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RHÖNITE FROM BIG BEND NATIONAL PARK, TEXAS

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ABSTRACT

A new occurrence of rhönite is reported from a mafic alkaline sill in west Texas. Chemical analysis and pleochroism closely match those of Rhön, Germany, but refractive indices, birefringence and $2V$ are greater than those previously recorded. X-ray powder data were obtained and a strong similarity to the aenigmatite pattern is observed. Unit cell refinement was made, using adjusted aenigmatite cell parameters and indexing. From the unit cell volume obtained (747.79 Å ±0.23), chemical analysis, and measured specific gravity (3.64 ± .04) it is calculated that the number of oxygens per unit cell is 40, and that the idealized formula is analogous to that of aenigmatite with the substitution of Ca, Al, and Mg for Na, Si, and Fe^{2+} respectively. The formula for the Big Bend rhönite is calculated to be $(Ca_{3.45}Na_{0.38}K_{0.01})$ $(Fe^{2+}{}_{3.60}Fe^{3+}{}_{1.91}Mg_{4.28}Mn_{0.06}Al_{0.15}Ti_{2.84})$ $(Si_{6.68}Al_{5.32})$ $(O_{39.4}OH_{0.6})$. No evidence was obtained concerning possible solid solution between rhönite and aenigmatite.

INTRODUCTION

Rhönite occurs in an alkaline mafic sill in Big Bend National Park, Brewster County, near Terlingua, Texas, and to the best of our knowledge this is the first reported occurrence of the mineral in the western hemisphere. The discovery was made during a general study, currently in progress, of the petrology of the alkaline rocks of the Terlingua region.

Preliminary identification was by optical properties in thin section (particularly strong absorption and characteristic pleochroism) and confirmation was made by wet chemical analysis (Table 4), in which the composition matches very closely that of rhönite from the Rhön region of Germany.

LITERATURE REVIEW

Rhönite was originally described by Soellner (1907) from the Southern Rhön region. His description included the chemical analysis of rhönite from a nepheline basanite (Table 4). Lacroix (1909) presented an analysis of rhönite from the Massif Central, Puy-de-Barneire, Saint-Sandoux, France (Table 4) where it occurs in several varieties of titanaugite-bearing nepheline dolerites. There it is found as crystals, up to several centimeters in length which are partially resorbed and rimmed with titanomagnetite. Ramdohr (1937), in a general discussion of rhönite in reflected light, emphasized its occurrence as small prisms intergrown with augite and magnetite in reaction structures on basaltic hornblende. Tomita (1934) described rhönite as an alteration product of kaersutite in analcime-bearing teschenites from the Oki Islands, Japan. According to Tomita (1934, p. 124), the peripheral alteration of kaersutite passes outward from rhönite through rhönite-aenigmatite, to aenigmatite. Yagi (1953, p. 786) described rhönite occurring as cores in zoned aenigmagite crystals from analcime-bearing syenites and monzonites from the Island of Sakhalin. In a restudy of the Rhön volcanics, Ficke (1961, p. 348) reported that rhönite occurs as an accessory mineral in many mafic volcanic rocks of the region (limburgites, basanites, hornblende basalts, basaltic tephrites, and pipe breccias). It is fine-grained (0.003–0.1 mm), and is the last of the mafic minerals to crystallize. Ficke also referred to a report of rhönite occurring in the Swabian Alps, Germany, where it is one of the last minerals to crystallize following melilite. Babkine *et al.* (1964) presented a microprobe analysis of rhönite from an inclusion of pyroxenite (17% titanaugite, 16% olivine Fa_{55}, 13% spinel) in a basalt from Haute Loire, France (Table 4). Scattered throughout the pyroxenite are small cavities filled with a glass which, according to Babkine *et al.*, was formed by the local melting of augite. From this glass rhönite has crystallized as small prisms not exceeding 0.02 mm in length. They also referred to rhönite occurring as an alteration product of hornblende in basalts of the region.

