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Sr, Nd, O and H isotopic ratios in Ascension Island lavas and plutonic inclusions; cogenetic origin

D. Weis [1,2,*], D. Demaiffe [1], S. Cauet [1,**] and M. Javoy [2]

[1] *Laboratoires Associés Géologie-Pétrologie-Géochronologie, Université Libre de Bruxelles, Avenue F.D. Roosevelt, 50, B-1050 Brussels (Belgium)*

[2] *Laboratoire de Géochimie des Isotopes Stables, Université de Paris VII, 2, place Jussieu, 75251 Paris Cedex 05 (France)*

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Eighteen basic rocks from Ascension Island (South Atlantic) give a mean $^{87}Sr/^{86}Sr$ ratio of 0.70311 ± 17 for both volcanics and plutonic inclusions. The late-stage differentiated rocks (rhyolites and granitic inclusions) have much higher $^{87}Sr/^{86}Sr$ ratios, up to 0.712. All these rocks display the same range of Nd isotopic compositions (ϵ_{Nd} values from 6.9 to 11.1 with a mean on 12 samples of 8.4 ± 0.6) implying a cogenetic relation between the two sequences. The D/H systematics lead to the same conclusion.

In the Nd-Sr diagram, the data plot close to the mantle array and show a positive correlation. This suggests a mixing between a depleted MORB-type mantle, i.e. the upper mantle, and a hot-spot with less depleted geochemical characteristics, i.e. the OIB mantle source.

The total range of $\delta^{18}O$ values lies between 4.8‰ for plagioclase cumulates and 6.7‰ for the most evolved rocks (peralkaline granites and comendites). The basic rocks have values around 5.3‰, typical of mantle-derived material. These oxygen data indicate that the high $^{87}Sr/^{86}Sr$ ratios in the most evolved rocks (both volcanic and plutonic terms) result from the combination of two different processes: incorporation of slight amounts (< 1%) of high-temperature altered oceanic crust by the magma in the late stages of the differentiation process and then in-situ Rb decay since the time of formation of these rocks. Both processes were very effective because of the high Rb and low Sr contents of these evolved rocks.

Oxygen isotope systematics in the Ascension Island granites and rhyolites indicate that a fractional crystallization process alone does not produce $\delta^{18}O$ values higher than 6.7‰, i.e. that the ultimate $\delta^{18}O$ enrichment, relative to the initial basic magma, is not greater than 1.5‰.

1. Introduction

The origin of granites and the possibility of generating them by direct fractional crystallization of mantle derived magma have been the subject of numerous debates for a long time. Modelling the genesis of granitic rocks is indeed much more difficult than that of basic rocks: the presence of accessory minerals [1] trapping the trace elements and especially the rare earths, strongly limits the straight forward application of trace element geochemistry for quantitative estimations. Moreover, granites typically outcrop within the continental crust where the exchange and/or interaction processes may be important and complex. Consequently, the primary geochemical characteristics of these granites are not easily defined, when this is not totally impossible. Therefore, the occurrence of granitic inclusions within the typical oceanic alkali-basalt differentiation suite [2,3] of Ascension Island presents a rare opportunity to study granites in the absence of continental processes.

Recent Pb, Sr and Nd isotopic studies [4–6] have shown that, in some cases at least, three components have been involved in the genesis of oceanic island basalts. There is no doubt that the lower mantle (oceanic island basalt source) and the upper mantle (mid-oceanic ridge basalt source) are two of these three mantle reservoirs. For the third component, in contrast, several hypotheses have been proposed: (1) a third mantle reservoir

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[*] FNRS "Chercheur Qualifié".

[**] FNRS "Aspirant de Recherche".

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[4], (2) reinjection of weathered oceanic crust [5], or (3) incorporation of sediments [6] along subduction zones. These different possibilities were invoked to explain the isotopic data of rocks falling off the so-called mantle array in the Nd-Sr diagram. In a recent compilation, White [7] has tentatively identified five geochemically distinct mantle reservoirs (or sources) to account for the Nd-Sr-Pb isotopic data of oceanic basalts.

Four analyses on lavas of intermediate and basic composition [8,9] of Ascension Island gave Nd and Sr isotopic compositions slightly below the mantle array [9–11], between the MORB data and the St. Helena data [12]. In addition, previous isotopic studies on two independent sample collections, Oxford collection [13,14] for Pb and Sr and Miami collection [15,16] for Pb, show oceanic characteristics for these isotopes, with ratios in the range of those of oceanic island basalts. They also indicate a common mantle origin for both the volcanics and the plutonic inclusions; moreover, the Pb data point to the existence of two possible mantle sources. The Sr isotopic study on Ascension Island samples by Harris et al. [14] has shown the basic rocks to have $^{87}Sr/^{86}Sr$ around 0.703, whereas the more evolved terms have ratios up to 0.712. These authors ascribed these high Sr isotopic ratios to contamination of the magma by Sr-rich (up to 400 ppm) oceanic sediments. Later, as an addendum to their first paper [17], they also consider two other possibilities: these evolved rocks either formed in an earlier stage of the island's evolution and were later incorporated in more recent material or they were contaminated by seawater at the time of their formation, as three of the four rocks with low Sr content and high $^{87}Sr/^{86}Sr$ ratios have corrected values, for in-situ Rb decay in 1.5 Ma, lower than the seawater value. Ascension Island is located on magnetic anomaly number 4 which corresponds to a maximum age of 7 Ma [18]. This gives an upper age limit for the Ascension Island rocks. K/Ar age determinations were only obtained on lavas [3,19] and give maximum values of 1.5 Ma. It has been suggested by Tilley [20] that the plutonic inclusions can be considered as the slowly cooled equivalents of the lavas and it seems therefore very improbable that they could have crystallized much earlier than the lavas.

Nd and Sr isotopic data have been obtained on ten and twenty-seven samples respectively, to define the characteristics of Ascension Island source and to precise the relations between the two groups of rocks, volcanic and plutonic. $^{18}O/^{16}O$ and D/H ratios have also been measured in order to find an explanation which gives a better answer to the problem of high $^{87}Sr/^{86}Sr$ ratios, because oxygen is a very good tracer of both seawater interactions and sediment contamination and hydrogen is sensitive to water-rock interactions.

