CARBONATITIC BARITES[1]

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ABSTRACT

Carbonatitic barites differ from those of noncarbonatitic origin in normally containing some Sr and in having a much wider total range of Sr content. Carbonatitic barites are late magmatic (rarely) to hydrothermal species, occurring commonly in late-generation carbonatites that are mineralogically and texturally complex. Ankerite, RE carbonates, fluorite, sulfides and quartz are common associates. Unlike early magmatic carbonatitic calcites and dolomites whose Ba and Sr contents vary regularly with relative age, barites from carbonatites show much greater variability in Sr content both among those of different paragenetic positions and within those of a single paragenetic type, further attesting to their formation by hydrothermal fluids in a declining-temperature environment and a locally variable Ba/Sr ratio in the solutions.

INTRODUCTION

For about six years, personnel of The Mineralogical Laboratory of The University of Michigan have been engaged in a systematic study of carbonatites, their geology and mineralogy. One phase of this work has been a detailed investigation of the composition of some of the more important carbonatitic minerals. The carbonate species were studied first (Quon, 1965; Quon and Heinrich, 1966), followed by the Ba-Sr sulfates. A study of carbonatitic magnetite is nearly complete, and one has been begun on the carbonatitic potash feldspars. This paper presents results on barite-celestite. Quon and Heinrich (1966; Quon, 1965) found a high degree of regularity in the relationship of minor element contents of the various carbonatitic carbonates to (1) the species, (2) the paragenetic position of the carbonate and, (3) in some cases, to the province.

A carbonate-rich rock may be classified as a carbonatite if it can be demonstrated that the carbonate species are of igneous derivation regardless of whether the body was crystallized from an intruded carbonatitic magma or was formed by a replacement between carbonatitic fluid and wall rock. Furthermore, in many complex carbonatites (*i.e.* multistage bodies) there was a gradation from magmatic to hydrothermal conditions, analogous to the formation of complex granitic pegmatites which contain both zonal (magmatic) and replacement (hydrothermal) bodies (Heinrich, 1966; Heinrich and Shappirio, 1966). In complex pegmatites the distinction between the two types of units is often possible, owing to difference in structure, mineralogy and texture. However, in carbon-

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atites, because of the similarities in mineralogy throughout the sequence and the ease with which earlier carbonatitic phases are retextured, a distinction between magmatic and hydrothermal units may be very difficult. One possible aid lies in the appearance of certain diagnostic species; another is the variation in composition of some multigenerational minerals.

The purpose of this study was, therefore, to learn something of the Sr contents of carbonatitic barites in the hope that this might give us an additional tool in determining the origin of the baritic carbonatites.

DEPOSITS STUDIED

Table 1 lists the barites studied and describes briefly their occurrence. All of these occurrences have been studied in the field, and for all of them the barite has been studied in the laboratory by means of thin sections. Not all the barites were suitable for analysis.

TABLE 1. GEOLOGY AND STRONTIUM CONTENT OF CARBONATITIC BARITES

Carbonatite	Wt. % SrO	Occurrence of barite	Reference
Magnet Cove, Ark.	0.7[1] 2.8[2]	Malvern deposit, a 60 ft.-thick replacement body of the Stanley shale, 2.5 mi. ENE of Magnet Cove complex. Quartz, calcite benstonite, pyrite, iron oxides are associates.	Parks and Branner, 1932; Brobst, 1958.
Amethyst, Fremont Co., Colo.	0.5[2] 0.65	Marginal zone 0.5–1 ft. thick essentially monomineralic. Barite is post-ferroan calcite.	Heinrich and Shappirio, 1966.
Goldie, Fremont Co., Colo.	0.14[2]	Irregular nodules with fluorite, cryolite and other rare alumino-fluorides of replacement origin.	Heinrich and Quon, 1963; Heinrich and Anderson, 1965.
Jim No. 6, Fremont Co., Colo.	0.35[2]	Euhedral metacrysts(?) 0.5–1.0 in. across, replacing fine-grained carbonate-hematite rock.	Dahlem, 1965.
Dreamer's Hope, Fremont Co., Colo.	0.1[2]	Grains and aggregates interstitial to dolomite; in breccia zones cutting dolomite.	Dahlem, 1965; Staatz and Conklin, 1966.
Klondike, Fremont Co., Colo.	0.1[2]	Interstitial aggregates in carbonate in composite body of feldspar rock and carbonatite.	Dahlem, 1965.

