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Ti-bearing aenigmatite from Djinga Tadorgal (Adamawa plateau) and Sao Tomé (Cameroon Line) phonolites: geochemical implications and application of the QUILF thermobarometer for the crystallization conditions

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ABSTRACT

The chemical and structural compositions of aenigmatite from Djinga Tadorgal and Sao Tomé phonolites were compared to those previously described in the felsic lavas and nephelinite from the Adamawa Plateau and Cameroon Line, in order to determine their crystallization temperatures and pressures. Chemical analysis results and the application of QUILF thermobarometer, indicated the equilibrium temperatures between 655 °C and 791 °C, oxygen fugacity (fO_2) below the FMQ buffer, pressures near 0.1 GPa and silica activities less than 1.0 for Djinga Tadorgal and Sao Tomé phonolites. The Ti-bearing aenigmatite from Djinga Tadorgal and Sao Tomé phonolites are a late and accessory mineral, resulting from the reaction between Ti-rich magnetite and Na_2O-rich magmatic liquid.

Keywords: Peralkaline lavas, mineral chemistry, silica activity, thermodynamic conditions.

INTRODUCTION

Djinga Tadorgal is a stratovolcano (altitude: 1747 m), located northern of the Congo Craton (western sector of the Adamawa plateau). Sao Tomé is an island (altitude: 2024 m) of the Gulf of Guinea (Figure 1), ocean sector of the Cameroon Line. Cameroon Line, recently considered as "Cameroon Hot Line" (Déruelle et al., 2007), and Adamawa plateau are tectono-magmatic structures, respectively oriented N30°E and N70°E (Figure 1). The Djinga Tadoral stratovolcano consisting of trachytic and phonolitic lavas, and several basaltic flow units (Mbowou et al., 2010), is a volcanic complex. Sao Tomé Island is essentially characterized by the occurrence of basaltic and felsic lavas associated with palagonotic tuffs (Caldeira et al., 2004).

Aenigmatite crystals were previously described in felsic lavas (trachytes, phonolites, rhyolites) and nephelinites from both Adamawa plateau (Ngaoundere) (Temdjim et al., 2004; Nkouandou et al., 2008) and

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"Cameroon Hot Line" [Oku (Lissom, 1991); Mts Bambouto-Bamenda (N'ni, 2004); Mt Bamenda (Kamgang et al., 2007); Rumpi Mt (Nkoumbou, 1990); Mt Etinde (Nkoumbou et al., 1995); Kapsiki plateau (Ngounouno et al., 1997); Benue Valley (Ngounouno et al., 2003)]. Firstly discovered and analyzed from the nepheline-bearing syenite by Breithaupt (1866); these crystals of the triclinic system and belonging to aenigmatite-rhönite group (Kunzmann, 1999) are rare in felsic alkaline or peralkaline lavas, produced by differentiation of basaltic magma in continental or oceanic domains (Grapes et al., 1979).

The objective of this first description of the aenigmatite from Djinga Tadorgal strato-volcano and Sao Tomé volcanic island was to compare their chemical and structural compositions, to those of felsic alkaline lavas and nephelinite from other parts of the Adamawa plateau and the Cameroon Line, in order to identify the petrologic implications and thermodynamic conditions of crystallization of this mineral.

MATERIALS AND METHODS

Mineral phases (aenigmatite, clinopyroxene, Fe–Ti oxides, feldspar, nepheline) analyses were performed on the electron microprobes CAMEBAX SX50 and SX100 at the 'Université Pierre-et-Marie Curie, Paris'. The measurements were made according to standard analyzed data, under the conditions expressed in kV (accelerating voltage), nA (beam current) and s (counting times at the peak). *Aenigmatite* (15 kV, 40 nA, 10 s for Si and Fe, 15 s for Ca and 20 s for all other elements, *clinopyroxene* (15 kV, 40 nA, 20 s for Si, Al, Fe, Mg, Ca, Na, Mn and 30 s for Ti and Zr), *Fe–Ti oxides* (15 kV, 40 nA, 40 s for Ti, Fe, Mn, Mg; 10 s for Si, 15 s for Cr and 30 s for Al), *feldspar* and *nepheline* (15 kV, 10 nA, 5 s for all elements).

Measurements correction was carried out using the "PAP" program (Pouchou and Pichoir 1991). Analyses are given in terms of oxides of the elements (wt. % = weight %).

