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# JARLITE: ORDER–DISORDER STRUCTURE SUGGESTED BY OPTICS, RESULTS OF NEW ANALYSES AND EMPIRICAL FORMULAE

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## ABSTRACT

Results of electron-microprobe analyses, X-ray data and density determinations of jarlite and "meta-jarlite" from Ivigtut, Greenland, are presented. Common jarlite, in crystals and grains, is composite; it has a core with straight boundaries and a mantle. The core shows a $2V_\alpha$ of $\leq 40°$; the mantle, in optical continuity with the core, shows a $2V_\alpha$ of approximately 80°. The core is rich in Sr and Ba, but nearly devoid of Mg and K. In the mantle, Mg and K are present in significant amounts. "Meta-jarlite", always developed as dendrites, is chemically similar to the mantle jarlite. Jarlite that is not composite, from the stenonite paragenesis, with $2V_\alpha$ between 5 and 10°, has a composition similar to that of jarlite in the core. Empirical formulae calculated from the new analytical data on the basis of 68 anions give 30 cations, in agreement with the nominal formula: $Na_2(Sr_{12}Na_2)Mg_2Al_{12}F_{64}(OH)_4$. The X-ray data, density determinations and calculated molecular weights support a formula based on 68 anions, but the calculated formula content cannot be accommodated in the structure in accordance with the model proposed. The empirical formula for jarlite in the core may be written $Na_2(Sr,Ba,Na)_{16}Al_{12}F_{68}$, assuming disorder between Mg and Sr sites. The empirical formulae for jarlite mantle and "meta-jarlite" show 1.5 to 2 atoms of Mg, and 14.5 to 15 atoms for the Sr sites. The core-mantle arrangement of jarlite and the different compositions suggest a shift during crystallization, from a disordered toward an ordered structure. The jarlite from the stenonite paragenesis seems to represent the disordered structure only.

*Keywords*: jarlite, electron-microprobe analyses, X-ray data, density, Sr aluminofluoride, empirical formulae, order–disorder structure, Ivigtut, Greenland.

## SOMMAIRE

Nous présentons les résultats d'analyses à la microsonde électronique, des données de diffraction rayons X et des déterminations de densité de la jarlite et de la "méta-jarlite" de Ivigtut, au Groënland. La jarlite commune, en cristaux et en grains xénomorphes, est composite, ayant un coeur idiomorphe entouré d'une bordure. Le coeur possède un $2V_\alpha \leq 40°$; la gaine, en continuïté optique avec le coeur, montre un $2V_\alpha$ d'environ 80°. Le coeur est riche en Sr et Ba, et quasiment sans Mg et K. Dans la bordure, Mg et K sont présents en quantités importantes. La "méta-jarlite", toujours sous forme de dendrites, est chimiquement semblable à la jarlite de la gaine. Une jarlite homogène fait partie de la paragenèse de la sténonite; elle possède un $2V_\alpha$ entre 5 et 10°, et une composition semblable à la jarlite du coeur. Les formules empiriques recalculées à partir des données nouvelles sur une base de 68 anions mènent à 30 cations, ce qui concorde avec la formule idéale: $Na_2(Sr_{12}Na_2)Mg_2Al_{12}F_{64}(OH)_4$. Les données de diffraction X et de densité, ainsi que les poids moléculaires calculés à partir des compositions, concordent avec une base de 68 anions, mais la formule calculée ne peut pas être accommodée dans une structure selon le modèle proposé. La formule empirique de la jarlite du coeur serait $Na_2(Sr,Ba,Na)_{16}Al_{12}F_{68}$ si nous supposons l'existence d'un désordre impliquant les sites Mg et Sr. La formule empirique de la jarlite de la bordure et de la "méta-jarlite" montre entre 1.5 et 2 atomes de Mg, et entre 14.5 et 15 atomes dans les sites du Sr. L'agencement coeur–bordure de la jarlite et les compositions distinctes font penser qu'il y a eu changement, lors de la cristallisation, d'une forme précoce désordonnée à une forme tardive plus ordonnée. Par contre, la jarlite de la paragenèse de la sténonite semble avoir cristallisé avec la structure désordonnée seulement.

