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Моделирование процесса формирования тройных комплексов эссенциального микроэлемента железа с рибофлавином и незаменимыми аминокислотами

Modeling of the formation of triple complexes of the essential trace element iron with riboflavin and essential amino acids

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Аннотация. *В данной работе проведено моделирование процесса формирования тройных комплексов эссенциального микроэлемента железа с рибофлавином и незаменимыми аминокислотами. Моделирование проводилось в программном обеспечении QChem с использованием молекулярного редактора IQmol. Для начала проводилось моделирование молекул незаменимых аминокислот: L-валина, L-лейцина, L-изолейцина, L-метионина, L-треонина, L-лизина, L-фенилаланина и L-триптофана. Моделирование проводилось путём взаимодействия атома железа с различными парами гетероатома азота и енольного кислорода рибофлавина, а также с карбоксильной группой и α-аминогруппой аминокислоты. В результате установлено, что взаимодействие микроэлемента железа с рибофлавином и незаменимыми аминокислотами является энергетически выгодным (ΔE > 2575 ккал/моль) и химически стабильным (0,075 ≤ η ≤ 0,138 эВ). На основе полученных данных определена наиболее вероятная конфигурация молекулярного комплекса – взаимодействие с рибофлавином через N_5 в пиразиновом кольце и енольный кислород, присоединённый к C_4 атому, в пиримидиновом кольце рибофлавина. Молекулярной системой, обладающий наибольшими значениями разницы полной энергии (ΔE = 2577.501) и химической жёсткости (η = 0.138 эВ), а значит наиболее энергетически*

выгодной и химически стабильной, является молекулярный комплекс валинаторибофлавинат меди, в котором взаимодействие железа с рибофлавином происходит через N_5 в пиразиновом кольце и енольный кислород, присоединённый к C_4 атому, в пиримидиновом кольце рибофлавина.

Ключевые слова: микроэлементы, железо, незаменимые аминокислоты, рибофлавин, моделирование

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Abstract. *In this work, we simulated the process of formation of triple complexes of the essential trace element iron with riboflavin and essential amino acids. The simulation was carried out in the QChem software using the IQmol molecular editor. To begin with, the modeling of the molecules of essential amino acids was carried out: L-valine, L-leucine, L-isoleucine, L-methionine, L-threonine, L-lysine, L-phenylalanine and L-tryptophan. Modeling was carried out by the interaction of the iron atom with various pairs of nitrogen heteroatom and enol oxygen of riboflavin, as well as with the carboxyl group and α-amino group of the amino acid. As a result, it was found that the interaction of the trace element iron with riboflavin and essential amino acids is energetically favorable ($\Delta E > 2575$ kcal/mol) and chemically stable ($0.075 \leq \eta \leq 0.138$ eV). Based on the data obtained, the most probable configuration of the molecular complex was determined - interaction with riboflavin through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring of riboflavin. The molecular system that has the highest difference in total energy ($\Delta E = 2577.501$) and chemical hardness ($\eta = 0.138$ eV), and therefore the most energetically favorable and chemically stable, is the copper valinatoriboflavinate molecular complex, in which the interaction of iron with riboflavin occurs through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring riboflavin.*

Keywords: trace elements, iron, essential amino acids, riboflavin, modeling

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Introduction. Iron is a very important trace element that affects a number of metabolic functions of the body: the level of hemoglobin and red blood cells in the blood directly depends on the amount of iron. Hemoglobin contains about 60% of all iron in the body, and its level affects the enrichment of the body with oxygen. Also, metabolic processes that depend on iron include: cholesterol metabolism, conversion of calories into energy and the fight against toxins [1 – 4]. Iron deficiency in the body affects the overall physical condition and immune system. With a slight deficiency, a person is subject to apathy, increased irritability and fatigue. The next stage of deficiency is iron deficiency anemia. With anemia, the following are possible: heart failure,

decreased immunity, leading to worsening chronic diseases, hair loss, increased fatigue, dizziness and a pale appearance. At high levels of this microelement, free radicals can be formed. Impaired iron metabolism in the body is the most common problem of diseases, from anemia to neurodegenerative diseases [5, 6].

