

Justification of applying collectors from the class of unsaturated tertiary alcohols in flotation of gold-bearing sulphide ores

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Given in the paper are the results of calculations of the simplest quantum-chemical parameters of the structures under consideration formed in the *mineral–flotation reagent* system by the method of molecular mechanics (MM2). Theoretical computations on the interaction mechanism between unsaturated alcohols and gold atoms in various energy states are formulated. There were determined the causes and ascertained the regularities of selective fixation of reagents with triple bonds, which are tertiary alcohols, on the surface of liberated grains of gold and gold-containing minerals. Presented are the results of practical flotation studies, which have showed that the use of acetylene reagents allows one to selectively obtain additional gold recovery from gold-bearing ores from 2.4 to 5.0% relative to the basic flotation indicators.

Key words: flotation, chemisorption, complex compounds, electronic structure, nucleophilic interactions, electrophilic interactions, reagents with dangling bonds, gold-bearing ore.

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Introduction

Reducing the level of irretrievable losses of metals during processing by transferring them into a potentially recoverable reserve, which can be more than 20%, may be achieved by selecting the optimal combination of reagents in flotation modes, that provides an additional extraction of target metals and, in that way, a reduction of their losses with regard to the basic level of recovery indicators [1–6].

Analysis of the results of our earlier studies has showed that the use of tertiary alcohols with triple bonds as additional collectors in the flotation modes of various types of non-ferrous and precious metal ores allows us to obtain in some cases a significant increase in gold extraction [7–9].

Obtaining stable results on selective additional gold extraction has required the formulation of theoretical points on the influence interpretation for DC and DC-80 alcohols with triple bonds on the flotation indicators of gold-bearing sulphide ores. The sulphide minerals surrounding gold contribute to additional recovery, however, we were interested in the interaction of acetylene reagent molecules with gold cations located in the structure of sulphide minerals.

The objective of the present research in the theoretical aspect was to identify the cause-effect relations

between the properties of reagents based on alcohols with triple bonds and metal cations of minerals as well as to establish the interaction regularities of reagents of the acetylene derivatives class with sulphide ore minerals during flotation.

Methods and discussion of the results of modeling quantum-mechanical parameters and investigating the interaction regularities of reagents of the acetylene derivatives class with gold atoms

Modeling and calculating elementary quantum-chemical parameters of the structures under study formed in the flotation system were performed applying the methods of molecular mechanics, semiempirical and ab initio quantum-chemical techniques with the use of ChemBio3D 11.0 package included into the ChemBioOffice 2008 suite of scientific applications (for Windows), developed by CambridgeSoft Corporation.

The MM2 technique was chosen as an intermediate step for preliminary structure optimization of the complex organic compounds. The MM2 (Molecular mechanics) technique is designed for modeling and calculating organic molecules. It takes into account the potential fields generated by all the atoms of the calculated system, in comparison with other methods of molecular mechanics [10].

Because of the collision of a DC molecule, which is a tertiary acetylene alcohol, with the surface of sulphide minerals containing gold cations, the formation of π -complexes can proceed in two ways, namely, nucleophilic and electrophilic. For calculated 2D models of DC molecules, in **Fig. 1** there are shown the ball-and-stick 3D models of (DC+Au) complexes in (HUMO) state (**Fig. 2, a**) and in (LUMO) state (**Fig. 2, b**).

For calculated 2D models of DC-80 reagent molecules (**Fig. 3**), there are shown the ball-and-stick 3D models of (DC-80+Au) π -complexes formed by a nucleophilic mechanism in the states: (HUMO) — **Fig. 4, a** and (LUMO) — **Fig. 4, b**.

