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Rb-Sr, Sm-Nd and Ar-Ar isotopic systematics of Antarctic nakhlite Yamato 000593

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Abstract: Isotopic analysis of the newly found Antarctic nakhlite Yamato (Y) 000593 yields a Rb-Sr age of 1.30 ± 0.02 Ga with an initial $^{87}Sr/^{86}Sr$ of 0.702525 ± 0.000027, a Sm-Nd age of 1.31 ± 0.03 Ga with an initial ε_{Nd} of $+16.0 \pm 0.2$ and an Ar-Ar isochron age of ≤ 1.36 Ga. The concordancy of these three ages and Rb-Sr and Sm-Nd initial isotopic signatures strongly suggest that Y000593 crystallized from low Rb/Sr, light REE-depleted source materials ~1.31 Ga ago. The crystallization age of Y000593 is compared with the age data of non-Antarctic nakhlites (Nakhla, Governador Valadares, Lafayette and Northwest Africa 998) and Chassigny. The initial Sr and Nd isotopic signatures suggest that Lafayette and Y000593 were co-magmatic or at least came from very similar magmas. Cosmogenic ^{36}Ar concentrations in Y000593 resemble those in other nakhlites. The similarities in crystallization and ejection ages and in petrologic features suggest the nakhlites were derived from similar source regions, and launch pairing of nakhlites and Chassigny.

The Rb-Sr data for Y000593 show that the isotopic system is disturbed by preterrestrial alteration of olivine. Although many of the acid-leached residues of mineral fractions fall along the 1.30 Ga Rb-Sr isochron, leached olivine does not. This indicates the lack of isotopic equilibrium between the olivine fractions and the secondary alteration phases. A tie-line between two olivine leachates provides a calculated "age" of 650 ± 80 Ma with an initial $^{87}Sr/^{86}Sr$ of ~0.70465, which gives a hint for the isotopic signatures of local brine as well as the timing of an aqueous alteration event on the Martian surface.

key words: Yamato 000593, nakhlite, Ar-Ar, Rb-Sr, Sm-Nd, chronology, crystallization, aqueous alteration, source material

1. Introduction

Nakhlites are unbrecciated, olivine-bearing clinopyroxenites that probably came from Mars (*e.g.*, McSween, 1994). A total of seven nakhlites have been identified, including four recent finds: Northwest Africa (hereafter NWA) 817 (104 g) and NWA 998 (456 g) from the hot desert of Morocco (Sautter *et al.*, 2002; Irving *et al.*, 2002),

† Deceased on January 9, 2004. His technical excellence will be missed by all coworkers.

three samples (Y000593, Y000749 and Y000802) from the Yamato Mountains areas of Antarctica (Kojima and Imae, 2001; Imae *et al*., 2002; Kojima *et al*., 2002; Misawa *et al*., 2003b) and Miller Range 03346 (715 g) from the Transantarctic Mountains of Antarctica (Satterwhite and Righter, 2004). Three Yamato samples, a total weight of ~15 kg, were collected in a small area within 1 km^2, suggesting that they are paired (Misawa *et al*., 2003b). Nakhlites have been subjected to lower peak shock pressures than other Martian meteorites (*e.g*., shergottites) and their original igneous textures are still well-preserved. Also, these meteorites have much older crystallization ages of ~1.3 Ga compared to ages of shergottites of ~0.18–0.47 Ga (*e.g*., Nyquist *et al*., 2001). The Yamato nakhlites contain pre-terrestrial low-temperature aqueous alteration materials, which provide information about the timing and nature of aqueous alteration on Mars (Bridges *et al*., 2003; Noguchi *et al*., 2003). In this report, we present Rb-Sr, Sm-Nd and Ar-Ar isotopic systematics for the largest Yamato nakhlite Y000593 and discuss the age correlation with other nakhlites, the nature of their source materials and the timing of formation of weathering products on Mars. Preliminary isotopic results for Y000593 have been given by Shih *et al*. (2002) and by Misawa *et al*. (2003a, c).

2. Samples and analytical procedures

Yamato 000593 is a coarse-grained rock and is extremely friable. The sample studied, Y000593,56, weighed ~2.3 g. One medium-sized fragment, plus fines, weighing ~0.5 g, was processed by gently crushing to grain size $<149\,\mu$m. This sample was coned and about half of the crushed material was taken as whole-rock samples (WR1 and WR). The rest of this sample and one large-sized fragment, weighing 1.4 g, were crushed and sieved into two size fractions, 149–74 μm and $<74\,\mu$m. Mineral separations were made from the coarser fraction using a Frantz isodynamic magnetic separator. At 0.5 ampere, we obtained a non-magnetic (NMag) sample, mainly composed of mesostasis materials including plagioclase. At 0.2 ampere, we obtained a clinopyroxene (Cpx) sample from the less-magnetic fraction. A high-purity clinopyroxene (Cpx2) sample was obtained by handpicking. From the more-magnetic fraction, two olivine samples were obtained by density separation using heavy liquids (Clerici's solutions) of 3.6 g/cm^3 and 3.95 g/cm^3. An olivine (Ol) sample was handpicked from the 3.6–3.95 g/cm^3 fraction and consisted of yellow olivine grains with dark inclusions (Fig. 1). The other olivine sample ($\rho>3.95$) was handpicked from the >3.95 g/cm^3 fraction and displayed adhering brown alteration products, which previously have been referred to as "iddingsite". The WR, Cpx, Cpx2, NMag, $\rho>3.95$ and Ol samples were washed with 2N HCl in an ultrasonic bath for 10 min to eliminate possible terrestrial contamination, if it exists, and Martian alteration products. Both the residues (r) and leachates (l) of these samples, plus unleached sample WR1, were analyzed for Rb, Sr, Sm and Nd following the procedures of Shih *et al*. (1999). The isotopic measurements were made on a Finnigan-MAT 261 multi-collector mass spectrometer following the procedures of Nyquist *et al*. (1994). The average values of $^{87}Sr/^{86}Sr$ for NBS 987 during the course of the study were 0.710237 ± 0.000014 ($2\sigma_p$, 6 analyses) and 0.710233 ±0.000026 ($2\sigma_p$, 14 analyses), normalized to $^{88}Sr/^{86}Sr=8.37521$. The $^{87}Sr/^{86}Sr$ results reported here were renormalized to the NBS 987 $^{87}Sr/^{86}Sr=0.710250$ (Nyquist *et al*.,

