CHEMISTRY OF BIOTITE AS A GUIDE TO THE NATURE OF MAGMAS, HAJJA GRANITOID COMPLEX. YEMEN REPUBLIC

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ABSTRACT

Biotites separated from Hajja granatic rocks, Yemen Republic, have been examined. The chemical data of 14 analysed biotites show that the biotites are ferrous iron varieties. The behaviour of major elements in the examined biotites discussed according to different variation diagrams and elemental ratios. The significance of $Fe^{(t)}$ ($Fe^{(t)}$+Mg) ratio as a relative measure of biotite crystallization suggests that the biotites may be formed under a temperature range of 815°–950°C. In the present study since Fe shows little variation with Al in biotites in both calc-alkaline and peralumios magmas, the substitution $2Al \rightarrow 3Fe^{2+}$ does not play an important role during the crystallization of biotites in these two types. On the other hand the substitution $3Mg \rightarrow 2Al$ is vital. The contrasting behaviour of Fe and Mg with respect to Al during the crystallization of biotites is partly governed by the various physicochemical conditions including the behaviour of volatiles in these magmas.

INTRODUCTION

Biotite is an important ferromagnesian mineral in most intermediate and felsic igneous rocks. Its potential to reveal both the nature and the physicochemical conditions of source magmas from which it formed is high. Igneous biotite can also be used to provide valuable petrogenetic information.

Althought numerous studies have been carried out on biotites from individual igneous complexes and granitoids in particular, very little work has been done to characterize biotite in the spectrum of igneous rocks or to find a possible tectonic linkage of biotite compositions to magma types from various sources and distinct petrogenetic histories.

Recently ABDEL-RAHMAN (1994) succesfully introduced several discrimination diagrams based on major-element composition of biotites in igneous rocks world-wide crystallized from three distinct magma types. He defined 3 magma suites, namely: 1. Alkaline complexes (A), 2. Peraluminous suites (P), 3. Calc-Alkaline complexes (C).

The present work is the first attempt to apply ABDEL-RAHMAN'S (op. cit.) scheme on biotites from Hajja granatic complex, Yemen Republic.

In a previous study the petrochemical characters and chemical classification of Hajja granitoid rocks were studied by SALEM et al. (1986).

The granatic rocks of Hajja *(Fig. 1)* comprise two field types represented by greyish white and pinkish granites. They are medium to coarse grained and may be porphyritic, composed essentially of quartz, alkali feldspar, plagioclase, biotite, in addition to fair amount of muscovite; iron oxide minerals epidote, sphene; magnetite and zircon are the common accessories. Biotite occurs as the dominant mafic mineral, it forms subhedral

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crystals ranging from 0.9 mm to 2.6 mm in length and from 0.3 mm to 1 mm in breadth. Biotite forms stout flakes with torn ends, sometimes exhibiting sub-parallel arrangement. It often displays pleochroism with X=straw yellow and Y=Z=dark brown.
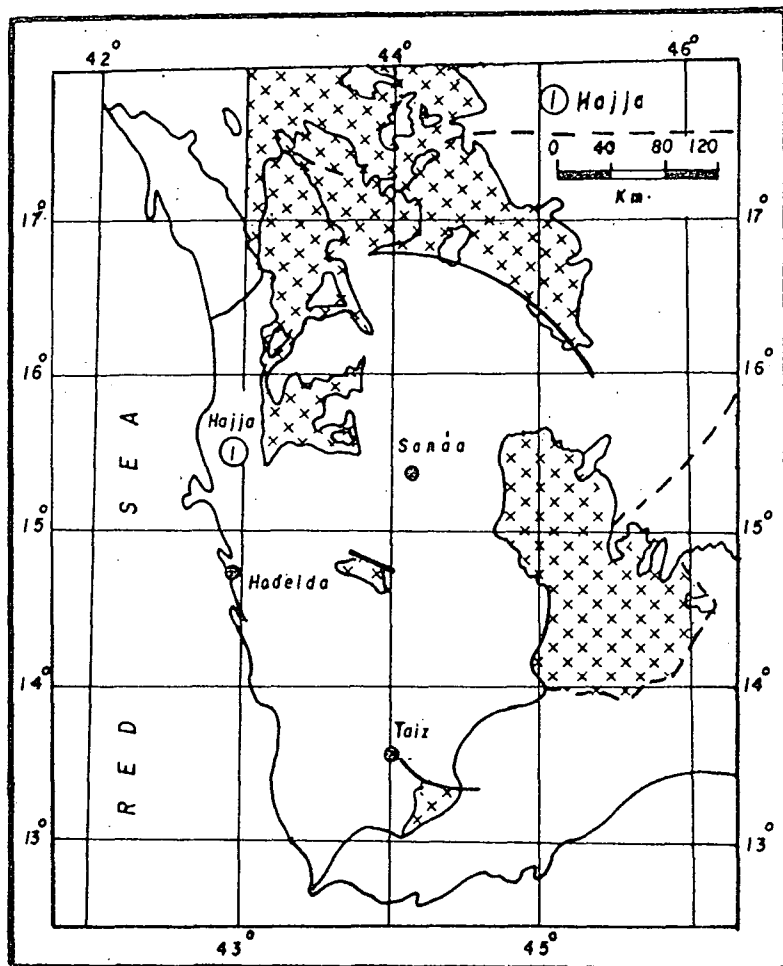

Fig. 1. Map showing distribution of the basement complex in the Yemen Arab Republic and Location of Hajja.

COMPOSITION OF BIOTITES

The chemical analyses[*] of the recovered biotites are given in Table 1 with some analyses from other localities for comparison. The chemical data are used in different

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TABLE 1

Chemical analyses of biotites from Hajja granitoids

Samples No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	A	B	C
SiO_2	37.40	38.27	37.92	38.74	39.79	38.78	38.77	35.85	34.75	37.64	37.70	38.40	37.25	38.90	35.40	35.99	36.90
Al_2O_3	14.30	13.18	13.67	13.33	11.81	13.53	13.47	16.60	15.85	16.42	15.90	15.66	16.88	16.32	16.66	14.94	115.40
Fe_2O_3	5.74	9.20	9.39	7.92	10.27	9.48	7.97	6.10	4.83	2.15	1.90	12.73	8.25	12.24	5.17	6.11	3.90
FeO	12.10	7.23	8.32	9.18	4.19	6.92	9.29	13.84	16.32	18.60	18.90	13.53	15.72	13.34	21.61	16.91	16.60
MnO	0.65	0.82	0.56	0.65	0.95	0.63	0.72	–	–	–	–	0.28	0.40	0.28	0.45	0.14	0.52
MgO	12.65	14.36	13.93	13.58	16.96	14.91	13.91	10.64	11.44	10.22	10.29	6.25	6.84	5.91	6.23	12.90	10.90
CaO	–	–	0.16	–	–	–	0.30	–	–	0.52	0.49	1.23	1.29	1.41	1.40	0.14	0.22
Na_2O	–	0.35	0.41	0.40	–	–	–	0.94	1.13	–	–	0.13	0.18	Trace	0.34	0.12	0.04
K_2O	9.53	9.95	9.25	9.59	10.04	10.00	9.66	10.13	9.54	9.48	8.89	6.78	7.13	6.78	6.34	7.81	9.10
TiO_2	4.34	3.39	2.94	3.25	2.35	2.38	2.58	2.90	2.61	2.51	2.72	2.52	3.42	2.40	2.18	2.73	2.90
H_2O	–	–	–	–	–	–	–	–	–	–	–	2.06	2.24	1.99	2.83	2.50	3.10
Total	96.79	90.75	96.52	96.95	96.35	96.60	96.67	96.99	96.47	96.99	96.81	99.60	99.60	99.57	98.99	99.43	98.98

