Economic Geology
Vol. 81, 1986, pp. 478–483

HIGH-TEMPERATURE FLUID INCLUSIONS AND THE ROLE OF THE BIOTITE GRANODIORITE IN MINERALIZATION AT THE PANGUNA PORPHYRY COPPER DEPOSIT, BOUGAINVILLE, PAPUA NEW GUINEA

C. J. EASTOE

Department of Geosciences, University of Arizona, Tucson, Arizona 85721

AND P. J. EADINGTON

Division of Mineralogy, C.S.I.R.O., P.O. Box 136, North Ryde, New South Wales 2113, Australia

Introduction

Porphyry copper mineralization at Panguna on the island of Bougainville in Papua New Guinea is located at the southern margin of the Pliocene Kaverong Quartz Diorite stock which intrudes andesites of Miocene age in the mine area. The geology of the deposit has been described by Fountain (1972), Baumer and Fraser (1975), and Baldwin et al. (1978). The orebody is centered on three smaller stocks of porphyry of quartz dioritic to granodioritic composition. Major features of the geology of the orebody are indicated in Figure 1.

The Leucocratic Quartz Diorite is intensely altered in its entirety to potassic and minor propylitic and sericitic assemblages. It lies at the center of a symmetrical cell of quartz-chalcopyrite mineralization defined by high fluid inclusion temperatures near the stock and bounded by halos of sulfide veins. The Biuro Granodiorite is weakly altered to argillic assemblages and does not appear to be the progenitor of or host to any significant copper mineralization. It is cut by a set of sparse quartz-chalcopyrite veins which also cut the Biotite Granodiorite, the most intensely altered of the stocks. The alteration assemblages are described below. The Biotite Granodiorite hosts largely subeconomic copper mineralization. Fountain (1972) considered it a likely progenitor of the main mineralization, within which it is centrally located. Eastoe (1978) found that there was no pattern of fluid inclusion data centered on the intrusion. It seems likely that the disseminated, subeconomic mineralization in the Biotite Granodiorite and small rich pods of chalcopyrite in marginal intrusion breccias were the only copper mineralization associated with the emplacement of the stock. Fluid inclusions in veins not attributable to the phases of mineralization mentioned so far yielded an asymmetrical pattern of temperature and salinity increasing toward the northern margin of the deposit. The pattern indicates that the progenitor of this earlier mineralization was the Kaverong Quartz Diorite. The mineralization was probably associated with the biotite alteration of the southern rim of the Kaverong Quartz Diorite. The biotitized zone is cut by the Biotite Granodiorite and the Biuro Granodiorite. The fluid inclusion data summarized here are discussed in detail by Eastoe (1978).

In this communication, we reexamine the role of the Biotite Granodiorite in the cycle of intrusion and hydrothermal activity which gave rise to the Panguna deposit, in particular in the light of some new fluid inclusion data from quartz phenocrysts.

The Biotite Granodiorite

The Biotite Granodiorite contains the most intensely altered rock in the Panguna mine and no fresh samples are available. Phenocrysts make up 40 to 65 percent of the rock and include andesine, quartz, hornblende, and magnetite, set in an aphanitic groundmass of quartz and alkali feldspar (Ford, 1976). Three styles of alteration have been described: (1) K silicate alteration (biotite, chlorite, K-feldspar, magnetite, anhydrite); (2) chlorite-sericite (gradational with the K silicate zone by increasing chloritization of biotite and sericitization of plagioclase); and (3) sericite-clay (chlorite present only as isolated flakes). The chlorite-sericite style includes a distinctive textural type in which 1-cm chloritic clots are set in a light-colored matrix (Ford, 1978). Oxygen isotope studies showed that sericite from these assemblages could have formed either from magmatic water at temperatures near 500°C or from meteoric water near 300°C (Ford and Green, 1977).