2. Geological setting

Ascension Island is formed by a large composite volcano located in the South Atlantic Ocean (7°57′S, 14°22′W), about 90 km to the west of the Mid-Atlantic Ridge and 50 km to the south of the Ascension fracture zone, which shifts the ridge 230 km to the east. The island covers an area of 93 km^2 and forms the upper part of a ≃ 3000 m high volcanic cone whose summit, Green Mountain, reaches 859 m above sea-level. A detailed description of the geology has been made by Daly [21]. The volcanic rocks show a wide-range of chemical compositions, typical of an oceanic alkali basalt differentiation suite [2,3,22], from olivine-rich basalt, through basalt, hawaiite, mugearite, trachyte to comendite. The basic samples include numerous coarse-grained blocks (referred to as "plutonic inclusions" in this paper) whose chemical compositions closely parallel those of the volcanics [2,3]: from olivine gabbro, diorite, monzodiorite to typical alkaline granite. However, from a mineralogical point of view, most gabbroic inclusions have tholeiitic affinities. These inclusions are entirely devoid of any metamorphic mineral or oriented structure. Their igneous origin [2,3,23] is substantiated by the occurrence of hypersolvus feldspar and empty acicular apatites as well as by plagioclase zonation and typical magmatic textures. Traces of local partial melting can be observed in some acidic inclusions [3].

3. Analytical techniques

Oxygen. Samples, crushed to < 200 mesh (the final step being in an agate mortar to obtain the best homogeneity) just before loading in Ni tubes, were reacted overnight at 550–600°C with bromine pentafluoride [24]. Oxygen was quantitatively converted to CO_2 on a carbon filament. Isotopic compositions were measured on a V.G. Micromass Ltd. 602C double collector mass spectrometer, with a CO_2 standard calibrated against silicate and carbonate oxygen international reference materials. The results are given in ‰/SMOW. Eighteen analyses of the silicate SF3 working standard (sandstone) give a mean value of 11.62 ± 0.06‰ (2 σ_m). NBS 28 was measured at 9‰ during this period. Each sample was analysed twice or more. The mean standard deviation of all replicate analyses on the 20 samples was 0.20‰.

Hydrogen. Hydrogen has been extracted following the procedure of Bigeleisen et al. [25]. Samples are outgassed under vacuum overnight at 150 °C. Hydrogen, extracted mainly in H_2O form by fusion in an induction furnace, is obtained by reduction on uranium at 700 °C. The precision is ±0.01‰ on water contents and ±3‰ on δD's.

Neodymium and strontium. For description of the Sr analytical techniques, the reader is referred to previous publications of our laboratory [26]. For Nd, samples (< 200 mg) were dissolved in a mixture 6 : 1 : 1 HF-HNO_3-$HClO_4$ in a teflon vessel. Separation of the REE was obtained with 4*N* HCl on a cation exchange column (Dowex 50W × 8) then Nd was separated with 0.3 *N* HCl on a column prepared following the Richard et al. [10] method adapted from Cerrai and Testa [27] (HDEHP(di(2ethylhexyl)orthophosphoric acid) on teflon powder). Each separation was repeated twice. Both types of columns were calibrated with ^{139}Ce. The total blanks for the whole procedure were always of the nanogram order, negligible in view of the Nd content of the samples. The Nd isotopic compositions were measured on double Re filament with a Finnigan MAT 260 mass spectrometer. Nd is run as metal. For each run, the 146, 143, 144 and 145 isotopes are measured together with the background at mass 147.5. The mean of the $^{145}Nd/^{144}Nd$ ratios for the 10 samples from the Miami collection is 0.34836 ± 1 (2 σ_m), all values normalized to $^{146}Nd/^{144}Nd = 0.7219$. The Nd isotope composition of the Johnson Matthey Nd oxide measured during this study was 0.51201 ± 4 and 0.34834 for the $^{143}Nd/^{144}Nd$ and $^{145}Nd/^{144}Nd$ ratios respectively. Because the $^{143}Nd/^{144}Nd$ value was significantly higher than the adopted value (0.51193) for that standard, a correction of -8×10^{-5} has been applied to all the samples (this corresponds to a ϵ_{Nd} correction of −1.6) for that ratio to account for this analytical bias (except for samples H81L and DS44 which were measured after the problem has been fixed). A duplicate of sample 10 has been analysed at the C.R.P.G. laboratory in Nancy and confirms the validity of that correction.

4. Results

Oxygen. We have measured $^{18}O/^{16}O$ ratios in 20 samples representing the whole petrologic suite for lavas and plutonic inclusions in Ascension Island. They are reported in Table 1. The $\delta^{18}O$ values are little variable and low, in the range of 5.2 to 6.8‰ for the lavas (except sample Ia) and 4.8 to 6.5‰ for the plutonic inclusions. The mean of the four basic lavas is 5.3 ± 0.08‰, i.e. in the lower part of the range of values reported for mantle-derived material [28–31]. Only the trachyte Ia has a very high $\delta^{18}O$ value (14.70‰) but also a

TABLE 1

Ascension Island rocks: $\delta^{18}O$ and δD values (both in ‰ relative to SMOW) and water contents (in %)

Sample	Petrographic type	$\delta^{18}O$	δD	H_2O^+
Lavas				
10	basalt	5.30 ± 0.01	−61	0.24
15150	basalt	5.36 ± 0.03	−52	0.35
63	hawaiite	5.23 ± 0.23	−35	0.41
77	basaltic tuff	5.28 ± 0.16	−31	0.60
Ia	trachyte	14.70 ± 0.26	−64	5.2
72	trachyte	5.55 ± 0.01	−73	0.17
76	trachyte	5.88 ± 0.08	−47	0.53
38	rhyolite	6.77 ± 0.04	−61	0.43
74	comendite	6.21 ± 0.25	−69	0.42
Plutonic inclusions				
25	gabbro	4.89 ± 0.22	−44	0.71
51	gabbro	5.46 ± 0.03	−77	0.48
19	hypersolvus monzodiorite	5.00 ± 0.18	–	0.29
23		4.81 ± 0.29	−59	0.27
31		4.89 ± 0.10	−46	0.30
18	subsolvus monzodiorite	5.70 ± 0.10	−38	0.52
24		5.23 ± 0.17	−68	0.27
41		5.14 ± 0.08	−70	0.23
2b	alkaline granite	5.76 ± 0.19	−82	0.40
17318 P	hypersolvus granite	5.58 ± 0.06	−79	0.17
17343	subsolvus granite	6.46 ± 0.29	−62	0.20

very high water content (5.2%) indicative of complex deuteric or meteoric alterations.

There are no striking differences between lavas and plutonic inclusions, except that in some of these latter rocks the $\delta^{18}O$ values are slightly lower than in the basic lavas, hence than typical mantle values. These samples correspond mostly to cumulate rocks (gabbros and hypersolvus monzodiorites).

We have not analysed mineral separates on those samples because there was no more material available. Sheppard ([32] and personal communication) analysed separated minerals from three Ascension Island acidic plutonic inclusions and indicated $\Delta_{QZ\text{-}KF}$ fractionation values around 1‰.

