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Evolution of Calciocarbonatite Magma: Evidence from the Sövite and Alvikite Association in the Amba Dongar Complex, India

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1. Introduction

CaCO₃-rich carbonatites seem to be the most common and abundant carbonatite in many complexes. The Amba Dongar complex too, is dominantly composed of sövite. Its dike equivalent-(alvikite I and II), though volumetrically less abundant, occur in association with sövite. Majority of sövites samples are composed essentially of CaCO₃ and MgO content is low (0.99% average) and goes up to maximum 3% in phlogopite-sövite. A small fraction of periclase was reported earlier from some sövite samples by Viladkar and Wimmenauer, [4]. In this paper I propose that sövites are crystallized from calciocarbonatite magma. Trace elements and REE distribution patterns suggest that sövite fractionated to Alvikite I and II. The C and O isotope data provides evidence to supports the mantle derivation of the carbonatite magma at Amba Dongar.

2. General geology

Amba Dongar carbonatite-alkalic rock diatreme is located on the western periphery of the Deccan flood basalt province in Gujarat State, Western India. This sub-volcanic diatreme consisting of different phases of carbonatites, nephelinites and phonolites, has intruded into the Bagh sandstone and overlying Deccan basalts sequence and has been dated at 65.5 Ma [1].

The outstanding geological feature of the Amba Dongar complex is the bold exposures and clear intrusive relationship between different carbonatite units. Sövite is the predominant type of carbonatite and forms a ring dike around the inner rim of carbonatite breccia (Fig.1). Apart from major ring dike, sövite also forms large and small plugs within the Bagh sandstone terrain. Ankeritic carbonatite intrudes sövite at several places as dikes and large oval plugs while, last in the differentiation sequence, sideritic carbonatite phase intrudes the ankeritic carbonatite as thin dikes, in the southwestern part of the ring structure. Alvikite (phase I), dikes equivalents of sövite, are widespread within as well as outside the ring structure. Alvikite phase II intrude sövite at some places along the ring dike. Nephelinite plugs and phonolite dikes occur in the outer periphery of the ring structure. Large reserves of hydrothermal fluorite deposits (11.5 million tones) are associated with the carbonatites. A

vertical zonation of fenitization developed with strongly sodic and sodi-potassic fenites at deeper levels and potassic fenites at higher levels [2] [3]. The central part of the ring structure is occupied by intrusive basalt which replaced the original plug of carbonatite breccia. This youngest intrusive event seems to have occurred at 41.7 Ma [3].

