

## Zoned hollingworthite from the Two Duck Lake intrusion, Coldwell complex, Ontario

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### ABSTRACT

Hollingworthite from a deposit rich in copper sulfides in olivine gabbro of the Two Duck Lake intrusion, Coldwell complex, Ontario, is complexly zoned. There are two main compositional trends: Ir- and As-rich hollingworthite is progressively zoned to pure hollingworthite (RhAsS) followed by partial dissolution, replacement, and overgrowth; the processes have caused the replacement of up to half of the Rh atoms by Os and Ru and, in one of the grains, of 0.4 of the As atoms by S. This type of zoning cannot result from simple exsolution of hollingworthite from interstitial magmatic sulfide; it is interpreted to reflect the effects of fluids that may have transported and precipitated different proportions of the Pt-group elements at different times.

### INTRODUCTION

Hollingworthite (RhAsS), defined by Stumpfl and Clark (1965), was found in Pt concentrates from the Driekop mine, South Africa. Other data on hollingworthite are derived from specimens from the same area (Tarkian and Stumpfl, 1975) and the Onverwacht mine, South Africa (Genkin, 1968). Hollingworthite is also associated with copper nickel sulfides at Noril'sk, USSR (Genkin, 1968; Genkin and Evstigneeva, 1986); Werner Lake, Canada (Rucklidge, 1969); Rathbun Lake, Canada (Rowell and Edgar, 1986); Kambalda, Australia (Hudson, 1986), and Siikakämä, Finland (Hänninen et al., 1986). Other occurrences are from Pt-group mineral (PGM) parageneses with magnetite and with serpentine and pentlandite interstitial to chromite in the Bird River Sill, Canada (Cabri and Laflamme, 1988); in chromite from Osthhammeren, Norway (Nilsson, 1990); in alpine-type ultramafic rocks (Distler et al., 1986); and in ophiolitic complexes (Tarkian and Prichard, 1987; Thalhammer and Stumpfl, 1988).

In the Coldwell complex, hollingworthite was found in close association with chalcopyrite in coarse-grained to pegmatitic olivine gabbro. The purpose of this paper is to elucidate the zoning of the hollingworthite crystals with respect to the whole paragenesis of the sulfide-bearing gabbroic host rock.

### GEOLOGICAL SETTING AND SAMPLE DESCRIPTION

The Coldwell alkaline complex, Ontario (Puskas, 1967; Mitchell and Platt, 1982), located on the north shore of Lake Superior (48°47'N, 86°30'W), is a Proterozoic (1.1 Ga) composite intrusion of silica-saturated and under-saturated plutonic rocks. It was emplaced into Archean metasedimentary, metavolcanic, and granitic rocks of the Superior Province. In the eastern part of the complex, the

arcuate belt of layered gabbroic rocks, the "Eastern Gabbro," is regarded as the earliest unit (Currie, 1980) and is host to disseminated copper nickel sulfides (Wilkinson, 1983). A Pt-group element (PGE) and copper deposit was outlined through recent exploration by Fleck Resources Ltd. (Dahl et al., 1986; Watkinson et al., 1986) in the Two Duck Lake intrusion that was emplaced into the contact area of the Eastern Gabbro with the Archean metavolcanic rocks. The host unit contains abundant inclusions of both rock types.

A disseminated pyrrhotite + chalcopyrite + pentlandite + cubanite assemblage enriched in PGE occurs in coarse-grained to pegmatitic gabbroic rocks. In sample 85-13-109.5, zoned hollingworthite occurs in a very coarse-grained sulfide-bearing olivine gabbro (Fig. 1A) composed of cumulus plagioclase ( $An_{63.9-53.6}$ ), postcumulus clinopyroxene (about  $Wo_{43}En_{40}Fs_{17}$ ), and olivine ( $Fo_{58}$ ); apatite and biotite (both F and Cl rich) are minor minerals. The crystallization order is plagioclase, clinopyroxene, olivine, followed by biotite, apatite, oxides, and sulfides. The sulfides are pyrrhotite, pentlandite, chalcopyrite, and cubanite. Pyrrhotite containing small amounts of Ni (0.3–0.42 wt%) varies from  $Fe_{0.85}S$  to  $Fe_{0.89}S$ . Pentlandite is relatively Co rich, with as much as 3.4% Co. Sample 43-290 (Fig. 1B) is also an olivine gabbro, but alteration is more pronounced with development of amphibole, chlorite, oligoclase, epidote, and calcite. Plagioclase ( $An_{57.6}-An_{46.6}$ ), clinopyroxene ( $En_{40.1-39.8}Wo_{45.3-41.2}Fs_{19.2-15.1}$ ), olivine ( $Fo_{59.1}-Fo_{57.8}$ ), biotite, hornblende, magnetite, ilmenite, and apatite are the magmatic minerals. Sulfides are composite assemblages of lamellar cubanite + chalcopyrite, enclosing anhedral and corroded pyrrhotite, and pentlandite. Chalcopyrite may also replace early magnetite (but not ilmenite lamellae) and pyrrhotite. Pentlandite occurs at the pyrrhotite-chalcopyrite interfaces. The habit of the sulfides, the reaction textures, the plagioclase

