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Study of Fluid Inclusions in Radioactive Mineralized Pegmatites, Dara Area, Northern Eastern Desert, Egypt

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دراسات المكتنفات السائلة للبجماتيت المتمعدن بمنطقة دارا شمال الصحراء الشرقية ، مصر

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Abstract

Numerous pegmatite pockets of unzoned and zoned types are the most important rock types from the radioactive point of view. They occur at the marginal parts and higher topographic level of G.Dara younger granites. Zoned pegmatites are composed of extremely coarse-grained milky quartz core, intermediate zone and wall zone. The alteration zone is found at the contact between quartz core and intermediate zone. It is recorded the highest radioactive values due to their mineral composition, as a result of alteration processes associated with radioactive minerals. Only two alteration zones (P1 and P2) has been studied in this paper.

The late magmatic alteration process (hematization, kaolinization, chloritization and fluortization) of the pegmatite resulted in the formation of chlorite, fluorite, clay minerals and carbonates (calcite) in the alteration zones as mineral assemblage. Opaque minerals are found as pyrite, iron oxyhydroxides and garnet.

Fluid inclusion studies by microthermometry were carried out on authigenic minerals (such as quartz and fluorite) in alteration zones (primary fluid inclusions). This study revealed that, at least two stages of the post-magmatic hydrothermal alteration are involved. The first stage is of high temperature, low saline fluids which characterized with hematization and and/or chloritization resulted from fluid-rock interaction with late magmatic fluids that very probably mixed with external low salinity fluids along brittle structure. The second stage is of low temperature high saline fluids characterized the fluoritized alteration due to consequent reaction with wallrocks and mixing with meteoric water.

Mixing of low salinity meteoric water with hot ascending saline hydrothermal solution leads to pH change and continuous interaction with wall rock. The change in pH plays the main role in remobilization

and precipitation of many rare metals such as U, Nb, Zr, Th, REEs + Y in minerals assemblage such as euxenite, zircon, thorianite, allanite, bastnesite and Y- fluorite. Heating of slightly saline meteoric water by convection acting on the pegmatites along shallow shear zone causes fluorite and uranium mineralization.

خلاصـــة

تسجل الأنواع العديدة من الجيوب البجماتيتة (النطاقية وغير النطاقية) الموجودة على الأجزاء الخارجية أو المستويات الطوبوغرافية العالية فى الصخور الحديثة بمنطقة دارا اعلى القراءات الإشعاعية . يتكون البجماتيت النطاقى من ثلاث نطاقات (نطـــاق لبـــى ونطـــاق متوسط ونطاق خارجى). يوجد بين النطاق اللبى والنطاق المتوسط نطاق متغيـــر يعطـــى تمعدنات لليورانيوم كنتيجة لوجود معادن إشعاعية مصاحبة لعمليات التغير . وهذا النطـــاق هو الهدف للدراسة فى هذا البحث

نتيجة تأثير عمليات التغير على النطاق المعنى يتكون الكلوريت والفلوريت والكاولين والكربونات (الكالسيت) بالإضافة إلى بعض المعادن العاتمة او القاتمة مثل البيريت والهيماتيت والجارنت.

أثبتت دراسة المكتنفات السائلة على معدنى الكوارتز والكلوريت بنطاق التغير آن هناك مرحلتين على الأقل من التغيرات فى مراحل المجما المتأخرة :الأولى كانت بفعل محاليل هيدروثرمالية ذات درجة حرارة عالية وملوحة قليلة وتتميز بتشكل الكاولينيت والكلوريت وتنتج هذه المحاليل من تفاعل سوائل الصخور مع سوائل المجما المتأخرة التى عادة ما تكون مختلطة مع سوائل قليلة الملوحة حول الكسور الضعيفة . النوع الثانى بفعل محاليل ذات درجة حرارة قليلة وملوحة عالية وتتميز بتكوين الفلوريت.

إن اختلاط السوائل الصاعدة الحارة مع مياه الأمطار يؤدى إلى تغير فى الأس الهيدروجينى الذى يلعب دورا هاما فى إعادة توزع وتوضع العديد من العناصر مثل اليورانيوم والثوريوم والزركونيوم و العناصر الأرضية النادرة بالإضافة إلى الفلوريت فى المعادن مثل الاوكزينيت والزيركون والثوريانيت والالنيت والبستنزيت والايتريوم فلوريت.

