

Thermodynamic Stability of Microcline in Hydrothermal Ore System of Malanjkhanda Copper Deposit, India

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Abstract: Mineral solubility calculations were used to establish the phase relations for the microclinization process at elevated temperature and pressure conditions. Mineral phases identified in the mineralization zone of the Malanjkhanda copper deposit, are the key component to construct hydrothermal alteration reactions by plagioclase, microcline, kaolinite, biotite, chlorite, and chalcopyrite. Theoretical modeling using standard state thermodynamic database suggested that the activity of K^+ ions regulate the microclinization process in the temperature range 300°C to 350°C with an insignificant consequence of pressure during a hydrothermal alteration in the Malanjkhanda copper deposit. Potash-rich hydrothermal solution promotes the deposition of ore minerals in Malanjkhanda copper deposit. Correlation of temperature with equilibrium kinetic and hydrothermal alteration processes are indicative of mineral-mineral and mineral-fluid stability in natural environments. The results of this study are very useful in crucial mineral exploration planning and developments of theoretical models of hydrothermal fluid-granite reactions.

Keywords: Granite, microcline, activity, alteration, Malanjkhanda.

1. Introduction

Geochemical processes in hydrothermal systems involve several mineral fluid reactions and affect the geochemical properties of granitic rocks¹. The chemical reactions depend on the species concentrations, the volume of the fluid, and the amount of mass transfer during hydrothermal alteration process². Mineral fluid reactions mechanisms of the granite proceed mainly by dissolution-precipitation processes³. This is resulting from the infiltration of hydrothermal fluid along microcracks which causes high mobility of metals and mineralogical changes in the granites⁴. Minerals in granites are

altered by the penetration of hydrothermal fluids along with cracks and fractures. Hydrothermal fluids with multiple ions exchange and complex reaction processes associated with active agents of acid alteration and ore-metal transport⁵.

Potassium feldspar (K-feldspar, KAlSi_3O_8) is a common mineral constituent in a wide variety of rocks whereas its natural host is granite. It has four polymorphs found in the natural rocks such as sanidine, orthoclase, adularia, and microcline⁶. Microcline is the lowest temperature triclinic polymorph⁷ of K-feldspar whereas sanidine, orthoclase, and adularia are moderate to high-temperature polymorphs⁸. The triclinic microcline is distinguished from other varieties of K-feldspar by its characteristic cross-hatched twinning or tartan pattern observed in petrographic thin section under a polarizing microscope⁹. Sometimes, microcline is occurring with untwined crystal which belongs to the monoclinic system⁶. Microcline is commonly formed by low-temperature metasomatic or hydrothermal or secondary alteration processes¹⁰ of potassium bearing primary silicate minerals such as feldspar, mica, etc. The process of formation of microcline is well-known as microclinization¹¹.

The Malanjkhanda Granitoid Complex is mainly composed of granite-granodiorite with regionally distributed grey and pink color granite predominantly confined to the mineralized quartz reef^{12,13} (Fig. 1a,b). Pink coloration observed in the granite around the mineralized quartz reef is due to the presence of K-feldspar megacrysts and these megacrysts are microclines^{14,15}. It is formed due to hydrothermal alteration of plagioclase in the grey granite¹⁶. In the Malanjkhanda copper deposit, silicate minerals are associated with sulfide ore phases which indicates they are equilibrated with the mineralizing hydrothermal fluids^{17,18}. This study has, therefore, focused on the microclinization processes in the granite caused by hydrothermal alteration during mineralization. This paper discussed the chemical and mineralogical changes in the granite ore system which is helpful in the understanding of thermodynamic stability of microcline in a natural environment.

