

Isotopic signature of hydrothermal sulfates from Carlin-type ore deposits

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ABSTRACT

Stable isotopes from sulfates of three origins in Carlin-type deposits (CTDs) from western North America fall into distinct clusters and can be utilized to discriminate between barite of Paleozoic (sedimentary exhalative) origin, barite of hydrothermal origin related to gold mineralization, and supergene alunite and jarosite of weathering origin. Although the latter are obvious from field relations, the distinction between hydrothermal and sedex barite is not always so clear. Sedex sulfate sulfur was probably reduced and mobilized during gold mineralization, equilibrated with hydrothermal fluids, and then precipitated as sulfate during the late stages of mineralization, with a distinctly different sulfur and oxygen isotope signature. Isotopic signatures of sulfates may, therefore, be utilized as a mineral exploration tool for CTDs.

1 INTRODUCTION

Sedimentary rock-hosted disseminated gold deposits, also known as Carlin-type gold deposits (CTDs), are among the most economically important types of ore deposits currently being sought and mined in the US. In excess of 100 million ounces of gold reserves have been identified in northern Nevada, the area hosting the largest proportion of these ore deposits (Teal and Jackson, 1997). Although high-grade portions of these deposits, such as at Mercur (Utah) and Getchell (Nevada), have been mined since the last century (Joralemon, 1951; Jewell and Parry, 1987), they were not recognized as a class until the discovery and development of Carlin as a bulk-tonnage mine between 1962 and 1965 (Hausen and Kerr, 1968).

Figure 1 shows the location of many of the major CTDs in western U.S.A. Although additional deposits which may be Carlin-type or similar deposits are known from locations as diverse as southern China (Ashley et al., 1991); southeast Asia (Garwin et al., 1995); and possibly Peru (Alvarez A. and Noble, 1988), the majority of deposits for which there are more than descriptive data available are located in the Great Basin of western North America.

2 DEPOSIT PARAGENESIS

Carlin-type deposits are commonly hosted in silty carbonates to calcareous siltstones and shales, although a number of other rock types may host ore locally (Arehart, 1996). Gold in Carlin-type deposits is closely associated with hydrothermally-generated arsenian pyrite, pyrite, and locally arsenopyrite (Wells and Mullens, 1973; Arehart et al., 1993a, b); lesser but significant amounts of gold occur

in jasperoidal quartz and minor amounts are associated with phyllosilicates and carbonaceous matter (Hausen and Kerr, 1968; Bakken and Einaudi, 1986; Hofstra et al., 1988). Temporally late in the gold stage are arsenic sulfides (realgar and orpiment), that slightly postdate the dominant alteration minerals such as kaolinite, sericite and quartz. Penecontemporaneously or somewhat later in the paragenetic sequence are barite, stibnite, and late calcite, commonly as open fracture fillings in silicified rocks. Late oxide minerals related to weathering, including the sulfates alunite and jarosite, are common above the primary ores

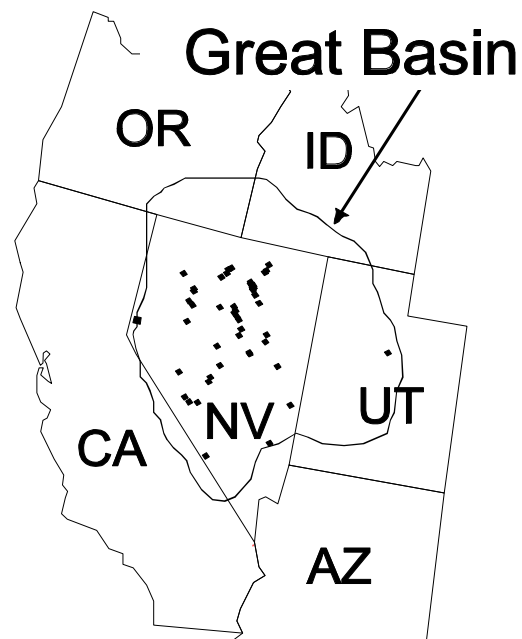


Figure 1. Location map of CTDs in western North America.

