فشار، حرارت و ترکیب سیالات در هاله دگرگونی کلیر، شمال غرب ایران: استفاده از تعادلات کاتی شامل در سنگهای رسی و کالک-سیلیکات

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چکیده: با نفوذ توده آذرین، نفلین سیپنت کلیر به داخل سنگهای رسی و آهکی کرتن، هالهی دگرگونی به ضخامت حدود یک کیلومتر ایجاد شده است. شش زون دگرگونی با کانه‌ای چون کردنیت، آکالی فلدسپار، سیلیمات و کوروندو در سنگها تشکیل شده است. سنگهایی که ترکیب شیمیایی مناسب داشته‌اند به علت حرارت توده به صورت بخشی ذوب شده‌اند. در سنگهای کالک-سیلیکات نیز نجوت گون کانه‌ای چون کردنیت، دیوپسیت، گارنت، اسکاپین‌یت، وولاسانتیت و وروزانتیت تشکل شده است. برای تخمین فشار دگرگونی در سنگهای رسی از پتروژنتیک گرد استفاده شد. نتایج محل مفاهیم در سنگهایی بر روی پتروژنتیک گردید. فشاری حدود ۳ کیلوبار را نشان می‌دهد. محاسبه محل مفاهیمی و اکسبتوسی مربوط به کالک-سیلیکات‌ها با فرض اکسبتوسی مساوی یک برابر تمام فازها و رس آنها بر روی نمودار حرارت-کسر مولی دی اکسید کربن، نشان می‌دهد که سر مولی دی اکسید کربن هنگام اوج دگرگونی برای با ۲/۲ بوده است که این مقدار تقیی مستقل از فشار دگرگونی است. محاسبه محل مفاهیم در سنگهایی بر روی نمودار حرارت-کسر مولی دی اکسید کربن با فرض انحلال جامد کانه‌ای و اکسبتوسی های غیر از یک، نتیجه استحیال را می‌دهد و مشخص می‌کند که ترکیب واقعی سیال در اوج دگرگونی مجاورتی با ترکیب معادل کسر مولی دی اکسید کربن برای ۲/۲ بی‌پیوند بوده است. گرمای اوج دگرگونی برای زون سیلیمات در سنگهای رسی ۴۵۰-۴۰۰ درجه سانتی‌گراد و برای زون وولاسانتیت-وروزانتیت در سنگهای کالک-سیلیکات‌ها ۲۰۰-۷۰۰ درجه سانتی‌گراد بوده است.
ویژه‌های کلیدی: سیلیمانتیت، کالک سیلیکات، فشار، دما، ترکیب سیالات.
Pressure, temperature and fluid composition in the Kalaybar aureole, NW Iran; Mineral equilibria in metapelitic and calc-silicate rocks

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Key words: pelites, calc-silicate, pressure, temperature, fluid composition, Kalaybar aureole

Abstract: Intrusion of the Kalaybar nepheline syenite into the Cretaceous pelitic and calcareous sediments has produced a thermal aureole up to 1km thick. Six mineralogical zones are developed in the pelitic rocks with minerals such as cordierite, K feldspar, sillimanite and corundum. Chemically suitable rocks are partially melted in response to the heat from the pluton. Five mineralogical zones are developed in the calc-silicate rocks with minerals such as tremolite, diopside, garnet, scapolite, wollastonite and vesuvianite. A petrogenetic grid was used for pelitic rocks to put constraints on pressure of metamorphism. Intersection of the reaction curves on the grid indicates a pressure of ca. 3 kbar for the contact metamorphism in the Kalaybar aureole. Calculation of the positions of the reaction curves for the calc-silicate rocks assuming unit activity for all phases, and plotting them on T-X$_{CO2}$ diagrams shows that the X$_{CO2}$ is about 0.2 and it is almost independent of pressure. Calculation of the position of the reaction curves assuming dilution of the phases due to solid-solution gives the similar results indicating that the actual composition of the fluid had a X$_{CO2}$ about 0.2 during peak metamorphic condition. The temperature was 700-750°C for sillimanite zone in pelites and 600-700°C for wollastonite-vesuvianite zone in the calc-silicates.
Introduction

