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LANTHANITE-(Nd) FROM SANTA ISABEL, STATE OF SÃO PAULO: SECOND BRAZILIAN AND WORLD OCCURRENCE

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ABSTRACT

Lanthanite-(Nd), known previously only from Curitiba (Paraná State, southern Brazil), has been found in calcrete of the Tertiary Caçapava Formation at Santa Isabel, São Paulo State, southern Brazil. Lanthanite-(Nd) occurs as veneers of platy crystals about 2 to 3 mm wide. The mineral is orthorhombic, space group *Pbnb*, *a* 9.490(7), *b* 16.94(1), *c* 8.941(7) Å, *V* 1437.1 Å^3, and $Z = 4$. The strongest six lines of the X-ray powder-diffraction pattern [*d* in Å(*I*)(*hkl*)] are: 8.530(100)(020), 4.765(50)(200), 4.246(60)(040), 3.258(70)(202), 3.036(80)(222), and 2.580(55)(242). The crystals are transparent, violet in natural light, and pale pink in artificial light. They have a white streak, pearly to vitreous luster, and uneven fracture. D_{meas} 2.82(3), D_{calc} 2.809 g/cm^3. Optically, the mineral is biaxial (–), with α 1.529(3), β 1.593(3), γ 1.617(3), $2V_x$ (meas.) 58°, $2V_x$ (calc.) 60°. The analytical formula, derived from results of a wet-chemical analysis, is $(La_{0.394}Nd_{0.390}Pr_{0.068}Sm_{0.059}Gd_{0.035}Y_{0.024}Dy_{0.009}Eu_{0.006}Ho_{0.005}Ce_{0.005}Er_{0.003}Tb_{0.002}Yb_{<0.0003})_2(CO_3)_2{\bullet}8H_20$, or simply, $(La,Nd)_2(CO_3)_3{\bullet}8H_2O$, with La ≈ Nd. Plots on a chondrite-normalized diagram show negative Ce and Eu anomalies.

Keywords: lanthanite-(Nd), hydrous La, Nd carbonate, second world occurrence, calcrete, São Paulo, Brazil.

SOMMAIRE

Nous avons découvert la lanthanite-(Nd), connue auparavant seulement a Curitiba (état de Paraná, dans le sud du Brésil), dans un encroûtement calcaire d'âge tertiaire de la formation Caçapava à Santa Isabel (état de São Paulo, aussi dans le sud du Brésil). Les cristaux en plaquettes atteignent de 2 à 3 mm de large. C'est une phase orthorhombique, groupe spatial *Pbnb*, *a* 9.490(7), *b* 16.94(1), *c* 8.941(7) Å, *V* 1437.1 Å^3, and $Z = 4$. Les six raies les plus intenses du cliché de poudre [*d* en Å(*I*)(*hkl*)] sont: 8.530(100)(020), 4.765(50)(200), 4.246(60)(040), 3.258(70)(202), 3.036(80)(222), et 2.580(55)(242). Les cristaux sont transparents, violette en lumière naturelle, et rose pâle en lumière artificielle. La rayure est blanche, et l'éclat, nacré à vitreux. La fracture est irrégulière. D_{mes} 2.82(3), D_{calc} 2.809. La lanthanite-(Nd) est biaxe négative, α 1.529(3), β 1.593(3), γ 1.617(3), $2V_x$ (mes.) 58°, $2V_x$ (calc.) 60°. Sa formule, d'aprés les données chimiques obtenues par voie humide, serait $(La_{0.394}Nd_{0.390}Pr_{0.068}Sm_{0.059}Gd_{0.035}Y_{0.024}Dy_{0.009}Eu_{0.006}Ho_{0.005}Ce_{0.005}Er_{0.003}Tb_{0.002}Yb_{<0.0003})_2(CO_3)_3{\bullet}8H_2O$, ou plus simplement, $(La, Nd)_2(CO_3)_3{\bullet}8H_2O$, avec La ≈ Nd. Par rapport aux abondances chondritiques, la lanthanite-(Nd) montre une anomalie négative en Ce et en Eu.

(Traduit par la Rédaction)

Mots-clés: lanthanite-(Nd), carbonate de La et de Nd hydraté, deuxième indice connu, encroûtement calcaire, São Paulo, Brésil.

