

Dadze T.P., Kashirtseva G.A. Solubility of gold and conditions of its deposition in sulfide-bearing systems.

Institute of Experimental Mineralogy kash@iem.ac.ru

Geological observations and theoretical calculations testify that the presence of gold in sulfide-bearing solutions, its transport and deposition are guaranteed by the interrelation of some basic parameters – Eh, pH, concentration of sulfide sulfur, T and P.

However, the explanation of the geochemical peculiarities of gold requires the experimental study of physical-chemical conditions of solubility, transport and concentration of this metal in the course of the hydrothermal process.

Gold solubility in sulfide-bearing solutions has been experimentally studied in the temperature range 200-350°C and a pressure along the line of saturated water vapor and 300 bar at different concentrations of sulfide sulfur in the solutions from 0.05 to 0.5 m. The forms of gold existence in sulfide-bearing solutions of various acidities were determined by the solubility method. The source of hydrogen sulfite was thioacetamide

(CH_3CSNH_2) which in acid and weakly acid solutions hydrolyzes with formation of hydrogen sulfite in stoichiometric amounts. The solutions with starting pH=7.4 and 8.96 were prepared with the addition of NaOH of low concentration into twice-distilled water. A series of experiments has been carried out with the addition of Na_2SO_4 into the system. For the solutions with the constant value of pH=1.44 to be prepared, HCl concentrations were calculated by the Shvarov's program [2] which at different thioacetamide contents provided given acidity at experimental parameters. Experiments were run in autoclaves of titanium alloy BT-8 20 cm³ in volume. The pressure in autoclaves was set by space factor the from PVT data for water. A plate of gold (99.99) 10x5x0.1 mm in size was hung on the obturator in the upper part of the autoclave. The content of gold in the solution after the experiment was determined by an atomic absorption method in the acetylene-air flame on the AAS-N apparatus at a wavelength 242.8 nm.

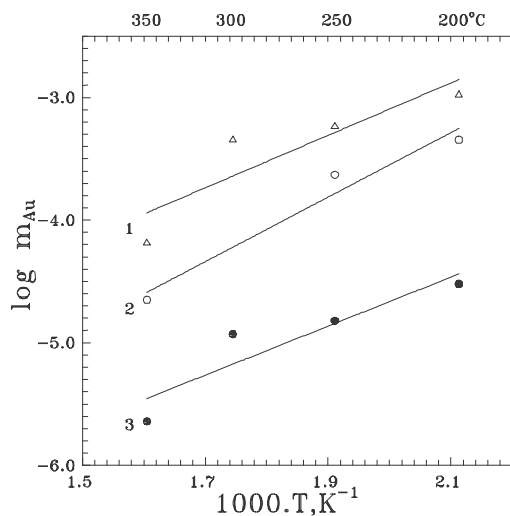


Fig.1. Dependence of gold solubility on temperature at P=300 bar. 1-0.5; 2-0.3; 3-0.05 m S sulfide

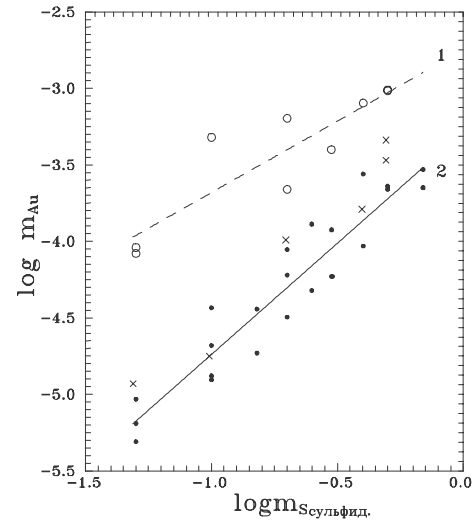


Fig.2. Dependence of gold solubility in sulfide-bearing systems at $\text{pH}_{\text{start}} = 7.4$, with (1) and without (2) addition of Na_2SO_4 at $T = 300^\circ\text{C}$ and $P = 300$ bar.

As we reported earlier [3-4] gold solubility depends on sulfide sulfur concentration in a solution, temperature, and pressure. Gold solubility increases markedly with increasing m_{sulfide} and decreases with temperature (fig.1). Gold solubility decreases with pressure, which agrees with literature data [5-6].