TABLE 1—*(continued)*

Carbonatite	Wt. % SrO	Occurrence of barite	Reference
Wet Mtns, Custer Co., Colo. Democrat Gulch, Custer Co., Colo.	0.035[1] 0.04[2]	Red to white, generally in coarse cleavable masses; less usually fine-grained to microcrystalline. Banded carbonatite.	Brobst, 1958. Christman *et al*, 1959.
Schindler, Fremont Co., Colo.	0.3[2]	In feldspathic thorite-bearing carbonatite as irregular masses.	Heinrich, MS rept.
Tuttle Ranch, Fremont Co., Colo.	0.06[2]	Irregular lenses and masses. Two generations: red granular and younger white cleavage masses. Quartz, hematitic feldspar, siderite, hematite, Cu sulfides, Th minerals are associated.	Christman *et al.*, 1953.
Haputa Ranch, Fremont Co., Colo.	0.08[2] 0.08	Quartz-barite-siderite-hematite carbonatites with Th minerals. Barite white to green in coarse cleavage masses.	Christman *et al.*, 1953.
Mountain Pass, Calif.	10.7[1] 1.4[2] 15.5 2.25 13.0 0.65 1.8 1.0	Up to 65%, average 20–25% of Sulphide Queen carbonatite as coarse tabular to ovoid grains. Also as fine-grained veinlets ± quartz and bastnaesite along fractures, even cutting coarse barite.	Olson *et al.*, 1954; Brobst, 1958
Rocky Point No. 5, Ravalli Co., Mont.	5.3[2] 4.1 5.1 7.0	Irregular to lensoid bodies up to several feet long concentrated in hangingwall half of carbonatite.	Heinrich and Levinson, 1961
Firesand River, Ontario	0.6[2]	Euhedra in calcite matrix associated with xenocrystic quartz grains and broken crystals.	Heinrich and Vian, 1966
Fen, Norway	0.55[1] 4.1[2]	A subordinate interstitial accessory in both sövite and rauhaugite.	Saether, 1958
Alnö, Sweden	0.03– 0.06[1] 1.4[2]	Late veins with marginal zones of barite (40%) calcite (40%) and fluorite with cores of 96–99% barite, the rest fluorite.	von Eckermann, 1948, 1950.

TABLE 1—(continued)

Carbonatite	Wt. % SrO	Occurrence of barite	Reference
Kaiserstuhl, Germany	0.19[1] 0.22 0.50	Closely associated with later vein carbonatites.	van Wambeke et al, 1964
Tundulu, Nyasaland	0.25[2]		This study.
Sukulu Hills, Uganda		As an accessory species in carbonatite; as barite-rich carbonatite breccia zones.	Davies, 1947, 1956
Ngualla, Tanganyika		Late quartz-calcite veins with barite, fluorite, chalcopyrite and galena cut "intermediate zone" sövite.	Heinrich, 1967
Mbeya (Panda Hill) Tanganyika	0.14[2]	Metacrysts in carbonatite breccia.	This study.
Shawa, Rhodesia		Barite veins cut serpentinized dunite core of the alkalic complex.	Johnson, 1961
Chilwa Island, Malawi		An accessory in ankeritic sövite (inter-mediate age); as a major constituent of late quartz-fluorite veins.	Garson and Smith, 1958
Amba Dongar, India		A major constituent of radioactive barite-fluorite-anhydrite carbonatite (intermediate age).	Sukheswala, Udas and Heinrich, 1965

[1] Sr analysis previously published.

[2] Sr analysis from this investigation.

Nearly all of the field relations as well as textural relations in hand specimens and in thin sections combine to give the following general picture of the paragenesis of carbonatitic barites:

1. They occur commonly and in abundance in mineralogically com-plex carbonatites, particularly those with RE minerals (carbonates and phosphates), fluorite, sulfides and quartz. If several types of carbonatite dikes occur together in a district, the baritic type is the youngest.

2. In mineralogically less complex carbonatites they are associated usually with younger carbonate species, such as ankerite, rather than with early Ba-Sr calcite.

3. In addition to mineral complexity many barite-rich carbonatites

are heterotextural, showing brecciation, fracturing and veining, as well as marked to extreme grain-size and grain-shape variations. Banding, lensoid structures, metacrysts and comb structures may be well developed in such bodies.

From this we conclude that most carbonatitic barites are late magmatic to hydrothermal, probably nearly always the latter. Certainly few, if any, belong to the early magmatic stage.

GEOCHEMISTRY OF Ba AND Sr IN CARBONATITES

Both Ba and Sr are abundant and widespread characteristic accessory elements of carbonatites and, indeed, of alkalic rocks in general. Their distribution in mineral species is as follows (Heinrich, 1966):

1. Minerals containing *both* Ba and Sr in minor to major amounts: calcite, dolomite, ankerite, apatite, barite, pyrochlore (pandaite); (rare species) celestite, burbankite.

2. Minerals containing chiefly Ba in minor to major amounts: biotite, phlogopite, orthoclase, melanite; (rare species) witherite, cordylite, alstonite, benstonite, barylite, baotite.

