HYDROTHERMAL REE MINERALIZATION IN THE AMBA DONGAR CARBONATITE COMPLEX, GUJARAT, INDIA

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Abstract

We have studied two groups of overprinted (altered) carbonatites at Amba Dongar, India: weakly overprinted and strongly overprinted. The mineral association of quartz, florencite-(Ce), strontianite, barite and sparse bastnäsite – parisite – synchysite typical of weakly overprinted carbonatites shows evidence of a high activity of OH^-, $(SO_4)^{2-}$, Al and Si in the fluid. Later, the fluid was characterized by an increased activity of F^-, $(SO_4)^{2-}$, REE and Si, reflected in the association of barite, fluorite, quartz and REE fluorcarbonates typical of strongly overprinted carbonatites. Hydrothermal minerals at the Amba Dongar were formed by re-equilibration and recrystallization of the primary minerals in the presence of OH^-, $(SO_4)^{2-}$, F^-, REE, Al and Si carried in solution by the hydrothermal fluid.

Keywords: hydrothermal mineralization, re-equilibration, recrystallization, carbonatites, REE fluorcarbonates, florencite-(Ce), Amba Dongar, India.

Sommaire

Nous avons étudié deux groupes de carbonatites remaniées et altérées à Amba Dongar, en Inde, qui montrent une surimposition légère ou une surimposition plus intense des assemblages primaires. L'association de quartz, florencite-(Ce), strontianite, barite et bastnäsite – parisite – synchysite (fluorcarbonates accessoires) typique des carbonatites légèrement remaniées témoignent d'une recristallisation en présence d'une phase fluide à activité élevée de OH^-, $(SO_4)^{2-}$, Al et Si. Par la suite, la phase fluide est devenue enrichie en F^-, $(SO_4)^{2-}$, terres rares et Si, ce qui a produit un assemblage de barite, fluorite, quartz et fluorcarbonates de terres rares typiques des carbonatites plus fortement remaniées. Les minéraux hydrothermaux à Amba Dongar se sont formés par ré-équilibrage et recristallisation des minéraux primaires en présence de OH^-, $(SO_4)^{2-}$, F^-, terres rares, Al et Si portés en solution par la phase fluide hydrothermale.

(Traduit par la Rédaction)

Mots-clés: minéralisation hydrothermale, ré-équilibrage, recristallisation, carbonatites, fluorcarbonates de terres rares, florencite-(Ce), Amba Dongar, Inde.

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INTRODUCTION

Field and textural observations of carbonatites indicate that mineralization in many cases is hydrothermal (Mariano 1989). Several examples of hydrothermal REE mineralization in carbonatites are known, including occurrences at Tundulu and Kangankunde, Malawi (Ngwenya 1994, Wall & Mariano 1996), Barra do Itapirapuã, Brazil (Andrade *et al.* 1999, Ruberti *et al.* 2002, 2008) and Khibina, Russia (Zaitsev *et al.* 1998). It is commonly unclear whether the REE minerals precipitated directly from hydrothermal solutions or formed during the re-equilibration or replacement of the primary (early-crystallized) minerals in the carbonatites.

In this paper, we focus on hydrothermally altered (overprinted) sövite at Amba Dongar, and also particularly on the mineralogy of the hydrothermally deposited phases. This carbonatite complex provides comparisons of primary sövite and the hydrothermally altered (overprinted) carbonatite that contains highly variable proportions of secondary minerals, such as quartz, fluorite, rare-earth fluorcarbonates [bastnäsite-(Ce), synchysite-(Ce) and parisite-(Ce)], florencite-(Ce), barite, strontianite and columbite. In addition to our presentation of new data on the primary and hydrothermal minerals at Amba Dongar, we speculate on the behavior of various elements of interest during hydrothermal activity.

GENERAL GEOLOGY OF THE AMBA DONGAR CARBONATITE COMPLEX

The general geology, geochemistry and mineralogy of the Amba Dongar complex have been described in numerous publications (*e.g.,* Viladkar & Dulski 1986, Viladkar & Wimmenauer 1992, Simonetti *et al.* 1995, Viladkar 1996).

The Amba Dongar carbonatite–nephelinite complex is a subvolcanic diatreme. It consists of a sövite ring dike that has an inner rim of carbonatite breccia (Fig. 1). The diatreme is surrounded by a fenite zone (as much as 300 m wide) in the surrounding Bagh sandstone. There is vertical zonation of fenitization, with sodic fenites at deeper levels and potassic fenites at the surface (Viladkar 1996). Sövite constitutes most of the total volume of exposed carbonatite, and appears to have been emplaced in several phases leading to the formation of the ring structure. This multiphase emplacement is indicated by the presence of xenoliths of older sövite in younger dikes or plugs of sövite. The sövite also forms small, circular to oblong plugs in the surrounding sandstone. The sövite shows a large variation in grain size and color; a coarse-grained variety is exposed in the inner part of the ring, whereas successive phases toward the outer margin are finer-grained. The sövite is extremely fine-grained at the sövite–sandstone contact.

On the basis of field observations, two phases of alvikite dikes have been recognized. Phase I is coeval with the main intrusion of sövite, and cross-cuts the sandstone, carbonatite breccia and pre-carbonatite basalt, whereas phase-II dikes were emplaced later. Ankeritic carbonatite, which is younger than the calcio-carbonatite, invades sövite at several outcrops in the form of dikes and plugs of varying size. The ankeritic carbonatite occurs in three distinct phases. The first phase consists of thin dikes intruding sövite and fenite; the second phase forms large plugs within the sövite ring-dike, whereas the last phase is characterized by high radioactivity owing to the high abundances of cerite, monazite-(Ce), and thorite (Viladkar 1996). Veins of sideritic carbonatite, which are found in ankeritic carbonatites, also contain cerite, monazite and thorite. Locally, the total REE content is well above 15 wt.% (Viladkar 1996). The carbonatite activity appears to have ended with fluorite mineralization.

Geochronological data have been reported by Deans & Powell (1968), Deans *et al.* (1973), Petibon (1994), Ray & Pande (1999) and Ray *et al.* (2000). However, the absolute age of emplacement of carbonatite – alkaline rocks in Chhota Udaipur Province is still controversial. Deans & Powell (1968) used the $^{40}Ar/^{39}Ar$ method to establish an age of 37.5 ± 2.5 Ma on pyroxenes from nephelinite. Deans *et al.* (1973) later obtained an age of 62 ± 2 Ma and 76 ± 2 Ma (K/Ar) on the K-feldspar from the potassic fenites. Later, Viladkar & Gruau obtained a Rb/Sr age of 41.7 ± 3.2 Ma for Amba Dongar using phlogopite from the phlogopite-bearing fenite (Viladkar & Gruau, unpubl. data, and Petibon 1994). The 41.7 ± 3.2 Ma age is definitely a reset age, and the later intrusion of part of central basalt core and number of dolerite dikes may be responsible for this event (S.G. Viladkar, in prep.). Ray & Pande (1999) and Ray *et al.* (2000) reported ages ranging from 64.7 ± 0.5 Ma to 65.5 ± 0.8 Ma.

