# **BULLETIN DE L'ASSOCIATION MINERALOGIQUE DU CANADA**



# JOURNAL OF THE MINERALOGICAL ASSOCIATION OF CANADA

Volume 33

August 1995

Part 4

The Canadian Mineralogist Vol. 33, pp. 709-721 (1995)

## CONSTRAINTS FROM FLUID-INCLUSION DATA ON THE ORIGIN OF THE JUBILEE CARBONATE-HOSTED Zn-Pb DEPOSIT, CAPE BRETON, NOVA SCOTIA\*

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## Abstract

Aqueous (Aq) and hydrocarbon (HC) fluid inclusions were studied in fibrous calcite (pre-ore), sphalerite, and coarse, anhedral calcite (syn- to post-ore) of the Jubilee Zn–Pb deposit, Cape Breton, Nova Scotia. The deposit is hosted by a limestone breccia at the top of the Macumber Formation (limestones) and under the Carrolls Corner Formation (evaporites). Aq inclusions from different minerals have similar temperatures of first melting (-62.1 to -46.7°C), indicating a  $H_2O$ -NaCl–CaCl<sub>2</sub> compositional system. Salinities (wt%), NaCl/(NaCl + CaCl<sub>2</sub>) weight ratios, and temperatures of homogenization (T<sub>h</sub>) of Aq inclusions range from 21 to 27%, 0.16 to 0.73, and 53 to 79°C for fibrous calcite, 15 to 24%, 0.26 to 0.64, and 59 to 228°C for sphalerite, and 12 to 24%, 0.17 to 0.72, and 66 to 224°C for anhedral calcite. The T<sub>h</sub> – salinity correlation suggests that a high-temperature and salinity. HC inclusions coexist with Aq inclusions. The T<sub>h</sub> of HC inclusions ranges from 34 to 89°C for fibrous calcite. Fluid pressures estimated from the intersection of isochores of coexisting Aq and HC inclusions range from 175 to 363 bars for the fibrous calcite, and from 160 to 398 bars for the anhedral calcite. The microthermometric data suggest that the Jubilee deposit formed from a hot, metal-carrying brine in a shallow environment (probably less than 1500 m) characterized by a low background temperature. Hydrocarbons migrated into the limestone breccia before the mineralization when the metal-carrying brine in greated through.

Keywords: Zn-Pb deposit, carbonates, aqueous fluid inclusions, hydrocarbon fluid inclusions, basinal brines, Jubilee, Cape Breton, Nova Scotia.

### SOMMAIRE

Des inclusions fluides aqueuses (Aq) et d'hydrocarbures (HC) ont été étudiées dans la calcite fibreuse (pré-minéralisation), la sphalérite, et la calcite grossière xénomorphe (syn- à post-minéralisation) du gîte à Zn-Pb de Jubilee, au Cap Breton, Nouvelle-Ecosse. Le gîte est encaissé par une brèche calcaire au sommet de la Formation de Macumber (calcaires), et en dessous

<sup>\*</sup> Geological Survey of Canada contribution number 31794.

de la Formation de Carrolls Corner (évaporites). Les inclusions Aq des différents minéraux ont des températures de première fonte semblables (-62,1 à -46,7°C), indiquant une composition dans le système H<sub>2</sub>O-NaCl-CaCl<sub>2</sub>. Les salinités (%, au poids), les rapports pondéraux de NaCl/(NaCl + CaCl<sub>2</sub>), et les températures d'homogénéisation (T<sub>b</sub>) des inclusions Aq varient de 21 à 27%, 0,16 à 0,73, et 53 à 79°C pour la calcite fibreuse, de 15 à 24%, 0,26 à 0,64, et 59 à 228°C pour la sphalérite, et de 12 à 24%, 0,17 à 0,72, et 66 à 224°C pour la calcite xénomorphe. La corrélation entre T<sub>h</sub> et la salinité suggère qu'un fluide à température et salinité élevées se serait mélangé d'abord avec un fluide de faible température et es alinité élevée, et ensuite avec un autre fluide de basses température et salinité. Des inclusions d'HC coexistent avec les inclusions Aq. La T<sub>h</sub> des inclusions d'HC varie de 34 à 89°C pour la calcite fibreuse, 46 à 87°C pour la sphalérite, et 31 à 238°C pour la calcite xénomorphe. La pression des fluides, dérivée par l'intersection des isochores des inclusions Aq et d'HC coexistantes, varie de 175 à 363 bars pour la calcite fibreuse, et de 160 à 398 bars pour la calcite xénomorphe. Les données microthermométriques suggèrent que le dépôt de Jubilee s'est formé à partir d'une saumure chaude métallifère dans un environnement de faible profondeur (probablement moins de 1500 m) ayant une faible température. Les hydrocarbures ont migré dans la brèche calcaire avant la minéralisation, ont réagi avec le soufire oxydé dérivé des évaporites, et ont produit un réservoir riche en soufire réduit ainsi disponible pour la minéralisation lors du passage de la saumure métallifère.

Mots-clés: dépôt de Zn-Pb, carbonates, inclusions fluides aqueuses, inclusions fluides d'hydrocarbures, saumures de bassins, Jubilee, Cap Breton, Nouvelle-Écosse.

