Chlorpotassium ferro-pargasite from Sesia-Lanzo marbles (Western Italian Alps): a record of highly saline fluids

DANIELE CASTELLI

Institut für Mineralogie und Petrographie, ETH-Zentrum CH-8092 Zürich, Switzerland

Permanent address: Dipartimento di Scienze della Terra dell'Università, via V. Caluso 37, I-10125 Torino, Italy

ABSTRACT. — In impure marbles of the internal Sesia-Lanzo Zone near Pont S. Martin (Aosta Valley), blue-green amphiboles developed during the re-equilibration of early-alpine eclogitic assemblages. These amphiboles, investigated by both electron microprobe analysis and single-crystal X-ray diffractometry, have compositions ranging from ferro-edenitic hornblendes to ferro-pargasites. The latter may contain up to 4.21 wt% of chlorine and 3.95 wt% of K₂O. Low Mg/(Mg + Fe²⁺) ratios allow high Cl = OH and K=Na substitutions in O(3) and A sites respectively.

The unusual crystal-chemistry is related to a particular composition of both solid and fluid phases involved in hydration reaction producing the Cl- and K-rich amphiboles.

Qualitative topology of related mixed volatile equilibria and estimates of available experimental data on H₂O-CO₂-KCl ternary fluids, at high temperatures and pressures, suggest that these amphiboles grew in equilibrium with a water-rich saline fluid, undersaturated respect to the KCl component.

Key words: Cl-amphiboles, saline fluids, metamorphism, western Alps.

Introduction

Potassium- and chlorine-rich calcic amphiboles have been found in few localities after KRUTOV (1936) described the first occurrence of dashkesanite (chlorpotassium hastingsite according to the I.M.A. nomenclature, LEAKE, 1978) in skarn rocks from Transcaucasia (U.S.S.R.).

Such amphiboles have been reported by LEELANANDAM (1969, 1970) in charnockitic rocks from Kondapalli (India), and JACOBSON (1975) has published additional data on dashkesanites from the type-locality and from St. Paul's Rocks (Equatorial Atlantic). Chlorine-bearing potassian hastingsites have been found by DICK and ROBINSON (1979) in a sphalerite skarn in southern Yukon. SHARMA (1981) and KAMINENI et al. (1982) recently described other chlorpotassium hastingsites from granulitic rocks of the Indian peninsula.

This paper is focused on the occurrence of calcic amphiboles - ranging in composition from chlorpotassium ferro-pargasites to potassian ferro-edenitic hornblendes - in the astroalpine Sesia-Lanzo unit (Western Italian Alps), and includes electron microprobe analyses and single-crystal X-ray diffraction data. The nature of the coexisting fluid phase is also discussed, providing new information about hydration reactions which involved the early-alpine eclogitic assemblages of the Sesia-Lanzo marbles.

Geological and petrographical setting

The internal sector of the Sesia-Lanzo Zone (Fig. 1) is a slice of astroalpine continental crust composed by high-grade paragneisses with minor amphibolites and marbles,
In the lower Aosta Valley, near Pont S. Martin (Fig. 1), impure marbles are strongly folded within eclogitic micaschists. Marbles occur as small lenses and/or discontinuous intercalations, centimeters to few meters thick, and may include smaller boudins of metabasites. The marbles also show a variety of chemical interactions with the country rocks. The chemistry of silicate phases and textural relationships (Fig. 2 and CASTELLI, 1987) allow one to reconstruct their complete metamorphic evolution.

Centimeter-scale pre-alpine porphyroclasts of colourless, twinned clinopyroxene (CpxI in Fig. 2) were partially re-equilibrated during the early-alpine eclogitic metamorphism and now exhibit thin rims of pale green clinopyroxene (CpxII) and garnet (Grt) + CpxII inclusions. Twin lamellae in CpxI, probably related to stress, can be up to 25 µm thick and develop parallel to the (100) of CpxI, showing the same optical characters and very small differences in chemical composition.

