The isotopic sulphur composition of two barite samples from Rose Mountain area near Faro, Yukon

Lee C. Pigage¹

Yukon Geological Survey

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ABSTRACT

The Rose Mountain area northwest of Faro is underlain by a succession of phyllites, cherts, sandstones, chert-pebble conglomerates and basalt belonging to the Slide Mountain Terrane. At least two, laterally extensive, massive barite horizons are interbedded with phyllite, bedded chert, sandstone and chert-pebble conglomerate of the Early Carboniferous Mount Aho formation, the lowermost unit in this succession. Samples from two barite horizons produced δ^{34} S values (‰) of +14.3 ± 0.2 (2 σ) and +13.8 ± 0.2 (2 σ), which are consistent with δ^{34} S values for coeval dissolved sulphate in seawater. Ambient seawater is therefore the most probable source of the sulphate in the barite. The results suggest barite precipitated from mixing of sulphate-poor hydrothermal fluids with ambient seawater rather than inversion of anoxic, stagnant, stratified basin waters.

RÉSUMÉ

La région du mont Rose, au nord-ouest de Faro, repose sur une succession de phyllites, de cherts, de grès, de conglomérats de galets de chert et de basalte appartenant au terrane de Slide Mountain. Une stratification entremêlée de phyllite, de chert lité, de grès et de conglomérat de galets de chert, appartenant à la formation de Mount Aho du Carbonifère précoce, forment les couches lithologiques les plus basses de la succession. De multiples horizons étendus de barytine et de chert phylliteux massifs sont interstratifiés avec ces roches sédimentaires. Des échantillons de deux horizons de barytine ont donné des valeurs (‰) de δ^{34} S de +14,3 ± 0,2 (2 σ) et de +13,8 ± 0,2 (2 σ), qui sont cohérentes avec les valeurs de δ^{34} S des sulfates du même âge dissous dans l'eau de mer. Par conséquent, l'eau de mer ambiante représente la source la plus probable de sulfates dans la barytine. Les résultats indiquent que la barytine n'a pas été précipitée par l'inversion des eaux de bassin anoxiques, stagnantes et stratifiées. Cependant, les valeurs de δ^{34} S résultantes permettraient une précipitation de la barytine résultant du mélange de fluides hydrothermaux à faible teneur en sulfates avec l'eau de mer ambiante.

¹lee.pigage@gov.yk.ca

INTRODUCTION

The isotopic composition of dissolved sulphate in modern ocean water is constant (δ^{34} S = +20.0‰) because of the rapid mixing of oceanic currents relative to the average residence time of sulphate in seawater (Claypool et al., 1980). This ratio has varied between +10‰ and +30‰ through geological time (Claypool et al., 1980); the variation results from a multitude of global processes, including climate change, volcanic activity, plate tectonics and ocean water circulation (Kampschulte et al., 2001). In earlier studies, this sulphur isotope variation in seawater sulphate through geologic time was tracked by measuring of isotopic compositions of evaporite calcium-sulphate deposits (gypsum and anhydrite) and barite deposits (Claypool et al., 1980). More recently, isotopic composition of trace sulphate dissolved in fossil shell material and whole-rock carbonate has also been used to refine geologic seawater sulphate compositions (Kampschulte et al, 2001) through geologic time.

Slide Mountain stratigraphy near Rose Mountain (NTS 105K/5 and 6) contains thin, laterally extensive, stratiform barite horizons. Sulphur isotope compositions for two barite samples from different horizons were measured during a regional compilation project of the Anvil District completed by the Yukon Geological Survey. An Early Carboniferous age for both barite horizons was also successfully determined from conodont collections obtained during the same project. This article presents the sulphur isotope data in the context of the fossil control, compares the isotope values to sulphur isotopes of coeval seawater, and briefly discusses how results are consistent with open oceanic circulation during precipitation of the barite.

SULPHUR ISOTOPE SYSTEMATICS

Sulphur has four stable isotopes 32 S (95.02%), 33 S (0.75%), 34 S (4.21%) and 36 S (0.02%) (Faure and Mensing, 2005). Isotopic sulphur composition is reported as the expression:

 $\delta^{34}S(\%) = [((^{34}S/^{32}S)sa - (^{34}S/^{32}S)std)/(^{34}S/^{32}S)std]*1000$

where "sa" represents the sample and "std" represents the standard. The reference sulphur isotope standard for this expression is the isotopic ratio of ${}^{34}S/{}^{32}S$ (= 0.0450) for the mineral troilite (FeS) in the Canyon Diablo meteorite (CDT). The value for CDT is very similar to the measured values of sulphur isotopes in mafic igneous rocks.