The chronological evolution of the complex, according to Viladkar (1996), can be summarized as follows: 1) intrusion of nephelinites and phonolites around the periphery of the ring structure, 2) emplacement of a large plug of carbonatite breccia, 3) intrusion of the sövite ring dyke and alvikites, and fenitization of the host sandstones, 4) emplacement of an ankeritic carbonatite plug within the sövite, followed by intrusion of thin veins of sideritic carbonatite in the ankeritic carbonatite, and 5) hydrothermal activity resulting in silicification and REE enrichment of some sövites and fluorite mineralization (11.6 million tonnes averaging 30% CaF$_2$).

The sövite samples used in this study were collected near the ankeritic carbonatite. A sample of sövite with cumulate apatite-(CaF) is from the northern part of the ring dike. This outcrop is cut by a nearby ankeritic carbonatite plug. The remaining samples are from the
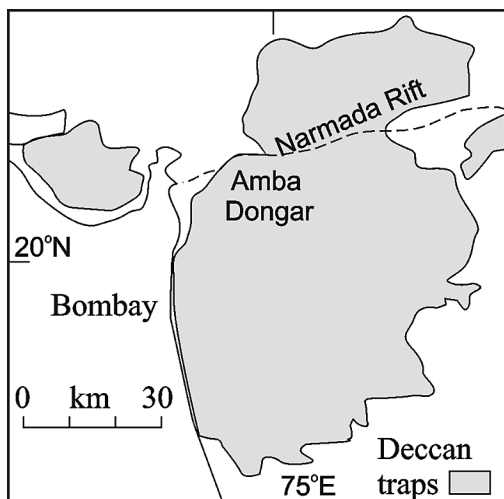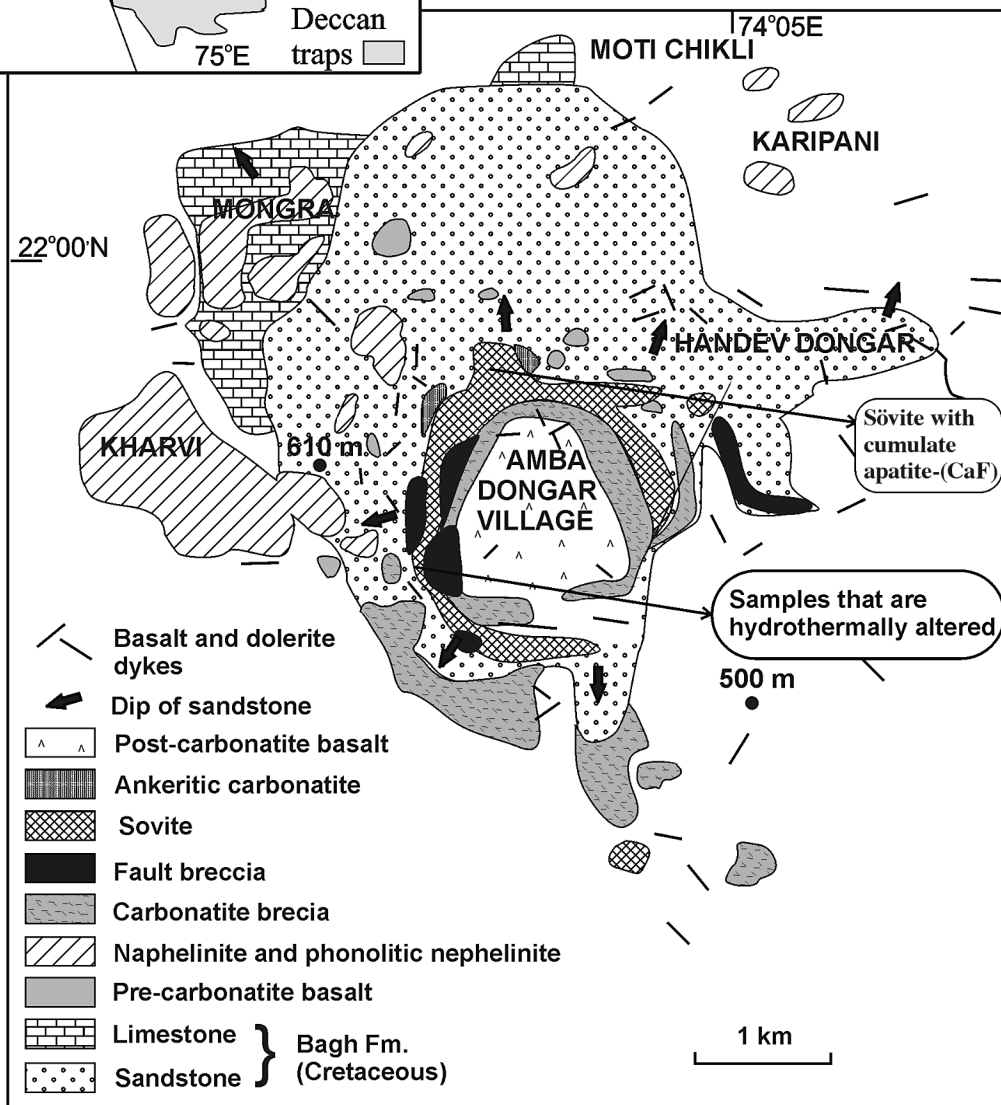
FIG. 1. Geological sketch of the Amba Dongar carbonatite complex. The locations indicate areas sampled to provide cumulate apatite-bearing sövite and hydrothermally altered sövites.

southern part of the sövite ring-dike along the contact with a large plug of ankeritic carbonatite.

ANALYTICAL METHODS

The minerals were analyzed using an electron microprobe (MAR–3 WDS microprobe system, Geological Institute, SD RAS, Russia), using natural apatite-(CaF) as a standard for P and Ca, albite for Na, wollastonite for Si, BaSO$_4$ for S, celestine for Sr, Cs$_2$ReCl$_6$ for Cl, synthetic rare-earth glass for La, Ce, Pr and Nd, almandine for Mn, diopside for Mg, Eg–79 for Fe, BaF$_2$ for F, rutile for Ti, and corundum for Al. The accelerating voltage was 20 kV, the beam current was 40 nA, and the beam size was 3–4 μm. The peak and background count-times were 20 s. A LEO–1430 scanning electron microscope with an IncaEnergy–300 energy-dispersive system (SEM–EDS) operated at 20 kV and 0.5 nA also was used.