INTRODUCTION

The type occurrence of lanthanite-(Nd), $(Nd,La)_2(CO_3)_3{\bullet}8H_2O$, is in a calcrete of the (Tertiary?) Guabirotuba Formation, Curitiba Basin, Curitiba, Paraná State, Brazil. On the basis of optical microscopy, Coutinho (1955) classified the mineral as "lanthanite". Fujimori & Bittencourt (1972) obtained cell parameters and DTA for the mineral. Microprobe and wet-chemical analyses by Ansell *et al.* (1976) and Cesbron *et al.* (1979) showed that the sample is comparatively richer in neodymium and poorer in cerium than lanthanite-(La). Roberts *et al.* (1980) furnished additional optical, physical and crystallographic data and, with I.M.A. approval, introduced the new name *lanthanite-(Nd)*. Working independently on material from the same occurrence, Fujimori (1981) and Svisero & Mascarenhas (1981) contributed additional optical, crystallographic, and chemical data. This paper describes the second Bra-

TABLE 1. OPTICAL PROPERTIES OF LANTHANITE-(Nd)

	1	2	3	4
α	1.514(calc.)	1.532(1)	1.517(calc.*)	1.529(3)
β	1.589(3)	1.590(1)	1.590	1.595(3)
γ	1.612(3)	1.614(1)	1.615	1.617(3)
2Vx meas.	60°	61°	-	58°
2Vx calc.	-	63.5°	-	60°

* Calculated on the basis of the 2V measured by Coutinho (1955).

1. Curitiba (Coutinho 1955)
2. Curitiba (Roberts et al. 1980)
3. Curitiba (Svisero & Mascarenhas 1981)
4. Santa Isabel (this paper)

TABLE 2. DISTRIBUTION OF RARE EARTHS IN LANTHANITE-(Nd) (wt.%)*.

	1	2
La_2O_3	35.6	38.6
CeO_2	<0.3	0.5
Pr_6O_{11}	9.8	7.0
Nd_2O_3	40.0	39.5
Sm_2O_3	7.5	6.2
Gd_2O_3	3.1	3.8
Tb_4O_7		0.2
Dy_2O_3	0.8	1.0
Ho_2O_3		0.6
Er_2O_3		0.3
Yb_2O_3		<0.05
ThO_2	<0.3	
Y_2O_3	~0.4	1.6
Eu_2O_3	1.1	0.60

* $RE_2O_3/\Sigma RE_2O_3$

1. Curitiba (analysis by Société Rhône-Poulenc)
2. Santa Isabel (this paper)

TABLE 3. CHEMICAL DATA FOR LANTHANITE-(Nd)

	1	2	3	4	5	6	7
La_2O_3	26.83	23.63	22.4	19.44	22.06	22.39	22.28
Ce_2O_3				0.03		0.02	0.29
Pr_2O_3		4.18	4.5	5.18	3.38	4.95	3.89
Nd_2O_3	27.70	26.57	22.2	21.84	24.09	21.18	22.77
Sm_2O_3		5.95	3.4	4.10	7.62	3.80	3.57
Eu_2O_3		0.90	0.6	0.60		0.66	0.37
Gd_2O_3		3.72	1.7	1.69		1.05	2.20
Tb_2O_3						0.14	0.13
Dy_2O_3				0.44		0.45	0.58
Ho_2O_3						0.07	0.33
Er_2O_3							0.20
Tm_2O_3							n.a.
Yb_2O_3						0.06	<0.02
Lu_2O_3							n.a.
Y_2O_3				0.22	0.20		0.94
ThO_2				0.03			n.a.
CO_2	21.74	25.61		22.15	43.30	25.0	21.43
H_2O	23.73			22.75		20.0	21.00
TOTAL	100.00	90.56		98.47		99.77	(100.00)

n.a.: not analyzed. Data expressed in wt.%.

1. Theoretical formula, $(Nd,La)_2(CO_3)_3.8H_2O$, Nd:La=1:1.
2. Curitiba (Ansell et al. 1976)
3. Curitiba (JCPDS card 30-678, average of 9 grains. Range: La_2O_3 18.5-28.0, Nd_2O_3 20.4-22.8 Geol. Surv. Canada)
4. Curitiba (Cesbron et al. 1979; original value for Eu_2O_3 of 1.64% in error; here corrected.)
5. Curitiba (Fujimori 1981, average of 2 samples)
6. Curitiba (Svisero & Mascarenhas 1981)
7. Santa Isabel (this paper)

zilian and world occurrence of lanthanite-(Nd), at Santa Isabel, São Paulo State.

