 3\text{H}_2\text{O}$	0.259	$0.235 \cdot 10^{-4}$	$0.260 \cdot 10^{-4}$
78	$\text{Ca}_9(\text{Si}_3\text{O}_9\text{H})(\text{Si}_2\text{O}_7\text{H})(\text{OH})_8 \cdot 6\text{H}_2\text{O}$	0.256	$0.204 \cdot 10^{-4}$	$0.220 \cdot 10^{-4}$
17	H_4SiO_4	0.340	$0.307 \cdot 10^{-4}$	$0.658 \cdot 10^{-4}$
82	$\text{Ca}_{10}\text{Si}_{12}\text{O}_{31}(\text{OH})_6 \cdot 18\text{H}_2\text{O}$	0.266	$0.212 \cdot 10^{-4}$	$0.231 \cdot 10^{-4}$
79	$\text{Ca}_9\text{Si}_6\text{O}_{18}\text{H}_2(\text{OH})_8 \cdot 6\text{H}_2\text{O}$	0.258	$0.201 \cdot 10^{-4}$	$0.216 \cdot 10^{-4}$

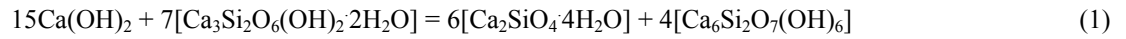
* The compound number is given in accordance with [9].

3. Results and Discussion

Let us consider the solid-phase chemical reactions of the interaction of compounds $\text{Ca}(\text{OH})_2$ (#19) and $\text{Ca}_3\text{Si}_2\text{O}_6(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ (#48). The bifurcation points during interaction of $\text{Ca}(\text{OH})_2$ and $\text{Ca}_3\text{Si}_2\text{O}_6(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ were

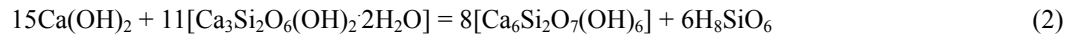
determined based on the triangulation of the $\text{CaO-SiO}_2\text{-H}_2\text{O}$ system (Figure 1) by connecting points #19 and #48 with a straight line. Line #19-#48 crosses lines #42-#69, #69-#18, #69-#39, #69-#78, #69-#79, #69-#17, #69-#82. The intersection points are the bifurcation points [9]. On this basis, the following reactions can be written at bifurcation points:

$$19 + 48 = 42 + 69$$



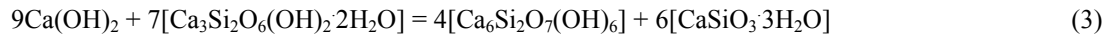
$$A_r = -0.005 \text{ eV.}$$

$$19 + 48 = 69 + 18$$



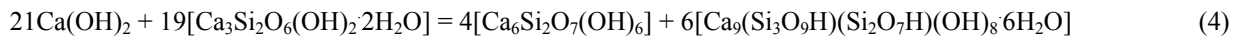
$$A_r = 0.054 \text{ eV.}$$

$$19 + 48 = 69 + 39$$



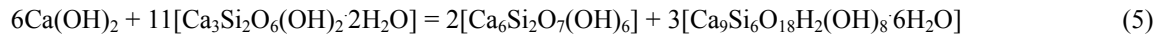
$$A_r = 0.002 \text{ eV.}$$

$$19 + 48 = 69 + 78$$



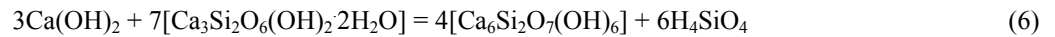
$$A_r = -0.001 \text{ eV.}$$

$$19 + 48 = 69 + 79$$



$$A_r = 0.001 \text{ eV.}$$

$$19 + 48 = 69 + 17$$



$$A_r = 0.083 \text{ eV.}$$

$$19 + 48 = 69 + 82$$



$$A_r = 0.009 \text{ eV.}$$

A_r is the chemical affinity of the reaction.

The "The Gibbs function normalized to the total number of electrons", referring to one electron, is interpreted as the

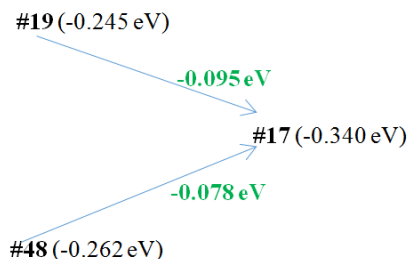
chemical potential (μ) of a substance

and is the driving force behind the chemical process. Then the chemical affinity for the reaction is determined according

to T.de Donde [14] as (eg, for reaction (1):

$$A_r = \mu_{19} + \mu_{48} - \mu_{42} - \mu_{69}$$

The chemical affinity sign indicates the possibility for the reaction to proceed from left to right (+) and from right to left (-). Based on this, it can be stated that reactions (2), (3), (5), (6), (7) proceed in the forward direction, and reactions (1) and (4) - in the opposite direction. Due to the fact, that the chemical potential and chemical affinity refer to one electron