3. Minerals containing chiefly Ba in minor to trace amounts: nepheline, alkalic amphibole.

4. Minerals containing chiefly Sr in minor to major amounts: (rare species): celestite, strontianite, ancylite, goyazite, svanbergite.

5. Minerals containing chiefly Sr in minor to trace amounts: nepheline, sphene, cancrinite, melilite, alkalic amphibole, plagioclase, thomsonite, eudialyte, perovskite.

Sr substitutes mainly for Ca in both silicates and nonsilicates, whereas Ba proxies primarily for K in silicates and for Ca in apatite and the carbonates and therefore has a more diverse mineral representation. The Sr/Ba ratio in the carbonatites and their associated alkalic rocks of the Alnö complex ranges, *in general*, from 0.01–0.43, with two exceptions. In sövite breccia it is 6.60 and in an apatite-rich dolomitic dike it is 14.0.

Quon and Heinrich (1966; Quon, 1965) found that carbonatitic calcites contain 0.004–0.4 (av. 0.106) percent Ba and 0.22–2.55 (av. 0.85) percent Sr. Sr is greater than Ba by a factor of 6.5 to 55, and similar ratios chacterize dolomites and ankerites of younger paragenetic positions. Thus, although carbonatites as a whole contain more Ba than Sr, much more of the latter is selectively fixed in early carbonate species, thereby enriching the residual carbonatitic fluids still further in Ba. For this reason independent Sr minerals are very rare in carbonatites. Celestite is reported only from the following localities:

Mountain Pass, California (barian)
Nano-Vara, U.S.S.R.

Tamazert, Morocco.

Strontianite also is very rare. Von Eckermann (1948) notes it at Alnö, as minute crystals in a biotite beforsite with accessory barite, chalcedony, apatite and pyrite. At Mountain Pass strontianite cements a breccia of barite-carbonate rock in the Sulphide Queen carbonatite and occurs in a vein in the Birthday No. 6 deposit. It "... appears to be one of the late vein minerals" (Olson *et al* 1954, p. 33). It also is reported as an exsolution product in a gray calcite of the Bearpaw Mountains, Montana (Pecora, 1962). Other localities are Nano-Vara, U.S.S.R.; Mbeya, Tanzania; and Kangankunde, Malawi.

Analytical Technique

Analyses were made with a Phillips X-ray spectrograph operating at 35 kV and 15 mA, at a fixed count of 2×10^5 on Sr Ka 25.1° 2θ.

A working curve was constructed with the data from standard samples made with reagent grade $BaSO_4$ and $SrSO_4$. The $BaSO_4$ was found to contain about 0.2 percent Sr and additional standards were prepared with a purer $BaSO_4$ ($<0.005\%$ Sr) precipitated from a weak solution of $BaCl_2$ by dropwise addition of 4:1 H_2SO_4. Appropriate quantities of $BaSO_4$ and $SrSO_4$ were weighed and wet-blended to make 0.01, 0.10, 1.00, 3.00 and 5.00 percent Sr standards. The working curve was obtained by plotting peak height minus background divided by background against strontium content. Analytical error is ±5 percent of the amount present. Sample purity was checked optically and by X-ray diffraction.

Sr Contents of Barites

Barite and celestite are isostructural, and a complete isomorphous series exists between the two (Grahmann, 1920; Starke, 1964). Several recent studies have concerned themselves with the Sr contents of barites of various origins (Gundlach, 1959; Werner, 1958; Starke, 1964; Brobst, 1958; Tischendorf, 1963; Puchelt and Muller, 1964; Tufar, 1965), but carbonatitic barites have not been investigated systematically. Barites very commonly contain some Sr, but varieties intermediate in the barite-celestite sereis have been reported but rarely. Palache *et al* (1951, p. 411), for example, state "Barite containing more than a few atomic percent of either Sr or Ca is uncommon . . . ." They cite a barite from Clifton, England with SrO 14.7 percent and a barian celestite from Greiner, Switzerland with SrO 28.27 percent; the first analysis was made in 1897, the second in 1886.

The Sr contents of carbonatitic barites are listed in Table 1, those of elected noncarbonatitic barites in Table 2.