3 STABLE ISOTOPE DATA

Sulfur isotope values reported from CTDs span a wide range, from -30 to +20‰ in sulfides and 0 to >35‰ in sulfates (Figure 2). It has long been observed that chemical and optical zoning may exist in many sulfides, and it has been shown recently that many sulfides are isotopically zoned as well (Eldridge et al., 1994). In CTDs, this isotopic zoning correlates with chemical zoning of As and Au in the pyrite (Arehart et al., 1993a, b). Pyrite having the highest $\delta^{34}\text{S}$ values also contains the highest concentrations of gold. These zones also contain high concentrations of As (but not all high-As zones contain Au). Calculated and measured $\delta^{34}\text{S}$ values of gold-bearing pyrite are near +20‰, whereas all non-ore pyrite is significantly lighter (Arehart et al., 1993b, Hofstra, 1997). Extremely low sulfur isotope values are represented by the latest stage marcasite or pyrite, which have $\delta^{34}\text{S}$ values as low as -30‰ (Arehart et al., 1993b).

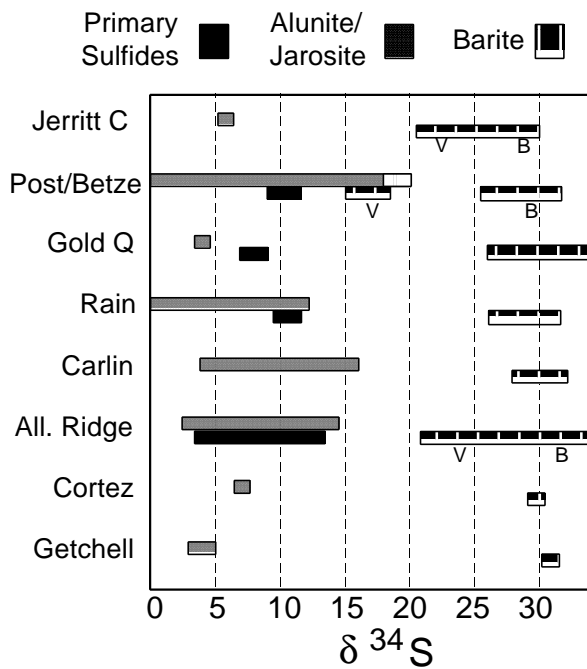


Figure 2. Sulfur isotope values of sulfides and sulfates in CTDs of the Great Basin. V=vein barite, B=sedex barite.

Sulfur and oxygen isotope values of sulfates that are spatially associated with the ore also are highly variable and fall into three distinct groups (Figures 2, 3). Some of these values are clearly related to primary sedimentary sulfate, which is abundant in the Paleozoic section in the Great Basin; the isotopic composition of these sulfates falls directly on the seawater evolution curve (Claypool et al., 1980), as would be expected. Other sulfates are clearly related to postore processes (e.g. alunite, Arehart et al., 1992) and have inherited their sulfur (and consequently their $\delta^{34}\text{S}$ values) through the process of near-quantitative oxidation of sulfides in the near-surface environment (Field, 1966; see Figure 2). When these non-hydrothermal sulfates are excluded from the data, the remaining sulfates

(barite) have $\delta^{34}\text{S}$ values ranging between +15 and +25‰ and $\delta^{18}\text{O}$ values near zero (Figure 3).

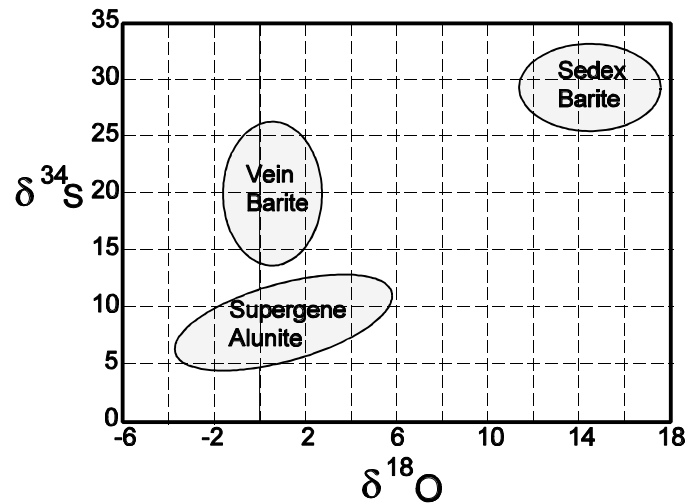


Figure 3. Sulfur and oxygen isotope values for sulfate minerals in CTDs.