(Traduit par la Rédaction)

*Mots-clés*: jarlite, analyses à la microsonde électronique, données de diffraction X, densité, aluminofluorure de Sr, formule empirique, ordre–désordre structural, Ivigtut, Groënland.

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## INTRODUCTION

Jarlite from the Ivigtut cryolite deposit (Bøgvad 1933) shows serious discrepancies among the formula assigned to it, the observed specific gravity, and the density calculated from X-ray data (Brosset 1942, Ferguson 1949). New analyses were carried out (Bøgvad 1951), but never published. Hawthorne (1983) published a study of the structure of jarlite based on samples collected in Ivigtut in 1942 (F.C. Hawthorne, pers. comm.) and used by Ferguson (1949). Owing to the pattern of mining of the deposit at that time, it is reasonable to assume that these samples came from the localities where Bøgvad obtained his original samples. Hawthorne (1983) proposed a new formula for jarlite: $Na_2(Sr, Na, \square)_{14}Al_{12}(Mg, \square)_2 F_{64}(OH,H_2O)_4$, with some additional replacement of Sr by Ca, Ba and K. In setting up the new formula, the chemical data of Bøgvad (1933) were supplemented with qualitative electron-microprobe data.

Following the discovery of the mineral acuminite, $SrAlF_4(OH)H_2O$ (Pauly & Petersen 1987), a closer investigation of the jarlite-bearing aggregates was undertaken, because acuminite occurs in such an aggregate. This led to the discovery of yet another new mineral: bøgvadite, $Na_2SrBa_2Al_4F_{20}$ (Pauly & Petersen 1988). The composition of these minerals inspired renewed work on jarlite utilizing the electron microprobe.

## OCCURRENCE OF JARLITE FROM IVIGTUT

Jarlite from Ivigtut was found in columnar aggregates, which form fissure fillings in the cryolite mass (Bøgvad 1933). The material at hand includes many of Bøgvad's samples and contains a number of specimens appearing as radiating aggregates. It was in a jarlite-bearing sample of the latter type that the mineral bøgvadite was first observed (Pauly & Petersen 1988).

The lamellae forming the columns and the radii are up to a few millimeters wide and may stretch across the decimeter-sized samples. They are delineated by seams, a few tenths of a millimeter wide, of cryptocrystalline topaz and fine-grained, spherulitic muscovite. The major constituent of the lamellae is jarlite, present in elongate, curved to fan-shaped grains. In thin section, viewed between crossed polars, jarlite may show a brush-like or wavy extinction reminiscent of spherulitic behavior. Strontian barite and, locally, bøgvadite occur scattered in the jarlite. Fluorite, 10–15%, and pyrite, rarely above 1%, also are present in the aggregates.

Crystal-lined cavities are particularly abundant in the outer parts of the radiating aggregates. The jarlite of the aggregates continues into the cavities, forming groups of fan-shaped crystals usually flattened on (100). Crystals of thomsenolite, and commonly also of ralstonite, are present in the cavities.

The present study has shown that the jarlite in lamellae as well as present as crystals is composite: in thin sections viewed between crossed polars, one generally observes a central area, a core, separated from the outer part, the mantle, of the grain or the crystal, by sharp, straight boundaries. Properly oriented sections reveal that the core of the jarlite has a small optic angle: $2V_\alpha \leq 40°$; grains showing a $2V_\alpha$ between 10 and 20° are commonly observed. The mantle part of the grain is in optical continuity with the core part; it shows the large optic angle usually attributed to jarlite ($2V_\alpha \approx 80°$). Bøgvad (1933) mentioned the observation of $2V_\alpha = 41–43°$ occasionally ascertained in crystals, but he did not investigate it further. Figures 1 and 2 show composite grains of jarlite and crystals as seen in secondary electron images on the microprobe. The core appears distinctly lighter grey than the mantle of the jarlite grains and crystals. The two are here treated separately because the mineral was originally established on jarlite crystals only (Bøgvad 1933).