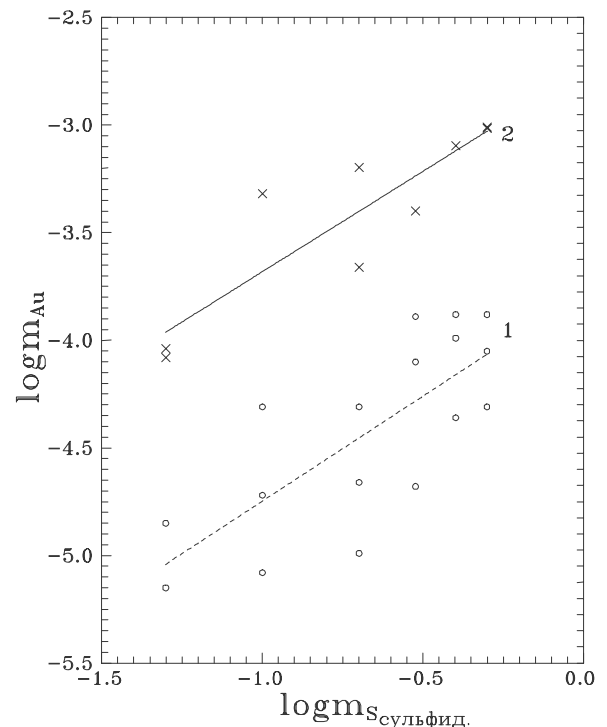


Fig.3. Dependence of gold solubility on m_{Sulfide} at different pH of a solution $T = 300^\circ\text{C}$ and $P = 300$ bar 1- $\text{pH}=1.44$; 2- $\text{pH}=7.52$

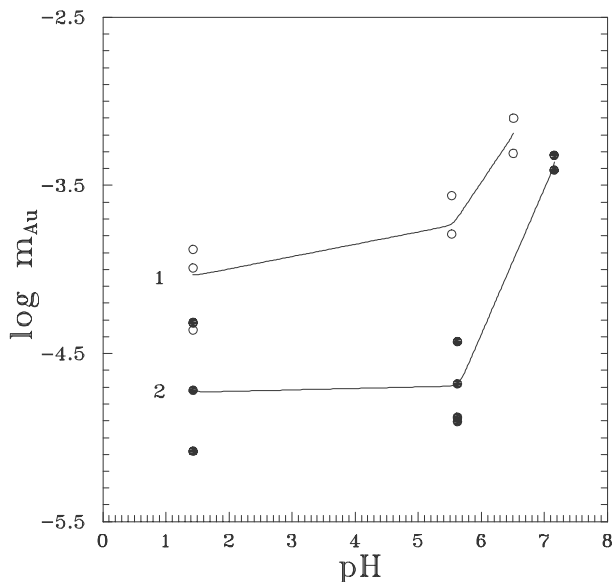


Fig.4. Dependence of gold solubility on pH of a solution. 1- 0.4; 2- 0.1 m S sulfide.

The effect of oxidation potential on gold transfer is rather complicated and should be considered alongside with changing pH and sulfide sulfur concentration in a solution. Changing sulfide sulfur concentration in its turn influences solution acidity. To study the effect of these relationships on gold solubility a series of experiments has been carried out at 300°C and 300 bar in sulfide-bearing systems with (and without) addition of 0.0469m Na₂SO₄ (fig.2). When Na₂SO₄ was added under experimental conditions pH increased from 5.5 to 7.5, Eh – from -0.620v to -0.840v. The calculations showed that after the addition of 0.0469m Na₂SO₄ into the system the concentration of sulfide sulfur in the system increased by 0.0468m. Fig.2 demonstrates an essential increase of gold solubility with the addition of Na₂SO₄ into the system.

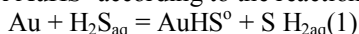
Presented in fig.3 is the dependence of gold solubility on sulfide sulfur concentration in the solutions with different pH at 300°C and 300 bar. Fig.3 clearly illustrates the effect of acidity on gold solubility in sulfide-bearing solutions. Curve 1 shows the results of the experiments with constant pH=1.44 and curve 2 – pH=7.5. Herewith, it should be pointed out that oxidation potential of the system changes essentially at pH=1.44, Eh= -0.144v, pH=7.52, Eh=-0.843v.

The equilibria of the dependence of gold solubility on sulfide sulfur concentration in a solution have been obtained at 300°C and 300 bar.

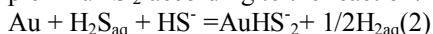
For curve (1) $\log m_{Au} = -3.772 + 0.975 \log S_{Sulfide} \pm 0.31$

For curve (2) $\log m_{Au} = -2.748 + 0.933 \log S_{Sulfide} \pm 0.10$

And, at last, fig.4 shows that gold solubility at experimental parameters up to pH<5 does not depend on pH of a solution, at pH>5 a sharp inflection was observed on the dependence curves of gold solubility on pH. In the former case this is most likely due to the fact that gold solubility proceeds with the formation of a neutral complex AuHS⁰ according to the reaction:



In the second case – with the formation of a charged complex AuHS₂⁻ according to the reaction:



From all above-stated the conclusion is obvious that the presence of gold in the hydrothermal solution, its transport and deposition are controlled by the interaction

of five major factors – pH, Eh values, total concentration of sulfide sulfur, temperature and pressure which is not in conflict with theoretical calculations and geological observations.

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