2. Geological Settings

The Paleoproterozoic Malanjkhanda granite (Fig. 1b) is a prominent unit of Bastar craton exposed in an area of about 1400 km², hosting the giant Cu-Mo-Au bearing arcuate shape quartz reef. Panigrahi *et al.*¹⁹ provided Pb-Pb zircon ages and reclassified in terms of Malanjkhanda Granitoid Complex-I (MGC-I: grey + pink granite) with the emplacement age of MGC-I unit is 2.48 Ga and Malanjkhanda Granitoid Complex-II (MGC-II: leucogranite) located towards

the south of copper mine near Birsa village. There are sporadic occurrences of microcline as impregnations in the mineralized quartz reef along with ore minerals^{13,15,17,18}. Fresh and unaltered megacrysts of microcline in the granite around mineralized zone enclosed along with patches of saussuritized plagioclase^{17,20}.

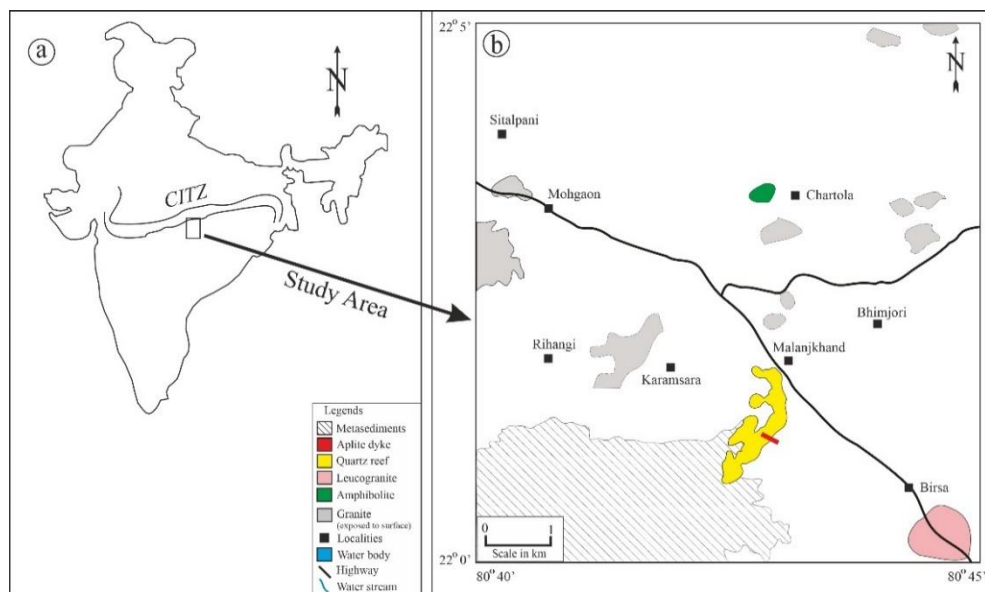


Figure 1. Map of the study area shows: (a) study area adjacent to the CITZ in the map of India and (b) local geological map of the Malanjkhanda copper deposit with sporadic occurrences of granitic outcrops.

Replacement relationship with saussuritized plagioclase and non-perthitic nature of microcline indicates precipitation of microcline from the aqueous solution after saussuritization of plagioclase during hydrothermal alteration processes in the Malanjkhanda granite^{15,21}. The presence of quartz megacrysts contains inclusions of saussuritized plagioclase and altered hornblende or biotite in the Malanjkhanda granite inferred quartz precipitation similar to that of microcline which post-dated the early hydrothermal alteration processes under similar physicochemical conditions¹⁴. Hydrothermal alteration processes in the hydrothermal ore system of Malanjkhanda copper deposit involve chloritization of hornblende and biotite with the concurrent saussuritization of plagioclase. All these observations strongly suggested that the presence of a regionally pervasive fluid phase brings about hydrothermal alterations in the early stage of mineralization which is followed by microclinization and silicification^{18,22}. Microclinization and silicification are pronounced because of precipitation of microcline and quartz respectively, in

the granite around the mineralized zone however incipient in other parts of the study area.