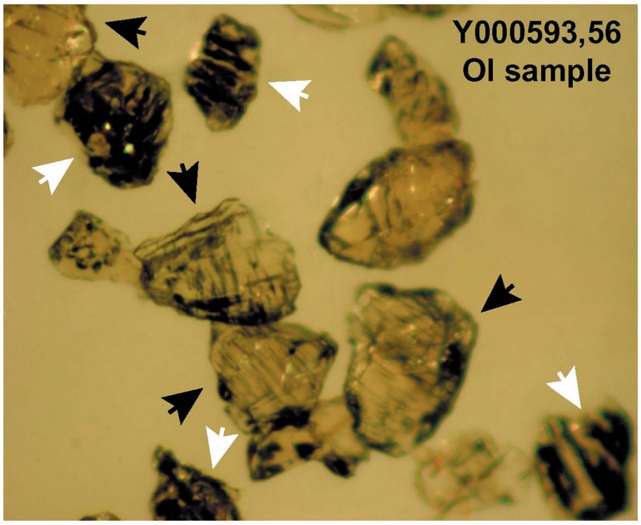

Fig. 1. Two olivine samples were obtained by density separation using heavy liquids (Clerici's solutions) of 3.60 and 3.95 g/cm³ (cf. Fig. 3). One (Ol) was handpicked from the 3.60–3.95 g/cm³ fraction and consisted of yellow olivines with lamellar inclusions (black arrows) as shown in the photograph. The other (>3.95 in Fig. 3) was handpicked from the >3.95 g/cm³ fraction and displayed adhering brown alteration products, which previously have been referred to as "iddingsite". Brownish alteration products (white arrows) are much less abundant in the Ol sample compared to the >3.95 sample. The field of views are ~0.6 mm.

1990). Because of the low Sm and Nd contents of the samples, Sm and Nd isotopes were measured as SmO^+ and NdO^+. The oxide-corrected $^{143}Nd/^{144}Nd$ values for standards are given in Table 2. The $^{143}Nd/^{144}Nd$ results for samples reported here were renormalized to $^{143}Nd/^{144}Nd = 0.511138$ for the Caltech Nd standard $n(\mathrm{Nd})\beta$ (Wasserburg *et al.*, 1981).

A 46.7 mg whole-rock sample from Y000593,56 was irradiated with fast neutrons to measure its ^{39}Ar-^{40}Ar age. The irradiation constant of $J = 0.02565 \pm 0.00012$ was determined from several NL-25 hornblende samples irradiated with the nakhlite sample. Argon was extracted from Y000593 by stepwise temperature release, and its isotopic composition was measured on a mass spectrometer. Additional experimental details were given by Bogard *et al.* (2000).

In order to investigate alteration effects on olivine, grains were handpicked from the separated olivine fractions and were imbedded in epoxy resin. The grain mount was polished and carbon-coated, and "iddingsite" veins in olivine were observed by back-scattered electron images using a scanning electron microprobe, model JEOL JSM5300 LV, at NIPR. Elemental distribution maps were taken by using K-α lines.

3. Results and discussion

3.1. Rubidium-strontium isotopic systematics

The results of Rb-Sr isotopic analysis for Y000593 are presented in Table 1. The Rb and Sr concentrations for four nakhlites are summarized in Fig. 2. Nakhla and Governador Valadares have similarly higher Rb and Sr contents than Lafayette, probably due to a higher olivine content in Lafayette. The Antarctic nakhlite Y000593 has similar Sr abundance as Nakhla and Governador Valadares, but among the nakhlites Y000593 has an intermediate Rb abundance. The near constancy of Rb and Sr in acid-leached and unleached WR samples, WR1 and WR(r), strongly suggests that the terrestrial weathering effects on Rb and Sr are less significant for this Antarctic nakhlite than for hot-desert Martian meteorites previously studied in the JSC lab. The Sr concentrations in the leachates from the Ol and ρ>3.95 samples are 5.6- and 2.4-times,

Table 1. The Rb-Sr analytical results for Y000593.

Sample[a]	wt (mg)	Rb (ppm)	Sr (ppm)	$^{87}Rb/^{86}Sr$[b]	$^{87}Sr/^{86}Sr$[c]
WR1	26.35	2.38	58.9	0.1172±14	0.704688±19
WR					
residue	26.30	2.44	61.9	0.1139±13	0.704633±11
leachate	3.90	2.00	33.0	0.1774±22	0.705506±12
Cpx					
residue	23.75	0.609	32.6	0.05407±56	0.703528±16
leachate	3.00	0.830	12.6	0.1895±23	0.705297±18
NBS 987 Sr standard: Sr^+ (6 analyses)[d]					0.710237±14[e]
Cpx2					
residue	17.51	0.147	22.4	0.01902±22	0.702886±30
leachate	0.57	1.38	18.8	0.2119±29	0.706032±26
NMag					
residue	1.45	18.3	752	0.07045±82	0.703790±18
leachate	0.46	14.5	275	0.1530±17	0.704699±21
Ol					
residue	7.58	0.0246	0.189	0.377±12	0.706650±21
leachate	1.64	0.0580	1.06	0.1585±86	0.706097±26
ρ>3.95					
residue	8.31	0.0536	1.25	0.1244±25	0.704465±11
leachate	6.70	0.0830	2.93	0.0823±15	0.705399±17
NBS 987 Sr standard: Sr^+ (14 analyses)[d]					0.710233±26[e]

[a] WR=whole-rock, Cpx=clinopyroxene, NMag=non-magnetic fraction, Ol=olivine, ρ>3.95=heavier olivine fraction.
[b] Uncertainties correspond to last figures and represent $\pm 2\sigma_m$ error limits.
[c] Normalized to $^{88}Sr/^{86}Sr$=8.37521 and adjusted to $^{87}Sr/^{86}Sr$=0.710250 of the NBS 987 Sr standard (Nyquist *et al*., 1990). Uncertainties correspond to last figures and represent the $\pm 2\sigma_m$ error limits.
[d] Sr standard data were obtained in different analytical sessions.
[e] Uncertainties correspond to last figures and represent $\pm 2\sigma_p$ error limits.

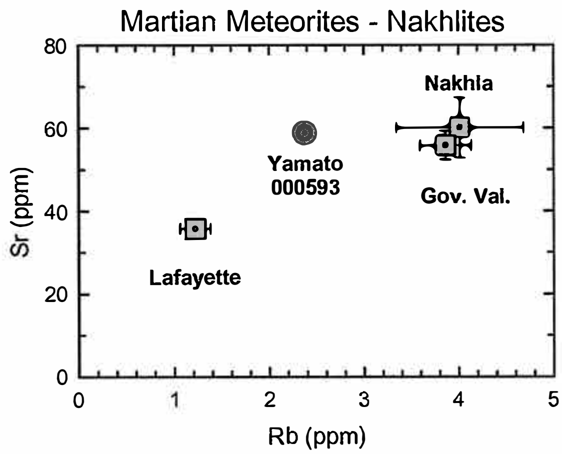

Fig. 2. Rubidium versus strontium abundance plot for nakhlites. Antarctic nakhlite Y000593 (circle) is the unleached WR1 sample from this study. Literature data for non-Antarctic nakhlites are shown by squares. Y000593 has the same Sr abundance as Nakhla and Governador Valadares but its Rb abundance is significantly lower than that in those two nakhlites. Lafayette has the lowest Rb and Sr among all nakhlites, perhaps due to its higher olivine and lower mesostasis content.

respectively, higher than those in their respective residues. This probably is related to the dissolution of minor aqueous alteration products (*e.g.*, "iddingsite" and carbonates) in the samples by the acid treatments.

