Gonnardite: Re-examination of holotype material and discreditation of tetranatrolite

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INTRODUCTION

Gonnardite was first proposed by Lacroix (1896) as a zeolite mineral found as radiated spherules in cavities of basalts from Gignat, Puy-de-Dôme, France, and named after the French mineralogist F. Gonnard. Successive investigation of both the type material (Hey 1932) and of new specimens from the type locality (Pongiluppi 1976; Nawaz 1988; Ross et al. 1992) have shown that the Gignat gonnardite is intimately associated with thomsonite and garronite. The gonnardite-thomsonite association is very common, although gonnardite can also be associated with phillipsite, chabazite, analcime, natrolite, or stilbite. Because pure gonnardite is uncommon, its crystal chemistry is confused, mainly because of biased chemical analyses carried out on polyphasic mixtures (Gottardi and Galli 1985). Chemical analyses of well-identified samples (Fig. 1: open circles), clearly show a broad distribution of the gonnardite samples along the natrolite-thomsonite compositional join. It is possible that several of the older analyses are biased by phase impurities or chemical inhomogeneities, nonetheless Figure 1 illustrates the wide chemical distribution of the mineral. The overall chemical trend corresponds to the partial Na4Si6 → Cu4Al2 thomsonite-like substitution in ideal natrolite (Nawaz 1988; Artioli and Torres Salvador 1991; Ross et al. 1992; Passaglia et al. 1992). The mineral described by the obsolete name of ranite (Mason 1957; Nawaz 1988) is here considered to be gonnardite.

The crystal structure of gonnardite has been much discussed. Structural studies on gonnardite have even been carried out on misidentified thomsonite crystals (Amirov et al. 1972). The crystal structure of a gonnardite sample from the Vevja Quarry, Tvedalen, Norway, was finally shown to be tetragonal I42d, and related to natrolite by Mazzi et al. (1986). Since then, the crystal structure of gonnardites from different localities has been refined from powder diffraction data (Artioli and Torres Salvador 1991). There is now little doubt that most gonnardites have a disordered natrolite framework and are not a polysomatic intergrowth of natrolite and thomsonite, as proposed in order to explain the chemical variability of the mineral (Ross et al. 1992). The role of Si,Al order in natrolite-group minerals was discussed by Alberti et al. (1995). The gonnardite atomic arrangement of framework atoms, Na cations, and water molecules in the channels is indistinguishable from the one reported for Ca-free tetranatrolite (Mikheeva et al. 1986; Rastsvetaeva 1995), and the chemical substitution and disorder of the Na and Ca cations in the cavities is reflected in the random presence in the structure of natrolite- and scolecite-like channels (Mazzi et al. 1986). The crystal chemical role of cation vacancies implied by the Na → Ca substitution has been discussed in detail by Ross et al. (1992).

The powder diffraction patterns of gonnardite and
tetranatrolite are essentially the same, except for small variations in the cell dimensions due to different chemical composition. Several authors in the past suggested that the two minerals have the same structure and they form a continuous compositional series (Mazzi et al. 1986; Nawaz 1988; Tschernich 1992; Alberti et al. 1995).

The present study was prompted by the lively discussion that took place during the work of the Subcommittee on Zeolite Mineral Nomenclature of the IMA Committee on New Minerals and Mineral Names. The discussion focused on the species status of gonnardite and tetranatrolite and left the definition of criteria for the distinction of the two mineral species unresolved. Tetranatrolite has been well characterized from the type and ideotype localities (Krogh Andersen et al. 1969; Chen and Chao 1980), but an adequate characterization of gonnardite from the type locality is lacking. The present study therefore re-investigates gonnardite samples from Gignat, France. The characterization involves specimens originally deposited at the British Museum by Lacroix and specimens collected by us from the type locality. We attempt to summarize the known chemical and crystallographic information on gonnardite and tetranatrolite to define the status of the two zeolite minerals.

**EXPERIMENTAL METHODS AND RESULTS**

Several zeolite-containing rock fragments were provided by A.M. Clark of the British Museum of Natural History from specimen no. BM1930-166 originally studied by Lacroix (1896) and by Hey (1932). Preliminary tests were performed by extracting very small amounts of zeolite filling material from each of the cavities in the basaltic rock. In two of the hand specimens only phillipsite, natrolite, and chabazite were identified by X-ray powder diffraction, whereas in a few cavities of the remaining fragments gonnardite was found in close association with thomsonite and natrolite.