In accordance with the first principle of thermodynamics during the transformation of components, their energetic change depends only on the initial and final states. Therefore, the possibility of converting the reaction components into products is initially considered. The chemical potential of the reaction components decreases in the cases of transformations #19-#17 (-0.095 eV), #48-#17 (-0.078 eV), #19-#69 (-0.005 eV). That is, these transformations proceed spontaneously from a phase with a high potential to a phase with a low potential. On the contrary, transformation #48-#69 (0.012 eV) cannot spontaneously proceed in the forward direction, but proceeds in the opposite direction.

Thus, transformations #19-#17, #48-#17, #19-#69 proceed in the forward direction, and transformation #48-#69 in the opposite direction. During these transformations, the system begins to produce entropy, for the first three transformations it is called entropy production, and for the opposite one - negentropy.

Negentropy is production of entropy for the reverse transformation. According to the second law of thermodynamics, production of entropy is always positive. Therefore, its value is not negative, but positive.

At the point of bifurcation, the system produces entropy caused by irreversible processes; the components of the system begin to transform into each other in both forward and reverse directions. The evolution of the system in forward or backward directions depends on the amount of entropy produced both in forward and backward directions. In all transformations, the production of entropy and negentropy exist in parallel. Negentropy acts opposite to the growth of entropy production of the system.

For all transformations, the temperature (T), thermodynamic force (F), entropy production (d_iS), total entropy (dS), entropy flux (d_eS), and rate constant (K) were calculated.

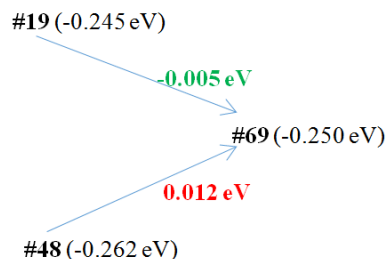
Affinity was calculated according to T.de Donde [14].

The temperature of the process of transformation of the initial component into a stable compound, accompanied by a decrease in the chemical potential, is defined as the difference in the "The Gibbs function normalized to the total number of electrons" of the components divided by the

and the elementary act of collision of the reaction components of a nonequilibrium system is described, stoichiometric coefficients are not taken into account.

Let us consider in more detail reaction (6), the data on the rest of the reactions are presented in Tables 3 and 4.

In this reaction, during interaction of #19 and #48, transformations are possible (chemical potentials of substances are indicated in parentheses):



"average electron heat capacity" of the resulting compound.

Thermodynamic force (F) and entropy production (d_iS) are determined in accordance with [3] by the following formulas:

$$F = (1/T_o - 1/T),$$

where T_o is the standard temperature (298 K).

$$d_iS = F \cdot A,$$

where A is the affinity.

The total entropy change for the transformation (dS) is defined as the difference between the "average electron entropies" of the compounds (Table 1), and the entropy flux is

$$d_eS = dS - d_iS.$$

K is the rate constant according to the transition state theory [15]

$$K = \chi \frac{k}{h} T e^{-\frac{\Delta E_a}{RT}} e^{\Delta S_a/R}$$

Where χ is transmission coefficient; k - Boltzmann constant; h - Planck constant; R - Gas constant; ΔE_a - activation energy; ΔS_a - activation entropy; T - temperature.

The parameters of individual transformations of the system components are given in Table 2.

Table 2. Parameters of individual transformations of the system components.

	#19-#69	#19-#17	#48-#17	#69-#48
A, eV	0.005	0.095	0.078	0.012
T, K	276	3094	2541	615
F, 1/K	$0.267 \cdot 10^{-3}$	$3.033 \cdot 10^{-3}$	$2.962 \cdot 10^{-3}$	$1.73 \cdot 10^{-3}$
d_iS , eV/K	$1.335 \cdot 10^{-6}$	$2.881 \cdot 10^{-4}$	$2.310 \cdot 10^{-4}$	$2.076 \cdot 10^{-5}$
dS , eV/K	$-0.041 \cdot 10^{-4}$	$0.43 \cdot 10^{-4}$	$0.447 \cdot 10^{-4}$	$0.024 \cdot 10^{-4}$
d_eS , eV/K	$-0.054 \cdot 10^{-4}$	$-2.451 \cdot 10^{-4}$	$-1.863 \cdot 10^{-4}$	$-0.1836 \cdot 10^{-4}$
K, 1/c	$5.741 \cdot 10^{12}$	$6.436 \cdot 10^{13}$	$5.285 \cdot 10^{13}$	$1.279 \cdot 10^{13}$

Transition to a stable state can only occur via instability. The hierarchy of the change of one stable state of the system by another causes a sequence of phase transitions from one structurally stable regime to another and is carried out

through the bifurcation point. This point is the threshold of stability of the thermodynamic branch. At this point, the individual transformations of the components pass into the conjugate transformation, where the chemical process begins.

