

**CHARACTERISTICS OF MINERALIZING FLUIDS IN THE  
BRALORNE-PIONEER MESOTHERMAL GOLD VEIN DEPOSIT:  
RESULTS OF A FLUID INCLUSION, STABLE ISOTOPE, AND  
THERMODYNAMIC STUDY\***  
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**INTRODUCTION**

Results from fluid-inclusion and stable-isotope studies, and thermodynamic modelling, have led to estimates of the pressure-temperature-composition (P-T-X) characteristics of mineralizing fluids in the Bralorne-Pioneer mesothermal gold vein deposits. Observed wallrock alteration mineral assemblages, and pressure-temperature (P-T) conditions estimated from fluid inclusions, were used to constrain a water-rock thermodynamic computer model. Stable-isotope studies confirm that significant interaction of wallrocks with the ore fluid took place. The "fault valve" hypothesis of Sibson *et al.* (1988) offers an explanation for fluid migration, and is consistent with the main features of the ribbon-banded, yet coarsely crystalline quartz veins at Bralorne.

The major gold-bearing veins at Bralorne strike about 110° azimuth and dip north at 70°, with slickensides plunging 45° east indicating that the last movement was reverse. Major ore shoots occupy somewhat less than 20 per cent of the veins and plunge steeply west, roughly perpendicular to the slickensides. The best host for veins seems to have been the competent Bralorne diorite (consisting of albite, hornblende and quartz) and the Cadwallader greenstone. The veins contain thin dark ribbons of fine-grained sulphide in massive milky quartz with minor calcite.

Hydrothermal alteration envelopes around the veins are extensive and grade outwards from intensely foliated quartz/ankeritic carbonate/sericite (+ fuchsite) to less sheared calcite/chlorite/albite to unshered epidote/calcite. Chemical studies of the alteration on a constant volume basis (based on Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>, which have remained relatively immobile), show that there has been addition of K<sub>2</sub>O, CO<sub>2</sub>, S, As and Au, while Na<sub>2</sub>O, FeO (total) and MgO have been strongly depleted close to the vein. SiO<sub>2</sub> and CaO are locally depleted and reconcentrated.

Disseminated pyrite, pyrrhotite and much lesser chalcopyrite occur within envelopes for up to several metres

from the veins. Arsenopyrite is confined to vein selvages. Minor amounts of sphalerite and especially galena appear to correlate with vein sections that are richer in gold. Traces of tetrahedrite and stibnite have been observed but tellurides have not. Gold is principally found as thin smeared flakes of the native metal in the black sulphidic septae of the strongly ribboned shear veins. Gold is only rarely found by itself in the quartz, usually in the rare extensional veins, where it forms extremely rich pockets.

**FLUID INCLUSION STUDIES**

Samples for fluid-inclusion studies were collected from quartz veins at surface and over a 2-kilometre depth extent; in general, the sites were the same as those sampled for oxygen isotope measurements (Figure 2-2-1a). Vein quartz forms

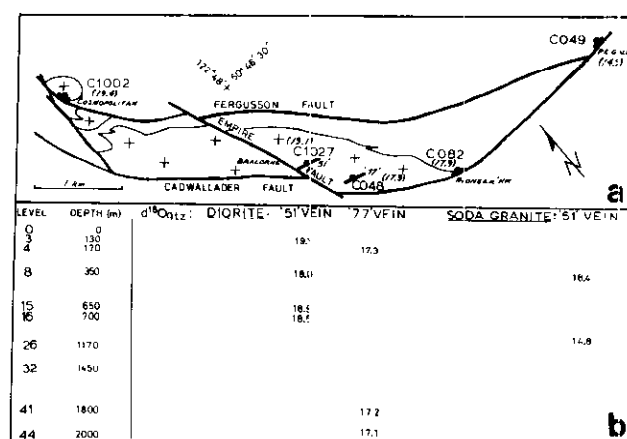


Figure 2-2-1a. Geographic zoning of oxygen isotope ratios of vein quartz from the Bralorne-Pioneer vein system. Plus symbols indicate extent of Bralorne intrusives, sample numbers are C1002, etc.; d<sup>18</sup>O values are in brackets, vein designation is in inverted commas, with vein strike and steep northerly dip indicated.

Figure 2-2-1b. Vertical zoning in oxygen isotope ratios in the Bralorne-Pioneer vein system is shown schematically, by variation in d<sup>18</sup>O quartz with reference to the mine levels.

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ehedral crystals up to 1 centimetre long that are outlined by concentric growth zones of minute (1-2 micron diameter) primary fluid inclusions. None of these inclusions were large enough for microthermometric measurements; instead, larger isolated (assumed primary) and fracture-controlled (pseudosecondary) inclusions in the 3 to 10 (rarely up to 30) micron range were studied. Particles of gold were not seen directly associated with any fluid inclusions, but the primary and pseudosecondary inclusions are assumed to have accompanied the main and subsequent stages of mineralization, respectively. The pseudosecondary inclusions are interpreted as later, based on their mode of occurrence and their lower temperatures of homogenization.

Preliminary fluid-inclusion data for the Bralorne deposit were reported in Leitch and Godwin (1987, 1988). Further fluid-inclusion studies, carried out with a United States Geological Survey gas-flow stage adapted by Fluid Inc., confirm the bulk of the results obtained with the Chaixmecca equipment, particularly the final homogenization temperatures ( $T_h$ ) and salinities from ice-melting temperatures ( $T_{mi}$ ). However, the fluid compositions of the fluids in the inclusions can now be characterized in greater detail. This is due to: (1) the increased visibility of the inclusions because of the better optics with the adapted stage, leading to recognition of additional phases, either optically or by observation of phase transitions, and (2) the increased precision of measurements of phase-transition temperatures by the "cycling" technique and the rapid response of the gas-flow system. This high precision is accompanied by a high degree of accuracy, since the thermocouple/readout unit is calibrated to better than  $0.4^\circ\text{C}$  from  $-56.6^\circ\text{C}$  to  $+660.4^\circ\text{C}$ .

Some of the scatter observed, for instance in the  $\text{H}_2\text{O}:\text{CO}_2$  ratios and final homogenization temperatures (Figure 2-2-2), is possibly due to necking of the inclusions after they formed. Such inclusions were avoided, but textural evidence for necking may not always be present. Useful information may still be obtained from these inclusions; Roedder (1979), demon-

strated that the true homogenization temperature will be intermediate to the homogenization temperatures of the necked parts.

