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Aspects of the Chemical Composition of the Birimian Gold Fluid

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Abstract

Fluid inclusion data, EPMA and geochemical studies of metamorphic chlorites from altered phyllites and graphitic phyllites were used to constrain the possible nature of the ore fluid involved in gold mineralization in the Birimian of Ghana. The results showed that the metavolcanics and metasediments rock groups in the Birimian gold belt were transformed by metamorphism and hydrothermal activity. The alteration processes involved in the transformation were sulphidation, sericitization, rutile formation, silicification, carbonation, carbonitization and of chloritization. Some of these processes were related to major gold mineralization in the country rocks. Minor alterations caused by the hydrothermal activity, which resulted in the mineralization are sulphidation, carbonation, tourmalization and chloritisation. It is inferred that gold was deposited under similar conditions as those of the hydrothermal activity. Calculated physico-chemical depositional conditions of the ore fluid, based on chlorite-pyrrhotite-pyrite and chlorite-albite relationships yielded fO_2 = -33 to -32; fH_2S = 1.5 to - 2.0; and pH= 5.8. Precipitation of gold in the country rocks occurred as gold-bearing sulphides, dominated by arsenopyrite, through interaction of the host country rocks with the mineralising fluid. Deposition of the gold occurred within major late-orogenic shear zones when the original mineralizing fluid decompressed, cooled and separated into CO_2-rich and H_2O-rich fluids.

Keywords: Alteration, hydrothermal, metamorphism, metasomatism, sulphide- gold mineralization

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1 Introduction

The Palaeoproterozoic rocks of the West African craton are defined by the Supergroup of the Birimian province. The Supergroup, formed 2.2-2.0 Ga ago, consists predominantly of granitic batholiths, metasedimentary sequences and metavolcanic greenstone belts. Since 1990, researchers have published several papers proposing various regional scale accretionary models for the evolution of the Birimian Supergroup and the Eburnean orogeny [1]. The Birimian lithostratigraphic succession starts with a major sequence of tholeiitic pillow basalts with intercalations of sediments, overlain by detrital and carbonate sediments, associated with calc-alkaline volcanics and plutons. A global scheme for the geodynamical evolution can be reasonably constrained using the characteristics of early tholeiitic plutono-volcanic and granitoid rocks [1].

The Paleoproterozoic Birimian sediments and volcanics in Ghana host most of the country's economically viable mineral deposits, including gold, diamond and manganese. The gold deposits and occurrences are set in the five parallel northeasterly volcanic belts, namely the Kibi-Winneba, Ashanti, Sefwi, Bui, and Bole-Nangodi belts as well as the north-trending Lawra belt. The volcanic belts consist of low-grade metamorphic lavas of predominantly tholeiitic composition [2] separated by basins consisting of isoclinally folded metasedimentary rocks composed of dacitic volcaniclastic rocks, wackes and volcanogenic argillites, which are derived from the belts [3, 4, 5].

The most important type of gold occurrence in Ghana is the mesothermal arsenopyrite-gold lode mineralization, commonly referred to as the Ashanti-type, typical of the Obuasi gold-field area considered the largest gold deposit in the region [6]. These deposits are largely confined to 'tectonic corridors' and they usually display complex, multi-phase structural features, which control the mineralization [6, 7]. Other deposits that also occur in Ghana are mesothermal gold-quartz vein deposits, disseminated gold-sulphide deposits and Tarkwaian paleoplacers [6]. However, the processes and mechanisms responsible for gold mineralization have not been studied in detail and only few studies have given attention to them. According to Eisenlohr et al. [8], ore fluid migration and gold deposition processes in most gold deposits worldwide are attributed to contrasting fluid-pressure/temperature regime between first and second order structures.

Limited wall-rock alteration in metavolcanic- and metasediment-hosted deposits and relatively extensive wall-rock alteration in granitoid-hosted deposits suggest that most of the lode gold mineralization such as those in the Blue Ridge of the southernmost Appalachians formed from fluids that were close to equilibrium with metavolcanic and metasedimentary rocks [9]. Stowell et al. [9] opined that silicate mineral thermobarometry of vein, selvage, and wall-rock mineral assemblages also do indicate that mineralization and regional metamorphism occurred at greenschist to amphibolite facies level. According to the authors, the association of gold mineralization with structurally controlled concordant and discordant quartz sulphide veins, and the temperatures and pressures of wall-rock alteration and regional metamorphism indicate that the distribution of gold is the result of metamorphism during deformation. In this paper, we use the results of microprobe analyses of metamorphic chlorites, geochemical data and fluid inclusion studies to discuss the possible nature of the ore fluid involved in gold mineralization in the Birimian terrane of Ghana.

2 Geological Setting

The rock units of the Birimian metasediments (i.e. Lower Birimian) are primarily phyllites, schists and greywackes (Fig. 1). There are also weakly metamorphosed tuffs, feldspathic sandstones, Mn- and Si-rich chemical sediments. The Birimian metavolcanics (Upper Birimian) (Fig. 1) contain mostly metamorphosed basaltic and andesitic lavas, hornblende-actinolite schists, carbonate-chlorite schists and amphibolites as well as phyllite, greywacke, conglomerate, quartz-sericite-schist, and mica schist.

Minor amounts of metamorphosed rhyolitic and dacitic lavas and tuffs, Mn-rich and Si-rich chemical sediments occur. Diabases and porphyries intrude the metavolcanics. The Birimian assemblage is 10-15 km thick [10], strikes generally NE, dips steeply to the southeast and is isoclinally folded. Intruded into the Birimian are two main granitoid types: one type being the Dixcove granitoid body occurs in smaller in masses and relatively Na-rich. According to Eisenlohr [11], this type of granitoid may be contemporaneous with volcanism of the Upper Birimian and is emplaced in the volcanic belts. The second type of granitoid intrusion is the Cape Coast type, which is voluminous in mass, rich in aluminium (peraluminous) and intrudes the Birimian volcanic/sedimentary assemblage.