Hydrogen. Eight volcanic rocks (from basalt to comendite) and eleven plutonic inclusions (from gabbro to alkaline granite) have been analysed. The δD's (reported in Table 1) range from −73 to −31‰ in the lavas and from −82 to −44‰ in the plutonic inclusions. The water contents (H_2O^+) vary respectively from 0.17 to 0.60% in the lavas (except for sample Ia which, as discussed above, has 5.2% water and will not be discussed further because of obvious alteration problems) and from 0.20 to 0.71% in the inclusions.

TABLE 2

^{87}Sr/^{86}Sr ratios and Rb and Sr concentrations (ppm) of Ascension Island lavas and plutonic inclusions

Sample	Petrographic type	Rb	Sr	^{87}Rb/^{86}Sr	^{87}Sr/^{86}Sr ± $2\sigma_m$	1.5 Ma [a]	7 Ma [a]	T_1 [b]	T_2 [c]
Lavas									
8	basalt	35	493	0.20	0.70298 5				
10	basalt	39	477	0.24	0.70298 7				
70	basalt	19.8 [d]	517	0.111	0.70306 8				
15150	basalt	24.0 [d]	534	0.130	0.70290 3				
63	hawaiite	35.4 [d]	722 [d]	0.142	0.70305 4				
64	hawaiite	66.3 [d]	1060 [d]	0.181	0.70339 3				
77	basaltic tuff	22.4 [d]	400 [d]	0.16	0.70320 6				
Ia	trachyte	89.1 [d]	17.8 [d]	14.4	0.70648 9	0.70617	0.70505	17.1	
72	trachyte	63	18.7 [d]	9.7	0.70396 12	0.70375	0.70299	6.9	
76	trachyte	145	54.1 [d]	7.8	0.71230 6	0.71213	0.71153	84.5	30.1
38	rhyolite	131	10.4 [d]	36	0.70824 6	0.70746	0.70462	10.1	
74	comendite	110	2.40 [d]	133	0.70620 15	0.70337	-	1.7	
Plutonic inclusions									
25	gabbro	0.84 [d]	526	0.0044	0.70300 6				
26	gabbro	0.49 [d]	563	0.0026	0.70370 7				
51	gabbro	0.93 [d]	160	0.017	0.70396 7				
59	gabbro	1.5 [d]	446	0.0097	0.70293 4				
Ib1	subsolvus monzodiorite	30	318	0.27	0.70297 6				
18		76	52	4.3	0.70588 10	0.70579	0.70545	47.3	
24		56	199	0.82	0.70292 4				
41		85	242	1.0	0.70303 9				
43		65	477	0.39	0.70282 9				
19	hypersolvus monzodiorite	91	264	1.00	0.70306 8				
23		72	55	3.8	0.70350 3	0.70342	0.70312	9.3	
31		65	43.6 [d]	4.3	0.70307 9				
2b	alkaline granite	114 [d]	4.9 [d]	67	0.71214 5	0.71071	0.70546	9.6	3.3
17318 P	hypersolvus granite	96	4.1	68	0.71028 13	0.70883	0.70351	7.5	1.3
17343	subsolvus granite	73	3.4 [d]	62	0.70814 56 0.70775 93	0.70682	–	5.8	

[a] Initial ^{87}Sr/^{86}Sr ratio calculated for the given age.

[b] T_1: time necessary to reach the measured ^{87}Sr/^{86}Sr starting from 0.703, in Ma.

[c] T_2: time necessary to reach the measured ^{87}Sr/^{86}Sr starting from 0.709, in Ma.

[d] Isotope dilution determination. Otherwise X.R.F.

Strontium. Twelve lavas and fifteen plutonic inclusions were analysed for Sr isotopic composition and Rb and Sr concentrations. The results are reported in Table 2. Basic rocks, either volcanic (basalt, hawaiite) or plutonic (gabbro, monzodiorite), have similar Sr isotopic composition with $^{87}Sr/^{86}Sr$ ratios around 0.703 (mean value of eighteen ratios: 0.70311 ± 17); they fall in the lower part of the oceanic basalt range of values (0.7026–0.7065 [33]). For the acidic rocks, the $^{87}Sr/^{86}Sr$ ratios can reach values as high as 0.712 for both volcanics and plutonic inclusions.

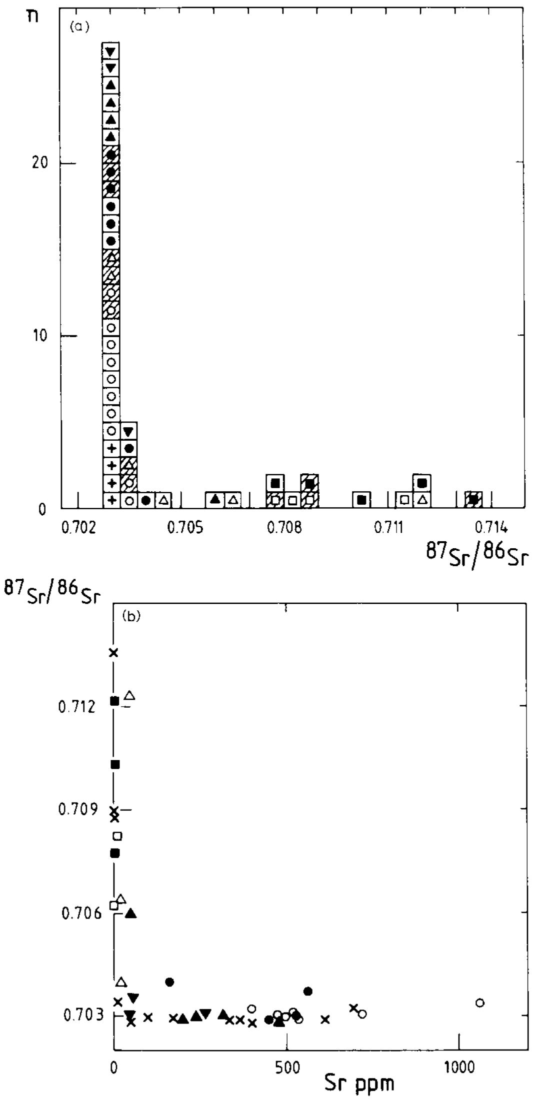

Fig. 1. (a) Histogram of $^{87}Sr/^{86}Sr$ ratios in Ascension Island rocks. Open symbols: lavas; closed symbols: plutonic inclusions. Circle: basic rocks, triangle: intermediate rocks, square: acidic rocks. The data of Harris et al. [14,17] are also reported with the same symbols for the different petrographic types but with hatched pattern. The four lavas analysed by O'Nions and Pankhurst [8] are given by the plusses. All the basic terms, either lavas or plutonic inclusions, have $^{87}Sr/^{86}Sr$ ratios around 0.703. (b) $^{87}Sr/^{86}Sr$ vs. Sr ppm for Ascension Island rocks. Symbols as in (a) for our data while the data of Harris et al. [14,17] are reported as crosses. There is a very large $^{87}Sr/^{86}Sr$ increase for Sr contents lower than 5 ppm.