Introduction

Gabal Dara area is bounded by latitudes 27° 52`and 28° 00` N and longitudes 32° 52` and 33° 03` E, covering an area of about 185 km^2. The investigated area is mostly covered by granitoid rocks forming high mountainous terrain reaching up to 1077m a.s.l (Fig.1).

Younger granites form the prominent mass of G. Dara. The marginal parts of G.Dara are characterized by the presence of numerous pegmatitic bodies mainly of unzoned type, while zoned pegmatites are located at the higher parts. Pegmatites are strongly related to the younger granites, but some of them are recorded invading the older granitoids especially on the peripheral zones of the granitic masses. The pegmatites occur as pockets lenses or small veins, sometimes circular and pod like. They are of limited dimension not exceeding 40 m^2. The pegmatites are abundantly encountered at the northern and eastern parts of G. Dara granitic mass and at different topographic levels.

Abdel Monem et al. (1988) stated that the younger granites forming the mass of Dara pluton could be regarded as representing a uniform radiometric unit with respect to their absolute potassium and equivalent uranium contents [2]. The structural conditions played an important role in the localization of the identified radiometric anomalies which was found closely related to the faulting pattern in the area [3].

Uranium and thorium are the main radioactive elements in the pegmatite bodies. Their concentration being a result of post magmatic processes. The minerals identified in the investigated pegmatites by XRD technique can be divided into non-radioactive and radioactive types, grouped in three main classes [4]:

(1)- Non-radioactive minerals such as haematite, goethite, pyrite, rutile and calcite

(2)- Radioactive minerals such as, euxenite, carnotite, curite, zircon and thorianite

(3)- Rare-earth radioactive minerals such as bastnaesite, allanite and associated fluorite.

The present study is concerned with the fluid inclusions of two-alteration zone of pegmatites of the G.Dara area, in order to estimate the physicochemical conditions of alteration processes.

Distribution of U and Th in the zoned Pegmatites

Concentration of the total soluble uranium ions in the samples was estimated using laser induced fluorescence technique following the scheme of [5]. Thorium was colourimetrically estimated using arzenazo III method following the scheme of [6].

Uranium and thorium distributions in the zoned pegmatites are studied in the present work in three zones named, wall zone, intermediate zone and altered zone (Table 1). It is observed that there is a systematic pattern for U and Th distributions within the different zones (Fig. 2). It is clear from this figure, that U and Th decrease from crosscut secondary altered zones toward wall zones. In the wall zone, the average of U content is 56.5 ppm, while Th average content is 47 ppm. These values reflect primary sites for the two radioactive elements where zircon, allanite and apatite are the main accessory minerals in this zone.

The intermediate zone which is in crosscut with altered zone, represents the most suitable host zone for radio-elements with progressively fewer occurrences noted in the secondary alteration zone. The average of both U and Th are 143.5 ppm and 105.5 ppm respectively. The high U and Th content is due to primary magmatic radioactive minerals and secondary alteration products due to fractionation, hydrothermal activity and weathering.

Secondary alteration zone represents the most populer host for the radioelement. Uranium average content in two secondary alteration zones is 233 ppm while Th average content is 205 ppm. The enrichment in both radioelements exhibits extensive replacement of primary mineral phases by secondary phases produced during late-stage hydrothermal activity that have been accentuated through microfracturing in the host granite. Secondary products include chlorite-replacing biotite, ferruginous clays replacing biotite and calcic plagioclase, and also hematitization as a result of chlorite alteration to carbonate and clay minerals. This alteration has increased the radio- element contents [7].

Fluid inclusion

Fluid inclusion provides geologist with what can only be observed as unique samples of the total spectrum of fluids that have interacted with an developed in the earth's crust and upper mantle throughout the whole of the geologic time. Historically, their most important contribution has been to understand the character, origin and evolution of the hydrothermal ore forming fluids and ultimately, ore genesis [7-9].

Fluid inclusions in ores and gangue minerals can be used to fingerprint certain type of ore forming fluids, to characterize particular ore mineral assemblages and to define areas where these fluids are most likely to concentrate [10-11].

In igneous rocks, fluid inclusions are especially common in the quartz of granitic rocks. The fluids trapped in granite quartz can either represent the earliest fluids from the cooling magma or later fluids developed during hydrothermal circulation. Both types of fluids developed may be trapped in apparently unaltered samples of rock [12] and it can prove exceedingly difficult to find a sample where the inclusion population only represents the magmatic fluid phase.