The rhönite occurrences in the literature indicate that the mineral is found principally in silica-poor, mafic to intermediate rocks. It is commonly accompanied by titanaugite and feldspathoids, and is often an alteration product of amphiboles.

Several authors have commented on the close relation between aenigmatite and rhönite. Fleischer (1936) discussed the formulas of the two minerals, and suggested (p. 348) that rhönite and aenigmatite form an isomorphous series analogous to anorthite and albite. In the Sakhalin occurrence Yagi noted that the rhönite cores are sharply bounded against aenigmatite, and he suggested that the two minerals form a discontinuous reaction series. However, Tomita's description of rhönite grading outward to aenigmatite implies a continuous solid solution. X-ray data, presented in a later section, strongly suggest that the two minerals are similar structurally.

OCCURRENCE

The intrusion in which rhönite is found is Location 159 on the geologic map of Big Bend National Park (Maxwell *et al.*, 1967, Plate II), and is situated about 1½ miles east of Maverick Mountain and the community of Study Butte. The sill is about 300 feet thick and is exposed laterally for 2000 feet. It is composed of two rock types: (1) an ultramafic rock termed ankaramite (Table 1), and (2) a rhönite-bearing melasyenite

TABLE 1. REPRESENTATIVE MODES

	Melasyenite	Ankaramite
Pyroxene	24	50
Olivine	2	14
Rhönite	11	
Biotite		2
Plagioclase	1	15
Alkali Feldspar	13	
Nepheline	8	
Analcite		8
Apatite	1	1
Zeolites	38	3
Opaque Mineral(s)	2	7
	100	100

which overlies the ankaramite. Field relations and mineralogical data suggest the melasyenite was formed by differentiation essentially *in situ*.

A representative mode of the melasyenite is shown in Table 1. The rock type has a variable texture, but is usually porphyritic with relatively large (3–6 mm) elongate phenocrysts of titanaugite, and smaller phenocrysts of rhönite, olivine (Fa_{65}), nepheline, and plagioclase (An_{55}), set in a microcrystalline groundmass of alkali felspar microlites and zeolites. The petrology and structure of the intrusion will be discussed in a subsequent paper.

The rhönite is found only in the melasyenite, but in this rock type it is characteristically present in significant amounts (5–15%). The phenocrysts of rhönite are prismatic and average 2 mm in length, although crystals up to 4 mm in length have been observed. Rhönite, titanaugite, and plagioclase crystallized early preceding nepheline and alkali feldspar. The rhönite is commonly intergrown with titanaugite, and rarely with olivine. All of the early formed mafic minerals show evidence of reaction with the later liquids; the olivine is strongly resorbed and the titanaugite has narrow, green, iron-rich rims. The rhönite commonly has reaction coronas of a granular opaque mineral, and is also replaced by an extremely fine-grained mixture of sphene (?), fibrous green chlorite (?), and a carbonate mineral.

OPTICAL PROPERTIES

The optical properties of the analysed rhönite are summarized in Table 2. The rhönite has a very strong absorption, and it is opaque in thin-section of normal thickness (30 μm). Where the sections are very thin ($\sim$10 μm), however, the mineral is seen to be strongly pleochroic

TABLE 2. PHYSICAL PROPERTIES OF RHÖNITE

Optical	
$\alpha=1.795\pm0.005$	dk. greenish brown
$\beta=1.806\pm0.005$	dk. reddish brown
$\gamma=1.83\ \pm0.01$	v. dk. reddish brown to opaque $X<Y<Z$
$\gamma-\alpha\cong0.035$	
$2V_z=66°$ (observed)	
$2V_z=65°$ (calculated)	
$r<v$ extreme	
Density 3.64 ± 0.04 (measured)	

and usually shows polysynthetic twinning. The optical properties of rhönite are difficult to determine precisely because of its strong absorption and extreme dispersion. The average $2V_z$ of seven grains from different rocks is 66° (range 62–73), determined by universal stage measurement of V using sodium light. Strong inclined dispersion together with dispersion of $2V$ caused one isogyre to be too weak for measurement. The isogyres were broad because of the unusually thin sections but locations of centers of isogyres and bisectrices were repeatable to ±1° and multiple readings were averaged in all cases using the basic technique of Fairbairn and Podolsky (1951). Optically zoned grains showed considerable diminution of $2V$ in a thin border region (values down to 47°) and the range of $2V$ observed in the main parts of the grains is thought to be a result of compositional variation.