Three U. S. districts are represented by a number of samples (Table 2):

	Range	Average
Wet Mountains, Colorado	0.035–0.65	0.2
Ravalli Co., Montana (Rocky Pt. No. 5 deposit)	4.1–7.0	5.4
Mountain Pass, California	0.65–15.5	5.79

TABLE 2. Sr CONTENTS OF SELECTED NON-CARBONATITIC BARITES

I. Previously reported

Locality or areas	Wt. % SrO (number of analyses)	Type of deposit	Reference
Wechselfenster, Austria	2.02, 2.05 (2)	Alpine veins ("lateral secretion" during metamorphism)	Tufar, 1965
Lower East Alpine Older crystalline, Austria	1.82, 1.95 (2)	Alpine veins	Tufar, 1965
Mesozoic, Semmering, Austria	0.38–2.68 (13) av. 1.62	Alpine veins	Tufar, 1965
Middle East Alpine crystalline, Austria	0.64–2.34 (3)	Alpine veins	Tufar, 1965
Gurktal Deche, Austria	0.85, 1.09 (2)	Alpine veins	Tufar, 1965
Paleozoic of Graz, Austria	0.30–1.86 (13) av. 0.57	Hydrothermal (Magmatic) (?)	Tufar, 1965
Graywacke zone, Austria	0.68–5.62 (6) av. 3.68	Alpine veins (?)	Tufar, 1965
Northern Calcareous Alps, Austria	3.20 (1)	Alpine veins (?)	Tufar, 1965
Upper Permian, Meggen, Germany	4.8 av. of 75 cm-thick layer 0.58	Syngenitic intra-formational sedimentary	Puchelt and Müller, 1964 Starke, 1964
Erzgebirge, Germany	1.19 (1494) (arith. mean)	Hydrothermal veins	Starke, 1964
Thuringer Forest, Germany	2.36 (177)		Starke, 1964
Harz Mtns., Germany	1.47 (88)		Starke, 1964
Black Forest, Germany	1.60 (56)		Starke, 1964
Ruhr Region, Germany	0.39 (90)		Starke, 1964

TABLE 2—(*continued*)

II. New analyses

Locality or areas	Wt. % SrO (number of analyses)	Type of deposit	Reference
Cheshire, Conn.	0.35		This study
Marquette Co., Mich.	3.3		This study
Pugh Quarry, Ohio	3.0		This study
Mineral Point, Wisc.	0.5	Hydrothermal	This study
Norman, Okla.	1.1	Sedimentary	This study
Gillman, Colo.	0.5	Hydrothermal	This study
Port Arthur Dist., Ontario	1.4		This study
Burgess Twsp., Larnark Co., Ont.	4.0		This study
Hull Twsp., Ottawa Co., Que.	4.2		This study
Frizington, England	0.1		This study
Gado, Rhodesia	1.1		This study

In the first two districts the barite is, in each, of a single paragenetic type, whereas at Mountain Pass at least three and possibly more generations of barite are represented. This suggests that within a single deposit the Sr content may vary strongly with paragenetic position, but also that within a single deposit or district one generation of barite may have highly variable Sr contents.

In comparison with hydrothermal, "lateral secretion" and sedimentary barites, carbonatitic barites display a much greater range in strontium content. Although many carbonatitic representatives have SrO contents (0.0%) well below the usual minimum for non-carbonatitic barites (0.%), they also may exceed by 2–3 (0.%) the maximum of their non-carbonatitic counterparts (.%).

DISCUSSION

Bolduan *et al* (1961) and Starke (1964) have demonstrated that the Sr contents of barites are a function of, first of all, the Ba and Sr con-

centrations of the solutions and secondly of the temperature of deposition. At a fixed Ba/Sr ratio the Sr content of the precipitated barite increases with increasing temeperature and similarly, at a fixed temperature, the Sr content increases with a decrease in Ba (or increase in Sr) content of the solutions (see Starke, 1964, Fig. 45, p. 60; Tischendorf, 1963).

In the sequence of formation of multiphase carbonatites with Ba-Sr sulfates, it has been determined that: (1) Barites are deposited relatively late, largely after magmatic calcite and dolomite both rich in Ba and Sr and in company with ankerite, siderite, late "pure" calcite and quartz and, (2) Barite is followed, in a few deposits, by deposition of celestite and strontianite.

Thus during the geochemical history of Ba and Sr in carbonatites the following declining temperature stages may be recognized:

1. Removal chiefly of small amounts of Ba in such early silicates as potash feldspar and the micas.

2. Removal of large amounts of both Ba and Sr in carbonate species, with much more Sr (6X to 55X) selectively abstracted. With declining temperatures the capaicity of the carbonates for isomorphous Ba and Sr decreases.

3. Up to this point the Ba/Sr ratio has increased markedly. Now barite containing variable and generally trace to minor amounts of Sr is deposited.

4. This reverses the Ba/Sr ratio, and a few deposits in which the initial Sr content was exceptionally high, celestite low in Ba may be deposited as the youngest sulfate.

The highly variable Sr contents of carbonatitic barites is in marked contrast to the regularity in the Ba-Sr contents of the older magmatic carbonate species. This affords additional support to the thesis that most carbonatitic barites are hydrothermal rather than magmatic in origin, and were formed in an environment in which equilibrium probably prevailed only within a restricted section of the deposit and over a markedly curtailed period of time.

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