

Compositions and Petrogenetic Significance of the Eudialyte Group Minerals from Sushina, Purulia, West Bengal

ANIKET CHAKRABARTY^{1*}, KAMAL L. PRUSETH² and AMIT KUMAR SEN³

¹Department of Geology, Durgapur Government College, Durgapur - 713 214

²Department of Geology and Geophysics, IIT Kharagpur, Kharagpur -721 302

³Department of Earth Sciences, IIT Roorkee, Roorkee - 247 667

*Email: aniket_chakrabarty@rediffmail.com

Abstract: The eudialyte-group of minerals (EGM) is one of the most important index minerals of the peralkaline (agpaitic) nepheline syenites. They crystallize in varied physico-chemical conditions ranging from the early-magmatic (orthomagmatic) to late-magmatic and even in the post-magmatic (hydrothermal) stage. In India, the only agpaitic nepheline syenite gneisses of the Sushina Hill region contain both late-magmatic as well as hydrothermal eudialytes. Compositionally these are Mn-Nb-Ca rich eudialytes and are comparable to the other EGM occurrences such as Ilímaussaq (Greenland), Tamazeght (Morocco), Mont-Saint Hilaire (Canada) and Pilansberg (South Africa). High Mn content (>6.5 wt.%) for both varieties of the Sushina EGM indicates that they are highly evolved in nature. In terms of the calculated site occupancy, particularly the [M(3)] and [M(2)], the Sushina eudialytes mimic some Pilansberg eudialytes. In addition to the eudialyte, the host nepheline syenite gneiss also contains an unknown Na-Zr-silicate (NZS) which is often found to be replacing both types of eudialytes. Compositionally these NZS can be tentatively represented as $\text{Na}_2\text{Zr}_2\text{Si}_6\text{O}_{17}$. These NZS are characterized by much higher Zr, but lower Mn and Nb concentrations compared to the associated eudialytes. Two distinct varieties of eudialyte and NZS indicate subtle changes in the alkalinity during their formations. The formation of the late-magmatic as well as hydrothermal eudialyte essentially took place at somewhat elevated pH conditions. The replacement or alteration of eudialytes by NZS indicates a decreasing pH condition. In terms of the chemical composition the late-magmatic eudialytes can be represented as a solid-solution series between the kentbrooksite-taseqite-aqualite while the hydrothermal eudialyte represents solid-solution between kentbrooksite-taseqite -Ce-zirsilite.

Keywords: Eudialyte-group of minerals (EGM), Na-Zr silicates (NZS), Agpaitic systems, Late-magmatic, Sushina Hill.

BACKGROUND INFORMATION AND OBJECTIVES

Eudialyte is an important index mineral for peralkaline nepheline syenites which are commonly termed as agpaitic nepheline syenites and were first reported from the Ilímaussaq peralkaline complex, South Greenland by Stromeyer (1819). The term ‘agpaitic’ corresponds to the peralkaline nepheline syenites which are characterized by complex Na-Zr-Ti minerals such as eudialyte, mosandrite (rinkite), astrophyllite, aenigmatite etc. (Ussing, 1912; Sørensen, 1997; Le Maitre, 2002; Marks et al. 2011). In contrast, syenitic rocks with relatively simpler mineralogy such as zircon, ilmenite, and titanite are termed as miaskitic nepheline syenites. There are many eudialyte bearing peralkaline nepheline syenitic complexes reported worldwide and the important localities include Ilímaussaq and Motzfeldt (Greenland), Tamazeght (Morocco), Pilansberg (South Africa), Mon-Saint-Hilaire (Canada) and Khibina

and Lovozero (Russia) (Marks et al. 2011, Schilling et al. 2011 and references therein).