For the reaction under consideration, the conjugated transformation begins with transformation #19-#69, which occurs at a lower temperature, in the second stage, transformation #48-#17 is added to it, then #19-#17. Here, the manifestation of the resonance mechanism based on the phenomena of cooperativity is possible, when the first reaction facilitates the passage of the second, the second reaction facilitates the passage of the third one, and so on. For reaction (1), two transformations are direct (#19-#42, #19-#62) two are reverse (#48-#42, #48-#69). For reaction (2) -

three straight lines (#19-#18, #48-#18, #19-#69), one reverse (#48-#69); for reaction (3) - two straight lines (#19-#39, #19-#69), two - reverse (#48-#39, #48-#69); for reaction (4) - two straight lines (#19-#78, #19-#69), two - reverse (#48-#78, #48-#69); for reaction (5) - two straight lines (#19-#79, #19-#69), two - backward ones (#48-#79, #48-#69); for reaction (7) - three straight lines (#19-#82, #48-#82, #19-#69), one - reverse (#48-#69). The transformations of the components occur both in the forward and reverse directions. There is not a single reaction where there would be no negentropy.

Taking into account the phenomena of cooperativity, the indicators for the conjugate transformation were calculated for all 7 reactions, both in the forward and reverse directions. The data are shown in Tables 3 and 4.

Table 3. Parameters of conjugate direct transformations.

Reactions	A, eV	T, K	F, 1/K	d _i S, eV/K	dS, eV/K	d _c S, eV/K	K, 1/c
1	0.012	292	0.069·10 ⁻³	0.828·10 ⁻⁶	-0.014·10 ⁻⁴	-0.0223·10 ⁻⁴	6.074·10 ¹²
2	0.12	1423	2.653·10 ⁻³	3.184·10 ⁻⁴	0.72·10 ⁻⁴	-2.464·10 ⁻⁴	2.96·10 ¹³
3	0.019	456	1.163·10 ⁻³	2.21·10 ⁻⁵	-0.009·10 ⁻⁴	-0.222·10 ⁻⁴	9.485·10 ¹²
4	0.016	386	0.765·10 ⁻³	1.224·10 ⁻⁵	-0.049·10 ⁻⁴	-0.171·10 ⁻⁴	8.029·10 ¹²
5	0.018	471	1.233·10 ⁻³	2.219·10 ⁻⁵	-0.053·10 ⁻⁴	-0.275·10 ⁻⁴	9.797·10 ¹²
6	0.178	2239	2.909·10 ⁻³	5.178·10 ⁻⁴	0.836·10 ⁻⁴	-4.342·10 ⁻⁴	4.657·10 ¹³
7	0.03	496	1.34·10 ⁻³	4.02·10 ⁻⁵	-0.018·10 ⁻⁴	-0.42·10 ⁻⁴	1.032·10 ¹³

Table 4. Parameters of conjugate reverse transformations.

Reactions	A, eV	T, K	F, 1/K	NegE, eV/K	d _i S _u , eV/K	dS, eV/K	d _c S, eV/K	K, 1/c
1	0.022	564	1.583·10 ⁻³	3.483·10 ⁻⁵	-3.400·10 ⁻⁵	-0.02·10 ⁻⁴	-0.328·10 ⁻⁴	1.173·10 ¹³
2	0.012	615	1.73·10 ⁻³	0.208·10 ⁻⁴	2.976·10 ⁻⁴	0.024·10 ⁻⁴	-0.184·10 ⁻⁴	1.275·10 ¹³
3	0.015	385	0.759·10 ⁻³	1.139·10 ⁻⁵	1.071·10 ⁻⁵	0.025·10 ⁻⁴	-0.114·10 ⁻⁴	8.008·10 ¹²
4	0.018	462	1.191·10 ⁻³	2.144·10 ⁻⁵	-0.92·10 ⁻⁵	-0.015·10 ⁻⁴	-0.229·10 ⁻⁴	9.61·10 ¹²
5	0.016	410	0.917·10 ⁻³	1.467·10 ⁻⁵	0.752·10 ⁻⁵	0.019·10 ⁻⁴	-0.1277·10 ⁻⁴	8.528·10 ¹²
6	0.012	615	1.73·10 ⁻³	2.076·10 ⁻⁵	4.9704·10 ⁻⁴	0.024·10 ⁻⁴	-1.836·10 ⁻⁵	1.279·10 ¹³
7	0.012	615	1.73·10 ⁻³	2.076·10 ⁻⁵	1.944·10 ⁻⁵	0.024·10 ⁻⁴	-1.836·10 ⁻⁵	1.279·10 ¹³

