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Chlorites from Italian Granitoid Rocks

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With 3 Figures

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Summary

Chemical, X-ray, optical, thermal and microprobe analyses have been carried out on six chlorites from Italian granitoid rocks formed as a result of deuteric action on biotites. The data show that five chlorites, spanning ripidolite-brunsvigite fields, are II*b* polytypes and therefore of high temperature origin. Only one sample is a thuringite, I*b* ($\beta = 90^\circ$) polytype, typical of low temperature.

The occurrence of this last polytype, never mentioned in the literature from granitoid rocks, suggests a wider possibility for using chlorite polytypism in studying the late magmatic phenomena.

Résumé

Chlorites de roches granitiques italiennes

On a fait des analyses chimiques, aux rayons X, optiques, termiques et à la microsonde électronique, de six chlorites, provenant de roches granitiques italiennes, dérivées par action hydrothermale sur des biotites. Les résultats montrent que cinq chlorites sont ripidolites-brunsvigites, du polytype II*b*, et donc, de températures élevées. Seulement une est une thuringite, polytype I*b* ($\beta = 90^\circ$), typique de températures basses.

La découverte de ce dernièr polytype, jamais cité dans la litérature, en roches granitiques, laisse prévoir la possibilité plus vaste d'employer le polytypisme des chlorites dans l'étude des phénomènes tard-magmatiques.

Introduction

Chlorite is a common, though admittedly minor, constituent of plutonic rocks and its origin is generally explained by deuteric phenomena acting on primary ferromagnesian minerals.

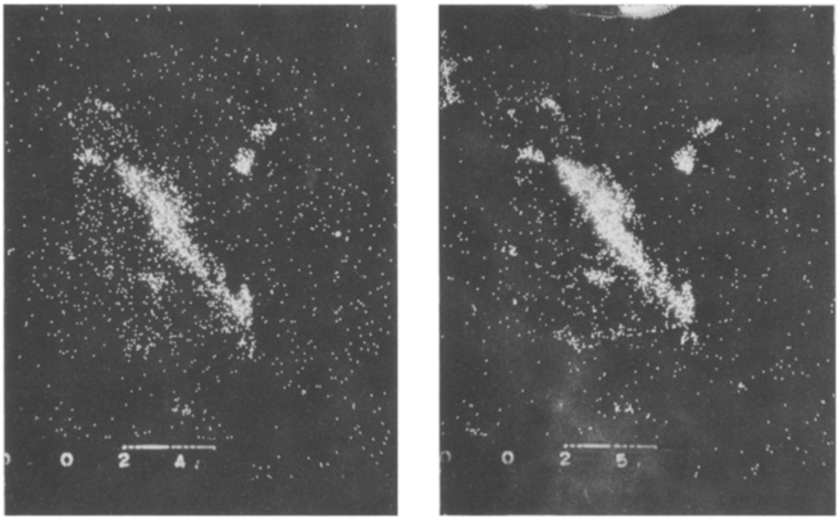
In previous reports (*Morten* and *Rossi*, 1971, 1972) the chlorites from the granitic rocks of the "dei Laghi" hercynic batholith have been studied and the authors stated that such minerals (1M, 2LC of the present work) had been formed as a transformation product of primary biotites via deuteric phenomena. It was also shown that the compositions of the chlorites and/or their association with others minerals (e. g. sphene) depend on the composition of the primary biotites. *Dodge* (1973) came to the same conclusion by studying chlorites from granitic rocks of the central Sierra Nevada batholith.

In the present work, the authors would like to expand the knowledge on the genesis of chlorites from granitoid rocks and set the chloritization phenomenon rather in a more general picture.

For this purpose, samples from several Italian outcrops (see Appendix 1) have been studied.

Chemical Study

The chemical compositions[1] of the selected chlorites and their atomic ratios calculated on the basis of 36(O, OH) and 28(O) are given in Table 1.



Ti Ca

Fig. 1. Ti and Ca$K\alpha$X-ray images (400×) in the CdA47 sample,showing the sphene distribution

The formulas are obtained from the analyses recalculated to 100% after substraction of the CaO, TiO_2 and SiO_2 amounts needed to form sphene. The presence of sphene closely associated with chlorites has been previously reported (*Morten* and *Rossi*, 1972) and reconfirmed in the present work by electron microprobe analyses (Fig. 1). The excess CaO and the Na_2O and

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[1] The separation and chemical techniques used are reported in a previous paper (*Morten* and *Rossi*, 1971).