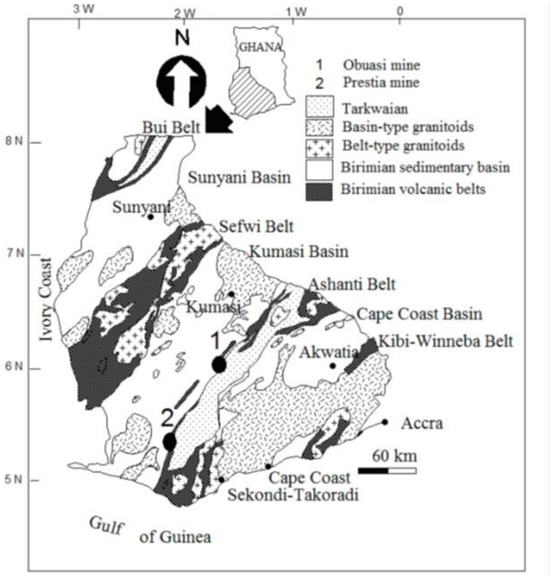

Figure 1: Geological map of Southern Ghana showing the area of study in the Birimian Supergroup, and the basin type granitoid. Samples were taken from (1) Obuasi and (2) Prestea (after Taylor et al. [5]).

3 Materials and Methods

Rock samples were collected from Prestea and Obuasi mines, prepared into thin sections, and studied for detailed mineralogical, microstructural and alteration features. Five of these samples were selected for microprobe analysis, using Cameca microbeam (Type Camebax MB) at University of Hannover operated at a 15KV accelerating potential with 18 na beam current.

One hundred and twenty (120) samples were analyzed at the Institute of Geosciences, Technical University of Braunschweig, Germany for major and trace elements (except for LOI, Au and Si) using a BAUSCH & LOMB induced couple plasma emissions spectrometer (Type 3520). LOI was determined thermogravimetrically while Au was by spectrophotometric with Rhodamin B. Si was determined on a PHILLIPS X-ray fluorescence spectrometer (Type PW 1410/10) using fused glass discs (2700 mg lithium tetraborate ($Li_2B_4O_7$), 300 mg lanthan oxide (La_2O_3) and 300 mg fused sample).

4 Results and Discussion

4.1 Alteration Processes

Post-formation changes of a rock, such as alteration, may be due to; (i) hydrothermal activity, (ii) metamorphism, or (iii) post-metamorphic deformation. Studies carried out in gold belts indicate that some of these events can be related to gold mineralization [12, 13, 14]. Though these processes can sometimes be very complex and superimposed, such as in the Norseman-Wiluna belt of Western Australia [15, 16, 17], petrographic studies on the Birimian suggest that it is possible to separate these events and determine which one was involved in gold mineralization [18, 19, 20].

In the formation of hydrothermal ore deposits through wall-rock alteration, the processes convert the initial mineral assemblages in the host rock to new minerals under different conditions of temperature, pressure and fluid composition. Thus, by establishing the mineral assemblages indicative of certain alterations (e.g. sulphidation), it is possible to decipher the conditions of temperature, pressure and fluid composition involved. Sometimes alteration in a rock may be due to the replacement of one mineral by another of different composition facilitated by interaction of the mineralising hydrothermal fluid with the rock [21]. This form of alteration is commonly known as metasomatism and an example is the addition of H_2O and CO_2 to rocks [22]. In metasomatic alterations, it is necessary to determine whether the constituents being subtracted have really left the system or whether they contribute volumetrically to the system as constituents of other minerals [23].

The processes responsible for the formation of the Birimian lode gold deposits were wall-rock alteration and metasomatism. In the Birimian terrane, wall-rock alteration often took the form of pyritization, arsenopyritization, sericitization, chloritization, silicification and carbonitization (i.e. formation of pyrite, arsenopyrite, sericite, chlorite, quartz veinlets and ankerite respectively) [24, 19]. Though the processes or mechanisms responsible for these alterations, namely hydrothermal activity, metamorphism or post-metamorphic deformation and their relations to the gold mineralization were not discussed by previous authors, several researchers have indicated the relationship between alteration processes and gold mineralization. For example, the West Bleida gold mineralization in the central Anti-Atlas in southern Morocco is linked to strong sericitization and silicification [25]. According to the authors, these altered rocks represent a proximal hydrothermal alteration halo around the West Bleida ore zones and that normative chlorite characterizes the metamorphic assemblage away from the ore zones.

Hirdes and Leube [18] used the spatial relationship of minerals in response to shearing and quartz vein emplacement to determine whether alteration was pre-, syn- or post-metamorphic. Using semi-quantitative x-ray diffraction data, they found certain hydrothermal minerals of secondary origin from two profiles at the Ashanti Mine. The minerals, which showed different distribution patterns, were chlorite, siderite, paragonite, ferroan dolomite/ankerite, muscovite/illite, kaolinite and gibbsite. Muscovite/illite and siderite were omnipresent and showed no spatial relationship with the zone of shearing and/or the quartz vein. Thus, the formation of muscovite/illite and siderite could not be said to be pre-, syn- or post-metamorphic. They further discovered that chlorite and muscovite/illite were less abundant in the immediate vicinity of shear zones or quartz veins, but more abundant outside these zones.

Based on similar line of argument, Hirdes and Leube [18] suggested that part of the sulphide formation was pre-metamorphic, and paragonite, ferroan dolomite, kaolinite and gibbsite were post-metamorphic (i.e. related to the hydrothermal activity which was connected to shearing and emplacement of gold-bearing quartz vein). The assemblages of local spatial alteration suggest that some lithological groups are more susceptible to certain types of alteration than others. Hirdes and Leube [18] therefore concluded that alteration conditions (i.e., pre- and post-metamorphic) are comparable to the low grade metamorphism in the country rocks.

4.1.1 Mineralogy of alteration

The Birimian metasediments comprise of phyllites, metagreywackes and siliceous rocks, the minerals in the unaltered metasediments were quartz, illitic and chloritic clay minerals and organic matter. Knowledge of alteration processes is deduced from (i) field, (ii) petrographic and (iii) geochemical data. Alterations in the Birimian rocks resulted in the formation of pyrite, arsenopyrite, sericite, chlorite, quartz veinlets (Fig. 2) and ankerite, both in the wall rocks and in the mineralized quartz reefs, suggesting a close temporal relationship between the alteration conditions and processes, the introduction and formation of gold. Some previous researchers [26] concluded that the formation of Mg-rich chlorite, graphite, sericite, quartz, tourmaline, sphene, with or without siderite and ankerite occurred at the peak conditions of metamorphism and consistently formed in the metamorphic plane of the metasediments.

