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Compositional variation of some rare earth minerals from the Fen complex (Telemark, SE Norway): implications for the mobility of rare earths in a carbonatite system

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ABSTRACT. Rare earth minerals in ankeritic ferrocarbonatite and associated magnetite ore, as well as in hematite–calcite–(dolomite) carbonatite (rødberg) derived from the ferrocarbonatite by post-magmatic alteration, have been studied by electron microprobe. The minerals found are monazite, parisite, and bastnäsite (identifications confirmed by X-ray diffraction) as well as an aluminous silicate with allanite composition, and possibly, synchysite. The rare earth element (*REE*) distribution patterns of the minerals change from light rare earth (*LREE*)-enriched in magnetite ore and ferrocarbonatite to less *LREE*-enriched, or middle *REE*-enriched in the most strongly re-equilibrated carbonatites. Two distinct mineral varieties, monazite-(Ce,Nd) and a Nd-fluorocarbonate, have been found in rødberg. The *LREE* were leached from their host minerals during re-equilibration of the carbonatite, most likely because of the influence of fluoride-bearing hydrothermal solutions.

KEYWORDS: rare earth minerals, monazite, allanite, carbonatite, rødberg, Fen complex, Norway.

WHOLE-ROCK data indicate that some of the carbonatite intrusive units in the early Cambrian Fen complex, Telemark, SE Norway (see Barth and Ramberg, 1966) are strongly enriched in the *REE*, and especially in the lighter elements of the group (Mitchell and Brunfelt, 1975; Möller *et al*., 1980). The highest *REE* contents have been reported from ankeritic ferrocarbonatite (formerly known as 'rauhaugite II', cf. Andersen, 1984) and in rødberg (hematite–calcite–dolomite carbonatite). Andersen (1984) has shown that the rødberg was derived from ferrocarbonatite by post-magmatic alteration processes which may be described as oxidation of ankerite and other ferrous minerals and selective dissolution of carbonates. The alteration processes were induced by the influx of groundwater-derived hydrothermal fluids into the intrusives from the surrounding Precambrian gneisses (Andersen, 1984). Reconnaissance La and Sm data on rødberg and its parent ferrocarbonatite indicate that the whole-rock La/Sm ratio, as well as the La-content, decreased during alteration (Andersen, 1984), suggesting that post-magmatic re-equilibration may be one of the factors controlling the *REE* distribution in carbonatites.

In the Fen carbonatites, total *REE* contents may amount to more than 15 000 ppm, i.e. 1.5 wt. % (Möller *et al*., 1980); the *REE* are therefore minor, rather than trace components. Reconnaissance data indicate that the *REE* are contained in a number of distinct phases. From semi-quantitative microprobe analyses, Åmli (1974) identified monazite and fluorocarbonates of the bastnäsite group as the main *REE*-bearing minerals in both ferrocarbonatite and rødberg. Analyses of mineral separates from ferrocarbonatite have further indicated that apatite has a total *REE* content approaching 1 wt. %, with a weakly *LREE*-enriched chondrite-normalized distribution pattern (Semb, 1971).

In order to understand the processes which control the behaviour of the *REE* elements in carbonatites during re-equilibration, information is needed on the behaviour of the *REE* minerals in a carbonatite system in various stages of alteration. Because the post-magmatic re-equilibration history of the carbonatites in the Fen complex is comparatively well understood (cf. Andersen, 1984), these rocks are well suited for a study of such processes. A study of the compositional variations of *REE* minerals from different rock units in the complex was therefore undertaken. The samples used were selected among the carbonatite and iron ore material described by Andersen (1983, 1984). Samples G 114–20 were collected from a *c*.2 m section through a comparatively unweathered, minor zone of rødberg (Andersen 1984, fig. 2), samples G 101–6 come from a section through

Table I. Petrography of carbonatite and ore samples from the Fen complex.