Table 1. *Chemical Analyses (Weight–%) and Atomic Ratios to 36(O, OH) and 28 (O). (M. Bondi Analyst)*

	1M	2LC	SR(*)	BR	CdA47	Sp5
SiO_2	26.72	28.37	24.08	24.16	25.07	24.06
TiO_2	0.14	2.46	tr.	0.50	1.72	1.23
Al_2O_3	21.07	19.33	19.48	17.39	13.76	14.64
Fe_2O_3	0.80	0.42	5.06	8.80	9.86	13.28
FeO	19.18	17.28	24.57	19.80	25.46	31.41
MnO	0.10	0.12	0.20	0.29	0.62	0.90
MgO	19.88	18.55	14.49	16.45	10.40	3.07
CaO	0.10	1.70	0.44	0.45	1.43	0.86
Na_2O	0.13	0.25	n.d.	n.d.	n.d.	n.d.
K_2O	0.09	0.24	n.d.	n.d.	n.d.	n.d.
H_2O^+	12.17	11.51	11.20	11.14	10.30	10.09
H_2O^-	0.13	–	0.40	0.82	0.80	0.93
Total	100.51	100.23	99.92	99.80	99.42	100.47

	36(O, OH)	28(O)	36(O, OH)	28(O)	36(O, OH)	28(O)	36(O, OH)	28(O)	36(O, OH)	28(O)	36(O, OH)	28(O)
Si	5.39	5.43	5.68	5.72	5.16	5.16	5.14	5.14	5.53	5.53	5.47	5.46
Al^{iv}	2.61	2.57	2.32	2.28	2.84	2.84	2.86	2.86	2.47	2.47	2.53	2.54
	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00
Al^{vi}	2.42	2.50	2.55	2.63	2.08	2.08	1.57	1.57	1.30	1.30	1.55	1.53
Fe^{3+}	0.12	0.12	0.07	0.07	0.82	0.82	1.43	1.43	1.73	1.73	2.36	2.36
Fe^{2+}	3.25	3.28	3.09	3.11	4.40	4.40	3.58	3.58	4.95	4.95	6.20	6.19
Mn	0.02	0.02	0.02	0.02	0.04	0.04	0.05	0.05	0.12	0.12	0.18	0.18
Mg	6.00	6.05	5.91	5.96	4.63	4.63	5.30	5.30	3.61	3.61	1.08	1.08
	11.81	11.97	11.64	11.79	11.97	11.97	11.93	11.93	11.71	11.71	11.37	11.34
OH	16.44		16.41		16.01		16.04		15.99		15.90	
Bruc.		+ 1.28		+ 1.14		+ 1.42		+ 1.43		+ 1.23		+ 1.29
Mic.		– 1.29		– 1.14		– 1.42		– 1.43		– 1.24		– 1.26

1 M – Mergozzo ripidolite (*Morten* and *Rossi*, 1971); 2LC – "La Colma" brunsvigite (*Morten* and *Rossi*, 1972); SR – sa Rubaria ripidolite; BR – Badde Rubos ripidolite; CdA47 – Cima d'Asta brunsvigite; Sp5 – Predazzo thuringite.

(*) *D'Amico* and *Emiliani* (1959) had previously studied a chlorite from sa Rubaria; there are some discrepancies between their and our results (particularly, they found a high CaO amount: 2.99%).

K_2O were ignored because there is no place in the chlorite structure for cations as large as Na^+, K^+ and Ca^{2+} (*Deer* et al., 1965; *Foster*, 1962; *Albee*, 1962).

The analysed chlorites were plotted according to the classification scheme proposed by *Foster* (1962). Their points, as well as those of the chlorites from granitic rocks of the central Sierra Nevada batholith (*Dodge*, 1973), fell mainly in the ripidolite-brunsvigite fields. Only the Sp5 sample fell in the thuringite field. An exceptionally iron-rich chlorite from plutonic rocks, such as the Sp5 specimen, has never as far as we know been reported in the literature. In other respects it shows remarkable similarities to chlorites from mineralized hydrothermal veins in Japan (*Shirozu*, 1958).