Methodology

Representative samples of two alteration zones (P1 and P2) in the studies pegmatite were chosen for the fluid inclusion study. In this study microthermometric measurements on authigenic minerals formed during alteration are carried out in order to estimate the physico-chemical conditions of alteration. Fluorite and quartz in (P_2) alteration zone and quartz of (P_1) alteration zone represent the studied minerals.

Double polished thin sections of selected samples (250-300μm) were prepared. Freezing/heating microthermometric measurements were carried out using a fluid inclusion adopted to USGS gas flow heating/freezing stage emanated to petrographic microscope, at Geol. Dept. Faculty of Science, Cairo University. Homogenization temperature (Th) is the temperature at which gas bubble can not be observed. All the fluids in the studied sample homogenize to liquid phase. Freezing run to -90^0 C followed by heating one enable us to record melting temperature (Te). The final ice melting temperature (Ti) enables calculations of fluid inclusion salinities. Table (2) shows Th, Ti, Te and salinity values of the two alteration zones of mineralized pegmatites (P1 and P2). Figure (3) shows the Th°C versus salinity for fluid inclusions in the studied samples.

Fluid inclusion petrography

Fluid inclusions are abundant in quartz and fluorite samples. A small number of fluid inclusions are trapped during the growth of surrounding host crystals and occur as isolated groups, sometimes confined within growth zone of quartz and fluorite crystals and these assumed to be primary. A great number of fluid inclusions is related to fractures and/or cleavage planes, and therefore, these inclusions were assumed to be secondary. Inclusions shape may vary from negative crystals to subspherical or irregular branched shapes (obviously necked down) and dimensions range from 10 to 20 mm. Primary fluid inclusions in quartz and fluorite minerals are two-phase, water-rich at room temperature, with constant liquid/ vapor ratio at~95:5. No microthermometric evidences of CO_2 have been found in any fluid inclusions described above. The results presented below are referred to the primary fluid inclusions (Fig. 4).

Results

Small size of most quartz inclusions is typical for low salinity fluid inclusions. However there is great range between the melting temperature (Te) values for fluorite and quartz. Te values in quartz of the both alteration zones range from –32.4°C to -44.1 °C coinciding with Mg, Na chlorite water system. Te values of fluorite in the second alteration zones (P2) range from –49.2°C to –59.1 °C which are much closer to eutectic for chlorite water system containing bivalent ions (notably Ca, Mg) than those containing only NaCl and / or KCl. The Te below –54.4 °C of pure system H_2O- CaCl- NaCl may be explained metastability [13]. The last ice melting temperature (Ti) shows different population. It ranges from –0.80 °C to –3.1 °C for quartz samples strongly indicate low salinity. Ti for fluorite sample ranges from –16.4 °C to –18.0 °C suggesting high salinity of the forming hydrothermal solution. Total salinities deduced from these temperature ranges from 1.4 to 3.9 Eq. Wt % NaCl (i –e the amount of NaCl which would produce an equivalent lowering) for quartz of both alteration zones and from 14.3 to 15.9 eq. Wt. % NaCl for fluorite of the second zones. All investigated inclusions homogenize into liquid state during heating runs, at homogenization temperature (Th) which represent minimum trapping temperature. Most of homogenization temperature (Th) range from 100 °C to 240 °C. These values of Th for quartz samples range from 240 °C to 185 °C while for fluorite samples it ranges from 104 °C to 135 °C

Discussion

The ascending hot fluids, after being acted on pre-enriched granitic volume, were alkaline and oxidizing when reached the shallow shear zone and caused intensive subsolidus alterations. The evidences of the alkaline nature of the hot-saline fluids are:

1- Precipitation of iron as (hydro) oxides (i.e. hematization) in (P_2).
2- Chloritization of primary silica in (P_2).
3- Metamictization of stable accessory minerals such as zircon and allanite in (P_2) and thorianite in (P_1).
4- Presence of both carnotite and euxenite in (P_2). Elevatorski (1978) suggested that, carnotite is associated with all types of hydrothermal solutions, but its colour change according to the type of hydrothermal solution. In strong acidic hydrothermal fluids (pH<4), carnotite occurs as yellowish brown colour, but in strong alkaline hydrothermal fluids (pH>10) occurs as cannary yellow colour. In the studied area, carnotite is cannary yellow colour [4] suggesting strong alkaline hydrothermal solution.
5- Alteration of the allanite by the effect of the alkaline hydrothermal fluids according to equation given by [15-16] as below

$$\text{allanite+ alkaline fluids} \longrightarrow \text{bastenasite + clay minerals + thorianite+ hematite}$$