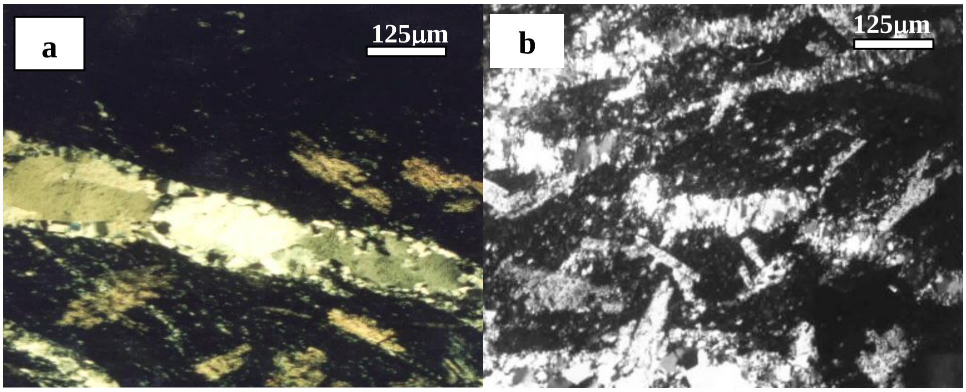

Figure 2: Photomicrographs of quartz micro veins. The areas in (a) where the width narrows up show the points of discontinuity and structural weakness that led to mobilization of gold, whereas (b) shows random mobilization of quartz as evidence that mobilization of gold takes place in areas where conditions of least resistance prevail.

But hydrothermal phase mineralization produced Fe- and Mg-rich chlorite, sulphide, sericite, siderite and ankerite, epidote and quartz after the prevalent metamorphism in the rocks.

The bulk composition of the Birimian metavolcanics is dacite to rhyodacite [27] and the original mineral assemblage is expected to include quartz, feldspars and biotite. Minerals produced consistently in the metamorphic plane of the metavolcanics are Mg- and Fe-rich chlorite, sericite, titanite, with or without siderite and ankerite; whereas the post-metamorphic minerals are sulphides, tourmaline, carbonates and chlorites (Fig. 3). A distinction can be made between the rock-mineral alterations due to metamorphism and those due to hydrothermal activity can be made by comparing minerals that are consistent with the metamorphic and post-metamorphic planes of both the metasediments and metavolcanics.

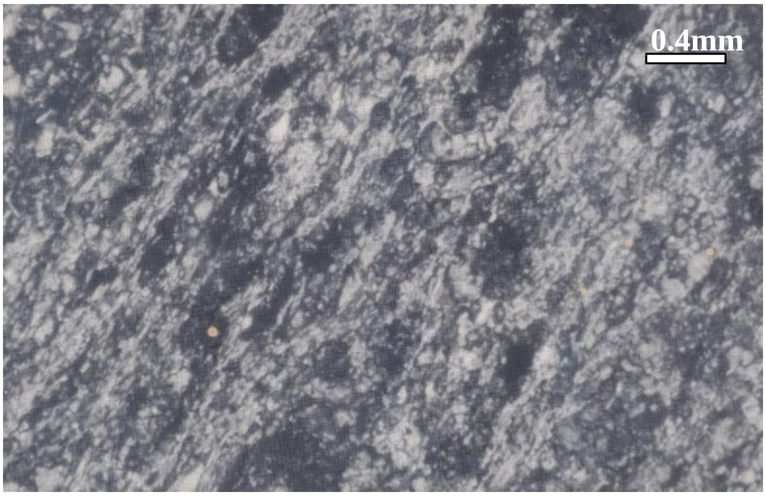

Figure 3: Photomicrograph of an intensely carbonated metavolcanic rock taken under cross-polars. The visible lineation is characteristic of the extent of alteration.

The comparison shows that Mg-rich chlorite, carbonates, titanite and sericite are regularly formed in metamorphic planes in the metasediments and metavolcanics [18], while hydrothermal minerals are sulphides, carbonates and Mg- and Fe- rich chlorites [18, 19]. The hydrothermal activity is inferred to be related to the gold deposition event, because the sulphides which contain the gold are in the post-metamorphic deformation planes.

4.2 Compositional Variation during Alteration

4.2.1 Geochemistry of alteration

One main objective of study of the geochemical data on ore deposits is to assess the alteration pattern, and extent using known two methods; either by statistical assessment of the chemical data (sometimes only by comparison of individual component values along a profile) or through the assessment of the elemental gains or losses using mass-balance calculations [23]. Hirdes and Leube [18] used the former approach to determine the extent and mode of the geochemistry of the alteration of rocks and minerals in the Ashanti mine. It was therefore concluded that depletion of Si (for a distance of ≈ 7.6 m) was accompanied by an increase in sericite (rise in Al, K) and carbonate (rise in CO_2, Ca, Mg), which correlated with gold. Rb and As also showed good correlation to Au. According to Hirdes and Leube [18], gold in the wall-rocks is positively correlated with Rb, K,As, Al, Bi, Pb, Cr, Ba and Zr (in decreasing order), whilst gold in the quartz vein is positively correlated with LOI (mainly as a function of carbonate), Na, Y, Mn, Ca, Cu and P (in decreasing order).

4.2.2 Mass-balance calculations

Several researchers [28, 29] have pointed out the need to consider the volume changes that accompany the various stages of alteration when interpreting geochemical data on lode gold deposits. This is because alteration in lode gold deposits is accompanied by deformation and or changes of volatile content [28], which tends to cause the absolute abundances of the mobile elements to change; immobile elements concentration changes occur either by dilution during addition of other components, or concentration will vary through the leaching of soluble components [28].

An inspection of tables of chemical analyses may not satisfactorily deduce chemical changes because; (i) the element oxides are constrained to a constant sum (100%), and (ii) the quantitative basis of the analyses is given under the assumption of a constant volume. The latter is particularly not true in view of the alterations which took place in the country rocks indicated earlier. Gresens [23] therefore suggested that knowing the volume relations enables one to calculate the actual gains and losses that occurred during such metasomatism. These mass-balance calculations involve finding the volume factor (f_v) of individual elements that behaved isochemically during the change from parent rock to altered rock. Establishing the original composition of the parent rock has often been problematic, in view of the dual metamorphism and hydrothermal activities. All the rocks have been altered to various degrees during the gold mineralization event. However, certain indicators can be used to indicate rocks which have been least altered based on the mineralogical composition and the similarity to rock types reported elsewhere in the Birimian in Ghana.

