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R.W. BOYLE
W.M. ALEXANDER
G.E.M. ASLIN

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SOME OBSERVATIONS ON THE SOLUBILITY OF GOLD

ABSTRACT

Solubility experiments confirm that gold can be transported in natural humic waters probably in the form of a metal-organic compound, as a chelate, or adsorbed to humic matter. In gossans and in auriferous sulphide (pyritic) deposits similar experiments indicate that gold can be transported in ferric sulphate solutions. Under hydrothermal conditions gold is readily soluble in alkali carbonate solutions containing H_2S , AsH_3 , or SbH_3 or combinations of these constituents.

RESUME

Des expériences de solubilité confirment que l'or peut être transporté dans des eaux humiques naturelles sous la forme d'un composé organo-métallique, soit un chélaté ou soit adsorbé à la matière humique. Dans les chapeaux de fer et les gisements de sulfures aurifères (pyritique) des expériences similaires nous montrent que l'or peut être transporté dans des solutions de sulfate ferrique. Dans des conditions hydrothermales, l'or est facilement soluble dans des solutions de carbonates alcalins contenant H_2S , AsH_3 , ou SbH_3 , ou un mélange de ces constituents.

INTRODUCTION

For some twenty years the senior author has been investigating the geochemistry of gold in all of its natural settings. Some of the results of this work have been published (Boyle, 1961, 1969; and Boyle and Jonasson, 1973). A forthcoming bulletin will cover the entire research on the element.

One problem in the geochemistry of gold has been of particular interest--namely, the solubility of gold in various natural solutions. Considerable work on this problem has been done in the past, particularly by Liversidge (1893), Stokes (1906), Harrison (1908), Lenher (1909, 1912, 1918), Hatschek and Simon (1912), Freise (1931), Fetzer (1934, 1946), Johnston and Leyland (1938), Ogryzlo (1935), Frondel (1938), Garces (1942), Smith (1943), Kreiter *et al.* (1959), Kelly and Cloke (1961), Shabynin (1966), Goni *et al.* (1967), Listova *et al.* (1966), Machairas (1967), Ong and Swanson (1969), Weissberg (1970), Curtin *et al.* (1970), Lakin *et al.* (1971), Vilor and Sarapulova (1971), Vilor and Skharupa (1971), Henley (1973), Seward (1973), Fisher *et al.* (1974) and Miller and Fisher (1974). The results of these various researches have shown that the principal soluble species of gold are gold hydroxide, $Au(OH)_3$ or $[AuO_2]^-$; halogen complexes of the type $[AuCl_4]^-$; various thio complexes of the type $[Au(S_2O_3)_2]^{3-}$; cyanide and thiocyanate complexes of the type $[Au(CN)_2]^-$ and $[Au(CNS)_4]^-$; and sulphide and polysulphide complexes such as $[AuS]^-$ and $[Au_2(HS)_2S]^{2-}$. Gold can also be transported by humic substances in which the gold appears to be adsorbed to the humic complexes forming some uncharacterized Au-humate. Colloidal gold is also well documented, the "Purple of Cassius" being known for centuries.

Our experiments on the solubility of gold have covered a wide range of natural solutions, some of which such as those containing chlorides, thiosulphates,

cyanides, thiocyanates, and alkali sulphides and bisulphides need not be considered here since they are well documented in the papers referred to above. We have been most interested in the solubility of gold in synthetic solutions representative of those that probably deposited auriferous quartz veins and those that have been responsible for the transport of gold in supergene processes. With respect to the first we have experimented with alkali carbonate and bicarbonate solutions saturated with hydrogen sulphide, arsine, stibine, and combinations of these substances. Concerning the supergene solutions, we have experimented with those containing humus or ferric sulphate, the former common in many surficial environments and the latter particularly characteristic of gossans and oxidizing gold deposits containing pyrite, pyrrhotite, and arsenopyrite. All of our experiments are essentially qualitative, the aim being to obtain a general estimation of the solubility of gold in the above mentioned solutions. The concentrations we have used in our experiments are perhaps greater than those encountered in nature; however, we consider that the greater concentrations compensate in part for the time factor in geology.

OCCURRENCE OF GOLD IN NATURE

Gold occurs in a number of ways in its various deposits, the chief modes being as the native metal and combined with tellurium in the various tellurides and with antimony in aurostibite, $AuSb_2$. In addition, gold is a submicroscopic and/or lattice constituent of a number of sulphides, arsenides, and sulphosalts. In minerals like tetrahedrite gold is a lattice constituent probably substituting in the silver sites; in minerals like pyrite and arsenopyrite some gold is present as microscopic blebs, but in some deposits there is also much gold in a submicroscopic form or present in lattice sites in these minerals.