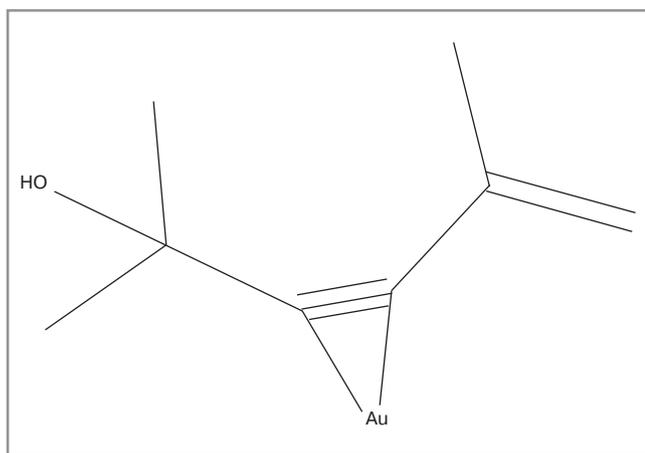


Fig. 1. Optimized computational 2D model of π -complex with DC reagent, formed by a nucleophilic mechanism

Table 1 aggregates calculated data of the total potential energies of π -complexes formed by (DC+Au) and (DC-80+Au) molecules, according to a nucleophilic mechanism.

Notation conventions for Table 1 are as follows: N is the selected molecular orbital in the HUMO/LUMO state, in which the ionization field is measured by energy in electronvolts (eV).

Calculated data of the total potential energies of complexes formed by (DC+Au) and (DC-80+Au) molecules listed in Table 1 show that they have a very close value.

A DC reagent can form electrophilic π -complexes according to the model shown in **Fig. 5**; the ball-and-stick 3D models of (DC-80+Au) π -complexes are shown in **Figs. 6, a, b**.

Table 2 represents calculated data of the total potential energies of (DC+Au) π -complexes formed by an electrophilic mechanism.

Comparison of calculated data of total potential energies of π -complexes showed that the energy of complexes formed by (DC+Au) molecules by an electrophilic mechanism (Table 2) is an order of magnitude lower than the energy indicators of π -complexes formed by (DC+Au) and (DC-80+Au) molecules by a nucleophilic mechanism (Table 1).

Our earlier instrumental spectral IR studies and quantum-chemical calculations have showed that chemical bonds are formed on the surface of floatable sulphide minerals as a result of adsorption of reagent molecules with triple bond [11].

All theoretical computations were formulated based on fundamental classical concepts accepted in chemistry and

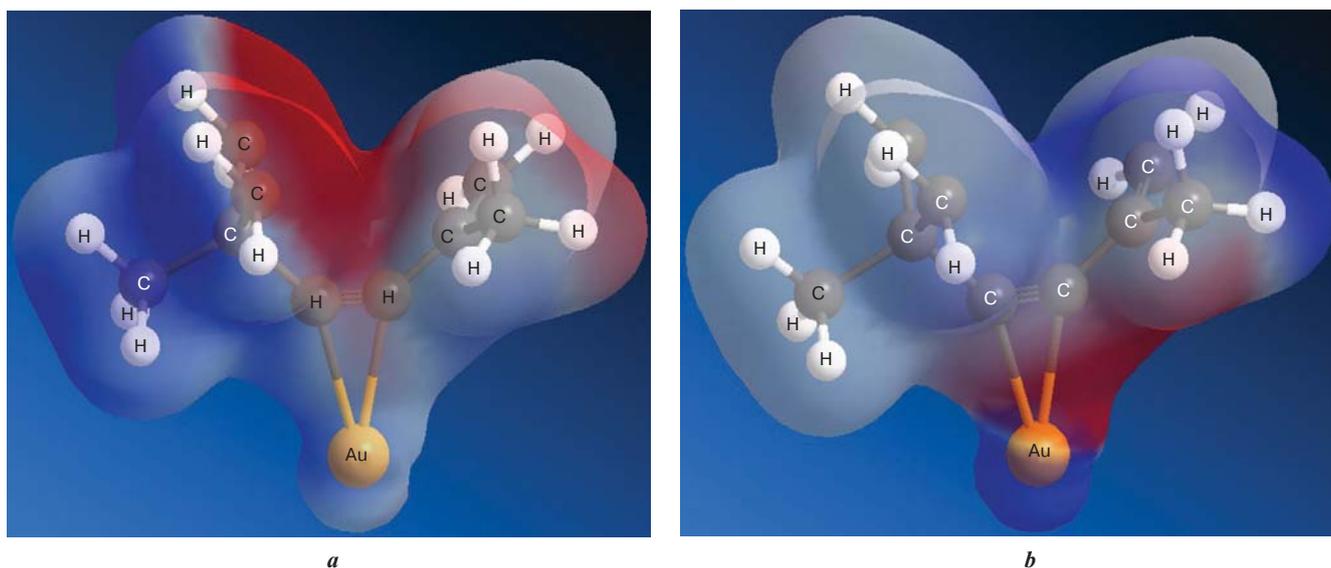


Fig. 2. Optimized computational 3D models of π -complexes with DC reagent formed by a nucleophilic mechanism in the states: *a* – (HUMO), *b* – (LUMO)