Contact metamorphic aureoles are natural laboratories that can demonstrate phase relations and metamorphic reactions with little complexity [8]. This is because pressure is almost constant during contact metamorphism, and contact meta-morphic aureoles do not experience complex multiple tectonic events as regional metamorphic rocks. Estimation of pressure of contact metamorphism can provide information about depth of intrusion of plutons which itself can give valuable information about condition of the earth crust during intrusion and contact metamorphism (e.g. [2]). Fluids play an important role in metamorphism and assessing their composition is very important for explaining some aspects of contact metamorphism. Presence or absence of fluids with different compositions, during metamorphism in one hand controls the rate and grade of metamorphism and in other hand influences the phase relations in the metamorphic rocks [2, 12, 18]. Fluids help heat conduction within the thermal aureoles. Difference in permeability of the country rocks may cause disturbances in distribution of metamorphic isograds [8]. The main factors in controlling the composition of fluids are dehydration and decarbonation because of metamorphic reactions, infiltration of an external fluid and exertion of fluids from the crystallising pluton. Study of calc-silicate rocks can help to understand the nature of fluids during metamorphism [18]. The main fluid during metamorphism of pelitic and basic rocks is H$_2$O whereas the fluids in metamorphism of calc-silicates are H$_2$O-CO$_2$ mixture [18].

Pressure and temperature (P-T) of contact metamorphism is addressed in this paper. Mineral equilibria in pelitic and calc-silicate rocks are employed to elucidate the P-T condition of the Kalabar aureole.

Fluid composition during metamorphism of calc-silicates in the Kalabar aureole is demonstrated using fluid-present reactions in these rocks. The method of investigation and the results are discussed.

Geology of the studied area

According to the geological subdivision of Iran, Kalaybar area is located within the western Alborz-Azarbaijan zone [13]. The oldest rocks in the Kalaybar area are Pre-Cretaceous regionally metamorphosed rocks with a lithology of phyllite and chlorite schist. Calc-silicate and pelitic rocks of Cretaceous are exposed at the northern and eastern parts of the aureole. Quaternary sediments cover the older rocks. Magmatism in the Kalaybar area can be divided into five main episodes. These are clinopyroxenite Plutonism, Cretaceous basic volcanics, analcime-bearing tephritic volcanism, nepheline syenite plutonism and Quaternary basaltic volcanism [1, 3, 10, 20]. The Kalaybar nepheline syenite pluton with an area of about 70 km$^2$ is the largest feldspathoid-bearing rock body in Azarbaijan and probably in Iran. Intrusion of this pluton is the main cause of thermal metamorphism in the Kalaybar area, which is studied here. This pluton is not homogenous and consists of variety of rocks including nepheline syenite,
syenite, nepheline gabbro, microdiorite, diorite and monzonite [1]. The intrusion of the rocks is related to the Eocene-Oligocene magmatism of Azarbaijan [10]. This intrusion has developed a thermal aureole up to 1km thick in the Cretaceous clac-silicate, pelitic and basic rocks (Fig. 1).

Fig. 1. Geological map of the northern part of the Karaybar aureole with sample localities.
Metamorphism of the pelitic rocks

Pelitic rocks are the most suitable rock types for metamorphic studies because they are very sensitive to pressure and temperature changes and produce new mineral phases due to changes in the metamorphic grade. Nineteen thin sections from the pelitic rocks within the Kalaybar aureole were studied in detail using, XRD and scanning electron microscope. Fig.1 illustrates the distribution of samples collected from the aureole. The following mineral parageneses were distinguished in the pelitic rocks (all parageneses include plagioclase and ilmenite as non-AFM minerals; abbreviations from Kretz [9]).

\[
\begin{align*}
\text{Qtz+Chl} & \quad (1) \\
\text{Qtz+Chl+Ms} & \quad (2) \\
\text{Qtz+Chl+Ms+Bt+Crd} & \quad (3) \\
\text{Qtz+Ms+Bt+Crd+Kfs} & \quad (4) \\
\text{Qtz+Ms+Bt+Crd+Kfs} & \quad (5) \\
\text{Qtz+Ms+Bt+Crd+Kfs} & \quad (6) \\
\text{Qtz+Ms+Bt+Crd+Kfs+Crd+melt} & \quad (7) \\
\text{Qtz+Ms+Bt+Crd+Kfs+Crd+Sil+melt} & \quad (8) \\
\text{Bt+Kfs+Crd+Sil+Crn (in quartz absent rocks)} & \quad (9) \\
\text{Qtz+Crd+Kfs+Opx+melt} & \quad (10)
\end{align*}
\]

These parageneses can be divided into six main groups. These groups are studied with more details below.

