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Experimental study of the change in the composition of gold-silver alloys under hydrothermal conditions in the presence of pyrite

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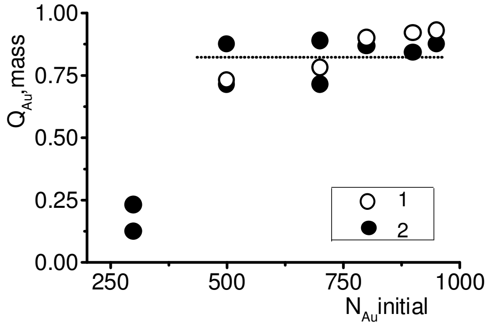
The study of conditions and forms of the mutual mobilization of gold and silver in hydrothermal processes of sulfide-formation is geochemically important. We have continued the experimental study [1] and obtained new experimental data (with autoclave method) on the interaction of fraction FeS_2 (Berezovsky deposit, size 1-2mm) and samples of synthesized Au-Ag-alloys ($N_{Au}= Q_{Au}$•1000=300-950, where Q_{Au} is a mass fraction of Au in the alloy) with solution of 0,5mNaCl+0,01mHCl (pH=2,03) at 300°C (P=500bar). Three fractions of experimental products which contain metals and sulfur (methods AA, SEM, iodometry, gravimetry) are distinguished: 1 – solution; 2 – reaction zone on the alloys surface; and 3 – pyrite with the new-precipitated phase. It is found that the enrichment of the solution in silver relative to gold (fraction 1) leads to stabilization of the composition of all alloys (fraction 2) at a level of Q_{Au}=0,75-0,90 (points 1 in the figure).



It is significant that the same Q_{Au} interval is typical still for the phase, new-precipitated on the pyrite surface (fraction 3, points 2 in the fig.) in the range N_{Au}(init)=500-950. For the sample of N_{Au}(init) =300, the revealed value of the mass fraction of gold in fraction 3 turns to be much lower (Q_{Au}=0,12-0,24) that is probably conditioned by the formation of the gold-bearing sulfide of silver.

The Q_{Au} value for Au-Ag-alloys in equilibrium with Ag_2S and FeS_2 predicted by a method of thermodynamic modeling ("HCh" code [2]) confirms to 0.84 value (broken line in the fig.).

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References

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Gold and arsenic interactions with pyrite in hydrothermal solutions

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The fate of gold and arsenic in hydrothermal systems containing pyrite is crucial for our understanding of processes related to hydrothermal ore deposition. Here we present X-Ray Absorption Spectroscopy (XAS) data from *in situ* hydrothermal mixing experiments where aqueous Au and As complexes have been reacted with hydrothermally produced pyrite at 25-250° C. The system set-up allows the anaerobic mixing of two fluids at high temperatures and liquid-vapour pressure and thus mimics a hydrothermal ore depositing environment. Such fluid mixing leads to precipitation of sulphides and the coeval removal of precious metals from solution. Pyrites precipitated from solution were equilibrated with aqueous gold (250-1000ppm) and arsenic (0.5-2wt%) solutions at temperatures between 25°C and 250°C and the reaction products characterised using XAS.

The high temperature (250°C) Au LIII edge XAS data for Au-only experiments show a first shell containing four S-Au distances of 2.42Å and a significant amount of information on Fe distances extending to 3rd and 4th shells. This data shows that gold is incorporated into the structure of pyrite and not adsorbed. At low temperatures (25°C) only the first two shells were present suggesting sorption as the dominant mechanism. Data from the edge region indicates that gold is present solely in its 1+ oxidation state and no Au^o was found. XAS data from high temperature experiments containing both Au and As show a best fit second shell with As-Au distances of 3.25Å.

Arsenic K-edge XAS measurements of the high temperature samples containing only arsenic have bond distances and scattering atoms very similar to that of arsenopyrite (3 x Fe @ 2.33Å and 1 x S @ 2.27Å). This suggests that arsenic is also incorporated into the structure at high temperatures. Conversely, the low temperature samples reveal larger bond lengths and less structure at greater distances from the central arsenic atom thus also indicating a sorption mechanism. The oxidation state of arsenic in all samples is also the same as arsenopyrite (As^{1-}).

The reaction of both gold and arsenic solutions with pyrite at high temperature leads to the incorporation of these metals into the structure of pyrite by a dissolution/reprecipitation or a diffusion mechanism. At low temperatures, sorption of gold and arsenic is the dominant mechanism. The structural incorporation of gold and arsenic into pyrite, demonstrated in this study, has important implications for ore deposits containing hydrothermally precipitated gold.