3. Malanjkhand Copper Deposit

The Malanjkhand copper deposit is a typical fossil hydrothermal deposit hosted in the Paleoproterozoic granite pluton in Central India^{14,21}. This mineralized granite is surrounded by older gneisses, metasedimentary rocks of Saussar and Sakoli Group; volcano-sedimentary sequences of Dongargarh Group, and sedimentary rocks of Chhattisgarh Group¹². There is no systematic alteration pattern observed in the granite around the mineralized quartz reef where microcline impregnation occurs in variable amounts (Fig. 1b). Fresh microcline also occurs in the stringers, pegmatites, and disseminated sulfide ores minerals¹⁶ (Fig. 2a). Since hydrothermal alteration processes in the Malanjkhand granitoid predates quartz-microcline precipitation which broadly overlapped with precipitation of sulfide ore minerals²³. Chalcopyrite is the dominant sulfide ore minerals that occurred along with fresh microcline megacrysts in the cleavage and fractures spaces without any accompanying alterations features. Fine-grained microcline veins along with quartz ($\pm$ chalcopyrite) are also present in minor amounts (Fig. 2b).



Figure 2. Field photos displays: (a) quartz vein in the K-feldspar rich granite and (b) sulfide ore minerals associated with quartz and microcline.

The mineralized reef and stringers ore contains negligible amounts of silicate minerals other than quartz and microcline with sporadic occurrences of epidote and chlorite in the mineralized zone. Quartz and microcline are the major silicate phases associated with all variety minerals in the Malanjkhanda deposit.

4. Petrography

The common petrographic features in the Malanjkhanda granite are megacrysts of microcline with crosshatched twinning and orthoclase with perthitic intergrowth in the mineralized quartz reef (Fig. 3a.) Hydrothermal alteration features are predominantly observed in the mineralized zone with occurrences of saussuritized plagioclase grains (Fig. 3b) that contain minute inclusions of secondary epidote and muscovite. Biotite and hornblende are the primary mafic minerals associated with microcline and chlorite in the mineralized zone (Fig. 3c). Chalcopyrite is the dominant copper ore mineral associated with the quartz reef along with pyrite (Fig. 3d) and other sulfide ore minerals¹⁵.

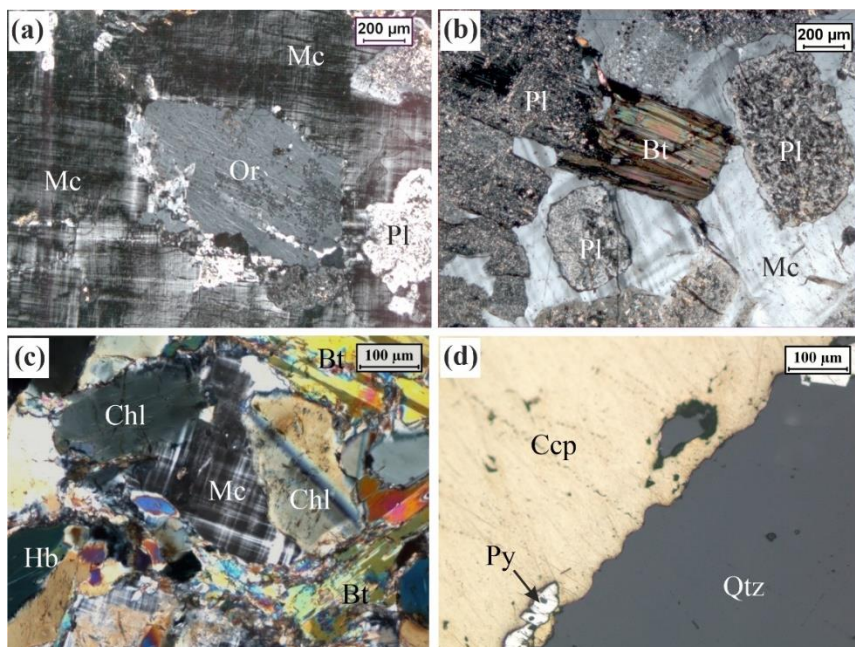


Figure 3. Petrographic microphotograph shows: (a) microcline megacryst with inclusion of orthoclase, (b) saussuritized plagioclase with biotite and microcline, (c) hornblende + biotite + microcline + chlorite mineral assemblage and (d) ore mineral assemblage chalcopyrite + pyrite + quartz. Mineral abbreviations are obtained in accordance with Kretz²⁴.