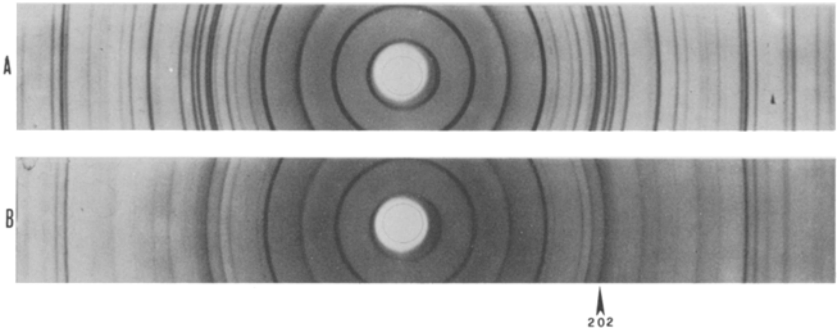

Fig. 2. Powder photographs of CdA47(A) representing the II*b* polytype, and of Sp5(B), the I*b*($\beta = 90°$) polytype

Table 1 also shows that the excess of charges in the brucite-like layer is balanced by the deficiency of charges in the mica-like layer, if cations are allocated as suggested by *Foster* (1962).

X-ray Study

The X-ray data have been obtained by means of diffraction patterns and powder photographs (Cu$K\alpha$, Fe$K\alpha$, 114.6 mm Debye-Scherrer cameras). The observed *d*-values, the relative intensities visually measured, Miller indices and cell parameters, calculated by least square refinement, are listed in Table 2. A comparison of the obtained data with those reported in the literature (*Shirozu*, 1958; *Brown* and *Bailey*, 1962; *Hayes*, 1970) shows that the 1M, 2LC, SR, BR and CdA47 samples are II*b* polytypes, while the Sp5 sample is a I*b* ($\beta = 90°$) polytype[2].

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[2] The X-ray single crystal data on the Sp5 sample confirm $\beta = 90°$.

Table 2. d_{obs} *Values, Relative Intensities and Cell Parameters*

	1M		2LC		SR		BR		CdA47		Sp5	
hkl	d_{obs}(Å)	*I*	d_{obs}(Å)	*I*	d_{obs} (Å)	*I*	d_{obs}(Å)	*I*	d_{obs}(Å)	*I*	d_{obs}(Å)	*I*
001	14.1	s	14.1	s	14.1	s	14.1	s	14.1	s	14.1	s
002	7.07	vs	7.08	vs	7.07	vs	7.07	vs	7.11	vs	7.07	vs
003	4.71	s	4.71	s	4.72	s	4.73	s	4.67	s	4.69	s
004	3.53	vs	3.54	vs	3.54	vs	3.54	vs	3.54	vs	3.52	vs
005	2.83	w	2.83	w	2.83	ms	2.83	w	2.82	w	2.83	m
200											2.72	m
131 $20\bar{2}$	2.59	ms	2.59	ms	2.61	ms	2.60	ms	2.61	ms		
201 $13\bar{2}$	2.54	ms	2.55	ms	2.55	ms	2.55	ms	2.55	ms		
132 $20\bar{3}$	2.44	ms	2.44	ms	2.46	ms	2.45	ms	2.46	ms		
202 $13\bar{3}$	2.38	ms	2.38	ms	2.39	ms	2.39	ms	2.39	ms	2.51	ms
133 $20\bar{4}$	2.26	ms	2.26	ms	2.27	ms	2.27	w	2.27	ms		
$13\bar{5}$ 204	2.00	s	2.01	s	2.02	s	2.01	s	2.01	s	2.14	w
007											2.02	w
$20\bar{6}$ 135	1.88	w	1.88	w	1.889	ms	1.887	w	1.887	mw		
$13\bar{6}$ 205	1.821	vw	1.82	vw	1.825	vw	1.825	vw	1.830	vw		
$13\bar{7}$ 206	1.659	vw			1.662	vw	1.665	vw	1.663	vw		
240 206											1.765	w
$20\bar{8}$ 137	1.563	ms	1.567	ms	1.566	ms	1.569	w	1.566	mw		
060 $33\bar{1}$	1.542	s	1.540	s	1.546	ms	1.546	w	1.544	s	1.553	s
062 331 $33\bar{3}$	1.508	w	1.506	w	1.513	w	1.513	vw	1.514	mw	1.517	ms
$13\bar{9}$ 208	1.392	ms	1.393	ms	1.393	ms	1.395	w	1.393	mw		
208 063											1.481	w
064											1.422	w
400 $26\bar{2}$	1.324	vw	1.324	vw	1.331	vw			1.329	w		
a(Å)	5.35 ± 0.01		5.339 ± 0.002		5.357 ± 0.003		5.356 ± 0.002		5.358 ± 0.002		5.39 ± 0.02	
b(Å)	9.25 ± 0.02		9.245 ± 0.003		9.277 ± 0.005		9.278 ± 0.004		9.282 ± 0.005		9.328 ± 0.009	
c(Å)	14.23 ± 0.03		14.281 ± 0.007		14.241 ± 0.008		14.265 ± 0.005		14.244 ± 0.008		14.15 ± 0.03	
β	97.0 ± 0.2		97.21 ± 0.04		97.23 ± 0.05		97.22 ± 0.03		97.21 ± 0.05		90.3 ± 0.3	