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Fig. 1. — Field occurrence of the impure marbles (black lenses) of the internal Sesia-Lanzo Zone near Pont S. Martin; the star indicates the outcrop of Cl- and K-bearing amphiboles. The inset shows the location of the enlarged map and the main tectonic units of the Western Italian Alps. SA: Southern Alps, SL: Sesia-Lanzo (dotted area is the internal sector), DBL: Dent-Blanche nappe; PN: Piemonte ophiolite nappe, GP: Gran Paradiso unit, MR: Monte Rosa unit, SB: S. Bernardo unit.

intruded by late hercynian gabbroids and granitoids (DAL PIAZ et al., 1972; COMPAGNONI et al., 1977).

It records a multistaged metamorphic history of alpine age ranging from 130 to 40 Ma (HUNZIKER, 1974; OBERHAENSLI et al., 1982). The main steps of this evolution are the development of early-alpine high pressure-low temperature assemblages successively re-equilibrated under blueschist and then, under greenschist facies conditions (COMPAGNONI, 1977). The P-T-t path of the Internal Sesia-Lanzo Zone is currently explained by subduction mechanism followed by decompressional stages (DAL PIAZ et al., 1972; COMPAGNONI et al., 1977; POGNANTE et al., 1980; LARDEAUX et al., 1982).

Fig. 2. — Textural relationships between pre-alpine clinopyroxene (CpxI) and alpine minerals in Pont S. Martin marbles (see text for discussion). Empirical formulae are also reported for some of the involved minerals. CpxI is a sodic augite very homogeneous in composition, with a little more alumina in the lamellae; early-alpine CpxII, coexisting with garnet (Grt), is definitely alumina- and sodium-richer.
Potassium- and chlorine-rich hornblendes (up to 350 \( \mu \text{m} \) long) grew during the first hydration of the eclogite assemblage, with the \( c \) axis co-planar to the lamellae of CpxI (Fig. 2). Despite their size, the amphiboles are easy to recognize because of the strong pleochroism ranging from very pale yellowish-brown to dark blue-green, and appear optically homogeneous. Crystals are locally coexisting with a biotite that also contains large amount of halogens (CASTELLI, 1987).

**Analytical techniques**

Chemical analyses on polished thin sections were carried out with an automatical ARL SEMQ electron microprobe, that uses six X-ray spectrometers and an X-ray dispersive analyzer (TN 2000 by Tractor Northern) simultaneously, with correction on-line for drift, dead-time and background (SOMMERAUER, 1981). An acceleration voltage of 15 KV and a sample current of 20 nA (on brass) were applied, yielding a beam size of 20 \( \AA \). Natural and/or synthetic oxides and silicates were used as standards, as well as monitors to check the accuracy of measurements. Detection limits and relative errors are listed in Table 1. A ZAF computer program has been used off-line to obtain full corrections for X-ray absorption, X-ray fluorescence by characteristic and continuous excitation, atomic number effect, backscatter and ionisation-penetration losses (GUBSER, 1975).

X-ray element maps and backscattered images have been made with a JOEL JSM-840 electron microscope connected on-line with a X-ray dispersive analyzer (TN 2000 by Tracor Northern), applying an acceleration potential of 15 KV and a sample current of 1.5 nA.

Chemical analyses were recalculated using a computer program (ULMER, 1986) that assigns cations to crystallographic sites assuming: \( N_{\text{cat}} = (\text{Ca} + \text{Na} + \text{K}) = 13.0 \).

\( \text{Fe}^{2+} \) and \( \text{Fe}^{3+} \) are computed assuming stoichiometry and charge balance. The \( \text{H}_2\text{O} \) wt\% is calculated from OH content derived from stoichiometry.

Amphibole single-crystals have been selected and examined on a Philips PW1100 four-circle diffractometer with graphite-monochromated Mo radiation. Data collection and crystallographic refinements have been carried out on two amphibole crystals according to the procedures described by UNGARETTI (1980) and by UNGARETTI et al. (1981, 1983). Crystal-chemical compositions have been obtained from site occupancies through the CORANF computer program, following the procedures described by CANNILLO et al. (1981) and by UNGARETTI et al. (1981, 1983).