In general agpaitic rocks are believed to be crystallize from an highly evolved mantle derived melt (Sørensen, 1992, 1997; Kramm and Kogarko, 1994; Marks et al. 2004) and are exceptionally rich in large ion lithophile elements (LILE) such as Na, K, and Li, Rare earth elements (REEs) and high field strength elements (HFSE) such as Nb, Ta, Ti, Zr, U and Th (Kogarko, 1980; Sørensen, 1992; Olivo and Williams-Jones, 1999). In the Ilímaussaq complex, the type area of the agpaitic rock, it has been found that the solidus of the agpaitic melt can be as low as 500° - 450°C (Markl and Baumgartner, 2002). In addition to this many agpaitic plutonic complexes around the world shows post-magmatic deuteric alteration (autometasomatic) subsequent to the ortho to late-magmatic event (e.g. Pilansberg Complex). During such alteration the early to late-magmatic minerals reacts

with the deuteritic fluids and form new mineralogical assemblages. Eudialytes usually form under conditions starting from the early magmatic (e.g. in Ilímaussaq, Khibina, Lovozero: Gerasimovsky et al. 1974; Pfaff et al. 2008) and continue up to late to post-magmatic activity (e.g. in Pilansberg Complex, North Qôroq, Greenland: Mitchell and Liferovich, 2006; Coulson and Chambers, 1996, Coulson, 1997). Variable conditions of formation coupled with great compositional variability make eudialytes an useful indicator of orthomagmatic to hydrothermal processes involved in the agpaite systems. EGM are also important as they are not compositionally destabilized by metamorphic fluids up to the amphibolite facies conditions and thus help in delineating the magmatic history even in the metamorphosed peralkaline rocks (Schilling et al. 2011).

In India only one such eudialyte bearing peralkaline complex is reported from the Sushina Hill Region, Purulia, West Bengal (Sushina eudialyte here after) (Chakrabarty, 2009; Chakrabarty et al. 2011). Earlier work on the Sushina eudialyte shows that these are essentially hydrothermal eudialyte rich in Mn, Nb, Sr and Ce. These eudialytes are hosted by the Mesoproterozoic nepheline syenite gneiss of the Chandil Formation of North Singhbhum Mobile Belt (NSMB) (Chakrabarty, 2009; Chakrabarty et al. 2011). The Chandil Formation is the upper most part of the NSMB and the various alkaline rocks such as carbonatite, alkali-pyroxenite and nepheline syenite gneisses are present along a 100 km long shear zone commonly termed as Northern Shear Zone. In addition to the agpaite nepheline syenite gneiss, a small body of miaskitic nepheline syenite (gneiss) is also exposed at the Sushina Hill region. Mineralogically this miaskitic rock is dominantly made up of albite, orthoclase, nepheline, Mn-rich biotite, magnetite and zircon (Chakrabarty, 2009). The eudialytes of the Sushina Hill area is yet to be properly characterized particularly in terms of their chemical composition. Moreover the structural formulae presented in the earlier contribution are somewhat arbitrary and need further revision. This contribution is in continuation with our previous work on the Sushina eudialyte. The main objectives of this study include presenting some new mineralogical data of the Sushina eudialyte, revising the structural formula of the reported eudialyte and demonstrating uniqueness with respect to other eudialyte occurrences.

CHEMICAL CHARACTERISTICS OF THE EGM

EGM are complex Na-Ca-Zr silicates with variable proportions of K, Sr, Fe, Mn, REE, Y, Nb, and Ti. The IMA (International Mineralogical Association) approved general

formula of EGM is $N_{15}[M(1)]_6[M(2)]_3Zr_3[M(3)][M(4)](Si_{24}O_{66-73})(OH)_{0-9}X_2$ (Johnsen et al. 2003; Pfaff et al. 2010) where:

N	= Na, K, Sr, Ca, REE, □ (vacancy), Ba, Mn ²⁺ , H ₃ O ⁺
[M(1)]	= Ca, REE, Mn ²⁺ , Fe ²⁺ , Na, Sr
[M(2)]	= Fe ²⁺ , Mn ²⁺ , □ (vacancy), H ₃ O ⁺ , Zr ⁴⁺ , Ta ⁵⁺ , Ti ⁴⁺ , K, Ba
[M(3, 4)]	= Si, Al, Nb ⁵⁺ , Ti ⁴⁺ , W ⁶⁺ , Na
Z	= Zr ⁴⁺ , Hf, Ti ⁴⁺ , Nb ⁵⁺
OH	= H ₂ O, OH ⁻ , O ²⁻ , CO ₃ ²⁻ , SO ₄ ²⁻ , SiO ₄ ⁴⁻
X	= Cl ⁻ , F ⁻ , OH ⁻