The determining factor in the bioavailability of microelements is the area of residence of a person. Differences in soil directly affect the content of microelements in food [7]. Certain regions of the world are deficient in various elements (iron, iodine, cobalt, manganese, copper, molybdenum, selenium, chromium and zinc). Their deficiency can be compensated with the help of food additives that increase the bioavailability of minerals [8 – 10]. Iron deficiency is most common among children and women of reproductive age [11, 12]. The problem of micronutrient deficiency, including iron, is very relevant and requires a solution. One of the ways to reduce element deficiency is the development of highly digestible and low-toxic complexes of various microelements created using the principle of chelation [13]. Chelation chemistry can be defined as an equilibrium reaction between a metal ion and a complexing agent, characterized by the formation of more than one bond between both reactants; As a result, a ring structure is formed into which the metal ion is included [14].

Amino acid and iron chelates are being developed as dietary supplements and therapeutics for the treatment of iron deficiency anemia. For example, iron bis-glycine chelate (*FeBC*) is the most studied and common form used as a supplement. It is best absorbed in milk, dairy products and wheat rolls [15, 16].

Currently, the method of theoretical research of molecules, nanomaterials, nanostructures, etc. is relevant. - computer modelling. Taking into account the operating conditions and environment of nanomaterials, the possibility of modeling and research at the development stage facilitates the prospect of creating various compounds [17, 18].

Thus, the purpose of this work is to model the formation of ternary complexes of the essential microelement iron with riboflavin and essential amino acids to determine the optimal molecular system.

Materials and research methods. *QChem* software using the *IQmol molecular editor*. The calculations were carried out on the equipment of the data processing center (Schneider *Electric)* of the North Caucasus Federal University. At the first stage, the modeling was carried out by interacting the iron atom with various pairs of nitrogen heteroatom and enol oxygen of riboflavin, as well as with the carboxyl group and α-amino group of the amino acid. To compare the results, modeling of individual molecules of essential amino acids was carried out: *L* -valine, *L* -leucine, *L* -isoleucine, *L* -methionine, *L* -threonine, *L* -lysine, *L* -phenylalanine and *L* -tryptophan. As part of the simulation, the values of the total energy of the molecular system (E*)*, the energy of the highest occupied molecular orbital (E $_{HOMO)}$, and the energy of the lowest unoccupied molecular orbital (E $_{LUMO)}$ were considered. Based on the data obtained, the values of the difference between the total energy of the amino acid and the ternary complex (Δ*E)* and chemical hardness (η*)*, calculated using formula 1 [19, 20]:

$$\eta = \frac{1}{2}(E_{LUMO} - E_{HOMO}). \qquad (1)$$

Research results and their discussion. The results of modeling ternary complexes of the essential microelement iron with riboflavin and essential amino acids are presented in Table 1.

Table 1. Results of modeling the interaction of zinc phosphate with biologically active substances

Molecular system	**Interaction of the iron atom with riboflavin**	**E, kcal/mol**	**ΔE, kcal/mol**	**EHOMO, eV**	**ELUMO, eV**	**η, eV**
1	**2**	**3**	**4**	**5**	**6**	**7**
Ferrous lysinatoriboflavinate	–	-496.481	–	-0.177	-0.024	0.077
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3072.932	2576.451	-0.221	0.052	0.100