Structural formulae of the apatite-(CaF) were calculated on basis of 13 atoms of oxygen. The proportion of the OH ion was calculated by normalization of the results to the ideal stoichiometry in the columns of anions (F + Cl + OH = 2 $apfu$). The structural formulae of florencite were calculated on the basis of 11 atoms of oxygen with XO_4 normalized to 2 $apfu$. The structural formulae for bastnäsite, synchysite and parisite were calculated on the basis of 4, 7 and 11 anions, respectively.

PETROGRAPHY OF THE CARBONATITES SAMPLES

The petrography and mineralogy of all rocks of the Amba Dongar complex were described in detail by Viladkar (1996). A summary of petrographic features

of sövites is given below. The terms "primary" and "secondary" in the following text are used to distinguish minerals formed at a magmatic stage from those formed by late- to postmagmatic hydrothermal fluids, respectively. A short description of fenites also is given below, as this information is important for a better understanding of the composition of the fluid phase.

The primary minerals in the sövite samples are calcite, apatite-(CaF), aegirine-augite, aegirine, magnetite, pyrochlore, phlogopite, zircon and niobian zirconolite. Calcite is the most abundant mineral (70–80%). Apatite-(CaF), magnetite, and pyrochlore are ubiquitous, and mostly form segregations from 2 to >10 vol.%. The crystals show many habits. They occur as rounded to oblong clusters (Fig. 2) or form perfect hexagonal prismatic crystals, some with pyramidal faces. Acicular prisms are not uncommon. In all types of carbonatite, pyrochlore occurs in variable amounts, but, in general, it is more abundant in sövite than in ankeritic carbonatite. Pyrochlore varies from pale golden-yellow, translucent to dark brown, almost opaque crystals. Zoning is very common in large grains; they consist of a dark core and a light margin. Reverse zoning also is not uncommon. Mica (mainly phlogopite) in coarse-grained sövite occurs as euhedral to subhedral crystals. Inclusions of apatite-(CaF), pyrochlore, and zircon are common in the mica. Occasionally, the phlogopite forms thin bands in association with abundant pyrochlore and niobian zirconolite, which show a parallel orientation. Viladkar (2000) showed that the compositional variation in mica, from phlogopite to tetraferriphlogopite, is related to the differentiation history of the carbonatitic magma at Amba Dongar.

Quartz, fluorite, strontianite, barite, florencite-(Ce), bastnäsite-(Ce), parisite-(Ce), synchysite-(Ce), mona-
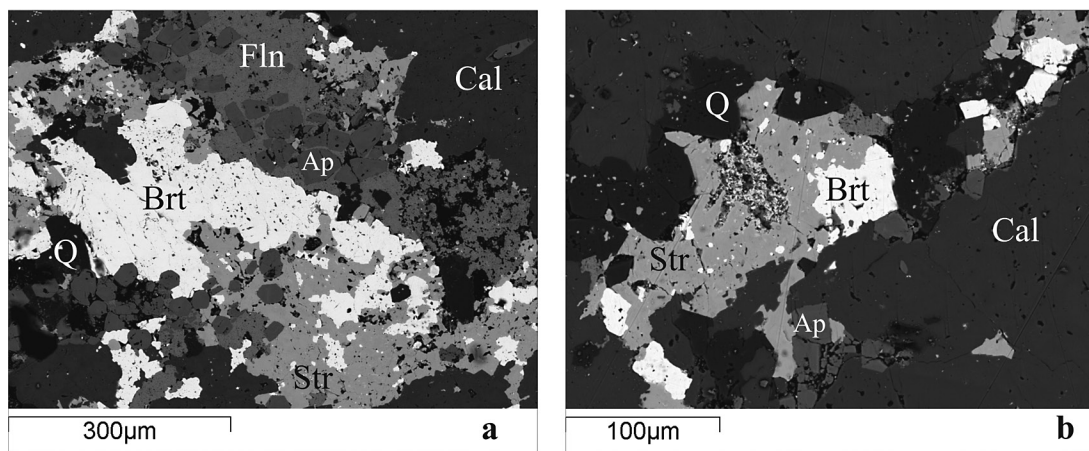

FIG. 2.   Association of barite (Brt), strontianite (Str), florencite-(Ce) (Fln) and quartz (Q) in weakly overprinted carbonatites. Other symbols: Cal: calcite, Ap: apatite-(CaF). BSE image.

zite-(Ce), columbite and ankerite were formed during the late stage. Chalcopyrite, dickite, galena, pyrite and, in some cases, fluorite are coated with quartz, iron oxide, and Mn oxides (Deans *et al.* 1972, Viladkar & Wimmenauer 1992). On the basis of the assemblages of secondary minerals, two groups of overprinted (altered) sövites have been identified: (I) weakly overprinted, (II) strongly overprinted.

Weakly overprinted carbonatites are microscopically similar to unaltered samples, but contain up to 2–3 vol.% of dispersed (and microveinlets) secondary florencite-(Ce), barite, strontianite, and quartz (Fig. 2). They also contain rare grains of rare-earth fluorcarbonates. Florencite ideally occurs only in weakly overprinted carbonatites; it replaces the apatite-(CaF) along cracks and at grain edges (Fig. 3). Strontianite forms

microveinlets and disseminations within calcite and microveinlets with barite, quartz, and florencite-(Ce) (Fig. 2). Bastnäsite-(Ce), parisite-(Ce) and synchysite-(Ce) are rare minerals in the weakly overprinted carbonatites. They form assemblages of acicular crystals and isometric grains in strontianite (Fig. 4a).