#### INTRODUCTION

The Jubilee Zn–Pb deposit on Cape Breton Island, Nova Scotia, is one of several base-metal deposits hosted by Early Carboniferous carbonates of the Late Devonian – Permian Maritimes Basin (Fig. 1). These deposits have been the subject of a number of recent studies aiming to understand their origin and relation to the evolution of the basin (*e.g.*, Ravenhurst *et al.* 1989, Burtt *et al.* 1994). According to previous studies, the Jubilee deposit shows similarities and differences with Mississippi-Valley-type (MVT) deposits (see Anderson & Macqueen 1988). Like MVT deposits, it is located near the edge of a basin, hosted by carbonates, controlled by a system of pre-existing porosity, and has a relatively simple mineralogy. Hydrocarbon-bearing fluid inclusions are present in ore and gangue minerals (Hein *et al.* 1993). Unlike most MVT deposits, it is hosted by limestone instead of dolostone (Paradis *et al.* 1993),



FIG. 1. Regional geological map of the southern margin of the Maritimes Basin (after Boehner *et al.* 1989) showing the location of the Jubilee and several other deposits: 1 Jubilee Zn-Pb deposit, 2 Pembroke Pb deposit, 3 Smithfield Pb-Zn deposit, 4 Gays River Zn-Pb deposit, 5 Walton Ba-Pb-Zn deposit. The inset map shows the regional setting of the Maritimes Basin (after Howie & Barss 1975).

and mineralization seems to have been strongly controlled by faults (Hein *et al.* 1993). Temperatures of fluid-inclusion homogenization range from  $82-98^{\circ}$ C in post-ore barite (Hein *et al.* 1993) to  $353-392^{\circ}$ C in sphalerite (Stewart 1978), which is markedly higher than in most MVT deposits.

Stable isotope studies (Armstrong et al. 1993, Fallara et al. 1994) have suggested that the Jubilee deposit formed from circulating basinal brines, and that hydrocarbons likely played an important role in the precipitation of sulfides. However, little is known about the composition of the brines, their relationship with each other, and with the hydrocarbons. This lack of knowledge about the fluids directly involved in mineralization seriously hinders our understanding of the mechanisms that produced the deposit. Fluid inclusions in hydrothermal minerals represent samples of the fluids that once existed in the mineralizing system, and can provide direct information about the composition of the fluids and the physicochemical conditions of the mineralizing processes. Fluid-inclusion studies of reconnaissance nature have previously been carried out (Stewart 1978, Hein et al. 1993). However, these studies measured homogenization temperatures of fluid inclusions, but did not report quantitative data on freezing temperatures. Consequently, the compositional systems and salinities of the fluids are unknown. Besides, the significance of the reported large range of homogenization temperatures remains an open question.

This paper presents a systematic microthermometric study of aqueous and hydrocarbon-bearing fluid inclusions from pre-, syn-, to post-ore minerals of the Jubilee deposit. The main objective of this study is to determine the compositional systems, salinities, and temperatures of the aqueous and hydrocarbon-bearing fluids present before, during, and after mineralization, and to evaluate the thermal regime, depth, and mechanisms of mineralization.

#### GEOLOGY OF THE DEPOSIT

The Jubilee deposit is located near an NNWtrending elongate dome (Fig. 2). The stratigraphic sequence, from bottom to top, consists of conglomerate and sandstone of the Horton Group, and limestone (Macumber Formation) and evaporites (Carrolls Corner Formation) of the Lower Windsor Group. A limestone breccia (<15 m) occurs at the top of the Macumber (Hein *et al.* 1988, Graves *et al.* 1990, Fallara *et al.* 1994) and hosts the deposit.

A number of NW-striking faults occur in the area. The Jubilee fault, which extends northwest from the dome, and the Road fault along the northeastern margin



FIG. 2. Geological map of the Jubilee Zn–Pb deposit and schematic distribution patterns of Zn + Pb grades multiplied by thickness in meters (%m; after Fallara 1995). Drill holes from which the samples of this study were collected are shown by filled circles.

of the dome (Fig. 2), are the most prominent. They are normal faults with very high angles of dip.

Mineralization is spatially associated with the Jubilee fault (the main ore-zone) and the Road fault (Fig. 2). Sulfides (pyrite, marcasite, sphalerite, galena and minor chalcopyrite) occur as cements in the breccia and as veinlets or disseminations in the matrix and in recrystallized fragments. Massive, high-grade ore locally replaces limestone (Fig. 2). The main gangue mineral is calcite; no dolomite has been found (Paradis *et al.* 1993). Minor gangue minerals include barite, anhydrite and gypsum. The total reserves are estimated at 0.9 million metric tonnes grading 5.2% Zn and 1.4% Pb (Hein *et al.* 1993).

The paragenetic sequence at the Jubilee deposit was studied by Paradis et al (1993) and Fallara *et al.* (1994), and is summarized as follows: (1) fibrous calcite, (2) pyrite and marcasite, (3) sphalerite, (4) galena, sphalerite and coarse, anhedral calcite, (5) coarse anhedral calcite, and (6) sulfates (barite, anhydrite and gypsum).

## SAMPLES AND PETROGRAPHY OF FLUID INCLUSIONS

Nine samples from five drill cores in the main ore zone, one sample from a drill core near the Road fault (Fig. 2), and a sample from the main showing on the surface, were prepared for fluid-inclusion study. All samples from drill cores in the main ore-zone contain fibrous calcite, pyrite – marcasite, sphalerite, galena, and anhedral calcite. The sample from the Road fault contains fibrous and anhedral types of calcite, but does not contain sphalerite. The sample from the surface showing contains massive, fine-grained sphalerite that replaces limestone; the sphalerite in this sample does not contain fluid inclusions suitable for microthermometric measurements.