As far as the latter polytype is concerned, *Brown* and *Bailey* (1962) and *Hayes* (1970) report maximum intensity values for the 202 reflection, while *Shirozu* (1958), for the same reflection, reports variable and not maximum intensity values, just as we found for the Sp5 sample (Table 2, Fig. 2). On the other hand *Hayes* (1970) found that in this polytype the *h*0*l* reflections – and 202 in particular – show a progressive weakening with a continued grinding, and concurrent development of a broad, diffuse diffraction band on the high angle side of the 202 line.

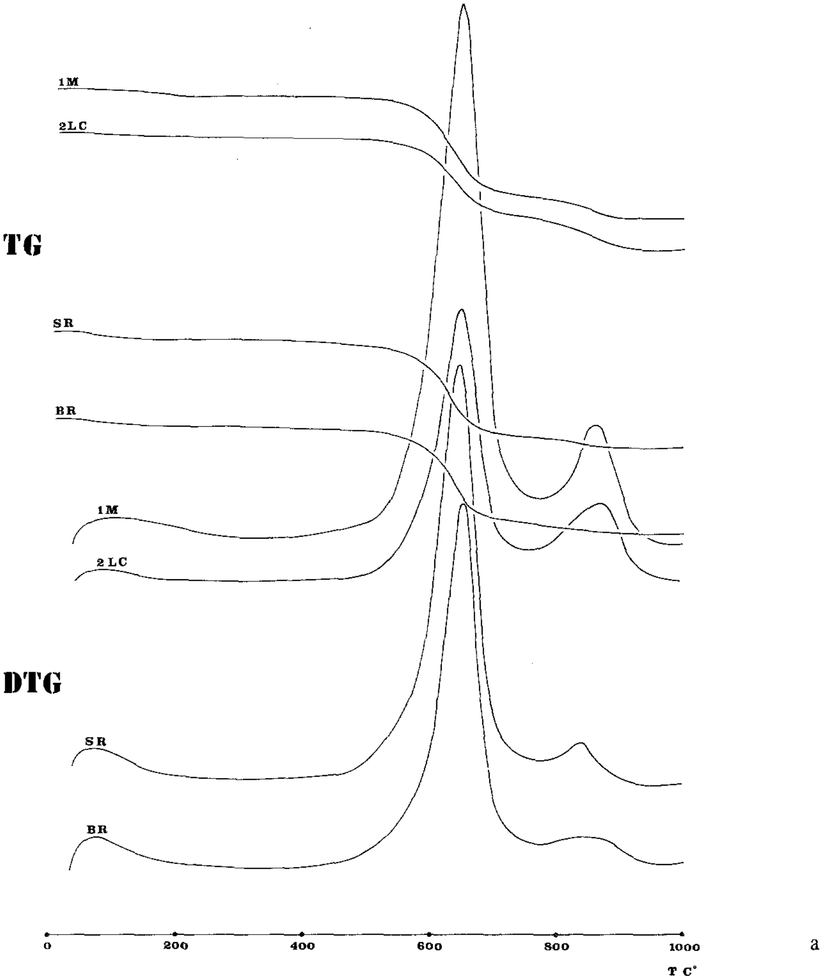

Fig. 3. TG and DTG curves. (a) In nitrogen atmosphere; (b) In nitrogen atmosphere and in air