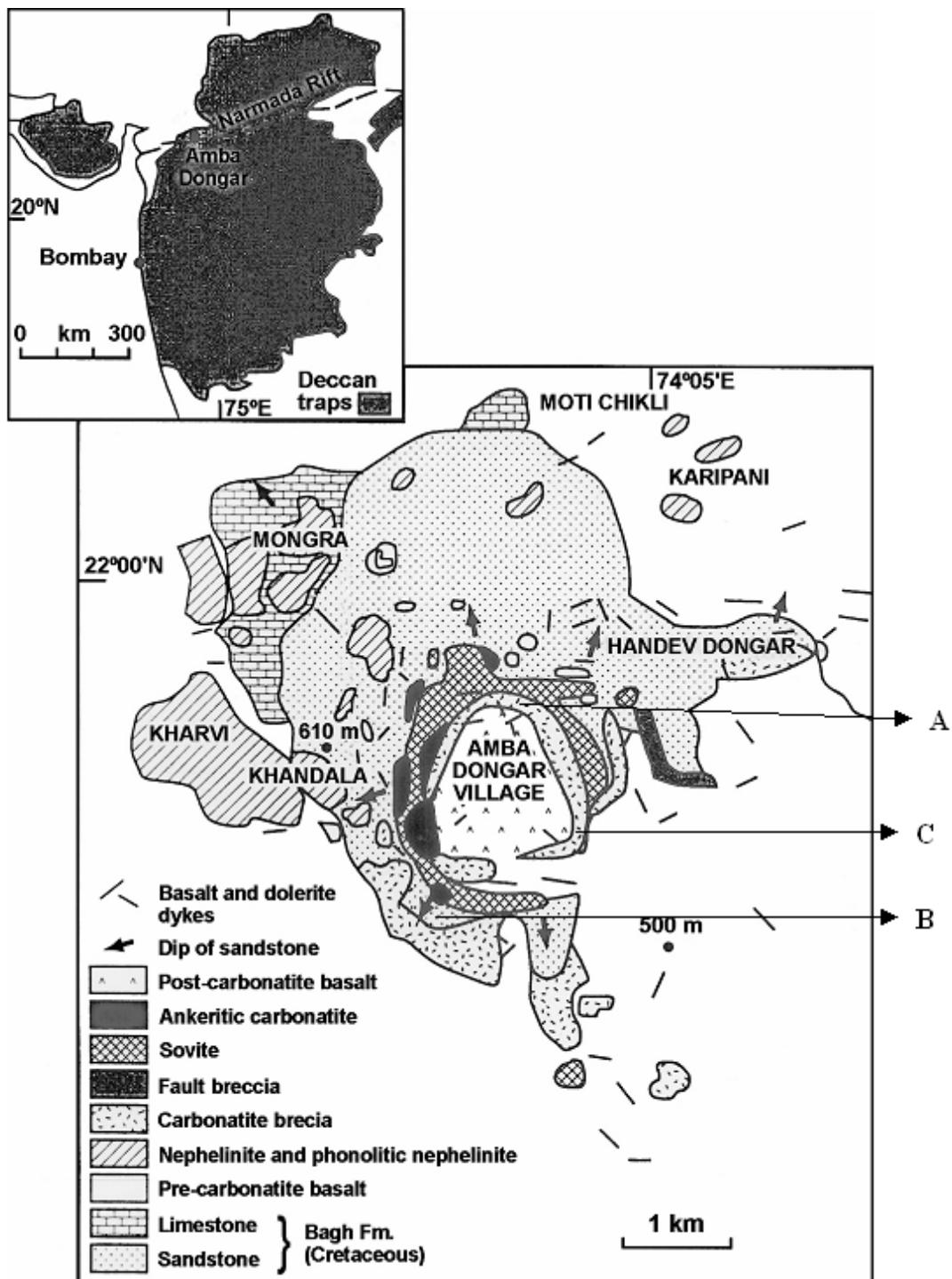


Fig. 1. Geological map of the Amba Dongar carbonatite diatreme with locations of Alvikite centres within sövite ring and outside the ring. A and C are in carbonatite breccia, B in Bagh sandstone.

It has been possible to establish the different phases of sövite intrusion with each phase marked by characteristic texture and mineralogy [3] [4]. The texture of sövite varies from coarse grained in the inner part of the ring in contact with carbonatite breccias to very fine grained in the outermost part of the ring in contact with the sandstone. Sövite also reveals excellent flow structure, and at several places large blocks of fenites are caught up within the flowage structure.

Early crystallization (segregations) of magnetite, apatite, pyrochlore and Nb-zirconolite occur as bands within sövite (Fig. 2). Silico-sövites (phlogopite-sövite and pyroxene-sövite) are not exposed at present level of erosion, however; many xenoliths of these rocks were encountered at deeper levels during mining operations and also in a tunnel driven through the sövite exposure.



Fig. 2. Banding (shown by an arrow) in sövite.

The alvikites vary in thickness from several centimeters to almost a meter. Detailed field investigation of numerous dikes suggests that there are at least two distinct intrusive phases; one either predating or contemporaneous with the main phase of sövite intrusion while the second (alvikite II) clearly postdates the sövite intrusion. In general, alvikites do not show any particular trend in their disposition and, they are located all over the central ring structure as well as outside the ring structure in sandstones and basalts. Three main centres of alvikite (Fig. 1) have been located in and around the ring dike of sövite. Alvikites of phase I intrude mainly carbonatite breccia (Fig. 3), sandstone and basalts while those of the second phase intrude sövite and fenite outcrops. Apart from these exposures alvikites were also exposed in fluorite mine workings and along road cuttings.

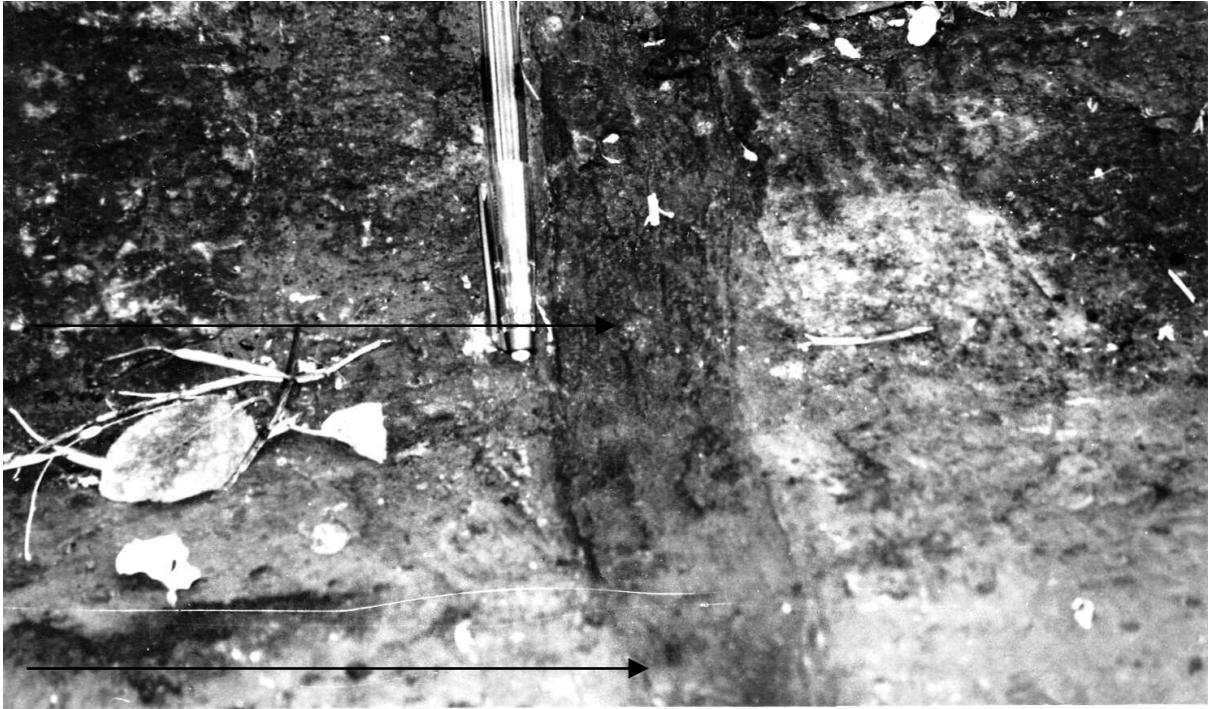


Fig. 3. Alvikite I (shown by an arrow) in carbonatite breccia.