As shown by the histogram of measured $^{87}Sr/^{86}Sr$ ratios (Fig. 1a), there is no difference in Sr isotopic compositions between the lavas and plutonic inclusions: this is true for both the basic and the acid rocks. The hypersolvus and subsolvus monzodiorites have also similar Sr ratios. The data of Harris et al. [14] who also observed high $^{87}Sr/^{86}Sr$ ratios in the most evolved lavas and plutonic blocks are reported on the same figure.

Neodymium. Six lavas and six plutonic inclusions covering most of the petrographic types (including two samples from the Oxford collection considered as representative of the most basic terms: H81L—a basalt, and DS44—an olivine-

TABLE 3

Nd isotopic composition and Sm and Nd concentrations (in ppm) in some of the Ascension Island rocks

Sample	$^{143}Nd/^{144}Nd$ $\pm 2\sigma_m$	ϵ_{Nd} [a]	Nd [b]	Sm [b]
Lavas				
H81L [d]	0.51306 1	8.2		
10	0.51306 2	8.2	64	16
	0.51305 [c] 3	8.1		
77	0.51313 6	9.6		
	0.51312 1	9.4		
15150	0.51308 5	8.6	39	10
74	0.51321 5	11.1	93	24
38	0.51306 1	8.2		
Plutonic inclusions				
DS44 [d]	0.51302 8	7.4		
25	0.51299 4	6.9		
41	0.51305 2	8.0		
19	0.51307 3	8.4		
	0.51304 5	7.8		
17318 P	0.51304 2	7.8		
17343	0.51309 1	8.8		

[a] The ϵ_{Nd} was calculated for a bulk earth value of $^{143}Nd/^{144}Nd = 0.512638$ [11].
[b] J.G. Schilling's data (INAA).
[c] Analysis done at the C.R.P.G. (Nancy).
[d] Samples given by C. Harris (Oxford collection).

plagioclase gabbro) were analysed for Nd isotopic compositions. The data are reported in Table 3. In contrast to the Sr isotopic data, all the analysed samples, either volcanic or plutonic, either basic or acid, have similar Nd isotopic compositions. The mean of $^{143}Nd/^{144}Nd$ ratios is 0.51310 ± 5 ($\epsilon_{Nd} = +9.0$) for six lavas and 0.51304 ± 3 ($\epsilon_{Nd} = +7.8$) for six plutonic inclusions, i.e. indistinguishable within errors. As already observed for their Pb isotopic compositions and although the range of values of both lavas and plutonic inclusions are comparable, it is clear that, individually, the inclusions have not been isotopically reequilibrated with their enclosing lava since they show distinct isotopic compositions. Four other lavas analysed earlier by O'Nions et al. [9] give a mean of 0.51301 ± 3 ($\epsilon_{Nd} = +7.3$). This is slightly lower than our values but still within errors and fall within the same trend in the Nd-Sr diagram. These positive ϵ_{Nd} values imply that the source region of the Ascension Island magma has a time-integrated LREE depletion.

5. Discussion

Considering the water contents and the D/H ratios (Table 1), two features are immediately apparent:

(1) The water contents are low in both categories of rocks, which is in agreement with the fact that these samples have been selected for their freshness. There is no significant difference between the plutonic inclusions and the lavas.

(2) The range of δD's is very similar for the lavas and the plutonic inclusions, although one could note a shift of about 10‰ towards higher δD's for the volcanics. On the average, in both lavas and plutonic inclusions, the acidic rocks are slightly lower in δD.

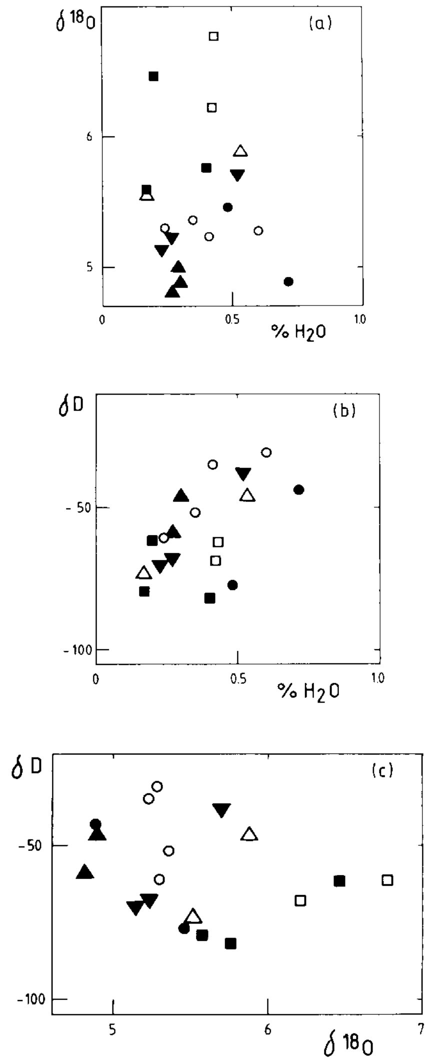

Fig. 2. (a) $\delta^{18}O$ (‰) vs. H_2O^+ (%) for Ascension Island rocks. Symbols as in Fig. 1. (b) δD (‰) vs. H_2O^+ (%): the data define a general positive correlation. (c) δD (‰) vs. $\delta^{18}O$ (‰): note the lack of any correlation.

If we try to correlate the three variables $\delta^{18}O$, δD and H_2O^+ with one another (Fig. 2a, b, c), one can notice that: (1) there is no correlation between $\delta^{18}O$ and H_2O^+; (2) there is a general positive correlation between δD and H_2O^+ for both types of rocks; (3) in a δD versus $\delta^{18}O$ diagram, the two types of rocks are rather significantly decoupled with lower $\delta^{18}O$ (or δD, or both) for the inclusions. The absence of correlation between $\delta^{18}O$ and H_2O^+ (Fig. 2a) stems from the fact that $\delta^{18}O$ variations can be associated with a variety of distinct phenomena among which magmatic differentiation and interaction with water at various temperatures. We shall see later that magmatic differentiation probably plays a major role. In contrast, D/H ratios are more sensitive to contamination phenomena because of larger relative differences between the possible reservoirs (e.g. mantle with values at −70 to −80‰ [37,38]) and seawater (with values around zero) which should lead to the positive correlation observed. Degassing of a water-rich magma shall deplete the residual dissolved water in deuterium [39] and this should also lead to a positive correlation.