There are ten IMA approved end members (Table 1) present within the EGM and great diversity in chemical composition exists between these end members. The structural formula calculation based on microprobe data for EGM is done on the basis of $\Sigma(Si+Zr+Ti+Nb+Al+Hf) = 29$ *afu* (atom per formula unit) (Johnsen and Grice, 1999). This standard procedure has been used for the generated as well as for all other comparative data of the eudialyte used in this work. In addition to the site occupancies, the structural formulae are also calculated according to the standard procedure described by Johnsen and Grice (199) and all the data set presented here are calculated accordingly.

ANALYTICAL TECHNIQUES

Eudialyte compositions were analyzed on polished thin sections using a CAMECA SX-100 electron microprobe operated in wave length dispersive mode at IIC, IIT Roorkee, using 20 nA beam current and an 15 kV of acceleration voltage with beam diameter <1 µm. Well characterized minerals as well as synthetic standards (Na & Si: Albite; K: Sanidine; Zr: ZrO₂; Nb, Ce and Sr: Glass standard supplied by CAMECA) were used for the standardization and calibrations of the instrument and the generated data were processed accordingly. Special attention was given to avoid the Na- loss during the analysis. This was done by first analyzing the Na (Na *Kα*: albite) with much reduced counting time (less than 60 s). The precision of replicate measurements of the standards was better than ±1% (relative). The elemental mapping of the selected eudialyte grains were done using SEM-EDAX (FEI: Quanta 200F) on the polished thin sections and the working condition was kept at 20Kv with spot size of 3 µm. It must be mentioned here that out of the different volatile matters present in the eudialyte structure we have analyzed only Cl and F ('F' content is below the detection limit by EPMA in our case). As other volatiles such as H₂O, OH⁻ can not be determined

Table 1. IMA approved end-members of the EGM

	N	M(1)	M(2)	Z	M(3)	M(4)	Si ₂₄ O ₆₆₋₇₃	(OH) ₀₋₉ X ₂
Eudialyte _{s.s}	Na ₁₅	Ca ₆	Fe ₃	Zr ₃	Si	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl, OH) ₂
Kentbrooksite	(Na, REE) ₁₅	Ca ₆	Mn ₃	Zr ₃	Nb	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (F, Cl) ₂
Alluaivite	Na ₁₆	[Ca(Mn)] ₆	Na ₃	Ti ₃	Si	Si	Si ₂₄ O ₇₄	Cl. 2H ₂ O
Aqualite	Na ₅ (H ₃ O ⁺) ₈ □ ₂	Ca ₆	□ ₃	Zr ₃	Si	Si	Si ₂₄ O ₆₆	(OH) ₉ Cl
Raslakite	Na ₁₅	Ca ₃ Fe ₃	[Na(Zr)] ₃	Zr ₃	(Si, Nb)	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl, OH) ₂
Oneillite	Na ₁₅	Ca ₃ Mn ₃	Fe ₃	Zr ₃	Nb	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (OH) ₂
Ce-Zirsilite	Na ₁₂ (Ce, Ca) ₃	Ca ₆	Mn ₃	Zr ₃	Nb	Si	Si ₂₄ O ₇₃	OH ₃ CO ₃ ·H ₂ O
(Mn, Ca) – ordered eudialyte	Na ₁₅	Ca ₃ Mn ₃	Na ₃	Zr ₃	Si	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (OH) ₂
(Mn, Na) – ordered eudialyte	Na ₁₅	Na ₃ Mn ₃	Fe ₃	Zr ₃	Si	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (OH) ₂
Taseqite	Na ₁₂ Sr ₃	Ca ₆	Fe ₃	Zr ₃	Nb	Si	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ Cl ₂

Data compiled from Johnsen and Grice (2003) and Pfaff et al. (2010)

by EPMA the structural formulae are calculated assuming that the (OH) and 'X' sites are filled up by these volatiles.