The various modes of occurrence of gold in its deposits are important in the solubility and migration of gold, particularly in supergene processes. Where the gold occurs in the native form in particles with diameters greater than 50μ the solubility of the metal is much less than when the element is released in a very finely divided form during the oxidation of minerals such as the tellurides, aurostibite, and auriferous pyrite and arsenopyrite which contain the metal as a lattice constituent or as submicroscopic particles. With the latter in mind, and since it seems probable that the source of gold for transport in both hydrothermal and supergene solutions would be largely in an extremely finely divided state, we have used gold as prepared below.

PREPARATION OF GOLD FOR SOLUTION EXPERIMENTS

The gold used in the solution experiments was precipitated in a fine (amorphous) state from sodium gold thiosulphate by reduction with stannous sulphate. The sulphates were thoroughly washed free from the gold (BaCl_2 test), and the gold was kept under metal-free water to prevent it from drying out and producing anomalous surface effects¹. An X-ray of the gold gave an amorphous pattern; X-ray fluorescent analyses showed only traces of tin in the gold sample.

RESULTS OF THE EXPERIMENTS (SUPERGENE PROCESSES)

The solutions tested for dissolution of gold consisted of specially prepared metal-free deionized water, humic solutions, and solutions containing sodium chloride, ferric sulphate, ferric sulphate and H_2SO_4 , and ferric sulphate plus H_2SO_4 and NaCl . An excess of amorphous gold was added to each of these solutions, and the solutions were allowed to stand for varying periods being agitated from time to time to ensure thorough reaction. The solutions were then progressively filtered and analyzed directly for gold by atomic absorption, using a Perkin-Elmer 303 spectrophotometer.

¹In various experiments carried out over a number of years the senior author has noted that there are marked differences in the solution properties of gold, some of which are of interest here. Nascent gold in a finely divided amorphous state as prepared above is relatively soluble in a number of solvents. If exposed to the air this gold becomes difficultly soluble; the same is true if the gold is left for any length of time in metal-free water. It appears that gold exposed to air and water becomes armoured against solution in some manner or other by a coating the nature of which is unknown. Furthermore, the state of the gold is important; the finely divided gold prepared as described above is much more soluble than gold filings or gold leaf. While not of particular interest in this study it is also notable that gold containing impurities such as silver is more soluble than gold of very high purity.

The 242.8 nm line was used with background correction and dilutions made on any samples indicating a concentration greater than 15 g/ml. The results are shown in Table 1 and discussed briefly below:

Metal-free water

Two samples of deionized metal-free water, to which an excess of amorphous gold was added, were allowed to stand with repeated agitation for nine days, filtered through millipore filters, and tested for soluble gold. Both samples showed undetectable amounts of gold.

Humic-rich solutions

Humic-rich solutions, such as those that occur in streams draining swamps, peat bogs, muskegs, etc., were prepared by reacting metal-free water for some 18 months with peat finely comminuted in a blender. The solutions obtained, containing approximately 0.5 per cent humic matter, were filtered through a No. 1 Whatman paper and tested for gold. None was detected. The humic solutions were then reacted with an excess of amorphous gold for 8 days with frequent agitation. Samples filtered through No. 1 Whatman paper contained 0.15μ g/ml Au, whereas those filtered through the millipore contained no detectable gold. Examination of the two solutions after filtration revealed that the one filtered through the Whatman paper retained much of the brown (colloidal?) material whereas that filtered through the millipore was quite colourless, all or most of the humic matter being removed. These results are entirely in agreement with those obtained by the senior author with humic substances in natural waters over several years, namely that the gold, silver, copper, and other metals are intimately associated with the humic matter being either chelated, organometallically bonded, or adsorbed. When the humic matter is removed the solutions are essentially relatively free of metals.

Sodium chloride solutions

Successive gold-free solutions - saturated, 50 per cent saturated, and 30 per cent saturated with sodium chloride - were prepared and reacted by constant agitation with excess amorphous gold for an hour, filtered through millipore filters, and tested for soluble gold. All three samples showed undetectable amounts of gold.