Sample	Rock type	Locality	Major and minor minerals	Whole rock La (ppm)	Whole rock Sm (ppm)
M 7	Magnetite ore	Tufte adit	Mt, py, qz, chl		
G 101	Ferro-carbonatite	W.Gruveåsen	Ank, chl, qz, py, mt		
G 105	"	"	" " " " "	2912	384
G 106	"	"	" " " " "		
G 113	Rødberg, (strongly altered)	N. Gruveåsen	cc, dol, hm	2914	578
G 114	Rødberg (weakly altered)	"	cc, dol, hm, ank(rel)	1339	268
G 118	Rødberg	"	cc, dol, apt, chl, hm	615	1196
G 119	Ferro-carbonatite	"	Ank, cc, mt, chl	1190	294
G 120	"	"	" " " "	2727	347

Most abundant minerals are underlined. Abbreviations: cc = calcite, dol = dolomite, ank = ankerite, qz = quartz, chl = chlorite, mt = magnetite, py = pyrite, hm = hematite. (rel): relict phase. Source of La and Sm data: Andersen (1984, Table 3).

unoxidized ferrocarbonatite. Sample M 7 is a coarse-grained magnetite–pyrite ore from the abandoned Søve niobium mines. Detailed sample descriptions are given by Andersen (1983, 1984). The most important petrographic features and whole-rock La/Sm data are given in Table I.

Analytical methods. The electron microprobe was used both for qualitative mineral identifications and for quantitative analyses. The minerals were analysed in polished and carbon coated thin sections, with an ARL-EMX microprobe equipped with a LINK energy-dispersive analyser. The raw data were reduced on-line by the commercial program ZAF4/FLS, allowing a maximum of fourteen elements in any run. Analyses of the *REE* standard glasses of Drake and Weill (1972) gave the following results (means in wt. % ± one standard deviation, literature value in parenthesis): La_2O_3 4.52 ± 0.18 (4.28); Ce_2O_3 4.18 ± 0.14 (4.00); Pr_2O_3 4.14 ± 0.28 (4.44); Nd_2O_3 4.12 ± 0.32 (4.26); and Sm_2O_3 3.91 ± 0.10 (4.26). The detection limits for individual *REE* oxides are estimated in the interval 0.5 to 1.0 wt. %. Further details of the analytical procedure are given by Andersen and Neumann (1985). It should be noted that accidental excitation of groundmass carbonates tends to decrease the sums of microprobe analses of fine-grained material like that presently studied.

X-ray diffraction patterns were made with 9 cm Debye–Scherrer cameras, using filtered Fe-$K\alpha$ radiation. The material used was picked from pieces of the samples under a binocular microscope and crushed in ethanol. In most cases, the powder of *REE* minerals was contaminated with other minerals of the carbonatite, leading to composite diffraction patterns.

Results

Identity and occurrence of the REE minerals. In the magnetite ore (sample M 7), *REE* minerals occur as comparatively large interstitial crystals, intergrown with quartz, and chlorite. Microprobe analyses show that the most abundant mineral is parisite, forming colourless to pale yellow, equant crystals. These are commonly rimmed by intergrowths of small, euhedral hematite crystals. Aggregates of elongated, brownish crystals with bastnäsite composition are found closely associated with the parisite. The differences in morphology between the two phases are illustrated in fig. 1. X-ray diffraction data confirm the identification of the minerals.

In the carbonatites, apatite is the most abundant *REE*-bearing mineral, forming subhedral prisms or small, equant grains dispersed in the carbonate groundmass. According to the microprobe data, other abundant *REE* minerals are monazite (confirmed by X-ray diffraction) and Ca-bearing members of the bastnäsite group. In the ferrocarbonatite these minerals occur as very fine-grained aggregates intergrown with chlorite and carbonates, and in crosscutting calcite veins. In the rødberg, *REE* minerals are found intergrown with the matrix phases; none have been observed in calcite veins formed during or after oxidation of the rock. Yellow or light brown colour, moderately high interference colours and, sometimes, fibrous habits usually distinguish them from the matrix phases of the carbonatite, but generally the small grain size