Strongly hydrothermally overprinted carbonatites have the highest abundances (up to 10–15 %) of quartz, fluorite and barite (Fig. 5). Ankerite occurs only in strongly overprinted carbonatites and is associated with quartz, fluorite and barite. The mineral makes up the borders of fluorite–quartz microveinlets (Fig. 5). The edges of the apatite grains were recrystallized and re-equilibrated during the intense hydrothermal overprint. Bastnäsite-(Ce) has not been detected in the strongly overprinted carbonatites. Synchysite-(Ce)

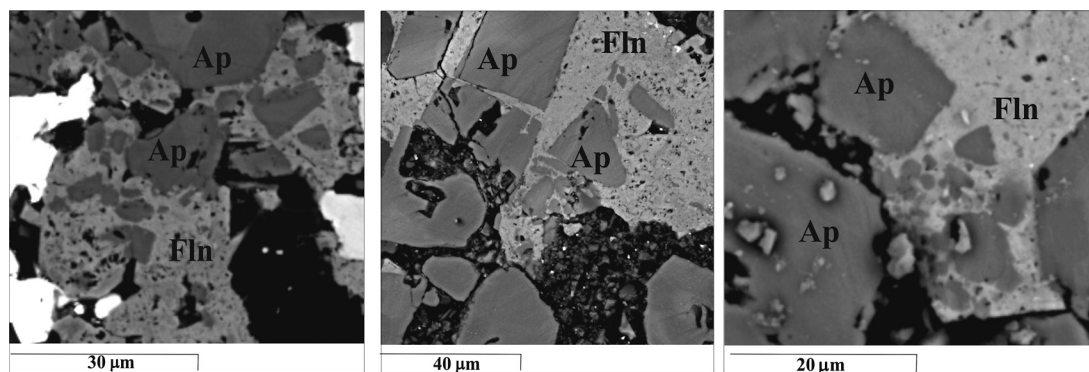FIG. 3. Replacement of apatite-(CaF) by florencite-(Ce). BSE images.
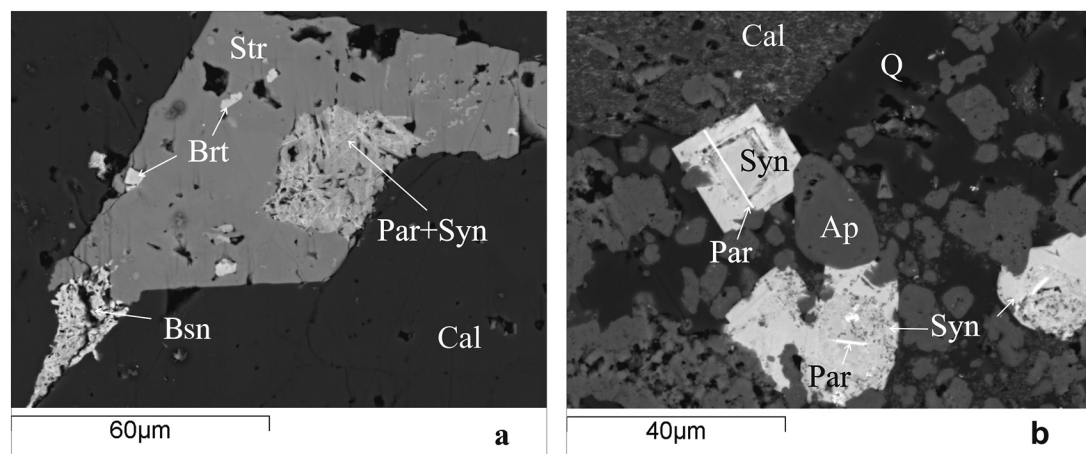

FIG. 4. a. Inclusions of REE fluorcarbonates [Bsn: bastnäsite-(Ce), Par+Syn: assemblages of parisite-(Ce) and synchysite-(Ce) grains] in strontianite, in weakly overprinted carbonatites. b. Crystals and grains of synchysite-(Ce) in strongly overprinted carbonatites. Note that parisite-(Ce) cuts the synchysite crystal. BSE images.

forms crystals and grains here (Fig. 4b), associated with quartz, barite and ankerite. Parisite-(Ce) cuts the synchysite crystals and grains.

The sodic fenites are composed of pyroxene, K-rich feldspar, quartz and albite. The pyroxene is aegirine-augite and occurs as prismatic grains. K-feldspar, quartz and albite fill the interstices. Minor minerals are pectolite, calcite, fluorite, rinkite with apatite-(CaF) and barite, pyrochlore, titanite, rutile, bastnäsite, parisite and synchysite as accessory minerals. The potassic fenites consist of K-feldspar (up to 80–90%), quartz and albite. Apatite-(CaF), pyroxene and rutile are minor minerals. The pyroxene is aegirine-augite, as in the sodic fenites, and forms idiomorphic grains and solid inclusions in K-feldspar. Zircon, monazite, florencite and barite are accessory minerals.
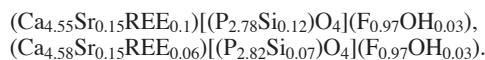
The latest hydrothermal fluorite mineralization occurs as widespread disseminations in brecciated host-rocks along the contact between sövite and country rock, but is mainly in the sövites. It also occurs in vugs of varying dimensions (a few centimeters to 0.7 meters), and forms the veins which at places are more than three meters wide. Vugs are commonly lined by aggregates of fluorite cubes set in a fine-grained matrix of fluorite and quartz. Many zones of mineralization are brecciated and carry fragments of host rock.

COMPOSITION OF THE MINERALS

Calcite contains strontium (up to 0.98 wt.% SrO), manganese (up to 0.60 wt.% MnO), iron (up to 0.35 wt.% of FeO) and, in some cases, magnesium (up to 0.25 wt.% MgO). Representative compositions of calcite are given in Table 1. In the weakly and strongly overprinted carbonatites, calcite is corroded. It contains no strontium (below detection limit; Table 1), probably as result of re-equilibration and recrystallization of the calcite in presence of hydrothermal fluids. The liberated strontium from calcite has been recycled to form strontianite. On the other hand, the calcite from fenites is enriched in strontium (up to 3.55 wt.% SrO) and manganese (to 3.84% MnO) (Table 1).

The apatite-group mineral is apatite-(CaF) containing the maximum F content allowable in the structure (up to 3.86 wt.%); compositions are typical of carbonatites, with an average of 3.30 wt.% SrO. There are varying amounts of SiO_2 and REE (Table 2), from 0.51 to 1.35 wt.% SiO_2 and from 1.09 to 3.15% REE_2O_3. A positive correlation between REE and SiO_2 is shown graphically in Figure 6. The REE–Si correlation is according to the britholite solid-solution (*e.g.,* Pan & Fleet 2002), for example:

$$(Ca_{4.55}Sr_{0.15}REE_{0.1})[(P_{2.78}Si_{0.12})O_4](F_{0.97}OH_{0.03}),$$
$$(Ca_{4.58}Sr_{0.15}REE_{0.06})[(P_{2.82}Si_{0.07})O_4](F_{0.97}OH_{0.03}).$$