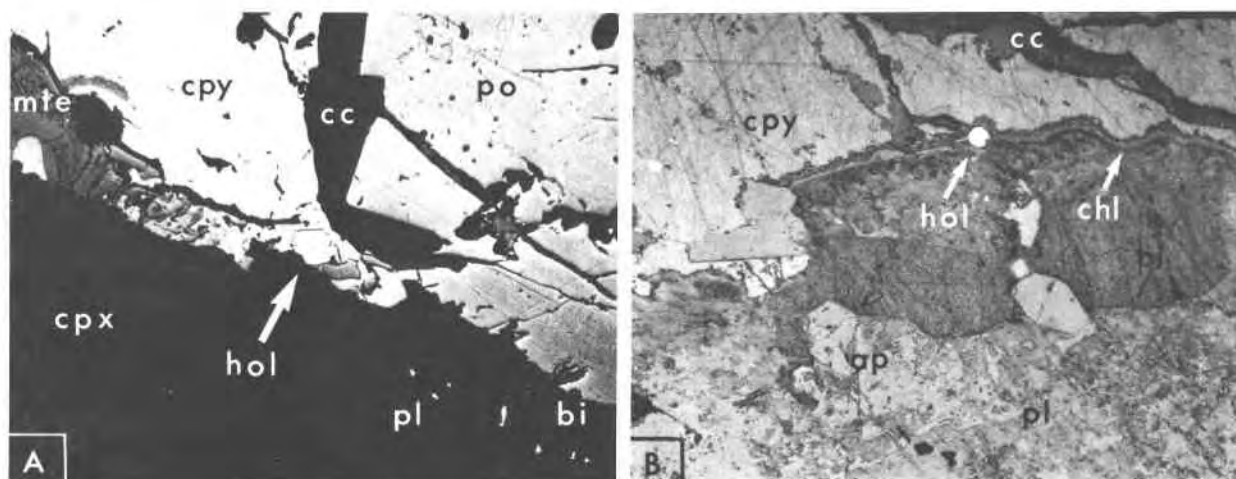


Fig. 1. Photomicrographs of sulfide-bearing olivine gabbro: (A) hollingworthite 1 ( $50 \times 65 \mu\text{m}$ ) and (B) hollingworthite 2 ( $30 \times 30 \mu\text{m}$ ). Abbreviations: pl, plagioclase; cpx, clinopyroxene; bi, biotite; ap, apatite; mte, magnetite; po, pyrrhotite; cpy, chalcopyrite; cc, calcite; chl, chlorite; hol, hollingworthite.

pseudomorphs, and the existence of Cu-rich minerals in cracks are compatible with late stage metasomatic replacement from a circulating Cu-rich fluid (Watkinson and Dahl, 1988).

The most common PGM are kotulskite and sperrylite. Kotulskite may be enriched in Bi or in Pb. The Pb-rich kotulskite is associated with zvyaginstevite and Au-bearing atokite. The Pd minerals are also present as complex intergrowths of palladoarsenide–nickeline–merseite II–majakite associated with Pd-bearing nickeline and unknown  $\text{Pd}_2(\text{Sb,As})$ .