There is no significant difference between plutonic inclusions and lavas. In that respect, we do not agree with the conclusions of Sheppard and Harris [40] that the lavas have not degassed. Their conclusion is based on the fact that the comendites show no significant vesicle concentration but Taylor et al. [41] have shown that vesicles can disappear upon compaction. It is highly unlikely that the magma which gave rise to the comendites contained as little as 0.4% water as suggested by Sheppard and Harris [40], if it results itself from the differentiation of a more mafic magma [3] which could originally contain at least half this amount of water [42,43]. With a differentiation degree of more than 80% (see below), the minimum initial amount of water in the magma at the comendite stage should be around 1%.

In fact, from our results on the whole suites of lavas and plutonic inclusions (and not only from five isolated results as in Sheppard and Harris [40]), the similarity between both suites is obvious and, whatever the initial state of the magma, it must be similar for both: if the initial water content can conceivably be mainly of mantle origin, it has probably been contaminated to various extents by water of oceanic origin, possibly up to the 8% announced by Harris (in [40]). Its δD should have been also much closer to zero. Then, in both lavas and plutonic inclusions, the δD range of -30 to -80‰ can be explained by various outgassing degrees, the outgassed water being enriched between 10 and 20‰ relative to the residual dissolved water [39].

In conclusion, the δD's and water contents of the lavas and plutonic inclusions support fully the idea that the two magmatic series derive from the same magma.

The δ^{18}O variations are correlated with a variety of geochemical parameters, among which:

—the ^{87}Sr/^{86}Sr ratios (Fig. 3a), themselves correlated with the ϵ_{Nd} values;

—the concentrations of LILE like Zr, Rb, La, etc. (Fig. 3b, c);

—the SiO_2 contents.

For all these correlations, there is the same distinction between the lavas whose variations in trace element cover the whole range and the plutonic inclusions, especially the cumulates, for

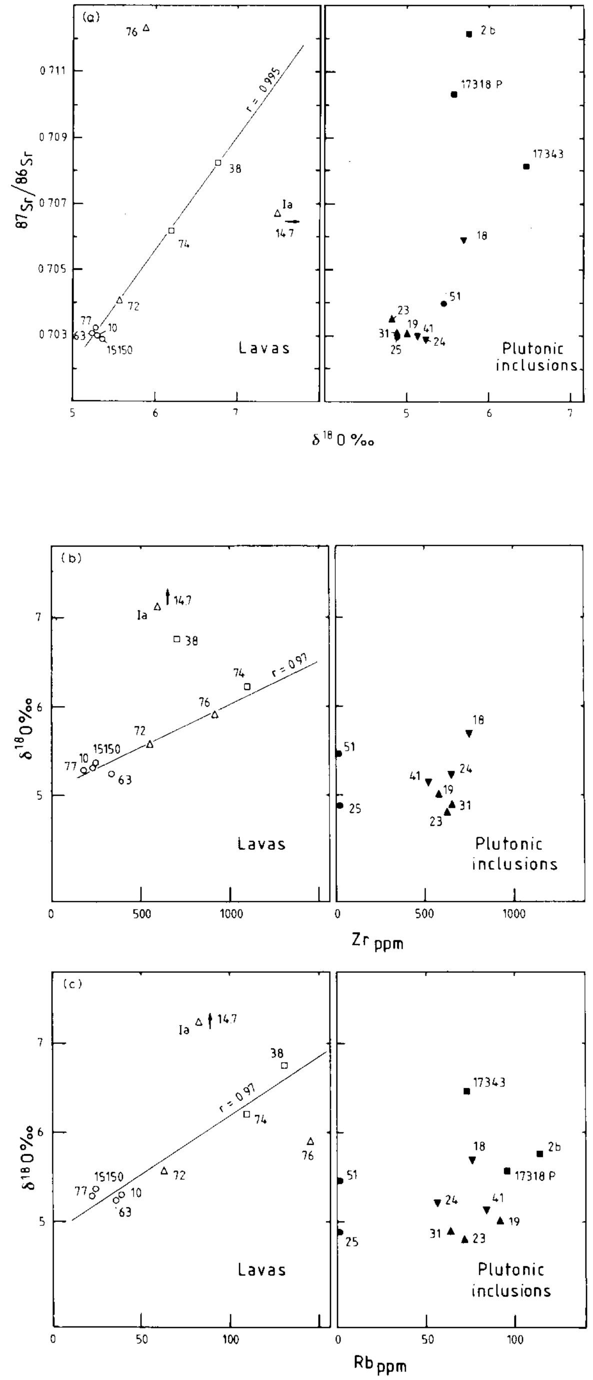

Fig. 3. (a) δ^{18}O vs. ^{87}Sr/^{86}Sr for Ascension Island rocks. Symbols as in Fig. 1. δ^{18}O vs. (b) Zr and (c) Rb (in ppm). Note the good correlation for the lavas while the variations in trace elements are much more restricted for the plutonic inclusions. Note, in general, the positive correlation of the δ^{18}O values with large-ion lithophile element contents as well as with ^{87}Sr/^{86}Sr ratios, i.e. with differentiation.

which the variations are much more restricted. These two groups of rocks will be discussed separately in the next two paragraphs concerning oxygen isotope systematics.

The correlations shown by the volcanic suite of rocks are readily explained by a fractional crystallization process (Fig. 4). Trace element modelling (J.G. Schilling, unpublished results) indicates that the extent of this differentiation reaches 80% of the original magma, from olivine basalt to alkali rhyolite (comendite). This is in agreement with the estimates of Harris [3] based on the least squares Wright and Doherty's [44] program for petrologic mixing. For olivine, clinopyroxene, plagioclase, Ti-magnetite and apatite, the mean $\delta^{18}O$ fractionation between magma and fractionating crystals would be +0.5‰. In all trace element diagrams, the most evolved lavas do not perfectly fit the correlations defined by the basic and intermediate terms of the volcanic suite. This is especially true for Rb which is very enriched (> 30%) and Zr, also enriched compared with the closed-system fractional crystallization model (Fig. 3b, c).