Jarlite found together with stenonite (Pauly 1962) is rather coarse grained, up to 10 mm across. It differs from the common jarlite in showing sharp, well-defined extinction between crossed polars. Grains, not composite, with small optic angles, $2V_\alpha = 5 - 10°$, are commonly observed. Jarlite bordering cavities and crystals of jarlite protruding into the cavities are composite, and the mantle parts show the usual, large $2V_\alpha$ ($\approx 80°$).

"Meta-jarlite" (Bøgvad 1933), a variety of jarlite (Ferguson 1949), was found as segregations in chiolite. It also was found in vein-like formations (Bøgvad 1951), up to 1 m long and 2-3 cm wide, squeezed in between coarse siderite and surrounding cryolite. In samples from this milieu, the "meta-jarlite" appears as greyish, botryoidal masses, in other samples as white, dense masses. This type of "meta-jarlite" occurs in the part of the quarry where stenonite was found. Scattered, small grains of stenonite are present in some of the samples of white, dense "meta-jarlite". In thin sections, "meta-jarlite" is always found developed as dendrites. In the vein-like "meta-jarlite", the dendrites stretch across the whole width of the vein. Scattered, thin veins of cryolite transect these otherwise rather pure masses of interfingering dendrites.

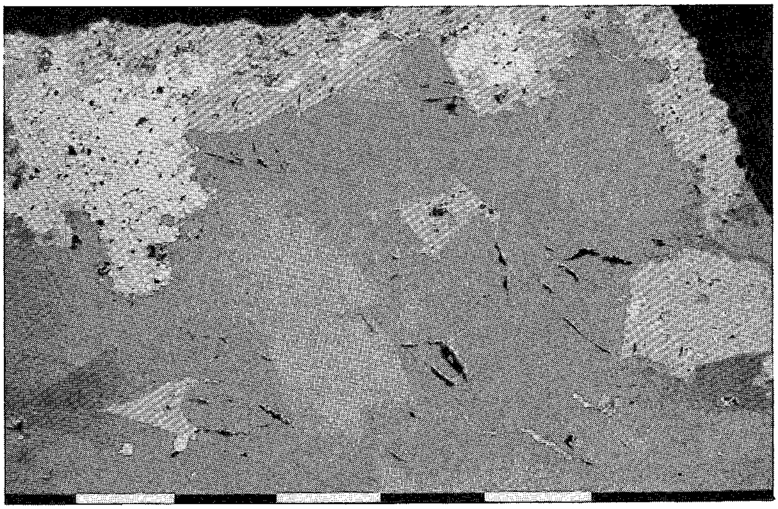

FIG. 1. SEM image: bøgvadite (light grey) with barite (white), jarlite core (grey) and jarlite mantle (dark grey). Bar = 0.1 mm.

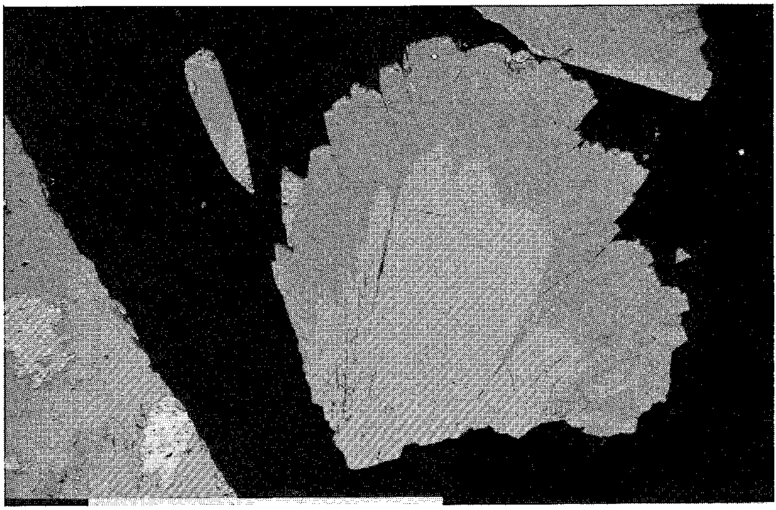

FIG. 2. SEM image: fan-shaped aggregate of jarlite crystals. Jarlite core light grey, jarlite mantle dark grey. Bar = 1 mm.