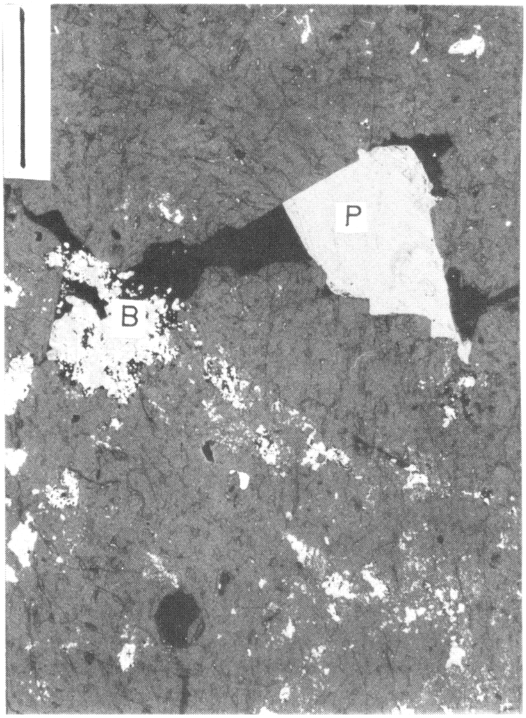

FIG. 1. Electron backscatter photomicrograph showing crystals of *REE* minerals in sample M 7. The crystals occur in interstitial cavities in a magnetite ore. Magnetite is dark grey in the photomicrograph. Voids or holes in the polished thin section are black. The light grey, nearly homogeneous grain in the upper middle of the frame is a crystal of parisite (P). Polycrystalline aggregates of prismatic bastnäsite crystals are seen in the lower part of the same interstitial void (B), and are also scattered in smaller cavities thoughout the area of the photomicrograph (white to light grey, heterogeneous). A Jeol JSM 840 electron microscope at the Centre for Industrial Research, Oslo, was used for the recording. Length of scale bar: 500 μm.

prevents identification of individual phases from optical properties. In one ferrocarbonatite sample (G. 106), accessory amounts of a fine-grained, strongly pleochroic, reddish-brown aluminous *REE* silicate were found. The mineral could not be isolated for X-ray studies.

Composition of the REE minerals. Quantitative microprobe analyses of *REE* minerals are listed in Table II, together with structural formulae. The data refer to means of several grains in each sample. The *REE* concentrations in apatite are too low to allow analysis with the microprobe technique applied. No *REE* heavier than Sm was present in quantities above the detection limits in any of the minerals analysed. As can be seen from Table II, the sums of the analyses tend to be low. The fluorine values cited in Table II are generally lower than expected from stoichiometric compositions, thus contributing to the low sums. Fluorine is, however, a problematic element in energy-dispersive microprobe analysis; the values given should not be considered better than semiquantitative. It should be noted that *REE* patterns calculated from the analyses are not affected by poor totals.

The cation distributions of bastnäsite and parisite from the magnetite ore are close to stoichiometric compositions (bastnäsite, $(REE)CO_3F$; parisite, $(REE)_2Ca(CO_3)_3F_2$) but include considerable amounts of iron, most of which must be divalent to satisfy the charge balance requirements. The fluorocarbonates from carbonatites also correspond to a parisite formula, with two exceptions. The mineral in sample G 118 has a Ca/*REE* ratio between those expected for stoichiometric parisite and synchysite [$(REE)Ca(CO_3)_2F$], and could possibly be better classified as synchysite from its composition. No X-ray diffraction data could be obtained to remove this ambiguity, due to the fine grain-size. The analysis from sample G 119 has an element ratio close to synchysite composition. This analysis is, however, characterized by particularly low fluorine, suggesting that the analysis may contain a considerable X-ray contribution from accidentally excited matrix calcite. Thorium is a minor constituent in most of the analyses, while yttrium could only be detected in fluorocarbonates from three samples.

The monazite is also nearly stoichiometric $(REE)PO_4$ and contains less iron than the fluorocarbonates. The Th content is similar to that in the fluorocarbonates, whereas yttrium was below the detection limit in all grains analysed. The high Ca and P recorded in G 119 are probably due to accidental excitation of apatite intergrown with the monazite.

The aluminous silicate found in sample G 106 has a composition agreeing with the formula of allanite [$(REE)_2(Al,Fe)_3Si_3O_{12}(OH)$]. In this mineral, no *REE* elements heavier than Nd could be detected.

The chondrite normalized distribution patterns of *REE* in the minerals (fig. 2) vary from strongly *LREE*-enriched in the ferrocarbonatite and magnetite ore, to less *LREE*-enriched in the rødberg. The different minerals in each sample have a strong tendency towards parallel distribution patterns. One sample, G 118, shows middle *REE*-enriched distribution patterns for the minerals. From the relative abundances of the elements (Table II), the minerals in this sample should be classified as distinct varieties: monazite-(Ce,Nd) and either synchysite-(Nd) or parisite-(Nd).