The majority of phase I alvikites show thin chilled contacts with the host rocks. In some dikes grain size variation from margin (smaller grains) to center (larger grains) is observed. However, the most conspicuous feature observed around alvikite dikes of this phase (whenever they intrude sandstone) is the feldspathization of country rock sandstone. The fenitization effects along alvikite-basalt contact are not so conspicuous except for a small increase in K (formation of K-feldspar), modal apatite and calcite in the host basalt.



Fig. 4. Alvikite II (shown by an arrow) in sövite.

Second generation alvikites (phase II) intrude sövite (Fig. 4) in the ring dike and some are located in exposures of fenites. Mineral zonation is clearly developed in some of these alvikites with margins exclusively rich in calcite while concentration of magnetite and pyrochlore in the central part of the dikes.

In Amba Dongar, along with intrusive phase of calciocarbonatite, the extrusive phase also occurs in the form of tuff (Fig.5) in the western part at Mongra [20].



Fig. 5. Carbonatite tuff beds (shown by an arrow) in Mongra, West of Amba Dongar.

3. Petrography and mineralogy

Sövite shows various textures such as coarse grained, medium grained and, fine grained porphyritic [6]. In porphyritic types, the euhedral calcite phenocrysts are embedded in fine grained groundmass calcite of a later generation. In silico-sövites primary phlogopite is the most abundant silicate mineral while diopsidic augite is next in abundance. Phlogopite occurs as euhedral to sub-hedral crystals which locally show resorption by surrounding calcite. Occasionally phlogopite forms thin bands with individual crystals showing parallel orientation. The majority of phlogopite crystals are homogenous and do not show any zoning. Accessory minerals in order of decreasing abundance are apatite, magnetite, fluorite, barite, galena, pyrochlore, niobian-zirconolite, zircon, bastnaesite and monazite.

Alvikites, on the other hand, are medium to fine grained equigranular with anhedral grains of calcite and ankeritic calcite (in alvikite II) showing interlocking texture. Porphyritic texture is not uncommon. Some dikes show good flow texture with platy calcite crystals aligned along the direction of flow. All alvikite dikes of phase I have mineralogy broadly similar to coarse sövites whereas the post-sövite alvikites (phase II) are characterized by

abundance of phlogopitic-biotite displaying strong zoning with pale brown core and dark brown rims. Oscillatory zoning is not uncommon. Dark brown to reddish brown tetraferriphlogopite occurs as small isolated flakes as well as rims of some strongly zoned crystals [7]. Octahedral magnetite is very common; sometimes it also forms a thin rim around pyrochlore.

Micas from sövites and alvikites were investigated in detail earlier and the details of their evolutionary history from sövite to alvikite are discussed in earlier publication [7]. The chemical differences shown by the two types of micas (Mg-rich in sövite and Fe-rich in alvikite II) and their evolution trends from sövite to Alvikite II are thought to reflect the chemical evolution of the calciocarbonatite magma from which they crystallized.

4. Analytical method

Major and trace elements were determined by X-ray fluorescence using a Philips PW 1450/20 instrument at the Mineralogische-Petrographisches Institute of the Albert-Ludwigs University, Freiburg im Br. Germany. Rare earth elements were determined by the Neutron Activation method at the B.A.R.C., Trombay, Mumbai (Note: where only Ce, La and Nd values mentioned in the analytical tables, these are the XRF analyses. Other REE analyses (La-Lu) are by the Neutron Activation method).

C and O isotopes were determined at the Biogeochemistry section of the Max-Planck Institute, Mainz, Germany using following method

Finely powdered (200 mesh-size) carbonatite samples were reacted for 72 hours with anhydrous (100%) phosphoric acid at 25^o C [8] [9] to ensure quantitative reaction of dolomite and ankerite components. Carbon and oxygen isotope ratios were determined for CO₂ on a VG PRISM mass spectrometer equipped with a three-collector system allowing the simultaneous collection of the masses 44, 45, and 46. Results were corrected for ¹⁷O after Craig [9] and are reported as δ¹³C and δ¹⁸O values relative to the Pee Dee Belemnite (PDB) and Standard Mean Ocean Water (SMOW) standards, respectively, with

$$\delta^{13}\text{C} = \left\{ \left[\frac{(^{13}\text{C} / ^{12}\text{C})_{\text{sa}}}{(^{13}\text{C} / ^{12}\text{C})_{\text{st}}} \right] - 1 \right\} * 1000(\text{‰})$$

and

$$\delta^{18}\text{O} = \left\{ \left[\frac{(^{18}\text{O} / ^{16}\text{O})_{\text{sa}}}{(^{18}\text{O} / ^{16}\text{O})_{\text{st}}} \right] - 1 \right\} * 1000(\text{‰})$$

were sa = sample and st = standard. Standard deviations of the measurements were usually smaller than ± 0.1 ‰.

5. Geochemistry

The present section is aimed mainly to show how the parent calciocarbonatite magma at Amba Dongar fractionated. Geochemical differences between sövite and alvikite (phases I and II) during their evolution are brought out using spidergram for trace elements and the chondrite normalized REE patterns. Representative major, trace and rare earth elements analyses of sövite, alvikite (phase I) and alvikite (phase II) are given in tables 1, 2 and 3 respectively.