The copper content of the intrusion averages 0.23 percent and attains ore grade (0.3%) near the contacts and at the center (Baldwin et al., 1978). The copper occurs as disseminated chalcopyrite and less commonly in association with irregular, anastomosing quartz veins, as well as in the late, sparse quartz-chalcopyrite veins that cut the intrusion. Within the breccias marginal to the Biotite Granodiorite, chalcopyrite occurs in vuggy areas intergrown with coarsely crystalline biotite, K-feldspar, quartz, and apatite.

The Biotite Granodiorite intruded the margin of the Kaverong Quartz Diorite, but its age relative to the other two porphyries is difficult to determine. Eastoe (1978) considered it to be older than the Leucocratic Quartz Diorite on the basis of a halo of pyrite veins which encircles the Leucocratic Quartz Diorite but cuts the Biotite Granodiorite (Fig. 1).

Inclusion Petrography of Quartz Phenocrysts

Quartz phenocrysts were examined in three samples of Biotite Granodiorite specimens (103001,

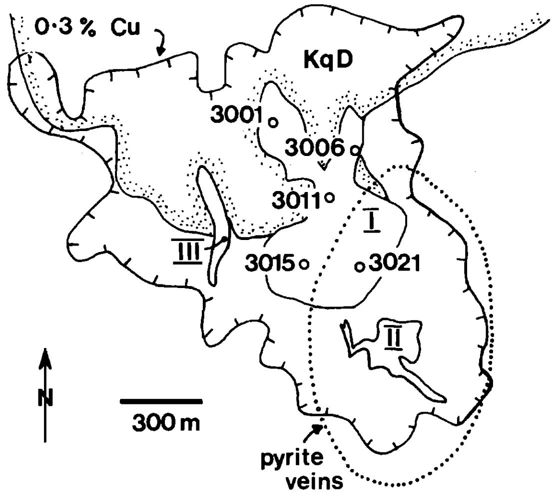

FIG. 1. Map of the Panguna deposit, outlined by the 0.3 percent copper grade contour. Major intrusive bodies are shown: the Kaverong Quartz Diorite (KqD, stippled margin), the Biotite Granodiorite (I), the Leucocratic Quartz Diorite (II), and the Biuro Granodiorite (III) stocks. Sample locations (last four digits of sample numbers) and the location of the halo of pyrite veins about the Leucocratic Quartz Diorite are given.

103011, and 103015), one of intrusive breccia (specimen 103006), and one of a quartz-feldspar porphyry dike (specimen 103021) from the central part of the biotite granodiorite. In each case, the phenocrysts were taken within a few millimeters of a quartz-Cu,Fe sulfide vein in which the fluid inclusions were also studied.

The phenocrysts contain inclusions of the following kinds: (1) type I, liquid-rich inclusions containing only liquid and a bubble; (2) type II, gas-rich inclusions with small liquid rims; (3) type III, salt-rich inclusions containing liquid, a bubble occupying 10 to 20 percent of the cavity volume, and daughter minerals including halite, sylvite, hematite, birefringent colorless salts, and opaque salts, some with the form of chalcopyrite; (4) type II–III, inclusions intermediate between types II and III; (5) opaque solid inclusions (rare); (6) isotropic transparent solid inclusions with internal bubbles; (7) and anisotropic transparent solid inclusions, some of low and some of high birefringence. Examples are shown in Figure 2. The fluid inclusions (1–4) range in size up to 60μ, with many between 15 and 25μ. The solid inclusions (5–7) range up to 100μ in size.

The phenocrysts in specimen 103001 are particularly rich in inclusions and the type III inclusions yielded microthermometric data of the greatest interest (see below). Therefore this sample has been studied in greatest detail. Type I inclusions in specimen 103001 occur along healed fractures and are secondary. Types II and III are primary, occurring as decorations on growth zones of the phenocrysts, rarely attached to solid inclusions, and commonly irregularly distributed through the centers of the phenocrysts. The growth zones are also abundantly decorated with transparent solid inclusions. Phenocrysts may exhibit no growth zone, or one, or even two concentric zones decorated with inclusions (Fig. 3C).