Fibrous calcite occurs as cement partly filling the secondary pores of the limestone breccia. It mainly predates pyrite - marcasite, and invariably predates sphalerite. Two petrographic types of sphalerite were distinguished in the samples, an anhedral type, very abundant, with brown to light yellow zonations that typically contain numerous tiny fluid inclusions, and a euhedral type, less abundant, yellow, and free of inclusions. The zoned sphalerite occurs as layers encrusting fibrous calcite and pyrite - marcasite, or as veinlets cutting fibrous calcite, pyrite - marcasite, and breccia fragments. The euhedral sphalerite postdates the zoned sphalerite, and occurs with galena and anhedral calcite, which fills the residual pores between breccia fragments. The anhedral calcite is partly contemporaneous with, but mainly postdates, zoned and euhedral types of sphalerite.

Fluid inclusions were studied in fibrous calcite (pre-ore), zoned sphalerite, and anhedral calcite (synto post-ore). Both aqueous and hydrocarbon-bearing fluid inclusions were observed in these minerals. Most aqueous fluid inclusions comprise a liquid and a vapor phase at room temperature (Fig. 3A). A few aqueous fluid inclusions comprise only a liquid phase. No daughter minerals were observed in any of the aqueous fluid inclusions. Hydrocarbon fluid inclusions comprise a liquid and a vapor phase as well at room temperature (Fig. 3B), and are distinguished from aqueous fluid inclusions by their color, freezing behavior, and fluorescence under blue light excitation. The liquid phase of hydrocarbon fluid inclusions is either brown to yellow-brown (Fig. 3B) or colorless (Figs. 3C, D); the former is more abundant than the latter. Some inclusions comprise both a brown hydrocarbon liquid and a colorless hydrocarbon liquid, plus a vapor phase (Fig. 3E). A few inclusions comprise an aqueous liquid, a hydrocarbon liquid, and a vapor phase (Fig. 3F).

In fibrous calcite, aqueous fluid inclusions are abundant and randomly distributed in three dimensions. These inclusions are likely primary according to the criteria of Roedder (1984). However, since these inclusions are densely distributed, it is possible that some of them are controlled by microfractures (secondary), but cannot be distinguished from the others in the thin sections. Some aqueous fluid inclusions are distributed along healed fractures crosscutting crystal boundaries. These secondary inclusions can readily be distinguished from the randomly distributed inclusions. Hydrocarbon fluid inclusions are scattered among the randomly distributed population of aqueous fluid inclusions, and are considered primary because no fracture control has been observed. All the hydrocarbon fluid inclusions in fibrous calcite are brown to yellow-brown. No colorless hydrocarbon-bearing fluid inclusions have been observed.

In zoned sphalerite, numerous aqueous fluid inclusions occur along growth zones defined by color zoning (Fig. 3G). These inclusions are primary, but are not amenable to microthermometric analysis because they are less than 1 µm in diameter. A number of aqueous fluid inclusions occur in clusters lacking fracture control, and some of them are relatively isolated. These inclusions are considered primary and distinguished from secondary inclusions in healed fractures cross-cutting crystal boundaries. Hydrocarbon fluid inclusions are much less abundant than aqueous fluid inclusions. They were observed within clusters of aqueous fluid inclusions, and are also likely primary. Both brown to yellow-brown and colorless hydrocarbon-bearing fluid inclusions were observed.

In anhedral calcite, aqueous fluid inclusions occur in clusters lacking fracture control or in short healed fractures terminating within individual crystals. Some inclusions are relatively isolated (Fig. 3A). These inclusions are likely primary or pseudosecondary.



FIG. 3. A: An isolated aqueous fluid inclusion in anhedral calcite; B: an isolated hydrocarbon fluid inclusions (yellow-brown, HC-1) in anhedral calcite; C: hydrocarbon fluid inclusions (colorless, HC-2) in sphalerite at room temperature; D: same inclusions as B at -36°C (note that the vapor bubble expanded and became irregular in form); E: an inclusion comprising a colorless hydrocarbon liquid (HC-2), a brown hydrocarbon liquid (HC-1), and a vapor phase (Vp), in anhedral calcite; F: an inclusion comprising an aqueous liquid (Aq), a brown hydrocarbon liquid (HC-1), and a vapor phase (Vp), in anhedral calcite; G: primary fluid inclusions distributed along growth zones of a sphalerite crystal.

Secondary inclusions in healed fractures crosscutting crystal boundaries are abundant. Hydrocarbon fluid inclusions are either scattered in clusters of aqueous fluid inclusions, or occur in healed fractures cross-cutting crystal boundaries. The former is likely primary, and the latter is secondary. Both brown to yellow-brown and colorless hydrocarbon-bearing fluid inclusions were observed in the clusters, whereas only brown to yellow-brown hydrocarbonbearing fluid inclusions were found in the healed fractures.

The petrographic study described above suggests that aqueous fluid and liquid hydrocarbon were coexistent during the precipitation of fibrous calcite, sphalerite, and anhedral calcite. They may have occurred as immiscible liquids and may have been trapped as separate aqueous and hydrocarbon fluid inclusions. Occasionally, both aqueous and hydrocarbon fluids were trapped in the same inclusion (*i.e.*, heterogeneous trapping, Fig. 3F).

Fluid inclusions that are likely primary or pseudosecondary, with the longest dimension ranging from 5 to 54  $\mu$ m (mainly >10  $\mu$ m), were selected for microthermometric analysis. Some secondary inclusions also were studied for comparison.