In these gonnardite occurs as white aggregates of fine fibers, often as radiating spherules overlying a thin whitish crust of amorphous or clay material. Commonly, the core of the spherules is composed of acicular gonnardite, while the outer rim is composed of overgrown thomsonite. The occurrence, association, and morphology are in close agreement with previous observations (Lacroix 1896; Hey 1932; Pongiluppi 1976; Ross et al. 1992).

The gonnardite material of two separate spherules was hand separated under the optical microscope, and the thomsonite rim was removed. The X-ray powder diffraction spectrum however revealed that a substantial amount of thomsonite is still present in the sample, because of the intimate association of the two phases. The diffraction patterns of the two gonnardite samples (G-BM1 and G-BM2) are shown in Figure 2, together with the pattern of a gonnardite sample from the type locality collected by us (G-GA: specimen no. 16-10-22/3 of the Zeolite Collection, Dipartimento di Scienze della Terra, Università di Modena), and with the calculated pattern of pure gonnardite, simulated using the structure model of Mazzi et al. (1986).

The high level of thomsonite impurity did not allow a reliable structure refinement of gonnardite to be carried out on the British Museum samples. Our sample contains a much lower thomsonite content. It was extracted from rocks of the same type, color, and general appearance as the original specimens, and on the basis of careful optical and X-ray data, we deem that this sample is as representative of the type gonnardite as the British Museum samples. Previous work on specimens collected from the type locality was carried out by Pongiluppi (1976).

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Quantitative analysis performed on the powder diffraction patterns (Fig. 2) by Rietveld full-profile refinement yielded the following results: G-BM1 = gonnardite 75 wt% and thomsonite 25 wt%; G-BM2 = gonnardite 84 wt%, thomsonite 16 wt%; G-GA = gonnardite 96 wt% and thomsonite 4 wt%. The quantitative analysis was performed by keeping the structure models of both phases fixed, and by optimizing the instrumental and profile parameters. Uncertainties are below 1 wt%.
contamination. Table 1 lists the mean chemical composition averaged on 13 point analyses. Individual point analyses are reported in Figure 1 (gray circles). The water content was determined by thermogravimetric analysis on 29.38 mg of sample using a Seiko SSC/5200 operating at 10 °C/min in the temperature range 18–1000 °C in air. The measured mass loss was 14.19 wt%, which was used in the normalization of the electron probe microanalysis data (Table 1, Fig. 1).

The powder diffraction spectrum of the G-GA gonnardite sample was then used for the Rietveld refinement of the structure: the diffraction data were collected on a Philips goniometer in Bragg-Brentano geometry, using graphite monochromated CuKα radiation, a step scan mode with steps in 2θ of 0.02°, a total for angular 2θ range of 15–140°, and counting time of 12 seconds per step.

Starting atomic coordinates were from Mazzi et al. (1986). The instrumental background was modeled by a Chebyshev polynomial with three coefficients, and the Bragg peak profiles were modeled by a pseudo-Voigt function corrected for asymmetry. Thomsonite was inserted as a second phase during the refinement, using the fixed structure model of Alberti et al. (1981). All computations have been performed using the GSAS program package (Larson and Von Dreele 1997).

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**Table 1.** Mean chemical composition of Gignat gonnardite

<table>
<thead>
<tr>
<th>element</th>
<th>oxide wt%</th>
<th>atoms p.f.u.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>43.17</td>
<td>11.499</td>
</tr>
<tr>
<td>Al</td>
<td>27.37</td>
<td>8.592</td>
</tr>
<tr>
<td>Fe³⁺</td>
<td>0.03</td>
<td>0.006</td>
</tr>
<tr>
<td>Na</td>
<td>8.73</td>
<td>4.508</td>
</tr>
<tr>
<td>Mg</td>
<td>0.01</td>
<td>0.007</td>
</tr>
<tr>
<td>K</td>
<td>0.02</td>
<td>0.004</td>
</tr>
<tr>
<td>Ca</td>
<td>6.45</td>
<td>1.841</td>
</tr>
<tr>
<td>Sr</td>
<td>0.01</td>
<td>0.002</td>
</tr>
<tr>
<td>Ba</td>
<td>0.02</td>
<td>0.002</td>
</tr>
<tr>
<td>H₂O</td>
<td>14.19</td>
<td>12.606</td>
</tr>
<tr>
<td>sum</td>
<td>100.00</td>
<td></td>
</tr>
</tbody>
</table>

Notes: Averaged on the 13 point analyses reported in Figure 1. Unit formula based on 40 oxygen atoms.