## SAMPLE SELECTION AND ANALYTICAL PROCEDURE

The samples used in the present electron-microprobe study were selected from the many hundred jarlite-bearing samples collected since 1932 in Ivigtut and at the dressing plant in Copenhagen. Doubly polished thin sections were made for microscopy in transmitted and reflected light. The latter, not marred by internal reflections, allowed observations for instance of barite and jarlite; also, they helped in the selection of areas for the following work. In the electron-microprobe examination, energy-dispersion spectra were used for qualitative characterization of jarlite and the other minerals present. Such characterization led to the final selection of targets for the quantitative analyses. These were carried out on a Philips model 505 scanning electron microscope equipped with a Microspec WDX 2–A spectrometer operating at 12 kV. The data were corrected with the NBS COR 2 Program (Henoc *et al.* 1973). The following standards were used: weberite (Na, Mg, Al), orthoclase, variety adularia (K), wollastonite (Ca), bøggildite (Pauly 1956) (Sr) and barite (Ba).

Analyses for F gave reasonable values, but because the recalculation program used could not accept determination of F simultaneously with the other elements, the analyses were carried out assuming all metals to be bound to F; in other words, the proportion of F was calculated. This shortcoming is presumably due to inappropriate absorption corrections for F.

The fluorides used as standards were checked mutually and against cryolite, chiolite and oxide standards. This step revealed that weberite contains 19.81% Na instead of 19.08%, and 0.28% K instead of 1.19% as given in Bøgvad (1938). The new percentage for Na was used when weberite was used as a standard. Incidentally, chiolite was found to contain 0.34% K; Stepanov (1963) had reported 0.35%.

Elements other than those mentioned were not detected; rare-earth elements, C (in carbonate) and S (in sulfate) were sought, in particular, but not detected.

## ANALYSES OF JARLITE AND "META-JARLITE"

### *Wet-chemical analyses*

The composition of jarlite crystals and "meta-jarlite" from chiolite (Bøgvad 1933) is given in Table 1, together with the repeated analytical results referred to by Bøgvad (1951), but hitherto not published; two unpublished results of analyses of white, dense "meta-jarlite" also are included in Table 1.

TABLE 1. COMPOSITION OF JARLITE FROM THE CRYOLITE DEPOSIT IVIGTUT, SOUTH GREENLAND, BY WET-CHEMICAL METHODS

| | Jarlite crystals | | "Meta-jarlite" from chiolite | | "Meta-jarlite" white, dense | |
|---|---|---|---|---|---|---|
| | A | B | C | D | E | F |
| Na | 3.23 | 3.08 | 3.54$^x$ | 4.41$^x$ | 3.27$^x$ | 3.52$^x$ |
| K | - | 0.85 | - | 0.69 | 0.55 | - |
| Li | 0.08 | bd | 0.04 | 0.07 | - | 0.009 |
| Mg | 0.90 | 0.77 | 1.38 | 1.23 | 1.26 | 1.28 |
| Ca | 0.55 | 0.18 | 3.20 | 3.09 | 0.22 | 2.22 |
| Sr | 35.60 | 37.81 | 28.70 | 29.85 | 34.00 | 32.99 |
| Ba | 0.99 | 2.88 | 2.25 | 2.01 | 5.51 | 3.54 |
| Al | 12.16 | 11.21 | 12.49 | 12.23 | 10.82 | 11.39 |
| Fe | 0.17 | - | 0.31 | - | - | - |
| MnO | - | - | - | 0.08 | - | - |
| F | 43.23 | 41.46 | 45.50 | 44.25 | 42.03 | (46.37)$^o$ |
| OH | 2.91 | 1.76 | 2.14 | 2.16 | 1.70 | - |
| Sum | 99.82 | 100.00$^+$ | 99.55 | 100.07 | 99.36 | |

$^+$Recalculated to 100% after deduction of 0.5% insoluble and 1% thomsenolite visually estimated to be present in the fraction prepared for analysis.