Table II. Compositions of rare earth minerals

a: Fluorocarbonates

	G 105	G 106	G 114	G 118	G 119	M 7 BAS	M 7 PAR
	Weight percent oxides						
CaO	8.51	9.18	8.62	13.93	8.43	0.71	8.30
FeO	0.21	2.00	2.89	1.84	9.25	1.68	1.47
La_2O_3	13.17	14.49	14.04	5.42	11.31	24.63	20.66
Ce_2O_3	28.12	28.67	27.80	19.55	23.33	33.44	29.54
Pr_2O_3	3.16	2.69	2.99	3.53	2.40	1.98	1.58
Nd_2O_3	10.62	9.94	10.44	21.18	9.66	6.52	5.88
Sm_2O_3	0.74	1.34	0.85	2.86	0.85	0.56	0.63
Y_2O_3	n.d.	0.85	0.23	0.39	n.d.	n.d.	n.d.
ThO_2	0.52	0.80	1.62	n.d.	1.74	0.64	0.71
F	4.1	4.3	7.2	7.5	1.5	7.4	5.5
S						0.40	0.60
-O=F	1.74	1.82	3.04	3.17	0.62	3.10	2.33
Sum	67.45	72.46	73.65	73.06	67.82	74.83	72.58
	Structural formulae based on 3.000 cations						
Ca	0.918	0.895	0.849	1.255	0.786	0.085	0.844
Fe	0.017	0.153	0.223	0.130	0.673	0.157	0.116
La	0.489	0.486	0.479	0.168	0.362	1.014	0.723
Ce	1.038	0.954	0.936	0.602	0.743	1.366	1.027
Pr	0.117	0.089	0.100	0.108	0.076	0.080	0.054
Nd	0.383	0.322	0.343	0.637	0.300	0.260	0.199
Sm	0.026	0.042	0.027	0.083	0.025	0.021	0.021
Y		0.041	0.012	0.017			
Th	0.012	0.017	0.034		0.034	0.017	0.016
F^-	1.32	1.24	2.10	2.00	0.41	2.60	1.66
S^{2-}						0.062	0.107

b: Monazite

	G 101	G 113	G 118	G 119	G 120
	Weight percent oxides				
CaO	n.d.	0.83	n.d.	15.22	0.74
FeO	2.12	n.d.	0.83	0.35	1.41
La_2O_3	13.97	15.51	6.88	11.42	17.98
Ce_2O_3	32.24	30.97	26.70	25.49	33.73
Pr_2O_3	3.88	3.17	4.82	2.43	3.29
Nd_2O_3	10.96	14.76	26.48	11.25	12.01
Sm_2O_3	1.28	2.12	4.08	1.54	1.06
ThO_2	1.79	0.77	0.20	n.d.	1.26
P_2O_5	25.48	28.35	29.81	33.00	26.92
Sum	91.72	96.48	99.80	100.70	98.40
	Structural formulae based on: 2.000 cations				
Ca		0.036		0.513	0.032
Fe	0.076		0.027	0.009	0.047
La	0.222	0.232	0.100	0.133	0.266
Ce	0.508	0.460	0.384	0.294	0.495
Pr	0.061	0.047	0.069	0.028	0.048
Nd	0.168	0.214	0.372	0.127	0.172
Sm	0.019	0.030	0.055	0.017	0.015
Th	0.018	0.007	0.002		0.011
P	0.928	0.974	0.991	0.880	0.914

c: Allanite

	G 106
	Weight percent oxides
SiO_2	30.48
Al_2O_3	13.85
MgO	0.29
FeO	15.63
CaO	9.22
La_2O_3	7.54
Ce_2O_3	14.92
Pr_2O_3	1.21
Nd_2O_3	3.37
Sum	96.51
	14.000 cations
Si	5.119
Al	2.923
Mg	0.077
Fe	2.341
Ca	1.769
La	0.498
Ce	0.978
Pr	0.079
Nd	0.216

