

## RADIATION PHYSICS OF BIOORGANIC COMPOUNDS: GLUCOSE

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It has been shown that radiation physics of bioorganic compounds (BOC) must take into account the stages of formation and colonization of metastable states in the process of relaxation of molecular fission fragments, the Lego effect. The theory of such radiation transformations requires the use of Ball and Stick structural models, as well as color statistics that record the topology of BOC molecules. The calculation, performed on the example of radiation destruction and ordering of glucose molecule atoms, demonstrates the possibility of forming 12 or more chemical compounds with different properties. The practical implementation of such compounds requires the use of different both irradiation conditions and post-radiation "cooling." The results obtained explain the results of BOS radiation experiments conducted on the M-30 microtron.

### INTRODUCTION

The interaction of nuclear radiation with matter is accompanied by processes of its destruction, ordering of the defect system, and the final state of radiation damage. Such transformations are valid for matter and any chemical and phase state and are determined by the scale of energy events – the interaction of ionizing radiation (nuclear scale, megaelectronvolt) and the ordering of the damage state (physico-chemical transformations, tens of electronvolt). However, the processes of ordering the defect state, which determines the radiation damage of a substance, significantly depend on its chemical and phase states. Currently, we can only speak of the radiation physics of crystalline materials as a complete science with the introduction of the concept of point-type radiation defects – such as Schottky, Frenkel, or areas of disorder [1, 2]. Such defects are considered as disturbances in the regular crystal lattice, and the study of their statistics and transformations forms the basis of solid-state radiation physics. However, this approach cannot be applied to materials that lack long-range atomic order, i.e., disordered or amorphous materials. This class of materials encompasses inorganic, organic, and bioorganic materials, as well as polymers, which are crucial for numerous practical applications and for understanding the nature of living matter under terrestrial radiation conditions. Since these materials are dominated by short-range order and the disordered state of atoms in the structure before and after radiation treatment, it is essential to select markers that determine the degree of their defectiveness.

In the first works on the radiation physics of polymer materials, the method of structural combinations based on click chemistry representations of stimulated switching of homo and hetero chemical bonds was used to explain the transformations of chalcogenide glasses (CG) under the influence of

external factors [3]. The foundations of an approach have been proposed that allows the evaluation of radiation-stimulated changes in the physicochemical parameters of glasses while preserving their polymer structure [4].

The radiation physics of organic structures has its own peculiarities due to their fragmentary structure, the presence of structural segments, complexes with different types of chemical bonds, and energy capacity [5]. In the future, we will consider bioorganic compounds (BOC) formed by a limited number of light chemical elements, which are called organogenic, such as carbon (C), oxygen (O), hydrogen (H), and nitrogen (N), which make up more than 90% of the total mass of bioorganisms. Terrestrial radiation plays a constant role in the existence of living matter, preventing its transition to an ordered state and serving as a source of new biological species during the evolution of biota [6, 7]. Unlike the CS discussed above, BOS are multicomponent compounds with a fragmentary structure containing structural segments and complexes with different types of chemical bonds. These can be strong bonds formed by organogenic atoms through covalent and ionic bonds, as well as weak bonds that give rise to van der Waals and London dispersion forces. Therefore, the radiation physics of such compounds, in addition to the switching of homo- and hetero-bonds during radiation transformations, must also consider other possibilities: the destruction of polymeric bioorganic structures, the formation of broken chemical bonds, as well as the creation of new chemical compounds and topological complexes. This paper considers the fundamentals of radiation physics of BOS using click chemistry methods and color statistics, which considers atoms with different bond energies as statistically non-equivalent [4]. Glucose was used as a model object for research, whose physicochemical parameters are well studied before and after radiation treatment [8–10].

## 1. THEORY

Unlike crystalline materials, where radiation defects cause local disturbances of long-range order and do not change their chemical composition, the situation is different for bioorganic materials. On the scale of a single molecule, the action of a high-energy particle causes significant destruction and the formation of an array of fragments upon its fission. Such fission fragments can be parts of the parent bioorganic molecule or the result of their click-chemical interaction, leading to the formation of molecular fragments of a different chemical nature that are absent in the original molecules. The general research scheme involves establishing all possible fragmentation channels, forming arrays of molecular fragments, and studying the thermodynamic ordering processes of the ensemble of structural clusters formed from these fragments. At the same time, for bioorganic materials, the result of ordering can be the restoration of their previous chemical composition, or the formation of new chemical compounds when molecular fragments are combined in ways that were not present in the initial composition.