– = Not detected
A = Biotite from Kadabora granites (KABESH and SALEM 1981)
B = Biotite from Ras Barud granites (KABESH et al., 1977)
C = Biotite from granodiorite, Sierra Nevada Batholith, California (DODGE et al., 1969)

petrochemical parameters and variation diagrams in order to elucidate the petrochemical and petrogenetic characteristics of the studied biotites.

Nockolds diagram

According to NOCKOLDS (1942) the biotite composition is different for different mineral associations, I-Biotites associated with muscovite and topaz, II-Biotites unaccompanied by other mafic minerals. III-Biotites associated with hornblende, pyroxene, and/or olivene. The chemical data are plotted on NOCKOLDS variation diagram (Fig. 2) which shows the fields (I, II, III); it is evident that the analysed biotites fall within field II of biotites unaccompanied by other mafic minerals.
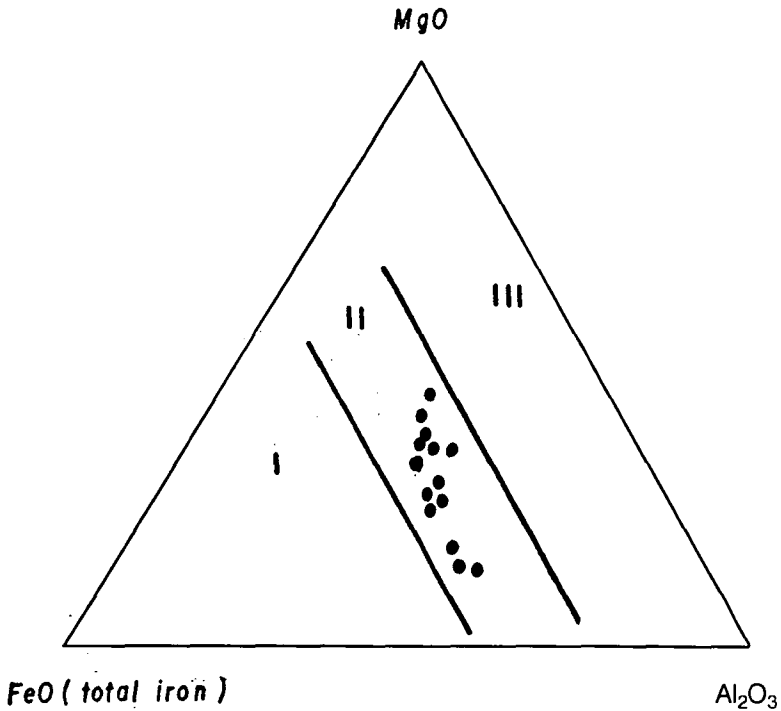
Fig. 2. Plots of biotites on the ternary diagram of NOCKOLDS (1947)
I – Biotites associated with muscovite, topaz etc. II – Biotites unaccompanied by other mafic minerals.
III – Biotities associated with hornblende, pyroxene and or olivine.

Heinrich variation diagram

The values $Fe_2O_3+TiO_2-MgO-FeO+MnO$ are taken into account. *Fig. 3*, it is evident that the examined biotites share the fields of magmatic and metamorphic metasomatic, which indicate that the Hajja granatic mass might have suffered some postmagmatic metasomatic processes.
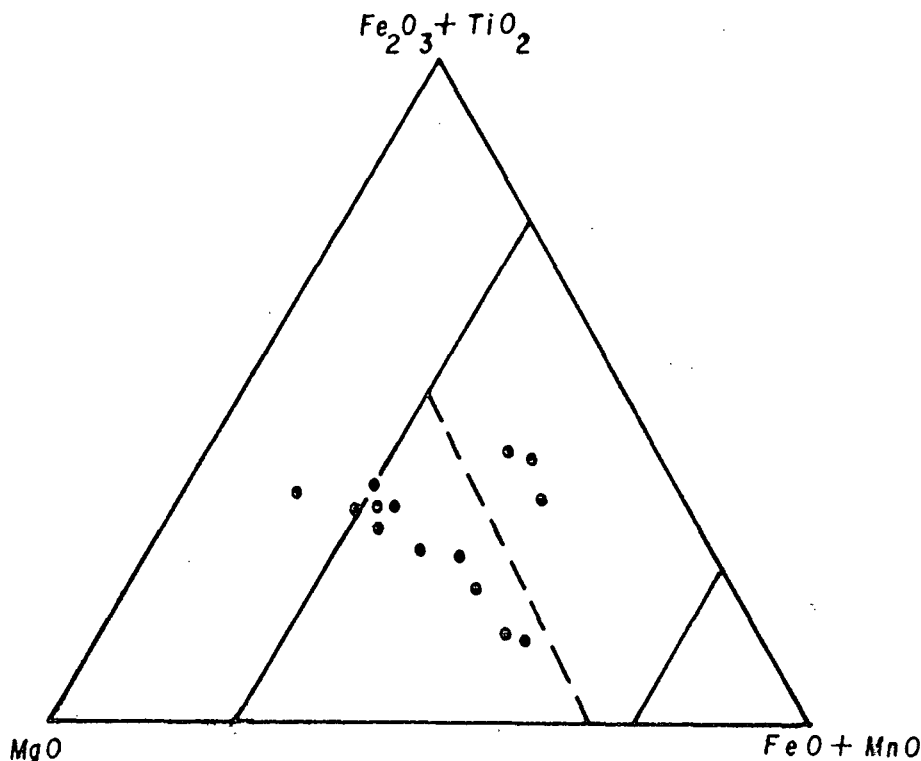
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Fig. 3. Heinrich variation diagram

STRUCTURAL FORMULAE OF BIOTITES

The chemical analyses (Table 2) are recalculated on the basis of 23 (O), (DEER et al., 1966), and the results are given in (Table 2) which gives the following picture: The coordinated large cations Ca, Na, K of X group have values reanging from 1,55–2,28, in all cases K cation constitutes over 75% of the total X values, while both Na, Ca contents in the biotites are low and sometimes are not present. The wide range of X values is due to the wide range of the alkalinity of the host granatic rocks. The Y group cations consist mainly of iron and magnesium, the relation between cations of the Y group is shown in *Fig. 4*. It is obvious that the studied biotites are magensio-ferrous rich varieties, the composition ranges from Mg biotites to Fe+2 biotites.