Comparison of entropy production for direct transformations and negentropy (NegE) shows that for reactions (2), (3), (5), (6) and (7), entropy production is greater than negentropy. The difference between entropy production and negentropy is called useful entropy production (d_iS_u):

$$d_i S_u = d_i S - \text{NegE}$$

By the value of the useful production of entropy, it can be stated that the energetically most probable are the passage of reactions (2), (3), (5), (6), (7) in the forward direction, caused by irreversible processes.

The change in useful production of entropy versus affinity is shown in Figure 2.

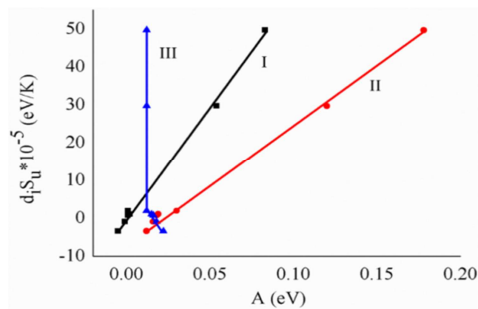


Figure 2. Dependence of the useful production of entropy on the affinity.

All changes in the useful production of entropy depending on the chemical affinity of the reaction (Figure 2, line I), depending on the affinity of direct transformation (Figure 2, line II) are characterized by straight-line dependences. In this case, the change in the useful production of entropy depending on the affinity of the direct transformation occurs in a wider range of changes in the affinity (from 0.01 to 0.18 eV). Whereas useful production from the affinity of the reverse transformation is characterized by a sharp decrease in it in a narrow range of affinity from (0.012 to 0.022 eV) along a sinusoidal curve (Figure 2, curve III). It should be noted that negentropy is a barrier to the growth of the entropy of the system. This barrier can be overcome only by reactions (2) and (6). This indicates the possibility of only reactions (2) and (6) occurring under real conditions (Figure 2, line I).

Entropy production and negentropy are considered together as the state of the system. Production of entropy leads to an increase in the entropy of the system, and due to negentropy - to a decrease in entropy.

The increase in the entropy of the system is associated with its production as a result of certain chemical reactions. Production of entropy begins under non-equilibrium conditions, it acts in the forward direction, and negentropy occurs immediately, but in the opposite direction. Production

of entropy causes formation of new structures, and negentropy is a force that destroys formation of new structures, restricts the random movement of elements, thereby counteracting the general tendency of increasing entropy, returning the system to its original state, which was characterized by strict order and direction. All this constitutes a negentropic field in all parts of the system and it objectively exists and its value can be calculated. Like entropy production, negentropy is related to chemical potential, thermodynamic strength, affinity, and other parameters. Negentropy is transferred from one system to another, is associated with the processes of self-organization, structuring, etc.

Negentropy is not an abstract quantity, it is associated with mass, energy, and characterizes the cause-and-effect relationships of processes occurring in matter. Negentropy as a material process can directly affect the energy and matter of the system and change them.

4. Conclusion

At the bifurcation point, the system produces entropy due to irreversible processes; the components of the system begin to turn into each other in both forward and reverse directions. The difference in the produced entropy, both in the forward and in the opposite directions, is the useful production of entropy, the value of which determines the evolution of the system in the forward or in the opposite directions. Entropy production in the opposite direction is negentropy. Production of entropy in both the forward and reverse directions in accordance with the second law of thermodynamics is a positive value. Therefore, negentropy is also positive. In all chemical reactions, production of entropy and production of negentropy are parallel processes; there is not a single reaction where there is no negentropy. Negentropy is opposed to the increase in production of the system entropy.

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Biography



Mailybi Aldabergenov is doctor of chemical science, professor. He works at "Modern Chemistry" LLP. He proposed a new concept for studying the nonequilibrium state on the basis of the "The Gibbs function normalized to the total number electrons". On the basis of the proposed principles, the mechanisms of interaction of system components at the level of elementary collisions are explained.

Technologies for obtaining polymer boron-phosphorus fertilizers "Foskabor", "Borkristalin", plant growth stimulators GuPhos have been introduced into the industry. He has published over 200 articles, 18 patents. Under his leadership, 3 doctoral, 14 candidate and 2 PhD dissertations were defended.