The ordering of the defect state of irradiated materials is an equilibrium process, the final state of which is determined by a set of thermodynamic parameters [11, 12]. For both crystalline and disordered materials, formally, the task of finding the final metastable states of a substance requires determining the minima of the isobaric thermodynamic function at a given temperature  $T$ .

While for irradiated crystalline substances, the ordering concerns a relatively simple system of point defects, such as vacancies or interstitial atoms, the concept of the defect state of BOM is much more complex. Radiation causes the molecular fragmentation of their structure, resulting in the formation of structural complexes containing clusters of organogenic atoms connected by stronger ionic-covalent bonds.

To perform model calculations of the probable channels of post-radiation fragmentation of BOS, it is convenient to use the Ball and Stick model [13], in which its chemical structure is represented as a set of balls (atoms) of different sizes, connected by directed segments (chemical bonds), whose binding energies are different. Based on the literature data [14, 15], the following hierarchy of energies of single chemical bonds of organogenic atoms of BOS can be obtained (the fraction of their breaking energy relative to O-H is given in parentheses):

$$\begin{aligned} E_{O-H} (1) > E_{C-H} (.93) > E_{N-H} (.85) > E_{C-C} (.8) > \\ E_{C-O} (.73) > E_{N-C} (.64) > E_{N-O} (.38). \end{aligned} \quad (1)$$

It should be noted that the equations (1) are approximate, as tabulated and, for example, calculated data for EC-O may differ within 10%. According to (1), atomic clusters of molecules formed after irradiation with BOS must contain complexes with hydroxyl groups (O-H), fragments of carbon skeletons (such as C-H and -C-C-), as well as nitrogen-hydrogen compounds where the nitrogen atom is bound to one or

two hydrogen atoms. The latter play an important role in the properties of proteins and other biologically important molecules. Unlike simple radiation defects in solids, the structural fragments formed in BOS are spatially significant, can contain 5–10 atoms, and the process of restoring the original structure and chemical compound as a result of click-chemical transformations of homo- and hetero-bonds is significantly complicated. The structure of the ensemble of molecular fragments formed during BOS irradiation may vary depending on the irradiation conditions: the type of nuclear particles, their energy, and the intensity of radiation. This factor also affects the nature of the initial array of fission fragments and the final formation of chemical compounds with properties distinct from those of the parent BOS. In the experiment, this ability was demonstrated using glucose samples, which, after irradiation, changed both their chemical properties, in particular the pH parameter, and their mass spectral composition [10, 16].

To obtain numerical results, let us consider the thermodynamic function  $F$  of the form:

$$F_i = U_i - T \cdot S_i. \quad (2)$$

As can be seen from (2), the minima of the thermodynamic function at a constant temperature  $T$  will be determined by a fixed sets  $\{ U_i, S_i \}$ , where  $U_i$  has the meaning of the internal energy of a finite metastable BOS, which may have the same chemical composition but a different structural formula and properties, and  $S_i$  is its configurational entropy, index  $i=1, n$  indicates the possibility of realizing not one, but a set of metastable states. Conventionally, these stages are shown in Fig. 1: from the formation of an ensemble of molecular fragments (EnMF) formed during the irradiation of the BOS, the structure of which depends on the irradiation conditions, the process of thermodynamic ordering of the ensemble during and after the action of radiation, and finally, the possibility of realizing a set of metastable states corresponding to compounds with other physico-chemical properties. The metastable states of BOS formed in the ordering process can be conveniently characterized by configuration coordinates  $\{ R_n \}$ .