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**Figure 2.** X-ray powder diffraction patterns of gonnardite. (a) calculated, pure gonnardite; (b), (c), and (d) measured, samples G-GA, G-BM1, and G-BM2 respectively. Stars indicate the Bragg peaks due to the thomsonite impurity.

Chemical analyses were performed on several small fragments of the spherules by electron probe microanalysis, using an ARL-SEMQ instrument, operated in wavelength dispersive mode at 15 kV, with a 10 nA sample current. The standards used were natural albite (Si, Al, Na), microcline (K), paracelsian (Ba), synthetic An₇₀ glass (Ca), Sr-anorthite (Sr), P140 olivine (Fe), Di₈₅-Jd₁₅ (Mg), and data acquisition and processing by the PROBE program (Donovan 1995). Each fragment was checked for purity by X-ray diffraction using a Gandolfi camera prior to the EPMA analysis, in order to exclude thomsonite contamination.
The refined atomic coordinates for gonnardite in space group $I4_1d$ are listed in Table 2. Refined cell parameters are $a = 13.2670(4)$ and $c = 6.6023(6)$. Final agreement indices are $R_w = 0.135$, $R_p = 0.101$, $\chi^2 = 18.6$ for 46 variables; Figure 3 shows the profiles resulting from the Rietveld refinement.

**DISCUSSION**

Although the Rietveld refinement of gonnardite from a slightly impure sample is of lower quality than the one originally performed with single crystal data, the results unambiguously show that the Gignat gonnardite can be adequately described using the structure model proposed by Mazzi et al. (1986) for a gonnardite sample from Norway. The framework has the NAT topology common to natrolite, mesolite, scolecite, and tetratrolite. The tetragonal symmetry is essentially due to the disordered Al, Si distribution over the framework tetrahedral sites, as long demonstrated by the relationship between framework disorder and deviation from tetragonal lattice in natrolite (Alberti and Vezzalini 1981; Alberti et al. 1995). Although in natrolite a partial Al, Si disorder in two out of the three independent tetrahedral sites only reduces the difference between the $a$ and $b$ cell parameters, in gonnardite and tetratrolite the average tetragonal symmetry is stabilized by (1) the statistical distribution of the cations and water molecules in the zeolitic channels, (2) the excess tetrahedral Al present in both frameworks, and (3) complete Al, Si disorder in all of the tetrahedral sites. Natrolite-type cation ordering in the tetrahedral sites is not possible for minerals having Si/Al < 1.5 on the basis of the Loewenstein’s avoidance rule (Alberti and Gottardi 1975; Alberti et al. 1995).

We would like to re-emphasize that gonnardite and tetratrolite, having the same space group, cannot be distinguished by analysis of crystallographic symmetry, cell parameters, or differences in their diffraction patterns.

The cations and water molecules found in the cavities of the Gignat gonnardite closely follow the arrangement proposed by Mazzi et al. (1986): there is one major cation position, which implies short-range order of the Na and Ca atoms along the channels, and statistical distribution over the crystal volume of natrolite (or tetratrolite)-like environments for the Na cations, and of scolecite-like environments for the Ca atoms. The presence of Ca in a channel implies that half of the cation sites are vacant, and an additional water molecule site is present in order to complete the Ca hydration sphere. The cation site was modeled by fixing the Ca content derived from the chemical analysis, and by freely refining the remaining site scattering power as Na atoms. Given the correlation between site occupancy factors and atomic displacement parameters, the resulting amount of Na (4.6 ± 0.2 atoms pfu) is in excellent agreement with the Na content derived from the chemical analyses (4.51 atoms pfu). Although a slight disorder in the cation positions has been shown to be possible in gonnardite (Artioli and Torres Salvador 1991), no evidence of split cation sites was found in the Gignat gonnardite. The total refined content of water molecules (13.6 ± 0.3 molecules pfu) is in reasonable agreement with the hydration content derived from thermal analysis (12.61 molecules).