The stability relation among various ore mineral phases was constructed using thermodynamic modeling^{15,23}. However, the involvement of microcline in the copper mineralization at Malanjkhand needs further investigations.

5. Hydrothermal Processes

The solubility of minerals and speciation of ore metals in hydrothermal solutions²⁵ is graphically depicting phase relationships, mineral solubilities, and fluid evolution²⁶. The internally evolved low-temperature and moderately high saline pervasive fluid phase in the surrounding Malanjkhand granite acquiring enough metal, silica, and potash through prolonged fluid-rock interaction^{14,18}. The copper ore mineral has been altered by percolating oxygenated and carbonated hydrothermal solutions through a series of geochemical reactions that produce the various ore assemblages²³. This caused a hydrothermal alteration, transportation of metal, silica, and potash to the fracture zone^{15,21}. It follows deposition of microcline and continued precipitation of quartz and ore minerals in the fracture zone on mixing of relatively higher temperature low-saline fluid phase of much smaller proportion evolved from subordinate leucogranite unit²². Overall, the microclinization processes in the hydrothermal ore system at Malanjkhand can be explained by the following mineralogical reactions involving aqueous species:

- (1) $\text{NaAlSi}_3\text{O}_8 + \text{K}^+ = \text{KAlSi}_3\text{O}_8 + \text{Na}^+$
- (2) $\text{CaAl}_2\text{Si}_2\text{O}_8 + 4\text{SiO}_2(\text{aq}) + 2\text{K}^+ = 2\text{KAlSi}_3\text{O}_8 + \text{Ca}^{2+}$
- (3) $10\text{Bt} + \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + 7\text{H}_2\text{O} + 10\text{H}^+$
 $= 6\text{Chl} + 14\text{SiO}_2(\text{aq}) + 5\text{KAlSi}_3\text{O}_8 + 10\text{K}^+$
- (4) $2\text{Bt} + 6\text{Cu}^+ + 12\text{H}_2\text{S} + 1.5\text{O}_2$
 $= 2\text{KAlSi}_3\text{O}_8 + 6\text{CuFeS}_2 + 6\text{H}^+ + 11\text{H}_2\text{O}$

where Bt = Biotite $[\text{K}(\text{Mg},\text{Fe})_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2]$

Chl = Chlorite $[(\text{Fe},\text{Mg})_5\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_8]$

Mineral assemblages observed under the microscope formed the basis of constructing mineralogical reactions between various phases. These reactions are used to derive equilibrium constant and solubility factor for reactions (1) to (4). The relationship between equilibrium constant and temperature ($T^\circ\text{C}$) is obtained from thermodynamic datasets SUPCRT92 assumed all calculations in the range from 25°C to 600°C at 0.5 to 2.0 kbar pressure (P) for the natural buffer limits of pH, $f\text{O}_2$, $f\text{H}_2\text{O}$, and $f\text{H}_2\text{S}$ ¹⁵.