Taking these observations into consideration, the pure phyllites at Prestea and Obuasi are classified as the least altered rock type in the metasediments at Prestea and Obuasi respectively; whereas the ankerite-bearing metarhyolites found only in Obuasi are classified as the least altered metarhyolites (excluding the siliceous rock and greywacke because they constitute rare lithology groups) in both Prestea and Obuasi. Fyon and Crocket [30] cautioned that LOI may not be an accurate measurement of total volatile content, particularly in sulphide-rich samples because the measurement of LOI is affected by changes in the oxidation state of iron. Accordingly, the LOI values are neither used as an indicator of CO_2 nor volatile content and are therefore excluded from the mass-balance calculations. The immobile elements Al and Sc are used to calculate the volume changes. Calculations for the volume changes and the gains and losses of the elements are presented diagrammatically in Fig. 4 (a-f). Figures 4a, b and c show the gains and losses in the ‘altered’- phyllite (ankerite-chlorite-, ankerite-, chlorite-phyllite). The diagrams reveal the following observations. Very little changes occur in Fe, Mn, Mg, Ca, Na, and K in the ‘altered’ phyllite. Though the altered phyllite generally shows little or no significant changes in the trace elements, significant changes can, however, occur depending on the extent of alteration. Figure 4d shows element changes in the graphite phyllite (i.e. those generally in the shear zones).

Major and minor elements depict trend similar to those of the ‘altered’ phyllites but elevated abundances occur in the trace elements (except for Cr). The graphite phyllite is sometimes also found together with the auriferous quartz veins in shear zones. Since the shear zones were initiated after metamorphism, it is inferred that changes in rocks should come from the later hydrothermal activity.

Hence, during the later hydrothermal activity (Fig 4d: graphite phyllite in shear zones) the trace elements were mainly added by the 'altering hydrothermal fluid' to the metasediments (and probably to parts of the country rocks). The mass-balance calculation diagrams for the metavolcanics are shown in Figs. 4e and f. In these diagrams, the major and minor elements show the same trend as in the metasediments (both the altered phyllites and the graphite phyllite in the shear zones).

There is enrichment in the trace elements (except for Ba, Sr and Pb which were lost to the alteration fluid from the country rocks). The similarity between the trace elements distribution pattern in the altered metavolcanics and the graphite phyllite (which is in the shear zone) suggests a close link between the gold mineralization in the metavolcanics and in the quartz veins.

4.3 Thermodynamics of Alterations in Host Rocks and Quartz Veins

4.3.1 Chlorite petrography

The chlorites occur as fine- to medium-grained grains (Fig. 5). They usually occur with rutile, sericite, quartz, carbonate, arsenopyrite and pyrite. Textural evidence of the Birimian chlorites shows both Mg-rich and Fe-rich types. Petrographically, no chlorite-muscovite intergrowth was seen in the studied samples. The two different types of chlorite observed apparently represent two distinct chlorite generations resulting from; (i) metamorphic and (ii) hydrothermal alterations.