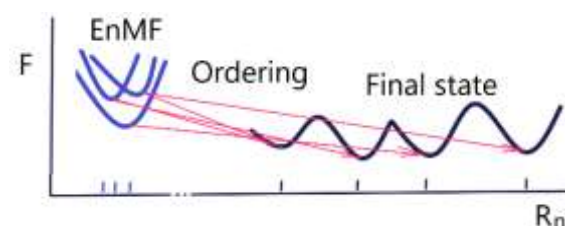


Fig. 1. Schematic representation of the evolution of an ensemble of molecular fragments of irradiated BOS (different irradiation conditions – different ensembles) through processes of thermodynamic ordering with the possibility of forming different sets of metastable states, identified by configuration coordinates  $\{ R_n \}$ . Such metastable states correspond to the possibility of creating different chemical substances from the initial composition of the BOS

In crystals, the configuration coordinates  $\{R_n\}$  can be related to the concentration of point defects in the structure, such as vacancies or interstitial atoms, and for polymeric materials, such as BOS, through the fixation of the energy states of their atoms. This problem can be solved within the framework of color statistics, which considers atoms as statistically non-equivalent when they have different binding energies in the BOS structure. Within the Ball and Stick model, the energy of a molecule  $U_i$  is found through the total energy of homo and hetero chemical bond breaks, which have different values, (1). A set of such bonds that determines the  $i$ -th metastable state of a molecule can be realized not by one, but by an array of colored atoms,  $\{k_j^i\}$ , which locally have different bond energies in the BOS and should be treated as statistically independent. Since this applies to organogenic atoms of the same type with varying energies of bond in the molecule, they can formally be distinguished by different conditional colors. Hereafter, the arrays,  $\{k_j^i\}$ , will be referred to as color sets (CS), and the statistics that allow such CS to be systematized are called color statistics [4]. Therefore, the BOS state can be recorded both as a chemical formula and as CS,  $\{k_j^i\}$ , which determines the topology of the atoms in the molecular structure.

The use of the Ball and Stick model and color statistics enables us to determine the values of the parameters  $\{U_i, S_i\}$  that are crucial for analyzing function (2). As mentioned above, the energy is determined by the sum of the energies of the breaks in the chemical bonds of the BOS, and the configurational entropy  $S_i$  will be expressed through  $\omega_i$ , – the degeneracy of the  $i$ -th metastable state with energy  $U_i$ , [17]:

$$S_i = \ln \ln \left( \sum_{j=1}^n \omega_j \right) = \ln \ln \left( \sum_{j=1}^n \frac{M!}{\prod_{j=1}^n k_j^i} \right), \sum_{j=1}^n k_j^i = M. \quad (3)$$

Here  $\{k_j^i\}$  – CS atoms for the  $i$ -th metastable state of a molecule having  $M$  atoms, the presence of the sum in (3) indicates that BOS is a non-additive system, when one of its states corresponds to an array of CS. The scheme for establishing color sets is as follows: organogenic atoms, whose immediate environment is determined by different ratios of chemical bonds of BOS atoms, are designated according to the following rule: a capital letter denotes the type of atom in an organic molecule (C, O, N, H), lowercase letters denote atoms in the immediate environment with which single chemical bonds are formed. For example,  $Ccchh$  describes a carbon atom with two covalent bonds to neighboring carbon atoms and two hydrogen atoms, while  $Och$  describes an oxygen atom bonded to carbon and hydrogen, and so on. Using this rule, you can get all possible color sets (CS) of organogenic atoms.

For carbon, C, whose valence is 4, the total number of colored atoms is 35, of which there will be CS in molecular structures where the carbon atom is connected to one or two more carbon atoms. Taking this condition into account, we obtain the following result:  $Ccenn$ ,  $Ccno$ ,  $Ccenh$ ,  $Ccco$  (ketone group),  $Ccch$ ,  $Ccchh$ ,  $Ccnnn$  (nitrile group),  $Ccnno$ ,  $Ccnnh$ ,  $Ccnno$  (amide group),  $Ccnoh$ ,  $Ccnhh$ ,  $Ccoo$  (carboxyl or ester

group),  $Ccooh$  (aldehyde group),  $Ccohh$ ,  $Cchhh$  – a total of 16 sets. For nitrogen, N, with a valence of 3, the total number of colored atoms is 20, of which only those CS in which N is bonded to carbon C can be considered promising for BOS:  $nccc$  (nitrile group),  $Nccn$ ,  $Ncco$ ,  $Nech$ ,  $Ncnn$ ,  $Ncno$ ,  $Ncnh$ ,  $Ncoo$ ,  $Ncoh$ ,  $Nchh$  (amino group or amide group) – 10 sets in total. For oxygen, O, a total of 10 sets of colored atoms can be obtained, of which only those in which oxygen is bonded to carbon are important:  $occ$  (ketone group),  $Ocn$ ,  $Oco$ ,  $Och$  (hydroxyl group) – a total of 4 sets. And finally, for hydrogen, H, which can form a CS with four atoms, three colored atoms are necessary for practical use:  $Hc$ ,  $Hn$ , and  $Ho$  (hydroxyl group).