Occurrence

Lanthanite-(Nd) occurs in sediments of the Tertiary (Eocene-Oligocene) Caçapava Formation, Taubaté Basin, São Paulo, Brazil. The outcrop is located at km 187.2 of the Presidente Dutra highway (BR-116), near the entrance to the town of Santa Isabel. Here, crystals of lanthanite-(Nd) are associated with dendritic crusts of manganese and iron oxides on white calcrete that fills irregular cm-wide fractures in green arenaceous mudstone. Thin crusts of white and pinkish calcite also were observed. The above association is similar to that found in the Guabirotuba Formation (Curitiba Basin), at the type locality. Similar and probably synchronous conditions of braided fluvial sedimentation, a semiarid paleoclimate, and tectonic events affected the Caçapava and Guabirotuba formations. The Taubaté and Curitiba basins apparently were generated by Early Tertiary rifting related to the separation of South America and Africa (Coimbra & Riccomini 1985).

Physical and Optical Properties

Lanthanite-(Nd) is found at Santa Isabel as veneers of platy crystals in fissures in calcrete. Very little material was recovered, and almost all was consumed during this investigation. The mineral is transparent, violet under natural light, pale pink under artificial light, has a pearly to vitreous luster, uneven fracture, white streak, and is not brittle. The mineral is nonfluorescent in both long- and short-wave ultraviolet light. Mohs hardness is 2.5 to 3. The mineral effervesces in dilute cold HCl. The width of individual plates is about 2 to 3 mm. The crystals are flattened on [010] and twinned on (101). The forms present are {001}, {010}, {100}, {101} and {121}. The crystals exhibit perfect {010} and good {101} cleavages. The average of three determinations of density, by heavy-liquid suspension of one grain, is 2.82(3) g/cm^3.

Optical properties were determined in white light. Lanthanite-(Nd) is colorless, biaxial negative, with $X = b$, $Y = c$ and $Z = a$. The β and γ indices were determined on flat-lying plates, using Cargille liquids checked to ± 0.0005 accuracy on an Abbé refractometer, at constant temperature. To obtain the α value, the same immersion method was applied following an additional preliminary step: under a binocular microscope, plates 0.2 to 0.3 mm wide and 0.1 to 0.15 mm thick were split with a razor blade along the {101} cleavage, the resulting halves adhering easily along their (010) plane. The two plates could then be made to stand vertically on a stable base, now 0.2 to 0.3 mm thick, on a gelatin-coated glass slide, thereby bringing the α index into a horizontal position. The slides were then covered with immersion oil (without a cover slip) and α was measured under