	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3073.235	2576.754	-0.233	0.033	0.137
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3073.180	2576.699	-0.192	0.045	0.119
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3073.038	2576.557	-0.185	0.000	0.093
Ferrous valinatoriboflavinate	–	-402.112	–	-0.249	0.016	0.133
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-2979.613	2577.501	-0.230	0.046	0.138
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-2979.225	2577.113	-0.234	0.035	0.135
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-2979.304	2577.192	-0.188	0.055	0.122
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-2978.970	2576.858	-0.179	0.017	0.098
Ferrous leucinatoriboflavinate	–	-441.397	–	-0.260	0.006	0.133
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3018.004	2576.607	-0.228	0.034	0.131
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3017.898	2576.501	-0.223	0.014	0.119
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3018.165	2576.768	-0.181	0.053	0.117
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3018.059	2576.662	-0.190	0.007	0.099
Ferrous isoleucinatoriboflavinate	–	-441.394	–	-0.247	0.018	0.133
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3018.335	2576.941	-0.231	0.043	0.137

1	2	3	4	5	6	7
Ferrous isoleucinatoriboflavinate	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3017.962	2576.568	-0.217	0.054	0.136
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3017.885	2576.491	-0.178	0.052	0.115
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3018.218	2576.824	-0.176	0.020	0.098
Ferrous methioninatoriboflavinate	–	-800.251	–	-0.232	0.006	0.119
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3376.764	2576.513	-0.237	0.036	0.137
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3376.684	2576.433	-0.219	0.052	0.136
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3376.630	2576.379	-0.187	0.045	0.116

	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3376.532	2576.281	-0.205	-0.009	0.098
Ferrous threoninatoriboflavinate	–	-438.015	–	-0.248	0.006	0.127
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3015.088	2577.073	-0.229	0.043	0.136
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3014.968	2576.953	-0.228	0.042	0.135
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3014.946	2576.931	-0.182	0.047	0.115
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3013.594	2575.579	-0.214	-0.064	0.075
Ferrous phenylalaninatoriboflavinate	–	-685.684	–	-0.195	-0.035	0.080
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3261.162	2575.478	-0.219	0.055	0.137
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3261.010	2575.326	-0.225	0.028	0.127
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3261.298	2575.614	-0.180	0.048	0.114

1	2	3	4	5	6	7
Ferrous phenylalaninatoriboflavinate	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3261.255	2575.571	-0.190	0.006	0.098
Iron tryptophanatoriboflavinate	–	-554.424	–	-0.240	0.002	0.121
	Through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3130.704	2576.280	-0.228	0.043	0.136
	Through N_3 and enol oxygen attached to the C_4 atom in the pyrimidine ring	-3130.226	2575.802	-0.224	0.004	0.114
	Through N_3 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3130.597	2576.173	-0.194	0.037	0.116
	Through N_1 and enol oxygen attached to the C_2 atom in the pyrimidine ring	-3130.577	2576.153	-0.172	0.019	0.096

Based on the data obtained, we can conclude that the interaction of the trace element iron with riboflavin and essential amino acids leads to a decrease in the total energy of the molecular complex (ΔE > 2575 kcal/mol) and an increase in the value of chemical hardness ($0.075 \leq \eta \leq 0.138$ eV), which indicates a high energy benefit and stability of the interaction process. It has also been established that ternary complexes of the microelement iron with riboflavin and essential amino acids, formed through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring of riboflavin, have increased chemical hardness compared to amino acids, which indicates greater probability of the formation of this interaction.

As a result of quantum chemical calculations, it was established that the optimal interaction of the microelement iron with riboflavin and essential amino acids is iron valinatoriboflavinate, in which the interaction of the iron atom with riboflavin occurs through N 5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring of riboflavin. A model of this molecular system is presented in Figure 1.