$^x$Na and Al too high due to contamination by cryolite.

$^o$F calculated. bd = below detection.
A & C from Bøgvad (1933), R.Blix analyst. B & D referred to in Bøgvad (1951), not published before, A.H.Nielsen analyst. E: E.-L.Mortensen analyst. F: Al determined by titration with CDTA, the other metals by atomic absorption spectrophotometry, Sinh Hy Nguyen analyst.

The differences between the composition of jarlite crystals (Bøgvad 1933) and that referred to by Bøgvad (1951), may be related to the procedures of heavy-medium separation used. Methylene iodide separation followed by treatment in Clerici solution was used by Bøgvad (1933), whereas the material for the repeated analyses was treated in methylene iodide only. The analytical data are of limited value because they do not distinguish between jarlite core and mantle (see below).

The analytical results of "meta-jarlite" from chiolite, 1933 *versus* 1951, are rather similar. They differ clearly from the compositions of the white, dense "meta-jarlite". In all four cases, Na is too high owing to contamination with cryolite. It is estimated that between 1 and 3% cryolite is present. Such amounts of cryolite may easily escape observations in the microscope if they occur as micrometric shards on edges and corners of the grains in the analytical fractions.

### *Electron-microprobe analyses*

Tables 2 and 3 show the results of the electron-microprobe analyses of jarlite and "meta-jarlite". The analyses of the core and the mantle

TABLE 2. ELECTRON-MICROPROBE DATA ON JARLITE FROM IVIGTUT CRYOLITE DEPOSIT, SOUTH GREENLAND

| | Jarlite crystals | | | | Jarlite from columnar aggregates | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | sample I | | sample II | | | |
| | core | | mantle | | core | mantle | core | | mantle | |
| | $X_4$ | s | $X_7$ | s | $X_2$ | $X_1$ | $X_6$ | s | $X_5$ | s |
| Na | 2.99 | 0.10 | 2.56 | 0.13 | 2.87 | 3.03 | 2.65 | 0.31 | 2.56 | 0.31 |
| K | 0.20 | 0.07 | 1.21 | 0.13 | 0.32 | 0.82 | 0.12 | 0.06 | 1.06 | 0.37 |
| Mg | 0.06 | 0.04 | 1.36 | 0.13 | 0.25 | 1.37 | 0.02 | 0.01 | 1.63 | 0.22 |
| Ca | 0.14 | 0.03 | 0.51 | 0.23 | 0.18 | 0.40 | 0.12 | 0.04 | 0.67 | 0.26 |
| Sr | 39.87 | 0.40 | 37.44 | 0.88 | 40.35 | 37.17 | 40.42 | 1.86 | 36.53 | 0.77 |
| Ba | 2.65 | 0.26 | 0.80 | 0.53 | 2.18 | 0.96 | 2.45 | 1.16 | 0.87 | 0.31 |
| Al | 10.72 | 0.25 | 11.05 | 0.22 | 10.49 | 11.07 | 10.80 | 0.24 | 11.17 | 0.35 |
| F$^+$ | 43.38 | 0.28 | 45.07 | 0.34 | 43.36 | 45.19 | 43.42 | 0.25 | 45.51 | 0.56 |
| $Z_{nn}$ | 107.- | | 105.- | | 108.- | 104.- | 91.- | | 88.- | |

$^+$F calculated; the analyses are normalized to 100%. $Z_{nn}$ = not normalized sum.
$X_n$ = average of n analyses; s = standard deviation.