The $^{87}Rb/^{86}Sr$ and $^{87}Sr/^{86}Sr$ data of one unleached whole-rock sample, WR1, and residues and leachates from WR, Cpx, Cpx2, NMag, $\rho > 3.95$ and Ol are shown in Fig. 3. If all these samples are included, they do not define a single linear array. However, samples WR1, WR(r), Cpx(r), Cpx2(r) and NMag(r) form a line in the $^{87}Rb/^{86}Sr$ and $^{87}Sr/^{86}Sr$ correlation diagram. These five data points yield an Rb-Sr age of 1.30 ± 0.02 Ga for $\lambda(^{87}Rb) = 0.01402\,Ga^{-1}$ (Minster *et al.*, 1982) with an initial $^{87}Sr/^{86}Sr$ of 0.702525 ± 0.000027 using the Williamson (1968) regression program. This Rb-Sr isochron age for Y000593 is in agreement with radiometric ages determined previously by various methods for four other nakhlites and Chassigny (see, Nyquist *et al.*, 2001; Carlson and Irving, 2004; Nakamura *et al.*, 1982b).

Data for the four leachates, WR(l), Cpx(l), Cpx2(l) and NMag(l) lie to the right of the primary Rb-Sr isochron. On the other hand, the olivine leachates, $\rho > 3.95$(l) and Ol(l), lie to the left of the isochron. These deviations are probably due to the presence of secondary aqueous alteration products in Y000593. This secondary alteration effect has been previously reported for Governador Valadares and Lafayette (Shih *et al.*, 1998, 1999). The leachate samples are more radiogenic and have higher Rb/Sr ratios compared to the corresponding residue samples, except for the olivine leachates: the olivine leachates are less radiogenic than the olivine residues and $\rho > 3.95$(l) shows a lower Rb/Sr signature compared to $\rho > 3.95$(r). Treatment of the olivine samples, $\rho > 3.95$ and Ol, with 2N HCl dissolved 45% and 18%, respectively, by weight and

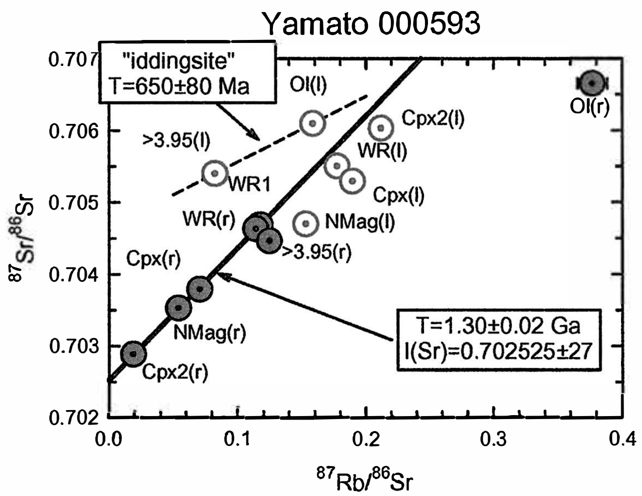

Fig. 3. Rubidium-strontium isochron diagram for Y000593. The data were fitted by the Williamson (1968) regression program and the isochron ages were calculated for $\lambda(^{87}Rb)=0.01402\,Ga^{-1}$ (Minster et al., 1982). Five data points, WR1, WR(r), Cpx(r), Cpx2(r) and NMag(r), yield an Rb-Sr age of 1.30 ± 0.02 Ga with an initial $^{87}Sr/^{86}Sr$ of 0.702525 ± 0.000027. This age for Y000593 is in agreement with radiometric ages determined previously by various methods for four other nakhlites and Chassigny (see, Nyquist et al., 2001; Carlson and Irving, 2004). Leachates of olivine, Ol(l) and $\rho>3.95$(l), define a possible secondary "iddingsite" isochron age of 650 ± 80 Ma with an initial $^{87}Sr/^{86}Sr$ of 0.70465 ± 0.00010 (see text). This age and initial Sr isotopic composition are similar to values found previously for Lafayette (Shih et al., 1998).

seemed to have effectively removed surficial alteration products. Calculated total contents of Rb from the combined data of the olivine leachate and residue samples are one order of magnitude lower than the Rb contents in the olivine separates from Nakhla (Gale *et al.*, 1975), which indicates that our olivine samples do not contain large amounts of alkali elements, even though the olivine sample, $\rho>3.95$, was associated with reddish-brown "iddingsite".

Figure 4 shows ages versus initial Sr isotopic compositions of nakhlites. The initial $^{87}Sr/^{86}Sr$ for Y000593 is within error limits of that of Lafayette (Shih *et al.*, 1998) and Chassigny (Nakamura *et al.*, 1982b) but slightly different from that of Nakhla (Papanastassiou and Wasserburg, 1974; Gale *et al.*, 1975) and Governador Valadares (Shih *et al.*, 1999). This suggests that Lafayette and Y000593 are co-magmatic. The slight discrepancy in the initial $^{87}Sr/^{86}Sr$ in nakhlites may be attributed to sampling of different flows of the same clinopyroxenite-dunite lithology on Mars. The low initial $^{87}Sr/^{86}Sr$ ratios of Y000593 and the other nakhlites suggest derivation from a low Rb/Sr mantle source region. There are small age differences among nakhlites and Chassigny, which could be partly due to aqueous alteration on the Martian surface (Nyquist *et al.*, 2001).

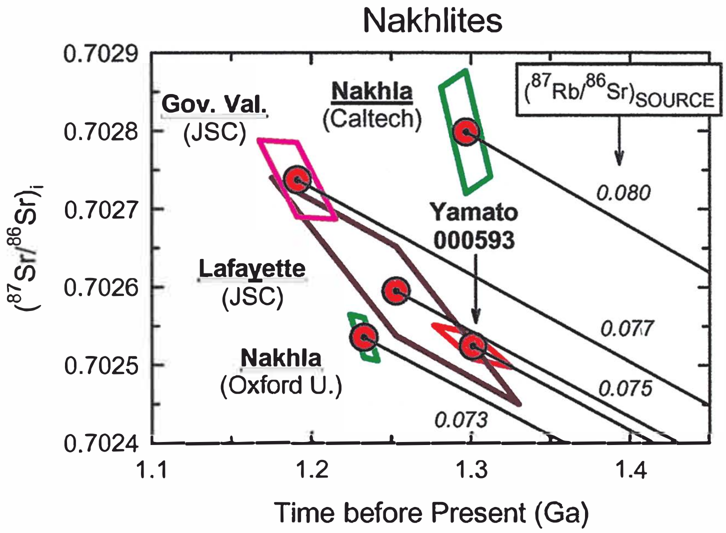

Fig. 4. Plot of ages versus initial $^{87}Sr/^{86}Sr$ ratios. Individual nakhlites are represented by parallelograms constructed from their age and initial $^{87}Sr/^{86}Sr$ parameters. The data show that they all crystallized within ± 0.1 Ga, and have initial $^{87}Sr/^{86}Sr$ ratios that vary by only 4 ε-units. Not all the nakhlite data overlap, suggesting that the nakhlites did not come from identical mantle sources, and/or they did not form at exactly the same time. However, single-stage model calculation indicates that Y000593 and Lafayette can be derived from a single source with $^{87}Rb/^{86}Sr = 0.075$. Governador Valadares and Nakhla may have been derived from slightly different sources. The calculated $^{87}Rb/^{86}Sr$ ratios required for single-stage evolution of the nakhlite sources are represented by the evolution lines. The small variations in age and initial $^{87}Sr/^{86}Sr$ ratios suggest that nakhlites represent different flows from very similar mantle sources. Data are from Papanastassiou and Wasserburg (1974), Gale et al. (1975), Shih et al. (1998, 1999) and this work.