Optical Study

The optical properties of the analysed chlorites are given in Table 3. For our data as compared with that determined by *Albee* (1962), the relationship existing between the optic sign and the refractive index is found to be on the whole the same in both cases, whereas the interference colours differ: the BR and SR samples (negative sign) exhibit abnormal brown interference colours, whereas the 1M and 2 LC samples exhibit normal interference

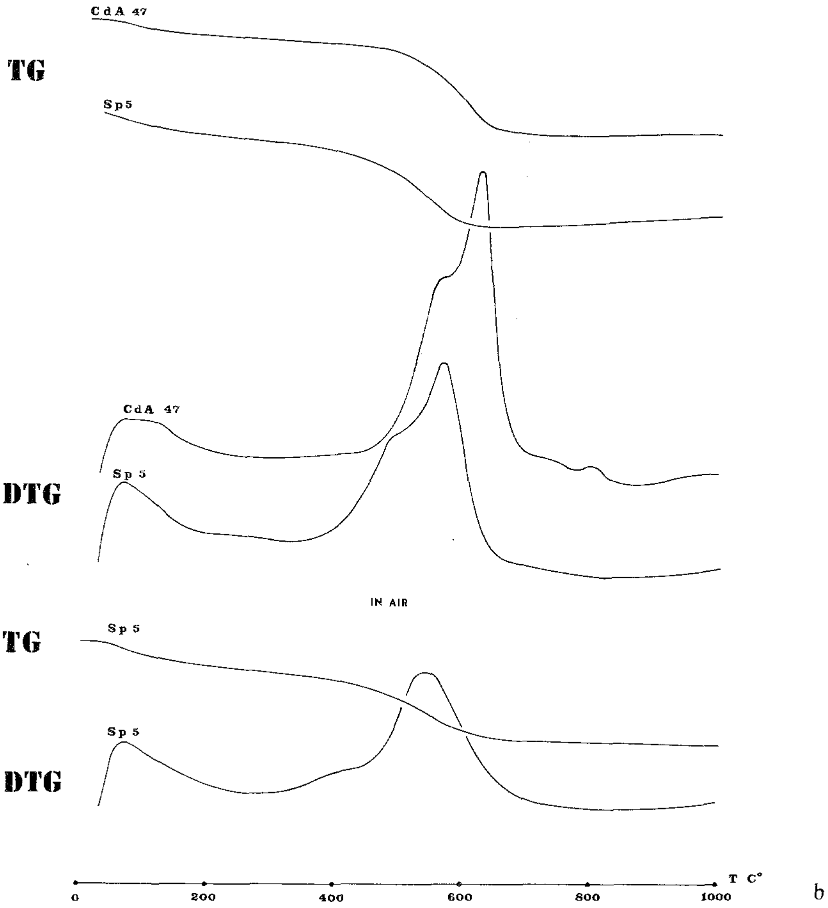

Table 3. *Optical Properties*

	1M	2LC	SR	BR	CdA47	Sp5
n (*)	1.610	1.605	1.629	1.627	1.636	1.665
$2V$	0°–10°	0°–10°	~0°	~0°	0°–10°	~0°
Sign	+	+	–	–	–	–
Pleoch.	$\alpha \simeq \beta$ = grass green	$\alpha \simeq \beta$ = light green	$\beta \simeq \gamma$ = grass green	$\beta \simeq \gamma$ = green	$\beta \simeq \gamma$ = green	$\beta \simeq \gamma$ = dark green
scheme	γ = pale yellow	γ = pale yellow	α = pale yellow	α = pale yellow	α = pale yellow	α = greenish yellow
	$\alpha \simeq \beta > \gamma$	$\alpha \simeq \beta > \gamma$	$\beta \simeq \gamma > \alpha$	$\beta \simeq \gamma > \alpha$	$\beta \simeq \gamma > \alpha$	$\beta \simeq \gamma > \alpha$
Int. col.	–	–	reddish-brown	reddish-brown	violet-blue	graysh-blue

(*) Refractive index values are ± 0.002; measurements made with Na_D light colours. The Sp5 sample – 1*b* ($\beta = 90°$) polytype – exhibits abnormal interference colours contrary to what *Shirozu* (1958) had stated.

