

MINERALOGICAL NOTES

WAIRAKITE FROM LOW-GRADE METAMORPHIC ROCKS ON ST. CROIX,
U. S. VIRGIN ISLANDS¹

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The calcium zeolite, wairakite, originally described from hydrothermally altered rocks at Wairakei, New Zealand (Steiner, 1955; Coombs, 1955) has since been discovered in low-grade metamorphic rocks near Mt. Rainier, Washington (Wise, 1959), and in altered volcanic rocks on St. Thomas, U. S. Virgin Islands (Donnelly, 1959). The present report describes an occurrence of wairakite and other Ca-Al silicates in low-grade metamorphic rocks of the Judith Fancy Formation from St. Croix, U. S. Virgin Islands (Whetten, 1961).

The Judith Fancy Formation, about 4,500 meters thick, consists largely of tuffaceous sedimentary rocks ranging from coarse breccias to fine siltstones, with local limestone beds of bioclastic origin. Generally poorly preserved foraminifera, radiolaria, corals and mollusks occur throughout the formation and indicate a marine origin and a Campanian Age (Late Cretaceous). Rocks of the Judith Fancy Formation appear to be slightly metamorphosed: pyroxene has altered to amphibole and chlorite, plagioclase has been albitized, volcanic glass has devitrified, and calcite, chlorite and epidote replace a variety of minerals. The metamorphic effects are probably caused in large part by increased temperature due to depth of burial, although contact-metamorphosed rocks are present adjacent to two plutons, and cleavage due to shearing during folding is locally well-developed in incompetent mudstone units in rocks older than the Judith Fancy Formation.

Wairakite, the only zeolite which has so far been found on St. Croix, has been identified by x-ray diffraction from a thin, very fine-grained limy unit in roughly the middle (?) of the Judith Fancy Formation. The zeolite makes up about 30% of a rock in which all of the major minerals are calcareous, including calcite (50%), clinzoisite (possibly zoisite) (10%) and prehnite (3%). In addition, chlorite, quartz and opal (as diatom frustules) are present. The grain size is between 10-60 μ for all minerals except prehnite, which occurs in spongy crystals up to 0.2 mm long, and chlorite, which is commonly smaller than 10 μ . Only clinzoisite shows good crystal faces, and there is no obvious preferred mineral

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orientation in the rock. Lacking a better name, this rock is called a wairakite-bearing limestone, although it is recognized that many of the minerals are not of sedimentary origin.

Calcite in the wairakite-bearing limestone is in part a primary mineral (recrystallized foraminifera are present), but much of it may be secondary. It occurs in veinlets with quartz and wairakite, and in other Cretaceous rocks on St. Croix calcite has clearly replaced large quantities of silicate minerals. Textural relations suggest that wairakite and prehnite have partially replaced calcite. Clinozoisite, because of its idioblastic character, is also considered to have developed *in situ*.

To obtain a wairakite concentrate, the wairakite-bearing limestone was pulverized and centrifuged in a bromoform-acetone mixture ($\rho = 2.35$ gm/cc). The light fraction consists of rounded, virtually isotropic ($n = 1.502 \pm .002$) wairakite grains, each surrounded by a thin discontinuous chlorite film, and in some cases enclosing tiny calcite crystals. The wairakite concentrate is estimated at 85–90% pure; the main impurities are chlorite, calcite and quartz. A partial micro-analysis compared with a chemical analysis of the pure mineral (Table 1) shows additional CaO and MgO in the St. Croix sample, which are undoubtedly from the impurities calcite and chlorite, respectively. The low Na₂O content in the St. Croix analysis is additional confirmation that the zeolite is wairakite, not analcime, its sodium analogue.

Wairakite apparently nucleates readily, and is easily produced synthetically. According to Fyfe (in Coombs *et al.*, 1959), the temperature at which it forms is highest when amorphous silica is used in the starting materials. Using silica gels, Koizumi and Roy (1960) and Ames and Sand (1958) have produced wairakite at temperatures as high as 400°–505° C., and as low as 260° C. However, it is likely that the lowest temperature at which anorthite will form (in the presence of quartz) is closer to the wairakite-anorthite equilibrium boundary.