#### ANALYTICAL METHODS

Homogenization temperatures of aqueous and hydrocarbon fluid inclusions, and melting temperatures of aqueous fluid inclusions, were measured using a U.S.G.S. heating-freezing stage that was calibrated to a precision and accuracy of  $\pm 0.1^{\circ}$ C with synthetic standard fluid inclusions provided by Fluid Inc. Homogenization temperatures were measured for each selected fluid inclusion in a chip of sample prior to cooling runs, in order to avoid the effect of stretching caused by fluid freezing.

The fluorescence spectrum of individual hydrocarbon fluid inclusions was measured using a Zeiss II photomicroscope, equipped with a HBO W/2 100 highpressure mercury lamp, a BG–12 excitation filter, a Zeiss FL prism, an oil immersion NEOFLUAR 100× objective, and an optovar set at 1.25×. The objective diaphragm apertures were 3, 6 and 10  $\mu$ m, and the measuring diaphragm aperture was 3  $\mu$ m. The excitation wavelength is 405 nm (blue light). The fluorescence spectra of the hydrocarbon fluid inclusions were calibrated against a uranyl glass GG17 standard at each 10 nm interval in wavelength ranging from 400 to 700 nm.

#### RESULTS

Microthermometric results of aqueous and hydrocarbon fluid inclusions from fibrous calcite, zoned sphalerite, and anhedral calcite are summarized in Table 1, and are described as follows. All the descriptions, unless otherwise specified, apply to the fluid inclusions that are likely primary or pseudosecondary on the basis of their petrography. The aqueous fluid inclusions in fibrous calcite classified as type P\* in Table 1 are considered primary based on  $T_h$  – salinity relationship, which will be discussed in detail in the next section.

## Aqueous fluid inclusions

Aqueous fluid inclusions from different minerals show similar temperatures of first melting, ranging from -62.1 to -46.7°C (Table 1). These first melting temperatures are compatible with those found in the system H<sub>2</sub>O-NaCl-CaCl<sub>2</sub>. The first melting temperatures of such system are usually within  $3^{\circ}$ C of the eutectic temperature (-52°C) if stable salt hydrates form during cooling, but they may be as low as -80°C if there are metastable salt hydrates (Davis *et al.* 1990). Minor amounts of other cations such as Mg<sup>2+</sup> and K<sup>+</sup> are possibly present, but cannot be examined in the present study.

Melting temperatures of hydrohalite (NaCl·2H<sub>2</sub>O), which was identified by its tabular shape and higher relief than ice, range from -37.6 to -24.9, -33.9 to -26.0, and -37.2 to  $-24.6^{\circ}$ C for fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite, respectively (Table 1). These temperatures are considered to be approximate because hydrohalite crystals cannot be seen clearly in the case of many inclusions. NaCl/(NaCl + CaCl<sub>2</sub>) weight ratios, estimated from melting temperatures of hydrohalite by using the H<sub>2</sub>O-NaCl-CaCl<sub>2</sub> diagram (Oakes *et al.* 1990) (Fig. 4), range from 0.16 to 0.73, 0.26 to 0.64, and 0.17 to 0.72 for fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite, respectively (Table 1).

		Tm-1st(- <sup>0</sup> C)		Tm-HH(- º C)		Tm-H2O(- ° C)	
Host	Туре	Range and Number	Mean and Std. Dev.	Range and Number	Mean and Std. Dev.	Range and Number	Mean and Std. Dev.
Fibrous Calcite	AQ: P AQ: P* AQ: S	48.3-62.1(25) 48.3-62.1(13) 49.4-54.1(4)	54.5(+/-3.3) 55.3(+/-3.8) 51.6(+/-1.9)	24.9-37.6(19) 26.2-37.6(9) 27.8-32.3(4)	31.2(+/-3.3) 30.9(+/-3.6) 29.8(+/-1.9)	9.9-34.3(28) 19.5-34.3(15) 8.7-9.7(4)	19.2(+/-7.6) 25.1(+/-4.7) 9.3(+/-0.5)
Sphalerite	AQ: P AQ: S	50.8-59.4(17) 56.4-57.2(2)	53.9(+/-2.6) 56.8(+/-0.6)	26.0-33 <b>.9</b> (9) 29.9-33.0(2)	29.5(+/-2.4) 31.5(+/-2.2)	10.6-25.3(17) 11.6-16.2(2)	14.7(+/-4.7) 13.9(+/-3.3)
Anhedral Calcite	AQ: P AQ: S	46.7-60.3(38) 50.3-55.8(8)	53.5(+/3.0) 53.1(+/-1.8)	24.6-37.2(26) 27.1-32.0(6)	29.9(+/-2.8) 29.5(+/-1.8)	7.6-27.0(31) 9.3-10.4(7)	12.0(+/-5.5) 9.9(+/-0.4)
		Th( 0 C)		Salinity (wt%)		NaCl/(NaCl+ CaCl2)	
Host	Туре	Range and Number	Mean and Std. Dev.	Range and Number	Mean and Std. Dev.	Range and Number	Mean and Std. Dev.
Fibrous Calcite	AQ: P AQ: P* AQ: S HC(br): P	41.1-204.8(46) 52.8-79.2(15) 80.7-89.8(4) 34.3-88.7(29)	90.5(+/-38.6) 66.1(+/-8.8) 86.0(+/-4.0) 51.2(+/-11.9)	14.0-26.8(28) 20.8-26.8(15) 12.7-13.8(4)	20.0(+/-4.4) 23.6(+/-1.9) 13.4(+/-0.5)	0.16-0.73(19) 0.16-0.73(9) 0.32-0.53(4)	0.38(+/-0.14) 0.40(+/-0.15) 0.44(+/-0.09)
Sphalerite	AQ: P AQ: S HC(br): P HC(cl): P	58.5-228.1(32) 76.4-90.2(4) 45.7(1) 55.8-87.1(2)	92.5(+/-38.8) 83.2(+/-5.8) 71.5(+/-22.1)	14.6-23.6(17) 15.5-18.9(2)	17.5(+/-3.0) 17.2(+/-2.5)	0.26-0.64(9) 0.29-0.42(2)	0.45(+/-0.12) 0.36(+/-0.09)
Anhedral Calcite	AQ: P AQ: S HC(br): P HC(cl): P HC(br): S	66.2-224.4(51) 70.3-87.3(9) 31.0-237.6(66) 106.1-121.3(2) 33.3-98.3(22)	101.1(+/-37.6) 77.4(+/-5.9) 88.1(+/-48.0) 113.7(+/-10.7) 54.1(+/-15.1)	11.6-24.3(31) 13.3-14.4(7)	15.2(+/-3.5) 13.9(+/-0.4)	0.17-0.72(26) 0.33-0.57(6)	0.44(+/-0.13) 0.45(+/-0.09)