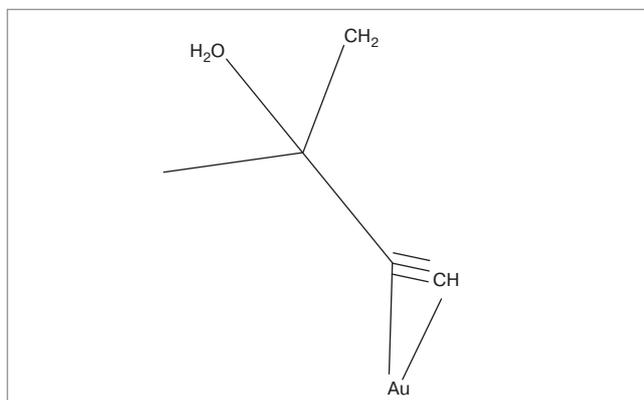


Fig. 3. Optimized computational 2D model of π -complex with DC-80 reagent, formed by a nucleophilic mechanism

physics. Based on the electronic structure, models of the proposed complexes for alkynes $[\text{Au}]\text{C}_2\text{H}_2$ and alkenes $[\text{Au}]\text{C}_2\text{H}_4$ were constructed, as well as the distinction in kind between the mechanism of their action in the process of flotation was formulated.

We believe that the reason for additional gold recovery during flotation with the use of unsaturated alcohols with triple bonds in the basic flotation modes is the specific behavior of alkynes, leading to the formation of π -complexes in the course of collision of reagent molecules with the surface of sulphide minerals containing gold cations.

Meanwhile, the mechanism of forming π -complexes, analyzed from the standpoint of electronic structure, has shown that the metal cation have to be in the ground state ($B^*=4$) according to the electronic formula 79Au :