The physico chemical conditions of these fluids were ever changing due to consequent reaction with wallrocks and mixing with meteoric water which leads to low temperature, high saline fluids. Accordingly, the hydrothermal fluids should have high saline fluids and should have developed a low pH. These conditions lead to reduction of iron oxide and formation of pyrite and most probably motivated the reduction and fixation of uranium from mineralized hydrothermal solutions.

$$2UO_2\ (CO_3)^{2-}{}_2 + 4FeS_2 + 2H_2O + 7O_2 = 2UO_2 + 2H_2CO_3 + 2Fe_2O_3 + 4SO_3^{2-}$$

Rapid cooling or dilution with colder water will cause the solution to become reactive with respect to hydrolytic alteration, boiling well tender them inert with respect to this process will cause deposition of additional calcite, at more shallow depth chlorite and kaoline minerals were deposited. This alteration assemblage is enclosed by rocks affected to varying degrees of alteration caused by interaction with descending recharge of essentially meteoric origin.

Accepting that the removal of solutes and vapor species from the cooling intrusion is a dynamic process. With the respective alteration zones following the advancing front of brittle fracturing to more deeper levels, it is unlikely that the alteration patterns will be preserved in such simple form of actual ore bodies. In addition to the "stretching out" of alteration zones to progressively deeper levels, overprinting and remobilization of components, in response to repeated episodes of intrusive activity and cooling [17-20], can be expected to lead to the rearrangement of alteration zones to patterns much more complex than the monogenetic ones.

Conclusions

The studied area is bounded by latitudes 27° 52` and 28° 00` N and longitudes 32° 52′ and 33° 03′ E, covering an area of about 185 km^2.

The investigated area is mostly covered by granitoid rocks forming high mountainous terrain reaching up to 1077m a.s.l.

Regarding to U and Th evaluation in the study area, pegmatites are the most important types. Zoned pegmatite records the highest radioactive level than unzoned pegmatite. Within the zoned pegmatites, it was noticed that U content decreases from inner zones toward outer zones. The alteration zones recorded the highest U and Th contents due to their mineral composition, as a result of alteration processes (hematization, kaolinitization, chloritization and fluortization), associated with radioactive minerals (zircon, euxenite, and thorianite), and rare earth radioactive minerals as (allanite, bastnaesite and fluorite).

Fluid inclusion studies of the investigated quartz and fluorite were conducted to elucidate the chemical characters of the hydrothermal solutions. They also allow us to depict the hydrothermal activities, acting on the pegmatites and their hosting Dara granitic pluton. It is possible to define two different acting solutions. The first one is marked by low to medium salinity, of hot circulating solutions acting under different physico-chemical conditions, while the second is marked by high salinity low temperature. The quartz seems to be linked to an early independent hydrothermal event during a reactivation on fracture system, which caused circulation of low salinity hot fluids. Since alteration zones are related to tectonic, variable fluid flux together with change in their physico – chemical potential of the fluid yield fluorite crystallization with variable salinity and temperature. The change in pH plays the main role in remobilization and precipitation of many rare metals such as U, Nb, Zr, Th, REE + Y in minerals assemblage such as euxenite, zircon, thorianite, allanite, bastnesite and Y- fluorite.

Mixing of low salinity meteoric water with hot ascending saline hydrothermal solution leads to pH change and continuous interaction with wall rock. Therefore the mechanism of fluorite precipitation is accompanied with a rather complicated series of alteration processes such as hematitizationn, fluoritization, silicification and kaolinization. Heating of slightly saline meteoric water by convection acting on the pegmatites along shallow shear zone causes fluorite and uranium mineralization.