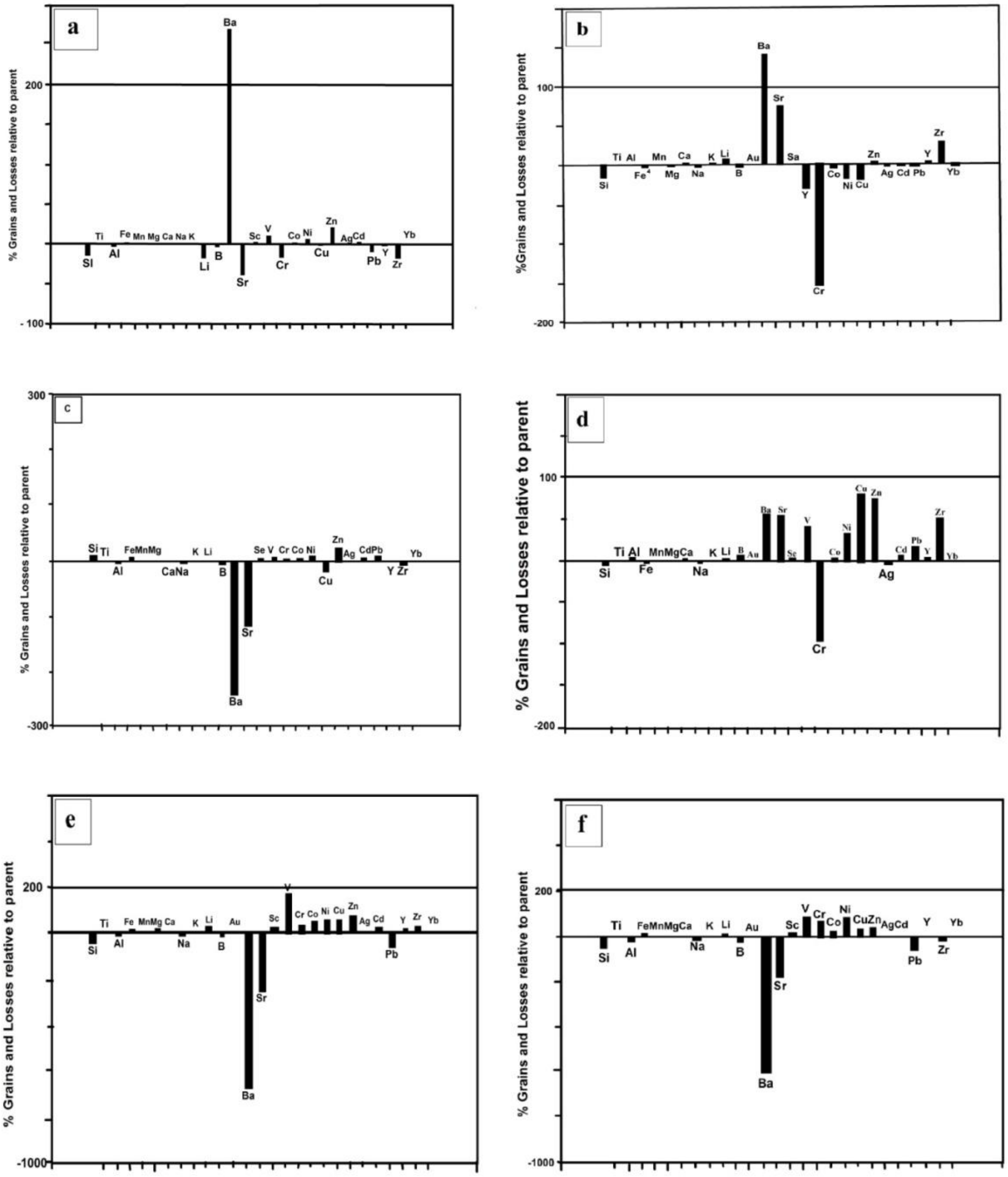

Figure 4: Enrichment and depletion diagrams of elements in the metasediments. (a): ankerite-chlorite phyllite, (b): ankerite-phyllite, (c): chlorite phyllite, (d): graphite phyllite and the metavolcanics; (e): ankerite chlorite metarhyollite, (f): ankerite metarhyolite. Gains and losses given as percentage compared to those for parent rock.

The hydrothermal chlorite with deep-blue interference colour occurs as post-tectonic, metasomatic alteration product either together with quartz at the rims of carbonate porphyroblasts or together with calcite forms a calcite-chlorite rock. The metamorphic chlorite is syn-tectonic and under the microscope is characterized by grey-green interference colour.

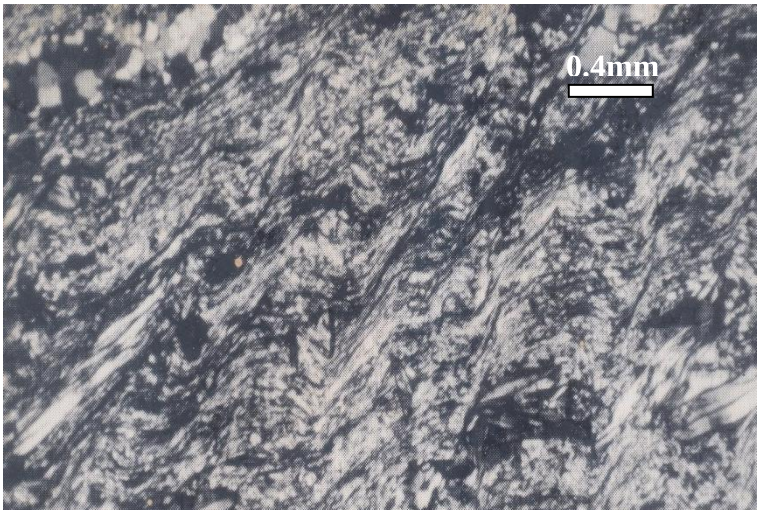

Figure 5: Photomicrograph showing single to tabular chlorite crystals in chlorite phyllite taken under cross-polars. Crenulation cleavages formed when foliation which existed in the rock was deformed into a series of small folds.

The well-crystallized metamorphic chlorite occupies the S_1 and S_2 planes and is younger than the hydrothermal chlorite. The metamorphic chlorite is characterized by the different shape and size (prismatic, 0.3 mm).

Since the alteration process in the Birimian resulted in the occurrence of a large number of different altered minerals in the country rocks and quartz veins [18, 19] it is possible to find a mineral system that can be used to determine the prevailing conditions of the alterations. In such instance, the chlorite mineral can normally be used because of its abundance, stability and syn- to post-metamorphic nature [31, 32]. Chlorites form solid solutions and show variable degree of substitution, where Al substitutes for Si in tetrahedral sites, while Mg substitutes for Fe in octahedral sites, thus introducing variability in the chemical composition of chlorites [33, 34, 35]. It has also been established that these variations are related to the bulk composition of the host rock; and increasing metamorphism and hydrothermal alteration processes usually record the physicochemical conditions of chlorite recrystallization [36, 37]. Also, because chlorites are commonly found in hydrothermal paragenesis, their non-stoichiometric characteristic has been widely used to place constraints on the thermodynamic conditions of ore deposits formation [36, 38]. Consequently, chlorites from Archaean gold deposits have been successfully modeled in the Fe-Mg-Al-Si-H_2O system to depict chemical conditions which existed during gold deposition [39, 40].

4.3.2 Chlorite geochemistry

Results of microprobe analysis of metamorphic chlorites from the Ashanti Mine are presented in Table 1. Though X-ray diffraction was not done on the chlorite, the negligible amount of K_2O, Na_2O and CaO and relatively low Si content in chlorites from the electron microprobe results (Table 1) strongly suggest that the chlorites are well-crystallized and are not interstratified or intergrown with other sheet silicate such as phengite, paragonite, margarite or smectite indicating total absence of diagenetic chlorite) [31].

Table 1: Electron microprobe analyses (wt. %) of metamorphic chlorites from Ashanti Mine