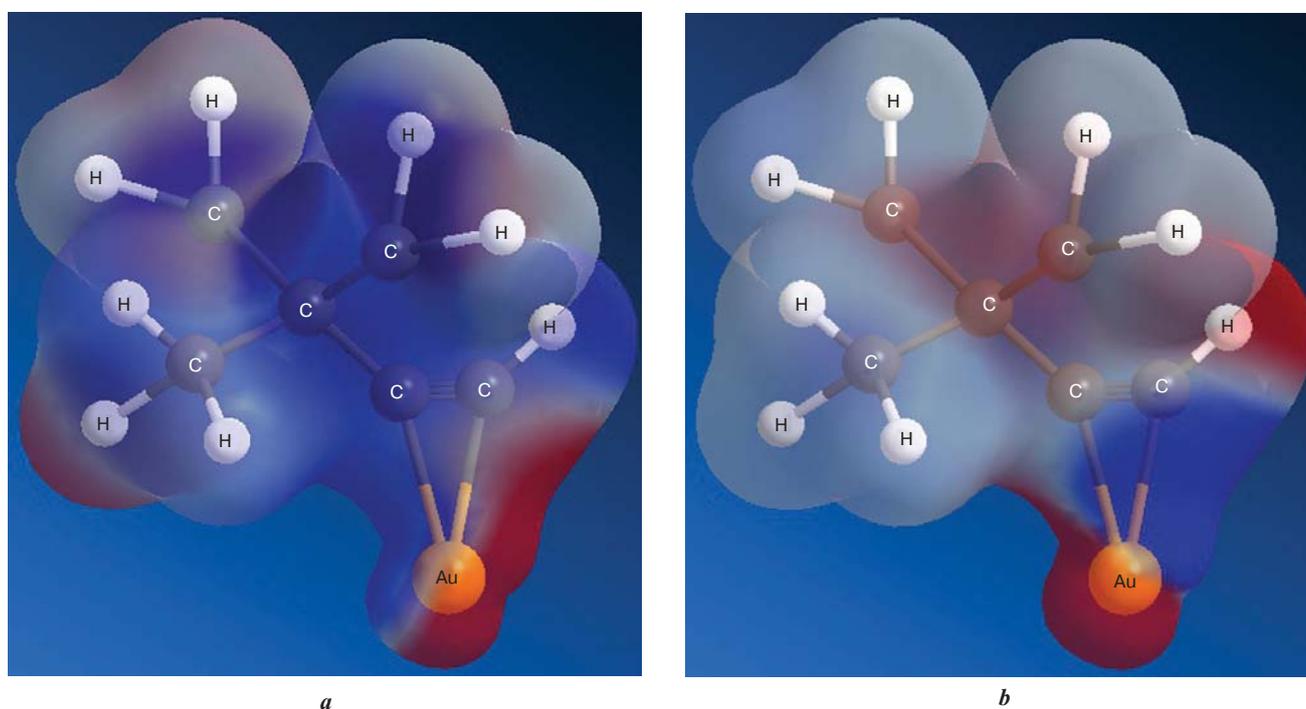


Fig. 4. Optimized computational 3D models of π -complexes with DC-80 reagent formed by a nucleophilic mechanism in the states: *a* – (HUMO), *b* – (LUMO)

Table 1

Values of calculated indices for total potential energy of π -complexes formed by a nucleophilic mechanism

Calculated parameters	Index value for complexes	
	DC+Au [N-31], [-5.683 eV]	DC-80+Au [N-24], [-5.703 eV]
Stretch	133.7206	133.1592
Bend	2437.9953	2433.1112
Stretch-Bend	-28.3894	-27.4191
Torsion	0.4823	1.2066
Non-1.4 VDW	-3.7237	-1.6617
14 VDW	4.2952	2.4300
Dipole/Dipole	0.9781	0.9926
Total energy, kcal/mol	2545.3584	2541.8187

$[\text{Xe}]5d^{10}6s^1p^2$ in order to form $[\text{Au}]\text{C}_2\text{H}_2$ complex. In this case, formation of π -complexes shown in Fig. 2, *a* and Fig. 3, *a* will proceed by a nucleophilic mechanism shown in Fig. 7, which has the structure of electron clouds in (HUMO) state.

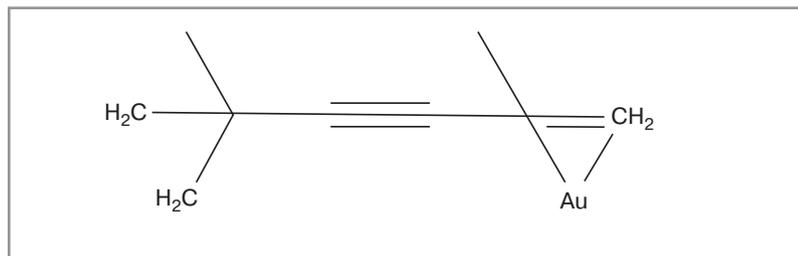
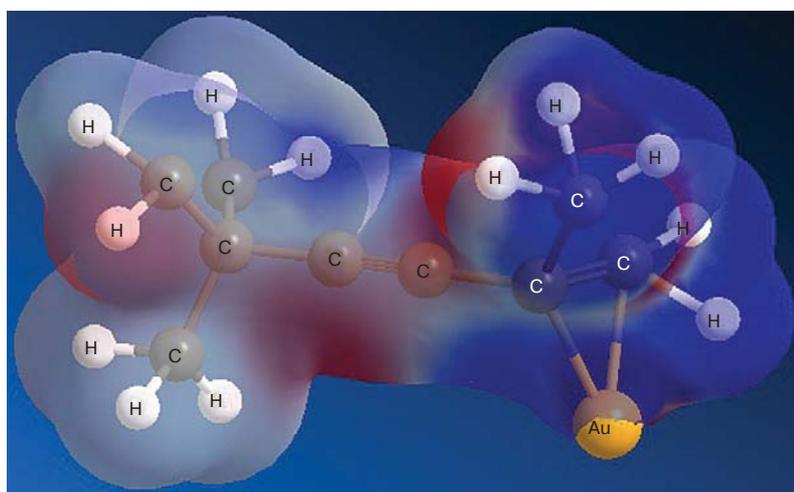
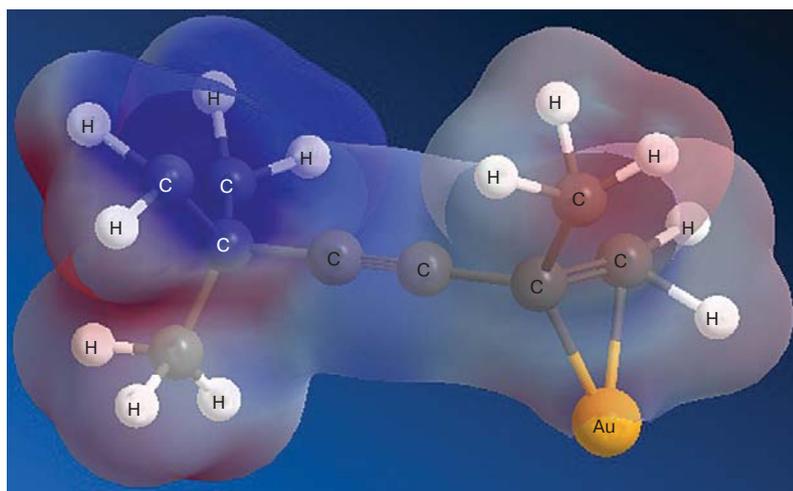