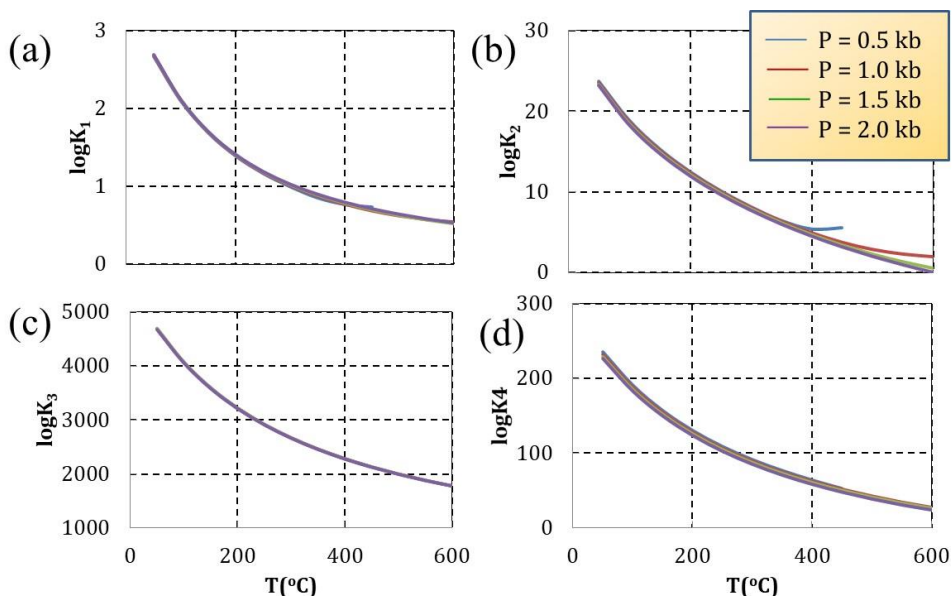


Figure 4. Temperature ($T^{\circ}\text{C}$) versus equilibrium constant diagram for mineralogical reactions (1) to (4) are calculated at different pressure in the range from 0.5 to 2.0 kbar.

The generalized thermodynamic properties of equilibrium reactions are taken from Johnson *et al.*²⁷ using the SUPCRT92 program with the *dslop04.dat* database. Considering activity of ideal phases is unity and activity of fluid or gaseous phases are taken as fugacity (f).

$$\log K_1 = \log(a_{\text{Na}^+}) - \log(a_{\text{K}^+})$$

$$\log K_2 = \log(a_{\text{Ca}^{2+}}) + 4\log(a_{\text{SiO}_2, \text{aq}}) - 2\log(a_{\text{K}^+})$$

$$\log K_3 = 10\log(a_{\text{K}^+}) + 14\log(a_{\text{SiO}_2, \text{aq}}) - 7\log(f_{\text{H}_2\text{O}}) + 10\text{pH}$$

$$\log K_4 = 11\log(f_{\text{H}_2\text{O}}) - 6\log(a_{\text{Cu}^+}) - 12\log(f_{\text{H}_2\text{S}}) - 1.5\log(f_{\text{O}_2}) - 6\text{pH}$$

Temperature versus equilibrium diagrams (Fig. 4a-d) suggested that the effect of pressure on the mineralogical reaction is very restricted variations observed in the range of 0.5 to 2.0 kbar and temperature range 25°C to 600°C necessitate for hydrothermal alteration in the Malanjhand copper deposit.

6. Microclinization

The late-stage hydrothermal alteration processes are characterized by kaolinite, chlorite, and pyrite saturation whereas albite and K-feldspar occur as a replacement of plagioclase²⁸. In the Malanjhand copper deposit, K-

feldspar occurs in the form of cavity lining, crosscutting fractures, overgrowths, and cavity fillings in the mineralized zone (Fig. 2a). Petrographic photomicrograph shows that K-feldspar is dominant of microcline type (Fig. 3a-c) and plagioclase typically forms clouded crystals (Fig. 3b). Physical observations on several parameters such as temperature, pressure, and activities of species in fluid show the most important factors that control the evolution of the hydrothermal system²⁹. Dissolution of feldspar in the hydrothermal solutions is enhanced by ion-exchange reactions and increase the rate of silica solubility³⁰. The calculated equilibrium constant for microcline indicates much higher solubility under hydrothermal conditions (25°C to 600°C). The thermodynamic properties of microcline and aqueous species that form between hydrothermal solutions and minerals are essential for quantitative interpretation of dissolution-precipitation reactions³¹. Mineral solubility calculations were performed to understand hydrothermal alteration processes at elevated temperatures and pressures to evaluate the formation of microcline in the Malanjhand copper deposit. The standard state adopted in this study for all mineral phases and hydrothermal solution in unit activity at the temperature (150°C to 400°C) and pressure (1 kbar) of mineralization^{15,18,20}. The natural buffer limits of f_{O_2} , $f_{\text{H}_2\text{O}}$, and $f_{\text{H}_2\text{S}}$ are considered unity for ideal species in the temperature range 25°C to 600°C. Assuming the silica activity ($a_{\text{SiO}_2,\text{aq}}$) is unity for all calculations at quartz saturation³² because of extensive mobilization of silica during the hydrothermal alteration processes in the Malanjhand copper deposit^{14,33}.