The solid inclusions in specimen 103001 have not been identified with certainty. The following data on composition are available:

1. The opaque inclusions yielded semiquantitative electron microprobe analyses indicating Cu,Fe sulfide composition.
2. The isotropic inclusions may consist of silicate glass but have not been analyzed.
3. One of the transparent inclusions of low birefringence is zoned alkali feldspar with a relatively K-rich core and an Na-rich rim (Table 1A).
4. One of the transparent inclusions of high birefringence gave the analysis in Table 1B. The grain was sufficiently large that the surrounding quartz should not have been excited. It has one refractive index of about 1.55 and a birefringence of about 0.1. Its identity remains unresolved.

Measurements on Type III Inclusions

Temperatures of disappearance of halite (T_{mNaCl}), bubble (T_h), and where possible, sylvite (T_{mKCl}) were measured for type III inclusions in quartz phenocrysts and veins in the five samples. Most of the T_m data were gathered on Chaixmeca apparatus at the University of Tasmania. T_h data were beyond the reach of that apparatus in some specimens, and in the case of sample 103001, were obtained on a modified Leitz 1350 heating stage at the C.S.I.R.O., North Ryde, New South Wales. The calibration curve (Fig. 4) used in determining the homogenization temperatures in 103001 depends on the melting points of $K_2Cr_2O_7$ (398°C) and NaCl (800.4°C). Extrapolation of the curve to 900° to 1,000°C gives a correction of 50° to 70°C to be added to readings. The calibration was checked at the time of writing, with the addition of the melting point of Au (1,064°C). By that time, the configuration of the thermocouple had been significantly modified, so that the two calibration curves are not strictly comparable. It is nonetheless relevant that corrections of the same sense were determined; they were larger by some tens of degrees near 1,000°C and were possibly nonlinear.

The vein data have already been presented (Eastoe, 1978) and are compared here with data for the phenocrysts (Fig. 5). In one case (103001) the salt-rich liquid included within the phenocrysts was quite distinct from that found within the adjacent vein, confirming that the inclusions in that phenocryst are not the result of contamination by later vein fluids. The same may be true in samples 103015 and 103021, and a larger data set might serve to make this clearer. Only a small number of suitable inclusions could be