Ferric sulphate solutions

Successive gold-free solutions - saturated, 50 per cent saturated, and 30 per cent saturated with ferric sulphate - were prepared and reacted by constant agitation with excess amorphous gold for an hour. Similar solutions, but with the addition of 2 ml of concentrated H_2SO_4 and with the addition of 2 ml of concentrated H_2SO_4 and 4 ml of saturated NaCl solution, were reacted with excess amorphous gold. In all cases as shown in Table 1 considerable amounts of gold were taken into solution, the largest amounts being present in the solutions containing H_2SO_4 . The presence of

TABLE 1. SOLUTION OF GOLD BY VARIOUS AGENTS

SUPERGENE PROCESSES

Type of solution	Concentration of salt, etc.	Filtration procedure	Gold present in solution ($\mu\text{g/ml}$)	Remarks
Metal-free water		50m μ millipore	0.00	Two 100 ml samples taken
Humic	0.5% humic matter	No. 1 Whatman 50m μ millipore	0.15 0.00	100 ml sample 100 ml sample
Sodium chloride	30% saturated	50m μ millipore	0.00	100 ml sample
	50% saturated	50m μ millipore	0.00	100 ml sample
	Saturated	50m μ millipore	0.00	100 ml sample
Ferric sulphate	30% saturated	50m μ millipore	0.07	100 ml sample
	50% saturated	50m μ millipore	0.14	100 ml sample
	Saturated	50m μ millipore	0.21	100 ml sample
Ferric sulphate +2ml H ₂ SO ₄	30% saturated	50m μ millipore	0.36	100 ml sample
	50% saturated	50m μ millipore	0.14	100 ml sample
	Saturated	50m μ millipore	0.07	100 ml sample
Ferric sulphate +2ml H ₂ SO ₄ + 2ml sat. sol. NaCl	30% saturated	50m μ millipore	0.18	100 ml sample
	50% saturated	50m μ millipore	0.07	100 ml sample
	Saturated	50m μ millipore	0.04	100 ml sample

HYPOGENE (HYDROTHERMAL) PROCESSES

Type of solution	Concentration of salt, etc.	Filtration procedure	Gold present in solution ($\mu\text{g/ml}$)	Remarks
Metal-free water		50m μ millipore	0.00	Two 100 ml samples taken
Sodium chloride	30% saturated	50m μ millipore	0.00	100 ml sample
	50% saturated	50m μ millipore	0.00	100 ml sample
	Saturated	50m μ millipore	0.00	100 ml sample
Potassium carbonate	30% saturated	50m μ millipore	0.00	100 ml sample
	50% saturated	50m μ millipore	0.00	100 ml sample
	Saturated	50m μ millipore	0.00	100 ml sample
Sodium carbonate	30% saturated	50m μ millipore	0.00	100 ml sample
	50% saturated	50m μ millipore	0.00	100 ml sample
	Saturated	50m μ millipore	0.00	100 ml sample
Sodium bicarbonate	30% saturated	50m μ millipore	0.54	100 ml sample
	50% saturated	50m μ millipore	1.00	100 ml sample
	Saturated	50m μ millipore	1.54	100 ml sample
Potassium carbonate +H ₂ S	30% saturated	50m μ millipore	30.00	100 ml sample
	50% saturated	50m μ millipore	45.50	100 ml sample
	Saturated	50m μ millipore	32.50	100 ml sample
Sodium carbonate +H ₂ S	30% saturated	50m μ millipore	21.00	100 ml sample
	50% saturated	50m μ millipore	41.00	100 ml sample
	Saturated	50m μ millipore	45.50	100 ml sample
Sodium bicarbonate +H ₂ S	30% saturated	50m μ millipore	5.50	100 ml sample
	50% saturated	50m μ millipore	7.50	100 ml sample
	Saturated	50m μ millipore	10.00	100 ml sample

HYPOGENE (HYDROTHERMAL) PROCESSES (cont'd.)