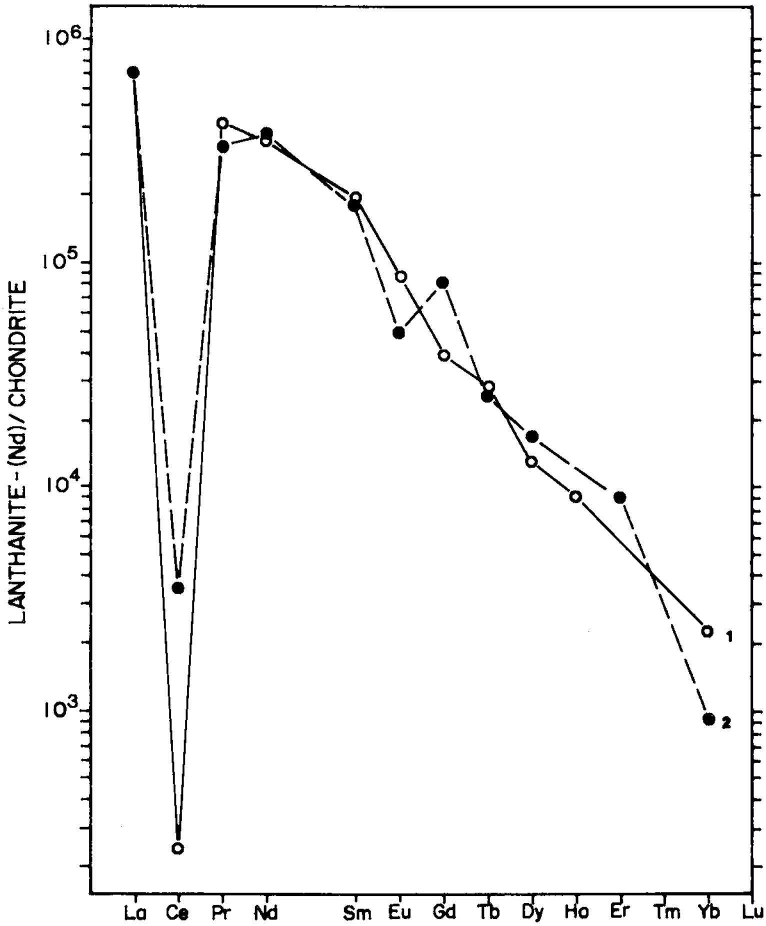

FIG. 1. Chondrite-normalized *REE* patterns for (1) Curitiba (Svisero & Mascarenhas 1981) and (2) Santa Isabel lanthanite-(Nd). Chondrite values are those of Taylor & Gorton (1977).

the polarizing microscope. The orientation was checked by means of interference figures. Optical properties of lanthanite-(Nd) are presented in Table 1.

CHEMICAL COMPOSITION

Relative proportions of rare-earth elements for the Santa Isabel lanthanite-(Nd) (Table 2, column 2) were determined by precipitation of *REE* and Y as oxalates later calcined to oxides. La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Yb and Y were determined by X-ray fluorescence spectrometry. Eu was determined by polarography. Tm and Lu were not determined. A separate hand-picked sample was used to determine concentrations of carbon and hydrogen on a Perkin Elmer Model 240 B Elemental Analyzer. C and H percentages were recalculated to CO_2 and H_2O. The total of *REE* oxides was considered to be 57.57 wt.% in order to total 100.00%. The chemical composition of the Santa Isabel lanthanite-(Nd) is compared to that of the Curitiba lanthanite-(Nd) in Table 3. Assuming two *REE* atoms, the empirical formula for Santa Isabel lanthanite-(Nd) is $(La_{0.394}Nd_{0.390}Pr_{0.068}Sm_{0.059}Gd_{0.035}Y_{0.024}Dy_{0.009}Eu_{0.006}Ho_{0.005}Ce_{0.005}Er_{0.003}Tb_{0.002}Yb_{<0.0003})_2C_{2.805}O_{8.610}\cdot 6.717H_2O$. Thus, the ideal formula is $(La,Nd)_2(CO_3)_3\cdot 7H_2O$ with La ≈ Nd, whereas in the generally accepted formula for lanthanite-(Nd) there are $8H_2O$ molecules. The calculated density for the analytical formula having $8H_2O$ and $3CO_3$ is 2.809 g/cm^3, whereas that assuming 6.717 H_2O and $C_{2.805}O_{8.610}$ is 2.663 g/cm^3. As the measured density is 2.82(3) g/cm^3, the former value is more reasonable. The discrepancies between the H_2O and CO_3 contents inferred from the chemical analyses and those necessary to obtain a satisfactory calculated density suggest analytical errors. Unfortunately, the

TABLE 4. X-RAY POWDER-DIFFRACTION DATA FOR LANTHANITE-(Nd)