TEXTURAL FEATURES OF THE SUSHINA EGM

The textures involving the eudialytes of the Sushina Hill region are very complex (Fig. 1). The host nepheline syenite gneiss is essentially composed of albite, orthoclase, aegirine and eudialyte. The secondary mineral assemblage is defined by the gonnardite, natrolite and some unknown Na-Zr silicates (Chakrabarty et al. 2011; Chakrabarty et al. in prep). These constituting minerals define a well developed but discontinuous gneissosity in the rock. The pink coloured eudialyte grains are set within the albite matrix (Fig. 1a). We report here two distinct textural mode of occurrence involving the Sushina eudialyte (Fig. 1a, b). The textural features indicate that majority of the eudialytes are essentially hydrothermal in origin (Fig. 1b) which is in agreement with earlier work (Chakrabarty et al. 2011) that concluded the Sushina eudialytes to be formed at the post-magmatic hydrothermal stage with extensive subsolidus low temperature alterations of the rock. However, careful petrological investigation reveals that some tiny but highly altered grains of eudialyte are present along with the hydrothermal eudialyte (Fig. 1a). These eudialytes are highly altered as evident from their low Na count indicating significant loss of Na due to alterations. Texturally such eudialyte grains are distinctively different from the hydrothermal ones, being smaller in size with prominent grain boundaries. These are likely the late-magmatic eudialytes. Most of the eudialyte grains (both late-magmatic and hydrothermal) are clustered with albite, aegirine ($\pm$ amphibole) and some unknown sodium- zirconium silicates

(NZS). These complex aggregates make it difficult to distinguish between late-magmatic and hydrothermal eudialytes using the normal petrological microscope. Such distinction can be brought out prominently in the BSE images (Fig. 1c-g, j). The late-magmatic eudialytes are relatively less pleochroic compared to the hydrothermal one. The effect of alteration in these eudialytes result in discoloration along the grain boundaries, a similar feature has been observed in some North Qôroq (Greenland) eudialytes (Coulson, 1997). The late-magmatic eudialytes grains are susceptible to alterations and frequently altered to serandite, NZS, titanite and pyrochlore (Chakrabarty et al. in prep) (Fig. 1c, d). Due to intense alterations, the individual grain boundaries are fused and appear to be a single grain in the BSE images (Fig. 1c). Late-magmatic eudialytes are, in general, characterized by their smaller size and fewer inclusions of other constituting minerals such as aegirine, albite etc. compared to hydrothermal ones (Fig. 1f). The most common secondary mineral present in association with the eudialyte grains is an unknown Na-Zr silicate (Fig. 1g). Elemental mapping of the late-magmatic eudialytes show that there are Zr and Nb rich zones present within the eudialyte (Fig. 1h-i). The brighter Zr rich zones are exclusive and represent the alteration or replacement of the eudialyte by Na-Zr silicates (NZS). Similar NZS rich zone is also reported in association with the hydrothermal eudialyte (Fig. 1j) (Chakrabarty et al. 2011). Such textural features indicate that replacement of the eudialytes by NZS took place at very late stage, after the formation of hydrothermal eudialyte. The Nb rich zones are probably representative of alteration of the late-magmatic eudialytes to pyrochlore or whôlerite like phases.