Fig. 5. Optimized computational 2D model of formation of π -complex with DC reagent by an electrophilic mechanism



a



b

Fig. 6. Optimized computational 3D models of π -complexes with DC reagent formed by a nucleophilic mechanism in the states: *a* – (HUMO), *b* – (LUMO)

The ground (HUMO) state is characterized by the nucleophilic substitution in the MO method, when electrons are transferred from nucleophile (C_2H_2) to the lowest free MO of the ligand (Fig. 7), and contact between the reagent molecule and metal cation is necessary. Additional energy is not required for the formation of a nucleophilic complex.

At the same time, the formation of $[\text{Au}]\text{C}_2\text{H}_2$ complex can also take place when the metal cation is in an excited ($B^* = 5$) state with the electronic formula ${}_{79}\text{Au}^*$: $[\text{Xe}]5d^96s^1p^1$. In this case, the formation will proceed according to the electrophilic mechanism shown in Fig. 8. The structure of electron bunches in (LUMO) state is shown in Fig. 2, *b* and Fig. 3, *b*.

The excited state of a metal atom is characterized by the electrophilic substitution reaction in the MO method, when electrons are transferred from the lowest unfilled MO of a ligand to unfilled MO of a nucleophile (C_2H_2). Formation of an electrophilic π -complex requires external additional energy so that electrons from $5d$ orbital can move to an unoccupied valency $6d$ orbital, and a complex could form upon collision with a reagent molecule, as shown in Fig. 8.

Formation of a π -complex for DC reagent can similarly proceed by an electrophilic mechanism not only with a triple, but also with a conjugated double bond existing in the structure of its molecules.

The nucleophilic model is constructed taking into account the substitution in the MO method, when electrons are transferred from a nucleophile (C_2H_2) to the lowest free MO of a ligand. The excited state is characterized by electrophilic substitution in the MO method, when electrons are transferred from the lowest unfilled MO of the ligand to the unfilled MO of the nucleophile (C_2H_2) [12].

The universal property of acetylene reagents is the priority of nucleophilic nature (reagents transfer electrons) of substitution with metal sulphides in the ground state, and electrophilic substitution (reagents receive electrons from transition metal atoms) with metal sulphides in the excited state, typical for alkene hydrocarbon atoms.

Based on the presented data, it is possible to formulate some differences in formation of MeS π -complex in (HUMO) and (LUMO) states: for the

(HUMO) state, the lower (close to the core) location of p -orbitals provide greater stability of the formed π -complexes than in the (LUMO) state by the amount of energy equal to $\Delta+1.2$ eV, and, besides, the arrangement of molecular orbitals in (HUMO) state ensures selectivity with respect to metal (in particular, gold) cations, what is relevant for providing additional selective recovery and separation of minerals in the flotation process.