Sample	1201SOV	1244SOV	1250SOV	1270SOV	187SOV	195SOV	Ave.	SD	CV%
SiO ₂	2.33	7.19	0.68	3.82	4.82	2.62	2.97	3.167	0.949
TiO ₂	0.96	0.15	0	0	0.41	0	0.09	0.182	1.889
Al ₂ O ₃	0.26	0.18	0	0.17	0.58	0.18	0.16	0.187	1.232
Fe ₂ O ₃	2.22	2.08	1.66	1.1	5.25	1.74	2.63	2.468	0.955
MnO	0.59	0.53	0.73	0.44	0.15	0.66	0.5	0.429	0.826
MgO	0.26	0.16	3.08	0.16	2.05	1.16	0.99	0.972	1.067
CaO	50	47.84	50.57	51.9	47.19	50	49.34	3.511	0.071
Na ₂ O	0	0.15	0	0.12	0	0	0.04	0.066	1.516
K ₂ O	0	0	0	0	0.16	0	0.06	0.066	1.232
P ₂ O ₅	0.6	3.47	0.45	1.04	3.99	0.19	1.69	1.469	0.855
CO ₂	41.89	36.3	41.52	40.2	33.32	41.74	39.76	2.814	0.071
Total	99.31	98.05	98.69	98.95	97.92	98.29	92.95		
Ba	770	3860	6580	920	980	4130	4080	3125	0.786
Sr	6100	3630	2560	5515	6310	2260	6871	6402	1.018
Nb	1070	250	190	110	80	490	535.88	1179	0.636
Zr	80	110	70	35	260	80	101.51	151.58	2.204
Y	50	230	60	130	80	110	124	77.64	1.427
Th	120	130	60	30	80	40	59	56.64	0.796
U	10	45	0	50	30	0	13.97	30.12	1.988
V	10	0	65		115	50	110.14	157	1.483
La	290	1110	662	1830	666	1080	1040.44	894.4	0.903
Ce	879	2460	1660	2980	1350	2800	2581.14	2697	1.117
Nd	233	550	375	410	446	695	572.05	459	0.862
Sm	34	125	47	44	58	177			
Eu	9.23	19.9	11.3	10.4	14.3	25.3			
Tb	3.09	8.61	3.59	4.32	4.15	7.95			
Yb	4.28	16	3.5	12.7	4.2	8.83			
Lu	0.55	2.33	0.4	1.78	0.55	0.87			

Table 1. Representative analyses of sövite from different parts of ring dike and plugs.

Sample	1 1272AD(I)	2 1292AD(I)	3 79AD(I)	4 86/A/48(I)	5 BH/15 /ALV	6 V/alk/ 68	Ave
SiO ₂	1.64	2.48	1.44	3.78	1.56	6.22	3.65
TiO ₂	0.00	0.00	0.11	0.02	0.11	0.13	0.09
Al ₂ O ₃	0.37	0.00	0.11	0.21	0.07	1.78	0.75
Fe ₂ O ₃	1.20	0.40	2.11	1.03	3.19	1.29	1.84
MnO	0.44	0.20	0.85	0.28	0.16	0.37	0.28
MgO	0.17	0.80	0.24	0.60	0.42	1.46	0.75
CaO	51.68	53.26	50.10	51.65	50.49	48.52	49.42
Na ₂ O	0.20	0.10	0.18	0.06	0.05	0.54	0.16
K ₂ O	0.00	0.00	0.00	0.10	0.00	0.59	0.37
P ₂ O ₅	0.30	0.00	0.47	0.15	3.24	0.09	0.87
CO ₂	42.04	42.10	40.78	41.50	39.80	38.90	40.48
Total	98.54	97.34	96.39	99.38	100.49	99.91	
Ba	2815.00	*	9850.00	211.00	485.00	210.00	2960.3
Sr	16410.00	*	5210.00	2407.00	4295.00	1282.00	6610
Nb	40.00	*	160.00	70.00	135.00	10.00	87.9
Zr	20.00	*	80.00	44.00	220.00	78.00	161.5
Y	175.00	*	155.00	70.00	40.00	33.00	226.8
Th	120.00	*	130.00	10.00	40.00	0.00	59.4
U	20.00	*	20.00	0.00	10.00	0.00	62.44
V	*	*	*	33.00	85.00	40.00	23.3
La	1070.00	354.00	1830.00	823.00	310.00	60.00	634.18
Ce	2140.00	680.00	3340.00	1430.00	690.00	120.00	1237.455
Nd	445.00	181.00	612.00	230.00	200.00	45.00	265
Sm	*	30.00	73.00	64.00	*	14.00	
Eu	16.30	7.51	16.20	7.93	*	4.00	
Tb	6.57	2.93	6.65	2.10	*	2.00	
Yb	11.80	6.37	8.60	1.70	*	1.50	
Lu	1.41	0.79	1.22	0.34	*	2.00	

Table 2. Representative analyses of alvikite I.