Variations in sulphur isotopic composition are caused by two kinds of processes (Faure and Mensing, 2005):

- 1) reduction of sulphate to hydrogen sulphide by anaerobic bacteria. During the reduction process, hydrogen sulphide is enriched in ³²S and the residual sulphate is enriched in ³⁴S;
- 2) isotopic exchange reactions occurring during exchange of sulphur between different ions, molecules and solids. During this process, ³⁴S is typically slightly enriched in compounds having the highest sulphur oxidation state and/or strongest bond strength.

Biogenic sulphate reduction (process 1 above) is the most important source of sulphur fractionation in nature. In an open system, fractionation is reasonably constant and depends on the physical parameters at the site of deposition. If sulphate availability is restricted (closed system), both remaining dissolved sulphate and reduced hydrogen sulphide have increasing δ^{34} S values with continuing biogenic reduction of sulphate and precipitation of sulphide minerals.

Sulphur isotope fractionation between gypsum mineral and dissolved sulphate in seawater during precipitation of the gypsum in modern sediments ranges from 0 to +2.4‰ (Strauss, 1997). Calcium sulphate therefore accurately reflects the dissolved sulphate isotopic composition of seawater at the time of precipitation. Recent work on marine barite indicates that barite also may accurately reflect dissolved sulphate compositions of contemporaneous seawater (Cecile et al., 1983; Paytan et al., 1998).

GEOLOGY FOR ROSE MOUNTAIN SAMPLES

Rose Mountain is located on the southwest flank of the Anvil District in central Yukon (Fig. 1), approximately 200 km northeast of Whitehorse and 14 km northwest of the town of Faro. Outcrop is extensive on ridges above tree line (approximately 1370 m) and is restricted to stream cuts and scattered ridge crests below that. Valley bottoms are typically covered with thin to thick glacial till. Overgrown exploration roads and outfitting trails extend into the area. Access is most readily accomplished by helicopter. Camps during the 1998 field season were placed using contract helicopter service based in Ross River.

The area lies within Tay River map sheet (NTS 105K). Regional geology was mapped by Roddick and Green



Figure 1. Location of Rose Mountain in Yukon. Modified form Wheeler and McFeely (1991).

(1961), and Gordey and Irwin (1987). More detailed studies by Tempelman-Kluit (1972) and Gordey (1990) were completed in response to exploration interest generated by the discovery of the massive sulphide deposits in the Anvil District. Jennings and Jilson (1986) and Pigage (2004) have also presented regional stratigraphic and structural summaries which have included this area.

Early exploration activity occurred dominantly on the north slopes of Rose Mountain and was focused primarily on lead-zinc targets because of the Anvil District massive sulphide discoveries. The two barite horizons discussed here were staked as the URN Claims by Cyprus Anvil Mining Corporation in 1977. The upper horizon was sampled in several locations in 1977 (Franzen, 1978) and 1981 (Read, 1982) to evaluate its industrial mineral potential. Exploration work on the URN claims is summarized in Yukon MINFILE (105K 106, Deklerk and Traynor, 2004).

Two or more barite horizons (Figs. 2 and 3) are contained within a uniformly southwest-dipping sequence of dark



Figure 2. Major tectonic/stratigraphic successions. Modified from Pigage (2004)

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grey to black, bedded chert, chert-pebble conglomerate, sandstone, and siliceous phyllite and sandstone belonging to the Mount Aho formation (Pigage, 2004). The Mount Aho formation ranges up to 1500 m thick, and has been informally divided into a lower pale green member and an upper dark grey member. Barite horizons occur in both members.

Mount Aho formation is the lowest unit in a continuous succession of three formations. Bedded cherts and phyllites of the Rose Mountain Formation conformably(?) overlie the Mount Aho formation and are in turn capped by basalts of the Campbell Range formation (Pigage, 2004). Tectonic affiliation of this assemblage has been controversial (Jennings and Jilson, 1986; Pigage, 1999 and 2004). Most recently it has been assigned to the Slide Mountain Terrane, an ocean-floor back-arc basin in Carboniferous-Permian time, which was subsequently partially subducted and amalgamated between ancestral North America and Yukon-Tanana Terrane (Tempelman-Kluit, 1979; Creaser et al., 1999; Pigage, 2004). The lower contact of Mount Aho formation is the Inconnu fault, the major northeast-verging thrust fault placing Slide Mountain Terrane onto ancestral North America.

Both members of the Mount Aho formation contain barite. The immediate host for the barite in both members consists of tan-weathering, pale cream, phyllitic bedded chert and argillite ranging up to 40 m thick. Barite is nodular to massive within bedded chert and argillite. Sulphide minerals are not visibly associated with the barite. Exploration work by Cyprus Anvil Mining Corporation (Franzen, 1978) and recent mapping by Pigage (2004) has confirmed that the barite horizons can be laterally traced along strike for several kilometres. The other major mineral associated with the barite is quartz (Read, 1982).