Representative chemical compositions and structural formulae obtained by electron microprobe analysis are listed in Table 2. Following plots have been made including all available quantitative data (52 spot analyses on the crystals). The site-populations deduced from crystallographic refinements are given in Table 3, together with the unit-cell parameters.

The details of the crystallographic analysis and the structural implication of the main isomorphous substitutions characterizing these amphiboles will be discussed in a separate paper (in prep.).

**Amphibole crystal-chemistry**

The chemical composition of the blue-green amphiboles strongly changes in different
crystals. Smaller chemical ranges are detectable within each crystal, although they do not present any regular zoning between core and rim. These amphiboles are characterized by more than 20 wt% of total FeO, high alumina amount (8.45-13.40 wt%) inversely related to silica (35.89 - 43.38 wt%); and are all calcic amphiboles (10.27 < CaO < 11.65 wt%) according to the I.M.A. classification (Leake, 1978), with the M(4) site containing only small amount of sodium. Composition ranges from ferro-paragisite to ferro-edenitic hornblende (Fig. 3).

The classification as pargasites instead of hastingsites has been assumed without direct determination of the ferric iron. The calculated Fe⁺⁺/Al⁺⁺⁺ ratios, which are lower than unity (in good agreement with the X-ray refinements), suggest a partial solid solution with the corresponding hastingsitic end-
The most important chemical features are the substitutions occurring at the A site and at the O(3) anion position.

Ferro-pargasites contain up to 3.95 wt% of K\textsubscript{2}O and 4.21 wt% of chlorine (Fig. 4), consequently the prefix \textit{chloropotassium} has to be used for them. Ferro-edenitic hornblendes, containing about 1.00 wt% of K\textsubscript{2}O, can be defined as potassium ferro-edenitic hornblendes (Leake, 1978).

These K and Cl contents are among the highest recorded in the literature; only in the type-locality of dashkesanite higher potassium and chlorine values were found (Krutov, 1936; Jacobson, 1975).

All amphiboles show a Cl ⇌ OH exchange strongly related to the Mg/(Mg\textsuperscript{2+} + Fe\textsuperscript{2+}) ratio and to the potassium content (Fig. 5a, 5b); the fluorine content increases up to 0.80 wt%.

![Figure 3](image_url)

**Fig. 3.** - Silicon content (at. p.u.f.) versus Mg/(Mg\textsuperscript{2+} + Fe\textsuperscript{2+}) ratio showing the compositional range of the analyzed amphiboles after Leake (1978); dots and crosses refer to data from electron microprobe analysis and X-ray refinement, respectively; (a): ferro-edenitic hornblendes, (b): ferroan pargasitic hornblendes, (c): ferro-pargasitic hornblendes, (d): ferro-pargasites.

![Figure 4](image_url)

**Fig. 4.** - Backscattered electron image (a) and X-ray maps of the same area showing potassium (b) and chlorine (c) distribution in a ferro-pargasite crystal inside CpxI (light grey and grey in (a) respectively). Scale bar is 30 \(\mu\text{m}\).
TABLE 3
Site-populations (atoms p.f.u.) and unit-cell parameters (e.s.d. in parentheses) determined by X-ray single-crystal diffractionmetry on two CI- and K-bearing amphibole crystals from SL1823 rock sample