Sample	1 1246/AD/II	2 1268(II)	3 1269AD(II)*	4 1288(II)	Ave
SiO ₂	8	4.77	5.19	1	4.11
TiO ₂	0.1	0.15	0.12	0.23	0.17
Al ₂ O ₃	0	0.05	0.04	0.04	0.07
Fe ₂ O ₃	8	10.21	7.9	9.3	8.19
MnO	1.5	0.74	1.43	0.5	0.84
MgO	0.6	0.41	0.45	0.8	0.5
CaO	45	40.2	44.48	46.3	45.08
Na ₂ O	0.1	0.12	0.15	0	0.07
K ₂ O	0.2	0	0	0	0.03
P ₂ O ₅	2.1	1.12	0.49	1.6	1.01
CO ₂	37.6	40.5	38.2	39	38.43
Total	100.2	98.27	98.45	98.77	98.26
Ba	12800	10810	8730	8200	8438
Sr	17510	3910	3210	6570	6059
Nb	1600	1890	580	1670	956
Zr	*	140	80	200	91
Y	180	120	130	80	112
Th	40	50	20	110	99
U	*	25	40	5	13
V	1055	200	135	0	269
La	2070	2140	2360	770	1508
Ce	8370	6010	5870	2250	4051
Nd	935	1050	1310	460	826
Sm	*	*	216	*	
Eu	*	*	34.6	*	
Tb	*	*	9.4	*	
Yb	*	*	3.5	*	
Lu	*	*	0.83	*	

Table 3. Representative analyses of Alvikite II.

On the CaO-MgO-Fe^t + MnO plot (Fig 6) there is clear separation between sövite and alvikite II (alvikite I plot in the same field as sövites). The alvikite II are richer in Fe on account of presence of ankeritic calcite in them. Most sövites and alvikites in Amba Dongar show MgO content below 1 wt % while those with MgO more than 1 wt% have either phlogopitic mica or periclase in them. Higher Fe₂O₃ contents of one sövite is due to high modal magnetite in this rock while the high Fe₂O₃ content of phase II alvikites can be attributed to a small amount of oxidized ankeritic calcite present in them.

The fractionation from sövite to alvikites (I and II) can be brought out more clearly on the spidergram for trace elements and chondrite normalized REE patterns. Plotting of average values of trace elements in sövites, alvikite (I) and (II) (Fig.7) show that incompatible trace elements gradually increase from alvikite I → sövite → alvikite (II), and as compared to alvikite I and sövites, alvikite II is enriched in all trace elements except Sr, Zr and Y.

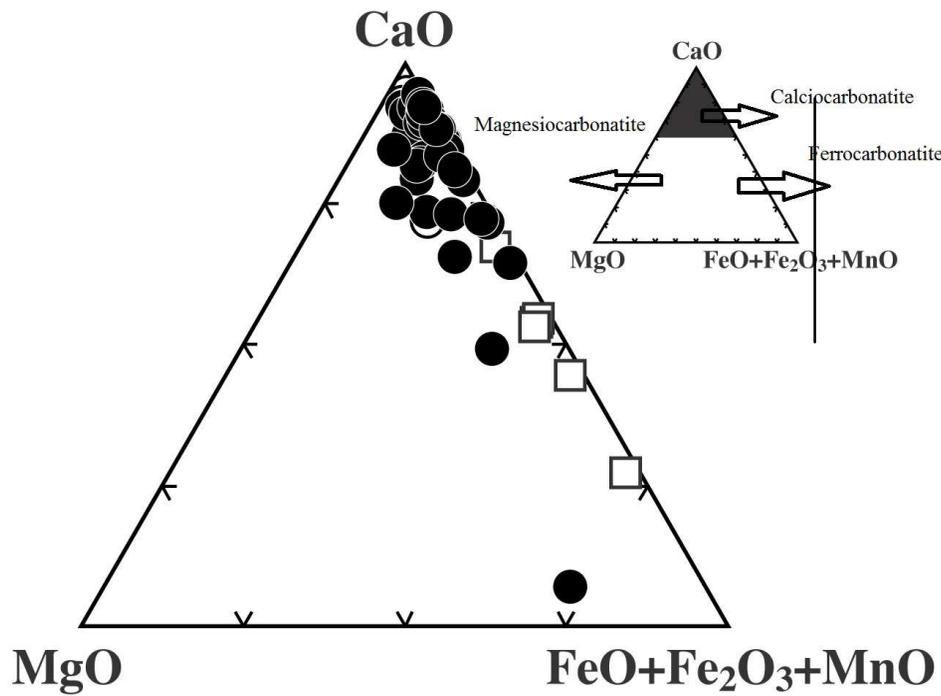


Fig. 6. Amba Dongar sövites (filled circles), Alvikite I (open circles) and Alvikite II (open squares) plotted on CaO-MgO-FeO+Fe₂O₃+MnO.