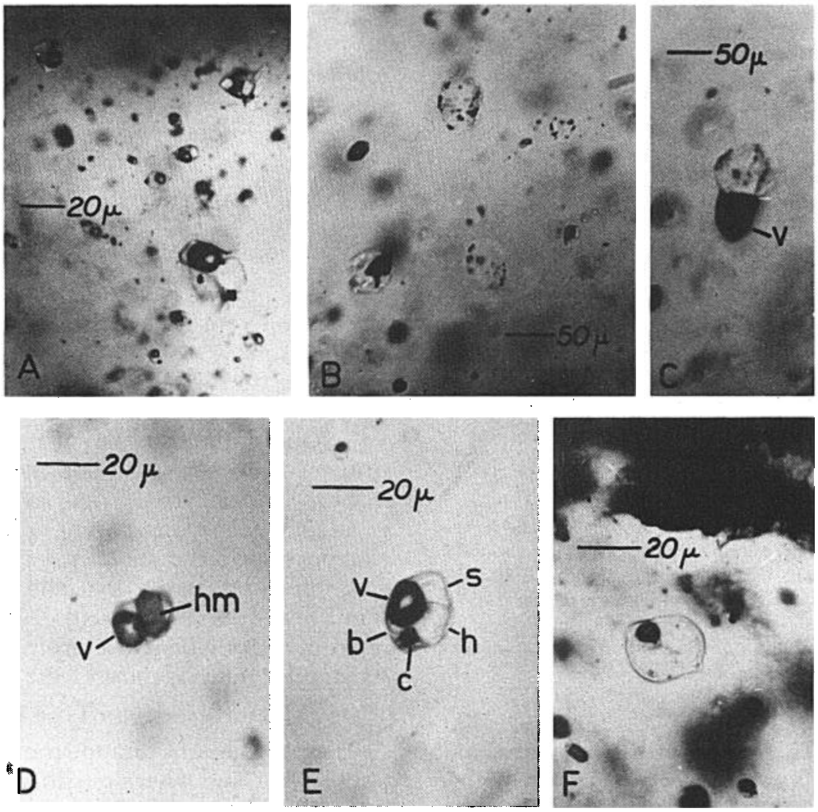

FIG. 2. Photomicrographs of inclusions from quartz phenocrysts in specimen 103001. A. Salt-rich inclusions, considered primary because they are scattered randomly through the host. B. Silicate glass inclusions with small bubbles (possibly shrinkage voids). The large vapor bubble attached to the lower left-hand inclusion may have been trapped vapor. C. A silicate (alkali feldspar?) inclusion with a vapor bubble (v) attached. D. A salt-rich inclusion containing a vapor bubble (v) and a large flake of hematite (hm). E = a salt-rich inclusion containing halite (h), sylvite (s), vapor (v), chalcopyrite (c), and a birefringent salt (b). F = a silicate glass(?) inclusion after heating to 1,000°C. Note the round bubble (contrast the inclusions in B) and the rim of melted material.

found in these specimens, however. The 103006 and 103011 vein and phenocryst fluids overlap in characteristics and may not be of different origins.

Because the material in 103001 was excellent and the salt-rich liquid clearly was not introduced from a vein, attention was focused on that specimen. The T_h data have a mode of approximately 1,000°C and a range of 940° to 1,080°C, except for one inclusion (not shown) which homogenized at 790°C. Note the difference in phase behavior between these and type

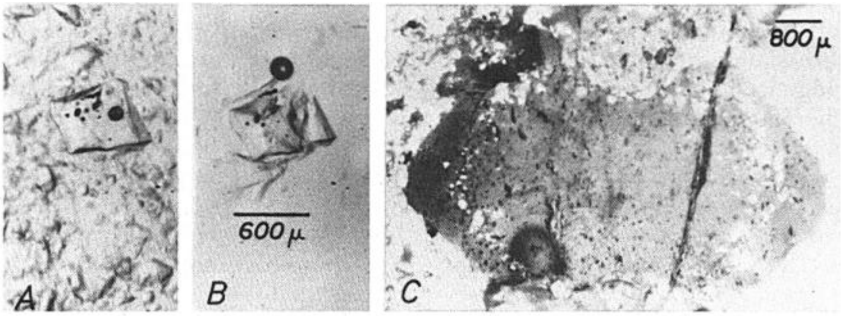

FIG. 3. Photomicrographs, specimen 103001. A. A chip from a quartz phenocryst bearing several type II inclusions. B. The same chip after a crushing experiment during which the largest inclusion was ruptured. C. A quartz phenocryst under crossed nicols, showing a growth zone decorated with birefringent solid inclusions.

TABLE 1. Electron Microprobe Analyses

A. A transparent solid inclusion of low birefringence in a quartz phenocryst

	Center		Rim	
	wt %	S.F	wt %	S.F.
Na_2O	7.14	1.23	11.23	1.91
Al_2O_3	19.39	2.04	20.41	2.11
SiO_2	67.02	5.97	66.81	5.87
K_2O	5.89	0.67	0.43	0.05
CaO	0.56	0.05	1.11	0.10
Total	101.10	9.96	100.60	10.05

S.F. = structural formula per 16 oxygens

B. A transparent solid inclusion of high birefringence in a decorated growth zone of a quartz phenocryst

	wt %	S.F.
SiO_2	65.9	8.1
Al_2O_3	3.2	0.4
FeO	0.3	0.0
MnO	0.3	0.0
CaO	16.3	2.1
Na_2O	1.8	0.4
Cl	0.1	0.0
Total	88.5	11.0