<table>
<thead>
<tr>
<th></th>
<th>AN-4</th>
<th>AN-2</th>
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<tr>
<td>Si</td>
<td>T(1)+T(2)</td>
<td>6.19</td>
</tr>
<tr>
<td>Al&lt;sup&gt;3+&lt;/sup&gt;</td>
<td>1.81</td>
<td>1.47</td>
</tr>
<tr>
<td>Al&lt;sup&gt;2+&lt;/sup&gt;</td>
<td>M(1)+M(2)+M(3)</td>
<td>0.60</td>
</tr>
<tr>
<td>Ti</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>Fe&lt;sup&gt;3+&lt;/sup&gt;</td>
<td>0.40</td>
<td>0.29</td>
</tr>
<tr>
<td>Fe&lt;sup&gt;2+&lt;/sup&gt;</td>
<td>3.16</td>
<td>2.65</td>
</tr>
<tr>
<td>Mg</td>
<td>0.82</td>
<td>1.53</td>
</tr>
<tr>
<td>Ca</td>
<td>M(4)</td>
<td>1.86</td>
</tr>
<tr>
<td>Na</td>
<td>0.14</td>
<td>0.15</td>
</tr>
<tr>
<td>Na</td>
<td>A</td>
<td>0.24</td>
</tr>
<tr>
<td>K</td>
<td>0.67</td>
<td>0.57</td>
</tr>
<tr>
<td>Cl</td>
<td>O(3)</td>
<td>1.02</td>
</tr>
<tr>
<td>OH,F</td>
<td>0.98</td>
<td>1.40</td>
</tr>
<tr>
<td>Mg/(Mg+Fe&lt;sup&gt;2+&lt;/sup&gt;)</td>
<td>0.21</td>
<td>0.37</td>
</tr>
<tr>
<td>a (Å)</td>
<td>9.922(1)</td>
<td>9.884(1)</td>
</tr>
<tr>
<td>b (Å)</td>
<td>18.219(2)</td>
<td>18.143(5)</td>
</tr>
<tr>
<td>c (Å)</td>
<td>5.360(1)</td>
<td>5.332(1)</td>
</tr>
<tr>
<td>B (°)</td>
<td>104.811(1)</td>
<td>104.861(1)</td>
</tr>
<tr>
<td>V (Å&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>936.7</td>
<td>924.2</td>
</tr>
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</table>

with decreasing of chlorine (Fig. 5c). In particular, ferro-edenitic hornblendes have the lowest contents in potassium and chlorine.

The interdependence of cation substitutions has been already observed and discussed in CI-bearing amphiboles (Leelanandam, 1969; Vielzeuf, 1982; Vanko, 1986), or can be recognized in published analyses (Jacobson, 1975; Dick and Robinson, 1979).

Volfinger et al. (1985) argue that the positive correlation between chlorine contents and \( X_{Fe} \) (\( X_{Fe} = \text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg}) \)) of some actinolitic amphiboles (Vielzeuf, 1982) is valid beyond the values \( X_{Cl} = 0.24 \), suggesting that this is related to the same structural behaviour observed in synthetic CI-bearing micas. Present data on natural ferropargasitic amphiboles show the same trend at higher chlorine contents. The calculated relationship: \( X_{Cl} = 1.53 X_{Fe} - 0.74 \) (correlation coefficient \( r = 0.98 \); see also Fig. 5a for a comparison) would fix at \( X_{Fe} = 0.48 \) the

Fig. 5. — Chlorine content versus Mg/(Mg+Fe<sup>2+</sup>) ratio (a), potassium (b) and fluorine (c) content (at. p.f.u.). Symbols as in fig. 3. The equations of the regression lines are: Mg/(Mg+Fe<sup>2+</sup>) = -0.31Cl + 0.51 (correlation coefficient \( r = 0.98 \)) in (a); K = 0.58Cl + 0.11 \( (r = 0.96) \) in (b); F = -0.35Cl + 0.42 \( (r = 0.91) \) in (c). See text for discussion.
minimum value which allows chlorine to enter
the O(3) site.

Discussion: chlorpotassium ferro-pargasite and fluid composition

Several strategies have been proposed in order to estimate the composition of fluids during metamorphic processes. One approach uses mineral-solution equilibria (FRANZ et al., 1981; FERRY and BURT, 1982), while others employ the crystal-chemistry of hydrated phases (MUNOZ and SWENSON, 1981; VOLFINGER et al., 1985; VOLFINGER and ROBERT, 1986), or fluid and solid inclusion studies (MERCOLI, 1982; TROMMSDORFF et al., 1985). Solid and fluid phase relations in the system CO₂-H₂O-NaCl have been recently discussed and applied to metacarbonate rocks (TROMMSDORFF and SKIPPEN, 1986; SKIPPEN and TROMMSDORFF, 1986).