Whole-rocks chemical analysis of phonolites (Table 2) was performed at the 'Centre de Recherches Pétrographiques et Géochimiques' (CRPG) of Nancy (France), where the samples were previously selected in order to limit superficial contamination, then crushed. Details of other analytical processes were presented elsewhere (Carignan et al., 2001). Major elements compositions were determined by inductively coupled plasma–atomic emission spectrometry (ICP-AES). Precision is 0.5% for major element oxides.

The program QUILF95 (Andersen et al., 1993) called QUILF thermobarometer (White et al., 2005; Ren et al., 2006) has been used in application for the Djinga Tadorgal and Sao Tomé phonolites, to determine their thermodynamic conditions of crystallization during the eruption. For each sample, two calculations were performed, one with pressure (P) fixed at 0.1 GPa while the silica activity ($aSiO_2$) was allowed to float (i.e., set as a trial value), and the other with $aSiO_2$ fixed at 0.1 and 0.2 while the temperature (T) was allowed to float. The minerals used (clinopyroxene, Fe-Ti oxide) for calculations, had not known subsolidus re-equilibrium as showed by their unzoned and homogeneous compositions (Table 2). The $aSiO_2$ (quartz saturation) varied with pressure and the temperature determined from clinopyroxene–Fe-Ti oxide equilibria was strongly pressure (P)-dependent. Thus, if either $aSiO_2$ or P is fixed, the other may be calculated if T is known with QUILF. Oxygen fugacity (fO_2) can also be calculated and even $\Delta FMQ = \log fO_2 - FMQ(T,P)$.