Experimental work by WONES and EUGSTER (1965) demonstrate the effect of oxygen fugacity on the formation and resultant composition of biotites, if it is assumed that substitution other than Fe → Mg will not materially influence biotite stability. Application of the experimental data to the studied biotites permits an evaluation of the variation during the course of crystallization of Hajja granitic magma. WONES and EUGSTER (1965) considered two contrastive trends of crystallization of biotites. Trend I representes a magma in which oxygen fugacity remains constant or somewhat increases whereas in trend II oxygen fugacity decreases.
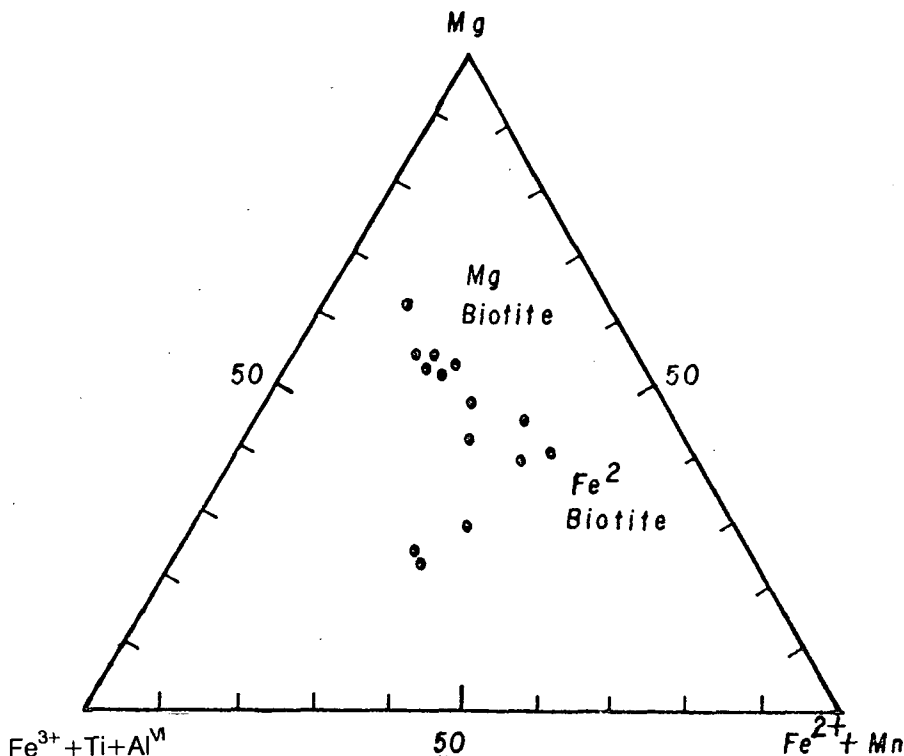
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Fig. 4. Relation between octahedral cations of the investigated biotites

The trend of the Hajja biotites within the compositional triangle Fe^{3+}, Fe^{2+}, Mg (*Fig. 5*) rather closely paralells WONES and EUGSTER'S (op. cit.) estamited composition of biotites solid solution in the ternary system KFe_3^{3+} $AlSi_3$ $O_{12}OH–KFe^{2+}_3$ $AlSiO_3O_{10}$ $(OH)_2–KMg_3$ $AlSi_3O_{10}$ $(OH)_2$, which are stable at oxygen fugacities controlled by individual buffer equilibria. The correlation suggests the composition of the examined biotites as defined by oxygen fugacities greater than those of the Ni–NiO buffer thus, the biotite composition suggests oxygen fugacities for the biotites of Hajja area as largely following a buffer curve during crystallization i.e. oxygen fugacities decreased with decreasing temperature. All the biotites analyzed belong to the assemblage potash feldspar – magnetite biotite, the buffering effect of magnetite was recognized in controlling oxygen fugacities in the crystallizing magma.

KENNEDY (1955) and OSBORN (1962) have suggested that the calc-alkaline or „Bowen trend" of late silica enrichment in contrast to the „Fenner trend" of late iron enrichment is caused by a relatively high constant oxygen fugacity during crystallization. However, because oxygen fugacity will decrease as temperature decreases i.e. was defined by buffering reaction, trend II of WONES and EUGSTER (1965).

The plotting of 100 Fe (total)/(Fc total+Mg) ratio of the studied biotites on the stability diagram (*Fig. 6*) according to EUGSTER (1965) reveals that these ratios of the analyzed biotites range between 32–69.96 which correspond to temperature about 815°C–950°C and oxygen frugacity of about $10^{-14}–10^{-8}$.

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TABLE 2

Structural Formulae of Biotites

| Formula No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Si	5.75	5.8	5.77	5.90	5.98	5.87	5.90	5.55	5.38	5.77	5.77	5.90	5.73	5.96
Al^{IV}	2.25	2.2	2.23	2.09	2.02	2.13	2.10	2.45	2.62	2.32	2.23	2.10	2.27	2.04
Z (sum)	8	8	8	8	7.99	8	8	8	8	8	8	8	8	8
Al^{VI}	0.34	.16	0.22	0.31	0.07	0.27	0.31	0.58	0.27	0.74	0.54	0.74	0.79	0.91
Fe^{3+}	0.75	1.17	1.20	1.00	1.29	1.20	1.02	0.80	0.64	0.28	0.25	1.47	0.95	1.41
Fe^{2+}	1.55	0.92	1.06	1.15	0.53	0.88	1.18	1.82	2.15	2.36	2.40	1.74	2.02	1.71
Mn	0.08	0.10	.07	.08	0.12	0.08	0.09	–	–	–	–	.04	0.05	.04
Mg	2.90	3.24	3.16	3.09	3.80	3.86	3.16	2.46	2.64	2.34	2.35	1.44	1.57	1.34
Ti	0.51	0.39	0.34	0.37	0.27	0.27	0.30	0.34	0.30	0.29	0.31	0.29	0.39	0.28
Y (sum)	6.13	5.99	6.04	5.99	6.08	6	6	6	6	6.01	5.85	5.72	5.77	5.69
Ca	–	–	0.03	–	–	–	0.31	0.58	0.27	0.74	0.54	0.74	0.79	0.91
Na	–	0.11	0.12	0.37	–	1.20	1.02	0.80	0.64	0.28	0.25	1.47	0.95	1.41
K	1.89	1.92	1.80	1.87	1.93	0.27	0.30	0.34	0.30	0.29	0.31	0.29	0.39	0.28
X (sum)	1.89	2.03	1.94	2.24	1.93									
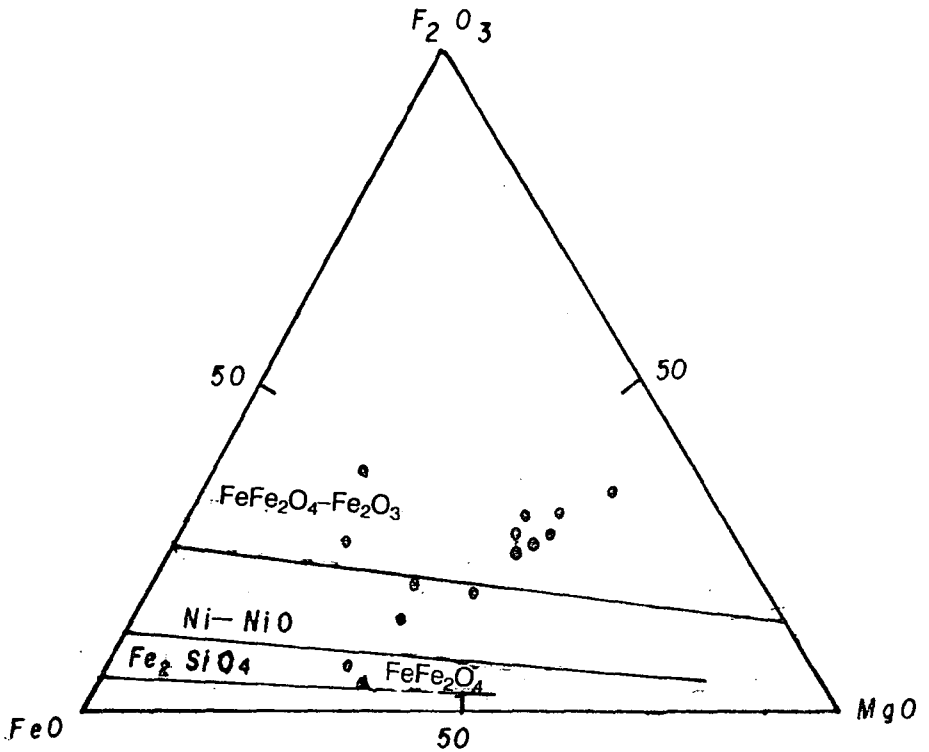
Fig. 5. Relation between Fe^{3+}, Fe^{2+} and Mg in the investigated biotites