The indices of refraction were determined in sodium light using the orthoscopic spindle stage technique described by Wilcox (1959, p. 1282). Perhaps fortuitously, $2V$ calculated from the indices agrees quite well with the observed value.

The optical properties of rhönite have been summarized by Tröger (1959, p. 65, 78), and Winchell and Winchell (1951, p. 478). It should be noted that the values for the indices of refraction and birefringence for the West Texas rhönite are greater than those listed for rhönite by Tröger (p. 65).

METHOD OF SEPARATION

To separate rhönite for analysis, the rock was pulverized to less than 230 mesh and the light fraction separated with tetrabromoethane. Magnetite was extracted from the rhönite-bearing heavy fraction by hand magnet. The remaining rhönite-titanaugite mixture was further split on a Franz isodynamic separator, and the rhönite was finally purified by centrifuging in Clerici solution. Examination of the purified material with the petrographic microscope revealed less than 1 percent impurities.

X-RAY ANALYSIS

Six runs of the purified rhönite were made at a speed of 0.50° 2θ/minute on a Norelco diffractometer equipped with an AMR monochrometer, using purified silicon powder ($a_0 = 5.4300$ Å, calibrated against Gibb's spinel standard) as an internal standard. Operating conditions were: 35 KV, 20 mA, scale factor of 2, multiplier of 1, and a time constant of 4. Two of us (MFC & JCB) each measured three of the patterns. In general, agreement was within 0.02° 2θ and the averaged result is given in Table 3, first column.

Comparison of the rhönite data with that of aenigmatite reveals similarities in 25 *d*-spacings and relative intensities (with most rhönite spacings shifted to longer values). Kelsey and McKie (1964, p. 990) noted that only the three lines of aenigmatite with the longest *d*-spacing were indexable due to shape and dimensions of the unit cell. However, their unit cell data reported in the A.S.T.M. powder diffraction index file (#16–377) has been indexed (by LGB), although only five of the 38 reported *d*-spacings were assigned unique indices.

Considering the X-ray data and the suggested chemical similarities between rhönite and aenigmatite (Fleischer, 1936) it was decided to attempt a preliminary unit cell determination using a least squares unit cell refinement computer program with a variable indexing option (Evans, Appleman and Handwerker, 1963). Starting parameters were based on those of aenigmatite adjusted for the percentage change of unit cell parameters of other silicates involving the same Ca+Al for Na+Si substitution that has been suggested for rhönite-aenigmatite (Fleischer, 1936). The five uniquely indexed aenigmatite lines left only one ambiguity for fixing starting indices. Both sets of ambiguous indices were tried and refinements differed by less than standard errors.

Since a single split of the sample was used, the true precision of the unit cell parameters probably is not as great as indicated by the standard errors shown in Table 3a. We feel that the best estimate of limits for the parameters is two to three times the reported standard errors. We conclude that the above mentioned cell, while not substantiated by single crystal data, provides a satisfactory starting point for future work.

SPECIFIC GRAVITY

Specific gravity was determined with fine (<230 mesh) grains in Clerici solution, centrifuging and with the method of Jahns (1939) using a small glass settling tank mounted on a petrographic microscope. The Clerici solution was calibrated both by index of refraction and by weighing an exact volume in a micropipette. There is a range of specific gravity from 3.60 to 3.76 and the average value for most grains is 3.64±.04. The