Results are summarized in Table 2-2-1 and in Figure 2-2-2. They show that the fluids contain a significant carbonic ( $\text{CO}_2 + \text{CH}_4$ ) component.

## CLASSIFICATION OF FLUID INCLUSIONS

**Type 1a** isolated primary inclusions in quartz, marked "P" in Figure 2-2-2, usually homogenize to liquid. They contain moderate amounts of carbonic fluid (0.1 to 0.4 mole fraction of the total contents of the inclusions, with a mode of 0.15), and consist of carbon dioxide and minor methane ( $X_{\text{CO}_2} = 0.10$ ,  $X_{\text{CH}_4} = 0.05$ ).

TABLE 2-2-1: SUMMARY OF FLUID INCLUSION CHARACTERISTICS FOR THE BRALORNE DEPOSIT (all temperatures in  $^\circ\text{C}$ ).

Type	Description
<b>Type 1a Primary</b>	<b>Three-phase, <math>\text{CO}_2</math>-bearing, <math>\text{CH}_4</math>-poor</b> ( $\text{H}_2\text{O}$ 0.8, $\text{CO}_2$ 0.1, $\text{CH}_4$ 0.05, $\text{NaCl}$ 0.03) <sup>1</sup> $T_h$ : range 235 to 425, average <sup>2</sup> 280 $T_{h\text{CO}_2}$ : range 15 to 27, mode <sup>3</sup> 20.5 $T_{m_{cl}}$ : range 8.2 to 11.0, mode 9.6 $T_{m_i(?)}$ : range -2 to -7, average -3.5 $T_{m_{\text{CO}_2}}$ : range -57.3 to -59.8, mode -58.8 $V/V+L$ : range 10-45 vol.%, mode 20 vol.% $r$ carbonic: $0.7\text{g}/\text{cm}^3$ $r$ bulk: $0.95\text{g}/\text{cm}^3$ $V_{bar}$ : 23 moles/ $\text{cm}^3$
<b>Type 1b</b>	<b>Three-phase, <math>\text{CO}_2</math>-<math>\text{CH}_4</math>-rich</b> ( $\text{H}_2\text{O}$ 0.45, $\text{CO}_2$ 0.25, $\text{CH}_4$ 0.25, $\text{NaCl}$ 0.03) $T_d$ : range 230 to 330, average 270 $T_{h\text{CO}_2}$ : range 3.3 to 9.0, mode 7.5 $T_{m_{cl}}$ : range 11.1 to 13.0, mode 12.4 $T_{m_i(?)}$ : not observable $T_{m_{\text{CO}_2}}$ : range -61.3 to -66.5, mode -63(?) $V/V+L$ : range 30-85 vol.%, mode 50 vol.% $r$ carbonic: $0.6\text{g}/\text{cm}^3$ $r$ bulk: $0.8\text{g}/\text{cm}^3$ $V_{bar}$ : 31 moles/ $\text{cm}^3$
<b>Type 2 Pseudosecondary</b>	<b>Two-phase, <math>\text{H}_2\text{O}</math>-liquid rich</b> ( $\text{H}_2\text{O}$ 0.95, $\text{CO}_2$ 0.05, $\text{NaCl}$ <0.01) $T_h$ : range 160 to 260, average 200 $T_{h\text{CO}_2}$ : range 20 to 31, mode 27.5 $T_{m_{cl}}$ : range 7.0 to 10.5, mode 9.5 $T_{m_i(?)}$ : range -0.5 to -2.5, average -1.8 $T_{m_{\text{CO}_2}}$ : range -56.6 to -57.5, mode -56.8 $V/V+L$ : range 5-15 vol.%, mode 10 vol.% $r$ carbonic: $0.7\text{g}/\text{cm}^3$ $r$ bulk: $0.97\text{g}/\text{cm}^3$ $V_{bar}$ : 20 moles/ $\text{cm}^3$
<b>Type 3 Secondary</b>	<b>Two-phase, <math>\text{H}_2\text{O}</math>-liquid rich</b> ( $\text{H}_2\text{O}$ 0.99, $\text{NaCl}$ <0.01) $T_h$ : range 120 to 180, average 150 $T_{m_i}$ : range -0.1 to -1.0, average -0.5 $V/V+L$ : 2-7 vol.%, mode 5 vol.% $r$ bulk: $1.00\text{g}/\text{cm}^3$ $V_{bar}$ : 18 moles/ $\text{cm}^3$

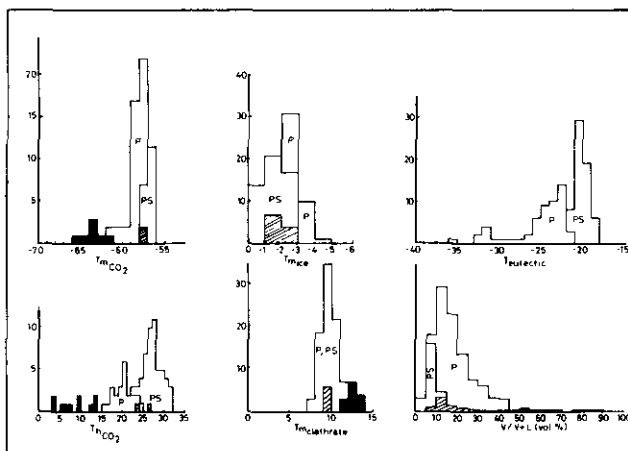


Figure 2-2-2. Summary of data for carbonic fluid inclusions in the Bralorne mesothermal gold-quartz vein deposit. P, PS, S correspond to primary, pseudosecondary, and secondary inclusions respectively; black areas to methane-rich (Type 1b) inclusions. All inclusions are in quartz except for hatched areas which are inclusions in calcite. Other abbreviations are in Table 2-2-1.

<sup>1</sup> Approximate mole fractions.

<sup>2</sup> Averaged from histogram modes in Leitch and Godwin (1988). Mean value is 280, standard deviation  $\pm 36$ , 276 measurements.

<sup>3</sup> Mode from histograms in Figure 2-2-1.