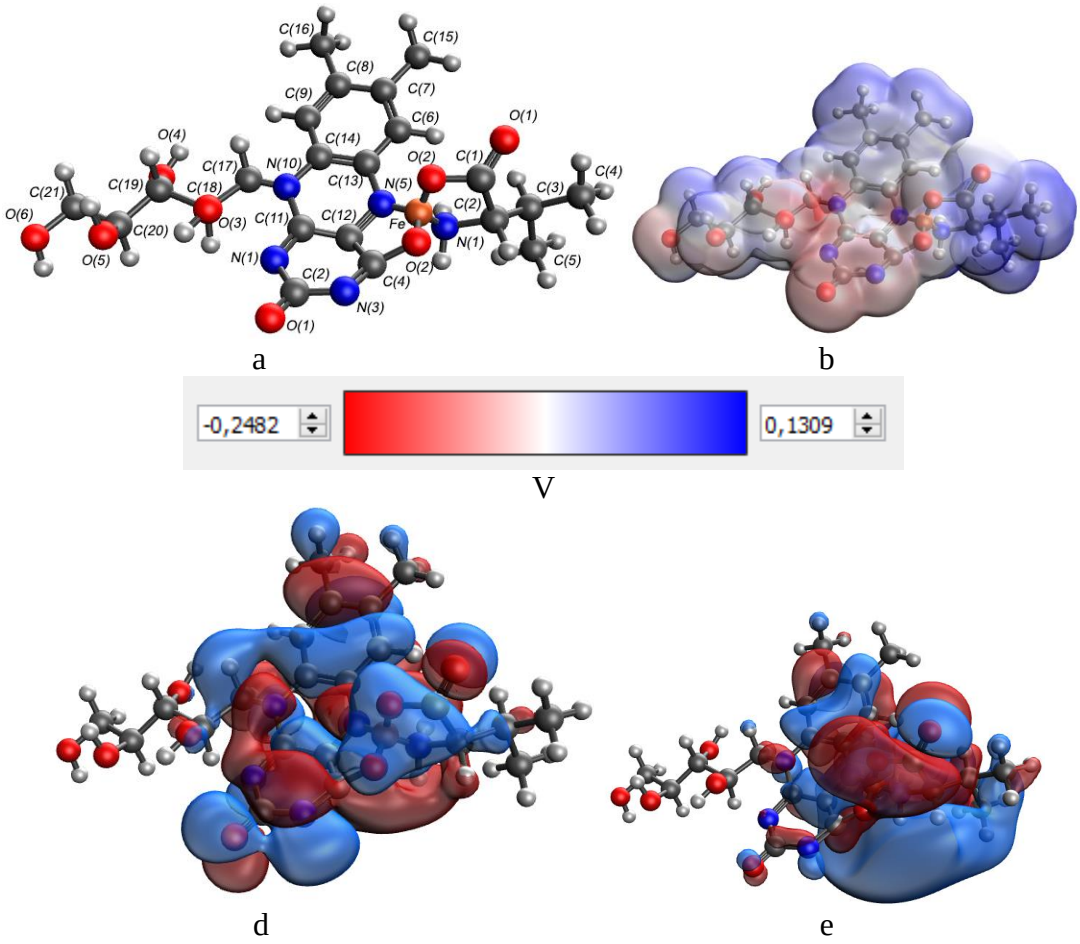

Figure 1. Results of modeling the molecule of iron valinatoriboflavinate, in which the interaction of iron with riboflavin occurs through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom in the pyrimidine ring of riboflavin:
a – model of a molecular complex; b – electron density distribution; c – gradient of electron density distribution; d – highest occupied molecular orbital; d – lowest free molecular orbital

Iron valinatoriboflavinate, in which the interaction of the iron atom with riboflavin occurs through N_5 in the pyrazine ring and enol oxygen attached to the C_4 atom, in the pyrimidine ring of riboflavin has the highest values of the difference in total energy (ΔE = 2577.501) and chemical hardness (η = 0.138 eV), as a result of which we can conclude that this interaction is more energetically favorable and chemically stable.

Conclusion. As a result of modeling the process of formation of ternary complexes of the essential microelement iron with riboflavin and essential amino acids, the effectiveness of the formation of stable chelate complexes was determined. It has been established that with the greatest probability the formation of chelate complexes occurs through the interaction of the iron atom with the carboxyl and α-amino group of the amino acid and through N_5 in the pyrazine ring and enol oxygen attached to *the* C_4 atom in the pyrimidine ring of riboflavin, and also that the optimal molecular complex is ferrous valinatoriboflavinate.

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