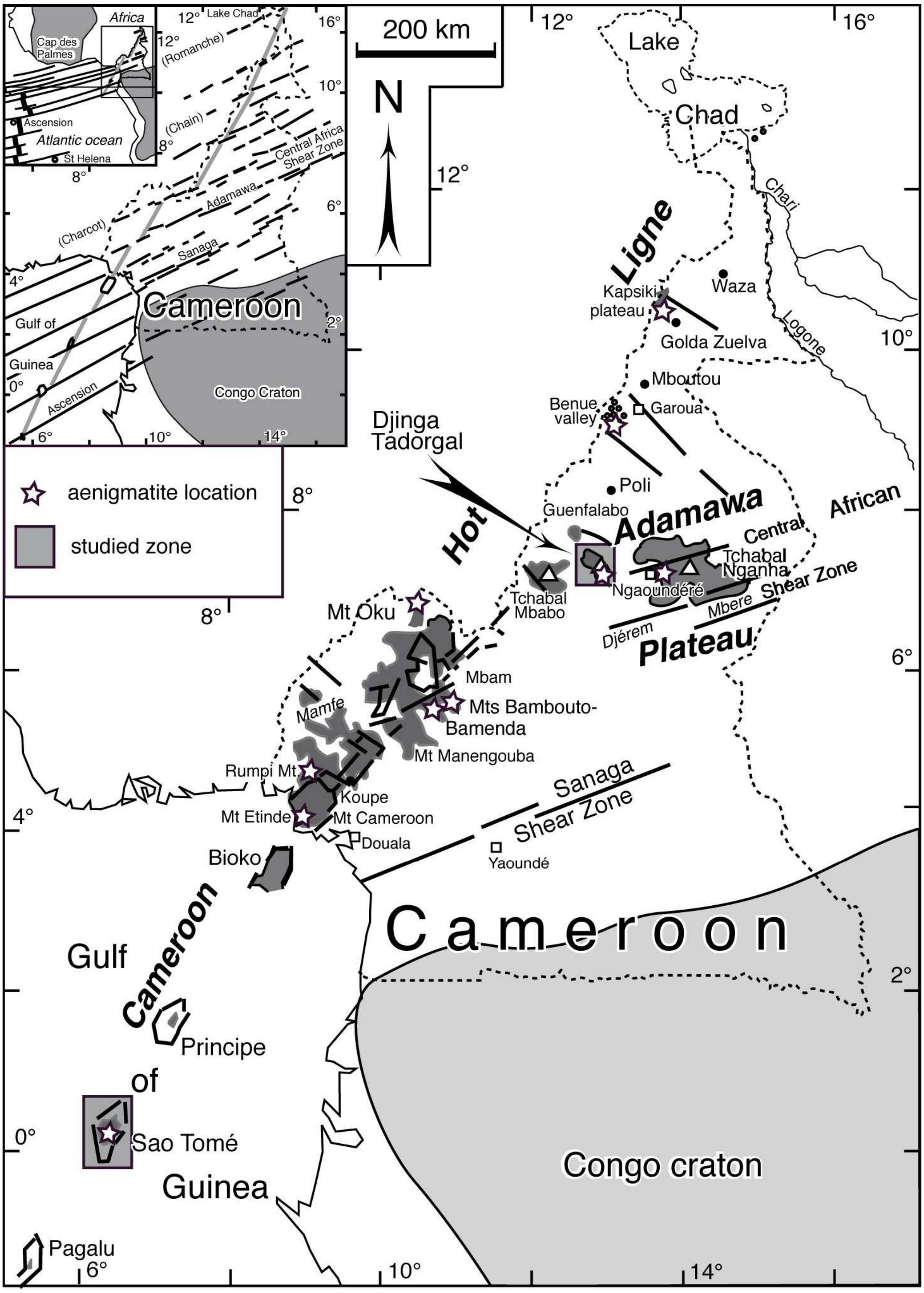

Figure 1: Location of the studied zone, sampling sites of aenigmatite-bearing rocks and tectono-magmatic settings of the "Cameroon Hot Line" and the Adamawa plateau (Déruelle et al., 2007).

RESULTS

Petrography of phonolites

The phonolites from Djinga Tadorgal and Sao Tomé are porphyritic laves with a fluidal aspect, which consisted of sanidine-anorthoclase (up to 4 vol.% = volume %), aegirine augite (~ 1.5 vol.%), aegirine (~ 0.5 vol.%), aenigmatite (~ 1 vol.%), nepheline (~ 3 vol.%) and Ti-magnetite (~ 2 vol.%) phenocrysts, in a groundmass (~ 88 vol.%) of the same microlitic crystals. Aenigmatite phenocrysts are reddish brown and show Ti-magnetite inclusions. Aenigmatite microlites occur sometimes as small interstitial crystals between the slats and microcrystals of aegirine and sanidine-anorthoclase.

Chemical characteristics of aenigmatite

The TiO_2- (up to 8.7 wt.%) and Na_2O- (up to 7.1 wt.%) rich aenigmatite from Djinga Tadorgal and Sao Tomé, showed high FeO*contents (up to 40.5 wt.%) and low CaO (< 0.7 wt.%). FeO* contents of aenigmatite from Oku reaching 42.8 wt.%. However, TiO_2 contents of the aenigmatite from the felsic lavas of Oku (TiO_2: up to 8.5 wt.%), Mts Bambouto (TiO_2: up to 8.1 wt.%), Rumpi Mt (TiO_2: up to 8.4 wt.%) and Ngaoundéré (TiO_2: 6.4 wt.%) are similar close to those from Djinga Tadorgal and Sao Tomé (Table 1). Na_2O contents for the aenigmatite from the Djinga Tadorgal and Sao Tomé (Figure 2) have more or less the same behavior as those of the whole Adamawa plateau (Na_2O: up to 7.0 wt.%) and "Cameroon Hot Line" (Na_2O: up to 7.6 wt.%; Table 1).

High Fe^{3+} (0.4—0.9 apfu) contents in aenigmatite from Djinga Tadorgal and Sao Tomé were characterized by the presence of Fe^{3+}-Tschermak (up to 4%), Fe^{3+}-aenigmatite (up to 23%) and Fe^{3+}-Al aenigmatite (up to 34%) components in their respective sites, like those of other aenigmatites of the Cameroon Line and the Adamawa Plateau. The aenigmatite is a silicate chain of theoretical formula: $X^{VIII}{}_2Y^{VI}{}_6Z^{IV}{}_6O_{20}$ with X = Na^+ , Ca^{2+} ; Y = Mg^{2+}, Fe^{2+}, Fe^{3+}, Ti^{4+}, Al^{3+}, Mn^{2+}, Cr^{3+}, Ti^{3+}, Ca^{2+}, Sb^{5+} Nb^{5+} As^{5+} and Z = Si^{4+}, Al^{3+}, Fe^{3+}, Be^{2+}, B^{3+}. The distribution of major elements of the Sao Tomé and Djinga Tadorgal aenigmatite is illustrated by the histograms (Figure 3), referring to the structural formula. Apart from the high Al^{IV} (up to 0.40 apfu) and Mg^{VI} (up to 0.24 apfu) contents in the aenigmatite from Sao Tomé and Djinga Tadorgal respectively, the behavior of all other major elements were similar, even for the formerly studied Cameroon Line aenigmatite phases than for the Adamawa Plateau.