Discussion

The present data confirm the findings of Åmli (1974), indicating monazite and fluorocarbonates of the bastnäsite group as the dominant *REE* minerals in the Fen carbonatites (in addition to apatite, which contains the *REE* as minor components). These minerals are found as minor or moderately abundant accessory minerals both in magnetite ore, ferrocarbonatite and in the rødberg formed from it by post-magmatic re-equilibration processes, and were thus stable throughout the crystallization and alteration history of the carbonatite. Some of the crystals found in the rødberg may have survived re-equilibration as such; others may have formed by recrystallization of precursors belonging to the ferrocarbonatite mineral assemblage. Apatite is a more abundant minor or accessory phase than monazite, fluorocarbonates and allanite in most of the samples studied; its total *REE* content is however below 1.0 wt. % and it is less enriched in light relative to heavy *REE* than any of the presently analysed minerals (Semb, 1971). The abundances and distribution patterns of the *LREE* in the whole-rock systems are therefore largely controlled by monazite and fluorocarbonates. The new results show that the minerals in the ferrocarbonatite generally have steeper *LREE*-enriched distribution patterns than their analogues in the rødberg. The only exception from this is the fluorocarbonate in sample G 114, whose *REE* pattern is comparable to those of the ferrocarbonatite minerals. This sample is, however, only weakly oxidized and is rich in ankerite relics (Andersen, 1984). The grains analysed in this sample may be relict members of the ferrocarbonatite assemblage, rather than equilibrated parts of the rødberg. The whole-rock La and Sm data summarized in Table I suggest that the La/Sm ratio of the whole-rock system decreased during rødberg formation, due to loss of La. The change of *REE* mineral compositions towards less *LREE* enrichment during re-equilibration must therefore be interpreted as a result of redistribution of elements between solid phases and a fluid phase.

Studies of *REE* distributions in minerals of the bastnäsite group indicate that they commonly show distribution maxima for La or Ce (Semenov,

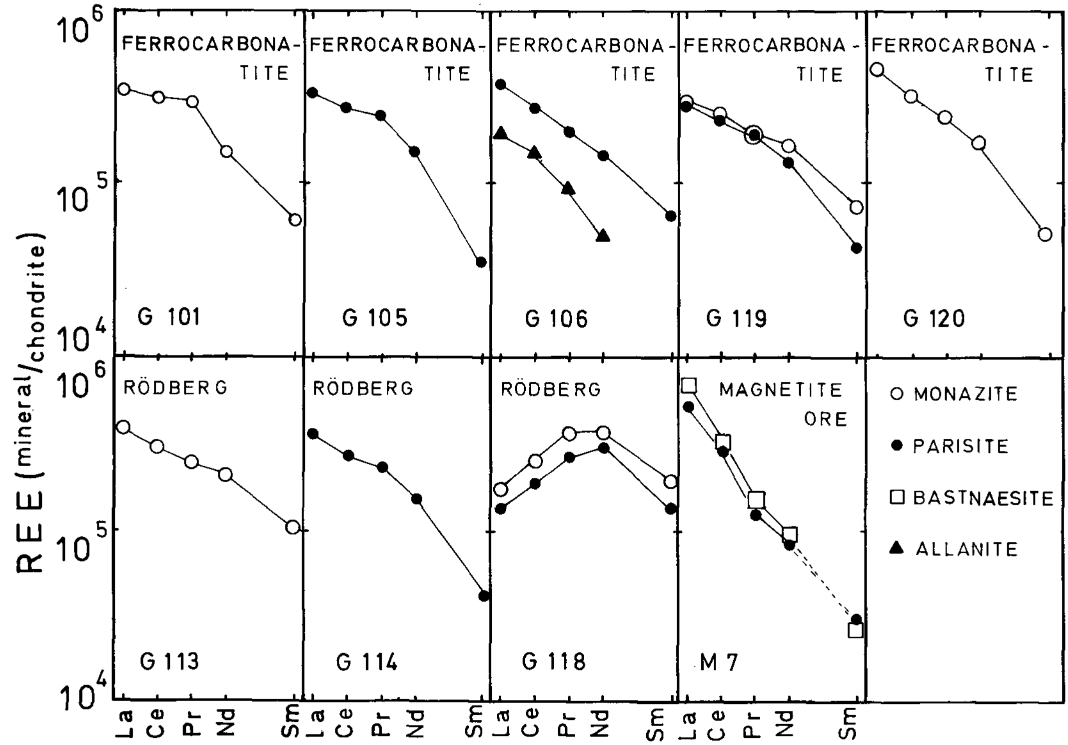

FIG. 2. Chondrite-normalized distribution patterns of the *REE* minerals in Fen carbonatites and magnetite ore. The data in Table II have been normalized to the nine chondrites value of Haskin *et al.* (1968), recalculated to weight percent oxides.