S.F. = structural formula per 24 oxygens, assuming the deficit in the total to be due to H_2O

III inclusions from the vein in 103001, where the T_h is less than the T_{mNaCl}. Note also that the T_h histogram has a high-temperature "tail" to the modal result. This part of the scatter of data could be due to the incorporation of small vapor bubbles in the inclusions at trapping. But the inclusions measured have vapor to cavity volume ratios similar to those of most type III inclusions in the phenocrysts, so that it is unlikely that the high modal T_h values themselves could be explained by incorporation of vapor at the time of trapping.

In the same phenocrysts, the isotropic transparent solid inclusions were observed to melt around the edges during heating experiments (Fig. 2F). The bubbles in such inclusions became rounded, coalesced, and in one case disappeared. Melting in one inclusion was observed at 950°C and must have begun at a lower temperature.

Crushing Experiment

The expansion of bubbles in type II inclusions in phenocrysts from specimen 103001 was observed as the host quartz was crushed under glycerine. A two- to three-fold increase in diameter was observed (Fig. 3A and B), corresponding to the presence of a gas at a pressure of about 10 bars at 25°C. The gas could be CO_2. (SO_2, another possibility, should have disproportionated at low temperatures.)

Discussion

Magmatic fluids

In three of the specimens, the quartz phenocrysts preserve salt-rich liquid which was not derived from nearby fractures. In specimen 103001, the petrographic evidence indicates that type II and III inclusions were of primary origin. The existence of mixed-type inclusions and the occurrence of both types II and III in single growth zones indicate that both salt-rich liquid and vapor were present with silicate liquid as the quartz phenocrysts grew. The salt-rich liquid is like that which deposited quartz, anhydrite, and Cu,Fe sulfides in veins at Panguna (Eastoe, 1978). Copper was already being deposited as copper-rich sulfide under magmatic conditions.

The high temperatures indicated by the fluid inclusions certainly lie within the range of magmatic temperatures. Similar high homogenization temperatures were reported for the tungsten-mineralized Mariktikan intrusion, U.S.S.R. (1,050°–1,120°C) by Reyf and Bazheyev (1977), and the Kal'makyr porphyry copper deposit, U.S.S.R. (860°C) by Sotnikov and Berzina (cited in Roedder, 1981).

The derivation of such fluids from a crystallizing granodiorite melt is consistent with present ideas on the composition and emplacement of calc-alkaline granitoids (Burnham, 1979). An external (ground water) origin for the nonsilicate fluids is untenable within the framework of Burnham's model. Ground water could not penetrate the carapace of an intrusion and gain access to the highly pressurized molten part.

The Biotite Granodiorite and mineralization

Despite the presence of copper-bearing fluid, abundant at least locally, the Biotite Granodiorite was not a significant mineralizer at Panguna. The absence of a stockwork centered on the intrusion suggests that fluid did not break out into the wall rocks. It is suggested that some magmatic fluid escaped upward

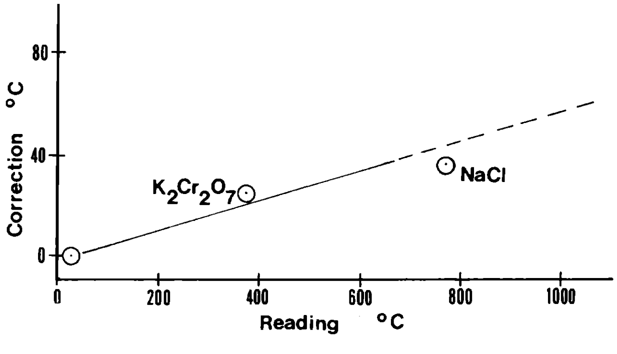

FIG. 4. Calibration curve used for the high T_h measurements. The corrections are added to the thermocouple readings.

