Dadze T.P., Kashirtseva G.A., Akhmedzhanova G.M. Complexation of gold in acid sulfide-bearing solutions at T=300°C and P=300 atm.

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This investigation was aimed at the study of gold solubility and the forms of its existence in acid sulfidebearing solutions under hydrothermal conditions.

Nowadays some authors suggest that in acid sulfidebearing solutions gold occurs in the form of neutral complexes AuHS^o (Renders and Seward, 1989; Shenbergar and Barnes, 1989; Pan and Wood, 1994; Gibert et al., 1998; Palianova et al., 1999) and HAu(HS)^o₂ (Wood et al., 1987; Hayashi and Ohmoto, 19991 etc.) forming by the reactions:

 $Au_{(S)} + H_2S = AuHS^{\circ} + 0.5H_2$ (1) (1)

 $Au_{(S)} + 2H_2S = Hau(HS)^{\circ}_2 + 0.5H_2$ (2) The main form of gold existence in near neutral solu-

tions is a charged complex $Au(HS)^{-}_{2}$ (Seward, 1973; Renders and Seward, 1989; Shenbergar and Barnes, 1989; Pan and Wood, 1994; Benning and Seward, 1996, Zotov et al., 1996; Dadze et al., 2000) forming by the reaction: Au(s) + H₂S + HS⁻ = Au(HS)⁻₂ + 0.5 H₂ (3)

The forms of gold existence in acid sulfide-bearing solutions have been determined by the solubility method at $T=300^{\circ}$ C and P=300 atm. Thioacetamide (CH₃CSNH₂) which in acid solutions hydrolyses to release hydrogen sulfide in stoichiometric amounts was used as a source of hydrogen sulfide. Solutions with constant pH=1.44 were prepared. For this purpose HCl concentrations were calculated which at different content of thioacetamide provided the given acidity at experimental parameters. The concentration of hydrogen sulfide changed from 0.05 up to 0.5 m. The experimental technique and analytical determination of gold in the solution are considered in detail in Dadze et al., (2000).

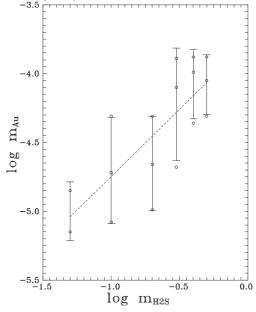


Fig.1. Dependence of gold solubility on hydrogen sulfide concentration in solutions with pH=1.44 at $T=300^{\circ}C$, P=300atm.

*
$$K_1 = \frac{m_{Au} \cdot a_{H_2}^{1/2}}{a_{H_2S}}$$
, average value $K_1 = 0.87 \cdot 10^{-5}$

Shown in fig.1 are experimental data on gold solubility depending on hydrogen sulfide concentration in the solution with constant pH=1.44 at 300°C and 300 atm. Gold concentration in the solution obviously increases with concentration of hydrogen sulfide. The equation of linear dependence of gold solubility in acid solutions on the concentration of hydrogen sulfide has been derived: $Logm_{Au} = -3.772 + 0.975 \ logm_{H2S} \pm 0.31$.

The slope factor of the solubility curve close to unit suggests that gold exists in acid sulfide-bearing solutions at experimental parameters mainly as a neutral complex AuHS^o, forming by reaction (1).

Table 1 presents the calculated constant of the reaction of AuHS^o formation based on experimental data on gold solubility and activity of the particles participating in the reaction, calculated by Shvarov's program (1999).

Gibbs energies (-32327 j/mole) and total constant of electrolytic dissociation (pK = 15.44) of the complex AuHS^o have been calculated from the constant (K₁ = $= 0.87 \cdot 10^{-5}$) and Gibbs energies of the reacting substances - at T=300°C, P=300 atm.

Table 1. Calculation of the reaction (1) constant from experimental data.

М	$m_{Au} \bullet 10^5$	activity o ticles	*K1	
CH ₃ CSNH ₂	exper.	H ₂ S(aq)	H ₂ (aq)	•10 ⁵
0.1	2.52	0.0991	0.00153	1.00
0.2	3.550	0.1973	0.00152	0.70
0.3	7.62	0.2939	0.00151	1.01
0.4	9.27	0.3892	0.00151	0.92
0.5	9.03	0.4835	0.00150	0.72

As we reported earlier (2000), at T= 300° C, P=300 atm and pH \geq 6.5: 7.5 gold occurs in sulfide-bearing solutions mainly as a charged complex Au(HS)⁻₂ forming by reaction (3).

Table 2. Concentration of gold complexes calculated from the constants of the reactions of their formation in solutions with pH= $5.5 \text{ K}_1=0.87*10^{-5}$, $\text{K}_3=1.86\cdot10^{-3}$.

М	activity of the particles	$*m \bullet 10^4$	**m•10 ⁴	$\Sigma m_{Au} \bullet 10^4$	$m_{Au} \bullet 10^4$

CH ₃ CSNH ₂	H ₂ S(aq)	HS	H ₂ (aq)	Au(HS) ₂	AuHS°	***calcul.	experim.
0.1	0.0977	0.001817	0.001806	0.078	0.200	0.278	0.210
0.2	0.1952	0.003105	0.001816	0.266	0.398	0.664	0.745
0.3	0.2916	0.004471	0.001936	0.5540	0.577	1.130	0.790
0.4	0.3870	0.005871	0.002075	0.933	0.739	1.6726	1.855
0.5	0.4814	0.007285	0.002211	1.395	0.891	2.286	2.240

<u>Note:</u> * $m_{Au(HS)_{2}^{-}} = \frac{K_{3} \cdot a_{H_{2}S} a_{HS^{-}}}{a_{H_{2}}^{1/2}}, * * m_{Au(HS)} = \frac{K_{1} \cdot a_{H_{2}S}}{a_{H_{2}}^{1/2}}, * * Sum of complexes m_{Au(HS)2^{-}} + m_{Au(HS)2^{$

Analysis of the literature and our data with a certain assurance allows to suggest that both complexes $Au(HS)_2$ and $AuHS^\circ$ are present in sulfide-bearing solutions in a weakly acid medium. Based on experimental data on gold solubility in sulfide-bearing solutions with pH=5.5 at T=300°C, P= 300 atm (Dadze et al., 2000) the amount of each complex has been calculated from the constants of the reactions of their formation and activities of the reacting particles.

The overall result of the calculated gold concentrations agrees well with the experimental data (table 2) which proves the reliability of the obtained constants of the complexation reactions.

However, our investigations allowed to conclude that in acid sulfide-bearing hydrothermal solutions a neutral complex AuHS^o is responsible for the gold transport.

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