**On the status of gonnardite and tetratrolite**

Under the current IMA rules, two mineral species can be distinct on the basis of their crystal structural and/or their crystal chemical differences. As discussed above, the present work and the published gonnardite and tetratrolite structure refinements clearly show that the two compounds have the same structure, apart from the small changes in the cation sites due to the slightly different chemical content. There is really no reliable crystallographic parameter useful for proper discrimination.

As far as the crystal chemistry is concerned, if we accept that tetratrolite is not just a disordered natrolite, but it can accept a substantial amount of Ca, Al substitution (Nawaz 1988; Ross et al. 1992), then the main criterion for distinction is based on the fact that tetratrolite minerals seem to show a different Ca, Al substitutional trend with respect to gonnardites (Ross et al. 1992).

As proposed by Ross et al. (1992) gonnardite minerals lie on the natrolite-thomsonite join, implying a Na$_2$Si$_2$ → Ca$_3$Al$_2$ chemical substitution (Fig. 1: NAT-THO join), while the tetratrolite minerals lie on the natrolite-“ideal tetratrolite” join, implying a Na$_4$Si$_2$ → Ca$_4$Al$_2$ chemical substitution (Fig. 1: NAT-TET join). It is clear that the two different substitution processes imply a different amount of cation vacancies in the channels, as discussed in detail by Ross et al. (1992). The tetratrolite points 8–12 in Figure 1 (corresponding to the points in Fig. 5 of Ross et al. 1992) however, only represent the Ca-rich tetratrolite minerals from Mont St. Hilaire and Magnet Cove analyzed by Ross and coworkers (Ross et al. 1992). If we insert in a single diagram all available chemical analyses of tetratrolite and gonnardite minerals (Fig. 1), it is hardly possible to distinguish well-defined chemical trends. In fact, published analyses of tetratrolite and gonnardite samples exist which are perfectly identical. For example, the chemical composition of the gonnardite from Tvedalen, Norway (Mazzi et al. 1986), is indistinguishable from tetratrolite 86-53A from Magnet Cove, Arkansas (analysis no. 3, Table 5: Ross et al. 1992); tetratrolite from Mont St. Hilaire, Canada (Chen and Chao 1980), is indistinguishable from gonnardite 85-16A from Magnet Cove, Arkansas (analysis no. 10, Table 3: Ross et al. 1992); tetratrolite from Lamo, Norway (Nawaz 1988), is identical to gonnardite 158913 from Kloh, Austria (analysis no. 6, Table 3: Ross et al. 1992); and gonnardite 85-16A from Magnet Cove, Arkansas (analysis no. 9, Table 3: Ross et al. 1992), is identical to tetratrolites from Kloh, Austria (Nawaz 1988), and from Gulacs Hill, Hungary (Alberti et al. 1982). Interestingly, even the type locality tetratrolite (Chen and Chao 1980) is compositionally closer to the gonnardite trend than to the so called “tetratrolite” trend. We therefore conclude the ana-

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**Table 2. Rietveld refined atomic parameters for Gignat gonnardite**

<table>
<thead>
<tr>
<th>atom</th>
<th>$x$</th>
<th>$y$</th>
<th>$z$</th>
<th>occupancy</th>
<th>$U_{iso}$ (Å$^2$)</th>
</tr>
</thead>
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<tr>
<td>T1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1.0</td>
<td>0.025(3)</td>
</tr>
<tr>
<td>T2</td>
<td>0.1319(3)</td>
<td>0.0512(3)</td>
<td>0.3845(8)</td>
<td>1.0</td>
<td>0.029(1)</td>
</tr>
<tr>
<td>O1</td>
<td>0.25</td>
<td>0.0949(7)</td>
<td>0.375</td>
<td>1.0</td>
<td>0.031(4)</td>
</tr>
<tr>
<td>O2</td>
<td>0.0903(5)</td>
<td>0.4478(5)</td>
<td>0.101</td>
<td>1.0</td>
<td>0.029(3)</td>
</tr>
<tr>
<td>O3</td>
<td>0.4373(5)</td>
<td>0.3721(5)</td>
<td>0.026</td>
<td>1.0</td>
<td>0.027(3)</td>
</tr>
<tr>
<td>Ca</td>
<td>0.3005(5)</td>
<td>0.25</td>
<td>0.125</td>
<td>0.58(2)</td>
<td>0.013(4)</td>
</tr>
<tr>
<td>Na</td>
<td>0.3005(5)</td>
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<td>0.125</td>
<td>0.23</td>
<td>0.013(4)</td>
</tr>
<tr>
<td>OW1</td>
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<td>0.3651(9)</td>
<td>0.375</td>
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<td>OW2</td>
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<td>0.235(2)</td>
<td>0.051(5)</td>
<td>0.35(2)</td>
<td>0.037(5)</td>
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lytical data as a whole do not show any distinct tetrNatrolite trend, but rather a wide distribution of compositions on both sides of the ideal natrolite-thomsonite join. The data also do not support the arbitrary distinction of compositional fields as proposed by Nawaz (1988). We conclude that the two proposed mechanisms for chemical substitution do not act independently, but rather each gonnardite sample is the result of a different degree of coupling between the two substitutional processes. Of course, this also implies a variable number of cationvacancies and water molecules in the channels.