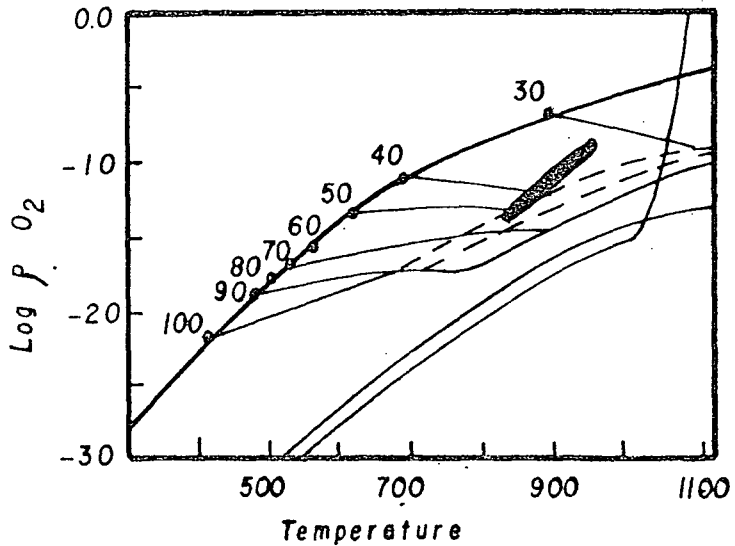*Fig. 6.* Biotite stability diagram of specific 100 Fe/(Fe+Mg) values as a function of oxygen fugacity and temperature at 20, 70 bars total pressure (after WONES and EUGSTER 1965). Heavily shaded area represents the investigated biotites.

Chemistry and crystallization conditions of the granitic magma essentially affect the composition of the biotites derived from this magma.

Al content plays an important role in the alkalinity of the magma and could be used as a factor controlling alkalinity, acidity and as an indicator for pressure prevailing during the process of crystallization.
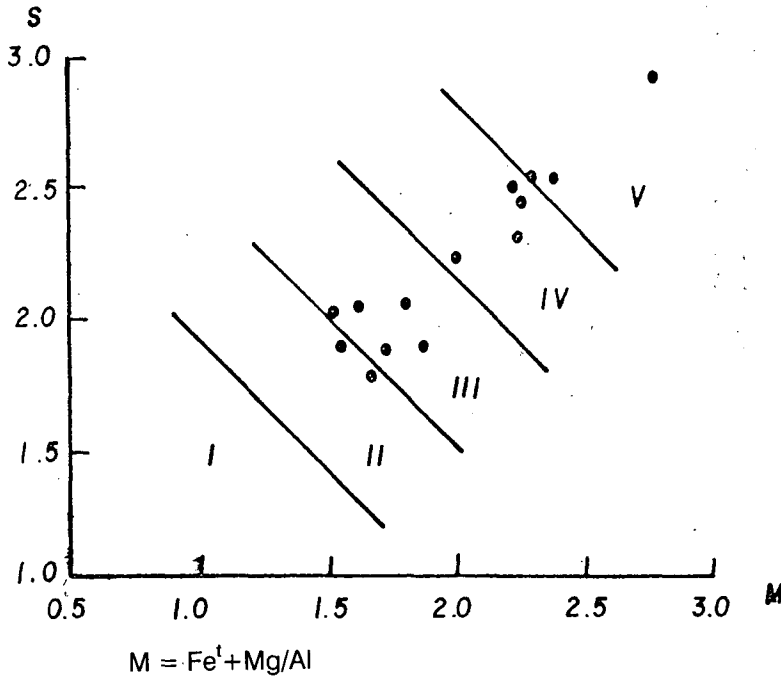
Fig. 7. Acidity-Alkalinity diagram of Hajja granitoids with biotite composition

On the diagram alkalinity–aluminosity of biotites (*Fig. 7*) the values Si/Al vs. (Mg+Fe)/Al was suggested by MARACHUCHIEV (1966). He divided the fields of crystallization of the biotites host rocks into five fields in order of increasing alkalinity:

I. Calcic magma, II. Calc alkaline magma,
III. Subalkaline magma, IV. Alkaline magma,
V. Peralkaline magma.

Plots of the examined biotites fall in the fields of subalkaline to alkaline granite. The above mentioned elements were used also by IVANOV (1970) for revealing the relationship and illustrating alkalinity and temperature of crystallization of biotites and water fugacity of crystallizing liquid. He suggested the following relationship

L=AlX100/Si+Al+Fe+Mg,
F=100FeO$^+$/(FeO$^+$+Mg).