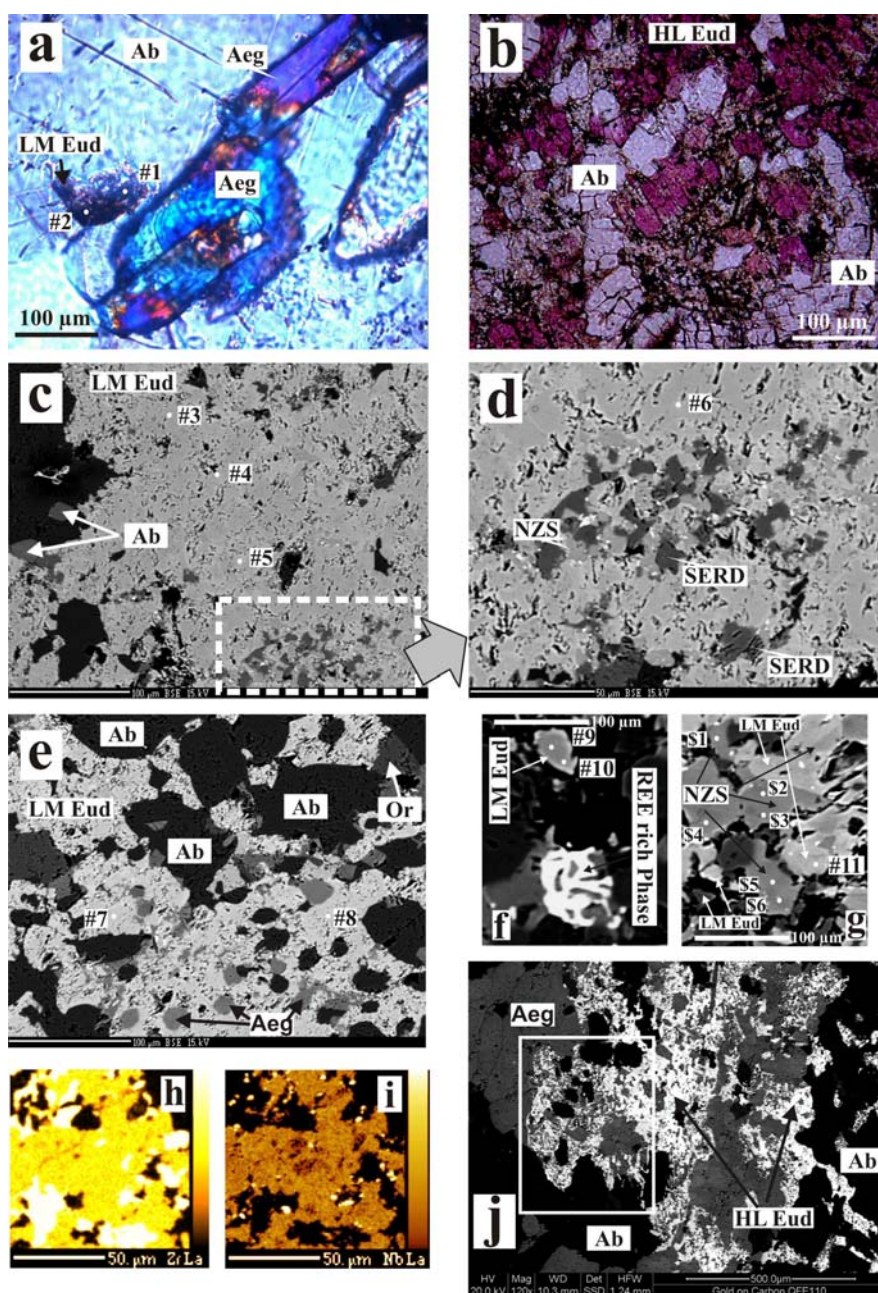


Fig.1. Photomicrograph and BSE images of the Sushina EGM showing different textures: **(a)** altered late-magmatic eudialyte set within the albite matrix and associated with aegirine; **(b)** hydrothermal eudialyte engulfing the pre-existing albite and forming complex zone composed of aegirine, albite and NZS; **(c-d)** late-magmatic eudialyte showing evidences of alterations to serandite; **(e)** highly altered late-magmatic eudialyte containing numerous inclusions of aegirine and albite. Intense alteration makes it difficult to distinguish the individual grain boundaries at places; **(f)** unaltered single grain of late-magmatic eudialyte set within the albite matrix. An unidentified REE rich phase is also present. Such phases are present as secondary minerals and probably formed with or after the formation of hydrothermal eudialyte; **(g)** late-magmatic eudialyte grains are being replaced by an unidentified Na-Zr silicate phase; **(h-i)** Zr and Nb elemental mapping of the late-magmatic eudialyte. The brighter zones in both the images indicate that the alteration of late-magmatic eudialyte to Zr and Nb rich phases. The alteration is more pronounced for Zr compared to the Nb. The Zr rich phases are representative of NZS while the Nb rich phases indicate alteration to pyrochlore and/or whölerite like phases; **(j)** BSE image of the hydrothermal eudialyte showing the evidences of corroding the albite and aegirine (white box). Abbreviations used: Ab: albite; Aeg: aegirine; Or: orthoclase; HL Eud: hydrothermal eudialyte; LM Eud: late-magmatic eudialyte and NZS: Sodium (Na)-Zirconium (Zr)-silicate. White dots with '#' and '\$' symbols indicate the point of analyses for the late-magmatic eudialyte and NZS respectively. The corresponding analyses are presented in Table 2 (eudialyte) and Table 4 (NZS).

CHEMICAL COMPOSITION OF THE EGM