Considering now the plutonic inclusions; the variations in trace element contents are less regular and distinctly smaller, especially for the quartz monzodiorites. These latter rocks, typical plagioclase cumulates, show a very narrow range of chemical compositions for both major and trace elements and are characterized by $\delta^{18}O$ values lower than those observed in mantle derived rocks [28–31]: the mean $\delta^{18}O$ value is 4.90 ± 0.10‰ for the hypersolvus monzodiorites and 5.36 ± 0.35‰ for the subsolvus monzodiorites. The plagioclase accumulation in these rocks (as shown petrographically but also by their geochemical characteristics: high Ba, positive Eu anomaly, etc. (J.G. Schilling, personal communication)) can easily account for these low $\delta^{18}O$ values. Basic magmas are characterized by small positive plagioclase-magma fractionations (also depending on the An content) but it is also possible that positive magma-plagioclase isotopic fractionation occurs [45] for an acidic magma, as should be the case here. The interpretation is that if the temperature is above 700 °C, as H_2O is richer in ^{18}O than silicates, the magma which contains more water than the crystals is enriched in ^{18}O. The $\delta^{18}O$ values in Ascension samples are distinctly higher than those observed in Icelandic rocks [46]. Slight interaction with high-temperature magmatic waters cannot be entirely excluded in view of these values. On the one hand, there is no obvious reason why this process should have affected only the plutonic inclusions of intermediate compositions and not all these rocks, but on the other hand, as these inclusions have cumulate textures, they do not represent very closely the composition of the magma and can have been affected by additional processes.

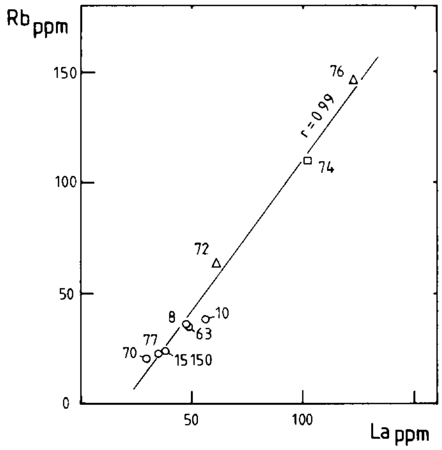

Fig. 4. Rb vs. La (in ppm) for Ascension Island lavas to show the linear fit of the data, indicating a fractional crystallization process.

It is not possible to quantify the fractional crystallization process for the plutonic suite in Ascension Island lavas because there are no sufficiently precise correlations in trace element diagrams. Nevertheless, as the geochemical characteristics of the plutonic inclusions fall within the range of those of the lavas and as they show similar variation trends, one can consider that they result from the same type of process, but with more complicated features because of the presence of crystals which can trap selectively some trace elements. The conclusions drawn for the lavas are then entirely applicable to the plutonic inclusions which can therefore be considered as their slowly cooled equivalents.

The purpose of this paper was not to discuss in detail the different explanations for the peculiar Sr isotopic compositions of the most evolved Ascension rocks. The same features were observed and discussed by Grant et al. [34] in the late-fractionated rocks of the alkali basalt-trachytephonolite suite of St. Helena and this discussion has already been presented for some Ascension samples by Harris et al. [14,17]. However, we will briefly review the different possibilities, as, in contrast to these previous publications, we can add oxygen data and the insight they can provide into that problem. Results of different calculations (similar in principle to those of Harris et al. [17]) have been reported in Table 2:

(1) First, the $^{87}Sr/^{86}Sr$ ratios were corrected for in-situ ^{87}Rb decay, assuming an age of 1.5 Ma for all the samples (which is as the maximum measured age of the lavas). These corrected values are only slightly lower than the measured ones, even for the most Rb-rich rocks and some of them still maintain values higher than the seawater ratio (0.70915).

(2) Assuming the age of 7 Ma, corresponding to the magnetic anomaly 4 [18] on which Ascension Island is situated, as the maximum age of the Island, the corrected Sr isotopic values for that time are still too high, up to 0.711.

(3) Third, the time necessary to reach the measured values starting from an initial $^{87}Sr/^{86}Sr$ ratio of 0.703 (mean value for the basic rocks) was calculated: the values range from 1.7 Ma for a comendite to 84.5 Ma for a trachyte and appear unrealistically high in view of the K-Ar age determinations and of the probable maximum age of Ascension Island. This rules out the possibility of a $^{87}Sr/^{86}Sr$ ratio increase within the magmatic chamber itself only as a result of chemical gradients developed in stratified magmatic chamber as was observed for the Bishop Tuff [35,36].

(4) The fourth calculation gives the time necessary for the most acidic rocks to reach the high Sr isotopic ratio starting from an initial ratio as high as that of the seawater $^{87}Sr/^{86}Sr$ value. With the exception of the trachyte Asc 76, two other samples need less than 3.3 Ma to reach these values, which is not incompatible with Ascension Island maximum age but which is nevertheless higher than the few K-Ar age determinations. It is to be noted, however, that these determinations were mostly obtained on intermediate and basic rocks [14,19], so that a priori one can not exclude the possibility that the acidic rocks crystallized earlier in the Ascension Island history and were later incorporated by the lavas. Moreover, only the top of a 3000–4000 m high volcanic cone has been sampled. However, both field and petrological arguments indicate the close genetic relationship between all the lavas forming a continuous differentiation suite from olivine basalt to comendite and between the lavas and the plutonic inclusions, except for some of the gabbroic inclusions of Dark Slope Crater ([14,16] and C. Harris, personal communication). The earlier crystallization of the acidic rocks appears then very unrealistic.

Consequently, one must consider that a combination of more than one of the following processes has played a role in the genesis of the most evolved Ascension Island rocks. A priori, three different processes may have acted in parallel or in addition to the in-situ Rb decay:

(1) Deep-sea sediment incorporation,

(2) Seawater contamination or interaction,

(3) Mixing with altered oceanic crust.

The first possibility which has also been proposed by Harris et al. [14,17] is not compatible with all the available isotopic data. Indeed, in that case, high $\delta^{18}O$ values should be observed because the sediments have much higher $\delta^{18}O$ values (as high as +25‰) [45–47]. The incorporation of only 5% of a sediment with a $\delta^{18}O$ value of 25‰ is sufficient to enhance the $\delta^{18}O$ value magmatic rock by 1‰. More radiogenic Pb isotopic compositions, in both $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios, would also be observed [48–50] as well as lower ϵ_{Nd} values (metalliferous sediments and Mn nodules are characterized by negative ϵ_{Nd} values [51,52].

The seawater contamination hypothesis has the advantage of not affecting the Pb and Nd isotopic systems because of the relatively low contents of both Nd and Pb in seawater. In that case, the oxygen isotopic system would show the following possible variations: lower $\delta^{18}O$ values in the case of high-temperature seawater interaction (> 400 °C [53]) or higher $\delta^{18}O$ values in the case of low-temperature processes ($\leqslant$ 400 °C). Since Ascension Island samples do not show any petrographic indication of either high- or low-temperature interaction, at least at an important scale, the seawater interaction hypothesis can also be rejected.