<sup>4</sup> Abbreviations:  $T_h$  = final homogenization temperature;  $T_d$  = decrepitation temperature;  $T_{h\text{CO}_2}$  = homogenization temperature of  $\text{CO}_2$ ;  $T_{m_{cl}}$  = final melting point of clathrate;  $T_{m_i}$  = final melting point of ice;  $T_{m_{\text{CO}_2}}$  = final melting point of  $\text{CO}_2$ ;  $V/V+L$  = ratio of vapour bubble to liquid;  $r$  = density;  $V_{bar}$  = molar volume.

**Type 1b** inclusions in quartz, shown in solid black in Figure 2-2-2, usually homogenize to vapour. They are vapour-rich and contain abundant methane and carbon dioxide (0.3 to 0.9 mole fraction carbonic component of the total contents, with a mode of 0.5), consisting of  $X_{\text{CH}_4} = 0.25$ ,  $X_{\text{CO}_2} = 0.25$ .

**Type 2** inclusions in quartz are pseudosecondary (marked "PS" in Figure 2-2-2). They homogenize to the liquid phase, and contain no methane and only minor carbon dioxide (mode of less than 0.1 mole fraction of the total contents), that is not often visible as a separate phase but may be inferred from clathrate melting temperatures. Primary inclusions in calcite (hachured in Figure 2-2-2) have similar characteristics to pseudosecondary inclusions in quartz.

**Type 3** inclusions in both quartz and calcite are secondary (S in Figure 2-2-2), and are localized along through-going fractures. They have no detectable carbonic behaviour and homogenize to the liquid phase.

## DENSITIES

Data in Table 2-2-1 and Figure 2-2-2 indicate that the carbonic (non-aqueous) portions of Type 1b (methane-rich) inclusions have lower densities (0.60) compared to Type 1a carbon-dioxide-rich inclusions (0.70) and to Type 2 inclusions (0.70). These densities are based on the temperatures of homogenization of the carbonic portion of the inclusion ( $T_{\text{HCO}_2}$ ), and data in diagrams from Swanenberg (1979) and Hollister (1981). The homogenization temperatures of the carbonic fluid, difficult to measure reliably for Type 1b inclusions because of the overlap with the clathrate melting temperatures, are well defined for Type 1a and Type 2. Bulk densities, equivalent mole fraction  $\text{CO}_2$ , and molar volumes ( $V_{\text{bar}}$ ) for the inclusions were estimated from Brown and Lamb (1986), assuming a density of 1.02 grams per cubic centimetre weight per cent for a 3 weight per cent equivalent NaCl solution (Potter and Brown, 1977) and using Swanenberg's method of computing equivalent  $\text{CO}_2$  for inclusions containing both methane and carbon dioxide.

## SALINITIES

Salinities of primary and pseudosecondary fluid inclusions at Bralorne are difficult to estimate because of variable amounts of both carbon dioxide and methane (compare Collins, 1979). The withdrawal of water attendant upon the formation of clathrates causes the aqueous solution to become more saline, depressing the melting point of ice. Thus the  $T_{\text{mi}}$  data presented in Leitch and Godwin (1988) for Type 1 inclusions (5 weight per cent NaCl equivalent), and for Type 2 inclusions (2 weight per cent NaCl equivalent), overestimates the salinity of the aqueous solutions (using the equation of Potter *et al.*, 1978). If only carbon dioxide is present, the salinity can be correctly estimated from clathrate melting temperatures (Bozzo *et al.*, 1973). A clathrate melting temperature of  $+9.5^\circ\text{C}$  (mode from P, PS peak in Figure 2-2-2), suggests 1 weight per cent NaCl equivalent for the Type 2 inclusions in quartz and calcite. Ice melting is not seen in the Type 1b methane-rich inclusions and the clathrate melting temperature ( $12.4^\circ\text{C}$ ) is well above  $10^\circ\text{C}$  (the presence of methane has an opposite effect on the clathrate melting temperature from that of dissolved salt), so the

method of Bozzo *et al.* is not applicable. Using the method of Linnen (1985), the positive correlation between mole fraction  $\text{CH}_4$  and  $T_{\text{mi}}$  in Type 1a and 1b inclusions suggests roughly equivalent salinities. The salinity of the Type 1 inclusions can therefore only be estimated to be between 1 and 5 weight per cent NaCl equivalent; hence a value of 32 weight per cent is assumed. Type 3 inclusions have ice melting temperatures just below that of pure water ( $-0.5^\circ\text{C}$ ), implying a salinity of about 0.8 weight per cent NaCl equivalent.

## COMPOSITIONS

Estimates of the salt composition of ore fluids at Bralorne were hampered by an inability to clearly see eutectic (first melting) temperatures. Vaguely detectable eutectic temperatures in inclusions in quartz (Figure 2-2-2) show modes at  $-20.5^\circ$ ,  $-23^\circ$  and possibly  $-32^\circ\text{C}$ . When compared to eutectic temperatures for different salt-water systems (for example, Roedder, 1984), these mean temperatures indicate that the fluids in Type 2 inclusions probably contained only NaCl, but that fluids in Type 1 inclusions may have also had minor amounts of KCl. This conclusion is supported by the computer modelling of the ore fluid (see below), which suggests a Na:K ratio of about 8:1. The uncertain eutectic temperature of  $-32^\circ\text{C}$  might indicate the presence of a divalent cation, such as magnesium (compare Crawford, 1981). Occasionally observed eutectic temperatures in primary inclusions in calcite (Leitch and Godwin, 1988) of about  $-26^\circ\text{C}$  may also indicate the presence of magnesium (or possibly calcium).

## PRESSURE ESTIMATES

Bulk density estimates of the inclusion fluids, and specifically for the carbonic portion of the various types of inclusion, serve to define isochores on a pressure-temperature (P-T) plot. Assuming pure  $\text{CO}_2$  in the carbonic fluid, the isochores may be projected to temperatures of entrapment (estimated from  $T_{\text{H}}$ ), providing estimates of entrapment pressures (Hollister and Burruss, 1976). Similarly, if temperatures of entrapment are estimated independently from sulphur isotope fractionations between coexisting galena-sphalerite pairs as  $350^\circ\text{C}$  for primary inclusions and  $250^\circ\text{C}$  for pseudosecondary inclusions (Leitch and Godwin, 1988), estimated entrapment pressures are about 150 and 75 megapascals (1.5 and 0.75 kilobars) respectively. Pressure estimates for entrapment at  $350^\circ$  and  $250^\circ\text{C}$  based on the simple assumption of pure  $\text{H}_2\text{O}$  fluids, are 100 and 50 megapascals respectively (Leitch and Godwin, 1988).