SULPHUR ISOTOPES FOR ROSE MOUNTAIN BARITE

Two samples of barite were collected from different barite horizons within the Mount Aho formation, one near the bottom and one near the top of the upper black to grey member. Locations of the samples are indicated in



Figure 4. Schematic stratigraphic column showing barite horizons with sample locations indicated. Based on geology from Pigage (2004).

Sample	98LP069	98LP082
Barite horizon	Upper horizon	Lower horizon
UTM East	576 481	578 008
UTM North	6 912 485	6 912 358
UTM Zone	8	8
Datum	NAD83	NAD83
S ³⁴ (‰, CDT)	+14.3	+13.8
2σ precision	±0.20	±0.20

Table 1. Isotopic sulphur composition of barite samples.

Figure 3; UTM coordinates are presented in Table 1. Figure 4 is a schematic stratigraphic section for the two samples indicating their relative location approximately 600 m apart stratigraphically, assuming a homoclinal southwest-dipping succession. Figure 4 also indicates approximate locations of two productive conodont samples which returned Early Carboniferous ages (Orchard, 2000).

SAMPLE 98LP082

Sample 98LP082 was collected from a barite horizon located in the pass at the south end of Cornice Ridge (Fig. 3). Medium crystalline, grey barite forms beds up to 10 cm thick (Fig. 5) within light cream to tan argillite and chert. Exposed surfaces weather to a very rusty brown, and the barite contains a poorly developed slaty cleavage. Overall thickness of the horizon is about 10 m, and bedding dips 20 degrees to the southwest. The barite horizon cannot be traced laterally along strike away from



Figure 5. Barite horizon at field station 98LP082. Massive barite occurs in tan chert and argillite. Pencil magnet is 12.5 cm long.

the pass. Metasedimentary rocks on both upper and lower contacts consist of black, siliceous argillite. The barite horizon occurs in the lowermost part of the upper member of the Mount Aho formation; within metres, the black siliceous argillite changes to interbedded maroon and green chert and argillite.

SAMPLE 98LP069

Sample 98LP069 was collected from an interbedded sequence of barite and light creamy white weathering chert and siliceous argillite (Fig 3) on a small ridge at the south end of Rose Mountain. Bedding dips 20 degrees to the northwest, and this horizon can be traced laterally, with discontinuous occurrences along strike for up to 5 km. The baritic horizon is at least 10 m thick. It is underlain by a dark grey to black chert-pebble conglomerate with abundant clasts of grey and black chert and overlain by a dark, dull grey, fine siltstone to silty argillite. The horizon is located near the top of the upper member of the Mount Aho formation. Two condont samples collected within short distances above and below this horizon (Fig. 4) returned Early Carboniferous ages (Orchard, 2000 in Pigage, 2004) with the upper sample being identified as Visean.

Sulphur isotope analyses were completed at the G.G. Hatch Isotope Laboratories, University of Ottawa, Ottawa, Ontario. The two barite samples were crushed, mixed with a set ratio of copper-oxide and quartz powders, and combusted at 1100°C. The resulting gases produced were cryogenically separated using a -80°C ethanol bath and -200°C liquid nitrogen. The amount of SO₂ generated was measured as it was collected in a breakseal pyrex tube. SO₂ in the tube was analysed on a triple collector VG SIRA 12 mass spectrometer. Two calibrated internal barite standards were run at the same time as the Rose Mountain barite samples. The calibrated standards were used to normalize the Rose Mountain barite data by linear regression. Routine precision of the analyses was $\pm 0.20\%$ (2 standard deviations).

Results of the analyses are presented in Table 1. Isotopic compositions are reasonably consistent. Without multiple analyses from each sample location, it would be difficult to delineate the range in δ^{34} S for a particular horizon.



Figure 6. Carboniferous isotopic sulphate composition for ambient seawater. Measured sulphate values for barite samples shown assuming Visean age. Modified from Kampschulte et al. (2001) using geologic time scale from Harland et al. (1990). Black rectangles=Rose Mountain samples with associated error, black dashed line=carboniferous seawater curve; thick, shaded line=error spread on curve.

DISCUSSION AND CONCLUSION

Kampschulte et al. (2001) recently published a detailed δ^{34} S curve for Carboniferous seawater (Fig. 6). The sulphur isotope variation is based on measured trace sulphate compositions within brachiopod fossils. The time scale of Harland et al. (1990) was used to translate fossil zones to absolute ages for tracking the sulphur isotope variation with time. Kampschulte et al. (2001) do not give an estimated error for their δ^{34} S curve; visual inspection of their data points indicate an estimated spread of ±1.5‰, which incorporates all of the spread in the sample data points. This "error" spread is indicated in Figure 6 as the shaded area around the calibrated curve.