3. 2. *Samarium-neodymium isotopic systematics*

The results of Sm-Nd isotopic analyses are presented in Table 2. The Sm and Nd concentrations of whole-rock samples of Y000593 and other bulk nakhlite samples are shown in Fig. 5. Yamato 000593 and Nakhla have distinctly higher Sm and Nd than Lafayette and somewhat higher Sm and Nd abundances than Governador Valadares. This is also consistent with high abundance of olivine, in which REE occur in low abundance, in Lafayette. The very high Sm and Nd abundances in acid leachates of Y000593 whole-rock and Cpx samples clearly suggest the presence of chlorapatite, a REE-carrier phase commonly found in nakhlites (Wadhwa and Crozaz, 1995), in Y000593.

The Sm-Nd isochron of nakhlite Y000593 is presented in Fig. 6. If all samples are included, they do not define a single linear array. However, six data points, including two leachates, WR(l) and Cpx(l), define a linear array corresponding to a Sm-Nd age of 1.31 ± 0.03 Ga for λ (^{147}Sm) $= 0.00654$ Ga^{-1} using the Williamson (1968) regression program. It is not clear why the acid residues of NMag and Ol deviate upward from the 1.31 Ga isochron (the Ol residue sample contains < 0.064 ng Sm and < 0.14 ng Nd);

the offset of these data and the leachate $\rho > 3.95(l)$ from the 1.31 Ga line is suggestive of disturbance.

The Sm-Nd age of 1.31 ± 0.03 Ga is in excellent agreement with the Rb-Sr age obtained from the same samples. The concordancy of Sm-Nd and Rb-Sr ages strongly suggests that Y000593 crystallized 1.31 Ga ago. Thus, the age datum for this new Antarctic nakhlite provides an additional evidence that nakhlites, and probably Chassigny as well, crystallized within a short period of time ~1.3 Ga ago. The initial ε_{Nd} value corresponding to the age for Y000593 is $+16.0 \pm 0.2$, which is close to that of Lafayette, but not within error limits of those reported for Nakhla and Governador Valadares (Fig. 7) and Chassigny (Jagoutz, 1996). In a similar manner to the initial $^{87}Sr/^{86}Sr$ of Lafayette and Y000593, the initial ε_{Nd} values of these rocks suggest that they were co-magmatic or at least came from very similar magmas. Nakhla and

Table 2. The Sm-Nd analytical results for Y000593.

Sample[a]	wt (mg)	Sm (ppm)	Nd (ppm)	$^{147}Sm/^{144}Nd$[b]	$^{143}Nd/^{144}Nd$[c]
WR1	26.35	0.889	3.82	0.14053±14	0.512168±10
WR					
residue	26.30	0.535	1.73	0.18661±19	0.512576±10
leachate	3.90	3.14	17.3	0.10980±12	0.511896±10
Cpx					
residue	23.75	0.558	1.78	0.18947±20	0.512574±10
leachate	3.00	1.16	6.36	0.11024±16	0.511912±10
Ames Nd standard: NdO^+ (5 analyses)[d]					0.511105±19[e]
Cpx2					
residue	17.51	0.472	1.49	0.19190±20	0.512619±12
leachate[f]	0.57				
NMag					
residue	1.45	0.307	1.40	0.13271±83	0.512196±11
leachate[f]	0.46				
Ol					
residue	7.58	0.00840	0.0184	0.276±15	(0.51356±13)
leachate[f]	1.64				
$\rho > 3.95$					
residue[f]	8.31				
leachate	6.70	0.103	0.540	0.11486±47	0.512038±15
Ames Nd standard: NdO^+ (22 analyses)[d]					0.511094±18[e]
Ames Nd standard: NdO^+ (6 analyses)[d]					0.511092±34[e]

[a] WR = whole-rock, Cpx = clinopyroxene, NMag = non-magnetic fraction, Ol = olivine, $\rho > 3.95$ = heavier olivine fraction.
[b] Uncertainties correspond to last figures and represent $\pm 2\sigma_m$ error limits.
[c] Normalized to $^{146}Nd/^{144}Nd = 0.724140$ and adjusted to $^{143}Nd/^{144}Nd = 0.511138$ of the Ames Nd standard (Wasserburg *et al*., 1981). Uncertainties correspond to last figures and represent $\pm 2\sigma_m$ error limits.
[d] Nd standard data were obtained in different analytical sessions.
[e] Uncertainties correspond to last figures and represent $\pm 2\sigma_p$ error limits.
[f] Not enough REE in the samples to measure.

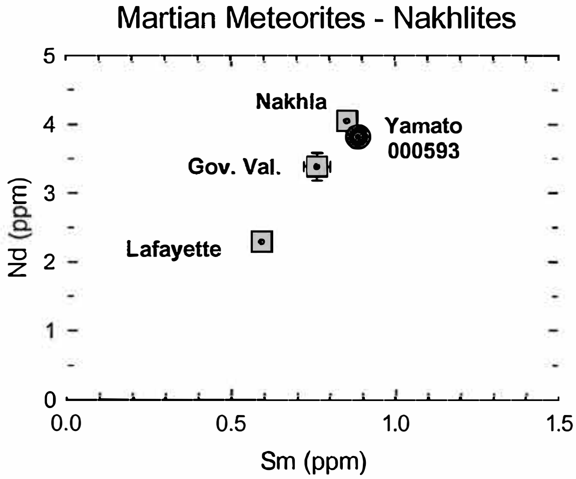

Fig. 5. Samarium versus neodymium abundance plot for nakhlites. Antarctic nakhlite Y000593 (circle) is the unleached WR1 sample from this study. Literature data for non-Antarctic nakhlites are shown by squares. Y000593 has the same Sm and Nd abundances as Nakhla. Lower Rb, Sr and REE abundances in Lafayette may be the results of a higher degree of partial melting of the source, or of post-eruption of olivine accumulation.