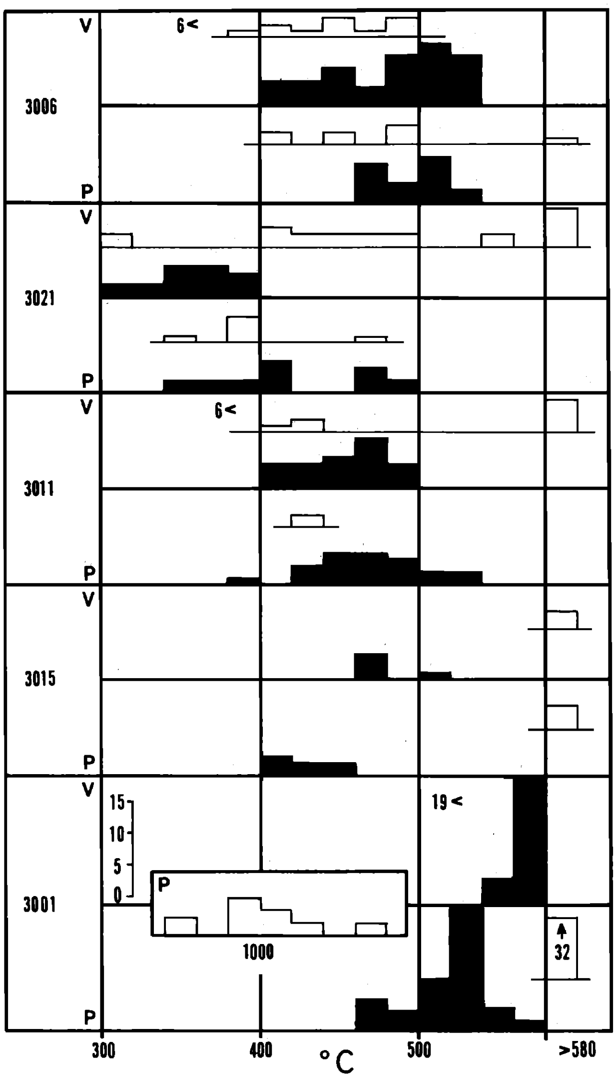

FIG. 5. Frequency histograms of $T_{h_{vapor}}$ (white) and $T_{m_{NaCl}}$ (black) data for salt-rich fluid inclusions. Specimen numbers are given at left. V = data for vein; P = data for phenocrysts. The symbol "6<" indicates 6 cases of $T_h < T_{m_{NaCl}}$, T_h not determined precisely. The inset in 103001 (P) represents T_h data for 13 inclusions determined on the modified Leitz 1350 stage.

along permeable parts of the marginal breccias which are now vuggy. Much magmatic fluid may have remained within the intrusion until subsolidus temperatures were reached, leading to the pervasive and unusual alteration and the disseminated mineralization. It is possible that stockwork-style fracturing and mineralization may have occurred around the Biotite Granodiorite above present levels of exposure. Ford (1976) considered it to have reached a higher level than the Kaverong Quartz Diorite and the Leucocratic Quartz Diorite.

Note that quartz phenocrysts of the Biuro Granodiorite also contain inclusions of types II and III, but these are sparse, completely absent in some phenocrysts and commonly of secondary habit in others. The Biuro Granodiorite is considered to be a nonmineralizer which did not produce significant fluids as it crystallized.

Comparison of measured temperatures with experimental data

Given the magmatic origin of the salt-rich liquid in phenocrysts of the biotite granodiorite, there remains the question of the meaning of the 1,000°C homogenization temperatures. These ought to correspond with experimental data for silicate melt-water systems but do not, as explained below.