1963; Clark, 1984); varieties which are enriched in yttrium and the heavy *REE* have, however, been described (Clark, 1984). Monazite, on the other hand, is always strongly enriched in the *LREE* but shows little tendency to discriminate between the individual members of that group (Jensen, 1967; Clark, 1984). These findings suggest that the actual *REE* distribution patterns of both of these minerals or mineral groups in general may be a result of an interplay of factors relating to the mineral's structure and to the element distribution in the geological environment.

The pattern of solubility of the *REE* in a fluid relates to differences in the stability of soluble complexes of the elements. The solubilities of *REE* oxides in pure water are low, and decrease from La to Lu, reflecting decreasing stability of aqua- or hydroxy-complexes (Felsche and Herrmann, 1970). Generally, the solubilities are increased by the presence of anionic ligands in the fluid. Inorganic ligands such as PO_4^{3-}, CO_3^{2-}, HCO_3^-, and Cl^- tend to form more stable complexes with the heavy elements, due to their smaller ionic radii (Humphris, 1984). Fluoride ion may be an exception to this pattern. Although fluoride complexes like $(NaYF_4)^0$ enhance the solubility of the heavy *REE* (Mineyev, 1963), thermodynamic data indicate that formation of $(REE)F^{2+}$ and $(REE)F_2^+$ complexes would lead to a uniform increase in oxide solubilities relative to those in pure water. La may also form an even more stable, neutral $(LaF_3)^0$ complex, further increasing its solubility (Alderton *et al.*, 1980). From the small differences in ionic radius between La^{3+} and Ce^{3+} (1.032 *vs.* 1.01 Å; Shannon, 1976) it seems possible that Ce may also be able to form a neutral fluoride complex, although this would be less stable than the corresponding complex of La. Due to this effect, the lighter elements would show the greater tendency to be partitioned into solution during interation between solid *REE*-bearing phases and fluoride-containing hydrothermal fluids.

The presence of fluoride in the late, oxidizing hydrothermal fluids in the Fen complex is indicated by the fact that fluorocarbonates have remained stable during re-equilibration, and by observations of fluorite as a minor phase both in the rocks

(Brøgger, 1921; Sæbø, 1971) and in cross-cutting veins formed during carbonatite re-equilibration (Andersen and Qvale, unpublished data). The evolution of the *REE* in monazite and fluorocarbonates during re-equilibration may therefore best be interpreted as a result of selective distribution of the lighter elements into fluoride-bearing hydrothermal solutions. Any new crystals of *REE* minerals formed at this stage of the rock's evolution would also have *REE* patterns reflecting the tendency of the *LREE* to be partitioned into solution.

Conclusions

The *REE*-rich carbonatites in the Fen complex contain a suite of *REE*-rich minerals (apatite, monazite, bastnäsite, parisite, and possibly synchysite and allanite). The *REE* distribution in monazite and fluorocarbonates varies from strongly La and Ce enriched in the ferrocarbonatite and associated magnetite ore, to less *LREE* enriched in the rødberg. Distinct compositional varieties enriched in middle *REE* (monazite-(Ce,Nd) and a Nd variety of a Ca-bearing fluorocarbonate) have been found in a sample of rødberg.

The differences in *REE* distribution between minerals from unoxidized and oxidized carbonatites are due to secondary removal of the *LREE* from their host minerals during post-magmatic carbonatite re-equilibration, in which the *LREE* were selectively partitioned into soluble fluoride complexes.

The present findings indicate that the *REE* elements may be mobile in a carbonatite system during re-equilibration, even though the phases which contain them remain stable throughout the alteration history. The extent to which the light and/or middle *REE* are partitioned into solution, depends on the nature and concentrations of anionic ligands present in the hydrothermal fluids. Fluoride ion increases the mobility of the lighter elements, probably because of the formation of neutral complexes such as $(LaF_3)^0$.

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