1		2		3		4		5			
d_{obs}(Å)	I/I_o	d_{obs}	I/I_o	d_{obs}	I/I_o	d_{obs}	I/I_o	d_{obs}	I/I_o	$d_{calc.}$	hkl
8.45	100	8.50	100	8.49	100	8.50	100	8.530	100	8.470	020
4.74	30	4.741	52	4.73	30	4.74	30	4.765	50	4.745	200
4.46	30	4.473	56	4.47	30	4.46	25	4.490	45	4.470	002
4.23	30	4.233	28	4.24	60	4.23	70	4.246	60	4.235	040
4.13	30	4.139	34	4.16	10	4.15	15	4.158	35	4.140	220
				4.03	10					4.069	211
3.95	30	3.953	32	3.95	10	3.96	15	3.974	30	3.954	022
		3.868	7							3.867	140
3.84	<10b					3.84	20	3.855	25		
		3.829	7							3.827	041
3.25	40	3.252	63	3.25	60	3.25	35	3.258	70	3.254	202
3.15	<10	3.160	7	3.15	10	3.15	10	3.157	10	3.160	240
3.07	<10	3.075	10							3.074	042
3.03	40	3.038	58	3.04	50	3.02	90	3.036	80	3.037	222
3.001	<10	3.004	14	3.00	50					3.005	151
		2.980	7							2.982	301
2.924	<10	2.926	7			2.93	10	2.929	5	2.925	142
		2.846	7							2.843	103
		2.823	12							2.823	060
2.814	10			2.82	60	2.81	40	2.825	45		
		2.810	9							2.813	321
		2.707	12							2.706	160
2.689	10			2.70	60	2.69	40	2.709	45		
		2.691	15							2.692	061
		2.638	6							2.637	331
2.576	20	2.579	30	2.58	60	2.58	50	2.580	55	2.580	242
		2.551	6							2.553	312
		2.531	6							2.534	340
		2.440	12							2.437	043
2.426	<<10b			2.43	40	2.42	15	2.431	8		
		2.424	13							2.426	260
		2.388	8							2.387	062
				2.39	20	2.37	<10				
		2.368	9							2.372	400
2.278	<10	2.282	21	2.28	20	2.28	10	2.283	5	2.285	420
2.232	<10	2.234	10							2.235	004
2.204	<10	2.202	7							2.205	342
										2.169	303
		2.167	26					2.167	8		
										2.168	243
2.162	10	2.161	6	2.16	20	2.16	10			2.161	024
										2.132	262
2.126	<10	2.132	9	2.12	20	2.11	10	2.126	<5	2.125	431
										2.117	080
		2.107	6							2.107	124
										2.096	402
2.091	<10	2.093	27	2.09	20			2.098	<5		
										2.096	271
										2.070	440
2.061	<10b	2.068	13	2.06	50	2.06	60	2.066	15		
										2.067	180
		2.050	9							2.054	352
2.029	10	2.032	29	2.03	20					2.034	422
		2.022	18							2.024	253
2.010	<10b			2.01	30	2.01	15	2.022	10	2.022	204
		2.013	16							2.017	441
		1.978	11							1.977	044
1.968	10			1.97	30	1.97	20	1.976	5		
		1.966	19							1.967	224
										1.935	144
1.931	<10	1.936	6	1.93	30	1.93	15	1.931	<5	1.934	280
										1.931	343
										1.914	082
				1.91	<10						
										1.905	362
										1.879	371
1.872	10	1.876	23	1.88	50	1.87	70	1.878	20	1.878	442
										1.876	182
										1.827	353
1.820	<10	1.825	12	1.82	20	1.82	25	1.822	5		
										1.825	244
		1.812	8							1.813	423
										1.782	452
				1.77	20	1.77	15	1.780	<5		
										1.780	461
				1.72	20	1.72	10	1.729	<5	1.726	083
				1.69	20			1.680	<5	1.678	135
										1.639	1.10.1
				1.64	30	1.64	30	1.639	<5		
										1.637	382
										1.601	503
						1.59	10	1.600	<5		
										1.598	424
						1.55	20	1.555	<5		
						1.51	10	1.518	<5		
						1.49	10	1.488	<5		
						1.47	15	1.470	<5		
						1.45	10	1.457	<5		
								1.421	<5		
						1.40	15	1.406	<5		
				1.38	20	1.37	20	1.380	<5		
								1.348	<5		
						1.33	10	1.337	<5		
								1.308	<5		
								1.291	<5		
				1.27	30	1.27	20	1.278	<5		
						1.26	15	1.265	<5		
								1.250	<5		
				1.19	30b	1.18	20				
								1.080	<5		
				1.04	50			1.046	<5		

b = broad line; d values below 1.600 allow multiple indexing

1. Curitiba (Ansell et al. 1976) 2. Curitiba (Cesbron et al. 1979)
3. Curitiba (Fujimori 1981) 4. Curitiba (Svisero & Mascarenhas 1981)
5. Santa Isabel (this paper)

small quantities of the mineral preclude additional analyses.