The values of Hajja biotites range from 15.3 to 23 for L) and from 32 to 69.96 for F, these values occupy the fields of moderate to high alkalinity and of high to moderate termpature of crystallization (*Fig. 8*).

$$L = 100 \times AL \; / \; Si + AL + Fe^t + Mg)$$
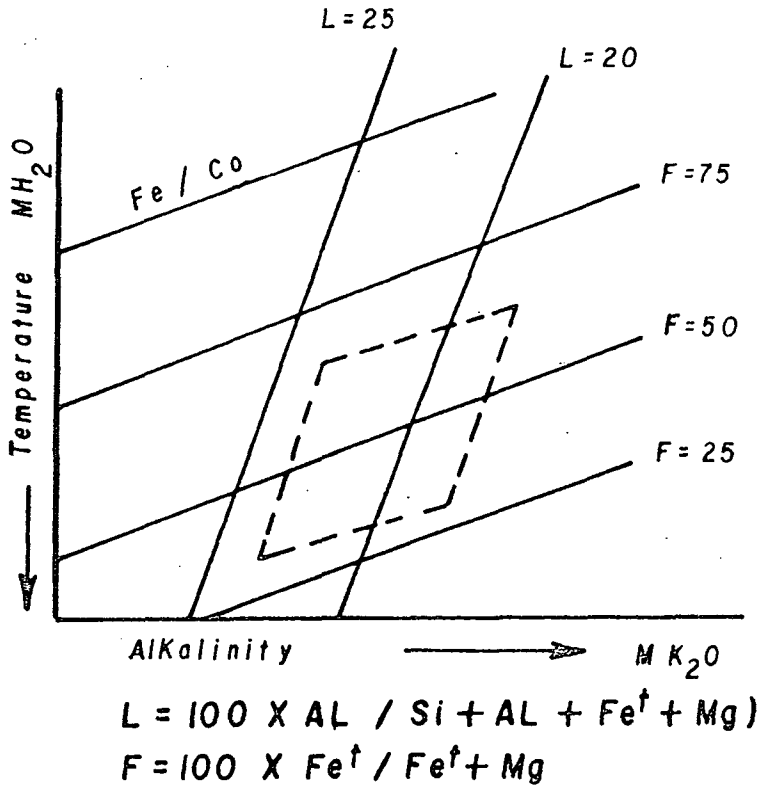$$F = 100 \times Fe^t \; / \; Fe^t + Mg$$

Fig. 8. Relation between water activity and potassium activity in granitoids of Hajja.

According to SMITH (1968) increasing temperature gives possibilities for Al to replace Si in the tetrahedral position while increasing pressure gives favourable condition for this replacement to take place on the octahedral coordination. Diagram (*Fig. 9*) illustrates the relation between Al^{IV} ad Al^{VI} coordinations.

The diagram shows that in the tetrahedral Al is not great from (2.020 to 2.618) while variations in octahedral Al are more or less clear from (0.02 to 0.92), this may indicate that these granites are allochtonous, where the pressure decrease through the magma ascending upwards intruding country rocks of different composition.

Relation of Pleochroism to chemistry

The formulae of biotites have been discussed by HAYAMA (1959, 1964) and ENGEL and ENGEL (1962). HAYAMA (1959) has correlated the change in the absorption colour of biotite to the behaviour of TiO_2 and $Fe_2O_3/(FeO + Fe_2O_3)$ values. He concluded that biotites of high Ti content and lower $Fe^{3+}/(Fe^{+2} + Fe^{+3})$ values are characterized by red brown absorption colour in Y and Z direction while those of lower Ti and higher $Fe^{3+}/Fe^{3+}/$ $(Fe^{+2} + Fe^{+3})$ values show brown absorption colours in Y and Z directions. The examined biotites are strongly pleochroic with X straw yellow and Y=Z dark brown (SALEM et al., 1986).
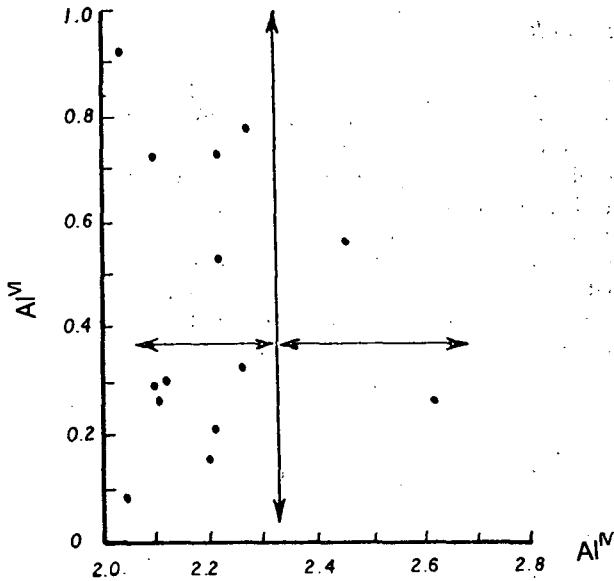
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Fig. 9. Content of Al in tetrahedral and octahedral positions in biotites of Hajja

According to the structural formulae Table 2 of the examined biotites the Ti/Fe^{+2} ratios are low whereas the ratios of $Fe^{3+}/(Fe^{3+}+Fe^{2+})$ are high. Therefore the Hajja biotites fall in the field of brown absorption or Y and Z direction (*Fig. 10*).
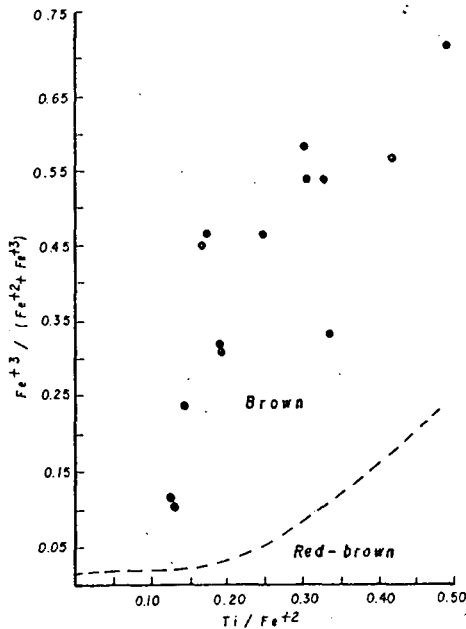

Fig. 10. Relation between the Y and Z absorption colours of biotites and their $Fe^{+3}/(Fe^{+2}+Fe^{+3})$ and Ti/Fe^{+2} ratios (after HAYAMA, 1959)

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This feature has been extensively reviewed by FOSTER (1960) HANZEN and WONES (1972) DYMEK (1983), HEWITT and ABRECTH (1986) AL-DAHAN et al. (1988). In the present work it is proposed to give only an outline of the main substitution mechanism govering tetrahedral and octahedral compositional changes.