TABLE 1. MICROTHERMOMETRIC DATA ON AQUEOUS AND HYDROCARBON FLUID INCLUSIONS

AQ: aqueous finid inclusion; HC(br); brown to yellow-brown hydrocarbon fluid inclusion; HC(cl): colorless hydrocarbon fluid inclusion; P<sup>\*</sup>: primary (including pseudosecondary) based on petrography; P<sup>\*</sup>: primary based on homogenization temperature – salinity relationship; S: secondary; Tm-1st: first-melting temperature; Tm-HH: melting temperature of hydrohalite; Tm-H<sub>2</sub>O: molting temperature of ice; Th: homogenization temperature.





Melting temperatures of ice range from -34.3 to -9.9, -25.3 to -10.6, and -27.0 to -7.6°C for fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite, respectively (Table 1). Melting temperatures of ice that indicate metastability, *i.e.*, as revealed by a vapor bubble appearing at the moment ice melts, are useless and are not reported in Table 1. Salinities were calculated using melting temperatures of ice and  $NaCl/(NaCl + CaCl_2)$  ratios with the equation 2 of Oakes et al. (1990). For fluid inclusions for which the melting temperature of hydrohalite was not measured, the average NaCl/(NaCl + CaCl<sub>2</sub>) ratio of fluid inclusions in the same mineral (0.38 for fibrous calcite, 0.45 for sphalerite, and 0.44 for anhedral calcite) was used to calculate salinities. The salinities obtained range from 14.0 to 26.8, 14.6 to 23.6, and 11.6 to 24.3 wt% for fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite, respectively (Table 1).

Temperatures of homogenization (to the liquid phase) range from 41.1 to 204.8, 58.5 to 228.1, and 66.2 to 224.4°C for fluid inclusions in fibrous calcite, sphalerite, and anhedral calcite, respectively (Fig. 5).

FIG. 5. Histograms of homogenization temperatures of aqueous fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite. The hatched area for fibrous calcite represent fluid inclusions that are most likely primary, on the basis of homogenization temperature – salinity relationship (see Fig. 8).



Secondary fluid inclusions in different host minerals show similar and relatively convergent microthermometric characteristics. Salinities, NaCl/(NaCl + CaCl<sub>2</sub>) ratios, and homogenization temperatures range from 12.7 to 13.8 wt%, 0.32 to 0.53, and 80.7 to 89.8°C for fibrous calcite, from 15.5 to 18.9 wt%, 0.29 to 0.42, and 76.4 to 90.2°C for sphalerite, and from 13.3 to 14.4 wt%, 0.33 to 0.57, and 70.3 to 87.3°C for anhedral calcite.

### Hydrocarbon fluid inclusions

Brown to yellow-brown and colorless hydrocarbon fluid inclusions display different behaviors upon cooling. The vapor bubble of brown to yellow-brown hydrocarbon fluid inclusions expands very slightly during cooling to liquid nitrogen temperature, and the liquid phase does not show an obvious phase-change. On the other hand, the vapor bubble within colorless hydrocarbon fluid inclusions expands greatly and becomes irregular upon cooling to about  $-5^{\circ}$ C (Figs. 3C, D), indicating that the liquid phase becomes frozen and shrinks.

Both brown to yellow-brown and colorless hydrocarbon fluid inclusions are strongly fluorescent under blue light excitation. Fluorescence spectra (Fig. 6) indicate that the colorless hydrocarbon fluid inclusions have a lower wavelength of spectral maximum ( $L_{max}$ ) (530 nm) than brown to yellow-brown inclusions (550–570 nm). Brown to yellow-brown hydrocarbon fluid inclusions with relatively large vapor bubbles have a higher  $L_{max}$  (570 nm) than those with relatively small vapor bubbles (550 nm).



FIG. 6. Fluorescence spectra of hydrocarbon fluid inclusions under blue light excitation. I. Randomly distributed hydrocarbon fluid inclusions from fibrous calcite (three inclusions); II. isolated and randomly distributed hydrocarbon fluid inclusions from anhedral calcite: IIa. brown to yellow-brown hydrocarbon fluid inclusions with relatively large bubble (five inclusions); IIb. brown to yellowbrown hydrocarbon fluid inclusions with relatively small bubble (six inclusions); IIc. colorless hydrocarbon fluid inclusions (one inclusion); III. secondary hydrocarbon fluid inclusions (brown to yellow-brown) from anhedral calcite (three inclusions).