TABLE 3. X-RAY POWDER DIFFRACTION DATA OF RHÖNITE

d (measured) (Av. of six runs)	d (calculated) unit cell refinement	h k l	I/I_0
8.14	[a]		24
7.50	[a]		7
6.48	6.47	$1\ \bar{1}\ 1$	4
4.832	4.834	$\bar{2}\ 2\ 0$	22
3.808	3.811	$2\ 0\ 0$	4
3.735	3.783(R1)	$\bar{1}\ \bar{1}\ 2$	11
3.468	3.464	$\bar{3}\ 2\ 0$	17
3.216	[a]		10
2.947	2.946	$\bar{1}\ 2\ 2$	100
2.831	2.830(R2)	$0\ 0\ 3$ $0\ \bar{2}\ 3$	10
2.783	[a]		31
2.691	2.686(R1)	$\bar{3}\ 0\ 1$	65
2.550	2.550	$1\ 1\ 2$	69
2.470	2.470	$0\ 2\ 2$	22
2.463	2.462	$2\ 0\ 2$	4
2.428	[a]		15
2.332	2.333	$\bar{2}\ 4\ 1$	20
2.108	2.107	$2\ 2\ 0$	4
2.092	2.093	$\bar{2}\ \bar{1}\ 4$	56
1.991	1.993(R2)	$4\ \bar{2}\ 2$ $2\ \bar{3}\ 4$	25
1.950	[a]		14
1.884	1.885	$0\ 1\ 4$	4
1.816	1.815(R2)	$0\ \bar{2}\ 5$ $4\ \bar{6}\ 1$	4
1.750	1.750	$\bar{3}\ 3\ 4$	9
1.725	1.725(R2)	$\bar{4}\ \bar{1}\ 2$ $4\ 1\ 0$	8
1.616	1.616	$2\ \bar{2}\ 5$	12
1.600	1.600	$0\ \bar{4}\ 5$	13
1.498	1.498	$\bar{2}\ \bar{4}\ 3$	38
1.495	1.495(R2)	$\bar{6}\ 5\ 3$ $3\ \bar{5}\ 5$	6
1.486	1.486(R2)	$7\ 5\ 0$ $\bar{3}\ 3\ 5$	17
1.468	1.468	$0\ 1\ 1$	17

[a] Not used in refinement because either lines were broad or 2θ varied more than 0.04° on six patterns.

R1—Rejects unit cell choice (refinement assignment) 2/25

R2—Rejection due to another line (lines) within 2θ tolerance (.04031 during 4th cycle) 6/25

TABLE 3a. UNIT CELL REFINEMENT SUMMARY

a_0	10.478 ± 0.002 Å	a^*	0.131288 ± 0.000024 Å^{-1}
b_0	10.923 ± 0.005 Å	b^*	0.127547 ± 0.000038 Å^{-1}
c_0	9.170 ± 0.002 Å	c^*	0.117783 ± 0.000022 Å^{-1}
α	$101°25.8 \pm 1.4$	α^*	$68° 52.0 \pm 1.1$
β	$96°56.7 \pm 1.7$	β^*	$70°50.7 \pm 1.3$
α	$129°41.4 \pm 0.9$	α^*	$47°4.7 \pm 0.9$

Volume $= 747.79 \pm 0.23$ Å^3
$a:b:c = 0.9593:1.0000:0.8395$

range in specific gravity supports the suggestion of compositional zoning shown by optics.

CHEMICAL ANALYSIS

Two grams of the purified rhönite were submitted for standard wet chemical analysis to the Japan Analytical Chemistry Research Institute. Results of the analysis are shown in Table 4. In addition, analyses of three rhönites described by Soellner (1907) (2), Lacroix (1909) (3), and Babkine *et al.* (1967) (4), an aenigmatite from Khibinite quarry, east of Kirovsk, Kola (Kelsey and McKie, 1964, p. 992) (5), and an average value from four recent analyses of aenigmatite (6) are included for comparative purposes.