The proportion of the britholite component in the apatite-(CaF) attains 9%. The light rare-earth elements in apatite-(CaF) are predominant, with HREE below detection. The average Ce:La ratio is 1.5–2. The content of SiO_2 in recrystallized apatite-(CaF) is below the detection limit. The amount of strontium is lower than in the primary apatite, and, in individual samples, the light REE attain 0.86 wt.% oxide (Table 2). In these cases, there is also about 0.3 wt.% Na_2O, indicating the usual coupled (REE + Na) for 2Ca substitution. The REE in apatite-(CaF) are incorporated according to the belovite solid-solution (*e.g.,* Pan & Fleet 2002), in
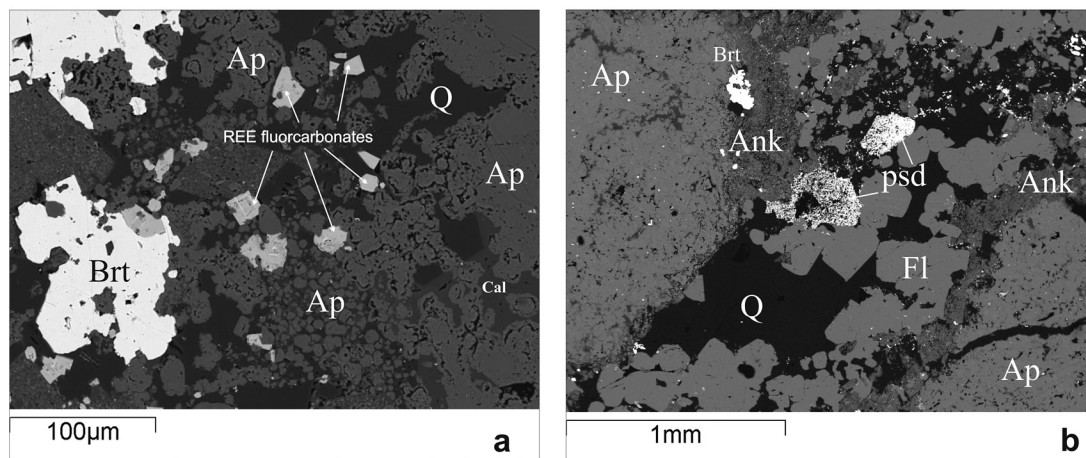

FIG. 5.   a. Recrystallization and re-equilibration of apatite-(CaF) in strongly overprinted carbonatites. REE fluorcarbonates [parisite-(Ce) and synchysite-(Ce)] are light grey. b. Ankerite borders (Ank) of fluorite (Fl) – quartz microveinlets with columbite–quartz assemblages (psd) after pyrochlore (?). BSE images.

TABLE 1. REPRESENTATIVE COMPOSITIONS OF CARBONATE MINERALS
FROM SÖVITES AND FENITES AT AMBA DONGAR

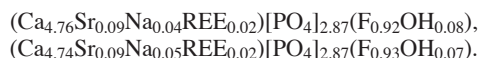|	primary calcite			recrystallized calcite			secondary ankerite			strontianite			calcite from fenite		
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
CaO wt%	54.07	54.56	53.85	55.09	55.19	55.15	31.02	32.8	32.35	2.88	3.82	4.87	53.43	46.3	48.57
MgO	0.18	0.11	0.09	0.11	0.13	0.13	1.27	1.26	1.14	n.d.	n.d.	n.d.	n.d.	n.d.	0.85
MnO	0.6	0.4	0.51	0.43	0.4	0.08	2.93	2.19	2.78	n.d.	n.d.	n.d.	1.16	3.84	1.16
FeO	0.35	0.32	0.32	0.36	0.46	0.43	21.42	21.43	23.12	n.d.	n.d.	n.d.	n.d.	0.68	1.88
SrO	0.63	0.33	0.54	n.d.	n.d.	n.d.	0.54	0.36	0.34	59.23	60.7	60.45	0.62	3.55	2.12
Total	55.8	55.7	55.3	56.0	56.2	55.8	57.2	58.0	59.7	62.1	64.5	65.3	55.21	54.37	54.6
$MgCO_3$ %	0.4	0.2	0.2	0.2	0.3	0.3	2.7	2.6	2.4						
$CaCO_3$	96.5	97.4	96.1	98.3	98.5	98.4	55.4	58.5	57.7	5.7	7.3	9.3	95.4	86.7	90.1
$MnCO_3$	1.0	0.7	0.8	0.7	0.7	0.1	4.8	3.6	4.5					1.9	6.4	1.9
$FeCO_3$	0.6	0.5	0.5	0.6	0.7	0.7	34.5	34.6	37.3					1.1	3.1
$SrCO_3$	0.9	0.5	0.8				0.8	0.5	0.5	94.0	92.0	89.8	0.9	5.3	3.1
Total	99.3	99.2	98.4	99.8	100.2	99.5	98.1	99.8	102.4	99.7	99.3	99.1	98.2	99.5	98.2

n.d.: not detected (below detection limit). Compositions are recast in terms of mol.% of the end members.

TABLE 2. REPRESENTATIVE COMPOSITIONS OF APATITE-(CaF)
FROM SÖVITES AND FENITES AT AMBA DONGAR

	Sövite						Fenite		
	Primary Fap			Recrystallized Fap					
	1	2	3	4	5	6	7	8	9
SiO_2 wt%	0.51	1.35	0.88	n.d.	n.d.	n.d.	0.44	n.d.	n.d.
CaO	51.2	50.02	50.91	54.26	53.34	53.25	53.29	55.17	53.69
Na_2O	n.d.	n.d.	n.d.	0.24	0.28	0.29	n.d.	n.d.	0.6
P_2O_5	40.11	38.73	39.68	41.45	40.88	41.42	42.44	44.19	41.87
SrO	3.17	3.06	3.01	1.81	1.93	1.91	1.98	n.d.	0.7
La_2O_3	0.66	1.08	0.61	0.15	0.25	0.19	n.d.	n.d.	n.d.
Ce_2O_3	1.03	2.11	1.29	0.38	0.53	0.56	n.d.	n.d.	n.d.
F	3.32	3.63	3.66	3.56	3.54	3.07	3.47	3.43	4.32
Total	100.0	101.9	99.9	101.8	100.7	100.7	101.6	102.8	101.2
$O=F_2$	1.39	1.53	1.54	1.5	1.49	1.29	1.45	1.44	1.81
Si *apfu*	0.043	0.115	0.074				0.036		
Ca	4.63	4.546	4.585	4.762	4.742	4.748	4.641	4.7	4.651
Na				0.04	0.05	0.05			0.094
P	2.866	2.781	2.824	2.875	2.871	2.918	2.921	2.975	2.866
Sr	0.155	0.150	0.147	0.086	0.093	0.092	0.093		0.033
La	0.021	0.034	0.019	0.005	0.008	0.006			
Ce	0.032	0.066	0.04	0.011	0.016	0.017			
F	0.886	0.974	0.973	0.922	0.929	0.808	0.892	0.863	1.105
OH	0.114	0.026	0.027	0.078	0.071	0.192	0.108	0.137	

n.d.: not detected. The proportion of OH was calculated by difference. Symbol: Fap: apatite-(CaF).