FIG. 7. Histograms of homogenization temperatures of brown to yellow-brown hydrocarbon fluid inclusions from fibrous calcite and anhedral calcite.

The fluorescence spectrum is a function of both the gross composition of the trapped hydrocarbons and their maturation state; aromatic and polyaromatic compounds are mainly responsible for the fluorescence properties (Guilhaumou *et al.* 1990). Higher values of  $L_{max}$  indicate higher thermal maturation and aromaticity (Khorasani 1987). However, no quantitative correlation between a fluorescence spectrum and a specific composition and maturity of the hydrocarbons has been established.

Temperatures of homogenization (to the liquid phase) of hydrocarbon fluid inclusions are summarized in Table 1; most of them were obtained from brown to yellow-brown hydrocarbon fluid inclusions in fibrous and anhedral calcite (Fig. 7). Only a few hydrocarbon fluid inclusions in sphalerite were measured for homogenization temperatures (Table 1). Homogenization temperatures of brown to yellow brown hydrocarbon fluid inclusions range from 34.3 to 88.7°C for primary inclusions in fibrous calcite, 31.0 to 237.6°C for primary inclusions in anhedral calcite, and 33.4 to 98.3°C for secondary inclusions in anhedral calcite.

#### DISCUSSION

The microthermometric data documented above can be used to infer the compositions, temperatures and pressures of the fluids existing in the hydrothermal system before, during, and after the formation of the deposit. The relationship between the distinct fluids and their role during mineralization, the thermal evolution of the hydrothermal system, the pressures of the fluids and corresponding depths, and the mechanisms of ore precipitation are discussed below.

### Multiple aqueous fluids and mixing

A wide range of salinities and homogenization temperatures of aqueous fluid inclusions is obvious (Table 1, Fig. 5). On a plot of homogenization temperature versus salinity (Fig. 8), it is shown that primary inclusions in fibrous calcite mainly plot in an area (A in Fig. 8) with homogenization temperatures ranging from 53 to 79°C and salinities ranging from 21 to 27 wt% (Table 1, type P\*). These inclusions are likely representative of the aqueous fluid present during the precipitation of fibrous calcite. However, some inclusions in fibrous calcite plot in areas that are dominated by fluid inclusions from sphalerite and anhedral calcite. These inclusions, although assigned to primary inclusions from petrographic judgement, are likely secondary inclusions that were superimposed on fibrous calcite during the precipitation of sphalerite and anhedral calcite.

Primary inclusions in sphalerite and anhedral calcite plot in a wide area varying from low salinity – low temperature (C in Fig. 8) to high salinity – high temperature (B in Fig. 8). These wide ranges can be explained by mixing of fluids. Two stages of mixing are envisaged. Firstly, a high-salinity high-temperature fluid (B in Fig. 8), which likely was the metal-carrying fluid, mixed with a high-salinity low-temperature fluid (A in Fig. 8), which saturated the pores before mineralization (*i.e.*, the fluid present during precipitation of fibrous calcite). This stage of mixing resulted in high salinities and intermediate temperatures, which define the subhorizontal mixing-line in Figure 8 (mixing 1). Secondly, perhaps during the waning stage of mineralization, the intermediate fluids produced by mixing 1 mixed with a third, low-temperature – lowsalinity fluid (C in Fig. 8). This stage of mixing constitutes the oblique line in Figure 8 (mixing 2). The proportion of the third fluid may have increased with time, and eventually dominated the hydrothermal system in the post-ore stage, as indicated by secondary inclusions in anhedral calcite, which plot in the lowsalinity, low-temperature area in Figure 8.

Low temperatures of the early fluid present during the precipitation of fibrous calcite (fluid A) and of the late fluid present after precipitation of anhedral calcite (fluid C) probably reflect conditions of burial of the deposit. The metal-carrying fluid (fluid B), however, was likely derived from a deep source characterized by high temperature and high salinity. The high salinity of fluid A may have been caused by addition of salts dissolved from evaporites during a reaction between oxidized sulfur in the evaporites and hydrocarbons, as will be discussed later.

#### Thermal regime of the hydrothermal system

The above discussion indicates that the hydrothermal system of the Jubilee deposit evolved from a low-temperature state before mineralization, through a high-temperature state during mineralization, and back to a low-temperature state toward the waning stage of and after mineralization. However, from Figures 5 and 8, it can be seen that the number of aqueous fluid inclusions in sphalerite and anhedral calcite with a high temperature of homogenization is limited. A question that arises is whether or not these high-temperature



FIG. 8. Homogenization temperature – salinity diagram of aqueous fluid inclusions from fibrous calcite, sphalerite, and anhedral calcite. Dashed lines show mixing trends (see the text for explanation).

inclusions truly record a high-temperature event. We argue in favor of the validity of these high-temperature data on the basis of several lines of evidence. Firstly, fluid inclusions with necking-down features have been avoided during selection of fluid inclusions for microthermometric measurements. Artificial stretching due to freezing of the fluid has been overcome by performing heating before freezing runs. Stretching due to natural heating does not seem to have happened because there is no evidence that the area has been heated to higher temperatures after the formation of the deposit. Secondly, high temperatures of homogenization of fluid inclusions similar to those measured in this study also were recorded in other base-metal deposits hosted by carbonates of Lower Windsor Group in Nova Scotia, e.g., 123–220°C for the Sugar Camp showing. Cape Breton Island (Chi & Savard 1995), 80-250°C for the Gays River deposit, 120-320°C for the Walton deposit (Burtt et al. 1994), 130-180°C for the Smithfield deposit, and 160-250°C for the Pembroke deposit (Ravenhurst et al. 1989) (see Fig. 1 for location of the deposits). Thirdly, higher temperatures of homogenization (353-392°C, 9 inclusions) in sphalerite from the main showing of the Jubilee deposit have been reported (Stewart 1978). Although we did not find fluid inclusions large enough for microthermometric study in the fine-grained replacive sphalerite from the main showing, and therefore were unable to confirm the accuracy of these high temperatures of homogenization, fluids that formed massive mineralization may have had an even higher temperature than those we measured.