The third hypothesis, that is mixing with altered oceanic rocks (or older altered basic Ascension rocks as they have isotopic features, especially Nd and Sr, in the range of MORB values) appears the most plausible. Pb and Nd isotopes will not be affected significantly by this process because there is no significant modification of Nd and Pb isotopic composition by alteration. The incorporation of high-temperature altered oceanic crust would have lowered the $\delta^{18}O$ values, as observed in Icelandic volcanics (variable and often low $\delta^{18}O$ values [54]). For Ascension Island rocks, this could a priori imply that the process occurred under relatively high-temperature conditions. A very clear relation between Sr concentrations and $^{87}Sr/^{86}Sr$ values is shown in Fig. 1b, that is constant isotopic ratios down to 5 ppm Sr and very steep increase of the $^{87}Sr/^{86}Sr$ ratios below that level. Comparable correlations also exist in the Rb/Sr or Rb versus $^{87}Sr/^{86}Sr$ diagrams. In the $^{87}Sr/^{86}Sr$ versus 1/Sr diagram, the Ascension acidic rocks define linear trends which indicate a $^{87}Sr/^{86}Sr$ contaminant ratio higher than 0.7075. It is important to underline the fact that, considering the very low Sr contents (< 5 ppm) of the high $^{87}Sr/^{86}Sr$ acidic rocks, the incorporation of less than 1‰ of altered oceanic crust (or older Ascension basic rocks—with Sr content around 100 ppm [5] and an $^{87}Sr/^{86}Sr$ ratio of 0.709) can account for the observed Sr isotopic ratios.

The mass balance calculations also show that the incorporation of 1% of altered crust with a $\delta^{18}O$ of 0.5‰ would only affect the $\delta^{18}O$ value of the rocks by 0.05‰, that is below the detection limit. If the $\delta^{18}O$ of the altered crust is 3‰, the corresponding shift would be 0.02‰. It explains also why no significant modifications are observed in the major or trace element contents.

If Pb and eventually Sr isotopic compositions could not by themselves entirely exclude a continental origin of the acidic rocks, such an origin would necessitate important re-equilibration processes between a plutonic inclusion and its host lava, for which there is no evidence. Indeed, if on the average, the plutonic inclusions and the lavas have very similar isotopic characteristics, there is not one to one correspondence between one inclusion and its host lava. Moreover, the positive ϵ_{Nd} values obtained for Ascension Island rocks, including the acidic terms, both plutonic and volcanic, argue strongly against any continental crust influence and/or contamination by oceanic sediments. Oxygen adds further evidence for a direct mantle origin of the acidic rocks. Hence it appears that $\delta^{18}O$ values of acidic rocks resulting from fractional crystallization alone are not greater than 6.7‰ (this work, [28,41]). Ascension Island peralkaline granites but also the rhyolites and comendites confirm that the $\delta^{18}O$ enrichment resulting from fractional crystallization is not greater than 1.5‰ relative to the initial value of the basic source. Moreover, the only acidic sample found on the moon (sample 12013) has $\delta^{18}O$ values between 5.95 and 6.34‰ [56], i.e. in the range of the lunar basaltic rocks.

Values higher than 7‰ [57], which correspond to a very large proportion of granites in continental environment, must be explained by other processes. If, as shown by Fourcade and Javoy [58], the $\delta^{18}O$ values of very old (> 3 Ga) granulitic rocks (from the Precambrian shield of In Ouzzal, Sahara) are low and homogeneous and can result from magmatic differentiation alone, the building of continental crust since then must have involved more and more addition of high ^{18}O oxygen from sedimentary or metamorphic rocks.

6. Source of the parental magma

All the data on Ascension Island together with those of O'Nions et al. [8,9] are reported in the Nd-Sr diagram (Fig. 5a). For comparison, some data on other oceanic rocks [12,55,59–63] were also reported. The comparison reveals several features worthy of comment:

(1) The Ascension Island data fall largely within the Nd-Sr mantle array, i.e. within the negative correlation observed for many oceanic rocks, either mid-oceanic ridge basalts (MORB) or oceanic island basalts (OIB). However, some islands fall distinctly off the array, such as St. Helena for example [4,12].

(2) The Ascension Island data fall in the upper part of this mantle array, characterized by the lowest $^{87}Sr/^{86}Sr$ ratios (< 0.7035) and the highest $^{143}Nd/^{144}Nd$ ratios ($\epsilon_{Nd} > +7$). They largely overlap the MORB field (particularly the Mid-Atlantic Ridge field [64,65]), which is quite unusual for oceanic island rocks. This implies that the source region of this alkali basalt differentiation suite is more strongly depleted in LILE than many other OIB sources.

(3) In detail, although within the mantle array, the $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ ratios in the

Ascension Island rocks appear positively correlated (Fig. 5b), while the intra-island isotopic variations in particular (i.e. the Azores) and the mantle array in general have Nd-Sr trends with negative slopes. The very steep positive slope for basic and intermediate rocks of Ascension Island is indicative of a much larger relative variation in Nd than in Sr isotopic ratios (Fig. 5b). The Ascension Island trend points towards the St. Helena group of data [12].

(4) The Nd and/or Sr isotopic compositions are not correlated with the Pb isotopic data [16]. The existence of two groups of rocks in the Ascension Island samples (the gabbroic inclusions and the basaltic tuff Asc77 of tholeiitic affinity on the one hand and all the other rocks, either volcanic or plutonic, on the other) as defined by the Pb isotopic compositions [14,16] is not shown by these two other isotopic systems.