#### ANALYTICAL METHODS

All analyses were performed on the CNRS–BRGM–University of Orléans Camebax electron microprobe with wavelength dispersive spectrometers and LiF, TAP, and PET analyzing crystals. Hollingworthite was analyzed with an acceleration voltage of 25 kV, a 20-s counting time, and a reference current of 40 nA using  $\text{OsL}\beta$ ,  $\text{IrL}\alpha$ ,  $\text{RuL}\alpha$ ,  $\text{RhL}\alpha$ ,  $\text{PtL}\alpha$ ,  $\text{PdL}\beta$ ,  $\text{AsL}\beta$ ,  $\text{CuL}\alpha$ ,  $\text{FeK}\alpha$ ,  $\text{NiK}\alpha$ , and  $\text{BiM}\alpha$  on metals standards. For  $\text{SK}\alpha$ , pyrite was used as a standard. The correction of the overlap of  $\text{RuL}\beta$  and  $\text{RhL}\alpha$

TABLE 1. Representative analyses of hollingworthite (wt%)

	1	2	3	4	5	6	7	8	9	10	11
Os	0.00	0.36	7.00	4.00	3.08	9.38	10.07	1.07	1.16	2.05	0.45
As	34.04	34.69	29.28	31.01	34.12	28.10	26.74	39.11	35.01	34.41	33.05
S	14.92	13.77	16.06	14.89	13.59	17.35	16.96	11.75	12.41	13.75	12.45
Ir	0.70	2.50	0.36	0.39	2.46	0.23	0.57	2.59	4.46	0.60	15.64
Cu	0.00	0.00	0.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ru	0.00	0.01	4.17	2.47	1.07	5.61	6.41	0.00	0.00	0.17	0.02
Rh	44.54	40.31	28.52	34.90	35.39	25.02	24.89	35.06	34.31	38.78	28.67
Pt	3.91	7.36	13.43	11.16	9.30	13.05	12.47	9.82	10.25	8.45	8.30
Pd	1.30	0.90	0.05	0.17	0.42	0.03	0.06	0.73	1.36	0.28	1.51
Fe	0.40	0.54	1.38	0.73	1.01	2.14	1.12	0.51	0.27	0.42	0.30
Ni	0.18	0.14	0.05	0.06	0.09	—	—	0.15	0.22	0.05	0.20
Total	99.99	100.58	100.38	99.78	100.53	100.91	99.29	100.79	99.45	98.96	100.59
<b>Formulae</b>											
Os	0.000	0.004	0.082	0.047	0.036	0.108	0.119	0.013	0.014	0.024	0.006
As	0.975	1.023	0.872	0.928	1.024	0.821	0.804	1.189	1.086	1.032	1.052
S	0.998	0.949	1.117	1.041	0.953	1.184	1.192	0.835	0.900	0.964	0.926
Ir	0.008	0.029	0.004	0.005	0.029	0.003	0.007	0.031	0.054	0.007	0.194
Cu	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ru	0.000	0.000	0.092	0.055	0.024	0.121	0.143	0.000	0.000	0.004	0.001
Rh	0.928	0.866	0.618	0.760	0.773	0.532	0.545	0.776	0.775	0.847	0.665
Pt	0.043	0.083	0.154	0.128	0.107	0.146	0.144	0.115	0.122	0.097	0.101
Pd	0.026	0.019	0.001	0.004	0.009	0.001	0.001	0.016	0.030	0.006	0.034
Fe	0.015	0.021	0.055	0.029	0.041	0.084	0.045	0.021	0.011	0.017	0.013
Ni	0.007	0.005	0.002	0.002	0.003	0.000	0.000	0.006	0.009	0.002	0.008

Note: Analyses 1–7 correspond to grain 1 of hollingworthite, 8–11 to grain 2.