The limited number of fluid inclusions with high temperatures of homogenization may be related to the fact that the high-temperature metal-bearing fluid was cooled rapidly because the ambient temperature of the deposit was considerably lower. Mixing of the hightemperature fluid with the low-temperature fluid present in pre-mineralization pores inevitably accelerated the cooling process. Many high-temperature fluid inclusions were likely trapped in sphalerite, but it seems that they belong to the group of inclusions that are too small for microthermometric measurements. The number of fluid inclusions studied neither reflects which temperature range was dominant during the main mineralization, nor the relative abundance of the respective fluid end-members.

The high temperatures of homogenization of the Jubilee deposit, and those of other base-metal deposits hosted by carbonates of Lower Windsor Group in Nova Scotia, are higher than those found in most MVT deposits (100–150°C, Roedder 1984, p. 416), but are similar to the Irish-type of carbonate-hosted base-metal deposits (100–240°C, Hitzman & Large 1986). It is likely that, as in the case of the Irish-type deposits, the Jubilee and other base-metal deposits of the Maritimes Basin formed in a more active tectonic and geothermal regime than typical MVT deposits. Clearly, the Jubilee

deposit was in a low-temperature burial setting before and after mineralization. Our on-going study of organic matter in the host rocks of the Jubilee deposit indicates that the host rocks did not attain thermal equilibrium with the high-temperature mineralizing fluid, probably owing to rapid cooling. Therefore, the Jubilee deposit represents a case in which the deposit formed from hot fluids in a cool background (Sangster *et al.* 1994).

## Fluid pressure and depth estimation

The depth of mineralization of the Jubilee deposit has been difficult to determine because the age of mineralization is unknown and therefore the thickness of cover at the time of mineralization cannot be estimated from stratigraphic reconstruction. Several authors (e.g., Narr & Burruss 1984, McLimans 1987, Walgenwitz et al. 1990) have used the intersection of isochores of coexisting aqueous and hydrocarbon fluid inclusions to estimate fluid pressures and corresponding depths. Petrographic observations indicate that aqueous fluids and liquid hydrocarbons coexisted as immiscible phases during the precipitation of fibrous calcite, sphalerite, and anhedral calcite, as well as after the precipitation of anhedral calcite in the Jubilee deposit. The homogenization temperatures of hydrocarbon fluid inclusions are generally lower than those of coexisting aqueous fluid inclusions (Table 1). This indicates that both aqueous and hydrocarbon fluids were liquid at the time they were trapped, because hydrocarbons have a higher bubble-point curve and a lower isochore slope in temperature-pressure coordinates than aqueous fluids, and the intersection of the two isochores lies in the field of liquid (e.g., Narr & Burruss 1984, Walgenwitz et al. 1990). If both isochores of coexisting aqueous and hydrocarbon fluid inclusions can be determined, their intersection can be used to estimate the pressure of trapping.

Isochores of aqueous fluid inclusions can be calculated by using the FLINCOR program (Brown 1989), with the fluid system being approximated by H<sub>2</sub>O-NaCl. The errors introduced by this approximation are negligible, according to the experimental data of Zhang & Frantz (1987). The isochores of hydrocarbon fluid inclusions, however, are difficult to determine, because the composition of the hydrocarbons is unknown. In practice, isochores of hydrocarbon fluid inclusions can be approximated by crude oils (e.g., Narr & Burruss 1984). In order to obtain a broad estimate of fluid pressure (and depth) of the Jubilee deposit, the minimum and maximum bubble-point pressures of crude oils reported by McLimans (1987) and Walgenwitz et al. (1990) were used to construct the isochores. These were located by extrapolation from the data of Walgenwitz et al. (1990), who have used the temperatures and pressures of decrepitation of hydrocarbon fluid inclusions to constrain the slopes of the isochores.



FIG. 9. Intersections of isochores for primary fluid inclusions in fibrous calcite (A), primary inclusions in anhedral calcite (B), and secondary inclusions in anhedral calcite (C). Aq1 and HC1 represent isochores for Mean - Standard Deviation, and Aq2 and HC2 represent isochores for Mean + Standard Deviation, for aqueous and hydrocarbon fluid inclusions, respectively. The lower and upper limits of the bubble-point curve of hydrocarbon fluid inclusions were taken from McLimans (1987) and Walgenwitz et al. (1990), respectively. The isochores of aqueous fluid inclusions were calculated by using the FLINCOR program (Brown 1989), with the fluid system being approximated by H<sub>2</sub>O-NaCl. The isochores of hydrocarbon fluid inclusions were extrapolated from the data of Walgenwitz et al. (1990). Shaded areas indicate possible range of fluid pressures.