1) Parageneses without biotite and cordierite (unmetamorphosed rocks)
These rocks are fine-grained and dark colored in hand specimen. There is no thermal effect on the rocks and they are fissile and are consisted of quartz, plagioclase, chlorite, muscovite and graphite.

2) Parageneses with biotite and cordierite
These rocks appear as dark coloured, hard hornfelses in the field and show fine-grained hornfelsic texture under the microscope, occasionally with porphyroblasts of biotite and cordierite in a matrix of fine-grained muscovite, plagioclase and cordierite. Biotite is the first contact metamorphic mineral in the rocks. The modal proportion of chlorite decreases and the modal proportion of biotite and cordierite increases as the distance from the contact decreases (the grade of contact metamorphism increases). The main mineral paragenesis in the lower grade rocks of this category is Qtz+Ms+Bt+Crd. As the grade of metamorphism increases alkali feldspar replaces muscovite and the paragenesis is Qtz+Bt+Crd±Ms+Kfs. Alkali feldspar appears as fine-grained crystals.

3) Parageneses with sillimanite
The main texture of these rocks is porphyroblastic with porphyroblasts of cordierite and sillimanite in a finer ground of quartz, biotite and alkali feldspar. The main paragenesis of the sillimanite-bearing rocks is Qtz+Bt+Crd±Kfs+Sil.

4) Parageneses with corundum and spinel and without quartz
These rocks show coarse and even-grained textures and appear as xenoliths within the nepheline syenite or very close to the contact. The main minerals are alkali feldspar, cordierite, spinel and corundum. Spinels are green coloured and have hercynitic composition [3].

5) Parageneses with orthopyroxene

These rocks occur in the vicinity of the igneous rocks, at the contact. Cordierite can be distinguished as dark spots on the rocks in the hand specimen. The main texture of the rocks under the microscope, is porphyroblastic with porphyroblasts of orthopyroxene and cordierite. The main paragenesis is Qtz+Crd+Bt+Kfs+Opx+melt.

6) Parageneses with melt

Chemically suitable pelitic rocks are partially melted within the Kalaybar aureole [3]. Melted parts include light domains (mesosomes) and dark domains (paleosomes). The main minerals in the mesosome parts are quartz and alkali feldspar with an igneous texture specially interstitial texture of quartz crystals and idiomorph to sub-idiomorph texture of alkali feldspars. Structural characteristics of the light domains (vein and droplet like features), textural differences between light domains and dark domains (light domains have igneous texture whereas the dark domains have hornfelsic texture and existence of granophytic texture within the light domains) and mineralogical composition of the light domain (mainly quartz and alkali feldspar) all agree that the light domains are actually partially melted mesosomes and the dark domains are unmelted paleoso-mes. Mineral parageneses of partia-llly melted rocks are:

\[
\begin{align*}
\text{Qtz} + \text{Kfs} + \text{Crd} + \text{Bt} + \text{melt} & \quad (11) \\
\text{Qtz} + \text{Kfs} + \text{Sil} + \text{Crd} + \text{Bt} + \text{melt} & \quad (12) \\
\text{Qtz} + \text{Kfs} + \text{Bt} + \text{Crd} + \text{Opx} + \text{melt} & \quad (13)
\end{align*}
\]

Table 1 includes mineral assemblages in the studied pelitic and semi-pelitic hornfelses from the Kalaybar aureole. Considering mineral parageneses, pelitic hornfelses in the Kalaybar aureole define six mineralogical zones, which are (I) biotite-cordierite zone, (II) andalusite–K-feldspar zone, (III) sillimanite zone, (IV) spinel-corundum zone (in the quartz absent rocks), (V) melt zone, and (VI) orthopyroxene zone. KFMASH minerals in the studied samples from the different zones of the Kalaybar aureole are plotted on the AFM diagrams in Fig. 2. Considering tie line arrangements on these diagrams, a set of metamorphic reactions can be written for the pelitic rocks. These reactions are summarised in Table 2.