	18/0B/310/9				28/OB/155/08			28/0B/155/09			38/0B/155/10		38/0B/155/12
SiO_2	25.58	27.31	28.43	26.54	26.0	28.71	28.32	26,43	28.6	30.06	24.26	24.22	27.46
TiO_2	0.04	0.03	0.0	0.02	0.05	0.0	0.06	0.04	0.02	0.03	0.1	0.05	0.07
Al_2O_3	23.41	23.58	23.1	23.91	22.97	23.31	23.56	23.91	22.75	22.9	22.68	22.15	20.68
FeO*	11.86	18.24	13.56	20.36	23.13	9.84	8.85	20.68	9.76	9.08	22.42	23.7	18.15
MnO	0.38	0.22	0.22	0.19	0.15	0.59	0.6	0.12	0.44	0.59	0.1	0.07	0.0
MgO	23.88	18.24	22.61	17.5	14.91	25.76	25.21	16.15	25.11	23.5	15.99	15.3	19.1
CaO	0.01	0.03	0.00	0.01	0.00	0.00	0.01	0.00	0.01	0.14	0.05	0.09	0.02
Na_2O	0.01	0.03	0.00	0.00	0.00	0.01	0.01	0.00	0.05	0.00	0.09	0.04	0.00
K_2O	0.03	0.00	0.00	0.01	0.00	0.01	0.02	0.00	0.05	0.16	0.03	0.01	0.10
Total	88.21	87.68	87.94	88.53	87.22	88.22	86.65	87.34	86.8	86.48	85.77	85.67	85.57
Structural Formulae calculated on the basis of 14 oxygen													
SI	2.77	2.75	2.78	2.68	2.71	2.76	2.76	2.71	2.79	2.92	2.58	2.6	2.84
AI	1.23	1.25	1.22	1.32	1.29	1.24	1.24	1.29	1.21	1.08	1.41	1.40	1.15
Z	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00
AI	1.44	1.55	1.45	1.53	1.53	1.40	1.46	1.6	1.41	1.55	1.43	1.40	1.37
Y	1.44	1.55	1.45	1.53	1.53	1.40	1.46	1.6	1.41	1.55	1.43	1.40	1.37
Fe^{2+}	0.96	1.54	1.11	1.72	2.01	0.79	0.72	1.77	0.8	0.74	1.99	2.13	1.57
Mn	0.03	0.02	0.02	0.02	0.01	0.05	0.05	0.01	0.04	0.05	0.01	0.01	0
Mg	3.45	2.74	3.3	2.63	2.32	3.69	3.66	2.47	3.65	3.41	2.53	2.45	2.95
Ca	0	0	0	0	0	0	0	0	0	0.01	0.01	0.01	0
Na	0	0.01	0	0	0	0	0	0	0.01	0	0.02	0.01	0
K	0	0	0	0	0	0	0	0	0.01	0.02	0	0	0.01
X	4,44	4.3	4.43	4.37	4.35	4.53	4.43	4.25	4.5	4.23	4.57	4.6	4.53
Fe/(Fe+Mg)	**0.22**	**0.36**	**0.25**	**0.40**	**0.46**	**0.18**	**0.16**	**0.42**	**0.18**	**0.18**	**0.44**	**0.47**	**0.35**
Si/Al	**1.04**	**0.98**	**1.04**	**0.94**	**0.96**	**1.05**	**1.02**	**0.94**	**1.06**	**1.11**	**0.91**	**0.93**	**1.13**
* Total Fe as FeO													
XSi, (Tl)	1	1	1	1	1	1	1	1	1	1	1	1	1
XSi, (T2)	0.385	0.38	0.39	0.34	0.36	0.38	0.38	0.36	0.4	0.46	0.39	0.3	0.42
XAl, (T2)	0.615	0.63	0.61	0.66	0.65	0.62	0.62	0.65	0.61	0.54	0.61	0.7	0.58
XAl, (0)	0.24	0.26	0.24	0.26	0.26	0.23	0.24	0.27	0.24	0.24	0.24	0.23	0.23
XFe, (0)	0.192	0.31	0.22	0.34	0.4	0.16	0.14	0.35	0.16	0.15	0,40	0.43	0.31
XMg, (0')	0.76	0.74	0.76	0.75	0.75	0.77	0.76	0.73	0.77	0.77	0.76	0.77	0.77
XMg, (0)	0.538	0.4	0.51	0.38	0.32	0.59	0.58	0.35	0.58	0.53	0.35	0.34	0.44
XOH, F	1	1	1	1	1	1	1	1	1	1	1	1	1

Though no systematic determinations of microprobe analyses were made across a profile to show whether Fe/Mg ratio of the chlorites varied consequent to disequilibrium, lack of optical features such as absence of reaction rims or zoning in the minerals strongly suggest equilibrium assemblages. The Fe/(Fe+Mg) ratio varies from 0.16 to 0.47, whiles the Si/Al ratio shows little variation, ranging between 0.91 and 1.13. Similarly, Al in the tetrahedral site (Al^{IV}) varies from 1.08 and 1.41 ions per formula unit. With the exception of FeO, there is little compositional variation of the chlorite samples, suggesting that the minerals were in equilibrium with the hydrothermal fluid [40]. The chlorites were modeled for the chamosite end-member, $Fe_5Al_2Si_3O_{10}(OH)_8$ given by Bryndzia and Scott [41] and for two Mg end-members, $Mg_6Si_4O_{10}(OH)_8$ and $Mg_5Al_2Si_3O_{10}(OH)_8$ [43], which enabled the calculations of the activities of the end-members to be done.

The chlorites data from the studied samples were plotted in the geochemical classification diagram of Hey [42]. (Figure 6). The plots show that the analyzed chlorite in sample 38/0B/155/12 has a pycnochlorite composition, while chlorites in sample 38/0B/155/10 have ripidolite composition. The chlorites in samples 18/0B/310/9, 28/OB/155/08 and 28/0B/155/09 have ripidolite and sheridanite compositions, with one chlorite from 28/0B/155/09 having a clinochlore composition. These results suggest that most of the chlorites in the samples are Mg-richer, as shown in Figure 7 in which selected chemical components of the chlorites are illustrated. Compared to the other samples, chlorites in 38/0B/155/10 are Si-poor. There is no defined trend between A^{IV} and A^{VI}. Total Al is negatively correlated with Si and Mg, whereas Fe displays a negative correlation with Mg. On the contrary, positive correlations exist between A^{IV} and F/FM (Fe/Fe+Mg) and Si and Mg (Figure 7).

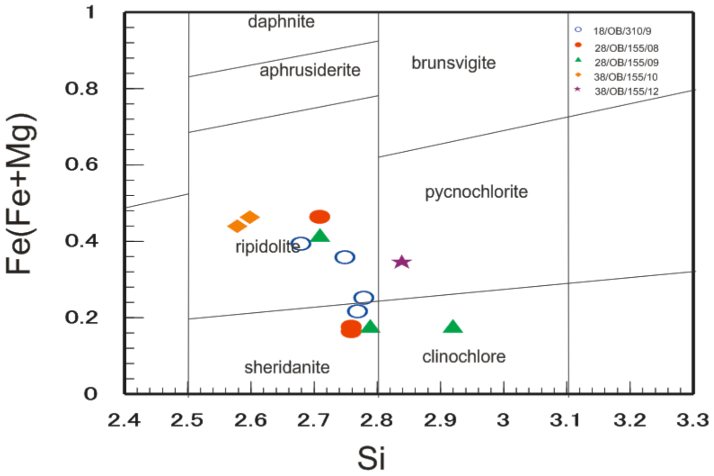

Figure 6: Geochemical classification of metamorphic chlorites from the study area (after Hey, [42]). Green triangle (sample 28/0B/155/09), filled red circle (sample 28/0B/155/08), and open blue circle (samples 18/0B/310/9) orange diamond (38/0B/155/10) and purple star (38/0B/155/12).