It is known that all chemical reactions proceed through the elementary stage of the formation of an "active complex" in the transition state, when chemical bonds in the starting substances have already been broken, and new ones are still at the stage of formation. Between the stages of initial and final states, it is necessary to overcome an energy barrier, the height of which corresponds to the activation energy of this process, therefore, the smaller the barrier value, the faster the chemical reaction proceeds [13].

It is necessary to note the new fundamental data obtained by theoretical physicists and confirmed by practical calculations on electronegativity. Therefore, for example, according to L. Pauling, it has indicators for Au = $2.54 \text{ eV}^{-1/2}$, Fe = $1.8 \text{ eV}^{-1/2}$ and S = $2.5 \text{ eV}^{-1/2}$ [14]; the refined values of electronegativity on the thermochemical scale are a dimensionless number for Au = 2.81, Fe = 2.32 and S = 3.44. In the new interpretation, all metals have a lower electronegativity than boron and hydrogen, which is in better agreement with chemical intuition than the Pauling values [15–16]. Electronegativity is a fundamental property of chemical elements that characterizes the prevalence of an atomic bond, if its value is less than 1.7, or an ionic bond, if its value is greater than 1.7.

The addition of electrons is accompanied by a decrease in

Table 2

Values of calculated indices for total potential energy of π -complexes formed by an electrophilic mechanism

Calculated parameters	Index value for π -complexes
	DC+Au
Stretch	27.8788
Bend	213.4842
Stretch-Bend	-11.2841
Torsion	42.5020
Non-1.4 VDW	-1.6636
14 VDW	3.0133
Dipole/Dipole	0.0480
Total energy, kcal/mol	273.9786

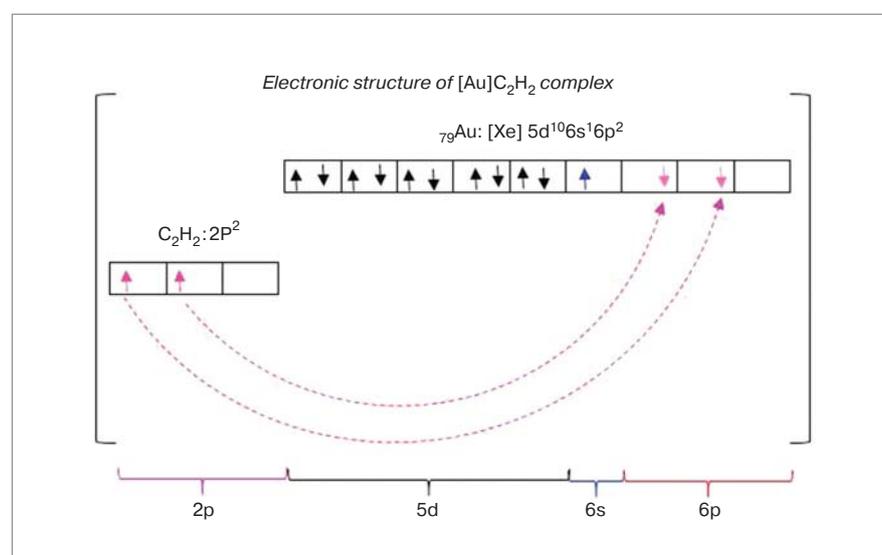


Fig. 7. Nucleophilic substitution in π -complex $[\text{Au}]\text{C}_2\text{H}_2$