Thermodynamic conditions of crystallization

QUILF thermobarometer results for T, P, fO_2, and $aSiO_2$ are summarized in Table 3 and Figure 4. It suggests equilibrium temperatures between 655 °C and 791 °C for Djinga Tadorgal and Sao Tomé samples, oxygen fugacity below the FMQ buffer, pressures near 0.1 GPa, and silica activity less than 1.0. These thermobarometric results are likely the magmatic conditions few times before the eruption of the Djinga Tadorgal and Sao Tomé phonolitic lavas and also likely the magmatic condition during the crystallization of aenigmatite (Figure 4). Thus, these magmatic conditions could be considered as the stability domain of the aenigmatite, which crystallized in the Djinga Tadorgal and Sao Tomé samples with the other phases as showed by the petrographic observations.

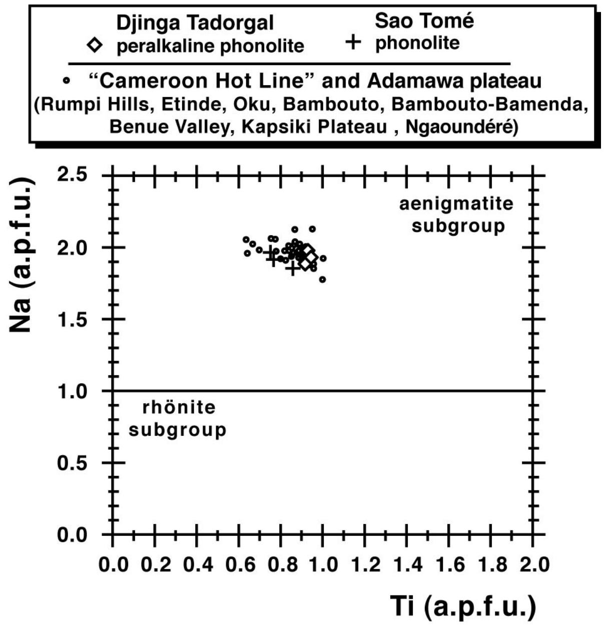

Figure 2: Differentiation of aenigmatite and rhönite subgroups. The compositions of the Djinga Tadorgal and Sao Tomé aenigmatite are represented in comparison to those of other areas of the Adamawa plateau and the "Cameroon Hot Line" (see text for references).