The hydrostatic pressure at the time of emplacement of the Biotite Granodiorite at its present level of exposure can best be estimated by the pressure at the edge of mineralization associated with the Leucocratic Quartz Diorite, 285 to 310 bars (Eastoe, 1982). The corresponding lithostatic pressure would have been 750 to 850 bars. It is probably necessary to consider a broader pressure range, in case the edge pressure represents a regime of pressures somewhat higher than hydrostatic (corresponding to a lower lithostatic pressure), or in case the magma was overpressured, or the phenocrysts trapped fluids below where they now occur. Let us consider the range 700 to 1,000 bars.

The data of Whitney (1975a) for synthetic quartz monzonite indicate that quartz cannot coexist with aqueous fluid at 1 kb at temperatures greater than 750° to 800°C. Furthermore, under these conditions quartz should appear very near the solidus. Data for 1 kb are not available for a granodiorite composition, but a comparison of data for 2 kb (Whitney, 1975b) suggests a similar limit. Therefore the temperatures indicated by the fluid inclusions are too high by more than 200°C.

There are some possible difficulties in the comparison:

1. NaCl was not present in Whitney's experimental systems.
2. The Biotite Granodiorite may have been more mafic than it now appears. Whitney's experimental systems contained no Fe or Mg. Small additions of FeO and MgO in silicate melt systems produce large shifts in the curves toward higher temperatures (compare diagrams of Wyllie, 1977). Because of the intense, pervasive alteration of the Biotite Granodiorite, it is not possible to evaluate this suggestion.
3. Pressures may have been lower than those considered. The decorated growth zones probably mark interrupted quartz crystallization and corrosion of the crystal surface, with the inclusions filling corrosion pits. Corrosion could be brought about by pressure increase, for example, during buildup of volatiles between episodes of explosive release of volatiles. In this case, most quartz growth and production of salt-

rich liquid would occur during intervening periods of relatively low pressure when the outer shell of the intrusion was fractured. The solidi become strongly pressure dependent below 1 kb but could not approach 1,000°C at realistic pressures, say greater than 500 bars.

4. If significant CO_2 was present, as suggested by the crushing experiment, the solidus and the liquidus may have been displaced to higher temperatures. The effect would be analogous to that demonstrated in andesitic magma by Eggler and Burnham (1973).

5. The fluid inclusion data may be at fault. There is, however, no evidence of leakage, and incorporation of vapor has been ruled out (above) as a means of generating the high T_h values. The host quartz has passed through the α-β transition, but the transition is rapid and reversible and should not affect the results. The calibration curve of the fluid inclusion stage was checked. Although significant error is probable at high temperatures, this seems unlikely to account for errors of as much as than 200°C.

At present, there seems to be no satisfactory explanation for the high homogenization temperatures. This is presented as an unsolved problem, perhaps indicating an imperfect understanding of fluid inclusion behavior or silicate-volatile systems at such high temperatures.

Implications for salt-water systems

The homogenization of type III inclusions at temperatures near 1,000°C demonstrates that immiscibility between aqueous vapor and salt-rich liquid persists to such high temperatures in natural systems. This is not as would be predicted by the application of figure 18 of Sourirajan and Kennedy (1962) for the system $NaCl$-H_2O. The extrapolated part of the diagram shows no immiscibility beyond 800°C. Figure 2B of Pichavant et al. (1982) suggests, albeit on the basis of rough estimations, that immiscibility should persist beyond 800°C. The natural system is, of course, much more complex in composition and the $NaCl$-H_2O system (if correctly depicted) is not a good model in this case.

Conclusions

Fluid inclusions of salt-rich liquid and vapor were trapped in quartz phenocrysts of the Biotite Granodiorite at Panguna. Textural evidence indicates that the fluid inclusions are primary. On the basis of T_h and T_{mNaCl} data, the salt-rich liquid is clearly different from that trapped in a nearby vein in one specimen and may also be different in two other specimens. Thus, there is evidence that the salt-rich liquid and the vapor are of magmatic origin and coexisted with silicate melt as the Biotite Granodiorite crystallized. The fluids bore copper but did not bring about significant mineralization beyond the intrusion. High homogenization temperatures, near 1,000°C, of the salt-rich inclusions cannot be reconciled with experimental data for artificial silicate melt-water systems at present.