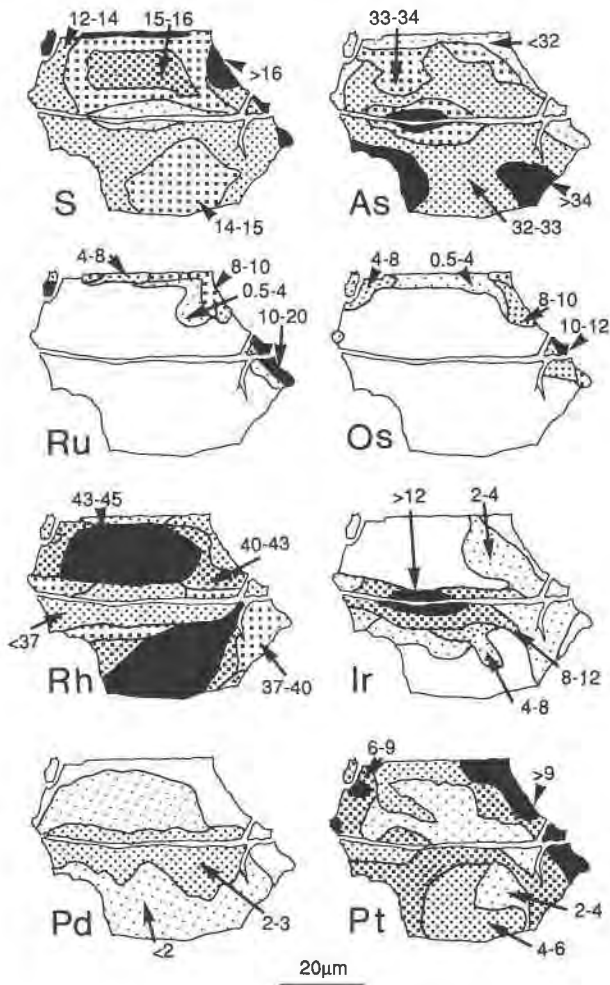


Fig. 2. Contour diagram, in weight percent for the different elements, drawn from microprobe analyses of the zoned hollingworthite grain 1.

observed for Ru-rich minerals (Ohnenstetter et al., 1986) was not deemed necessary for the low to very low Ru zones of hollingworthite. All data were corrected with the ZAF program MBXCOR of Henoc and Tong (1978). Backscattered images were performed on Cambridge Stereoscan 200 SEM at the BRGM and at the Geological Survey of Canada, and the X-ray scanning was performed on the Camebax microbeam. To compare the X-ray scanning with the probe data, 137 analyses were performed on a  $50 \times 65 \mu\text{m}$  zoned euhedral hollingworthite and 80 on a second hollingworthite ( $30 \times 30 \mu\text{m}$ ). Typical compositions of hollingworthite are listed in Table 1 and illustrated in Figures 2–6; all data are reported by Ohnenstetter (1990).

#### HOLLINGWORTHITE ZONING

Hollingworthite is isometric with a cobaltite structure (Hulliger, 1963), as are irarsite (IrAsS) and platarsite (PtAsS). Osarsite (OsAsS) and ruarsite (RuAsS) are