The presence of broken chemical bonds, for example, when a C-C bond breaks, increases the capacity of the CS by additionally introducing atoms with dangling bonds, for example,  $Cc\bar{c}hh$ , denotes a carbon atom of the fission fragment that has lost a chemical bond with a neighboring atom of the carbon skeleton.

Thus, the role of radiation is reduced to reformatting homo and hetero chemical bonds and CS, and the task is reduced to establishing possible metastable states at the final stage of ordering irradiated materials. Further analysis can be conveniently carried out using the example of the radiation ordering of glucose molecules.

## 2. STRUCTURAL MODELS OF THE ORDERING OF IRRADIATED GLUCOSE

Glucose  $C_6H_{12}O_6$  is a convenient model object for studying radiation fragmentation of BOS. The choice of glucose/sucrose, which are essential representatives of complex carbohydrates, is due to their widespread usage in medicine and the food industry. In energy metabolism, glucose is the most important source of energy for biota, and in plants – for the production of cellulose for use in cell walls. It exists in linear and cyclic modifications and has a number of isomers. As can be seen in Fig. 2,a, the linear form of glucose contains an aldehyde (-CHO), five hydroxyl groups, and a primary alcohol (-CH<sub>2</sub>OH); its structure is described by CS  $\{Ccohh, 4 Ccch, 1 Cc2oh, 5 Och, 1 O2c, 7 Hc, 5 Ho\}$ .

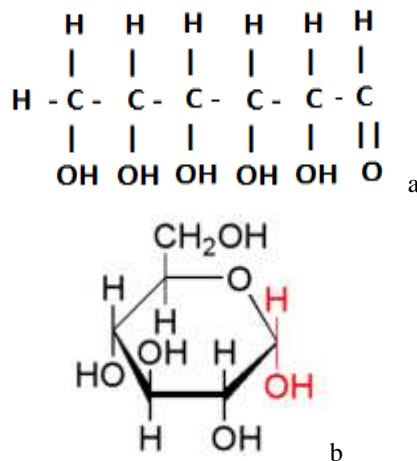


Fig. 2. Structural formulas of  $D$ -glucose,  $C_6H_{12}O_6$ : a – chain; b – cyclic. Fisher's representation of chemical compounds was used [18]

As mentioned above, we should proceed from the fact that radiation leads to the destruction of the glucose molecule and the subsequent rearrangement of its fragments. Each stage of rearrangement leads to the formation of a metastable structure, the energy state of which is determined not only by homo- and hetero-bonds, but also by bonds broken by radiation. Let us introduce the following notation:  $x_1$  is the number of C-C bonds;  $x_2$ , respectively, is C-O;  $x_3$  is C-H; and  $y_1$  is O-H. For glucose, as shown in Fig. 2,  $x_1 = 5$ ,  $x_2 = 7$ ,  $x_3 = 7$ , and  $y_1 = 5$ . Considering the presence of "broken" bonds, for example, for carbon atoms –  $x_{11}$ , or oxygen atoms –  $y_{11}$ , formed under the action of radiation, all possible combinations of their homo- and hetero-bonds are determined from the system of equations:

$$\begin{aligned} 2x_1 + x_2 + x_3 + x_{11} &= 24, \\ x_3 + y_1 &= 12, \\ x_2 + y_1 + y_{11} &= 12. \end{aligned} \quad (4)$$

Within the Ball and Stick model, each combination  $\{x_i, y_j\}$  that satisfies (4) determines the bond energy of

the molecular structure,  $E$ , since it is additive in terms of the number of chemical bonds:

$$E = E_{C-C} \cdot x_1 + E_{C-O} \cdot x_2 + E_{C-H} \cdot x_3 + E_{O-H} \cdot y_1. \quad (5)$$