Type 1b (vapour-rich), and to a lesser extent Type 1a inclusions, decrepitate at temperatures ranging from  $230^\circ$  to  $330^\circ\text{C}$  (most decrepitated before homogenizing). Decrepitation over such a small temperature range is consistent with rapid pressure increases in carbonic fluids at temperatures above  $250^\circ\text{C}$  (Malinin, 1974). Since internal pressures of up to 120 megapascals (1.2 kilobars) are required to decrepitate 12 to 13 micron size inclusions in quartz, and up to 270 megapascals for smaller inclusions (Leroy, 1979), a minimum trapping pressure of at least 100 megapascals is implied for the 3 to 10 micron Bralorne inclusions, in agreement with the pressure estimates from projection of isochores.

The pressure-temperature conditions of entrapment of the primary and pseudosecondary fluids at Bralorne can also be evaluated using solubility relationships for H<sub>2</sub>O and CO<sub>2</sub> in salt solution (Bowers and Helgeson, 1983a). Fluids in Type 1a inclusions, with mole fractions of XCO<sub>2</sub> = 0.1 and XCH<sub>4</sub> = 0.05, salinities of about 3 weight per cent NaCl equivalent, and Th less than or equal to 300°C, would have been supercritical at pressures above 150 megapascals. If the 0.95 gram per cubic centimetre isochore is projected to the solvus for these inclusions at the estimated trapping temperature of 350°C, the pressure of entrapment is constrained to 175 megapascals (Bowers and Helgeson, 1983b). For primary Type 1b fluids, with mean carbonic mole fraction of 0.5, estimated mean bulk density of 0.8 gram per cubic centimetre and salinity of 32 weight per cent NaCl equivalent, decrepitation temperatures of 230° to 300°C give minimum estimates of internal (and, therefore trapping) pressures of 150 megapascals.

TABLE 2-2-2  
SUMMARY OF PRESSURE ESTIMATES  
FOR THE BRALORNE DEPOSIT

Fluid Type	Primary		Pseudosecondary
	(1a)	(1b)	(2)
L&G (1988)	1.0 (100)		0.5 (50)
Leroy (1979)	>1.0 (>100)	>1.0 (>100)	
H&B (1975)	1.5 (150)		0.75 (75)
B&H (1983b)	1.75 (175)	1.5 (150)	1.0 (100)

References: L&G=Leitch and Godwin (1988); H&B=Hollister and Burruss (1975); B&H=Bowers and Helgeson (1983b) Pressures in kb; bracketed figures are in megapascals

The presence of occasional vapour-rich, methane-rich Type 1b inclusions with the far more abundant vapour-poor, methane-poor Type 1a inclusions raises the possibility of trapping at subcritical temperatures. However, Type 1b inclusions are so rare in comparison to Type 1a that most trapping was probably at or above the solvus. Also, continued fault movement at lower temperatures could have produced heterogeneous populations of inclusions when earlier inclusions were broken.

Similarly, Type 2 pseudosecondary fluids, with XCO<sub>2</sub> = 0.05, mean salinity of 1 weight per cent NaCl equivalent, and mean Th of 200°C, would have been supercritical above 75 megapascals (Bowers and Helgeson, 1983a). If the 0.97 gram per cubic centimetre isochore is projected to the solvus for these inclusions at the estimated trapping temperature of 250°C, the pressure of entrapment is constrained to 100 megapascals (Bowers and Helgeson, 1983b).

In summary, pressures of entrapment that consider the combined CO<sub>2</sub>-CH<sub>4</sub> compositions of the fluids (Bowers and Helgeson, 1983a,b) imply higher pressures (from 100 to 175 megapascals) than those that consider the inclusions as pure carbon dioxide (Hollister and Burruss, 1975) at 75 to 150 megapascals, or from the pressure corrections necessary to correlate homogenization temperatures of aqueous inclusions with temperatures from sulphur isotopes (50 to 100 megapascals). Estimated pressures of entrapment for pseudosecondary and primary fluids are compared in Table 2-2-2.

## EVOLUTION OF MINERALIZING FLUIDS

There appears to be a progression in fluid composition at Bralorne from Type 1 to Type 2 to Type 3. Early, Type 1 primary fluids were high temperature (350°C), carbon dioxide and methane rich (XCO<sub>2</sub> + XCH<sub>4</sub> = 0.15), with low salinities (approximately 3 weight per cent NaCl equivalent). Later, Type 2 pseudosecondary fluids were lower temperature (250°C), with less carbon dioxide (XCO<sub>2</sub> = 0.05), no methane, and had lower salinities (less than 1 weight per cent NaCl equivalent). Type 3 (secondary) fluids were coolest (180°C), with carbon dioxide below the detection limit of 0.03 mole fraction, but the same low salinity as the pseudosecondary fluids. The evolution of the fluids can be explained by a simple mixing of hotter, more saline, carbon dioxide and methane-rich fluid with progressively greater amounts of cold, more dilute meteoric water over time, as the hydrothermal system waned.

The estimated pressures of entrapment are lower for Type 2 pseudosecondary inclusions than for Type 1 primary inclusions. Also, the geothermal gradient was probably lower (10°C per kilometre) at the time of entrapment of the pseudosecondary fluids than it was for the primary fluids (30°C per kilometre; Leitch and Godwin, 1988). These two features suggest that the deposit had been partly unroofed and the rock mass had cooled by the time of entrapment of the fluids in the pseudosecondary, and finally secondary, inclusions.

If the higher pressures estimated above for the primary and pseudosecondary inclusions (Table 2-2-2), which are consistent with calculations of Leroy (1979), are correct, then the early (main) mineralizing event took place at depths of 6 to 10 kilometres from fluids of Type 1. Later mineralization took place at depths of 2 to 6 kilometres from fluids of Type 2. These pressures and depths are comparable to those estimated by Smith *et al.* (1984: 130 to 290 megapascals, or 5 to 12 kilometres) for a similar mesothermal gold vein deposit at Timmins, Ontario. These figures have since been questioned by Brown and Lamb (1986), who suggested much higher pressures (350 to 800 megapascals; 15 to 30 kilometres depth), based on extrapolation to elevated temperature and pressure of isochores calculated from molar volumes at room temperature, rather than from molar volumes at the elevated conditions. Such pressures seem unrealistically high for the Bralorne camp, if the geothermal gradient was as high as 30°C per kilometre (Leitch and Godwin, 1988).