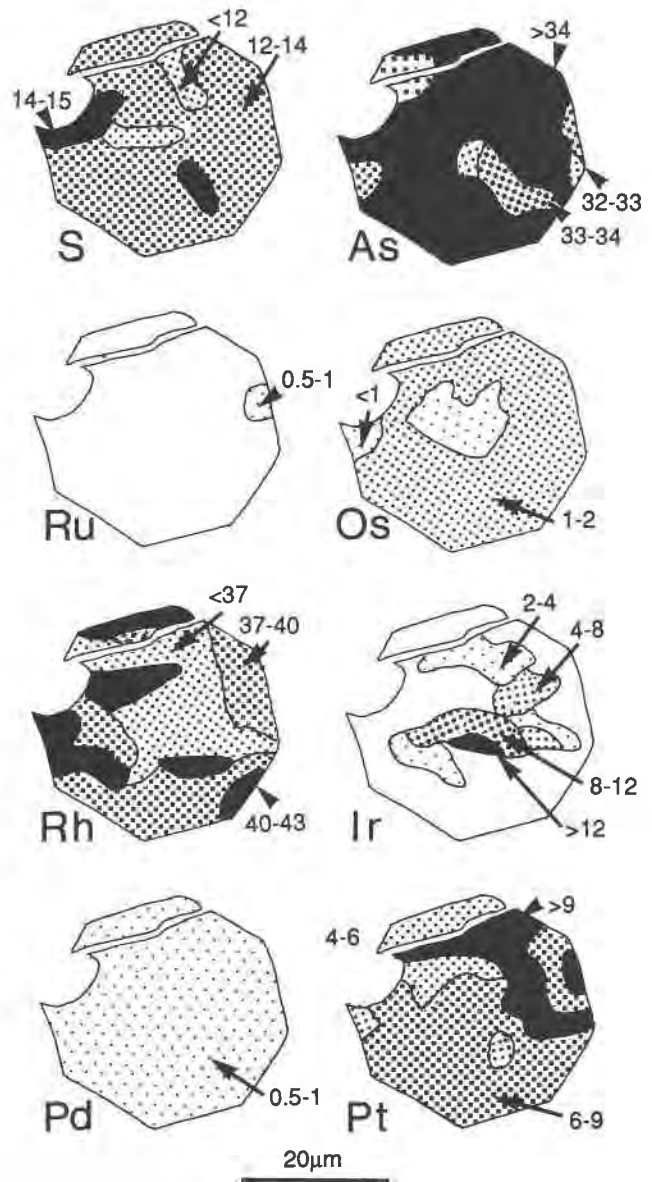


Fig. 3. Contour diagram, in weight percent for the different elements, drawn from microprobe analyses of the zoned hollingworthite grain 2.

monoclinic (Snetsinger, 1972) with arsenopyrite-type structure (Hulliger, 1964). No Pd equivalent is known. The different crystal structures of the MeAsS minerals and the similar sizes of the relevant atoms indicate that Ir and Pt could substitute for Rh more easily than would Os and Ru in hollingworthite. Laurite ( $\text{RuS}_2$ ) and erlichmanite ( $\text{OsS}_2$ ) have the same  $Pa3$  space group as does hollingworthite but with the pyrite structure (Leonard et al., 1969; Snetsinger, 1971).

As with other hollingworthite (Cabri, 1981), that from the sulfide-bearing olivine gabbro at Coldwell is marked by zoning as shown by contoured diagrams (Figs. 2 and 3). Growth zoning is modified by late dissolution and