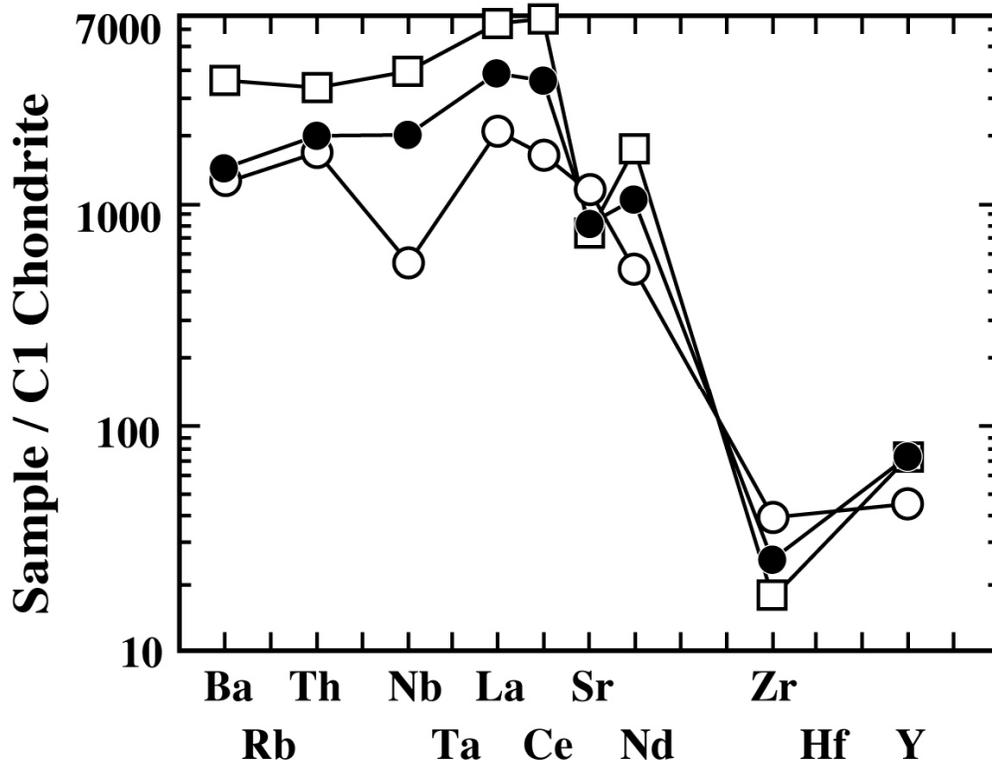


Fig. 7. Spidergram showing trace element variation in averages of Amba Dongar Alvikite I (open circle), Alvikite II (open square) and sövites (filled circle).

Chondrite normalized plots of sövite (two different phases, coarse and fine grained) and alvikites (I and II) of Amba Dongar are shown in Fig. 8. Alvikite II is strongly enriched in all REE in comparison to Alvikite I and sövites. Steep slope of LREE/HREE, similar and parallel REE patterns point to fractionation during evolution of these two types of carbonatites in Amba Dongar.

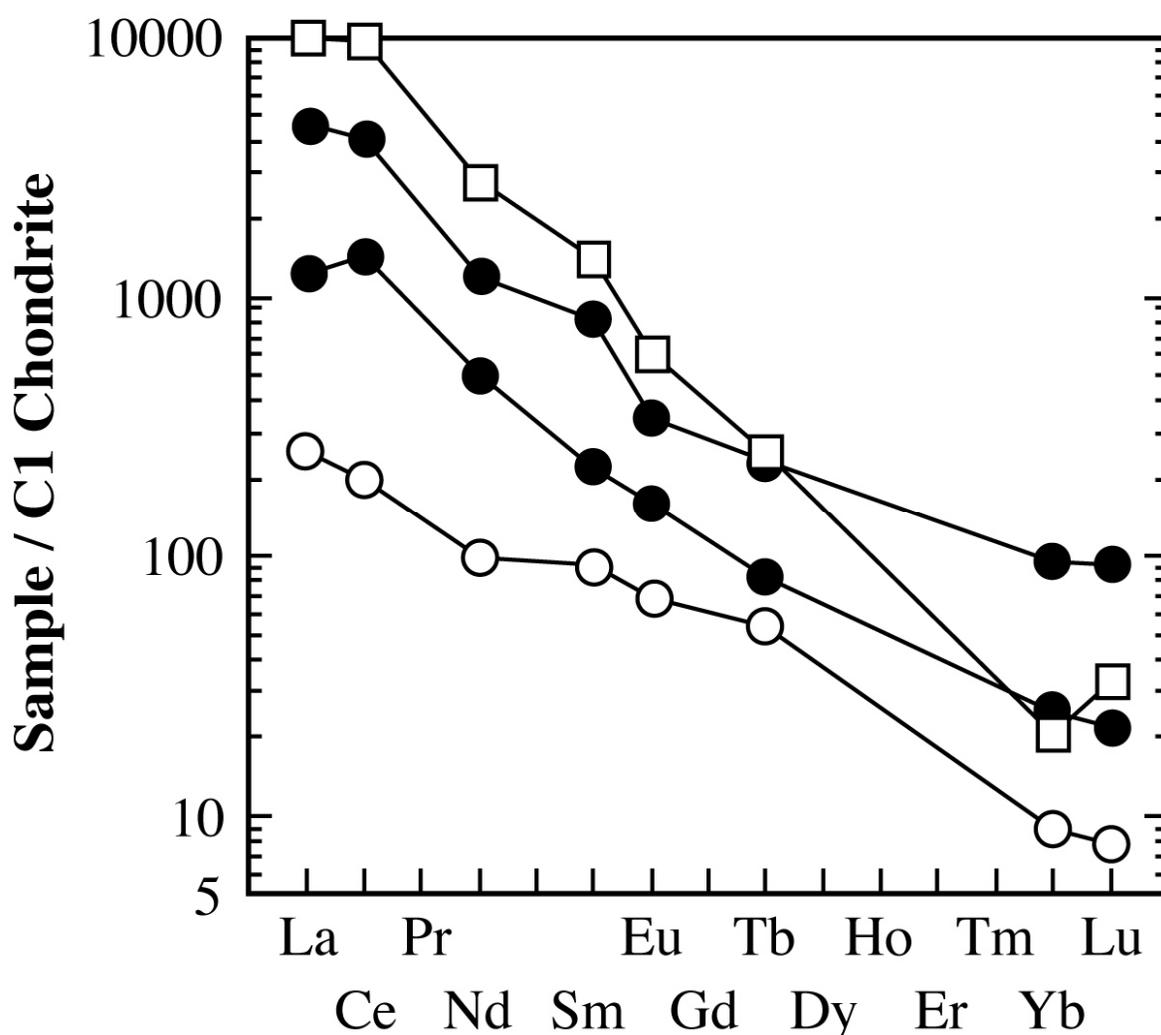


Fig. 8. Chondrite normalized REE distribution patterns of Amba Dongar sövites and alvikites; Open circle – Phase I alvikite, Open square -- Phase II alvikite, Filled circle - 2 phases of sövites (coarse and fine grained).