The close relation between rhönite and aenigmatite has already been noted. Kelsey and McKie (1964, p. 995) suggested the idealized formula:

$$Na_4(\overset{2+}{Fe}_{10}Ti_2)Si_{12}O_{40}$$

for aenigmatite and pointed out that the rhönites described by Soellner and Lacroix contained larger amounts of Al, Ca, Mg, and Fe^{3+} than aenigmatite. Fleischer (1936, p. 364) stated that rhönite is a calcium-aluminum variety of aenigmatite, although he based his formula on 42 oxygens/unit cell. At least a partial solid solution series between aenigmatite and rhönite was implied (via a coupled substitution of Na+Si for Ca+Al). Zies (1966, p. 204), in a study of cossyrite (aenigmatite) from Pantelleria, noted that his analysis closely approached the idealized formula of Kelsey and McKie.

Based on the average of the measured specific gravity (3.64), chemical analysis, and unit cell volume (747.79 Å^3) from the unit cell refinement the number of oxygens per unit cell is 40.57 for the West Texas rhönite. If one assumes 42 oxygens per unit cell (Fleischer, 1936) the specific gravity calculates to 3.79, far outside the possible error in measuring the specific gravity. Therefore we assume 40 oxygens and 28 cations per unit

TABLE 4. COMPARISON OF RHÖNITE AND AENIGMATITE ANALYSES

	1	2	3	4	5	6
	Rhönite	Rhönite	Rhönite	Rhönite	Aenigmatite	
	Big Bend, West Texas	Rhön, Germany	Saint-Sandoux, France	Haute-Loire, France	Kola	"Average" Aenigmatite
SiO_2	24.82	24.42	30.90	29.8	39.62	40.76
TiO_2	9.09	9.46	8.04	10.2	9.66	8.93
Al_2O_3	17.24	17.25	17.65	13.6	0.64	0.62
Fe_2O_3	9.48	11.69	6.80	21.20	4.64	2.90
FeO	15.98	11.39	15.20		33.92	36.87
MnO	0.26	tr	nd	0.1	2.46	1.62
MgO	10.67	12.62	9.08	14.4	1.65	0.90
CaO	11.97	12.43	12.20	11.4	0.44	0.36
Na_2O	0.72	0.67	0.76	nd	7.20	7.09
K_2O	0.02	0.63	0.61	nd	0.04	0.06
H_2O^+	0.35	nd	0.20	nd	0.05	—
H_2O^-	0.06	nd	nd	nd	nil	—
P_2O_5	0.03	nd	nd	nd	nd	—
Cl	nd	nd	nd	nd	0.02	—
F	nd	nd	nd	nd	nil	—
	100.69	100.56	101.44	100.7	100.34	100.11

(1) Analyst: H. Asari, Japan Analytical Chemistry Research Institute

(2) Analyst: M. Dittrich, in Soellner (1907)

(3) Analyst: Pisani, in Lacroix (1909)

(4) Electron microprobe analysis by J. C. Babkine, et al. (1967)

(5) Aenigmatite from split of same sample as used for single crystal determination of unit cell. Analyst, J. H. Scoon, in Kelsey and McKie (1964).

(6) Average of 4 recent analyses: 2A and 3A from Carmichael (1962, p. 96); 1 and 2 from Kelsey and McKie (1964, p. 992).

cell for comparing rhönite and aenigmatite. This is just four times the formula proposed by Machatschki (1953).

Table 5 contains structural formulas of the three rhönites 1, 2, & 3 from Table 4. Placement of the ions in their respective sites follows the reasoning of Kelsey and McKie (1963, p. 993). Kelsey and McKie concluded that $(OH)^-$ is not an essential constituent of aenigmatite; a similar statement concerning rhönite must await structural analysis.

If, as Fleischer suggested (1936, p. 346–348), rhönite is related to aenigmatite by substitution of Ca+Al for Na+Si and if both end members contain the same number of anions, the data reported in Table 5 suggest the following generalized formula:

$$\overset{X}{(Ca, Na, K)_4}\overset{Y}{(Fe^{2+}, Fe^{3+}, Mg, Al, Mn, Ti)_{12}}\overset{Z}{(Si, Al)_{12}}O_{40}.$$

Kelsey and McKie (1963, p. 993) concluded (based on the average of the four recent aenigmatite analyses, 6 of Table 4) that the *Y* cations can be written as $(Al, Fe^{3+}, Mg)_{0.84}\,Ti^{1}_{1.92}\,Fe^{2+}_{8.79}\,(Mn, Ca)_{0.46}$ which corresponds to an average of 28.4 charges contributed by the *Y* cations. For the three rhönites, the charges contributed by the *Y* cations are 29.4 (1), 29.8 (2), and 28.2 (3) for an average of 29.2.