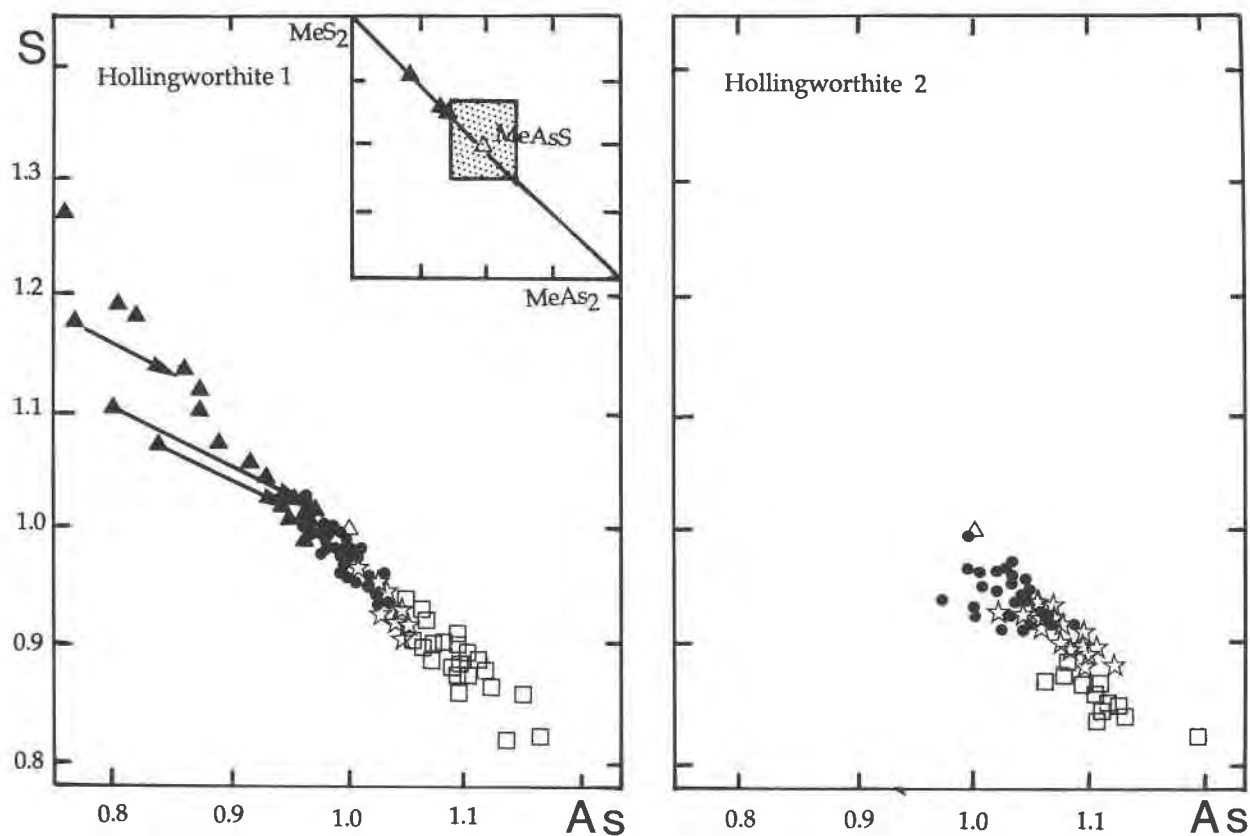


Fig. 4. S vs. As (atomic percent) in hollingworthite 1 and 2. Dots, nearly pure hollingworthite; filled triangles, Ru- or Os-enriched hollingworthite; squares, Ir-rich hollingworthite; stars, Pt-rich zones; open triangles, stoichiometric RhAsS. Inset shows location of rectangles and outlying data.

replacement along cracks and at the edges of the grains. Nearly pure hollingworthite replaces Ir-rich zones in the center of the crystals. Both may be replaced by late Pt-enriched hollingworthite, especially along cracks. It appears that after these two major replacement stages, which exist in both grains, a new influx of Pt followed by Os

and Ru influxes modified the previous minerals especially at the rims of hollingworthite 1 (Fig. 2).

In hollingworthite 1, the core has low contents of Fe (0–0.5%), Ni (0.1–0.3%), and Pd (2–3%) with Ir content as high as 23 wt%. Pt values (2–4%) are also low and no Os and Ru were detected. In the Pt-enriched zones, Fe

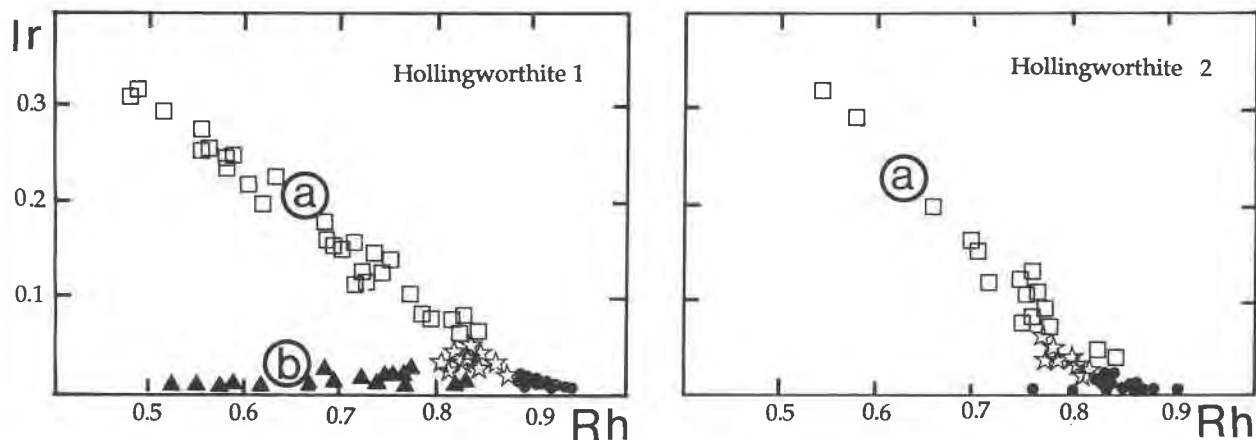


Fig. 5. Ir vs. Rh in hollingworthite 1 and 2 (atomic percent). Note two trends for grain 1: a = negative correlation between Ir and Rh and b = Ru-enriched hollingworthite (laurite-erlichmanite–hollingworthite solid solution) at constant Ir. Symbols are as those in Figure 4.