As mentioned before Al plays a dominant role in the nature of Fe^{2+} Mg, Ti substitutional relations. Some of these relations have been studied to demonstrate this feature in the examined biotites, the most clear substitution is the relation between the four cations Ba Al^{IV} SiK, the binary diagram (*Fig. 11*) shows a good relation which indicate the process of substitution through th crystallization of the magma forming biotites. It is obvious that by lowering temperature and oxygen frugacity the SiK pair is substituting Ba Al^{IV}.
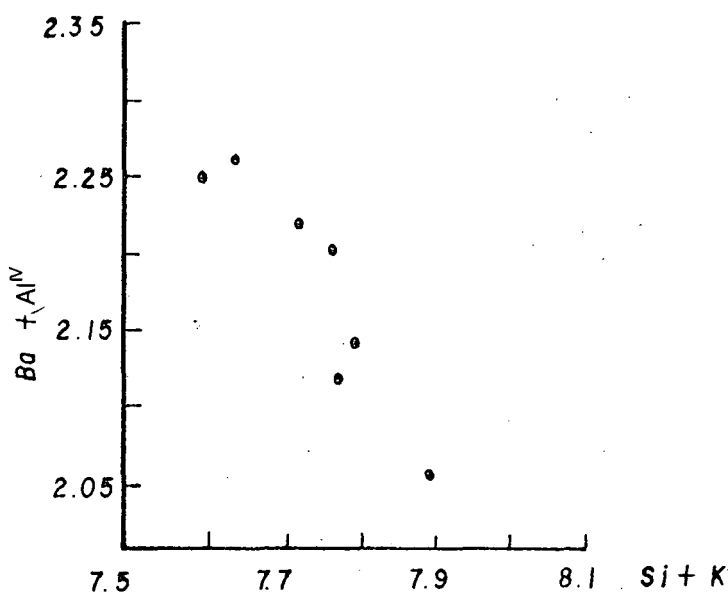

Fig. 11. Plots of Ba+Al^{IV} against Si+K

There is also a good relation in substitution between Al^{VI} and Mg ion (*Fig. 12*) which have greater values at high temperature and are by decreasing temperature. Ti cation is correlated with both Al^{IV} and Al^{VI} but with no clear relation with both cations of Alumina (*Fig. 13 and 14*). On the other hand if we divided the relation of Al^{IV} into two values Al^{IV} < 0.5 and , Al^{IV}>0.5, shows a positive relation in the first portion while in the second where Al^{IV}>0.5 a negative relation exists. This means that the Al content in biotite is the main factor in most of the substitutional processes, because of the dual role of Al ions, since Ti is slightly higher with low oxygen fugacity.

As regards the relation between Ti and Mg it is clear from the diagram (*Fig. 15*) that there is moderate correlation except at high values of Mg which means high temperature.

The following two relations are regarded as the dominant Ti substitution mechanism for Mg-biotites, which are suggested by ABRECHT and HEWITT (1988), FOLEY (1989).
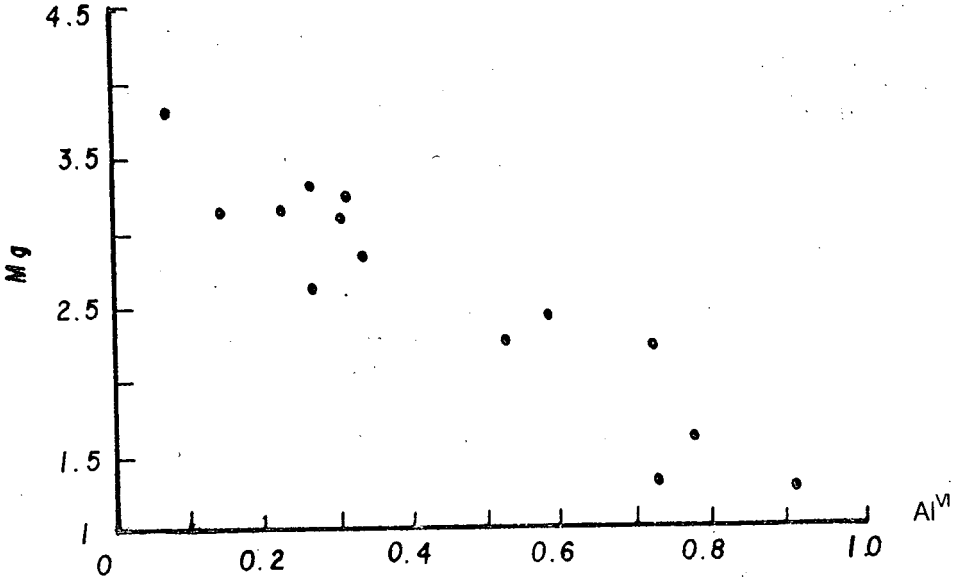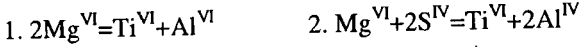
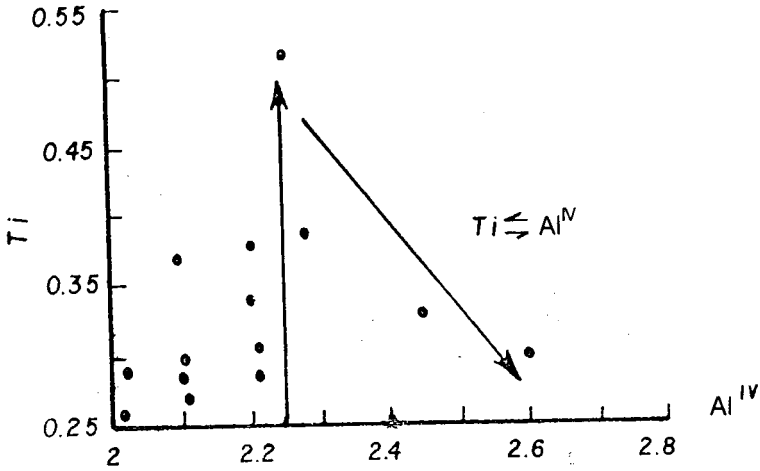1. $2Mg^{VI} = Ti^{VI} + Al^{VI}$ 2. $Mg^{VI} + 2S^{IV} = Ti^{VI} + 2Al^{IV}$