Here  $E_{C-C}$ ,  $E_{C-O}$ ,  $E_{C-H}$  and  $E_{O-H}$  are the energies of breaking single chemical bonds C-C, C-O, C-H, and O-H, which are tabulated in [14, 15] and whose values can be taken as 3.61, 3.31, 4.2, and 4.52 eV, respectively. The value of  $E$ , determined in (5) with accuracy to a constant value, reflects the value of the internal energy  $U_i$ , a parameter important for calculating the thermodynamic function  $F$  and establishing the probability of realizing the metastable state of a biomolecule. Another parameter in formula (2) is its configurational entropy,  $S_i$ , which requires establishing  $\{k_i^j\}$  a set of colored organogenic atoms for the  $i$ -th metastable state. Given a combination of chemical bonds of the molecule  $\{x_i, y_j\}$ , which satisfies the system of equations (4), such CS can be established from the solution of the following system of equations:

$$\begin{aligned} 2x_1 &= 2Ccc2o + 2Ccchh + Cc2oh + 2Cccoh + Ccohh + Cchhh + 2Ccooo; \\ x_2 &= 2Ccc2o + 2Cc2oh + Cccoh + Ccohh + 3Ccooo, \quad x_2 = Ohc + 2 \cdot O2c + Ooc; \\ x_3 &= 2Ccchh + Cc2oh + Cccoh + 2Ccohh + 3Cchhh, \quad Hc = x_3; \\ y_1 &= Och, \quad y_1 = Ho; \\ Ooc &= n3 - Och - O2c. \end{aligned} \quad (6)$$

Here both the presence of broken bonds  $x_{11}$ ,  $y_{11}$ , and the atoms that have them are neglected. However, even with such simplifications, systems (4) and (6) can give non-physical solutions, allowing the formation of topologically disconnected molecular structures.

Another task is to exclude solutions involving mutual switching of double bonds of the C=C and O=O type. Therefore, equations (6) must be supplemented with filters that allow such non-physical solutions to be excluded. In particular:

for carbon atoms (Ccccc, Ccch, Cccco, Ccchh, Cccho, Cccoo) that form double bonds:

$$\begin{aligned} &\text{for odd Ccccc:} \\ 4Ccccc &= 3Cccch + 3Cccco + 2Ccchh + 2Cccho + \\ &\quad + 2Cccoo + Cchhh + Cchho + Cchoo + Ccooo. \\ &\text{for odd Ccch:} \\ 3Cccch &= 4Ccccc + 3Cccco + 2Ccchh + 2Cccho + \\ &\quad + 2Cccoo + Cchhh + Cchho + Cchoo + Ccooo. \\ &\text{for odd Cccco:} \\ 3Cccco &= 4Ccccc + 3Cccch + 2Ccchh + 2Cccho + \\ &\quad + 2Cccoo + Cchhh + Cchho + Cchoo + Ccooo. \\ &\text{for odd Ccchh:} \\ 2Ccchh &= 4Ccccc + 3Cccch + 3Cccco + 2Cccho + \\ &\quad + 2Cccoo + Cchhh + Cchho + Cchoo + Ccooo. \\ &\text{for odd Cccho:} \\ 2Cccho &= 4Ccccc + 3Cccch + 3Cccco + 2Ccchh + \\ &\quad + 2Cccoo + Cchhh + Cchho + Cchoo + Ccooo. \\ &\text{for odd Cccoo:} \\ 2Cccoo &= 4Ccccc + 3Cccch + 3Cccco + 2Ccchh + \\ &\quad + 2Cccho + Cchhh + Cchho + Cchoo + Ccooo \\ &\text{for oxygen atoms (Ooo) that form double bonds:} \\ &\text{for odd Ooo} \\ 2Ooo &= Oco + Oho. \end{aligned} \quad (7)$$

The color sets obtained by solving (6) with restrictions (7) allow us to calculate both the configuration entropy (3) and the thermodynamic function (2) for each molecular structure formed from fragments of the initial glucose molecule. The results of such calculations, performed for molecular structures

formed during the radiation destruction of glucose and retaining its carbon chain containing  $x_1 = 5$  C-C bonds, are given in Table. Within the Ball and Stick model, the bond energy  $E$  of all these structures, calculated using formula (5), provides an estimate of 93.2 eV, with the entropy value varying between 33.5 and 36 J/K.