TABLE 5. FORMULAS OF ANALYZED RHÖNITE

	X	*Y*	*Z*
(1)	$[Ca_{3.45}Na_{.38}K_{.01}]$	$[Fe^{2+}_{3.60}Fe^{3+}_{1.91}Mg_{4.28}Mn_{.06}Al_{.15}Ti_{1.84}]$	$[Si_{6.68}Al_{5.32}](O_{39.4}OH_{.6})$
(2)	$[Ca_{3.56}Na_{.35}K_{.22}]$	$[Fe^{2+}_{2.55}Fe^{3+}_{2.35}Mg_{5.03}Ti_{1.90}]$	$[Si_{6.54}Al_{5.44}]O_{40}$
(3)	$[Ca_{3.40}Na_{.38}K_{.20}]$	$[Fe^{2+}_{3.30}Fe^{3+}_{1.33}Mg_{3.52}Al_{1.44}Ti_{1.57}]$	$[Si_{8.03}Al_{3.97}](O_{39.6}OH_{.4})$
(1)	$\Sigma X = 3.84$	$\Sigma Y = 11\ 85$	$\Sigma Z = 12.00$
(2)	$\Sigma X = 4.13$	$\Sigma Y = 11.83$	$\Sigma Z = 11.98$
(3)	$\Sigma X = 3.98$	$\Sigma Y = 11.17$	$\Sigma Z = 12.00$

1. Big Bend, Texas
2. Rhön, Germany
3. Saint-Sandoux, France

If, as suggested by the analyses of rhönite, Ca approaches 4, and if the *Y* cation substitutions are neglected for simplicity, then the composition of an intermediate member of Fleischer's proposed substitution series would be given by the expression

$$(Ca_{1-x}(Na + K)_x)_4 Y(Al_{1-x}Si_x)_4Si_8O_{40}$$

Substitution of the appropriate values of x from Table 5 enables a crude evaluation of the proposed scheme. In all cases, the amount of Si predicted is greater than that observed. This discrepancy is particularly large for (1) and (2); 8.39 versus 6.68 and 8.57 versus 6.54 respectively. If Fleischer's cell is used, the same order of discrepancy is observed.

Available information thus suggests that if rhönite and aenigmatite are members of a solid solution series the mechanism of substitution is more complex than coupled Si+Na for Ca+Al. Based on admittedly limited data, it is proposed that if Al is present in amounts greater than predicted (i.e. Si is less than predicted) from the simple formula given above, then the charge deficiency is balanced by substitutions in the *Y* cation site. Comparisons of charges contributed by the *Y* cations with agreement between predicted and observed Al and Si contents suggest the possibility of such a scheme. The two highest Al and lowest Si cases show 29+ charges instead of the theoretical 28. Consideration of interrelated substitutions on the *Y* sites will necessitate a more complex model than that given by the substitutional formula used above. Ernst (1962) synthesized aenigmatite during a study of the system Na-Fe-Si-O-H. It appears that even titanium is not an essential constituent of aenigmatite.

In conclusion, we agree with Fleischer that rhönite and aenigmatite represent a substitutional series. The details of cation substitution must await further study and there may be a number of different substitutional schemes for the rhönite-aenigmatite series.

Chemical analyses of natural aenigmatites and rhönites suggest that there is only limited solid solution between these minerals at magmatic temperatures. All analyses closely approach end members; no analyses of minerals of intermediate composition have been reported. In this regard a microprobe study of the zoned crystals reported by Tomita (1934) and Yagi (1953) would be of interest.

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