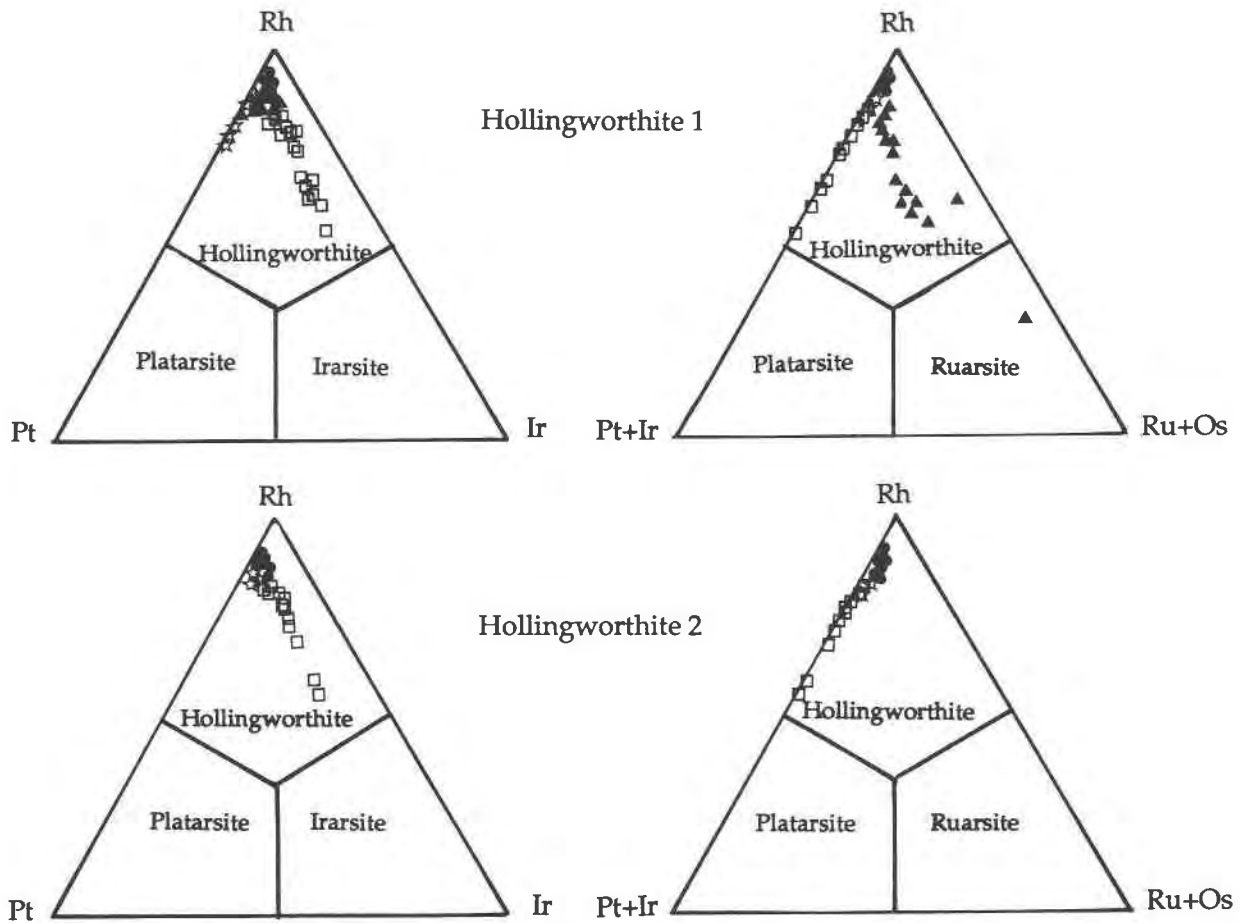


Fig. 6. Rh-Pt-Ir and Rh-(Pt + Ir)-(Ru + Os) triangular plots after Tarkian (1987) for the two hollingworthite grains (atomic percent). Symbols are as those in Figure 4.

increases slightly (0.5–1%), whereas Rh decreases. This stage is marked by overgrowth of a Pt-enriched zone (up to 13.8%) followed by Os- (up to 10%) and Ru- (up to 9.2%) enriched zones, whereas Pd, Ni, As (20–32%), and Rh (<30 wt%) decrease. The composition of the thin rims is generally modified by analytical border effects that cause low totals of the analyses (95–97%). Close to chalcopyrite, an apparent increase in Fe and Cu in the rim results from contamination from the surrounding mineral. In the second hollingworthite (Fig. 3), the core is Ir rich (22.9 wt%) with Ni and Fe values about 0.3%. However, Pt content is high (near 10%), whereas Ru and Os are less than 2%. Partway to the edge, Pt-enriched zones have lower Ir and Rh contents, and Fe and Ni values are similar to those in the core area. The rim is enriched in Pt (about 9%) and poor in Ni (<0.1 wt%).