Type of solution	Concentration of salt, etc.	Filtration procedure	Gold present in solution ($\mu\text{g/ml}$)	Remarks
Sodium carbonate +AsH ₃	30% saturated	50m μ millipore	17.50	100 ml sample
	50% saturated	50m μ millipore	20.00	100 ml sample
	Saturated	50m μ millipore	23.70	100 ml sample
Sodium bicarbonate +AsH ₃	30% saturated	50m μ millipore	3.10	100 ml sample
	50% saturated	50m μ millipore	6.10	100 ml sample
	Saturated	50m μ millipore	12.50	100 ml sample
Sodium carbonate +H ₂ S+AsH ₃	30% saturated	50m μ millipore	29.00	100 ml sample
	50% saturated	50m μ millipore	50.00	100 ml sample
	Saturated	50m μ millipore	63.00	100 ml sample
Sodium bicarbonate +H ₂ S+AsH ₃	30% saturated	50m μ millipore	1.70	100 ml sample
	50% saturated	50m μ millipore	2.10	100 ml sample
	Saturated	50m μ millipore	2.90	100 ml sample
Sodium carbonate +SbH ₃	30% saturated	50m μ millipore	1.80	100 ml sample
	50% saturated	50m μ millipore	7.30	100 ml sample
	Saturated	50m μ millipore	13.30	100 ml sample
Sodium bicarbonate +SbH ₃	30% saturated	50m μ millipore	0.00	100 ml sample
	50% saturated	50m μ millipore	0.60	100 ml sample
	Saturated	50m μ millipore	1.20	100 ml sample
Sodium carbonate +H ₂ S+SbH ₃	30% saturated	50m μ millipore	29.00	100 ml sample
	50% saturated	50m μ millipore	40.00	100 ml sample
	Saturated	50m μ millipore	66.00	100 ml sample
Sodium bicarbonate +H ₂ S+SbH ₃	30% saturated	50m μ millipore	26.00	100 ml sample
	50% saturated	50m μ millipore	38.80	100 ml sample
	Saturated	50m μ millipore	46.20	100 ml sample

NaCl appears to depress the solubility of the metal. These data indicate that amorphous gold is soluble in ferric sulphate solutions alone, and more so when free H₂SO₄ is present.

RESULTS OF THE EXPERIMENTS
(HYPOGENE PROCESSES)

The synthetic hypogene solutions consisted of various carbonate solutions saturated with H₂S, AsH₃, SbH₃, and combinations of these constituents. These were reacted with amorphous gold at room temperature, and the soluble gold was determined as described above.

Carbonate solutions

Sodium and potassium carbonate solutions were reacted for an hour by constant shaking with excess amorphous gold. Both solutions were found to be essentially free of soluble gold after filtration through millipore filters. Sodium bicarbonate solutions (gold free) on the other hand were found to contain significant amounts of gold after the same treatment as shown in Table 1.

Numerous solutions of sodium and potassium carbonate and bicarbonate were prepared and consider-

able amounts of amorphous gold were added. These solutions were successively treated with hydrogen sulphide, arsine, stibine, and the various gases combined as shown in Table 1. The gases were bubbled through the carbonate solutions with constant stirring for periods ranging from 15 minutes to an hour or more. In most cases all of the amorphous gold added was dissolved and hence the solubility limits while high are unknown.

The results obtained from the qualitative experiments with alkali carbonate solutions indicate extensive solution of gold when the solutions contain H₂S, AsH₃, or SbH₃, or various combinations of these gases. The state of the dissolved gold in such solutions when H₂S or sulphide ion is present would seem to be as a soluble sulphide of the type [AuS]⁻ or as thioarsenites, thioarsenates, thioantimonites, or thioantimonates of the type [Au(AsS₃)]²⁻ and [Au(SbS₃)]²⁻. Production of these complexes in the alkali carbonate solutions evidently takes place first with the formation of alkali sulphide or bisulphide, thioarsenate, thioantimonate, etc. followed by the dissolution of the gold as the various soluble complexes. When only AsH₃ or SbH₃ are present the soluble complexes are evidently gold arsenides and stibnides such as [AuAs]²⁻ and [Ausb]²⁻ or more complicated polymerized and hydrolyzed species.

Our interest in determining the approximate solubility of gold in alkali carbonate solutions containing H₂S, AsH₃, SbH₃, or their analogous ions is that these solutions are evidently largely responsible for the transport and deposition of gold in veins and other deposits under endogenic conditions. This is based on the general observation that carbonatization accompanies many types of endogenic gold deposits, and that the element is nearly universally associated with S (sulphides mainly pyrite), As (arsenopyrite), and Sb (stibnite and sulphosalts) in such deposits. There is also much sericitization (introduction of K) or albization (introduction of Na) in shear zones, faults, etc. containing gold deposits.

CONCLUSIONS

We conclude that under supergene conditions gold may be transported by humic waters, the metal being intimately associated with the humic matter probably forming an adsorbed phase or some uncharacterized type of organometallic compound or chelate. In deposits where sulphides, especially pyrite, pyrrhotite, and arsenopyrite are oxidizing, gold can be transported in ferric sulphate solutions. Under hydrothermal conditions gold is readily transported in solutions containing H₂S, AsH₃, or SbH₃ or combinations of these constituents. Having regard to the chemical similarity of Se and Te to S we can assume that gold may also be transported where H₂Te and H₂Se or their analogous tellurides and selenides are present in solution.

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