In terms of mineral composition (Table 2) both the late-magmatic as well as hydrothermal eudialytes are rich in Mn (5.71-7.27 wt.% and 7.17-7.65 wt% respectively) and Nb (1.81-4.20 wt.% and 3.98-4.69 wt% respectively) (Fig. 2). However, both the variants are almost free of Fe and Al. The high Mn-content of these eudialytes suggests that these are late-post magmatic in origin (Pfaff et al. 2008, Schilling et al. 2011). In terms of Fe-Mn systematics the Sushina eudialytes are found to be highly evolved in nature and represent Mn-rich analog of Nb-poor eudialyte i.e. relatively manganoan eudialytes. There is a significant difference observed between the hydrothermal and late-magmatic eudialyte, the latter ones contains more volatile matters compared to the hydrothermal ones. Moreover, the reported hydrothermal eudialytes are essentially rich in Ce and Sr (Chakrabarty et al. 2011) while the late-magmatic eudialytes

are totally free of these elements. The studied eudialytes are compared with the some other eudialyte bearing peralkaline feldspathoid syenites reported from various localities (Fig. 2 and references there in). Sushina eudialytes are characterized by relatively higher concentrations of Mn and Ca compared to the all other EGM reported globally and only comparable with some late-post magmatic eudialyte of the Pilansberg, Tamazeght and Mont-Saint Hilaire peralkaline complexes (Fig.2). The Mn content of the Sushina EGM is relatively high and only comparable with the Pilansberg eudialytes while the Nb content shows wide variations and are comparable with some Tamazeght, Ilímaussaq and Pilanesberg EGM (Fig. 2). The REE (in our case only Ce) and Sr concentration of the hydrothermal eudialytes also match well with the eudialytes of the Pilansberg complex while in other localities the Sr concentration are found to be much higher. The Na, Ti, Zr, Si and Cl content do not show any significant variations

Table 2. Representative electron microprobe data of EGM from Sushina Hill Region (The # symbol indicates corresponding points presented in Fig. 1)