Empirical formulae calculated from the analyses assuming 68 anions (Hawthorne 1983):

Jarlite crystals, core molecular wt. 2978
$Na_{3.87}(Sr_{13.55}Ba_{0.57}Ca_{0.10}K_{0.15})_{\Sigma 14.37}Mg_{0.07}Al_{11.83}F_{68}$
Jarlite crystals, mantle molecular wt. 2867
$Na_{3.19}(Sr_{12.25}Ba_{0.17}Ca_{0.36}K_{0.89})_{\Sigma 13.67}Mg_{1.60}Al_{11.74}F_{68}$

Jarlite sample I, core molecular wt. 2979
$Na_{3.72}(Sr_{13.72}Ba_{0.47}Ca_{0.13}K_{0.24})_{\Sigma 14.56}Mg_{0.31}Al_{11.59}F_{68}$
Jarlite sample, mantle molecular wt. 2860
$Na_{3.77}(Sr_{12.13}Ba_{0.20}Ca_{0.29}K_{0.60})_{\Sigma 13.22}Mg_{1.61}Al_{11.73}F_{68}$

Jarlite sample II, core molecular wt. 2976
$Na_{3.43}(Sr_{13.73}Ba_{0.53}Ca_{0.09}K_{0.09})_{\Sigma 14.44}Mg_{0.02}Al_{11.91}F_{68}$
Jarlite sample II, mantle molecular wt. 2839
$Na_{3.16}(Sr_{11.84}Ba_{0.18}Ca_{0.47}K_{0.77})_{\Sigma 13.26}Mg_{1.90}Al_{11.75}F_{68}$

TABLE 3. ELECTRON-MICROPROBE DATA ON "META-JARLITE" SAMPLES I,II,III AND JARLITE FROM THE STENONITE PARAGENESIS IVIGTUT CRYOLITE DEPOSIT, SOUTH GREENLAND

| | "Meta-jarlite" | | | | | | | Jarlite | |
|---|---|---|---|---|---|---|---|---|---|
| | from chiolite | | white, dense | | | | botry-oidal | from stenonite paragen. | |
| | | | I | | II | III | | | |
| | $X_{16}$ | s | $X_{11}$ | s | $X_2$ | $X_2$ | $X_2$ | $X_{11}$ | s |
| Na | 3.28 | 0.14 | 3.04 | 0.12 | 2.89 | 3.05 | 2.97 | 3.25 | 0.06 |
| K | 0.49 | 0.11 | 0.55 | 0.09 | 0.50 | 0.62 | 0.62 | 0.30 | 0.04 |
| Mg | 1.28 | 0.15 | 1.04 | 0.08 | 1.57 | 1.45 | 1.68 | 0.38 | 0.07 |
| Ca | 2.85 | 0.29 | 0.44 | 0.05 | 0.73 | 1.15 | 1.10 | 0.28 | 0.07 |
| Sr | 31.05 | 0.85 | 33.69 | 0.47 | 34.27 | 33.92 | 34.61 | 32.76 | 0.56 |
| Ba | 2.58 | 0.77 | 5.46 | 0.46 | 5.09 | 2.56 | 2.38 | 8.63 | 0.60 |
| Al | 11.77 | 0.16 | 11.19 | 0.13 | 10.58 | 11.46 | 11.10 | 10.97 | 0.18 |
| $F^+$ | 46.70 | 0.26 | 44.59 | 0.19 | 44.37 | 45.79 | 45.54 | 43.44 | 0.26 |
| $Z_{nn}$ | 99.- | | 96.- | | 103.- | 95.- | 89.- | 98.- | |

$^+$F calculated; the analyses are normalized to 100%. $Z_{nn}$ = not normalized sum.

$X_n$ = average of n analyses; s = standard deviation.

Empirical formulae calculated from the analyses assuming 68 anions (Hawthorne 1983). NOTE: formulae are arranged according to molecular weight.

"Meta-jarlite from chiolite molecular wt. 2767

$Na_{3.95}(Sr_{9.80}Ba_{0.52}Ca_{1.97}K_{0.35})_{\Sigma 12.64}Mg_{1.46}Al_{12.07}F_{68}$

"Meta-jarlite" white, dense III molecular wt. 2821

$Na_{3.74}(Sr_{10.92}Ba_{0.52}Ca_{0.81}K_{0.45})_{\Sigma 12.70}Mg_{1.68}Al_{11.99}F_{68}$

"Meta-jarlite" botryoidal molecular wt. 2837

$Na_{3.67}(Sr_{11.21}Ba_{0.49}Ca_{0.78}K_{0.45})_{\Sigma 12.93}Mg_{1.96}Al_{11.67}F_{68}$