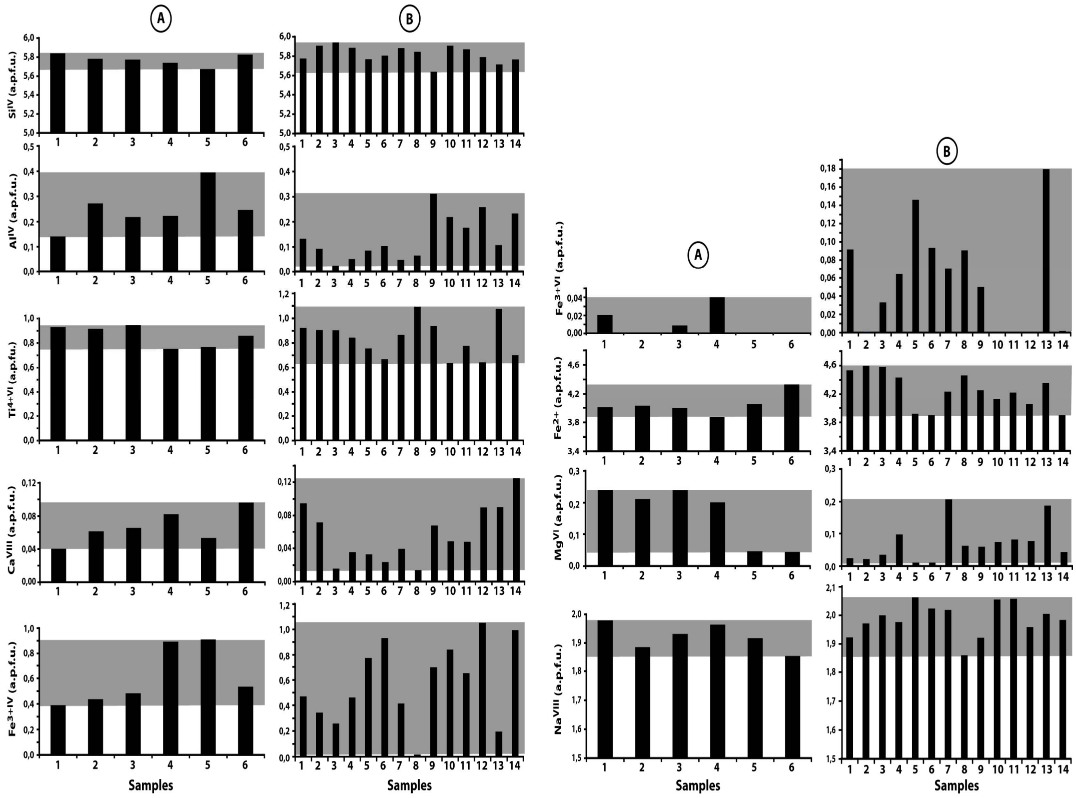

Figure 3: Distribution of major elements in the histograms, refering to the structural formula of aenigmatite. (a) Djinga Tadorgal and Sao Tomé, (b) other areas of the Adamawa plateau and the "Cameroon Hot Line".

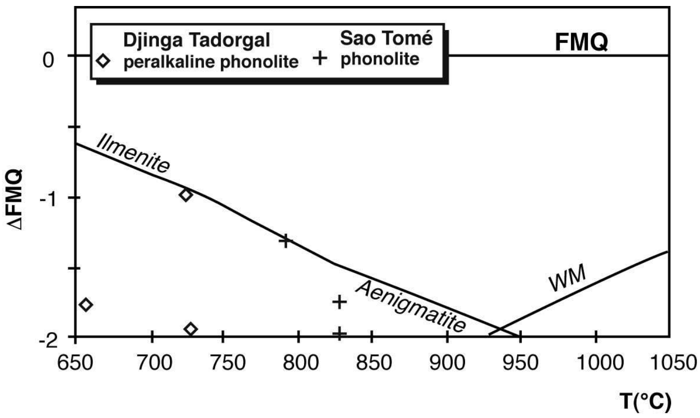

Figure 4: Temperature and oxygen fugacity data from the Djinga Tadorgal and Sao Tomé phonolites with the aenigmatite—Ilmenite and wüstite—magnetite (WM) stability curves (White et al., 2005) plotted relatively to the FMQ buffer (ΔFMQ; Frost et al., 1988).

Table 1: Representative chemical compositions and structural formula (formula proportions based on 14 cations and 20 oxygens) of aenigmatite from Adamawa Plateau and the "Cameroon Hot Line" lavas.