**Metamorphism of calc-silicates**

These rocks are fine-grained grey coloured in the Kalaybar aureole. Porphyroblasts of scapolite and garnet can be distinguished in the hand specimens in the rocks close to the contact. The first effect of thermal metamorphism is recrystallisation of calcite grains. Tremolite is developed in the outer aureole, which is replaced by diopside in the higher grades.
Epidote in low-grade rocks and anorthitic plagioclase in the higher-grade rocks are other minerals. Garnet appears as isotropic minerals both in form of bands and as idioblastic crystals. Fig. 3 illustrates Back Scattered Electron images (BSE) from garnet, pyroxene and anorthite in the Kalaybar calc-silicates. Semi-quantitative analyses of the minerals using an EDS analyser, combined with SEM, indicates that garnets are mainly grossularite-rich, pyroxenes are essentially diopside, plagioclases are An-rich and scapolites are meionite-rich. As it is clear from these images, none of the minerals shows compositional zoning. Scapolite shows poikiloblastic texture with inclusions of calcite, garnet and diopside. Wollastonite is formed in the rocks close to the igneous contact and has an acicular to tabular texture. Vesuvianite (idio-crase) app-ears in the rocks adjacent to the contact. Quartz is present almost in all studied calc-silicates. The main mineral assemblages in the calc-silicates of the Kalaybar aureole are as follows (abbreviations from Kretz [9]):

\[
\begin{align*}
\text{Cal+Qtz+Ep} & \quad (14) \\
\text{Cal+Qtz+Ep+Tr} & \quad (15)
\end{align*}
\]

Table 1: Mineral assemblages in the pelitic rocks of the Kalaybar aureole. ∗ = Major phase, ○ = Minor phase, × = XRD, L = Partial melting.

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Pressure, temperature and fluid composition ….
Table 2: Metamorphic reaction pelitic rocks. Mineral assemblages are shown with regular font and reactions are shown in bold. Non-AFM minerals (e.g., plagioclase) are not included.

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<td>Sil Zone</td>
<td>Ms+Qtz=Kfs+Als+H2O</td>
<td></td>
</tr>
<tr>
<td>Opx Zone</td>
<td>Qtz+Kfs+Crd+Sil</td>
<td>Qtz+cal+Kfs+Crd+L</td>
</tr>
<tr>
<td>Spl-Crn Zone</td>
<td>Bt+Sil=Spl+Crd+Kfs+H2O</td>
<td>Quartz-absent rocks at high grades</td>
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</table>

_diagram:_

- Cordierite-Biotite Zone
- Andalusite-K-Ndthropol Zone
- Stilaminie Zone
- Melt Zone
- Orthopyroxene Zone
- Spinel-Conchondite Zone
Fig 2 Phase relations and metamorphic reaction for pelitic rocks of the Kalaybar aureole. SFM diagram is used for quartz-besent rocks.

![SFM diagram for quartz-besent rocks](image)

Fig 3 Back scattered images from calc-silicates of the kalaybar aureole. a) Minerals in sample RH64, garnet (Grt), clinopyroxene (Cpx) and clacite (Cal). b) Minerals in sample RH42, Garnet (Grt) and anorthite (An).

Cal+Qtz+Cpx
Cal+Qtz+Cpx+Pl
Cal+Qtz+Cpx+Grt
Cal+Qtz±Grt+Scp
Cal+Qtz+Cpx±Grt+Scp
Cal+Qtz±Pl+Cpx+Grt+Scp
Cal+Qtz+Cpx+Scp+Ves+Wo
Cal+Qtz±Pl+Cpx+Grt+Wo+Scp

Table 3 includes the mineral assemblages in the studied calc-silicates. These mineral assemblages define five mineralogical zones in the calc-silicates, which are: (I) epidote-tremolite zone, (II) diopside zone, (III) garnet-scapolite zone, (IV) wollastonite zone and (V) wollastonite vesuvianite zone. Fig. 4 illustrates the mineral parageneses in the calc-silicates from the
kalaybar aureole plotted on the ACM diagram indicate a set of metamorphic reactions, which are summarised in Table 4.