Unfortunately, these Cl- and K-bearing amphiboles do not provide enough information to exactly define the solid-fluid system. In fact, despite GILBERT'S paper (1966) on ferro-pargasite stability, there are very few experimental data available on iron- and halogen-rich minerals coexisting, at high temperatures and pressures, with an halogen-rich fluid phase. Nevertheless, some general considerations may be, at least qualitatively, discussed.

Petrographical setting (Fig. 2) has shown that the reaction producing these amphiboles took place inside CpxI, along the (100) twin lamellae which were the preferential surfaces for the pyroxene-fluid interaction: such textures are characteristic when pyroxenes enter the stability field of amphiboles in the presence of fluid (i.e. YAMAGUCHI et al. 1978; VELEN and BUSECK, 1981).

No direct P-T estimations are available on hydration reactions following the early-alpine metamorphism in marbles of the Sesia-Lanzo Zone. Usually, these reactions are located in mafic and/or pelitic rocks of the Internal Sesia-Lanzo Zone along the P-T-t path between the stability fields of the early-alpine parageneses — at pressures higher than 14-15 Kb at T ≈ 550°C — and the greenschist late-alpine re-equilibration at P ≤ 6 Kb, T = 400-500°C (UNGARETTI et al. 1983, with references therein).

Textural relationships indicate that the Cl-K-amphiboles developed after the breakdown of the early-alpine CpxII (being in the stability field of Na-amphiboles at pressure lower than 10-12 Kb and higher than 5-6 Kb; see UNGARETTI et al. 1983, Fig. 12) and before the greenschist facies minerals enveloping CpxI + CpxII. Thus, considering the high dP/dT ratio of the trajectory followed by these rocks, 8 Kb and 500°C are here assumed to be the P-T conditions for the growth of Cl-K-amphiboles.

Modal analysis of the assemblage sketched in Fig. 2 suggests that CpxI and CpxII contain enough sodium and particularly alumina to produce the Cl-K-bearing amphiboles, while potassium, chlorine and fluorine have been necessarily provided by a fluid phase, according to an equilibrium as:

\[
CpxI (± CpxII) + Cl-K-fluid = Cl-K-Amp + Cc + Qz
\]  

(1)

The presence of chlorine-rich fluids has been already suggested in order to explain other occurrences of Cl-bearing amphiboles (JACOBSON, 1975; DICK and ROBINSON, 1979; VIELZEUF, 1982; VANZO, 1984, 1986; VOLFINGER and ROBERT, 1986). Furthermore, in other siliceous marbles of the Sesia-Lanzo Zone (outcropping few kilometers south of Pont S. Martin) saline fluid inclusions have been recognized in omphacite (CASTELLI, 1987).

However, an exact evaluation of the salinity of the fluids is almost impossible. For example, using the experimental data of VOLFINGER et al., (1985, Table I, page 40) one could assume that the Cl- and K-richest amphiboles of Pont S. Martin marbles have grown in equilibrium with a H₂O-KCl binary fluid largely oversaturated respect to the KCl component; but, despite of careful microprobe analysis, no solid salt inclusions has been detected.

VOLFINGER and ROBERT (1986) already pointed out such discrepancies between available data on synthetic and natural minerals with high chlorine contents, stressing
the role of acidity and P-T conditions in changing properties of solutions. Furthermore, retrograde metamorphic reactions that hydrate solid phases are processes that concentrate salt components in the liquid phase (SANFORD, 1981; TROMMSDORFF and SKIPPEN, 1985, 1986).

The Cl(F)- and K(Na)-rich amphiboles of the marbles of the Sesia-Lanzo Zone have a rather wide compositional range, possibly reflecting a large chemical spectrum of fluid-rock interaction.