## OXYGEN ISOTOPE STUDIES

One hundred oxygen isotope analyses (including 11 duplicates) of mineral separates and whole-rock samples from 15 different vein systems in the deposit were obtained, in order to: establish lateral and vertical zonations within the deposit; derive estimates of the temperature of alteration and mineralization independent of the fluid inclusion and sulphur-isotope studies; examine fluid-wallrock interactions and water:rock ratios; and estimate the isotopic compositions and origin of the mineralizing fluids. These isotopic data are presented in Tables 2-2-3 and 2-2-4. Vein quartz was sampled at surface over a distance of almost 6 kilometres from the Cosmopolitan vein at the northwest end, to the P.E. Gold



TABLE 2-2-4  
DUPLICATE OXYGEN ISOTOPE DATA  
FOR THE BRALORNE-PIIONEER DEPOSIT

Sample No.	Mineral <sup>1</sup>	Original d <sup>18</sup> O	Duplicate d <sup>18</sup> O	Average d <sup>18</sup> O	Variation (±)
8-51B(FW)	qz	18.04	17.90	17.97	0.07
C111-28	qz	18.31	18.42	18.36	0.05
15-51(C)	qz	18.86	19.00	18.93	0.07
16-51	qz	18.10(E)	18.44(C)	18.27	0.17
C118-11	qz*	15.70	15.33	15.52	0.18
C049	qz	14.93	14.84	14.90	0.04
C082	qz	17.87	18.03,18.11	18.00	0.12
C1027	qz	19.19	19.08	19.13	0.06
C128-20	qz	17.24	17.03	17.13	0.10
C116-3/16	qz	17.20	17.39	17.29	0.09
C033-1/2	se/fu	14.81(se)	14.58(fu)	14.69	0.11

Mean variation = 0.09

<sup>1</sup> Abbreviations: qz = quartz, se/fu = sericite or fuchsite.

for vein quartz (+14.9) in the southeast to higher d<sup>18</sup>O (+19.4) in the northwest (Figure 2-2-1a). However, samples from the Pioneer mine and from the 77 vein at the Bralorne mine are almost within analytical error. The d<sup>18</sup>O of quartz from the P.E. Gold vein is lower than any of the other surface samples. The observed geographic variation in d<sup>18</sup>O of quartz may reflect spatial variations in d<sup>18</sup>O of the mineralizing fluids, or variations in temperature from hottest at the southeast to coolest at the northwest. Mineralogical data (biotite is more common towards the southeast: Leitch and Godwin, 1988; Joubin, 1948; Nordine, 1983) and structural data suggesting a deeper level of exposure to the southeast (Joubin, 1948) are consistent with a geographic variation in temperature of vein formation, at the level of exposure sampled.

A similar variation in d<sup>18</sup>O of quartz, where d-values decrease with depth, is suggested by data in Figure 2-2-1b. Highest values of d<sup>18</sup>O are found at greater elevations in the system (Cosmopolitan vein at surface); lowest values are in the deepest levels (41 and 44 levels, 1800 to 2000 metres depth).

A detailed traverse across the 51 vein on 16 level (samples 1 to 7 in Table 2-2-3) was made to test for small-scale variations in the vein quartz. However, there was no significant change across the 20-centimetre width sampled, so the vein quartz at Bralorne appears to be relatively homogeneous in terms of its d<sup>18</sup>O.

## GEOOTHERMOMETRY

Mineral pairs of quartz-sericite and quartz-chlorite were analysed from veins and wallrocks to estimate the temperature of mineralization based on oxygen isotope fractionations. Other mineral pairs (quartz-albite and quartz-hornblende) were analysed to estimate the temperature of emplacement of the host Bralorne intrusions. All these mineral pairs were observed intergrown with each other and are assumed to have formed in equilibrium. Temperatures of mineralization, calculated with the relevant fractionation equation, are presented in Table 2-2-3. In the following discussion, the term "unaltered" is used for rocks that are not

visibly hydrothermally altered (C093A and C094A), although their isotopic ratios and lower greenschist mineral assemblages show that they are not truly fresh.

For **quartz-sericite**, the equation of Bottinga and Javoy (1975) was used rather than that of Clayton *et al.* (1972) combined with that of O'Neil and Taylor (1969), because the latter approach gave unreasonably low temperatures. Calculated temperatures vary from 360° to 405°C for milky quartz from the Bralorne 51 vein on 8 level (350 metres depth), 86 vein on 26 level (1200 metres depth), and Pioneer HW Main vein (at surface). A sample of clear quartz (Bralorne 51 vein on 8 level) and associated, but not intergrown, mica yielded a high temperature of 470°C. This quartz is paragenetically distinct, forming clear euhedral crystals that also have anomalously high homogenization fluid-inclusion temperatures (Leitch and Godwin, 1988). The quartz-sericite pair from the 79 vein (41 level) gives an unrealistically high temperature of 845°C. However, contamination of the sericite by fine-grained quartz is likely since the separate was not hand-picked and the sericite d<sup>18</sup>O value of 16.0 is 2 to 3 per mil higher than all other sericites. For altered wallrocks, estimates of mineralization temperature range from 530° to 560°C for the 51 vein on 15 level (650 metres below surface) to 560°C for the same vein on 8 level (Table 2-2-3).

With the quartz-chlorite equation of Wenner and Taylor (1971) most of the **quartz-chlorite** pairs analysed give either unrealistically high temperatures or are reversed (provide no valid temperatures). Only one sample, C002FW4, from altered wallrock around the 51 vein on 8 level, gave a geologically reasonable temperature of 325°C. Isotopic disequilibrium indicated between the quartz-chlorite vein minerals may stem from crystallization at different times and/or re-equilibration of the chlorite with subsequent fluids at lower temperatures.

Using the **quartz-albite** pair (Matthews *et al.*, 1983), a weakly altered sample, 10 metres from the 51 vein on 15 level (C033-9), gives a temperature of 290°C, which appears to be strongly isotopically reset towards lower temperatures by the hydrothermal alteration. Unaltered "fresh" wallrock (C093A) gives a possibly magmatic quartz-albite temperature of 730°C.