**Table 3** Mineral assemblages in the calc-silicate rocks of the Kalaybar aureole.  
× = Major phase, ○ = Minor phase, ⊙ = Xenolith in the igneous rocks,  
* = Analysed by XRD, • = Studied by SEM.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>G.R.</th>
<th>Dol,Cal</th>
<th>Qtz</th>
<th>Pl</th>
<th>Cpx</th>
<th>Grt</th>
<th>Wo</th>
<th>Scp</th>
<th>Ves</th>
<th>Ep</th>
<th>Tr</th>
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<td>×</td>
<td>×</td>
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<td></td>
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<td>×</td>
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<td>×</td>
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</tr>
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<td></td>
<td>∈</td>
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<td>×</td>
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<td>∈</td>
<td>×</td>
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<td>×</td>
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<td>∈</td>
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<td>O</td>
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<td></td>
<td>∈</td>
<td>×</td>
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</tr>
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<td>O</td>
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</tr>
</tbody>
</table>

**Geothermometry, geobarometry and XCO2 calculations**
In order to estimate the pressure and temperature of contact metamorphism, and calculation of fluid composition (XCO2 and XH2O in the fluid), mineral equilibria approach was applied to the pelitic and calc-silicate rocks of the Kalaybar aureole.

Estimation of pressure using the pelitic rocks: The estimation of pressure of contact metamorphism was carried out in the pelitic rocks using a petrogenetic grid. There are three possible melting reactions for the pelitic rocks, which are labelled as reactions 2, 5 and 6 on the petrogenetic grid presented in Fig. 5. Reaction 2 is proposed by Tuttle and Bowen [19], reaction 5 is studied by Seifert [16] and reaction 6 is suggested by Shaw [17]. These melting reactions intersect two other reaction curves on the grid (reactions 3 and 4), which their position on the P-T grid is calculated by the means of THERMOCALC software [6, 7, 14]. These reactions are confirmed by mineral parageneses and AFM diagrams (Table 2 and Fig. 2). All melting reactions occur in the sillimanite zone of the Kalaybar aureole (marked with small arrows on the Ky = Sil and And = Sil curves on Fig. 5). The positions of aluminosilicate reaction curves are calculated using THERMOCALC. Also position of the curves proposed by Holdaway [4] and Richardson et al. [15] are provided for comparison. Intersect of curves 3, 4, 5 and 6 on the P-T grid of Fig. 5 indicates a pressure of ca. 2.5 –3 kbars.
for the Kalaybar aureole. This pressure is marked with a large arrow on Fig. 5 (pressure is constant during contact metamorphism). Because of errors

Table 4 Metamorphic reactions in calc-silicates of the Kalaybar aureole.

<table>
<thead>
<tr>
<th>Metamorphic grade</th>
<th>Mineralogical Zone</th>
<th>Metamorphic assemblages and reaction</th>
</tr>
</thead>
</table>
| Low              | Epidote-tremolite Zone | Cal+Qtz+Pl+Ep  
|                  |                        | Cal+Qtz+Tr+Ep  
|                  |                        | Chl+Cal+Qtz=Tr+Ep+H2O+CO2  
|                  |                        | Cal(Dol)+Qtz+H2O=Tr+Cal+CO2  |
|                  | Diopside Zone          | Cal+Qtz+Ep+Grt  
|                  |                        | Cal+Qtz+Cpx+Pl  
|                  |                        | Ep+CO2=An+Cal+H2O  
|                  |                        | Tr+Cal+Qtz=Dol+Di+CO2+H2O  |
|                  | Garnet-scapolite Zone  | Cal+Qtz+Cpx+Grt+Scp  
|                  |                        | An+Cal+Qtz+Di=Grt+CO2  
|                  |                        | Cal+An+Qtz=Scp  |
| Medium           | Wollastonite Zone      | Cal+Qtz+Cpx+Pl+Wo+Scp+Grt  
|                  |                        | Grt+Cal+Qtz=Scp+Wo  
|                  |                        | Grt+Qtz=An+Wo  
|                  |                        | Qtz+Scp=Wo+An+CO2  |
| High             | Wollastonite-vaselinianite Zone | Cal+Qtz+Cpx+Ves+Wo+Scp  
|                  |                        | Di+Grt+Cal+H2O=Wo+Ves+CO2  |

in thermodynamic data used for construction of the internally consistent thermodynamic dataset [6], the concluded pressure is subjected to about ±1 kbar uncertainty.