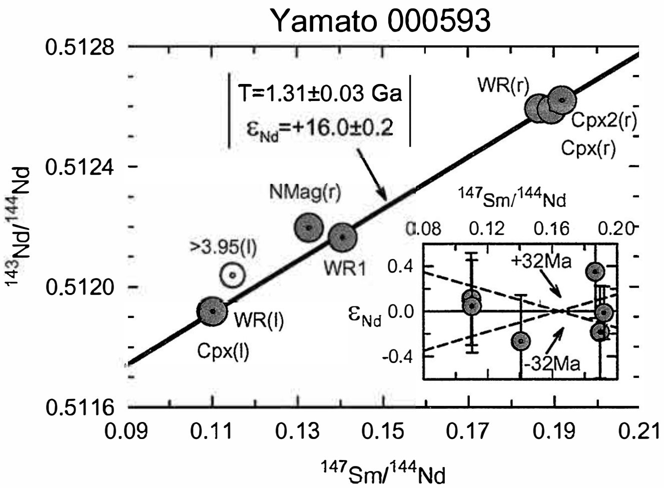

Fig. 6. Samarium-neodymium isochron diagram for Y000593. The data were fitted by the Williamson (1968) regression program and the isochron ages were calculated for $\lambda(^{147}Sm) = 0.00654\,Ga^{-1}$. The initial $^{143}Nd/^{144}Nd$ ratio is expressed in ε-units, using the notation and reference parameters of DePaolo and Wasserburg (1976) and Jacobsen and Wasserburg (1984). If all samples are included, the data do not define a single linear array. However, six data points, including two leachates, WR(l) and Cpx(l), define a linear array corresponding to a Sm-Nd age of 1.31±0.03 Ga. The obtained Sm-Nd age of 1.31±0.03 Ga is in excellent agreement with the Rb-Sr age (Fig. 3). The concordancy of Sm-Nd and Rb-Sr ages shows that Y000593 crystallized 1.31 Ga ago. A calculated age using three residues, WR(r), Cpx(r) and Cpx2(r), and the untreated WR1 sample, is 1.33±0.05 Ga. The calculated age becomes younger with a much large error (1.16±0.30 Ga) if all four residues, NMag(r), WR(r), Cpx(r) and Cpx2(r), are used. The reason why NMag(r) is desplaced from the isochron is unknown. We cannot exclude the possibility of analytical effects since this sample contained small amounts of REE.

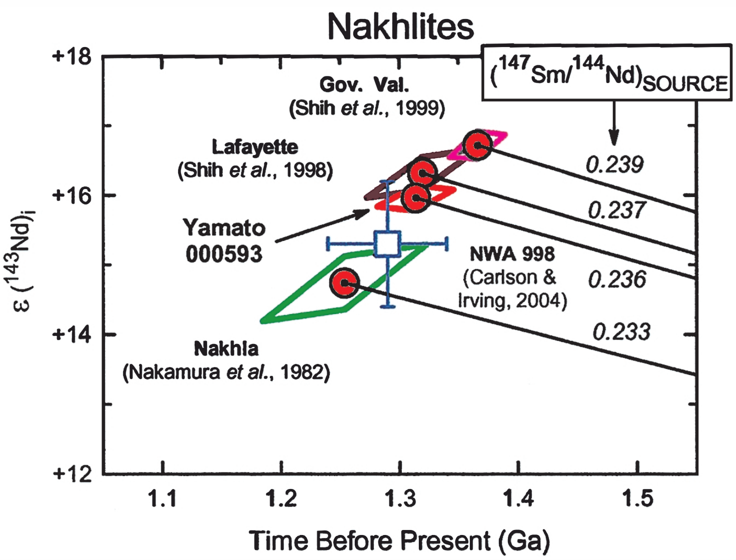

Fig. 7. Ages versus ε-Nd plot for nakhlites, represented by parallelograms constructed according to their ages and initial ε-Nd parameters. As in the T-I (Sr) plot (Fig. 4), not all nakhlite data overlap, suggesting that the nakhlites did not come from identical mantle sources at the same time. However, they all crystallized within ±0.1 Ga with initial ε-Nd values varying by only 2 ε-units. These small variations in ages and initial ε-Nd values suggest that nakhlites represent different flows from very similar light REE-depleted mantle sources. A single-stage model calculation shows that $^{147}Sm/^{144}Nd$ variations in nakhlite sources were small but distinct, as represented by different calculated evolution lines. Data are from Nakamura et al. (1982a), Shih et al. (1998, 1999), Carlson and Irving (2004) and this work.

Governador Valadares appear to be from "similar" but distinct sources.

3.3. ^{39}Ar-^{40}Ar age

The results of Ar isotopic analysis for Y000593 are presented in Appendix 1. The ^{39}Ar-^{40}Ar age spectrum and the K/Ca ratios as a function of cumulative release of ^{39}Ar (produced in the reactor from ^{39}K) are shown in Fig. 8. Most of the ^{39}Ar and ^{40}Ar were released in a narrow temperature peak over ~600–800°C. For the first ~84% of the ^{39}Ar release the K/Ca ratio varies only between 0.9 and ~0.3. We interpret this phase to be the mesostasis, which contains most of the alkali element inventory of Y000593 (*e.g.*, the non-magnetic fraction of Table 1). For temperatures releasing the last ~16% of the ^{39}Ar, the K/Ca ratio decreases rapidly to values of ~0.001 and lower. This phase may represent small inclusions contained within olivine and pyroxene (*e.g.*, Harvey and McSween, 1992). The total Ar-Ar age is 1.40 Ga. The age spectrum rises to a maximum age of ~1.5 Ga at ~10% ^{39}Ar release and then steadily declines at higher temperatures. We attribute the small age decrease over ~95–100% ^{39}Ar release to degassing of recoiled ^{39}Ar from surfaces of mafic mineral grains. We attribute the lower observed ages for the first ~10% ^{39}Ar release to loss of ^{40}Ar by terrestrial

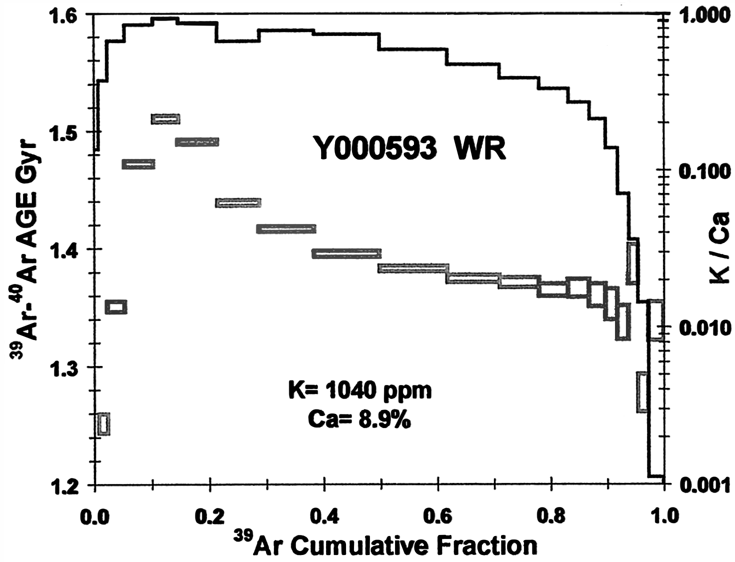

Fig. 8. Plot of ^{39}Ar-^{40}Ar ages (rectangles, left scale) and K/Ca ratios (stepped line, right scale) against cumulative release of ^{39}Ar for stepwise extractions of Y000593 whole-rock. Determined K and Ca concentrations are also given.