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Table 1: U, Th and Th/U for the different zones of pegmatites

	Wall zones			Intermediate zones			Alteration zones		
	W1	W2	Average	I1	I2	Average	P1	P2	Average
U	47	66	56.5	120	167	143.5	266	200	233
Th	44	50	47	111	100	105.5	250	160	205
Th/U	0.94	0.76	0.85	0.92	0.60	0.76	0.90	0.80	0.85

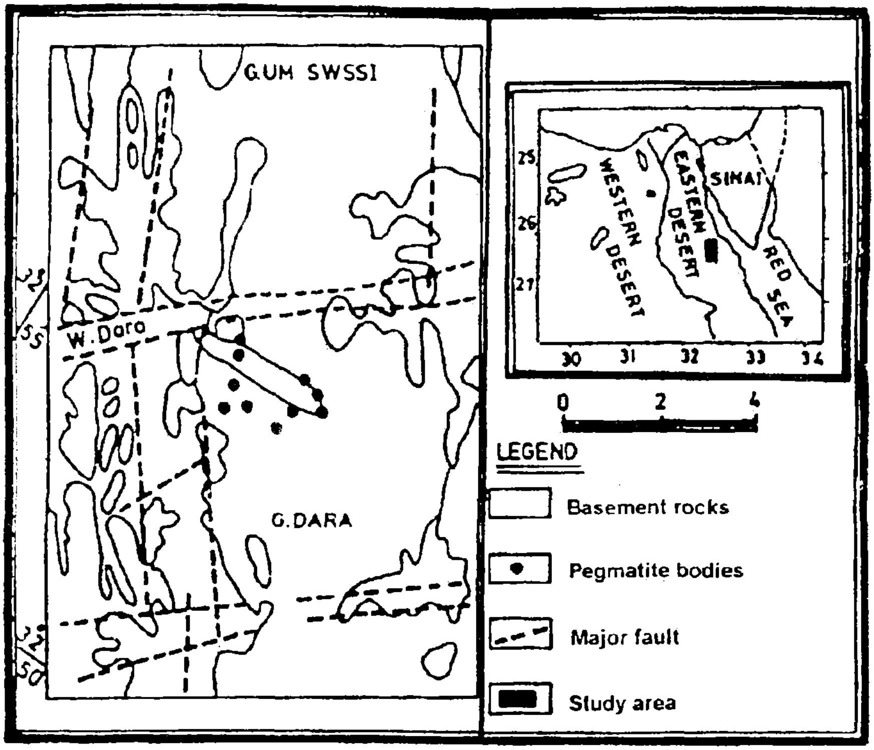

Fig. 1: Geologic map of G. Dara area, Northern Eastern Desert, Egypt.