A qualitative estimation of the fluid composition can still be obtained using isobaric-isothermal plots of simplified systems (SKIPPEN, 1974). Thus, mixed volatile equilibria in the CaO-MgO-SiO₂-H₂O-CO₂ system has been calculated at the P-T conditions corresponding to the growth of the Cl-K-bearing amphiboles (Fig. 6a).

The related chemical potential diagram (Fig. 6b) allows to consider the salt component in a ternary fluid system (SKIPPEN and TROMMSDORFF, 1986), and also to draw the

Fig. 6. — a: Natural logarithms of the fugacities of H₂O and CO₂ at 500°C and 8 Kb. The solid curve represents the binary supercritical fluid in H₂O-CO₂, computed with the modified Redlich-Kwong equation of KERRICK and JACOBS (1981); the labels of the two dots on the curve indicate different molar fractions of carbon dioxide at Pₗₑᵥ = P(H₂O) + P(CO₂). The straight lines show the topology of mixed volatile equilibria in the system CaO-MgO-SiO₂-H₂O-CO₂, calculated using the thermodynamic data of PERKINS et al. (1986); (di: diopside, tr: tremolite, cc: calcite, do: dolomite, qz: quartz). The activity of calcite in equilibrium (1) has been corrected to plot it through the invariant point (SKIPPEN, 1974). b: CO₂ versus H₂O chemical potential diagram, at 500°C and 8 Kb, showing schematically phase elements in the H₂O-CO₂-KCl ternary system. Solid curve labels as in Fig. 6a. The dot along the H₂O-KCl binary corresponds to the saturation point calculated using the experimental data of CLARK (1959) and POTTER et al. (1975), modelled according to the H₂O-NaCl data of GUNTER et al. (1983). The pattern of the sylvite + liquid curve (dashed curve) is derived from that indicated at lower pressure by SKIPPEN and TROMMSDORFF (1986). The equilibrium here shown is that reported in text, and corresponds to equilibrium (1) in the simplified system of Fig. 6a. The heavy section of the straight line is the range of H₂O-CO₂ potentials discussed in text.
pattern of equilibrium (1) reported above. The lack of solid salt inclusions suggests that the composition of the fluid coexisting with the chlorpotassium ferro-pargasites was within the \( H_2O-CO_2 \) potential region along the modified equilibrium (1), between the curve of total pressure and the sylvite + liquid curve at 500°C and 8 Kb.

Conclusions

The occurrence of chlorpotassium ferro-pargasites and related potassic ferro-edenitic hornblendes in the Sesia-Lanzo Zone is, at the present, the first in the Alps. Cl- and K-bearing amphiboles grew during hydration reactions involving marbles of the internal sector of the Sesia-Lanzo Zone which suffered the high pressure-low temperature metamorphism of early-alpine age (Hunziker, 1977; Compagnoni et al. 1977).

Chemical analyses and X-ray refinements provide new data about the crystal-chemistry of pargasitic amphiboles when large substitution of halogens occurs, showing that the high iron content is responsible for the partitioning of chlorine in the solid phase in respect to the fluid.

The potassium substitution in the A site was supposed to take place with increasing pressure (Hinrichsen and Schurmann, 1977), but it is also strongly related to the increasing of the Fe/Mg ratio (Volfinger et al. 1985 and present data).

Like previous descriptions found in the literature (Leelanandam, 1969, Jacobson, 1975; Dick and Robinson, 1979; Sharma, 1981; Kamineni et al., 1982; Wanko, 1984, 1986) the Cl-K-bearing amphiboles occur in metamorphic rocks characterized by strong interaction of minerals with a multicomponent fluid.

Petrographical setting, the absence of solid salt inclusions and qualitative estimates in the \( CaO-MgO-SiO_2-H_2O-CO_2-KCl \) system, suggest that at 500°C and 8 Kb the composition of the ternary fluid coexisting with chlorpotassium ferro-pargasites plots in the region of undersaturated fluid, closer to the \( H_2O-KCl \) binary.

Further experimental investigations on the distribution of halogens in hydrous silicates and coexisting saline fluids at high temperatures and pressures are needed to provide better constraints on the composition of chloride brines in metamorphic rocks.

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