Color sets of molecular structures that can be formed during the radiation destruction of glucose for  $x_1=5$ . The molecular structures formed when considering the bond energy hierarchy (1), corresponding to glucose and fructose molecules, are highlighted in bold

Ccchh	Ccooh	Ccohh	Ccohh	Cchhh	Ccooo	Cccoo	Och	Occ	Ooc	Hc	Ho	S
1	0	0	1	1	0	3	5	1	0	7	5	34.9
0	0	1	0	2	0	3	5	1	0	7	5	34.2
1	0	1	2	0	0	2	5	1	0	7	5	35.3
0	0	2	1	1	0	2	5	1	0	7	5	35.3
<b>0</b>	<b>0</b>	<b>3</b>	<b>2</b>	<b>0</b>	<b>0</b>	<b>1</b>	<b>5</b>	<b>1</b>	<b>0</b>	<b>7</b>	<b>5</b>	<b>34.2</b>
2	1	0	1	0	0	2	5	1	0	7	5	35.3
1	1	1	0	1	0	2	5	1	0	7	5	36
1	1	2	1	0	0	1	5	1	0	7	5	36
0	1	3	0	1	0	1	5	1	0	7	5	34.9
<b>0</b>	<b>1</b>	<b>4</b>	<b>1</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>5</b>	<b>1</b>	<b>0</b>	<b>7</b>	<b>5</b>	<b>33.5</b>
2	2	1	0	0	0	1	5	1	0	7	5	35.3
1	2	3	0	0	0	0	5	1	0	7	5	34.2

As can be seen, this model calculation uses a significantly larger capacity CS, – 12, than the number of colored atoms in a glucose molecule, 7. This enables the expansion of the range of molecular structures formed from glucose fragments. Such structures differ in their configurational entropy (S), which determines the topology of the arrangement of atoms in molecular structures. Estimates indicate that the entropy term in (2) accounts for a few percent of the energy term, which determines the prevalence of the final molecular structure. The CS of glucose and fructose are highlighted in bold in the table. These chemical compounds differ in their entropy terms, and their CS have more Ccohh and Ccooh atoms, which form energy-intensive bonds between atoms. Such CS groups form the topology of the molecular structure and are markers of biochemical functions that determine their dominance in the metabolic processes of biota.

### CONCLUSIONS

The proposed theoretical approach enables us to study the radiation modification of BOS by examining the representation of the ordering of radiation fragments and the destruction of organic molecules, as well as the formation of metastable states. It has been shown that the probability of realization of the final states of BOS will be determined by the structure of the array of fragments of molecule division, which depends on the method of radiation treatment of the material (type of nuclear particles, their energy spectrum, intensity, etc.) and the nature of the ordering for the occupation of the corresponding metastable state of the substance. The classification of such states and the identification of their peculiarities are possible using structural models, such as Ball and Stick, and color statistics, which enable the determination of the near-order of BOS atoms and the topological features of their structure. It has been shown that BOS belong to the class of non-additive systems, where the degeneration of their energy state is determined by an array of CS atoms that record the

topology of the structure and the configuration of the atoms.

These possibilities are considered using the example of irradiated glucose, where the ordering of the array of molecule fragments leads to the possibility of forming new chemical compounds. It is demonstrated that determining stable chemical configurations formed from glucose cleavage fragments, the process is similar to the Lego game, necessitates an examination of energy and entropy factors, as well as the topology of homo- and hetero-bonds of organogenic atoms. This theory explains the results of radiation experiments on the M-30 microtron, which recorded changes in the physicochemical and mass spectrometric indicators of irradiated glucose [9, 10].

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## РАДІАЦІЙНА ФІЗИКА БІООРГАНІЧНИХ СПОЛУК: ГЛЮКОЗА

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Показано, що радіаційна фізика біоорганічних сполук (БОС) має враховувати етапи утворення та заселення метастабільних станів у процесі релаксації фрагментів поділу молекул (ефект Лего). Теорія таких радіаційних перетворень потребує використання структурних моделей Ball і Stick, а також кольорової статистики, яка фіксує топологію молекул БОС. Розрахунок, проведений на прикладі радіаційної деструкції та упорядкування атомів молекули глюкози, демонструє можливість утворення від 12 та більшого числа хімічних сполук із різними властивостями. Практична реалізація таких сполук потребує використання різних режимів опромінення та післярадіаційного «охолодження». Отримані результати дозволяють пояснити результати радіаційних експериментів БОС, які проведені на мікротроні М-30.