Oxide wt. %	Late-magmatic eudialyte This work											Hydrothermal eudialyte Chakrabarty et al. (2011)		
	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	P1	P2	P3
SiO ₂	51.57	49.56	45.43	46.87	50.05	46.04	48.40	47.70	47.90	48.33	45.37	48.89	48.91	48.39
Na ₂ O	8.26	8.67	8.32	8.56	8.75	8.41	8.16	7.81	8.89	9.44	7.50	8.39	8.62	8.80
CaO	13.73	13.86	12.73	11.63	13.85	13.33	13.35	13.21	13.46	13.15	15.54	13.36	13.18	13.41
Al ₂ O ₃	0.10	0.02	0.04	0.34	0.03	0.03	0.02	0.01	0.02	0.05	0.02	0.06	0.02	0.00
K ₂ O	0.26	0.37	0.53	0.44	0.29	0.52	0.54	0.53	0.39	0.48	0.51	0.56	0.51	0.53
FeO	0.00	0.00	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.90	0.90	0.76	0.85	0.94	0.66	0.75	0.67	0.86	0.73	0.53	0.69	0.60	0.68
MnO	5.71	6.14	7.01	6.37	5.74	7.27	7.14	7.08	6.26	6.70	6.89	7.39	7.65	7.17
Nb ₂ O ₅	1.81	3.33	3.90	3.23	2.64	3.87	4.20	4.08	3.30	3.37	3.87	4.02	3.98	4.69
ZrO ₂	12.80	12.12	11.27	11.68	12.50	11.56	12.05	12.04	11.86	12.20	10.93	11.17	11.16	11.09
Ce ₂ O ₃	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.04	2.00	2.53
TiO ₂	0.44	0.05	0.05	0.07	0.10	0.01	0.02	0.00	0.08	0.03	0.02	0.06	0.08	0.07
SrO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.52	1.39	1.37
Total	95.58	95.02	90.09	90.04	94.89	91.70	94.63	93.13	93.02	94.48	91.18	98.15	98.10	98.73
O=Cl	0.20	0.20	0.17	0.19	0.21	0.15	0.17	0.15	0.19	0.16	0.12	0.16	0.14	0.15
Corrected Total	95.38	94.82	89.92	89.85	94.68	91.55	94.46	92.98	92.83	94.32	91.06	97.99	97.96	98.58
$\Sigma(\text{Si}+\text{Zr}+\text{Ti}+\text{Nb}+\text{Al}+\text{Hf}) = 29 \text{ afu}$ (Johnsen and Grice, 1999)														
Si	25.31	25.20	24.96	24.95	25.26	24.97	24.97	24.96	25.14	25.08	25.07	25.20	25.22	25.07
Na	7.86	8.55	8.86	8.83	8.56	8.84	8.16	7.92	9.05	9.50	8.03	8.38	8.62	8.84
Ca	7.22	7.55	7.50	6.63	7.49	7.75	7.38	7.41	7.57	7.31	9.20	7.38	7.28	7.44
Al	0.06	0.01	0.03	0.21	0.02	0.02	0.01	0.01	0.01	0.03	0.01	0.04	0.01	0.00
K	0.16	0.24	0.37	0.30	0.19	0.36	0.36	0.35	0.26	0.32	0.36	0.37	0.34	0.35
Fe	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.75	0.78	0.71	0.77	0.80	0.61	0.66	0.59	0.76	0.64	0.50	0.60	0.52	0.60
Mn	2.37	2.64	3.26	2.87	2.45	3.34	3.12	3.14	2.78	2.94	3.22	3.23	3.34	3.15
Nb	0.40	0.77	0.97	0.78	0.60	0.95	0.98	0.97	0.78	0.79	0.97	0.94	0.93	1.10
Zr	3.06	3.00	3.02	3.03	3.08	3.06	3.03	3.07	3.03	3.09	2.94	2.81	2.81	2.80
Ce	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.38	0.38	0.48
Ti	0.16	0.02	0.02	0.03	0.04	0.00	0.01	0.00	0.03	0.01	0.01	0.02	0.03	0.03
Sr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.45	0.41	0.41
Sum Cations	46.62	47.98	49.02	47.64	47.70	49.29	48.02	47.82	48.66	49.07	49.82	49.80	49.89	50.27
O	72	73	74	72	73	74	73	73	73	74	75	74	75	75