$$(Ca_{4.76}Sr_{0.09}Na_{0.04}REE_{0.02})[PO_4]_{2.87}(F_{0.92}OH_{0.08}),$$
$$(Ca_{4.74}Sr_{0.09}Na_{0.05}REE_{0.02})[PO_4]_{2.87}(F_{0.93}OH_{0.07}).$$

A similar compositional evolution of apatite has been described by Toledo *et al.* (2004) in the Catalão I alkaline–carbonatitic complex, Goias, Brazil, where silica, strontium and rare-earth elements were found to decrease from igneous to hydrothermal apatite.

The apatite-(CaF) from fenites contains strontium, in some cases, silica and sodium (see Table 2).

Florencite-(*Ce*) is enriched in strontium (up to 5.43 wt.% SrO). Minor amounts of calcium, iron and SO_3 are found in the mineral (Table 3). In addition, silica was detected in many cases, up to 1.95 wt.%. Average Ce/La, Ce/Nd and La/Nd values are 1.07, 7.56 and 7.17, respectively. Calcium and Sr substitute for the REE, causing an inverse relation between Ca + Sr and REE, as illustrated for example by:

$$(Sr_{0.27}Ca_{0.04}REE_{0.74})(Al_{2.93}Fe_{0.04})(P_{1.9}S_{0.1}O_4)(OH)_6.$$
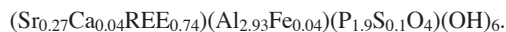
Florencite-(Ce) occurs rarely in carbonatites, as at Kangankunde and Tundulu, Mbeya (McKie 1962), Wigu, Tanzania (McKie 1962), East Sayan, Russia (Somina & Bulakh 1966), Tomtor, Russia (Entin *et al.* 1991), Mt. Weld, Australia (Lottermoser 1990). In all cases, the mineral crystallized during late-stage processes or under supergene conditions. Crystals of florencite from fenites are zoned; the core is richer in REE than the rim. The mineral is enriched in strontium similar to florencite from sövite (Table 3).

contrast to the case of fresh samples, where the REE–Si correlation is according to the britholite solid-solution. A positive correlative relationship between REE and Na is confirmed, for example:
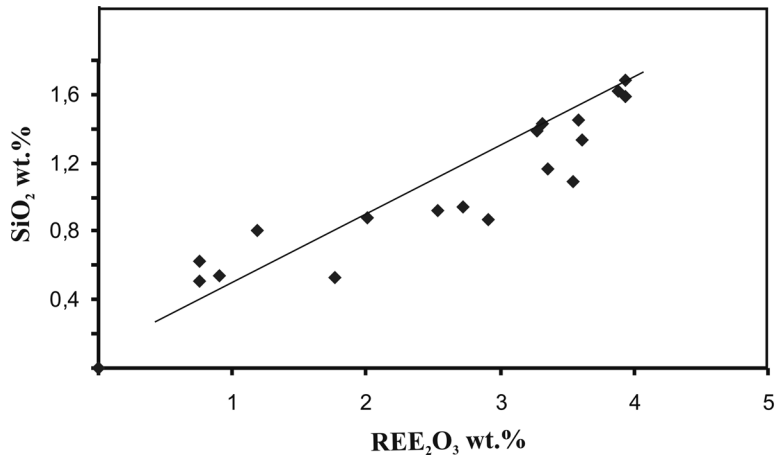
Fig. 6. REE_2O_3–SiO_2 correlation in apatite-(CaF).

TABLE 3. REPRESENTATIVE COMPOSITIONS OF FLORENCITE-(Ce)
FROM SÖVITES AND FENITES AT AMBA DONGAR

	Sövite					Fenite, rim			Fenite, core		
	1	2	3	4	5	6	7	8	9	10	11
FeO wt%	0.85	2.31	0.56	0.51	1.53	2.03	0.51	0.39	4.49	n.d.	0.49
SiO_2	n.d.	1.95	n.d.	n.d.	0.75	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Al_2O_3	29.67	27.86	28.88	29.57	29.11	31.54	29.53	30.67	28.2	29.74	29.77
CaO	0.52	0.71	0.41	0.79	0.81	n.d.	0.42	0.29	n.d.	0.23	n.d.
BaO	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.5	n.d.	1.55	1.41	1.0
P_2O_5	27.62	25.98	26.12	27.81	26.57	28.21	26.08	26.84	26.84	28.56	28.12
SrO	5.45	4.57	5.43	5.47	4.67	13.36	12.64	11.77	5.68	6.14	4.83
La_2O_3	10.56	10.56	8.33	11.93	11.53	3.91	3.87	5.31	7.29	7.01	10.02
Ce_2O_3	11.49	10.13	12.32	12.21	12.17	5.89	4.94	7.04	10.62	10.85	11.9
Pr_2O_3	0.86	n.d.	1.09	n.d.	n.d.	n.d.	n.d.	0.8	1.14	1.32	1.22
Nd_2O_3	1.5	1.11	1.96	1.53	1.02	1.32	1.16	1.48	2.34	2.91	2.31
SO_3	1.09	1.08	1.48	1.17	1.23	4.71	3.91	4.69	n.d.	n.d.	n.d.
Total	89.7	86.3	86.6	90.9	89.4	90.9	84.6	89.3	88.2	88.2	89.7
Si *apfu*		0.17			0.06						
Al	2.89	2.88	2.93	2.85	2.93	2.71	2.78	2.75	2.93	2.9	2.95
Fe	0.06	0.17	0.04	0.03	0.11	0.12	0.03	0.02	0.33		0.03
Ca	0.05	0.07	0.04	0.07	0.07		0.04	0.02		0.02	
Ba							0.05		0.05	0.05	0.03
P	1.93	1.93	1.90	1.93	1.92	1.74	1.77	1.73	2.00	2.0	2.0
S	0.07	0.07	0.10	0.07	0.08	0.26	0.23	0.27			
Sr	0.26	0.23	0.27	0.26	0.23	0.57	0.59	0.52	0.29	0.29	0.24
La	0.32	0.34	0.26	0.36	0.36	0.11	0.11	0.15	0.24	0.21	0.31
Ce	0.35	0.33	0.39	0.37	0.38	0.16	0.14	0.20	0.34	0.33	0.37
Pr	0.03		0.03					0.02	0.04	0.04	0.04
Nd	0.04	0.03	0.06	0.04	0.03	0.03	0.03	0.04	0.07	0.09	0.07

Strontianite contains an average of 3.61 wt.% CaO (Table 1).