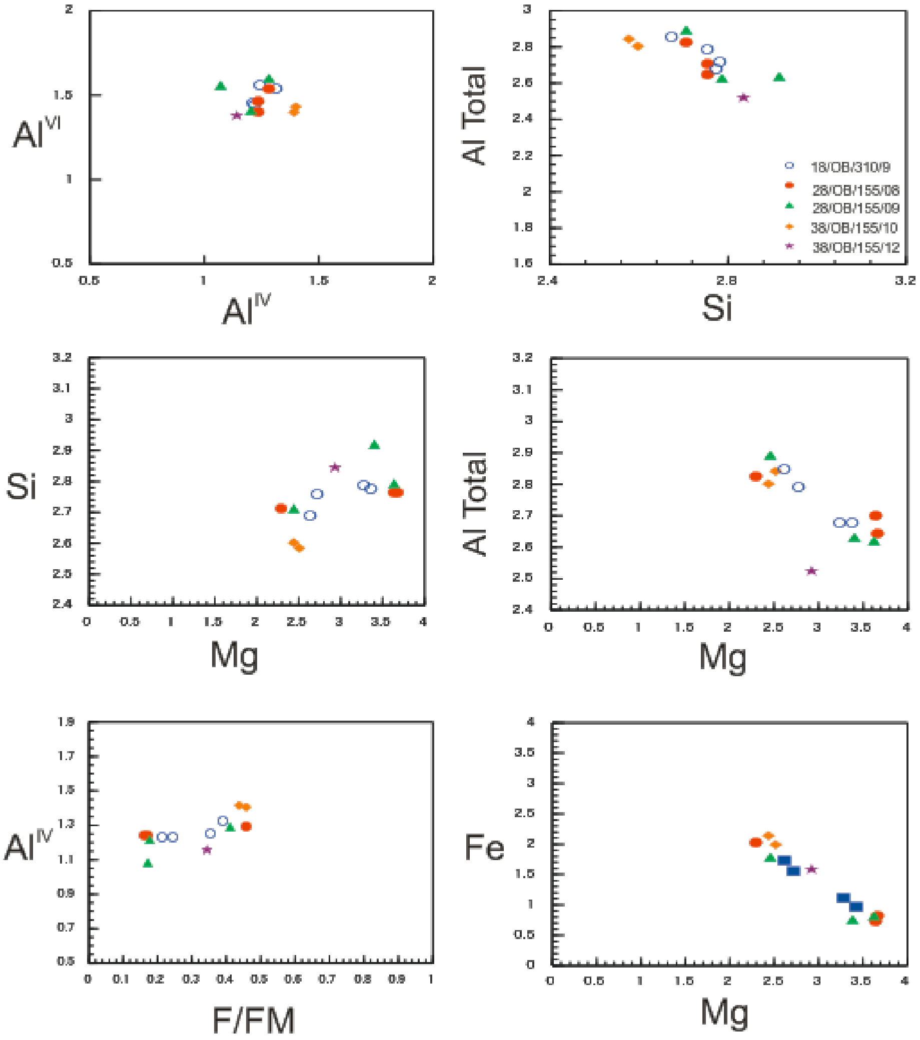

Figure 7: Compositional diagrams of the metamorphic chlorites analyzed by EMPA. Cations are expressed in a.p.f.u.

4.4 Chlorite-pyrrhotite-pyrite Relationship

Chlorite-pyrrhotite-pyrite equilibrium assemblage was used in the thermodynamic calculations based on the petrographic interpretation of the silicate mineral and ore phases, as this assemblage was seen to be in texturally stable coexistence and related to gold mineralization. Using the TWQ computer programme [44] and thermodynamic parameters for the minerals of interest, the four isothermal reactions below (equations 1 to 4) gave univariant points for the recognition of the fugacities of oxygen and hydrogen sulphide. The sources of the thermodynamic parameters for the sulphides were Helgeson et al. [44] and Scott [45] for the quartz and albite, respectively.

$$25/3FeS_2 + Fe_5Al_2Si_3O_{10}(OH)_8 + 5/6Mg_6Si_4O_{10}(OH)_8 =$$
$$40/3FeS + Mg_5Al_2Si_3O_{10}(OH)_8 + 10/3SiO_2 + 20/3H_2S + 25/6O_2 \qquad (1)$$

$2FeS_2 + 2H_2O = O_2 + 2H_2S + 2FeS$ (2)

$5H_2S + Fe_5Al_2Si_3O_{10}(OH)_8 + 5/6Mg_6Si_4O_{10}(OH)_8 =$
$5FeS + 10/3SiO_2 + 25/3H_2O + Mg_5Al_2Si_3O_{10}(OH)_8$ (3)

$40/3H_2O + 10/3SiO_2 + 5FeS_2 + Mg_5Al_2Si_3O_{10}(OH)_8 =$
$10H_2S + 5/3O_2 + Fe_5Al_2Si_3O_{10}(OH)_8 + 5/6Mg_6Si_4O_{10}(OH)_8$ (4)

Empirical chlorite geothermometers after Cathelineau and Nieva [46] showed a temperature of 360^oC for the regional metamorphism of the Birimian [47]. Chlorite-pyrrhotite-pyrite equilibrium assemblage was used in the thermodynamic calculations based on the petrographic interpretation of the silicate mineral and ore phases, as this assemblage was seen to be in stable textural coexistence and related to gold mineralization.

Fugacities of oxygen (fO_2) and hydrogen sulphides (fH_2S) calculated at 410°C and 3 kbar using a fluid composition of 0.7 mole H_2O and 0.3 mole CO_2 [26, 48] yielded low log fO_2 and log fH_2S (Table 2). These indicate relatively reduced conditions for the mineralizing fluid.

4.4.1 Chlorite-albite relationship

The chlorite-albite relationship is used together with the salinity from fluid inclusion studies [48] to calculate the likely pH of the fluid. The relationship at 410°C and 3 kbar is given as follows:

$½NaAlSi_3O_8 + 5Mg_6Si_4O_{10}(OH)_8 + ½H^+ = Mg_5Al_2Si_3O_{10}(OH)_8 + 38SiO_2 +$
$2H_2O + ½Na^+$ (5)

From Berman [43], log $a(Na^+/H^+)$ for the above relationship has a constant value of 5, where ‘a’ is the activity.