Although this third group of sulfates are clearly hydrothermal, in most cases barite veins are later than the bulk of gold mineralization. Commonly, barite forms late vein-filling crystals along with quartz, stibnite, and calcite, typically growing in open fractures.

4 ISOTOPIC COMPOSITION OF HYDROTHERMAL WATER

Oxygen isotope values for the hydrothermal fluid(s) must be inferred (calculated) from measurements made on ore-related alteration phases. The most common alteration phase on which oxygen measurements have been made is quartz (jasperoid). Jasperoid $\delta^{18}\text{O}$ values range widely between deposits and even within deposits (O'Neil and Bailey, 1979; Holland et al., 1988; Hofstra et al., 1988; Arehart, 1996). At Jerritt Canyon, there is a strong correlation between $\delta^{18}\text{O}$ of jasperoid and gold grade (Hofstra et al., 1988), and Holland et al. (1988) documented higher $\delta^{18}\text{O}$ values of jasperoids from ore deposits vs. non-economic prospects. Most investigators have interpreted the most positive oxygen values as being representative of the ore-bearing fluid (or at least the most exchanged fluid). Based on measured $\delta^{18}\text{O}$ values, temperatures between 200 and 250°C, and equilibrium fractionation between quartz and water, the calculated oxygen isotope composition of the fluids responsible for several CTDs ranges from -10 to +10‰.

5 ORIGIN OF HYDROTHERMAL SULFIDES AND SULFATES

There are several potential sources of sulfur for the hydrothermal fluids which created CTDs; these include sedimentary rock-hosted sulfides and/or sulfates; igneous rock-hosted sulfides (no significant sulfates are known

from these rocks in Nevada), and sulfur from potential magmatic fluids. Both sedimentary and igneous rock-hosted sulfides have $\delta^{34}\text{S}$ values (ca. -5 to +9‰) that are significantly lower than the ore sulfides. No simple mechanism is available that can shift these lower values to the higher ones recorded in ore-stage sulfides. Similarly, magmatic sulfur generally has values of $0 \pm 5\%$, and is not easily shifted to higher values. The most reasonable source for hydrothermal sulfur in CTDs of the Great Basin is through reduction of sulfate sulfur (Arehart et al., 1993b).

The ore-generating fluid initially had $\delta^{34}\text{S}$ values near +20‰ (note that gold deposition occurred very early in the paragenesis) but evolved through time to lower values, as

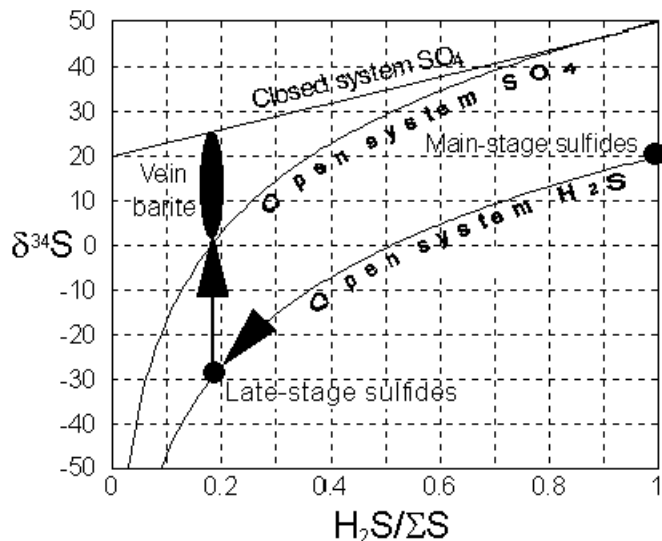


Figure 4. Schematic Rayleigh fractionation of main-stage sulfides to produce late-stage sulfides, followed by oxidation of ore fluids to produce late-stage barite.

recorded in latest-stage marcasite (-30‰). A most reasonable mechanism that explains this shift is Rayleigh fractionation of the initial fluid during ore depositional processes. A hypothetical model of the evolution of hydrothermal sulfur is shown in Figure 4.