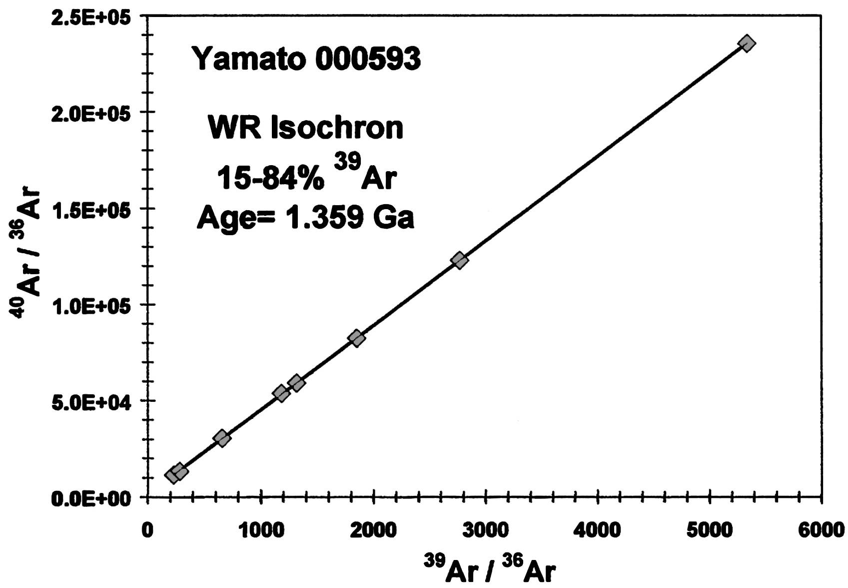

Fig. 9. Isochron plot of $^{40}Ar/^{36}Ar$ versus $^{39}Ar/^{36}Ar$ for nine extractions releasing 15–84% of the ^{39}Ar. The isochron age is 1.359 ± 0.020 (2σ) Ga and the $^{40}Ar/^{36}Ar$ intercept is 1502 ± 159. We interpret the isochron age as the most reliable ^{39}Ar-^{40}Ar age of Y000593 (see text). Higher apparent ages shown in Fig. 8 for individual extraction temperatures indicate the presence of excess ^{40}Ar.

weathering. There is no indication in these Ar data for a Martian weathering age of ~0.7 Ga, as has been suggested for some other nakhlites (Swindle and Olson, 2004).

Those Ar-Ar ages above a value of ~1.3 Ga, occurring over ~10–90% of the ^{39}Ar release, have been elevated by the presence of a trapped Martian Ar component. Radiogenic ^{40}Ar in Y000593 can be resolved from trapped Ar by use of a $^{40}Ar/^{36}Ar$ *versus* $^{39}Ar/^{36}Ar$ isochron plot. But for this isochron to be accurate, cosmogenic ^{36}Ar must first be subtracted. We used minimum values in the $^{36}Ar/^{37}Ar$ ratios to estimate cosmogenic ^{36}Ar for each extraction (Garrison *et al*., 2000) and subtracted these concentrations from measured ^{36}Ar values. Figure 9 gives the resulting isochron plot for eight extractions releasing 15–84% of the ^{39}Ar. This isochron is highly linear (R^2= 0.99998); its slope corresponds to an age of 1.359±0.020 (2σ) Ga; and its $^{40}Ar/^{36}Ar$ intercept of 1502±159 confirms the presence of a trapped Martian Ar component. Our sample released ~2.6×10^{-10}cm^3 STP/g of trapped Martian ^{36}Ar. This is >50% of the total ^{36}Ar released up through 87% of the ^{39}Ar release, beyond which degassing of cosmogenic ^{36}Ar greatly dominates.

The Ar-Ar isochron age is slightly older than the Rb-Sr age (1.30±0.02 Ga) and Sm-Nd age (1.31±0.03 Ga) of Y000593 reported here. This Ar-Ar age is also slightly older than Ar-Ar ages we previously reported for Governador Valadares (1.32±0.08 Ga; Bogard and Husain, 1977) and Chassigny (1.32±0.14 Ga; Bogard and Garrison, 1999) and than Ar-Ar ages for Nakhla (1.323±0.022 Ga) and Lafayette (1.322±0.020 Ga) reported by Swindle and Olson (2004). Swindle and Olson (2004) also reported small excesses of ^{40}Ar at low to intermediate extraction temperatures for Nakhla and Lafayette. Although a few radiometric age determinations of nakhlites have given values around 1.36 Ga (Nyquist *et al*., 2001), we believe that the older Ar-Ar age for Y000593 may have been produced by trapped Martian Ar. The derived trapped Martian $^{40}Ar/^{36}Ar$ ratio of ~1500 in Y000593 is consistent with this ratio for the recent Martian atmosphere deduced from shock glass in shergottites (Bogard and Garrison, 1999). However, our Y000593 sample apparently contains additional ^{40}Ar beyond this atmospheric ^{40}Ar. We suggest that this excess ^{40}Ar is a radiogenic component that was present in the nakhlite melt, rather than being introduced into the meteorite after its formation. This conclusion would be consistent with the presence of trapped ^{40}Ar in mesostasis, rather than in mafic minerals, as mesostasis is the latest phase to solidify.

3.4. *Ejection age and launch pairing of nakhlites and Chassigny*

The average cosmic-ray exposure (CRE) age of three Yamato nakhlites (Y000593, Y000749, Y000802) is 12.1±1.0 Ma (Okazaki *et al*., 2003). This age is consistent with the CRE ages for Nakhla (10.75±0.40 Ma), Governador Valadares (10.0±2.1 Ma), Lafayette (11.9±2.2 Ma) and NWA 817 (9.7±1.1 Ma) (summarized in Nyquist *et al*., 2001; Marty *et al*., 2001).

Our whole-rock sample of Y000593 contained 1.18×10^{-8}cm^3/g of cosmogenic ^{36}Ar, most of which was released over 1100–1350°C. However, ^{38}Ar has a significant component produced in the reactor from ^{37}Cl. Most of the ^{38}Ar releases in the temperature range 800–1000°C, from a phase, likely a phosphate, distinct from those containing most of the K and Ca. Assuming a reasonable cosmogenic $^{38}Ar/^{36}Ar$ ratio of 1.5, our sample of Y000593 contained ~1.77×10^{-8}cm^3/g of cosmogenic ^{38}Ar. This

^{38}Ar concentration using the production rate from Eugster *et al.* (1997) is similar to cosmogenic ^{38}Ar concentrations of 1.76–2.10$\times 10^{-8}$cm^3/g reported for other nakhlites, Nakhla, Lafayette and Governador Valadares (Eugster *et al.*, 1997).