For **quartz-hornblende** (Bottinga and Javoy, 1975), hornblende from unaltered Bralorne diorite (C093A), well removed from the main veins, gives a similar temperature (340°C) to that from quartz-hornblende in C033-9, 10 metres from the 51 vein on 15 level, which gives 350°C. Both these figures are extrapolated slightly outside the listed range of the equation. However, as for the quartz-albite pairs, they imply a high degree of interaction of the wallrock with the altering fluids, (that is: a high water:rock ratio). The interaction is apparently more marked for the hydrous minerals, chlorite and hornblende, than for albite.

## IMPLICATIONS FOR WATER:ROCK RATIOS

Samples from five detailed traverses across altered wallrocks at several levels within the mine, were analysed for whole-rock oxygen isotope ratios to test for isotopic zoning. Distances of samples from the vein are in Table 2-2-3; the

width and style of the alteration envelopes are described in Leitch and Godwin (1988). Along two of these traverses, oxygen isotope measurements were also made on minerals.

Whole-rock and mineral oxygen isotope ratios increase as the veins are approached (Table 2-2-3). For instance, in the series C033-1 to 10, taken through altered diorite near the 51 vein on 15 level, there is a progression from  $d^{18}\text{O} = +10.6$  in unaltered rock (estimated by C093A), to  $+18.9$  immediately adjacent to the vein. The minor reversal between 7 and 8 is due to a subsidiary fracture off the main vein. The  $d^{18}\text{O}$  varies in a similar way among samples from the other two series in diorite host rocks (51 vein on 8 level, C002FW1 to FW4; 51 vein on surface, C1027HW1 to HW3).

Whole-rock  $d^{18}\text{O}$  values are also changed for at least 3.5 metres from the 51 vein on 8 level where it is hosted by a rock rich in quartz (soda granite). Values remain relatively constant at about 13.7 to 13.9 over this interval, compared to the unaltered rock at 12.7, and a vein  $d^{18}\text{O}$  of 18.4. There is thus a clear pattern of widespread re-equilibration of the whole-rock  $d^{18}\text{O}$  values in diorite and soda granite, due to hydrothermal alteration, for up to 10 metres from the veins.

Oxygen-isotope gradients are apparently not as pronounced at deeper elevations (lower mine levels) as for upper levels. On 41 level, 1600 metres below surface, around the 77 vein, the C116-23 to 18 series shows that there is a small increase from a value of 11.7 at 10 metres from the vein, to a plateau value of about 13.0 within 5 metres of the vein. Vein quartz here has a  $d^{18}\text{O}$  value of 17.1. On 44 level, 1800 metres below surface, the C128 series shows very little or no variation within the limits of measurement:  $d^{18}\text{O}$  whole-rock values remain at about 11 to 12. Both these traverses are in much more mafic wallrock than the typical diorite. These more mafic variants contain less than 5 per cent quartz, and are principally composed of large amounts of clinopyroxene, hornblende, or their alteration products (chlorite).

Quartz in the altered envelopes on 8 and 15 levels has a high  $d^{18}\text{O}$  compared to quartz in the unaltered rock. Quartz  $d^{18}\text{O}$  values increase systematically towards the veins (that is 51 vein on 15 level, C033 series, from 18.9 to 14.3; Table 2-2-3). Similarly, for the C002FW1 series around the 51 vein on 8 level, the variation is 18.0 to 14.3. The other major rock-forming minerals in the diorite, plagioclase and hornblende, show increases in  $d^{18}\text{O}$  toward the veins (to within 10 metres) as long as they are stable. The variation in  $d^{18}\text{O}$  of plagioclase is from 13.8 to 14.5, and for hornblende from 6.2 to 8.3.

In summary, the whole-rock  $d^{18}\text{O}$  patterns in hydrothermally altered wallrock envelopes may be largely due to the amount of quartz added during the alteration of diorites and soda granites, which as shown, is shifted towards higher  $d^{18}\text{O}$  values closer to the vein. In the more mafic host rocks at deeper levels, the lack of alteration quartz may explain the smaller shifts observed in whole-rock  $d^{18}\text{O}$  values. The strong shifts in  $d^{18}\text{O}$  values for both mineral separates and whole-rock samples imply high water:rock ratios during the alteration, a feature common to other carbonate alteration zones around mesothermal gold quartz veins (Kerrick, 1983; Taylor, 1987).

## OXYGEN-ISOTOPE COMPOSITION OF THE ORE FLUID

Oxygen isotope compositions of mineralizing fluids were calculated from isotopic analyses of vein quartz, trapping temperatures estimated from fluid-inclusion studies, and the quartz-water fractionation equation of Clayton *et al.* (1972). Calculated values of  $d^{18}\text{O}$  fluid are given in Table 2-2-3.

The  $d^{18}\text{O}$  of the ore fluid appears to have been reasonably constant at 131 per mil from bottom to top of the vein system. Two quartz samples, from the P.E. Gold vein and the 51 vein on 26 level have lower  $d^{18}\text{O}$  fluid values. The vertical spatial variation in  $d^{18}\text{O}$  of vein quartz (decreasing  $d$ -values with depth) is thought due to a gradual increase in temperature with depth rather than variation in  $d^{18}\text{O}$  fluid.