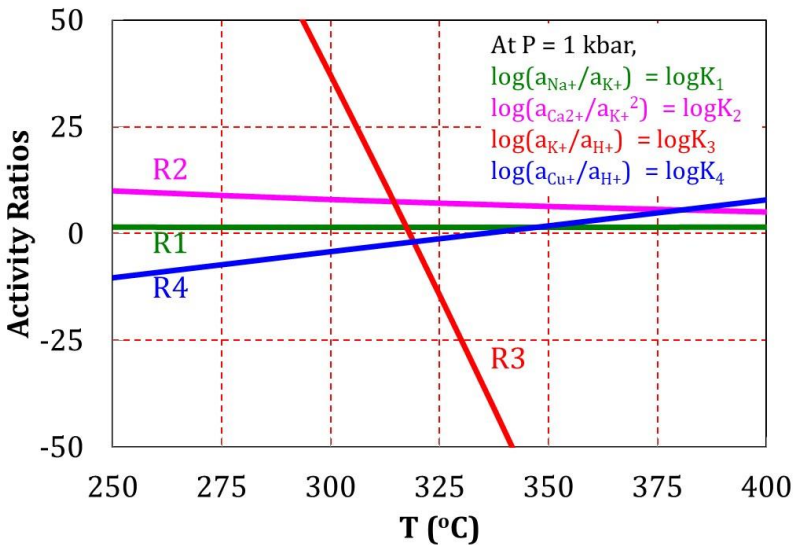


Figure 5. Temperature ($T^{\circ}\text{C}$) versus activity ratios diagram for the mineralogical reactions related to microclinization.

Previous studies also demonstrated excess silica crystallized to form various types of quartz textures^{34,35} and accessory minerals³⁶. All other minerals in the hydrothermal system are represented by saturation limits as a function of $\log(a_{\text{Na}^+}/a_{\text{H}^+})$, $\log(a_{\text{K}^+}/a_{\text{H}^+})$, $\log(a_{\text{Ca}^{2+}}/a_{\text{H}^+})$, $\log(a_{\text{Cu}^{2+}}/a_{\text{H}^+})$ and $\log(a_{\text{Ca}^{2+}}/a_{\text{K}^+}^2)$, which correspond to the descriptive variables in the stability diagrams³². The activity of K^+ ions ($\log(a_{\text{K}^+}/a_{\text{H}^+})$) dynamically control the microclinalization process in the temperature range 300°C to 350°C with the negligible effect of pressure. Potash-rich hydrothermal solution energetically assisted copper mineralization in the Malanjkhand quartz reef. Therefore, activity ratios of different species are calculated at 1 kbar pressure to construct the stability diagram (Fig. 5) for hydrothermal alteration processes responsible for copper mineralization.

7. Conclusions

The present study suggested that the microcline occurs in variable proportions in the Malanjkhand granite, quartz reef, stringers, and disseminated sulfide ore-minerals in the mineralization zone. Quartz and microcline are associated with all varieties of minerals in the Malanjkhand copper deposit. Hydrothermal alteration processes are involved in the mobilization of metal ions, silica, and potash to the fracture zone. Further, late-stage hydrothermal processes necessitate deposition of microcline, precipitation of quartz and sulfide ore minerals in the fracture zone responsible for the formation of mineralized quartz reef. The activity of K^+ ions ($\log(a_{\text{K}^+}/a_{\text{H}^+})$) strongly influenced the microclinalization process in the temperature range 300°C to 350°C with the negligible effect of pressure during the hydrothermal alteration processes in the Malanjkhand copper deposit.

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