Acknowledgments

The authors are grateful to D. Cox, E. Theodore, and D. H. Green for their helpful comments on this work.

February 20, June 20, 1985

REFERENCES

Baldwin, J. T., Swain, H. D., and Clark, G. H., 1978, Geology and grade distribution of the Panguna porphyry copper deposit, Bougainville, Papua New Guinea: ECON. GEOL., v. 73, p. 690–702.

Baumer, A., and Fraser, R. B., 1975, Panguna porphyry copper deposit, Bougainville, *in* Knight, C. L., ed., Economic geology of Australia and Papua New Guinea, 1. Metals: Australasian Inst. Mining Metallurgy Mon. 5, p. 855–866.

Burnham, C. W., 1979, Magmas and hydrothermal fluids, *in* Barnes, H. L., ed., Geochemistry of hydrothermal ore deposits: New York, Wiley, p. 71–136.

Eastoe, C. J., 1978, A fluid inclusion study of the Panguna porphyry copper deposit, Bougainville, Papua New Guinea: ECON. GEOL., v. 73, p. 721–748.

—— 1982, Physics and chemistry of the hydrothermal system at the Panguna porphyry copper deposit, Bougainville, Papua New Guinea: ECON. GEOL., v. 77, p. 127–153.

Eggler, D. H., and Burnham, C. W., 1973, Crystallization and fractionation trends in the system andesite-H_2O-CO_2-O_2 at pressures to 10 kb: Geol. Soc. America Bull., v. 84, p. 2517–2532.

Ford, J. H., 1976, A geochemical and stable isotope study of the Panguna porphyry copper deposit, Bougainville: Unpub. Ph.D. thesis, Univ. Queensland, 213 p.

—— 1978, A chemical study of alteration at the Panguna porphyry copper deposit, Bougainville, Papua New Guinea: ECON. GEOL., v. 73, p. 703–720.

Ford, J. H., and Green, D. C., 1977, An oxygen- and hydrogen-isotope study of the Panguna porphyry copper deposit, Bougainville, Papua New Guinea: Geol. Soc. Australia Jour., v. 24, p. 63–80.

Fountain, R. J., 1972, Geological relationships in the Panguna porphyry copper deposit, Bougainville Island, New Guinea: ECON. GEOL., v. 67, p. 1049–1064.

Pichavant, M., Ramboz, C., and Weisbrod, A., 1982, Fluid immiscibility in natural processes: Use and misuse of fluid inclusion data, I. Phase equilibria analysis—a theoretical and geometrical approach: Chem. Geology, v. 37, p. 1–27.

Reyf, F. G., and Bazheyev, Y. D., 1977, Magmatogenic chloride solutions and tungsten mineralization: Geochem. Internat., v. 14, p. 45–51.

Roedder, E., 1981, Natural occurrence and significance of fluids indicating high temperature and pressure, *in* Rickard, D. T., and Wickman, R. E., eds., Chemistry and geochemistry of solutions at high temperatures and pressures: Oxford, Pergamon Press, p. 9–40.

Sourirajan, S., and Kennedy, G. C., 1962, The system $NaCl$-H_2O at elevated temperatures and pressures: Am. Jour. Sci., v. 260, p. 115–141.

Whitney, J. A., 1975a, Vapor generation in a quartz monzonite magma: A synthetic model with application to porphyry copper deposits: ECON. GEOL., v. 70, p. 346–358.

—— 1975b, The effects of pressure, temperature and X_{H_2O} on phase assemblage in four synthetic rock compositions: Jour. Geology, v. 83, p. 1–27.

Wyllie, P. J., 1977, Crustal anatexis: An experimental review: Tectonophysics, v. 43, p. 41–71.