Location	**Djinga Tadorgal**			**Sao Tomé**			**Oku**		
Lava type	**Peralkal. Phonolite**			**Phonolite**			**Rhyolite**		
Sample	**DT113**			**ST44**			**LJ04E**		
SiO_2 (wt.%)	40.42	40.05	40.10	39.58	38.73	39.93	39.95	40.54	40.96
TiO_2	8.56	8.44	8.73	6.90	6.96	7.82	8.48	8.25	8.26
Al_2O_3	0.83	1.60	1.29	1.30	2.29	1.43	0.78	0.54	0.15
FeO*	36.58	37.00	37.30	39.67	40.53	39.88	42.13	40.57	40.21
MnO	4.09	4.17	3.96	2.99	2.98	2.91	1.02	1.20	1.69
MgO	1.11	0.98	1.11	0.93	0.21	0.20	0.11	0.10	0.16
CaO	0.26	0.40	0.43	0.53	0.34	0.62	0.61	0.46	0.10
Na_2O	7.07	6.74	6.92	6.99	6.75	6.56	6.86	6.98	7.11
K_2O	0.04	0.43	0.00	0.07	0.43	0.07		0.02	0.08
Sum	98.96	99.80	99.84	98.95	99.23	99.42	99.94	98.66	98.72
Si (apfu)	5.838	5.780	5.773	5.738	5.672	5.827	5.775	5.906	5.941
Ti	0.930	0.917	0.945	0.752	0.767	0.858	0.922	0.904	0.901
Al	0.141	0.272	0.218	0.222	0.395	0.246	0.133	0.093	0.026
Fe^{3+VI}	0.389	0.436	0.483	0.893	0.911	0.535	0.469	0.344	0.259
Fe^{3+IV}	0.020	0.000	0.009	0.040	0.000	0.000	0.092	0.001	0.033
Fe^{2+}	4.009	4.029	3.999	3.875	4.052	4.330	4.532	4.597	4.585
Mn	0.500	0.510	0.483	0.367	0.370	0.360	0.125	0.148	0.208
Mg	0.240	0.211	0.239	0.201	0.046	0.044	0.024	0.022	0.035
Ca	0.040	0.061	0.066	0.082	0.053	0.096	0.094	0.072	0.016
Na	1.979	1.886	1.932	1.964	1.917	1.854	1.923	1.971	1.999
K	0.008	0.080	0.000	0.013	0.080	0.013		0.004	0.015
Rhönite (mol.%)	2.0	3.1	3.3	4.1	2.7	4.8	4.7	3.6	0.8
Fe^{3+}-Al aenigmatite	10.1	21.1	15.2	14.0	34.2	15.0	3.8	2.1	1.0
Fe^{3+}-Tschermak.	2.0	0.0	0.9	4.0	0.0	0.0	9.2	0.1	3.3
Fe^{3+} aenigmatite	7.0	8.3	5.5	24.8	23.3	14.2	7.8	9.6	9.9
Ideal aenigmatite	78.8	67.5	75.1	53.1	39.8	66.1	74.5	84.6	85.0

Peralkal. : peralkaline; FeO*: total Fe as Fe^{2+}

Location	**Benue**	**Bamboutos**	**Bamenda**	**Rumpi Mt.**			**Mt. Etinde**	**Ngaoundéré**
Lava type	**Peralkal. Trachyte**	**Phonolite**	**Peralkal. phonolite**	**Phonolite**			**Néphélinite**	**Phonolite**
Sample	**MU1**	**B5**	**CA317**	**CN44**			**M146**	**NG16**
SiO_2 (wt.%)	39.69	41.40	40.37	38.17	40.47	40.83	40.06	39.56
TiO_2	6.90	8.09	10.23	8.42	5.79	7.16	10.04	6.37
Al_2O_3	0.50	0.29	0.50	1.79	1.27	1.04	0.64	1.36
FeO*	39.84	39.73	36,75	40.51	40.66	40.52	39.67	40.20
MnO	4.03	2.04	4.26	2.15	1.98	1.94	1.27	3.32
MgO	0.05	0.98	0.34	0.27	0.34	0.38	0.88	0.20
CaO	0.21	0.26	0.14	0.43	0.31	0.31	0.59	0.80
Na_2O	7.32	7.33	7.56	6.71	7.26	7.38	7.25	7.02
K_2O	0.03	0.03		0.00		0.00	0.05	
Sum	98.57	100.14	100.17	98.45	98.08	99.56	100.45	98.83
Si (apfu)	5.768	5.882	5.843	5.639	5.907	5.871	5.713	5.765
Ti	0.754	0.865	1.093	0.936	0.636	0.774	1.077	0.698
Al	0.086	0.048	0.066	0.312	0.218	0.176	0.108	0.234
Fe^{3+VI}	0.775	0.414	0.015	0.698	0.842	0.649	0.196	0.995
Fe^{3+IV}	0.146	0.070	0.091	0.050	0.000	0.000	0.180	0.002
Fe^{2+}	3.920	4.236	4.463	4.256	4.121	4.223	4.355	3.901
Mn	0.496	0.246	0.537	0.269	0.245	0.236	0.153	0.410
Mg	0.011	0.207	0.062	0.059	0.074	0.081	0.187	0.043
Ca	0.033	0.040	0.014	0.068	0.048	0.048	0.090	0.125
Na	2.062	2.019	1.860	1.922	2.054	2.057	2.004	1.983
K	0.006	0.006		0.000		0.000	0.009	
Rhönite (mol.%)	1.6	2.0	0.7	3.4	2.4	2.4	4.5	6.2
Fe^{3+}-Al aenigmatite	5.3	0.8	5.2	24.4	17.0	12.8	1.7	10.9
Fe^{3+}-Tschermak.	14.6	7.0	9.1	5.0	0.0	0.0	18.0	0.2
Fe^{3+} aenigmatite	24.6	13.5	0.0	6.4	36.4	22.6	0.0	30.2
Ideal aenigmatite	53.9	76.7	85.0	60.8	44.1	62.2	75.8	52.5

Table 2: Representative compositions of clinopyroxene and Fe-Ti oxide from Djinga Tadorgal and Sao Tomé phonolites.