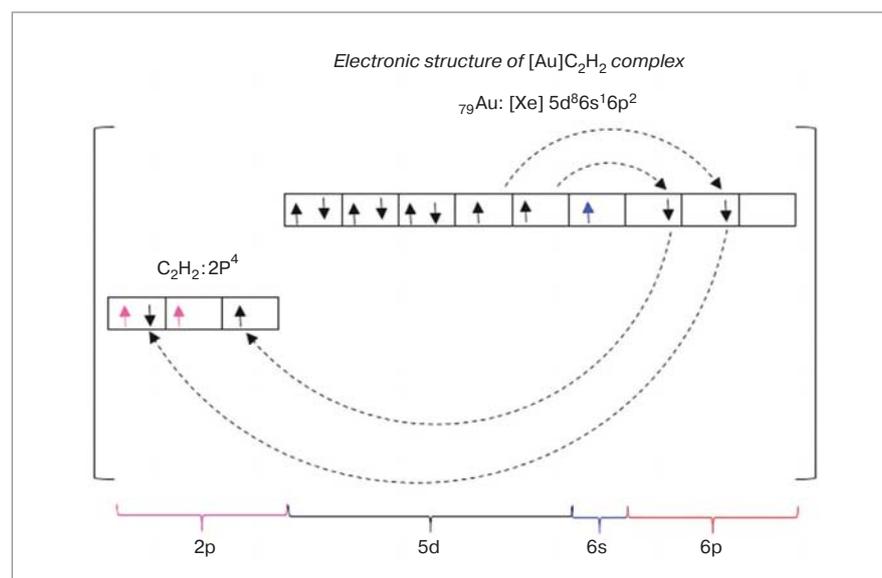


Fig. 8. Electrophilic substitution in π -complex $[\text{Au}]\text{C}_2\text{H}_2$

oxidation degree of gold atoms; these restrictive conditions for alcohol compounds with triple bonds determine their selectivity with respect to floatable minerals. The refined values of electronegativity help to more accurately predict the result of the interaction of reagent molecules with the surface of floatable minerals.

When reagents interact with floatable minerals, it is necessary to take into account such an important property of some metals as the skip of electrons from an s -sublevel of the outer layer to a d -sublevel of the previous layer, which is a deviation from the general rules for most elements in the filling of $1s, 2s, 2p, 3s, 3p, 4s, 3d$, etc. electronic shells [17–18].

This is caused by the fact that it is more energy-advantageous for such atoms when there is a half or fully filled sublevel $p^3; p^6; d^5; d^{10}; f^7; f^{14}$ in the atom [19].

In the atoms of elements in which the electron shell structure is close to the aforesaid, premature filling of the d -sublevel can be observed due to an irregular skip of an electron from the external s -sublevel to the underlying (pre-external) d -sublevel (regular skips).

This rearrangement proceeds in the atomic valence band (AVB) of the electron shell, which can be rearranged during the formation of a chemical bond. AVB includes, as a rule, an external s -sublevel (ns) and the sublevel that is filled in for this element. For gold, the theoretical structure of the atom is $5d^9 6s^2$, whereas the actual one is $5d^{10} 6s^1$.

Experimental verification and results of process approbation of the use of collecting reagents based on acetylene hydrocarbons during flotation of gold ores

The selective effect of DMIPEC reagent in relation to gold was shown during the flotation of copper ores

of planned processing of the Novy Sibay and Nizhnyaya Zalezhd deposits, where the increase in gold extraction averaged 2.5–5.0%; during the flotation of gold ore of the Akbakaysky mining and processing enterprise (Republic of Kazakhstan), the quality of the concentrate was improved by 20 g/t; an additional recovery by almost 10.6% was obtained; the flotation rate has increased by 65% [20].

Application of DMIPEC reagent in the flotation mode of the Northern Aktash deposit gold-antimony ore has showed a positive effect on the recovery rates and flotation time (Table 3).

Additional gold extraction amounted to (from the operation): 26.3% into the rougher flotation concentrate, 2.4% into the total concentrate.

Subsequent studies of the selective action of reagents of the unsaturated alcohols class with triple bonds during flotation of the Bereznyakovsky deposit gold-quartz low-sulphide ore and the Olympiadinsky deposit gold-bearing ore were carried out using DC-80 reagent.

The use of DC-80 acetylene reagent in the process of flotation of the Bereznyakovsky deposit gold-quartz low-sulphide ore allows to significantly (up to 6%) reduce the loss of potentially recoverable metal reserves in the intercycle flotation (the research was carried out in common with the Institute of Comprehensive Exploitation of Mineral Resources Russian Academy of Sciences and Nosov Magnitogorsk State Technical University in 2021) [21].

The results of the DC-80 reagent selective action during the flotation of the Olympiadinsky deposit gold-bearing ore are shown in Table 4.