Colours and the pleochroic scheme are in agreement with the data found in the literature.

Thermal Study

The thermal study was carried out by means of TG analyses, using a Dupont 951 thermogravimetric analyzer, in a nitrogen atmosphere and in the air, with a heating rate of 50° C per minute.

TG and DTG curves are reproduced in Fig. 3. They show that (1) the dehydration of the brucite-like and mica-like layers takes place at two different temperatures; (2) the dehydration peak of the mica-like layer decreases, disappearing in the Sp5 sample; (3) the dehydration peak of the brucite-like layer shows the same inflexion in the CdA47 and Sp5 samples.

Regarding the first point: the dehydration taking place at two different temperatures in ripidolites is neither in agreement with what *Caillére* and *Hénin* (1960 , 1963) stated, nor with what has been previously found studying the 1M sample by TGA with a less sensitive apparatus (*Morten* and *Rossi*, 1971).

Regarding the second point: it is worthwhile emphasizing the inverse relationship between the 2nd peak intensity/1st peak intensity ratio and the Fe^{2+} content. The decrease in the intensity of the second peak, up to the point of disappearing in the Sp5 the Fe^{2+}-richest sample, is therefore due mainly to the oxidation of the Fe^{2+} at the expense of the mica-like layer oxydriles.

Regarding the third point: the first peak inflexion (CdA47 and Sp5 samples) indicates a reduction in the weight-loss caused by Fe^{2+} oxidation at the expense of the brucite-like layer oxydriles. The two chlorites in which this phenomenon occurs are the Fe^{2+}-richest samples and consequently moderate amounts of Fe^{2+} are present in the brucite-like layer. The proof is shown by the fact that the inflexion disappears when the test is carried out in air. This is because the Fe^{2+} oxidation then takes place at the expense of the O_2 in the air (Fig. 3 b).

Conclusions

From the obtained data, we may deduce that chlorites from granitoid rocks have formed as a result of deuteric phenomena acting on primary ferromagnesian minerals, mainly biotites. Their composition and the close association with others minerals is influenced by the composition of the primary minerals (*Chayes*, 1955; *Schwartz*, 1958; *Morten* and *Rossi*, 1972; *Dodge*, 1973).

Such chlorites are Fe-rich members, generally II*b* polytypes, and therefore of high temperature origin (*Brown* and *Bailey*, 1962; *Hayes*, 1970). Nevertheless, we have found a chlorite (Sp5) of the I*b* ($\beta = 90°$) polytype which, as far as we know, has never been mentioned in the literature from granitoid rocks. This polytype, typical of low temperature, has been considered up to now to be peculiar to diagenetic environment (*Hayes*, 1970) and/or to low temperature metamorphism (*Brown* and *Bailey*, 1962).

Since the Sp5 sample occurs in most deuterized Predazzo granite facies (*Paganelli* and *Tiburtini*, 1964; *Del Monte* et al., 1965; *Borsi* and *Ferrara*, 1967; *Lucchini* et al., 1971; *Morten* and *Rossi*, 1974), the presence of this I*b* ($\beta = 90°$) polytype shows that deuteresis in the Predazzo granite took place at a low temperature, under 150–200°C, assuming therefore this value as a boundary to the conversion to II*b* polytype (*Hayes*, 1970).

So, it is possible to state that the deuteric phenomenon which causes the primary ferromagnesian minerals-chlorite transformation takes place in a wide range of temperatures and that such chlorites are not necessarily the II*b* polytype.

Chlorite polytypism therefore can be used as a geothermometer in the study of deuteric phenomenon.