Fig. 12. Plots of Mg against Al^{VI}

Fig. 13. Plots of Al^{IV} against Ti

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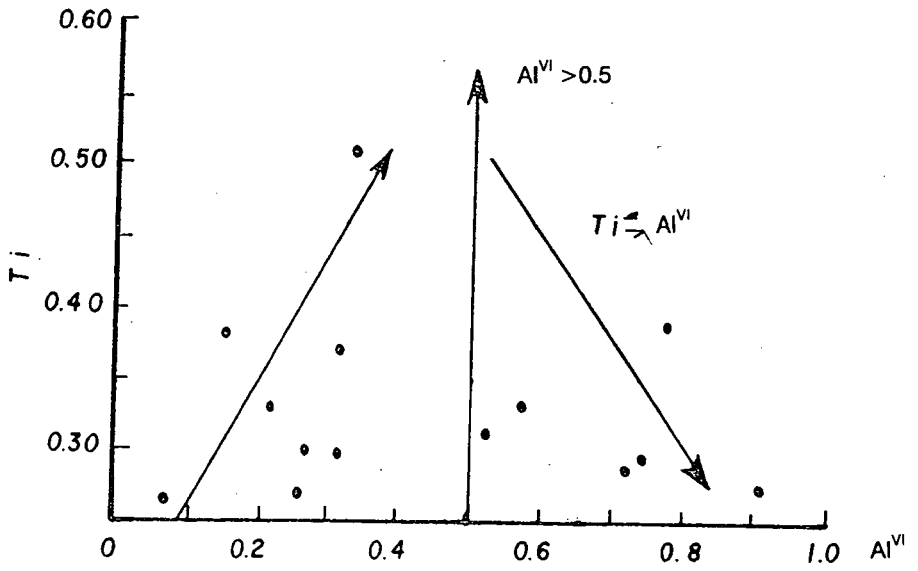
Fig. 14. Plots of Al^{VI} against Ti
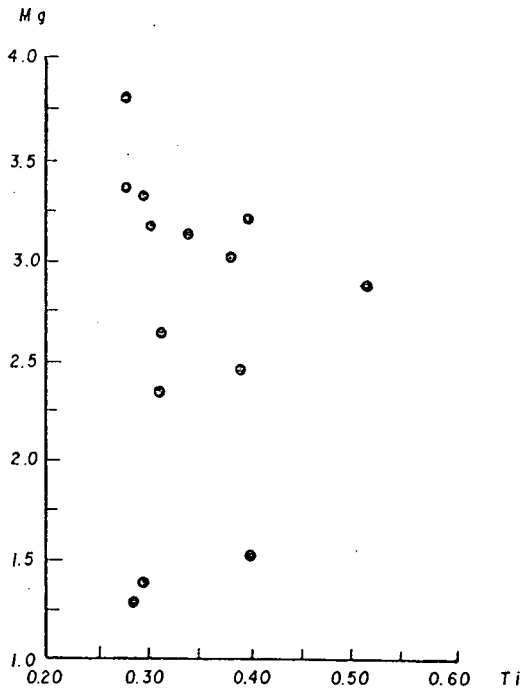

Fig. 15. Plots of Ti against Mg

Discrimination on the basis of biotite composition can be summarized as follows:

1. Discrimination $FeO^t-Al_2O_3$:– the FeO^t Al_2O_3 diagram (*Fig. 16*) shows that there is virtually no overlap between the 3 magmatic fields A, P, C only 3 samples plot in field P. FeO^t shows slight variation with increasing Al_2O_3 this may suggest that the substitution $2Al \rightarrow 3Fe^{2+}$ is not vital during biotite crystallization in calc alkaline magmatic systems.

2. Discrimination in $MgO-Al_2O_3$: – However some overlap occurs between a few biotite samples (3) in peraluminous (P) and calc-alkaline (C) rocks. Biotites which occupy fields P and C exhibit a general trend showing a gradual increase in MgO with decreasing Al_2O_3 (*Fig. 17*). This negative relation shows a substitution between Mg and Al in octahedral sites $3Mg \rightarrow Al$ within biotites in both gneous groups.

3. Discrimination in FeO^t-MgO: In this $MgO-FeO^t$ diagram *Fig. 18* biotites plot in calc-alkaline orogenic field (C) are moderately enriched in Mg (FeO^t/MgO 1.63 on average) occupying a narrow sector (C) with an $FeOt-MgO$ negative trend. Few biotites (3 samples) plot below that general trend within a separate field (P) this is because they are significantly depleted in Mg (wt% MgO 6.33 on average) compared with biotites in calc alkaline suite (–wt% MgO=13 on average). The well defined trend observed in (*Fig. 18*) reveals the fact that the $FeOt \rightarrow MgO$ substitution governs' biotite composition in calc-alkaline (C) magmas, but it it less important for biotites in peraluminous rocks (P).
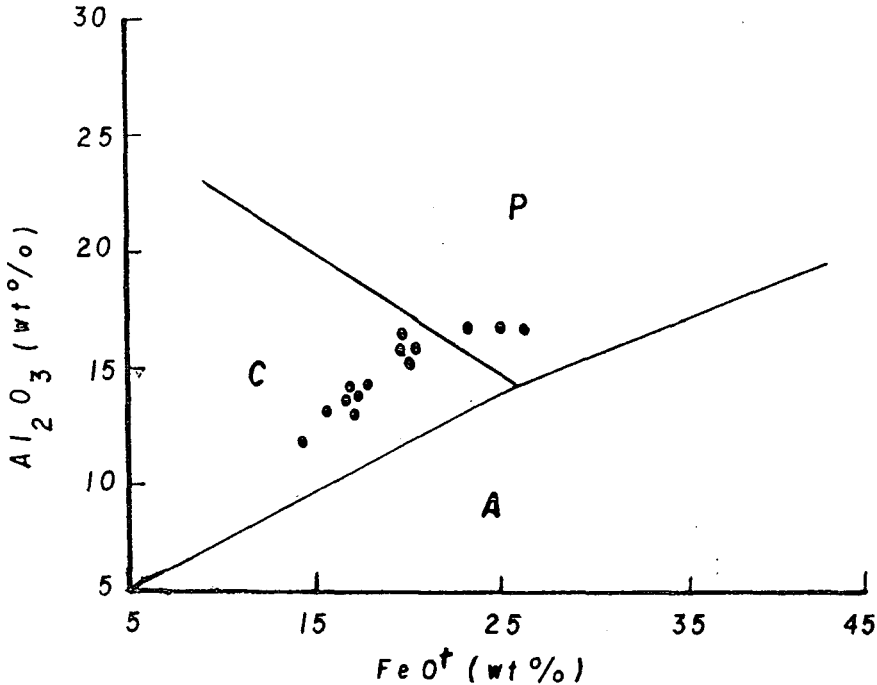
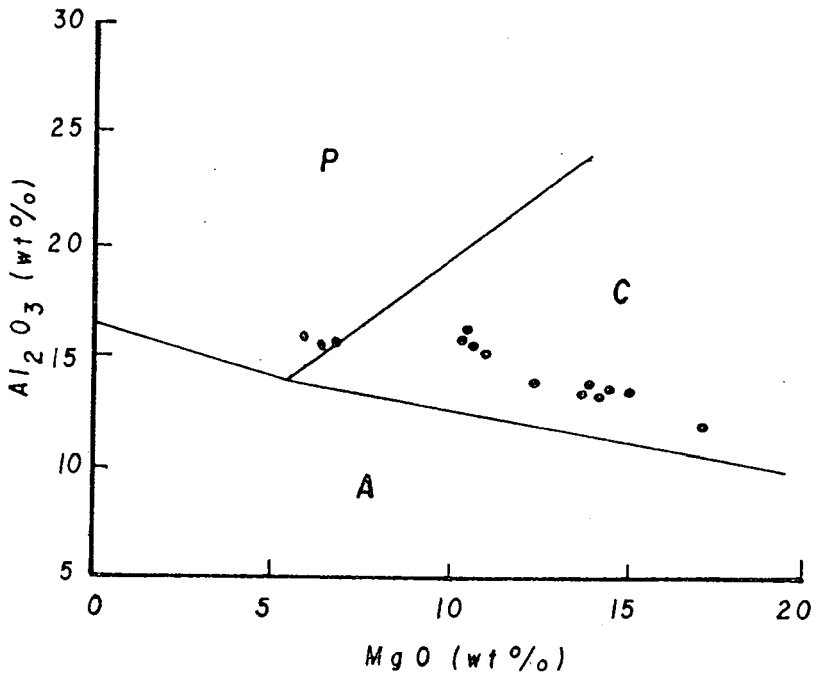Fig. 16. $FeO^t - Al_2O_3$ biotite discriminant diagram (A – alkaline, P – Peraluminous, C – Calcalkaline)