Ankerite contains minor Mg, Mn and Sr (Table 1).

Parisite-(Ce) and *synchysite-(Ce)* from weakly overprinted sövites are enriched in Th and Y (Table 4).

The light rare-earth elements are predominant among REE. The Ce/La, Ce/Nd and La/Nd values are similar for the bastnäsite-(Ce), parisite-(Ce) and synchysite-(Ce). In *bastnäsite-(Ce)*, they are 1.34, 5 and 3.74, respectively. In parisite-(Ce), they are 1.34, 5.32 and

3.94. In synchysite-(Ce), they are 1.51, 4.32 and 2.87, respectively. Parisite-(Ce) and synchysite-(Ce) from strongly overprinted carbonatites have higher Y, Th and lower Sr (Table 4) than those from weakly overprinted carbonatites.

A notable feature of synchysite-(Ce) is the higher amount of Y, compared to other carbonatites worldwide, where yttrium has not been observed: Mountain Pass, California, U.S.A. (Olson *et al.* 1954, Castor 2008), Kangankunde, Malawi (Wall & Mariano 1996), Bayan-Obo, Inner Mongolia (Chao *et al.* 1997) and Yuzhnoe and Arshan, Russia (Ripp *et al.* 2000).

Barite and *Ba-rich celestine* occur in both weakly and strongly overprinted carbonatites. Barite contains up to 2.82 wt.% SrO, and up to 1.04 wt.% CaO, and Ba-rich celestine contains as much as 1.97 wt.% CaO (Table 5).

Columbite-(Fe) + quartz mixtures (Fig. 5) may be pseudomorphs after pyrochlore (?) in the strongly overprinted carbonatites; in most carbonatites, columbite has been described as a replacement mineral after pyrochlore (*e.g.,* Kapustin 1980). However, there are no remnants of the original pyrochlore in the strongly overprinted carbonatites. Columbite-(Fe) contains up to 4.46 wt.% TiO_2 and up to 1.06 wt.% MnO, and little V (up to 1.40 wt.% oxide).

DISCUSSION

Roedder (1973) studied the fluid inclusions in fluorite of Amba Dongar and concluded that the fluorite deposits related to the carbonatites were formed from low-salinity (0.8–1.8 wt.% equiv. NaCl), low-temperature (100–150°C) CO_2-bearing aqueous fluids. Williams-Jones & Palmer (2002) based their study of fluid inclusions on apatite in sövites and on quartz from surrounding fenitized sandstones; they showed that the fluid evolution in the Amba Dongar carbonatite started with the exsolution from the magma of a sulfate- and potassium-rich, CO_2-CH_4-bearing aqueous fluid. Later, at relatively shallow depths, a second solute-rich aqueous fluid exsolved, with lower values of K/Na and S/Cl, and significant concentrations of Ca, Al and Si.

We believe that the mineral association of quartz, florencite-(Ce), strontianite, barite and sparse of REE fluorcarbonates typical of weakly overprinted carbonatites results from high activity of OH^-, $(SO_4)^{2-}$, Al and Si in the fluid. Thereafter, during evolution, the chemical composition of fluid was modified and characterized by a increased activity of F^-, $(SO_4)^{2-}$, REE and Si, based on the mineral association (barite, fluorite, quartz and REE fluorcarbonates) of the strongly overprinted carbonatites.
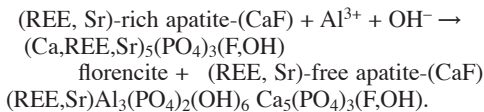
TABLE 4. REPRESENTATIVE COMPOSITIONS OF BASTNÄSITE-(Ce), SYNCHYSITE-(Ce), AND PARISITE-(Ce) IN THE AMBA DONGAR SÖVITES

	Weakly overprinted sövite						Strongly overprinted sövite			
	Bsn 1	Bsn 2	Pst 3	Pst 4	Syn 5	Syn 6	Pst 7	Syn 8	Syn 9	Syn 10
CaO wt%	4.73	2.89	8.42	7.38	17.00	15.13	10.24	19.53	18.47	18.54
SrO	1.61	1.57	1.47	1.27	1.01	1.11	n.d.	0.75	n.d.	1.03
Ce_2O_3	31.79	32.85	27.86	28.42	21.67	23.00	29.17	21.85	19.20	19.41
La_2O_3	23.35	25.04	20.66	21.03	14.57	16.82	11.76	9.70	10.88	8.13
Pr_2O_3	2.26	1.93	1.71	2.07	1.57	1.59	2.38	2.07	1.34	1.97
Nd_2O_3	6.46	6.47	5.29	5.29	5.50	5.15	6.83	7.33	7.35	7.09
Y_2O_3	n.d.	n.d.	n.d.	n.d.	1.67	1.23	2.56	2.12	7.37	1.02
ThO_2	1.51	2.44	2.67	2.38	2.73	2.85	3.54	2.32	n.d.	8.50
F	7.28	5.74	6.67	7.20	6.15	5.75	7.37	5.57	6.55	5.71
Total	78.98	78.93	74.75	75.03	71.87	72.63	73.86	71.23	71.16	71.40
-O=F_2	3.06	3.05	2.80	3.02	2.58	2.41	3.09	2.34	2.75	2.39
Ca *apfu*	0.17	0.11	0.83	0.73	0.91	0.82	1.00	1.05	0.97	1.00
Sr	0.03	0.03	0.08	0.07	0.03	0.03	-	0.02	-	0.03
Ce	0.39	0.42	0.94	0.96	0.40	0.43	0.97	0.40	0.34	0.36
La	0.29	0.32	0.70	0.71	0.27	0.31	0.39	0.18	0.20	0.15
Pr	0.03	0.02	0.06	0.07	0.03	0.03	0.08	0.04	0.02	0.04
Nd	0.08	0.08	0.17	0.17	0.10	0.09	0.22	0.13	0.13	0.13
Y	-	-	-	-	0.04	0.03	0.12	0.06	0.19	0.03
Th	0.01	0.02	0.06	0.05	0.03	0.03	0.07	0.03	-	0.10
F	0.77	0.64	0.97	1.05	0.97	0.92	1.06	0.88	1.01	0.91
OH	0.23	0.36	1.03	0.95	0.03	0.08	0.94	0.12	-	0.09

Structural formulae are based on 4, 7 and 11 anions for bastnäsite-(Ce), synchysite-(Ce) and parisite-(Ce), respectively. n.d.: not detected. Symbols: Bsn: bastnäsite-(Ce), Pst: parisite-(Ce), Syn: synchysite-(Ce).