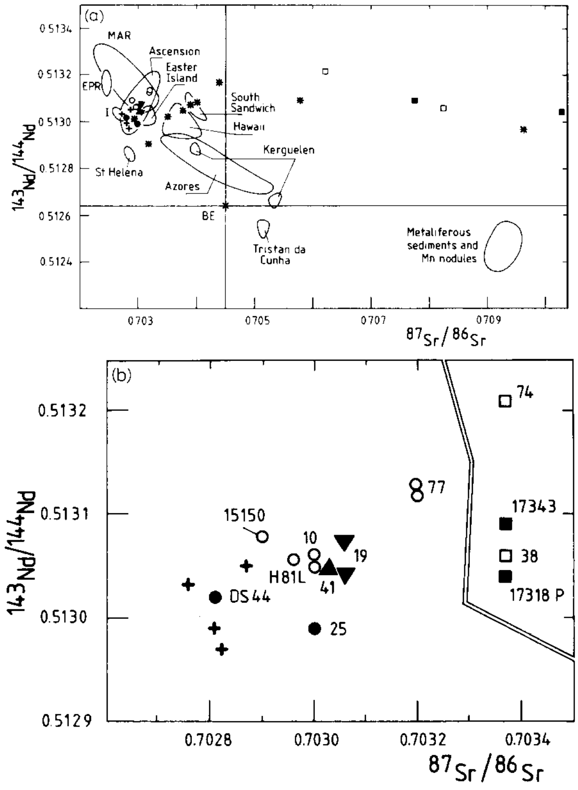

Fig. 5. (a) $^{143}Nd/^{144}Nd$ vs. $^{87}Sr/^{86}Sr$ ratios for Ascension Island lavas and plutonic inclusions. Symbols as in Fig. 1. Comparison with other data on oceanic basalts, either MORB or OIB [4,5,9,12,59–62] and on sediments [51,52,63]. Altered basalts are represented by the asterisk [63]. (b) Same diagram, but with an expanded scale to show the positive correlation of these ratios amongst Ascension Island rocks.

The observed trends in the Nd-Sr and Pb-Pb diagrams are probably best explained in terms of mixing between two end-members which have to be defined. One of the components involved has geochemical features comparable to those of MORB source; i.e., $\epsilon_{Nd} \simeq +10$; $^{87}Sr/^{86}Sr \simeq 0.7030$; $^{206}Pb/^{204}Pb \simeq 18$; $^{207}Pb/^{204}Pb \simeq 15.55–15.60$ and $^{208}Pb/^{204}Pb \simeq 38.2$. On the other hand, a component with lower $^{143}Nd/^{144}Nd$ ratios ($\epsilon_{Nd} < 10$) but with about the same or slightly lower $^{87}Sr/^{86}Sr$ ratios is needed. This source might have geochemical characteristics close to those of St. Helena (but with less extreme isotopic compositions). In the Pb-Pb diagram, the same situation is observed: one of the two groups of samples defined by Weis [16] which comprised most of the Ascension Island samples has indeed Pb isotopic ratios intermediate between those of MORB [50,65,66] and those of St. Helena Island [66,67]. It is to be noted, as also recognized by White [7], that the origin of such St. Helena-type source still remains enigmatic.

One cannot explain the peculiar features of the isotopic data of Ascension Island by the incorporation of oceanic sediments. In that case, one should expect a shift of the data to higher $^{87}Sr/^{86}Sr$ ratios, a trend with a negative slope in the Nd-Sr diagram and distinctly higher $^{207}Pb/^{204}Pb$ ratios. The isotopic data discussed above suggest that material from at least two different sources have been involved in the genesis of Ascension Island rocks. In this context, a general model for the genesis of Ascension Island could be established in the following way: when the island was closer than now to the Mid-Atlantic Ridge, an early magmatic episode of tholeiitic nature generated the gabbroic inclusions and the basaltic tuff 77 (coming from Dark Slope Crater, known to be the earliest active center of Ascension Island [68] and from Middelton's Ridge). With time, the spreading activity of the Mid-Atlantic Ridge continuing, the island was progressively remote from the ridge, so that the hotspot component gained increasingly more importance and was responsible for the genesis of the alkali basalt differentiation suite with its intermediate and acidic rocks, both volcanic and plutonic.

This model has several advantages in explaining the isotopic heterogeneities within Ascension Island lavas, especially in the Pb isotopic system, and interestingly confirms the model recently proposed by Hamelin et al. [65] to explain the isotopic variations observed in the northern segment of the Mid-Atlantic Ridge.

Dupré and Allègre [6] and Dupré [69] have recently shown the importance of mixing processes between the lower and upper zones of the mantle. The Ascension Island data are in agreement with this process. Nevertheless, the Ascension Nd-Sr-Pb isotopic compositions are intermediate between those observed for the two large-scale mantle domains defined for the oceanic island and continental alkali basalt source (the lower mantle). This is not surprising as Ascension Island is geographically located close to the boundary of the two domains (see fig. 3 in Dupré and Allègre [6]), the South Atlantic and southern Indian Ocean islands on the one hand and the North Atlantic and eastern Pacific Ocean islands together with the continental alkali basalts from Africa (Kenya and Hoggar) on the other hand.

7. Conclusions

(1) The $\delta^{18}O$ values in Ascension Island lavas and plutonic inclusions range from 4.8‰ for the plagioclase cumulates to 6.7‰ for the most evolved rocks, i.e. granite and alkaline rhyolite. The basic rocks have values around 5.3‰, characteristic of mantle-derived material.

(2) The mean of $^{87}Sr/^{86}Sr$ ratios for eighteen basic rocks is 0.70311 ± 17 whereas the late-stage differentiated rocks (rhyolites, comendites and alkaline granitic inclusions) have ratios up to 0.712. These high $^{87}Sr/^{86}Sr$ ratios result from the combination of two processes which have acted in parallel: the incorporation of small amounts ($< 1\%$) of high-temperature altered oceanic crust (or older altered Ascension basic rocks) enhancing the $^{87}Sr/^{86}Sr$ ratio of the magma in the late stages of the differentiation process followed by the in-situ Rb decay since the time of formation of the rocks. This latter process is particularly effective because of the high Rb enrichment of this alkaline magma.

(3) The Ascension Island lavas and plutonic inclusions have indistinguishable Nd, Sr and H isotopic compositions, implying a common origin for their parental magma. There is no evidence for continental crust involvement in the genesis of the acidic rocks; the plutonic inclusions have an oceanic origin and can be considered as the slowly cooled equivalents of the lavas. The positive ϵ_{Nd} values for both volcanics and plutonic inclusions suggest also a time-integrated REE-depleted mantle source.

(4) Pb-Sr-Nd isotopic systematics point to a mixing between a MORB-type, upper mantle component and a plume-type (OIB), lower mantle component with relatively radiogenic Pb and unradiogenic Nd and Sr (i.e. with geochemical characteristics quite comparable to those of St. Helena rocks).

(5) The oxygen isotope systematics in these samples have implications for the application of isotopic geochemistry to granite petrogenesis:

(a) the $\delta^{18}O$ values of acidic rocks resulting from fractional crystallization processes alone are not greater than 6.7‰ implying a $\delta^{18}O$ enrichment relative to the parental basic magma not greater than 1.5‰. Higher enrichments usually observed for granites in continental environment imply necessarily other processes, such as alteration and/or crustal contamination;

(b) one must be very careful when utilizing the initial Sr ratio of alkaline anorogenic complexes to define the source region of the parental magma [70,71] as relatively high $^{87}Sr/^{86}Sr$ values for granitic rocks do not necessarily reflect a continental origin.

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