"Meta-jarlite" white, dense I molecular wt. 2898

$Na_{3.83}(Sr_{11.14}Ba_{1.15}Ca_{0.32}K_{0.41})_{\Sigma 13.02}Mg_{1.24}Al_{12.02}F_{68}$

"Meta-jarlite" white, dense II molecular wt. 2912

$Na_{3.66}(Sr_{11.39}Ba_{1.08}Ca_{0.53}K_{0.37})_{\Sigma 13.37}Mg_{1.88}Al_{11.42}F_{68}$

Jarlite, stenonite paragenesis molecular wt. 2977

$Na_{4.21}(Sr_{11.12}Ba_{1.87}Ca_{0.21}K_{0.23})_{\Sigma 13.43}Mg_{0.46}Al_{12.09}F_{68}$

TABLE 4. PHYSICAL PROPERTIES AND GLADSTONE-DALE CONSTANTS OF "META-JARLITE" AND JARLITE

| | "Meta-jarlite" from chiolite A | | Jarlite from the stenonite paragenesis B | | Jarlite core sample II C | |
|---|---|---|---|---|---|---|
| a (Å) | 15.826 | (1) | 16.0460 | (9) | 16.039 | (2) |
| b (Å) | 10.795 | (1) | 10.9708 | (7) | 10.915 | (1) |
| c (Å) | 7.2720 | (5) | 7.2809 | (6) | 7.2593 | (5) |
| β | 102.115 | (6) | 101.734 | (6) | 101.73 | (1) |
| V (Å$^3$) | 1214.63 | | 1254.93 | | 1244.32 | |
| Mol.wt. | 2767 | | 2977 | | 2976 | |
| $D_{calc.}$ g/cm$^3$ | 3.782 | | 3.939 | | 3.971 | |
| $D_{meas.}$ g/cm$^3$ | 3.77 | | 3.89 | | 3.96 | |
| $K_c$ | 0.1112 | | 0.1079 | | 0.1078 | |
| $\frac{\alpha+\beta+\gamma}{3}$ | 1.431 | | 1.439$^x$ | | 1.434$^x$ | |
| $K_p$ | 0.1140 | | 0.1114 | | 0.1094 | |

$K_c$ calculated by means of the constants for simple F compounds (Pauly 1982).

$^x$: α estimated. A & B see Table 3, C see Table 2.

in the composite jarlite were made in pairs, as far as possible: the analyzed areas, 40 × 50 μm, were chosen opposite each other 50 μm from the mutual boundary. They are clearly of different compositions. Concentrations of Mg and K in the jarlite core are very low, approaching the limits of detection, whereas the jarlite mantle shows significantly higher values for Mg and K. Sr and Ba are distinctly higher in the core than in the mantle.

The composition of "meta-jarlite" from chiolite (Table 3) seems to be rather similar to that obtained by wet-chemical analyses of this type of "meta-jarlite", apart from the influence from cryolite contamination in the latter (Table 1). The compositions of the other types of "meta-jarlite" illustrate variations in the Ba/Sr ratio similar to what was found in the wet-chemical analyses (Table 1).

The composition of jarlite from the stenonite paragenesis shows the highest Ba/Sr ratio of all the samples analyzed. The Mg and K values are small, closer to those found for the jarlite core samples than to any of the others.

An indication of the amount of (OH) can be obtained from the Gladstone–Dale constants for the samples listed in Table 4. The $K_C$ values for these samples were calculated as if all metals were bound to F, using the constants for simple F compounds (Pauly 1982). Calculations show that $K_C = K_P$ if it is assumed that 2 to 3 atoms of F are replaced by (OH). The results of wet-chemical analyses (Table 1) indicate that 3 to 4 atoms of F are replaced by (OH).