A different discrimination method between tetrNatrolite and gonnardite was proposed by Alberti et al. (1995) on the basis of the Si/Al ratio in the framework: tetrNatrolite must have tetragonal symmetry and Si/Al ≥ 1.5. In such a case, natrolitetype order in the tetrahedral sites is possible in principle, and the framework disorder is entirely due to the peculiar genetic process, possibly related to the dehydration of paranatrolite (Chao 1980). On the basis of the combined crystallographic-chemical parameters, only four samples could be considered true tetrNatrolites: the minerals from Ilmaussaq, Gronland (Krogh Andersen et al. 1969), Mont St. Hilaire, Canada (Chen and Chao 1980), Lovozero, Russia (Guseva et al. 1975), and Khibiny, Russia (Kiseleva et al. 1997). These samples (nos. 1, 3, 2, and 13 respectively) are those lying above the natrolite-scolecite join in Figure 1.

Keeping in mind that the genetic process cannot be taken as a basis for species discrimination and identification purposes, we note that by adopting the discrimination parameters proposed by Alberti et al. (1995), it is not possible to characterize gonnardite or tetrNatrolite without appropriate analysis of both crystal symmetry and framework chemistry.

Two further points need to be considered in the discussion about the status of gonnardite and tetrNatrolite as separate minerals species. First, following the recent recommendation of the Subcommittee on the Nomenclature of Natural Zeolites (Coombs 1997), a substantial range in composition in a zeolite does not warrant the status of different mineral species, unless accompanied by change in topologically distinctive tetrahedral framework (Rule 1a). Zeolites having the same topologically distinctive tetrahedral framework constitute a series when they display a substantial range in composition (Rule 1b).

Based on the evidence here and on the recently proposed rules, we suggest that almost all gonnardite and tetrNatrolite minerals reported in the literature can be considered the same mineral. Possible exceptions are the four samples discussed above and having tetragonal symmetry and framework Si/Al ≥ 1.5. Although it is advised that two mineral species are not distinguished on the basis of framework Si/Al ratio (Rule 3), an exception has already been made in the case of clinoptilolite-heulandite (Coombs 1997), and tetrNatrolite-gonnardite would constitute a further exception.

We here prefer close adherence to the proposed rules, and suggest that no arbitrary chemical distinction is made between tetrNatrolite and gonnardite. Because of priority, the gonnardite name should be retained for all minerals exhibiting tetragonal I42d symmetry and the natrolite-type framework, and the tetrNatrolite name should be discredited. Given the wide chemical composition range, and under the novel recommendations of the Subcommittee on Zeolite Mineral Nomenclature (Coombs 1997), gonnardite ought to be regarded as a compositional series (Rule 1b). Individual species in a zeolite mineral series with varying extraframework cations are to be named by attaching to the series name a suffix indicating the chemical symbol for the extraframework element that is the most abundant in atomic proportions (Rule 5). All reported gonnardite analyses show Na as the prevalent cation, and should be named gonnardite-Na. It is at present doubtful that gonnardite could possibly have Ca as the prevailing cation, and therefore the gonnardite-Ca end-member exist, because a mineral with such a composition will likely exhibit the thomsonite framework. However it is possible in principle to find a gonnardite having near equal proportions of Na and Ca cations.

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REFERENCES CITED


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