Minerals	**Clinopyroxene**		**Fe-Ti oxide**	
Sample	**DT113**	**ST44**	**DT113**	**ST44**
SiO_2 (wt.%)	51.510	51.770	0.010	0.000
TiO_2	2.340	2.420	23.210	22.320
Al_2O_3	0.630	0.690	0.070	0.140
FeO	21.40	24.910	69.520	70.410
MnO	1.810	1.200	5.340	4.900
MgO	3.090	0.100	0.090	0.250
CaO	9.400	7.200	0.000	0.000
Na_2O	8.490	9.410		
ZrO_2		1.320		
Total	98.670	99.020	98.240	98.020
En (mol.%)	0.287	0.013		
Wo	0.456	0.421		
Hem (mol.%)			0.564	0.580
Gk			0.003	0.093

Table 3: Results of QUILF thermobarometer calculations for Djinga Tadorgal and Sao Tomé samples.

Location		Djinga Tadorgal			Sao Tomé		
En (mol.%)		0.287	0.287	0.287	0.013	0.013	0.013
Wo		0.456	0.456	0.456	0.421	0.421	0.421
Hem (mol.%)		0.564	0.564	0.564	0.580	0.580	0.580
Gk		0.003	0.003	0.003	0.093	0.093	0.093
T(°C)	Input	727	727	725	827	727	827
	Calc.	655	725			791	
P(GPa)	Input	0.100	0.100	0.100	0.100	0.100	0.100
	Calc.	0.100	0.100			0.100	
$aSiO_2$	Input	0.100	0.100	0.800	0.200	0.200	0.400
	Calc.			0.178	0.172		0.297
$\log fO_2$	Input	-20.000	-18.200	-15.000	-10.000	-16.000	-15.000
	Calc.			-14.900	-18.195		-14.985
ΔFMQ		-1.941	-1.934	-0.962	-1.981	-1.287	-1.748

Table 4: Distribution of major elements in aenigmatite-bearing phonolites from Djinga Tadorgal and Sao Tomé.

Lava types	**P. Phonolite**	**Phonolite**
Sample	**DT113**	**ST44**
SiO_2 (wt.%)	62.64	61.68
TiO_2	0.60	0.35
Al_2O_3	18.00	18.26
Fe_2O_3*	3.02	3.73
MnO	0.19	0.21
MgO	0.30	0.31
CaO	0.93	1.25
Na_2O	7.30	6.78
K_2O	6.07	5.46
P_2O_5	0.10	0.06
L.O.I	0.60	1.29
Sum	99.75	99.38
Na_2O+K_2O	13.37	12.24
TiO_2/Fe_2O_3*	0.20	0.09
D.I.	93.42	89.11
P.I.	1.03	0.93
mg#	0.26	0.17

DT: Djinga Tadorgal; ST: Sao Tomé; Fe_2O_3*: total Fe as Fe^{3+}; L.O.I.: Loss on ignition; D.I.: Differentiation index; P.I.: Peralkaline index.

DISCUSSION

High TiO_2 contents in the studied aenigmatite reflects a late crystallization. The low Ca (0.040—0.096 apfu) and Al (0.141—0.395 apfu) contents in the chemical structure of the Djinga Tadorgal and Sao Tomé aenigmatite could be the result of the combined effect of the low temperature of crystallization and the high silica activity. The high Fe^{3+} contents characterized by the presence of Fe^{3+}-Tschermak, Fe^{3+}-aenigmatite and Fe^{3+}-Al aenigmatite, indicate fO_2

conditions, lower than those of the FMQ buffer. Low oxygen fugacity (fO_2) favors crystallization of Ti-aegirine. A sudden increasing of fO_2 in felsic rocks would produce instability of aenigmatite and growth of Na contents in clinopyroxene, with crystallization of aegirine and aegirine augite (Njonfang and Nono, 2003). This suggests that the change of fO_2 conditions during the crystallization, may explain the chemical and structural features of the Na-rich pyroxene (aegirine-augite, aegirine) and the aenigmatite crystals in felsic rocks.