The measured δ^{34} S values for the Rose Mountain barite samples are also shown in Figure 6. The Rose Mountain barites have δ^{34} S values consistent with the calibrated values of coeval dissolved seawater sulphate during Visean (and Serpukhovian) time. When combined with the Early Carboniferous (Visean?) age from the conodont assemblages, these results indicated that ambient, oxygenated seawater is a reasonable source for the sulphate in the barite.

Stratiform massive barite deposits form as chemical sediments, either from hydrothermal fluids or from diagenetic or cold-seep fluids (Paradis et al., 1998; Koski and Hein, 2003). Hydrothermal barite is commonly associated with sulphide minerals (pyrite, galena, sphalerite), and commonly forms a zoned deposit with sulphide minerals being vent-proximal and barite being distal. Depositional environment is shallow- to deep-water and occurs within intracratonic or continental margin, fault-controlled marine basins. Sulphur isotope composition of barite precipitated at 21°N, at the East Pacific Rise, is the same as that of ambient seawater sulphate, and indicates the source of sulphur for barite deposition is seawater sulphate (Zierenberg et al., 1984). Lydon et al. (1985) argued from solubilities of barite, calcite, sphalerite, quartz and amorphous silica that Devono-Mississipian sulphide-absent barite deposits in Selwyn Basin precipitated from lower temperature hydrothermal fluids (100°C formational waters) relative to sulphide-bearing barite deposits (>200°C formational waters).

Diagenetic or cold-seep barite deposits are thought to result from precipitation of barite at an anoxic-oxygenated seawater interface. An oxygenated-anoxic interface may develop locally in the sub-seafloor or may develop as a result of stratification of a marine basin with reduced waters at the ocean floor in the basin and oxygenated waters occurring higher in the water column. Barium remains in solution in the reduced waters, but precipitates when exposed to overlying or nearby oxygenated waters. Inversion of stratified basin waters would provide a mechanism for mixing of fluids and formation of extensive barite deposits. Sulphur isotope compositions of barite from these deposits is commonly heavier than contemporaneous seawater sulphate, due to the isotope fractionation associated with bacterial reduction of sulphate in the reduced anoxic seawater. Goodfellow and Jonasson (1986) noted that in Lower Paleozoic Selwyn Basin, barite δ^{34} S values are heavier by +2‰ to +20‰ relative to contemporaneous seawater sulphate. They attributed these heavier values to anoxic, stratified basinal waters which periodically underwent inversion with subsequent rapid deposition of barite. Cecile et al. (1983) noted that barite deposits with heavy δ^{34} S values in the western Canada Cordillera were from thin or nodular barite deposits in organic-rich sediments. They hypothesized a cycling barite precipitation-dissolutionreprecipitation process in a zone encompassing both reducing and oxidizing conditions. Some of the released sulphate during the dissolution process is bacterially reduced to hydrogen sulphide, thereby fractionating the remaining sulphate to heavier δ^{34} S values.

The preliminary δ^{34} S values of + 14.3‰ and +13.8‰ for Rose Mountain barites (2 samples) indicates that ambient seawater is the most probable source of sulphate in the barite. Further, the δ^{34} S values from the barite are not more positive than contemporaneous seawater sulphate, suggesting that the barite did not precipitate as a result of diagenetic oxidation-reduction reactions or inversion of anoxic, stratified basin waters. The measured δ^{34} S values therefore suggest the alternative process of precipitation of barite from mixing of low-temperature, sulphate-poor hydrothermal fluids with ambient seawater.

Physical characteristics of the barite horizons are inconclusive. The 5-km lateral extent for the upper horizon suggests barite precipitation is not occurring at a single vent source, and perhaps is occurring at a basinwide change in barium solubility. The absence of visible organic material (creamy tan colour) of the immediate host for the barite horizons points to local oxygenated conditions or alteration in a more general reducing basin (black cherts and argillites).

Lydon et al. (1985) suggested that stratiform barite deposits and barite-sulphide deposits within Selwyn Basin both formed from exhalation of hydrothermal fluids at the sea floor, with sulphide-bearing deposits precipitating from higher temperature fluids sourced from deeper levels of the crust. The deposit models for these two types of stratiform barite have the same processes involved in their genesis. The occurrence of stratiform barite deposits in Selwyn Basin therefore indicates that genetic parameters are also broadly appropriate for the presence of stratiform barite-sulphide deposits in other parts of Selwyn Basin. Similarly, the presence of barite deposits in the Rose Mountain area indicates that the Slide Mountain stratigraphy is a viable exploration target for barite-sulphide deposits, as well as barite deposits.

It must be remembered that these results are preliminary and are based on only two δ^{34} S measurements. For more rigorous determination of depositional environment and possible source areas for barium-rich fluids, barite should be more extensively sampled for sulphur isotope studies to verify continued correspondence to coeval seawater sulphate. Chemical analysis of the barite samples and determination of Sr isotope ratios further help distinguish between different types of barite (Paytan et al., 2002).

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