Three major stages of crystallization may be pointed out in the two hollingworthite crystals (Fig. 4): (1) crystallization of Ir- and As-rich hollingworthite (close to the irsarsite composition), (2) partial resorption of this phase and precipitation of nearly pure hollingworthite, and (3) resorption along the edges of the grain and in cracks to generate Pt- and Fe-enriched hollingworthite as well as

enrichment in Pd along the main transverse cracks, growth of an outer rim enriched in Pt, or in Os and Ru, or in all three, and depletion in Pd and Ni.

#### SUBSTITUTION IN HOLLINGWORTHITE

Two main substitutions in hollingworthite are defined (Figs. 5 and 6). The first, observed in both grains, is irsarsite-hollingworthite substitution with replacement of Ir by Rh coupled with an increase of the S-As ratio. The transition is clear in Figure 5 from a composition that is Ir rich toward nearly pure RhAsS (Begizov et al., 1976). The second, observed toward the rim of one grain, corresponds to the substitution of laurite-erlichmanite for hollingworthite. The Ru and Os enrichments are also accompanied by an increase in S (Fig. 4). Our data indicate a wide range of solution between hollingworthite and laurite marked by the replacement of 0.4 formula units of As by S (Fig. 4) and half the Rh by Ru and Os (Fig. 5). The As-S substitution covers a range from  $\text{MeAs}_{0.40}\text{S}_{1.60}$  to  $\text{MeAs}_{1.18}\text{S}_{0.82}$ , with most of the analyses close to the line  $\text{MeS}_2\text{-MeAs}_2$ . The three analyses that plot outside the general trend on the left part of Figure 4 are contaminated by chalcopyrite; they fall on the trend after

chalcopyrite subtraction (arrows). The As-S substitution is characterized by a strong S depletion. The highest As value in hollingworthite ( $\text{MeAs}_{1.36}\text{S}_{0.64}$ ) is from Feather (1976). Irsite and osarsite from the Witwatersrand may have similarly high As values (Feather, 1976). However, there is little or no As-S substitution exhibited either by the  $\text{MeS}_2$  series [laurite-erlichmanite (Cabri, 1981),  $\text{IrS}_2$  (Ren and Deng, 1973), and  $\text{RhS}_2$  (Feather, 1976)] or by the  $\text{MeAs}_2$  series [iridarsenite (Harris, 1974), anduoite (Yu and Chou, 1979), omeiite (Ren et al., 1978), and sperrylite (Feather, 1976; Cabri et al., 1977)]. Cabri and Laflamme (1988) have also described an overgrowth of laurite on hollingworthite.

### CONCLUSION

The precipitation of highly zoned hollingworthite is clearly later than the crystallization of sulfide-bearing magma in the Two Duck Lake intrusion, Coldwell complex. Hollingworthite is spatially related to veins and alteration assemblages of iron chlorite + epidote + actinolite + calcite, as are other PGM in the Two Duck Lake rocks, and it is characterized by compositional variation in response to the chemical change of fluid with time. Two successive main substitutions are defined between irsrite or Ir-rich hollingworthite and hollingworthite and between laurite-erlichmanite and hollingworthite. During these events, As decreased more than 0.4 formula units, whereas S increased in response to an increase of  $f_{\text{S}_2}$  with time. Variations in hollingworthite compositions thus reflect variations in the composition of fluid, perhaps as saline fluid became enriched in S by the breakdown of pyrrhotite; they cannot be readily explained by exsolution of PGM from magmatic sulfides. Fluids caused some alteration of original silicates and oxides, modified the original magmatic sulfides through replacement of the Fe minerals by chalcopyrite, and precipitated Pt-group minerals (Watkinson and Dahl, 1988).

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