6. C and O isotopes

Stable isotopes of carbon and oxygen have proved useful in differentiating primary igneous carbonatites from marine carbonates. Relative to the PDB standard, mantle derived carbon has a $\delta^{13}\text{C}$ range of -7 to -5 ‰, and relative to the SMOW standard, mantle oxygen has a $\delta^{18}\text{O}$ range of +5 to +8 ‰ [10] [11]. However, various types of carbonatites (e.g., calcitic, dolomitic, ankeritic) may have evolved from a set of source values typical of the above range, and have undergone fractional crystallization and subsequent magmatic hydrothermal or meteoric alteration in diverse environments subsequent to their

emplacement. Therefore, it is customary to divide carbonatites broadly into two categories, primary and secondary. In the former variety, variations in both the isotope ratios (viz., $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) occur mainly due to fractional crystallization; the $\delta^{13}\text{C}$ values range from -9 to -1 ‰, while $\delta^{18}\text{O}$ ranges from $+5$ to $+15$ ‰ [12] [10]. In the second (altered) variety, the $\delta^{18}\text{O}$ values can increase up to $+30$ ‰, close to that of marine carbonates [13].

The data obtained on C and O isotopes of sövite, alvikites (I & II) supports the observation that calciocarbonatite magma at the Amba Dongar carbonatites complex, is mantle derived as majority of sövite samples and a few alvikites have retained the primary mantle signature. Accordingly, it is seen that the majority of sövite samples and few alvikite I and II plot in the field of mantle derived primary carbonatites in Fig. 9 [14]. Two samples of alvikite I fall within the mantle box while, remaining plot close to the box. Majority of alvikite II plot outside the box and this trend can be explained by process of fractionation. The sövite samples which lie farther away from the mantle box are mostly medium to fine grained sövites. The arrow shows the relation between phenocryst in sövite (star) and the