Total Cu recovery was 9.25% in the basic mode and 11.01% in the mode with DC-80 reagent; an additional extraction of gold was 1.76%. It should be noted that

Table 3
Flotation indicators of the Northern Aktash deposit gold-antimony ore

Product name	Yield, %	Content, g/t		Recovery, %		Reagent consumption
		Au	Sb	Au	Sb	
Rougher flotation concentrate	6.1	10.2	2.86	30.7	64.4	<i>Basic experiment.</i> Rougher flotation: Pb(NO ₃) ₂ – 150 g/t, CuSO ₄ – 300 g/t, BPX – 200 g/t, T-92 – 40 g/t
Scavenger flotation concentrate	26.6	3.88	0.269	51.0	26.4	
Total concentrate	32.7	5.05	0.75	81.7	90.8	
Tailings	67.3	0.55	0.037	18.3	9.2	
Initial	100.0	2.02	0.27	100.0	100.0	
Rougher flotation concentrate	14.4	8.4	1.42	57.0	74.0	Rougher flotation: Pb(NO ₃) ₂ – 150 g/t, CuSO ₄ – 300 g/t, BPX – 200 g/t, DC – 40 g/t
Scavenger flotation concentrate	22.1	2.6	0.225	27.1	18.0	
Total concentrate	36.5	4.88	0.71	84.1	92.0	
Tailings	63.5	0.53	0.035	15.9	8.0	
Initial	100.0	2.12	0.28	100.0	100.0	

Table 4
Indicators of flotation of the Olympiadinsky deposit gold-bearing ore

Products	Yield, %	Content, g/t	Recovery, %	Conditions
		Au		
Basic experiment				
Concentrate of the 1 st rougher	3.16	21.20	5.49	BPX – 220 g/t Flotanol C07 – 8 g/t CuSO ₄ – 70 g/t Pulp density – 29.65 % Time: – of rougher flotation – 10 min, – of scavenger flotation – 15 min.
Concentrate of the 2 nd rougher	1.69	14.10	1.95	
Concentrate of the 1 st cleaner	3.61	3.92	1.16	
Concentrate of the 2 nd cleaner	0.78	10.20	0.65	
Final tailings	97.1	2.25	51.0	
Feed ore	100	4.28	100	
Flotation with DC-80 reagent				
Concentrate of the 1 st rougher	4.18	28.10	6.51	BPX – 110 g/t Flotanol C07 – 8 g/t DC-80 – 110 g/t CuSO ₄ – 70 g/t Pulp density – 29.65 % Time: – of rougher flotation – 10 min, – of scavenger flotation – 15 min.
Concentrate of the 2 nd rougher	3.19	17.40	3.08	
Concentrate of the 1 st cleaner	2.95	5.23	0.86	
Concentrate of the 2 nd cleaner	0.69	14.70	0.56	
Final tailings	97.1	2.25	50.4	
Feed ore	100	4.26	100	

in the case of flotation with DC-80 reagent, the consumption of the main collector – butyl potassium xanthate (BPX) – was reduced by 50%. It is noteworthy that application of DC-80 reagent during flotation does not have a deleterious effect on the further bacterial leaching of concentrates.

Conclusion

The results of the presented studies have made it possible to put forward a hypothesis that allows us to objectively interpret the additional selective extraction of gold-containing sulphide minerals, from 2.4 to 5.0%, in the case of inclusion of reagents based on alcohols with triple, as well as triple and conjugated double bonds, into the reagent modes of flotation.

The most significant conclusion from the results of the conducted investigations is that the molecules of reagents with triple bonds have a selective ability to form π -complexes with metal atoms of floatable sulphide minerals. Such π -complexes, when metal atoms are in the “basic” energy state, are formed by a nucleophilic mechanism. When metal atoms are in an “excited” energy state, π -complexes are formed by an electrophilic mechanism. Thus, a rise in the flotation activity of sulphide minerals and gold, which provides an increase in metal extraction, is associated with the selective action of reagents with triple bonds, which are tertiary alcohols, with respect to the atoms of sulphide metals in various energy states.

Fulfilled calculations of the simplest quantum-chemical parameters of the structures under study, the analysis of the electronic structure of π -complexes formed by electrophilic and nucleophilic mechanisms, as well as formulated theoretical assumptions about the selective interaction between molecules of unsaturated tertiary alcohols and ions of floatable minerals are confirmed by the results of numerous flotation experiments. The use of such reagents with triple bonds increases the sulphide ore flotation efficiency, which will provide an increase in the extraction of target metals due to the flotation of the previously unrecoverable part of minerals.

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