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Appendix I

Sample 1M

Mergozzo, Monte Montorfano, Novara, hercynic dei Laghi batholith. The "green" granite host rock shows a granular holocrystalline hypidiomorphic texture with modal composition: quartz = 12–15%; plagioclase = 48–50%; chlorite + muscovite = 26–32%; others = 2–5% and chemical composition: SiO_2 = 62.14%; TiO_2 = 0.40%; Al_2O_3 = 18.20%; Fe_2O_3 = 2.34%; FeO = 2.07%; MnO = tr; CaO = 0.36%; MgO = 4.80%; Na_2O = = 4.75%; K_2O = 2.18%; H_2O^- = 0.12%; H_2O^+ = 3.15%. The chlorite appears in rounded fibrous aggregates and/or concentrated in irregular lamellae (*Gallitelli*, 1938).

Sample 2LC

La Colma, Alzo, Novara, hercynic dei Laghi batholith. The granite host rock shows a granular hypidiomorphic holocrystalline texture with modal composition: quartz = 30.3%; feldspars = 56.3%; chlorite = 10.4%; others = 3.0%. The chlorite, pseudomorphous after biotite, appears in ondulose and strained lamellae. Along the cleavage traces many leucoxenic granulations can be noticed (sphene) (*Morten* and *Rossi*, 1972).

Sample SR

Riu de sa Rubaria, Nuoro, hercynic Sardo-Corso batholith. The syenite-lamphrosyenite host rock has mainly hypidiomorphic texture, partly poikilitic with modal composition: microcline perthite = 22.5%; albite = 41%; muscovite = 2.5%; chlorite = 31%; sphene = 2%; apatite = 0.5%; Fe-ores = 0.5% and chemical composition : SiO_2 = 51.89%; TiO_2 = 0.80%; Al_2O_3 = 19.49%; Fe_2O_3 = 1.91%; FeO = 7.96%; MnO = 0.05%; MgO = 4.87%; CaO = 1.28%; Na_2O = 4.92%; K_2O = 3.87%; H_2O^+ = 3.10%; H_2O^- = 0.19%. The chlorite can be found in regular lamellae or more frequently arranged in flake-like crystals (*Emiliani* and *D'Amico*, 1959).

Sample BR

Badde Rubos, Monte Ortobene, Nuoro, hercynic Sardo-Corso batholith. The host rock is a pegmatite granite with large feldspars, with modal composition: quartz = 33.5%; microcline = 29%; plagioclase = 30%; muscovite = 3.5%; biotite (chlorite) = 3.5%; others = 3.5% and chemical composition : SiO_2 = 71.55%; TiO_2 = 0.24%; Al_2O_3 = 15.31%; Fe_2O_3 = 1.89%; FeO = 0.29%; MnO = 0.03%; MgO = 0.55%; CaO = 1.36%; Na_2O = 3.46%; K_2O = 4.84%; H_2O^- = 0.14%; H_2O^+ = 0.65% (*D'Amico*, 1959). The chlorite appears in flake-like crystals or pseudomorphous after biotite.

Sample CdA47

Cima d'Asta, Trentò, hercynic Cima d'Asta batholith. The granite host rock has a granular holocrystalline hypidiomorphic texture, with modal composition: quartz = 2.82%; alkali-feldspars = 3.41%; plagioclase = 31.0%; biotite (chlorite) = 6.2%; others = 0.5% (*D'Amico* and *Del Monte*, 1969). The chlorite is pseudomorphous after biotite with exsolution of leucoxenic granulations (sphene) (*Gandolfi*, 1967).

Sample Sp5

Predazzo, Trento, triassic Predazzo-Monzoni intrusive complex. The host rock is a "medium grained albite granite" and "texture is holocrystalline, moderately granular and hypidiomorphic" with modal composition: quartz = 33.0%; perthite = 40.0%; plagioclase = 23.5%; others = 3.5% and chemical composition: SiO_2 = 73.02%; TiO_2 = –; Al_2O_3 = 14.92%; Fe_2O_3 = 0.50%; FeO = 0.92%; MgO = 0.10%; MnO = 0.01%; CaO = = 0.98%; P_2O_5 = 0.09% (*Paganelli* and *Tiburtini*, 1964). The chlorite appears distributed in pseudomorphous lamellae after biotite.

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