The similarities in crystallization and ejection ages and petrographic features suggest that all the nakhlites were ejected from Mars in the same impact event. Including the dunite Chassigny, which shares the same CRE (11.3$\pm$0.6 Ma) age and crystallization (~1.3 Ga) age as the nakhlites (Nyquist *et al.*, 2001), and assuming the three Yamato nakhlites represent a single fall, at least six, and probably seven, nakhlite-Chassigny meteorite falls were launched from Mars in a single event. These observations suggest a "large" event.

3.5. *Disturbance of isotopic systematics*

Yamato nakhlites contain pre-terrestrial secondary minerals (*e.g.*, Bridges *et al.*, 2003; Noguchi *et al.*, 2003) as is the case with non-Antarctic nakhlites (Bunch and Reid, 1975; Reid and Bunch, 1975; Berkley *et al.*, 1980; Boctor *et al.*, 1976; Treiman *et al.*, 1993; Gillet *et al.*, 2002, see Bridges *et al.*, 2001). Bridges and co-workers described siderite-clay intergrowths of secondary mineralization in Y000593,54 (Bridges *et al.*, 2003). As shown in Fig. 10, we found C- and Ca-rich veins in one olivine separate (Ol). The outer veins contacting olivine contain C and Ca, suggesting this phase is different from siderite but is probably calcite. These alteration phase differences imply that Martian weathering products are heterogeneously distributed in Y000593. It has been suggested that aqueous alteration on the Martian surface was responsible for the observed disturbance of the Rb-Sr system in Nakhla at or near 1.3 Ga (Gooding *et al.*, 1991). We suggest the weathering products influenced some previous Rb-Sr age determinations (Papanastassiou and Wasserburg, 1974; Shih *et al.*, 1998; Misawa *et al.*, 2003c).

Nakhlite olivines in general contain symplectite exsolution consisting of magnetite and augite, which could have been produced by the decomposition of Fe^{3+}-bearing olivine at relatively high temperature (~800°C) and high fO_2 conditions (Mikouchi *et al.*, 2000) or by subsolidus olivine oxidation (*e.g.*, Champness, 1970). Aqueous alteration veins cross-cut symplectites, indicating "iddingsite" formed after the symplectite (Noguchi *et al.*, 2003). Symplectite alteration did not occur at the cross-cut portion of "iddingsite" (Noguchi *et al.*, 2003). This fact suggests that symplectite survived and was little affected during the "iddingsite" formation. Narrow veins of secondary mineralization in olivine (Fig. 10), probably including calcite, could be a sink of Sr. Textural relations indicate that [olivine+symplectite] and ["iddingsite"+calcite] have not been equilibrated.

3.6. *Did nakhlite "iddingsite" record a final aqueous alteration event on Mars?*

A tie line between two olivine leachates, $\rho>3.95$(l) and Ol(l), provides a calculated Rb-Sr "age" of 650$\pm$80 Ma with an initial ^{87}Sr/^{86}Sr of 0.70465$\pm$10. This "age" is in good agreement with the oldest K-Ar age of 670$\pm$91 Ma for the "iddingsite" sample from Lafayette (Swindle *et al.*, 2000) and with the Rb-Sr age of 673$\pm$65 Ma for HCl leachates from the "iddingsite"-rich samples (Shih *et al.*, 1998) from Lafayette. If we assume that two olivine leachates, $\rho>3.95$(l) and Ol(l), represent "iddingsite", and that

(a)

(b)

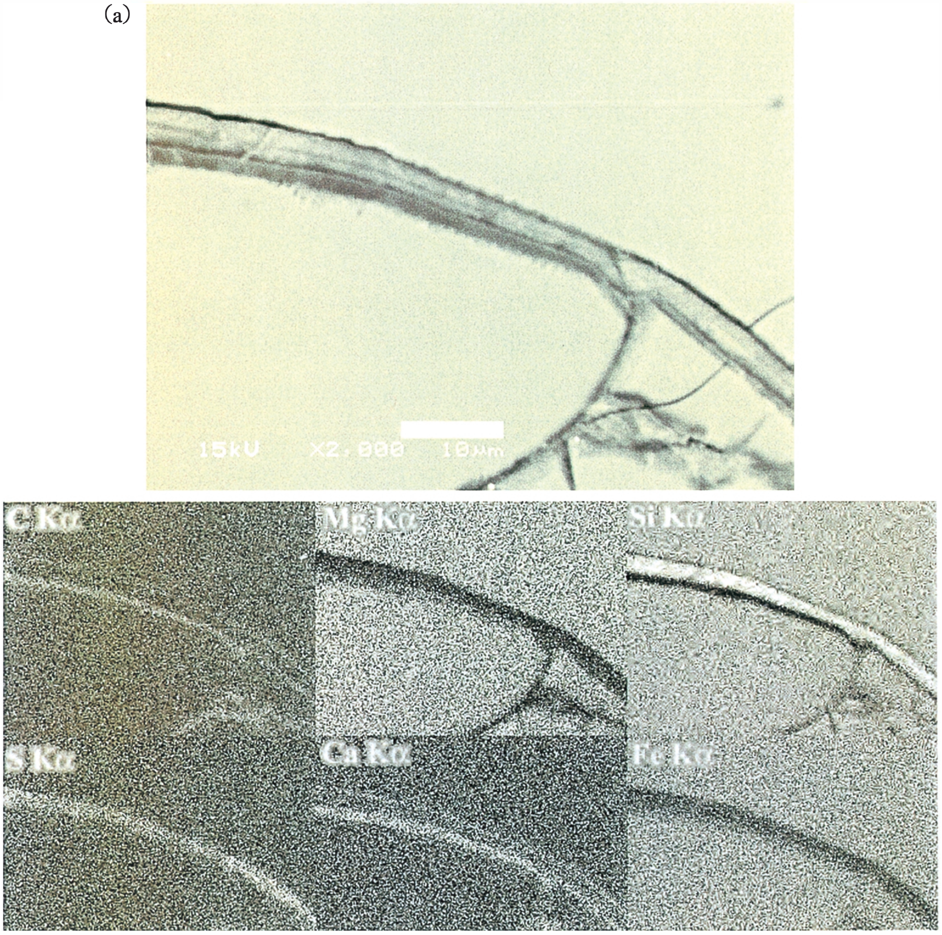

Fig. 10. (a) Backscattered electron image of Y000593 olivine showing veins of aqueous alteration. Scale bar = 10 μm.
(b) X-ray maps of C, Mg, Si, S, Ca and Fe. Inner alteration veins contain Si and S. Outer thin veins which directly contacting olivine are enriched in C and Ca, and contain minor amounts of Mg and Fe. The elemental distributions indicate the presence of carbonates (outer veins directly contacting olivine) and sulfates (inner veins). Some clay minerals as noted by Bridges et al. (2003) and Noguchi et al. (2003) also may be present.

contributions of Rb and Sr from olivine during 2N HCl leaching are negligible (*i.e.*, olivine did not contain a significant amount of incompatible elements, in particular, Rb and Sr) even though leachates are mixture of "iddingsite" and olivine, the young "age" may represent the formation age of "iddingsite". That is, during a weathering episode, Sr might have added. "Iddingsite" in Y000593 might then have formed as final brine evaporates possessing the relatively high $^{87}Sr/^{86}Sr$ of ~0.7046. A comparable $^{87}Sr/^{86}Sr$ (~0.7042) was also obtained from the HCl-leachates of Lafayette "iddingsite" (Shih *et al*., 1998).