TABLE 1. CHEMICAL DATA FOR WAIRAKITE FROM ST. CROIX, VIRGIN ISLANDS (S. SHAW, ANALYST) AND WAIRAKEI, NEW ZEALAND
Calcite, quartz, and chlorite are impurities in the St. Croix sample.

	St. Croix (partial micro-analysis)	New Zealand (data from Steiner, 1955)
Na ₂ O	1.63 wt. %	1.06 wt. %
K ₂ O	0.17	0.16
CaO	16.01	11.7
MgO	0.96	—

Coombs (1960), using powdered mixes of the minerals kaolin, calcite and quartz, which may approximate the materials which produced the St. Croix wairakite-bearing rock, formed wairakite in the temperature range of 273°–347° C., with at least a trace of calcite present at the end of each run. Anorthite was first formed at 295° C. It is possible that this latter temperature is close to the highest temperature at which wairakite will form under equilibrium conditions. It should be noted that the St. Croix wairakite-bearing limestone apparently did not reach equilibrium, as a small amount of isotropic material ($n=1.45$) resembling opal is present in the form of diatom frustules (identified by Dr. Hsin-Yi Ling). The presence of four calcic phases may be further evidence of disequilibrium.

At Wairakei, N. Z., wairakite forms over a temperature range of 200°–250° C. (Steiner, 1955) at comparatively shallow depths and low pressure. The lower temperature may well be the minimum temperature at which wairakite will form.

Assuming a geothermal gradient of 30° C./km (Turner and Verhoogen, 1960, p. 436), temperatures on the order of 200°–300° C. would be reached at depths of approximately 7,000–10,000 meters. It is impossible to estimate the burial depth of the wairakite-bearing limestone with any degree of accuracy, but a minimum figure would be on the order of 1,500–3,000 meters, or more than two to six times less than would be required by a normal gradient. An alternative preferable to deep burial and subsequent erosion is an abnormally high geothermal gradient for the St. Croix area, as is predicted for the occurrence of wairakite by Coombs' (1961) schematic equilibrium diagram for certain possible reactions involving Ca-Al silicates. The presence of metamorphic amphibole after pyroxene (virtually unknown in other zeolite-bearing low-grade metamorphic rocks), epidote, and prehnite in association with wairakite, is compatible with a higher temperature gradient than is normal in zeolite mineral facies rocks.

In either case, the presence of wairakite indicates a relatively high temperature of formation, which is probably outside of the stability fields of the more common zeolites, and satisfactorily explains why zeolites are not more extensively developed on St. Croix. This has been somewhat of a puzzle, considering the widespread zeolitization of rocks of similar composition and age in nearby Puerto Rico reported by Otálora (1964).

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THE GROWTH OF CROCOITE SINGLE CRYSTALS

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The generally accepted methods of growing single crystals are: a. from saturated solutions, b. from the fused state, c. from the vapor phase, d. the hydrothermal method. Lead chromate was found insoluble in every solvent investigated (in water its solubility product is only 1.8×10^{-14}) with the exception of strongly acidic solutions. It starts to decompose at 100–200° below its melting point where its vapor pressure is still very low. As a result, there is only one work published on the growth of lead chromate single crystals. O'Connor and Buchanan (1952) grew large (5×3 mm) PbCrO₄ crystals by slow evaporation, at 80° for 12 weeks, of a 0.05 M solution (2 liters) of Pb(NO₃)₂ in 1N HNO₃, to which was added, daily for 6 weeks, 50 ml of 0.05 M K₂Cr₂O₇ solution. Further growth, using these crystals as seeds, was never achieved.

In connection with our hydrothermal research program we investigated several possibilities of growing large single crystals of lead chromate.

Hydrothermal method. Solubility measurements were attempted with lead chromate in a platinum reactor up to 450° C. and 20,000 psi. In each case, all the PbCrO₄ which dissolved was decomposed.

Chemical reaction in solution. PbCl₂ was placed on the bottom of a glass tube, b, in Fig. 1; the upper part of the tube was separated from the lower part by a rubber plug, a, through which passed a glass tube. Na₂Cr₂O₇ crystals, c, were placed above the plug and, to slow down the diffusion of the CrO₄⁼ ions, the upper part of the tube was filled with glass wool, d. The amount of Pb⁺⁺ ions present was limited by the small solu-

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