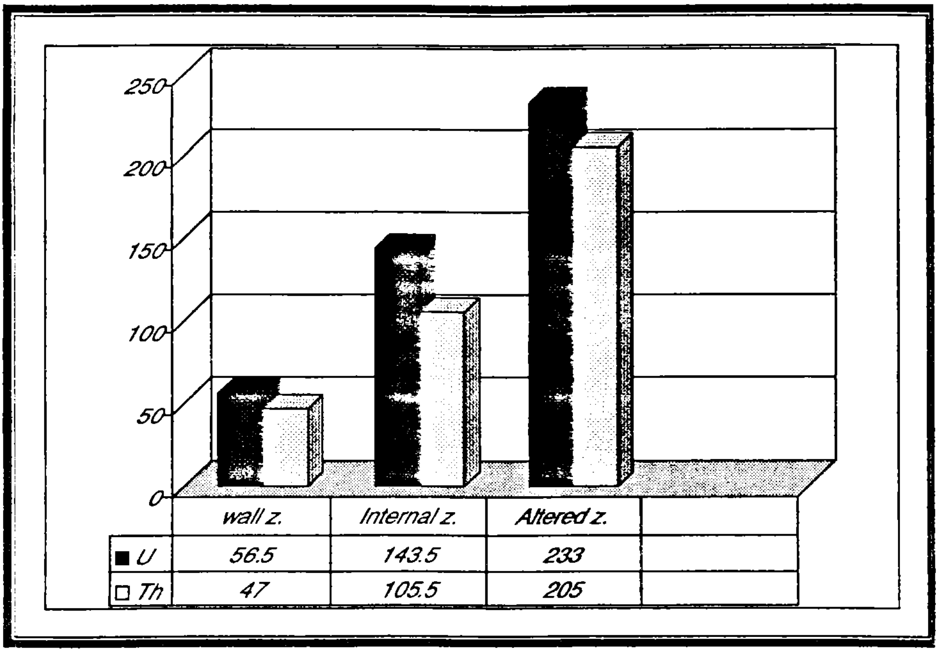

Fig. 2: Average of U and Th distribution in different zones of pegmatites

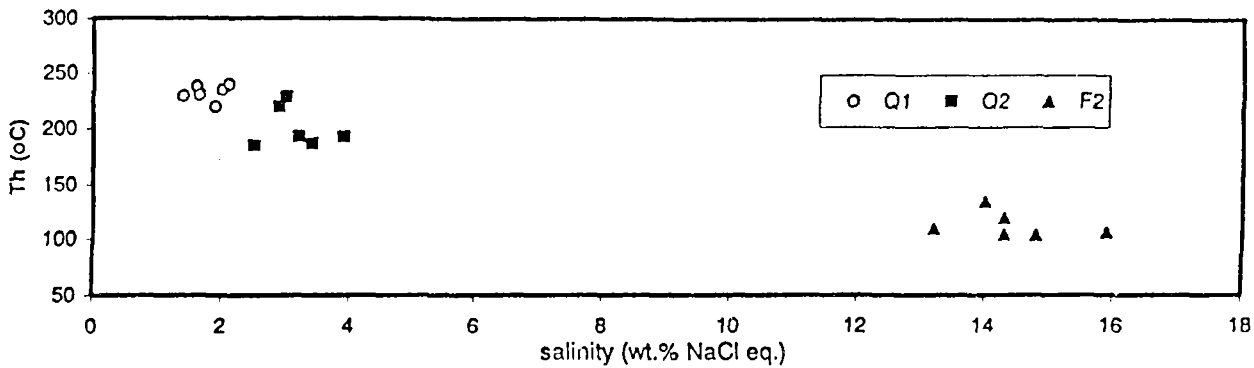

Fig. 3: Th (°c) Vs. salinity for fluid inclusions in the studied samples. circles: samples from quartz of PI, squares: samples from quartz of P2, triangles: samples from fluorite of P2

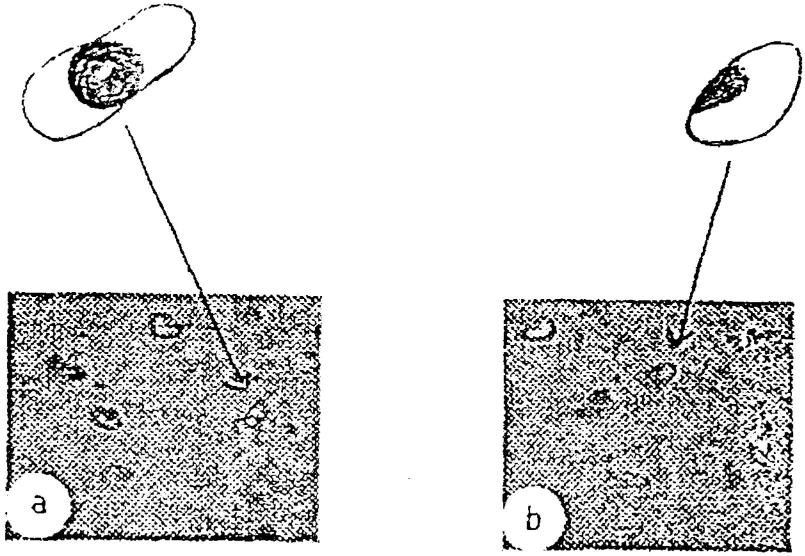

Fig. 4: Photomicrographs and sketch view of fluid inclusions in quartz.

(a) : quartz from first alteration zone (P1)

(b) : quartz from second alteration zone (P2)

Table (2) : Homogenization temperature (Th), melting temperature (Te), Ice temperature (Ti) and salinity values of the fluid inclusion for two alteration zones of mineralized pegmatites(P1 and P2)

	1						2						3					
Th	238.1	229.7	235.5	230.7	240	220	143.1	185	192.9	220.4	187	229	106.6	105	110	135.2	120	104
Ti	-0.8	-0.9	-1.1	-0.8	-1.3	-1.0	-2.8	-2.7	-3.1	-2.4	-2.6	-2.5	-18.4	-17.7	-16.4	-16.9	-17.7	-18
Te	-36.4	-35.8	-38.4	-32.4	-33.7	-34.2	-37.9	-38.7	-43.2	-35.3	-40.1	-44.1	-59.1	-49.2	-55.1	-56.1	-57.3	-58.1
Salinity Eq. Nacl%	1.6	1.4	2.0	1.6	2.1	1.9	3.2	2.5	3.9	2.9	3.4	3.0	15.9	14.3	13.2	14.0	14.3	14.8

1-Quartz of first alteration zones (P1),

2-Quartz of first alteration zones (P2),

3-Fluorite of second alteration zones (P2)

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