The originally published $Eu_2O_3/\Sigma RE_2O_3$ and Eu_2O_3 wt.% of the Curitiba lanthanite-(Nd) studied by Cesbron *et al.* (1979) are 3.00 and 1.64, respectively. Such values are much higher than those found in other samples of lanthanite-(Nd). We have obtained from the Société Rhône–Poulenc a copy of the results of the analyses furnished to Cesbron *et al.* (1979) in which Eu_2O_3/RE_2O_3 is 1.1 (Table 2, column 1) and not 3.00 as published by Cesbron *et al.* (1979). The recalculation for Eu_2O_3 results in a value of 0.60 wt.%, which more closely approximates the data of other authors.

The chondrite-normalized *REE* pattern of Santa Isabel lanthanite-(Nd) (Fig. 1) shows a strongly fractionated, light-*REE*-enriched linear pattern with negative Ce and Eu anomalies, particularly strong for the former; the pattern of Curitiba lanthanite-(Nd) (Svisero & Mascarenhas 1981) also is characterized by a pronounced negative Ce anomaly, but does not display a Eu anomaly. The Ho value for Santa Isabel lanthanite-(Nd) was interpolated between the values for the adjacent elements Dy and Er because the Ho value obtained from chemical analysis seems to be in error. The analytical result for Ho_2O_3 is 0.33 wt.%, whereas the chondrite-normalized *REE* pattern suggests a value near 0.09.

Calculations using the Gladstone–Dale relationship yield a K_P of 0.2057 and a K_C of 0.1975 for the analytical formula using constants reported by Mandarino (1976); hence $1-(K_P/K_C)$ is -0.0415, indicating good compatibility (Mandarino 1979). If the amounts of CO_2 and H_2O are in fact greater than those indicated from the chemical analysis, as the measured density suggests, the compatibility is even better.

X-RAY DATA

Lanthanite-(Nd) is orthorhombic, space group *Pbnb*, $Z = 4$. X-ray powder-diffraction data for Santa Isabel lanthanite-(Nd) were obtained utilizing

TABLE 5. CELL PARAMETERS OF LANTHANITE-(Nd)

	1	2	3	4	5
a (Å)	9.470(7)	9.476(4)	9.468(8)	9.460(5)	9.490(7)
b (Å)	16.902(7)	16.940(8)	16.91(1)	16.884(9)	16.94(1)
c (Å)	8.929(6)	8.942(4)	8.923(7)	8.905(5)	8.941(7)
V(Å^3)	1429.2	1435.4	1429.0	1422.3	1437.1

1. Curitiba (Ansell et al. 1976)
2. Curitiba (Cesbron et al. 1979)
3. Curitiba (calculated from Fujimori 1981)
4. Curitiba (Svisero and Mascarenhas 1981)
5. Santa Isabel (this paper)

Cu$K\alpha$ radiation (λ 1.5418 Å) and a 114.6-mm Debye–Scherrer camera. Relative intensities were estimated visually. The results are compared with those for the Curitiba lanthanite-(Nd) in Table 4, and unit-cell parameters refined from powder data are shown in Table 5. The relation *a*:*b*:*c* calculated from unit-cell parameters is 0.5602:1:0.5278.

Possible Genesis

Hypotheses for the possible origin of the Curitiba (Coutinho 1955, Cesbron *et al.* 1979) and Santa Isabel lanthanite-(Nd) are highly speculative. The present authors contend that lanthanite-(Nd) was probably precipitated from sulfate-rich, *REE*-bearing solutions that reacted with carbonate in the calcrete. The presence of sulfides and sulfates in several outcrops of the Caçapava Formation (Atencio 1986) suggests that such solutions did exist. The rare earths may have been released by dissolution of heavy minerals found in the host sediments or in the metamorphic and pegmatitic basement. Rare-earth elements may have remained absorbed in laterized soils, claystone, calcrete, organic material, or even in manganese oxide. On the other hand, the Santa Isabel lanthanite-(Nd) occurrence is adjacent to the Parateí fault, and the caliche of the Resende Basin is located on a structural high; these features suggest the possibility of a hydrothermal influence on the caliche and the lanthanite-(Nd). Neither of these hypotheses has been substantiated.

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