If Rayleigh fractionation occurs because of oxidation of H_2S in either open or closed systems, resultant $\delta^{34}\text{S}$ values of sulfate in the fluid also will decrease through time. Depending upon the relative effectiveness of equilibrium vs. non-equilibrium processes, the $\delta^{34}\text{S}$ value of sulfate in solution will fall between these two extremes. Therefore, any sulfate minerals that are deposited will have $\delta^{34}\text{S}$ values between 0 and 25 ‰, if formed from the same or similar solutions which deposited latest-stage sulfide having $\delta^{34}\text{S}$ values of -30 ‰ (Figure 4).

In fact, this process is probably what has been recorded in hydrothermal barite from CTDs. Late-stage barite from the Post/Betze orebody was deposited in the same veins and breccias as light sulfides, and is depleted in ^{34}S relative to early non-vein barite (Arehart et al., 1993b). Although this barite could have formed as a result of quantitative oxidation of the original hydrothermal solutions, this

alternative is considered unlikely because of the significant difference in their positions in the paragenetic sequence. In addition, the available evidence indicates that barite was not stable at this point in the evolution of the hydrothermal system. Barite deposited early in the paragenetic sequence (essentially remobilized sedex barite) has $\delta^{34}\text{S}$ values in excess of 25‰. Similar trends in $\delta^{34}\text{S}$ values of barite are observed at Alligator Ridge (Ilchik, 1990), Gold Quarry, and Rain (Howe et al., 1995).

The late hydrothermal barite also has oxygen isotopic compositions consistent with derivation from late hydrothermal fluids, which were probably closer to unexchanged meteoric water in character than were the main-stage fluids. Early gold-rich fluids were more exchanged with the wallrocks through which they traveled; there is a positive correlation between oxygen isotope values and gold grade in jasperoids (Hofstra et al., 1988; Holland et al., 1988). In contrast, water-rock ratios for the latest stage fluids were most likely very high and therefore more meteoric in character. In addition, many models for CTD ore deposition (Hofstra et al., 1988; Kuehn and Rose, 1995; Arehart, 1996) implicate fluid mixing in these deposits; such late fluid mixture(s) also are likely to have a significant non-exchanged component. Presuming that these late fluids are represented by the lower values of fluid compositions as recorded by jasperoidal quartz (-10 to -5‰), then late barite deposited at 225 °C should have $\delta^{18}\text{O}$ values between -4‰ and +1‰, which is in agreement with measured values (Figure 3).

6 IMPLICATIONS FOR MINERAL EXPLORATION

The presence of two isotopically distinct types of barite could provide important clues to CTD mineralization in the Great Basin. Although in many cases there are textural differences between sedex and hydrothermal barite that allow identification, there are some samples for which the textural criteria may not apply. In these cases, isotopic measurements provide an important discriminator between the two types of barite. Because barite mineralization may extend beyond easily recognizable rock alteration in some CTDs, stable isotopes could be useful in identifying hydrothermal systems that may host gold. Whether this type of relationship holds for CTDs in other parts of the world is as yet undetermined as there are insufficient isotopic data available.

7 SUMMARY

Stable isotopic measurements on sulfates associated with CTDs yield three distinct groups of samples (sedex barite, hydrothermal barite, and supergene alunite and jarosite) that are easily discriminated on the basis of S and O isotopes. Available data are consistent with deposition of sedex barite from Paleozoic ocean waters during primary sedimentation. Reduction of this sedex barite during hydrothermal circulation yielded an ore fluid having $\delta^{34}\text{S}$ values of approximately +20‰ from which primary gold-bearing pyrite was deposited. Late sulfides and vein barite

were deposited from a hydrothermal fluid that had undergone Rayleigh fractionation (sulfides) and late-stage oxidation (barite). Supergene sulfates (alunite, jarosite) formed as the result of weathering of the primary sulfide ores.

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