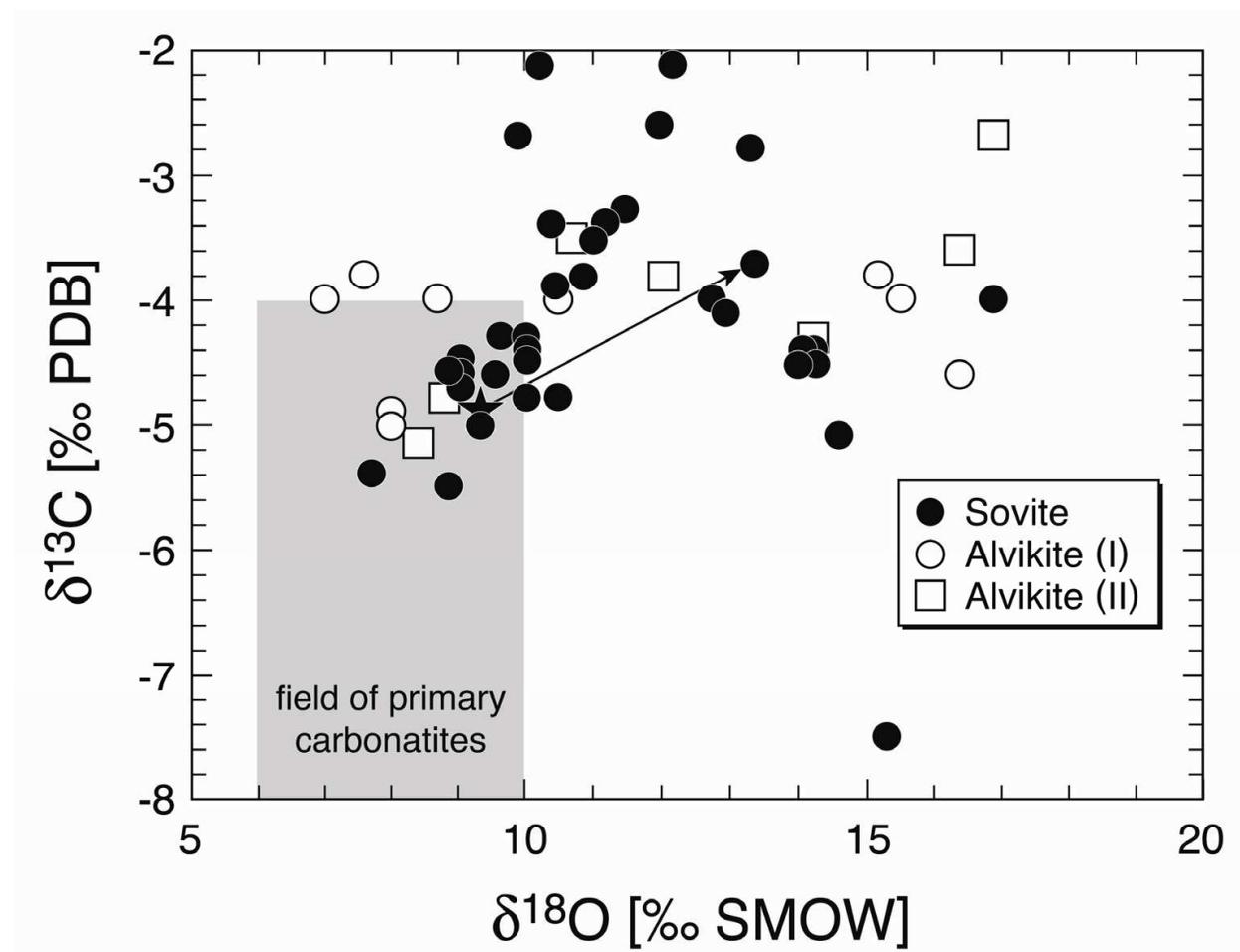


Fig. 9. C and O compositions of sövite, alvikite I and alvikite II of Amba Dongar complex. Open circle – Phase I alvikite, Open square -- Phase II alvikite, Filled circle – 2 phases of sövites (coarse and fine grained).

groundmass sövite. Coarse-grained sövite, in general, has lower $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values compared to fine grained ones, indicating that the fractional crystallization of the carbonatite magma initiated with the crystallization of coarse-grained sövite. As fractionation proceeded, the magma continuously became enriched in water (i.e., the $\text{CO}_2/\text{H}_2\text{O}$ ratio decreased), at which time crystallization of fine-grained sövites started, thus explaining their higher $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values.

It thus can be concluded that the primary carbonatites of Amba Dongar are interpreted to have evolved from a carbonatite parent magma of $\delta^{18}\text{O} = 8\text{‰}$ and $\delta^{13}\text{C} = -5.3\text{‰}$. The initial molar ratio of H_2O to CO_2 that best explains the trend is ~ 0.9 , with a crystallization temperature of $\sim 800^\circ\text{C}$. The secondary carbonatites in Amba Dongar appear to be altered mainly by a hydrothermal fluid with $\text{H}_2\text{O}/\text{CO}_2$ ratio between 0.001 and 0.1 at temperatures between 100 and 200°C . This model also applies to the secondary carbonatites found in carbonatite dikes from Amba Dongar [15].

7. Discussion

There has been an ongoing debate for many years about whether calciocarbonatite magmas exist or whether calcitic carbonatites are cumulate rocks derived from the crystallization of dolomitic carbonatite magma. While this may be true in some cases the present study of Amba Dongar dikes argues very strongly for the existence of a calcitic carbonatite magma (calciocarbonatite) magma, although somewhat magnesian in its early stages.

In Amba Dongar field relations between sövite and two phases of alvikites and petrographic observations such as tabular plates of calcite aligned along flowage, porphyritic texture with phenocrysts of calcite provide convincing evidence to assume that these carbonatites are crystallized from CaCO_3 -rich melt. The surface exposures of different generations of carbonatites are very clear and injection of alvikite II into sövite provides indisputable evidence of their origin that they being younger than the main sövite intrusion. With the help of geochemistry of major, trace and rare earth elements the fractionation of calciocarbonatite magma from sövite to two generations alvikite is well documented in the earlier section.

Early experimental studies of Wyllie and Tuttle [16] showed that calcite can crystallize as a liquidus phase at a temperature around 650°C at 0.1 GPa pressure.

According to Gittins [17] and Bailey [18] carbonatites can be precipitated from primary calciocarbonatite magma. Bailey listed 8 examples effusive calciocarbonatite [18], (Table 2, p. 6) and majority of them erupted from mantle as primary melt. Mariano and Roedder [19] studied products of Kerimasi carbonatite volcano and concluded that calcite was a primary phase in some flows. It is worth mentioning here that in Amba Dongar too an extrusive phase of calciocarbonatite magma occurs in the form of well bedded tuff and C and O isotopes of an unaltered tuff plots in field of mantle derived primary carbonatites [20]. Jago and Gittins [21] and Gittins and Jago [22] suggest, from experimental evidence, that a small amount of fluorine in carbonatite magma can reduce the temperature of crystallization of calcite which allows crystallization of calcite at atmospheric pressure

and conclude that the most of the calcite in extrusive carbonatite is primary magmatic. Dalton and Wood [23] demonstrated experimentally the formation of primary calciocarbonatite magma at low pressure. This is further supported by experimental results of Wyllie and Lee [24]. Additional evidence of existence of primary calciocarbonatite comes from occurrence of globules of calcite in mantle xenoliths which has been considered as evidence for the formation of immiscible CaCO_3 -rich melts in the mantle [25].

It thus appears that calciocarbonatite magma was the primary magma in Amba Dongar and during its evolution differentiated to alvikite I and II. That such calciocarbonatite magma was initially more magnesian is evident from the presence of phlogopite-sövite and periclase bearing sövite [4].

8. Acknowledgements

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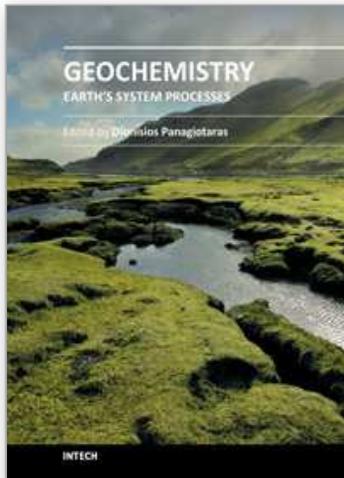
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