The small-scale variation above, is similar to the regional lateral variation in observed  $d^{18}\text{O}$  of quartz described by Nesbitt *et al.* (1987). Deposits are zoned from the high-temperature Bralorne-type gold-quartz veins with  $d^{18}\text{O}$  quartz =  $17.5 \pm 1.0$ , to intermediate temperature antimony-silver-gold veins with  $d^{18}\text{O}$  quartz =  $21.0 \pm 1.0$ , to low-temperature mercury deposits with  $d^{18}\text{O}$  quartz =  $29.0 \pm 2.0$ , by deposition from the same or similar deeply circulating, highly evolved ( $d^{18}\text{O}$ -shifted) fluids of around 11.52 per mil (corrected to the estimated trapping temperature of  $350^\circ\text{C}$ ), which is within analytical uncertainty of the value obtained for Bralorne in this study. The strong enrichment in  $d^{18}\text{O}$  values of quartz, combined with D/H studies of fluid inclusions, led Nesbitt *et al.* (1985, 1987) to propose that the ore-forming fluids were composed simply of meteoric water that had circulated deeply during regionally extensive transcurrent faulting. The calculated  $d^{18}\text{O}$  fluid of  $+13$  per mil at Bralorne is virtually identical to the values for the Coquihalla deposits near Hope, British Columbia and the Mother Lode deposits in California (Taylor, 1987), and is in the middle of the range for metamorphic waters. However, the concept of "metamorphic water" must be clarified, as its original isotopic definition (Taylor, 1974) is not meant to imply water of dehydration (*see* Taylor, 1987). Inasmuch as most water/rock reactions involve either sea water or meteoric water, the evolved meteoric water hypothesis seems reasonable but might be further constrained by data from hydrogen isotope studies (in progress). For example, in similar deposits in the Mother Lode of California, sericite and mariposite associated with alteration and gold mineralization formed from waters with somewhat higher  $d\text{D}$  than waters in some fluid inclusions in adjacent quartz veins, indicating mixing of waters with different evolutionary histories (*see* Taylor, 1987).

The hypothesis of meteoric water (Nesbitt *et al.*, 1987) *is* the principal ore fluid does not explain the abundance of carbon dioxide in the ore fluids. Such high carbon dioxide contents are consistent with fluids produced by metamorphic devolatilization at amphibolite to granulite facies, and have been interpreted as metamorphically derived fluids (Kerrick, 1983; Colvine *et al.*, 1984). A more complex, mixed origin for hydrogen-oxygen-carbon-sulphur in the mineralizing fluids must be considered for fluids circulating along principal structural and tectonic boundaries. For example, emplacement of mafic magmas along major tectonic zones provides a source of carbon dioxide isotopically similar to that found in the Mother Lode (Taylor, 1987).

## THERMOCHEMICAL MODELLING OF WATER/ROCK REACTION

### FLUID CHARACTERISTICS

The chemical characteristics of the ore-forming fluid at Bralorne were modelled using the PATH computer program (Helgeson *et al.*, 1970; Perkins, 1980). Thermodynamic data from Helgeson (1969) were used for the aqueous species and from Helgeson *et al.* (1978) for solid phases. Gold chloride complexes were included but not thiosulphide complexes, although if the latter were included the results would not change except to increase the total gold in solution (Seward, 1973). The program models the ore fluid by progressive titration, or step-wise water/rock reactions, first calculating the fluid that would be in equilibrium with the observed alteration assemblage, then "reacting" the fluid with "fresh" rock of the observed composition. The program writes detailed reactions for each step in the process, considering as many as 25 reactants and as many products.

Any model for mineralization at the Bralorne deposit must account for the strong, widespread carbonate alteration accompanying ore deposition. The typical mineral assemblage in altered wallrock includes quartz, muscovite, dolomite, calcite, albite, chlorite, pyrite and native gold. Thermodynamic data for arsenic compounds are not available, so the implications of the arsenopyrite commonly seen in the vein assemblage cannot be assessed. The ore fluid in equilibrium with the observed alteration assemblage is characterized in Table 2-2-5. Except for the carbon dioxide fugacity, the characteristics are similar to those described by Helgeson and Garrels (1968) for deposition of gold, pyrite and quartz in response to temperature drop. The high carbon dioxide content is dictated by the carbonate alteration in the present study, supporting gold deposition by reaction with the wallrock. The gold content of the ore fluid predicted by our model (roughly 0.1 to 0.2 ppb), is also much lower than previous estimates. Our estimate is much closer to the average from mineralized areas of 0.1 ppb, measured by McHugh (1988). Such low predicted gold contents emphasize the importance of understanding gold depositing mechanisms, rather than transporting mechanisms, in order to adequately explain the formation of large gold deposits like Bralorne.

In order to model chemical reaction between wallrock and ore fluid, the Bralorne diorite was assumed to comprise an original mineral assemblage of 60 per cent albite, 6 per cent tremolite and 34 per cent ferrosilite (thermodynamic data are not available for hornblende, a compositionally complex mineral). The temperature and pressure constraints applied were constant at 300°C and 100 megapascals (selected to be as close as possible to the fluid inclusion information without being out of the range of the experimental data in PATH). The alteration mineralogy predicted by the program fits well with the sequence observed outwards from the veins at Bralorne. Close to the vein, the model predicts dissolution of the starting minerals (albite and tremolite + ferrosilite), and precipitation of major amounts of quartz, dolomite and muscovite, minor amounts of pyrite, and minute amounts of native gold. Gold precipitation is most favoured in the early steps of the process; the bulk of the gold (about two-thirds of the total) is deposited immediately adjacent to the vein as the

TABLE 2-2-5  
CHARACTERISTICS OF THE ORE FLUID AT THE  
BRALORNE DEPOSIT, PREDICTED BY THE PATH PROGRAM

Temperature = 350°C <sup>1</sup>		Pressure = 1.75 Kb <sup>1</sup>	
pH = 4.5 (slightly acid at 350°C and 1.75 Kb)			
Na/K = 8:1	[Na] = 0.4 m	[K] = 0.05 m	[Cl] = 0.5 m
(Total salinity = 0.5 m, approximately 3 wt. %)			
$f_{\text{CO}_2} = 10^{2.5}$	$f_{\text{CH}_4} = 10^{0.5}$	$f_{\text{O}_2} = 10^{-30}$	$f_{\text{S}_2} = 10^{-7}$
[Fe <sup>++</sup> ] = $1 \times 10^{-7}$ m	[Mg <sup>++</sup> ] = 0.003 m	[Ca <sup>++</sup> ] = 0.01 m	
[S <sup>-</sup> ] = $10^{-10}$ m	[Au <sup>+</sup> ] = $10^{-10}$ m (0.1 ppb, as AuCl <sub>2</sub> <sup>-</sup> )		

<sup>1</sup> Estimated from fluid inclusion studies

carbon dioxide bearing fluids react with the wallrock. Gold precipitation is predicted to decrease sharply as soon as graphite (in trace amounts) becomes stable. Further from the vein, chlorite becomes stable (approximated thermodynamically by talc, for which data are available), and further out still, albite becomes stable where muscovite is no longer stable. Concurrent with these changes, as the fugacity of sulphur in the fluid drops, the stable sulphide becomes pyrrhotite rather than pyrite, and the precipitation of gold is no longer favoured. Where magnetite becomes stable (furthest from the vein), gold is dissolved. The model thus predicts a strong correlation of gold with pyrite and an absence of significant gold from pyrrhotite or magnetite-bearing assemblages.