Thus, $\log a_{Na+} - \log H^+ = 5$
$-\log H^+ = 5 - \log a_{Na+}$

The calculated pH value of 5.8 is compatible with the presence of sericite, a common potassium aluminosilicate mineral, which indicates a near-neutral pH value.

Table 2: Fugacities of oxygen (fO_2) and hydrogen sulphides (fH_2S).

Log fO_2	Log fH_2S
-32.2	1.8
-31.8	1.7
-32.0	1.8
-31.6	1.6
-31.5	1.5
-32.4	1.9
-32.5	2.0
-31.6	1.6
-32.4	1.9
-32.3	1.9
-31.5	1.5
-31.5	1.5
-31.6	1.6

5 Discussion

5.1 Nature of the Ore Fluid

The mineral assemblages in the graphite-chlorite-ankerite phyllite suggest aqueous, oxidizing, near neutral, moderate temperature hydrothermal fluids interacted with the wall rocks. Fluid inclusion studies on the auriferous quartz veins indicate a parent homogeneous H_2O-CO_2 fluid with traces of CH_4-N_2 fluid formed at 370°C-410°C at pressures less than 2-4 kbar which gave rise to CO_2-rich fluids and H_2O-fluids. The salinity of CO_2-rich fluids was about 5 wt % NaCl equiv. as against roughly 16 wt % for the H_2O–fluids [26].

5.2 Transport and Deposition of the Gold

The parameters for the Birimian auriferous fluid are helpful in constraining the transport as well as deposition mechanisms. Although gold can be present in solution as Au^+ or Au^{3+}, Seward [49] concluded that at high temperatures the Au^+ is more dominant and gold in hydrothermal systems is more likely to be transported as Au^+ complexes. Also according to Seward [49], the dominant species in near-neutral pH fluids is $Au(HS)_2^-$, which is therefore expected to be predominant in the Birimian auriferous fluid of pH 5.8. In this paper, the discussion on gold deposition is focused on two types of ore body, namely, the disseminated sulphide lode gold-ore type (i.e. mineralization in the wall rocks) and the quartz vein ore type.

Philips et al. [50] and Gilligan and Foster [51] suggested that an efficient mechanism for gold deposition particularly in iron-rich wall rocks, is the destabilization of the $Au(HS)_2^-$ complex through sulphidation of iron in the wall rocks. They argued that by this process gold deposition is accompanied by growth of pyrite, pyrrhotite or arsenopyrite. Though rocks such as high-Fe tholeiitic basalts and banded iron-formation have not been reported in the Birimian in Ghana [18], the presence of significant amount of ankerite/siderite and gold-rich zones in the tuffaceous bodies (i.e. the second mined ore body next to the quartz veins), which are characterized by lots of gold-rich arsenopyrite does not exclude an active role of sulphidation mechanism towards the deposition of gold in the wall rocks.

From the mineral assemblages, it is clear that the gold deposition was also related to fluid mixing and fluid-wall rock interactions which constitute a sequence of hydrothermal alteration events associated with shear-deformation. The first stage of fluid interaction with the host rock(s) was followed by more intense shearing and infiltration of mainly auriferous hydrothermal fluid that replaced most of the older alteration minerals and deposited the mineral assemblages observed in the chlorite-graphite-ankerite phyllite/ankerite metarhyolite. This ore stage alteration process involved variable degrees of silicification, sulphidation, rutile-formation, sericitization, carbonation, carbonatization and different stages of chloritisation. SiO_2, Mg, Fe, Ca, C and B were liberated and deposited in fractures to form quartz/or carbonate/or tourmaline veins.

Furthermore, if gold is transported as $Au\ (HS)_2^-$ then precipitation of gold can occur as a result of: (i) decrease in pH, (ii) decrease in temperature or (iii) decrease or increase in fO_2 [52, 53]. At the Anglogold Ashanti mine in Ghana, the fO_2 calculated for two depths at ≈300 m (sample 18/OB/310/9) and ≈900 m (samples 38/OB/155/10 and 38/OB/155/12) shows virtually no changes at constant pH values. This suggests that the precipitation of gold would be influenced by falling temperature.

Another important gold deposition mechanism is phase separation [54, 55]. This mechanism was very efficient in view of the phase separation which occurred in the Birimian fluid and subsequently led to the separation into CO_2-rich and H_2O-rich fluids. This separation may have caused the destabilization of the gold complexes and the gold deposition. This mechanism is interesting for the Birimian fluids, considering the fact that the CO_2-rich fluid type was dominant in the auriferous quartz veins in the prominent mines (e.g. Ashanti and Prestea Mines), whereas the H_2O-rich fluid type dominated in the quartz veins gold prospects such as those in Kubekrom [26]. The fH_2S values of $10^{1.5}$ -$10^{2.0}$ bars show the importance of aqueous-reduced sulphur species and the reducing nature of the depositional environment, which is indicated by large amount of carbonaceous matter.

6 Conclusion

Field observations and petrographic studies of altered and graphite phyllites suggest that the metasedimentary and metavolcanic rocks in the Birimian gold belt were transformed mainly by metamorphism and hydrothermal activities. Interaction of hydrothermal fluid with the host rocks produced episodes of hydrothermal alteration associated with shear-deformation, resulting in mineral ore-forming mineral assemblages gold mineralization.

Chlorite-pyrrhotite-pyrite and chlorite-pyrite relationships allowed the calculation of fugacities of oxygen and hydrogen sulphide and pH. The values of fO_2 of $10^{-31.5}$ to $10^{-32.5}$ bars, of fH_2S of $10^{1.5}$ to $10^{2.0}$ bars and pH of 5.8 were used to constrain the gold transport and deposition mechanism that indicates that the gold was transported mostly as $Au\ (HS)_2^-$ complex in a highly reducing, near neutral fluid. Gold precipitation in the wall rocks occurred when the gold complex was destabilized by the phase separation of the fluid into CO_2-rich and H_2O-rich phases during falling temperature regimes in the growth of quartz veins, arsenopyrite, pyrite and pyrrhotite coupled with the formation of ankerite and siderite.

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