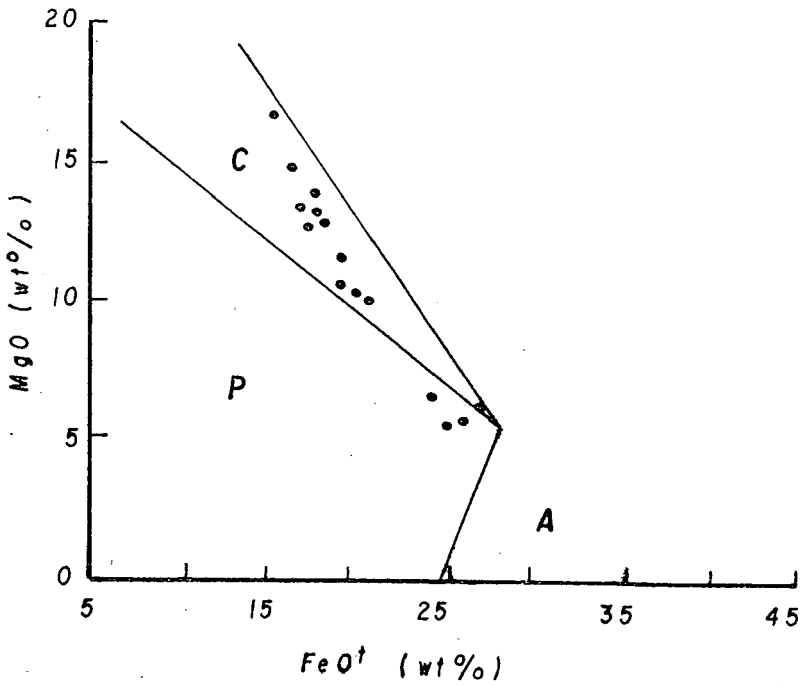*Fig. 17.* MgO–Al₂O₃ biotite discriminant diagram (symbols A, P, C as in *Fig. 16.*)

Fig. 18. FeO⁺ – MgO biotite discriminant diagram (symbols A, P, C as in *Fig. 16.*)

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Trace elements in ppm of biotites from Hajja granitoids with some localities for comparison, and some elemental ratios of the examined biotites

Sample	1	2	3	4	5	6	7	A	B	C
Sn	10	50	30	15	5	10	5	–	–	–
Ga	50	15	15	70	70	100	50	95	90	44
V	50	70	30	30	150	20	50	90	100	180
Cu	50	20	15	500	50	20	10	–	25	50
Co	20	50	20	15	70	50	30	60	40	34
Ni	30	70	20	20	10	20	5	95	25	10
Zr	50	200	500	1000	1000	1000	50	–	57	60
Sr	50	50	70	70	70	70	50	6	6	20
Ba	100	200	200	100	300	100	50	450	340	–
Pb	5	20	10	–	20	10	10	–	–	–
103 Ba/K	1.3	2.41	2.6	1.26	3.6	1,2	0.62	–	–	–
Ni/Co	1.5	1.4	1	1.3	0.1	0,4	0.15	–	–	–
103 Ni/Mg	0.39	0.81	0.24	0.24	0.1	0,22	0.06	–	–	–

A, B, C as in TABLE 1

Trace elements distribution

Some trace elements for seven samples of the examined biotites are determined and given in (Table 3), with some analyses of biotites from Egyptian granites.

Distribution of trace elements depend mainly on the presence of this element in the magmatic source from which the biotite crystallized. Many studies confirmed that enrichment of one or another element is used as indicator or pathfinder for metallogenic potentilalites, or petrogenesis of the host rocks of the biotites.

In the case of the investigated biotites, the number of samples analyzed limited the possibilities to give a good picture for the course of trace elements relations and distribution within the examined biotites. Some elemental ratios are calculated and given in (Table 3). The comparison of the trace elements values with other Egyptian biotites (KABESH et al., 1981) show that the examined biotites are similar to biotites from non mineralized pink granatic rocks. The comparison with biotites of Ras Barud (KABESH et al., 1977) shows that the present biotites have less values in the following trace elements (Ga, Cr, Ni, Ba, Sn, V) and higher values in (Cu, Zr, Sr).
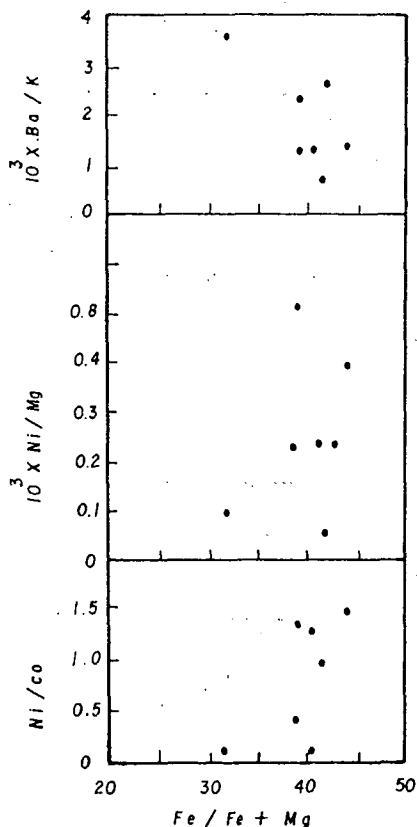

Fig. 19. Plots of Fe/Fe+Mg against some elemental ratios for biotites of Hajja granites

The ratio $FeO^t/(FeO^t+MgO)$ has been plotted against different elemental ratios such as $10^3 \times Ba/K$, 10^3 Ni/Mg, Ni/Co; (*Fig. 19*) it is clear that Ba/K ratio decrease with increasing the former value which means increasing of K content with increasing Fe total values and decreasing oxygen fugacity and temperature as well as increasing acidity of the examined biotites while the value Ni/Mg increases i.e. increasing Ni and decreasing Mg which is compatible with the previous conclusion. Ni increase may be attributed to contamination or hybridization with some basic xenolithes or magma with the granitoid magma, the same thing applies with the Ni/Co value. The value $Ga \times 10^3$ Al is compared with some Russian studies, BARABANOF (1985) which were found to be about 6 for granodiorites; this means that the biotites of Hajja were possibly derived from magma of granodioritic composition.

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