## EMPIRICAL FORMULAE FOR JARLITE

The empirical formulae (Tables 2, 3) are calculated from the analytical results on the basis of 68 anions, as proposed by Hawthorne (1983). The number of cations amounts to 30.15 on average in these formulae, in agreement with the 30 cations shown in the nominal formula (Hawthorne 1983): $Na_2(Sr_{12}Na_2)Mg_2Al_{12}F_{64}(OH)_4$. The deviations in the structure, mentioned by Hawthorne (1983) as indicating the "presence of additional components (other cations or vacancies)" are apparently connected with "other cations", not with vacancies.

The validity of a formula based on 68 anions is supported by the agreement among X-ray data, density determinations and molecular weights calculated from the compositions of: "meta-jarlite" from chiolite, jarlite from the stenonite paragenesis, and the jarlite core sample II (Table 4).

The nominal formula has 12 atoms of Al. The five analyses (Table 3) were performed together and give as an average 11.97 atoms of Al. The analysis of "meta-jarlite" white, dense II (Table 3) was performed together with the analyses of jarlite crystals and jarlite sample I (Table 2), and they give as an average 11.66 atoms of Al. This is presumably due to analytical errors.

The structure proposed by Hawthorne (1983) requires two atoms of Na in 6-fold coordinated sites, and "the remaining Na" placed in 9- to

10-fold coordinated sites with Sr, together with Ba, Ca and K, resulting in 14 atoms in these sites.

In the jarlite cores (Table 2), Mg hardly appears as a constituent, and their empirical formulae show that addition of the remaining Na to the atoms assigned to the Sr sites leads to a number of atoms exceeding 14: 16.24, 16.28 and 15.87.

Although Mg is reported to be present in significant amounts in the other analyses of jarlite, their empirical formulae (Tables 2, 3) show that the addition of the remaining Na to the atoms assigned to the Sr sites increases their number to more than 14 atoms: in jarlite mantles: 14.86, 14.99 and 14.42 (Table 2); in "meta-jarlite": 14.59, 14.44, 14.60, 14.85 and 15.03 (Table 3); in jarlite from the stenonite paragenesis: 15.64 (Table 3). The analysis of the latter was performed together with the first four analyses of "meta-jarlite" in Table 3. It deviates from these and the other analyses in giving more than four atoms of Na. Mg and K are present, but in small amounts; its empirical formula is closer to the formulae of the jarlite core than to those of "meta-jarlite" or jarlite mantle.

## DISCUSSION

The formula content calculated from the analyses (Tables 2, 3) cannot be accommodated in the structure in accordance with the model proposed by Hawthorne (1983), unless disorder to some extent is assumed for the octahedrally coordinated Mg sites and the 9- to 10-fold coordinated Sr sites. Extensive disorder seems indicated for the structure of the jarlite core, where the octahedrally coordinated Mg sites are practically devoid of Mg. This seems also to apply to the jarlite from the stenonite paragenesis.

The analytical results on the jarlite mantles and on the various samples of "meta-jarlite" give a more complicated picture because they, on recalculation, show from 1.5 to 2 atoms of Mg, although all of them yield more than 14 atoms, from 14.5 to 15, for the Sr sites. In spite of analytical errors and uncertainties, the trend toward a surplus of atoms for the Sr sites and the insufficient numbers of Mg atoms for the Mg sites seem real.

The composite nature of jarlite, with straight boundaries between the core and the mantle, suggests definite changes during its crystallization. The optical continuity of the two parts indicate uninterrupted growth. It is assumed that the changes were of a physical nature, such as changes in T or P, because no indications of renewed influx of fluids were observed in the material studied. The core–mantle boundary thus seems to indicate a shift from a disordered structure to an ordered structure. In the latter, to which "meta-jarlite" also seems to belong, the degree of order appears to be somewhat imperfect.

The anomalous optical properties exhibited by jarlite (less pronounced in the core than in the mantle) and the dendritic development of "meta-jarlite" may reflect imperfections in the crystals induced during their formation.

The jarlite from the stenonite paragenesis shows none of these anomalies. The conditions for crystallization were obviously favorable in this milieu because of its $CO_2$ content. As the empirical formula of this jarlite is close to those of jarlite core, it seems that the jarlite from the stenonite paragenesis also represents the disordered structure.

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