However, the Ar-Ar age spectrum for Y000593 gives little support to the younger Rb-Sr age of ~650 Ma suggested by the Rb-Sr data for the olivine leachates. Formation of secondary minerals such as "iddingsite", invoked to explain this younger Rb-Sr age, is expected to occur much more readily on alkali-poor olivine than on alikali-rich materials such as plagioclase and mesostasis. Thus, one might not expect to see younger Ar-Ar ages for only a modest amount of Martian chemical weathering because Ar age spectra is dominated by Ar release from mesostasis and plagioclase. Nevertheless, recent ^{39}Ar-^{40}Ar data of whole-rock samples of Nakhla and Lafayette were interpreted as revealing a low-temperature disturbance, suggesting weathering and "iddingsite" formation at a time much later than the crystallization of the rock (Swindle and Olson, 2004). Those analyses showed much larger ^{39}Ar recoil effects compared to Y000593. Although the interpretation of the Ar data for Nakhla and Lafayette is consistent with the present and previous Rb-Sr results on nakhlite olivines, we believe that the Ar data for Nakhla and Lafayette are just as well interpreted as resulting from diffusive loss of ^{40}Ar and ^{39}Ar recoil effects. Thus, we cannot conclude unambiguously from this study that the aqueous alteration recorded in nakhlites occurred in a single event.

4. Conclusions

Isotopic analysis of Y000593 yields a Rb-Sr age of 1.30$\pm$0.02 Ga with an initial $^{87}Sr/^{86}Sr$ of 0.702525$\pm$0.000027, a Sm-Nd age of 1.31$\pm$0.03 Ga with an initial ε_{Nd} of +16.0$\pm$0.2 and an Ar-Ar age of $\leq$1.36 Ga. The concordancy of these three ages and Rb-Sr and Sm-Nd initial isotopic signatures suggests that Y000593 crystallized from low Rb/Sr, light REE-depleted source materials ~1.31 Ga ago. The crystallization age of Y000593 is comparable to the age data of non-Antarctic nakhlites (Nakhla, Governador Valadares, Lafayette and NWA 998) and Chassigny. The initial Sr and Nd isotopic signatures suggest that Lafayette and Y000593 were co-magmatic or at least came from very similar magmas. The similarities in crystallization and Mars ejection ages and in petrologic features suggest the nakhlites were derived from similar source regions, and experienced launch pairing with Chassigny. The Rb-Sr data for Y000593 show that the isotopic system was disturbed by pre-terrestrial alteration, but lack of identifiable corresponding effect in the Ar data prevents unambiguous identification of the ~650 Ma apparent age with a single event.

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Appendix 1. Argon isotopic data.

Temp (°C)	^{39}Ar (ccSTP/g)	Age (Ga) ± error	K/Ca ± error	^{40}Ar/^{39}Ar ± error	^{38}Ar/^{39}Ar ± error	^{37}Ar/^{39}Ar ± error	^{36}Ar/^{39}Ar ± error
325	1.25E-09	1.078	0.1356	31.87	7.467	4.055	0.0694
		0.019	0.0035	0.75	0.243	0.104	0.0032
425	2.77E-09	1.252	0.3726	39.03	1.447	1.476	0.0169
		0.009	0.0050	0.35	0.018	0.020	0.0009
500	5.29E-09	1.351	0.6700	43.45	0.598	0.821	0.0048
		0.006	0.0074	0.20	0.004	0.009	0.0004
560	8.65E-09	1.472	0.8489	49.17	0.211	0.648	0.0030
		0.005	0.0088	0.13	0.002	0.007	0.0002
600	7.71E-09	1.510	0.9309	51.07	0.128	0.591	0.0022
		0.006	0.0097	0.14	0.002	0.006	0.0002
630	1.18E-08	1.491	0.8677	50.09	0.129	0.634	0.0052
		0.005	0.0088	0.09	0.002	0.006	0.0004
655	1.30E-08	1.439	0.6630	47.57	0.110	0.830	0.0046
		0.005	0.0068	0.12	0.001	0.009	0.0003
680	1.68E-08	1.417	0.7778	46.50	0.077	0.707	0.0024
		0.005	0.0080	0.10	0.001	0.007	0.0002
705	2.02E-08	1.395	0.7316	45.51	0.076	0.752	0.0018
		0.005	0.0076	0.12	0.001	0.008	0.0002
730	2.09E-08	1.383	0.5860	44.92	0.102	0.939	0.0020
		0.005	0.0060	0.11	0.001	0.010	0.0002
750	1.63E-08	1.375	0.4718	44.54	0.156	1.166	0.0021
		0.005	0.0050	0.14	0.002	0.012	0.0003
770	1.23E-08	1.371	0.3847	44.39	0.278	1.430	0.0022
		0.006	0.0042	0.19	0.002	0.016	0.0003
790	9.07E-09	1.365	0.3282	44.09	0.524	1.676	0.0024
		0.007	0.0038	0.25	0.005	0.019	0.0004
810	6.45E-09	1.367	0.2687	44.17	0.933	2.047	0.0027
		0.009	0.0034	0.35	0.011	0.026	0.0006
835	5.08E-09	1.361	0.2114	43.89	1.607	2.601	0.0040
		0.011	0.0030	0.44	0.023	0.037	0.0008
860	3.73E-09	1.353	0.1379	43.53	2.790	3.989	0.0075
		0.014	0.0023	0.59	0.042	0.067	0.0008
895	3.44E-09	1.337	0.07057	42.83	4.791	7.793	0.0166
		0.015	0.00128	0.64	0.080	0.141	0.0016
930	2.83E-09	1.387	0.03611	45.11	5.352	15.23	0.0308
		0.017	0.00072	0.78	0.129	0.31	0.0057
980	3.24E-09	1.277	0.01432	40.16	7.081	38.40	0.0714
		0.016	0.00029	0.69	0.136	0.77	0.0069
1090	4.53E-09	1.338	0.00111	42.88	11.03	494.2	0.7674
		0.017	0.00002	0.74	0.20	9.8	0.0571
1200	3.30E-10	1.88	0.00004	71.7	33.3	12300	18.6
		0.97	0.00004	59.4	27.6	10200	15.6
1350	6.53E-11	1.98	0.00004	78.0	35.1	12600	18.8
		3.08	0.00011	199	89.9	32300	48.2
1550	1.31E-11	4.31	0.00013	387	13.3	4330	6.86
		1.82	0.00014	430	14.9	4810	7.70

Columns give: 1) extraction temperature in ℃; 2) ^{39}Ar concentrations; 3) Ar-Ar age; 4) K/Ca ratio; and 5–8) measured isotopic ratios, appropriately corrected for blanks, decay and reactor interferences. One σ uncertainties are given below individual values.