The actual precipitation of the gold in the constructed PATH reaction model involves reduction of the aurous gold, Au<sup>+</sup>, in the AuCl<sub>2</sub><sup>-</sup> complex, to native gold, Au<sup>0</sup>, by donation of an electron. This process seems to be controlled by a concurrent oxidation reaction, of S<sup>-</sup> in H<sub>2</sub>S, to sulphur in pyrite, FeS<sub>2</sub>, which may be thought of as S<sub>8</sub>. This is suggested by the strong correlation in the model between Fe<sup>++</sup> consumption and pyrite production on the one hand, with gold produced on the other. Consideration of charge balance requirements for Equation 2 (Table 2-2-4) shows that in this reaction, the small extra amount of S<sup>2-</sup> (in H<sub>2</sub>S) being oxidized to S in pyrite (thought of as S<sup>-</sup>) is the same as the small amount of gold being precipitated. As soon as carbon (graphite) becomes stable (C<sup>4+</sup> + 4e<sup>-</sup> = C<sup>0</sup>), the competition for available electrons appears to sharply reduce the possibility of precipitating gold.

### COMPARISON OF PREDICTED AND OBSERVED MINERAL ASSEMBLAGES

Many of the features predicted above correspond to observed mineral assemblages. In the altered envelopes there is a transition outwards from a quartz-sericite-ankerite-pyrite zone to a chlorite-albite-calcite zone. Amorphous carbon is seen occasionally in altered wallrocks but is never abundant. Only occasionally, in a late-stage carbonate alteration that is black with fine amorphous carbon, does it form as much as 1 per cent of the rock.

Assay data at the mine show that gold concentrations drop off sharply immediately outside the veins. From hand-specimen and microscope studies, the bulk of the gold is in the thin black ribbons of the quartz veins, not in the milky white



quartz itself. In the ribbons, which are black due to finely divided pyrite (carbon has not been identified here), the gold is closely associated with the pyrite, plus arsenopyrite, sericite and minor ankerite. Pyrite and other sulphides are almost never seen in the quartz itself; the bulk of the sulphides are in the adjacent wallrocks and in the dark ribbons. Pyrrhotite is occasionally present in the altered wallrocks, but there is insufficient data to confirm the predicted inverse relationship between gold and pyrrhotite. Magnetite is not found in any rocks at Bralorne except as primary grains in the postmineral Bendor intrusives.

In detail, there is an empirically observed correlation between higher gold values and the presence of galena, and to a lesser extent, sphalerite (Joubin, 1948; Dolmage, 1934; Leitch and Godwin, 1988). Microscopically this is seen as gold, up to several hundred microns across, intergrown with galena in some of the Bralorne-Pioneer specimens, whereas the gold inclusions in pyrite and arsenopyrite tend to be smaller (less than 15 microns). The richest gold ore shoots are in diorite, peripheral to soda granite bodies, (Campbell, 1975; James and Weeks, 1961). This has formerly been explained by suggesting either a genetic relationship of ore to the soda granite (now disproved by isotopic dating: Leitch and Godwin, 1988), or by brittleness of the soda granite causing an inability to sustain large openings. However, the PATH model suggests that the more abundant iron in the diorite, the favoured host at Bralorne, and in the greenstone, the favoured host at Pioneer, could increase the precipitation of pyrite, and hence of gold.

#### FLUID MIGRATION AND MINERAL DEPOSITION

Any explanation for ore deposition at Bralorne must account for two notable features of the quartz veins: dark ribbons within the veins that contain the bulk of the gold; and syntaxial, coarse milky quartz crystals up to 1 centimetre long outlined by growth zones of minute primary fluid inclusions. Although many random orientations exist, these crystals are occasionally perpendicular to the ribbons or walls of the veins. This observation is supported by the strong induced piezo-electric-response of the quartz, which indicates alignment of the c-axes of many grains (M.M. Gomshei, personal communication, 1988).

The dark ribbons in the quartz imply a cyclic process, with repeated fracturing and precipitation of quartz. The coarse quartz crystals imply open-space growth under conditions of high fluid pressure. This might at first seem to be in conflict with the mesothermal environment of deposition (4 to 8 kilometres depth) where confining pressure is normally lithostatic. However, repeated fracturing and deposition of quartz at high hydrostatic pressures may be explained by the "fault-valve" model of Sibson *et al.* (1988). This model proposes that a geopressed reservoir of fluids derived by metamorphic devolatilization at amphibolite or granulite grade is developed by ponding below the ductile-brittle transition zone. This transition at the base of the seismogenic zone (the zone in which earthquakes are mainly concentrated) roughly corresponds to the transition to greenschist facies. Pore pressures become very high because steeply-dipping (55-75°) faults in a horizontally compressive stress field are in an inappropriate orientation for slip; the pressures

are a maximum for structures dipping at about 57 degrees (Sibson *et al.*, 1988). In order to permit slip, and therefore rupture and fluid flow along the steep faults which host the Bralorne veins, the pore pressure must have exceeded the lithostatic pressure by the amount of the cohesive strength of the already-cemented faults. Such extremely high fluid pressures provide the environment for open-space coarse crystal growth at depths of over 4 kilometres. When rupture occurs, fluid flows into the open space of the fault where the sudden drop in pressure may promote deposition of quartz (Walther and Helgeson, 1977) and sulphides (Helgeson and Lichtner, 1987). Fluid flow would tend to die away slowly in such a system over a period of a few months (Sibson, 1981), possibly allowing time for coarse crystal growth.

As mineral deposition occurs and the fracture becomes sealed, pressure can rise again, leading to a repetition of the process. Such a cyclic process may explain the ribboning in the Bralorne veins. Each ribbon of quartz would have associated with it a black layer of intensely altered host rock consisting of pyrite, arsenopyrite, sericite and occasional ankerite, with gold, that formed the vein wall before rupture took place and the process was repeated again. The PATH model predicts that the bulk of the gold would be precipitated in the immediate envelope to the vein, so that incorporating even a sliver of wall rock into the vein would include most of the gold.

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# NOTES