Isochores were calculated for primary aqueous and brown to yellow-brown hydrocarbon fluid inclusions in fibrous and anhedral calcite and for secondary aqueous and hydrocarbon fluid inclusions in anhedral calcite (Fig. 9). Isochores were not calculated for fluid inclusions in sphalerite because only a few hydrocarbon fluid inclusions were measured for homogenization temperatures. In order to evaluate the possible range of fluid pressures, mean values ± standard deviations of homogenization temperatures and salinities of aqueous and hydrocarbon fluid inclusions (Table 1) were used: 66.1  $\pm$  8.8°C (T<sub>h</sub> of Aq), 51.2  $\pm$  11.9°C (T<sub>h</sub> of HC), and 23.6  $\pm$  1.9 wt% (salinity of Aq) for primary inclusions in fibrous calcite;  $101.1 \pm 36.7$ °C (T<sub>h</sub> of Aq), 88.1  $\pm$  48.0°C (T<sub>h</sub> of HC), and 15.2  $\pm$  3.5 wt% (salinity of Aq) for primary inclusions in anhedral calcite, 77.4  $\pm$  5.9°C (T<sub>h</sub> of Aq), 54.1  $\pm$  15.1°C (T<sub>h</sub> of HC), and  $13.9 \pm 0.4$  wt% (salinity of Aq) for secondary inclusions in anhedral calcite. The shaded areas in Figure 9 indicate possible ranges of fluid pressures. The fluid pressures thus calculated range from 175 to 363 bars during precipitation of fibrous calcite, from 160 to 398 bars during the precipitation of anhedral calcite, and from 190 to 441 bars after this episode.

Because the Jubilee deposit was overlain by a thick sequence of evaporite of the Carrolls Corner Formation that constitutes an impermeable cover, the fluid pressure in the system was possibly close to lithostatic. Assuming a lithostatic system with a rock density of  $2.7 \text{ g/cm}^3$ , the depths corresponding to the fluid pressures calculated above range from 661 to 1371 m during the precipitation of fibrous calcite, 604 to 1503 m during precipitation of anhedral calcite, and 718 to 1666 m after this episode. It is likely that the Jubilee deposit formed at a depth less than 1500 m. These results agree with recent studies of hydrocarbon source-rocks showing that the Horton and basal Windsor Groups sediments a few tens of kilometers west of our study area did not pass the oil window conditions, i.e., did not get buried deeply (e.g., Fowler et al. 1993).

## Mechanisms of sulfide precipitation

The precipitation of sulfides may be caused either by a drop in solubility (due to temperature drop, fluid – rock interactions, etc.) if the metals were transported together with reduced sulfur, or by mixing of a metalcarrying fluid with a reduced sulfur-enriched fluid (Sverjensky 1981). Anderson (1975) pointed out that if metals and reduced sulfur were transported together, the solution must be very acid. Hayashi et al. (1990) demonstrated that zinc sulfide complexes cannot be responsible for the significant transport of zinc to form economic deposits. Sulfur isotopes of sulfides of the Jubilee deposit indicate that sulfur was likely derived from the evaporites of the Carrolls Corner Formation (Fallara 1995).

The presence of primary hydrocarbon fluid inclusions in fibrous calcite suggests that liquid hydrocarbons migrated into the high-permeability zone represented by the limestone breccia before mineralization. Carbon isotope data suggest that the carbon of fibrous calcite was mainly derived from oxidation of hydrocarbons (Armstrong *et al.* 1993, Fallara *et al.* 1994). It is likely that hydrocarbons reacted with sulfate (Anderson & Garven 1987) or thiosulfate (Spirakis & Heyl 1993) derived from the evaporites within and overlying the limestone breccia, producing oxidized carbon and reduced sulfur.

The reduced sulfur was possibly preserved in a reservoir due to the sealing of the overlying evaporites. At a later time, the metal-carrying fluid migrated into the reservoir, mixed with the pre-existing fluid, reacted with the reduced sulfur, and precipitated Zn and Pb sulfides.

### CONCLUSIONS

The microthermometric data of fluid inclusions from fibrous calcite (pre-ore), sphalerite, and anhedral calcite (syn- to post-ore) indicate that the Jubilee deposit probably formed in a shallow (less than 1500 m) burial environment with low background temperature (about 50-90°C), into which was introduced a hot (up to 230°C), metal-carrying brine during mineralization. The evolution of the mineralizing system can be summarized as follows. At some time in the premineralization period, liquid hydrocarbons migrated into the limestone breccia and reacted with oxidized sulfur (sulfate or thiosulfate or both) derived from the evaporites, producing a reservoir enriched in reduced sulfur and a high-salinity (21-27 wt%) aqueous fluid, from which fibrous calcite precipitated. During the major period of mineralization, a basemetal-carrying aqueous fluid of high salinity (about 24 wt%) and high temperature (up to 230°C) entered the reservoir of reduced sulfur and hydrocarbon, mixed with the pre-existing fluids and cooled rapidly, and precipitated Zn-Pb sulfides. Toward the waning stage of mineralization, another fluid, characterized by relatively low salinity (about 13-14 wt%) and low temperature (about 70-90°C), affected the mineralizing system, and eventually dominated the hydrothermal system in the post-ore stage.

#### ACKNOWLEDGEMENTS

This study was financially supported by the Canada – Nova Scotia Cooperation Agreement on Mineral Development. We thank Don Sangster, Suzanne Paradis, Dan Kontak and Francine Fallara for helpful discussion, Jean-Claude Bérubé for the doubly polished thin sections, Luce Dubé and Yvon Houde for drafting, and Francine Fallara for providing Figure 2. We are particularly grateful to Greg Lynch, Normand Tassé, and two anonymous reviewers for their critical review of the first draft of the manuscript. Editorial suggestions by Robert F. Martin have helped in clarifying the text.

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- Received August 3, 1994, revised manuscript accepted March 9, 1995.