Estimation of X_{CO2} and temperature using calc-silicate rocks. In order to undertake geothermometry and X_{CO2} calculations, multiple equilibrium approach was employed. Sample RH68 from the calc-silicates of the Kalaybar aureole contains the largest number of minerals in textural (and thermodynamical) equilibrium (paragenesis 22). Three or four reactions can be written for the end-members of these minerals (depending on T-X_{CO2} window). The reactions, which are determined using THERMOCALC are as follows:

Cal+Di+Wo+Scp+H2O = Ves+CO2  
Cal+Qtz+Di+Scp+H2O = Ves+CO2  
Cal+Qtz = Wo+CO2  
Qtz+Ves+CO2 = Di+Wo+Scp+H2O  

Assuming unit activity for all end-members, the positions of the equilibrium curves for the above reactions were calculated for three nominal pressures 2, 4 and 6 by THERMOCALC. The results are plotted on T-X_{CO2} diagram in Fig. 6. As it is obvious from this figure, intersection of the equilibrium curves defines the X_{CO2}. The X_{CO2} is about 2.5 and it is almost independent of pressure (i.e. in all three pressures the X_{CO2} is about 2.5). Microprobe
analyses of phases in calc-silicates of Kalaybar aureole are not available, therefore the calculation of the exact activities of the end-members is not possible. Among the phases in sample RH68, clinopyroxene and scapolite may show solid-solution. The positions of the reaction curves were calculated for diopside activities of 0.8 and 0.5 and scapolite activities (meionite) of 0.8 and 0.5 at pressure of 3 kbar.
**Fig 4** Phase relation and metamorphic reaction for calc-silicates of the Kalaybar aureole.

![Phase relation and metamorphic reaction for calc-silicates of the Kalaybar aureole.](image)

**Fig 5** Estimation of pressure of contact metamorphic in the Kalaybar aureole using petrogenetic. The pressure is about 3 kbars.


**Fig. 7** illustrates the results. In both cases ($a_{Di} = 0.8$ and 0.5; $a_{Me} = 0.8$ and 0.5) the $X_{CO2}$ is about 0.2, which is very close to the mole fraction calculated for pure phases (unit activities). The temperature for sample RH68 is 650-700°C. Temperature for sample RH68 is 650-700°C.
Figure 6 Estimation of $X_{CO_2}$ and temperature of metamorphism in wollastonite-vesuvianite zone of the Kalaybar aureole (Sample: RH68). The equilibrium curves for the reactions intersect at $X_{CO_2}$ about 0.25, which is independent of pressure. Assuming a pressure of 2-4 kbars, the temperature for this zone is 600-700°C.
Conclusions and discussion

The pressure of contact metamorphism in the Kalaybar aureole was calculated by means of petrogenetic grid and multiple equilibrium approach. The estimated pressure is ca. 3 kbar. This pressure is equivalent to 9 km depth of burial. The possible error is ±1 kbars, which corresponds to ±3 km.
depth. The maximum temperature estimated here is 700-750°C. The estimated temperature has a local significance because the thermal gradient in a contact aureole is very steep but the calculated pressure has a regional importance indicating that either the thickness of the continental crust in the Kalaybar aureole was about 9km during the emplacement time or there has been a tilting during the intrusion of the pluton i.e. the contact exposed in the northern part of the aureole (where the pressure estimation is done) have been deeper than contacts at the southern part. More studies are required to elucidate this point.

Considering mineral assemblages in the studied rocks, it is very likely that fluids during contact metamorphism were of a binary composition of H2O-CO2. Absence of volumetric amounts of graphite and absence of Cl and F-bearing minerals demonstrate that other fluid species were not present in contact metamorphic processes in the Kalaybar aureole. Occurrence of paragenesis Cal+Qtz+Cpx+Scp+Ves+Wo in the Kalaybar aureole, defines a $X_{CO2}$ about 0.2. In other words the fluid was rich in H2O ($X_{H2O} = 0.8$). This finding has important applications in further studies on nature of contact metamorphic reactions (fluid-present or fluid-absent) and on nature of partial melting processes within the Kalaybar aureole.

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References