On the basis of a generally antipathetic relationship between aenigmatite and FeTi-oxides, Nicholls and Carmichael (1969) proposed that aenigmatite crystallization is stabilized by the reaction of pyroxene and Ti-rich oxides with Na-rich melt. They recognized a “no-oxide field” in T–fO_2 space in which aenigmatite is stable but the oxides are not. The aenigmatite can replace Fe-Ti oxides, which crystallize simultaneously or later with the sodic clinopyroxene. The phonolites from Djinga Tadorgal and Sao Tomé are alkali rich (more or less peralkaline) lavas (Na_2O+K_2O: 12.2—13.4 wt.%; P.I. = [$(Na_2O+K_2O)/Al_2O_3$]: 0.93—1.03; Table 4), but do not contain ilmenite and arfvedsonite crystals. Overall, there is a simple relationship between whole-rock peralkalinity and the degree of (Al+Ca) substitution. Mahood and Stimac (1990) noted that aenigmatite phenocrysts contain more Na and less Al and Ca as whole-rock P.I. increases. The development of peralkaline liquids (Ronga et al., 2010) is considerable when the fO_2 is below FMQ buffer at the temperatures below 900 °C. Abnormally low values of Ti/Fe in felsic rocks are sometimes caused by the crystallization of Ti-bearing aenigmatite, as evidenced by the low values of TiO_2/FeO^* ratio (0.09—0.20; Table 1) for the Djinga Tadorgal and Sao Tomé phonolites.

The Djinga Tadorgal and Sao Tomé aenigmatite could therefore be the product of the reaction between aegirine and Ti-magnetite.

$4(NaFeSi_2O_6)$ + Ti-(Fe_3O_4) <==>
aegirine Ti-magnetite

$Na_2Fe_5TiSi_6O_{20}$ + $(Na_2Si_2O_5, 2FeO)$ + $1/2(O_2)$.
aenigmatite melt gas

However, Na-rich pyroxene (aegirine, aegirine augite) are known to occur at low temperature (< 600 °C) and under low fO_2. According to White et al., (2005) which described the stability of aenigmatite relative to ilmenite (Ti-rich oxides) as a function of T and fO_2, proposed the equation below :

$Na_2Si_2O_5 + 4SiO_2 + (2Fe_2O_3 + FeTiO_3)$ =
melt quartz ilmenite s.s.

$Na_2Fe_5TiSi_6O_{20}$ + O_2
aenigmatite gas

It appears that aenigmatite stability is a function of both fO_2 and $aSiO_2$. Although the stability field of aenigmatite expands at lower fO_2, it also requires relatively high silica activities, particularly at T >750 °C, and is overall more stable at lower temperatures (Macdonald et al., 2011). The crystallization of Ti-bearing aenigmatite is therefore probably the result of the reaction between the Ti-rich oxides and Na_2O-rich magmatic liquid, as attested by the presence of Ti-magnetite inclusions in Djinga Tadorgal and Sao Tomé aenigmatites.

Conclusion

The characteristics of the Ti-bearing aenigmatite from Djinga Tadorgal and Sao Tomé phonolites are more or less similar to those of the felsic lavas from other parts of the Adamawa plateau and the Cameroon Line. Aenigmatie is a late and accessory mineral, exclusively present in more or less peralkaline rocks (Avanzinelli et al., 2004). As showed by QUILF calculations, the Djinga Tadorgal and Sao Tomé phonolites were erupted at very low fO_2 below FMQ buffer, after the occurrence of Ti-bearing aenigmatite. Ti-bearing aenigmatite is the product of the reaction between Fe—Ti oxides and Na_2O rich magmatic liquid. It stability is most likely temperature- and fO_2-dependent, at ~ 0.1 GPa. fO_2 affect clinopyroxene stability and composition. As discussed, Aegirine and other Na-bearing phases such as aenigmatite played an important role in the formation of Djinga Tadorgal and Sao Tomé phonolites. Aenigmatite was consequently stable in the studied phonolites under low fO_2 conditions, low temperature (< 800 °C) and low pressure (~0.1 GPa).

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