We further observe that there were two important ways for formation of hydrothermal minerals at Amba Dongar: (1) re-equilibration and recrystallization of the primary (early-crystallized) minerals, such as apatite-(CaF), calcite, pyrochlore, releasing elements to form secondary minerals like strontianite, florencite-(Ce), and columbite, and (2) introduction of key elements from hydrothermal solution into the carbonatites to form quartz, barite, ankerite, and REE fluorcarbonates.

We speculate that the primary apatite-(CaF) was leached by Al-bearing hydrothermal fluid leading to the formation of florencite-(Ce). The replacement of apatite-(CaF) by florencite-(Ce) may be represented by a hypothetical reaction of the type:

$$(REE, Sr)\text{-rich apatite-(CaF)} + Al^{3+} + OH^- \rightarrow$$
$$(Ca,REE,Sr)_5(PO_4)_3(F,OH)$$
$$\text{florencite} + \quad (REE, Sr)\text{-free apatite-(CaF)}$$
$$(REE,Sr)Al_3(PO_4)_2(OH)_6 \quad Ca_5(PO_4)_3(F,OH).$$

Similar replacement of apatite-(CaF) and monazite-(Ce) by florencite-(Ce) has been described by Nagy *et al.* (2002) in metamorphic rocks of the Sopron Hills, Eastern Alps.

We propose that aluminum was transported as a major constituent of the hydrothermal fluids that led to the formation of florencite. Evidence of high activity of aluminum at Amba Dongar can be seen in the feldspar-rich fenites (Deans *et al.* 1972, Viladkar 1996). More-over, according to Williams-Jones & Palmer (2002), the fluid inclusions in apatite from sövites contain significant concentrations of Al (up to 46 at.%.). The carbonatitic magma in Amba Dongar did contain minor Al, which is evident in presence of phlogopite in some sövites. The Kalkfeld carbonatite complex, in Namibia, can be considered a good example of the important role of aluminum as well, because alkali feldspar is a major constituent of fenites there, and the fluid inclusions from minerals of those carbonatites contain albite and cryolite as daughter minerals (Bühn & Rankin 1999).

Another possible example of replacement of a primary mineral to form secondary minerals at Amba Dongar involves the re-equilibration and recrystal-lization of calcite with the formation of strontianite, where strontium is liberated from the primary calcite, producing Sr-poor calcite in altered sövites. Part of the strontium probably was introduced by hydrothermal solutions. In addition to Sr, the residual fluid was characterized by a high activity of F^-, $(SO_4)^{2-}$, REE, Si and Al.

The formation of REE carbonates from hydro-thermal systems related to carbonatites has been discussed by Hogarth *et al.* (1985), Zaitsev (1996), Andersen (1986), and Andrade *et al.* (1999). These authors emphasized the important role of postmagmatic alteration and re-equilibration processes in the origin of hydrothermal REE minerals. Furthermore, the REE could be accumulating in a residual fluid, because there is in general a low mineral/melt partition coefficient for the REE (Sokolov 2005). In hydrothermal systems, the REE can migrate as complexes; it has been demon-strated both empirically (in field studies, references cited) and experimentally (*e.g.*, Balashov & Krigman 1975, Flynn & Burnham 1978, Bilal & Becker 1979, Bulakh *et al.* 1998), that the anions F^-, Cl^-, OH^-, CO_3^{2-}, SO_4^{2-} and PO_4^{3-} can be effective ligands for complexing the REE. The mineralogy of our samples suggests that these metals most likely migrated with F^-, resulting in the formation of REE fluorcarbonates.

Silicification is a common feature of carbonatite-related REE deposits (*e.g.*, Wall & Mariano 1996, Andrade *et al.* 1999, Ripp *et al.* 2000). According to Andrade *et al.* (1999), quartz from the hydrothermal association in the Barra do Itapiratã carbonatites, in Brazil, formed as a result of the assimilation of the host granite. Barker (2001) concluded that activity of silica generally is low in carbonate magmas, but increases during fractional crystallization and as a result of assimilation. At Amba Dongar, silica prob-ably was released during the fenitization of sandstone to form pure K-feldspar fenites, and transported in hydrothermal solutions. Moreover, on the basis Nd, Sr and Pb isotopic data, Simonetti & Bell (1995) proposed that silica saturation was due to interactions between a cooling fluid derived from the carbonatitic melt and the continental crust.

Assimilation can take place at deeper levels in the crust. The presence of silica in the carbonatitic magma at Amba Dongar is evident from the primary silicate minerals (pyroxene, mica). It is reasonable to assume that SiO_2 was concentrated in the residual fluids. Williams-Jones & Palmer (2002) documented the significant concentrations of Si in the carbonatitic fluid at depths of 3–5 km. Silica, like REE, can accumulate in residual fluid because there is a low mineral/melt parti-tion coefficient in the primary minerals (calcite, apatite). It can also be transported as complexes in a carbonatitic system, leading to increased silica in residual fluids.

There is isotopic evidence of low-temperature hydro-thermal activity at Amba Dongar complex (Simonetti & Bell 1995, Viladkar & Schidlowski 2000). Unusual oxygen isotope compositions of sövites at Amba Dongar may be due to low-temperature hydrothermal activity involving ^{18}O-depleted groundwater.

TABLE 5. REPRESENTATIVE COMPOSITIONS OF Ba, Sr SULFATES IN THE AMBA DONGAR SÖVITES

	Barite						Ba-rich celestine	
CaO wt%	1.04	0.33	0.40	0.53	0.44	0.57	1.97	1.44
BaO	64.98	63.60	64.57	62.87	63.33	63.88	24.80	19.17
SrO	1.46	2.07	0.00	2.82	1.41	1.47	44.20	47.25
SO₃	34.37	34.93	34.65	35.04	33.90	33.98	28.25	30.91
Total	101.9	100.9	99.6	101.3	99.1	99.9	99.2	98.8

Conclusions

Two principal conclusions emerge from this study.

(1) Hydrothermal minerals at the Amba Dongar were formed by re-equilibration and recrystallization of primary carbonatite minerals, with elements introduced by hydrothermal solution.

(2) There is evidence that OH^-, $(SO_4)^{2-}$, F^-, REE, Al and Si were important components of the hydrothermal fluid.

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