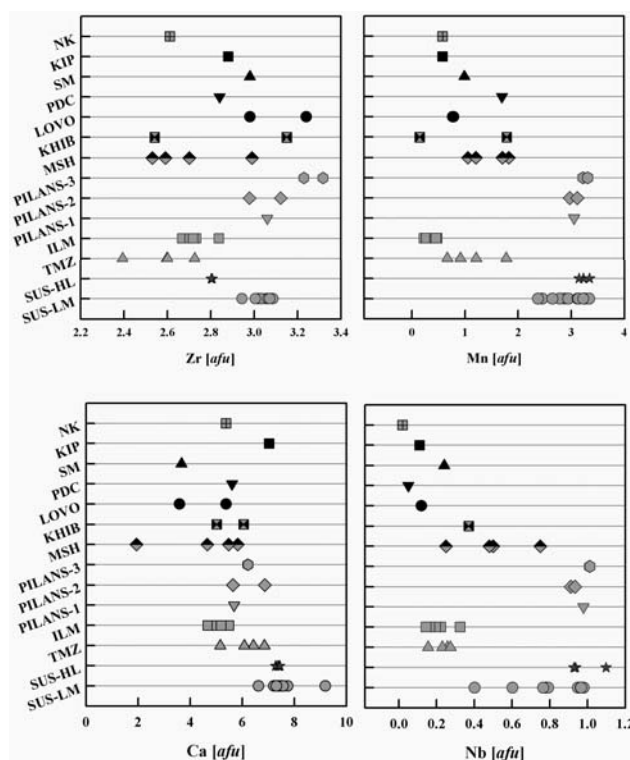


Fig.2. Variation of Zr, Mn, Ca and Nb (in atom per formula unit: *afu*) in EGM of the Sushina region with respect to some other EGM occurrences reported world-wide (see text for the explanations). Locations and data source: (1) SUS-LM: Sushina late-magmatic eudialyte (this work); (2) SUS-HL: Sushina hydrothermal eudialyte (Chakrabarty et al. 2011); (3) TMZ: Tamazeght, Morocco (Schilling et al. 2009); (4) ILM: Ilímaussaq, Greenland (Pfaff et al. 2008); (5) PILNS: Pilansberg, South Africa (Mitchell and Liferovich, 2006) (1: ortho-magmatic eudialyte, 2: late-magmatic eudialyte and 3: hydrothermal eudialyte); (6) MSH: Mont-Saint Hilaire, Canada; (7) KHIB: Khibina, Russia; (8) LOVO: Lovozero, Russia; (9) PDC: Pocos de Caldas, SE Brazil; (10) SM: Saima, North China; (11) KIP: Kipwa, SE Canada; (12) Nora Karr, Sweden (6-12: Schilling et al. 2011 and references therein).

between the two types of the Sushina eudialytes. A recent review on the compositional variability of the EGM by Schilling et al. (2011) presented a wide range of compositions from all the EGM bearing complexes of the world. In comparison to all other EGM bearing complexes, the Sushina eudialytes are characterized by very high concentrations of Ca (Fig. 3) while all other elements such as Nb, Mn, Na, Ti, Zr and Cl are falling well within the range of the observed EGM in other localities. The notable feature of the Sushina eudialyte is that the lower limit for Nb, Mn and Ca are very high compared to the all other EGM bearing complexes (Fig. 3) and comparable with the EGM of Pilanesberg, Ilímaussaq and Tamazeght complexes. The

Nb concentration is significantly higher in the hydrothermal eudialytes which are probably the most Nb-rich eudialytes reported so far (Table 2 comp: P3). In contrast, the late-magmatic eudialytes are most Ca rich similar to those reported from the Pilanesberg complex (by Olivo and Williams-Jones, 1999; Mitchell and Liferovich, 2006). Majority of the Sushina eudialytes are characterized by low Na content compared to the other known eudialyte bearing complexes (Fig. 3). Na loss is the first indication of the eudialyte alteration as evidenced at Ilímaussaq and North Qôroq (Pfaff et al. 2008; Coulson, 1997) and the low Na content in our analyzed eudialytes can be attributed to the alteration of the Sushina eudialytes at post-magmatic stage. This is consistent with our observation that the least altered late-magmatic eudialytes (Fig. 1f) that show higher concentrations of Na (Table 2, comp: 9, 10) compared to the highly altered ones (Table 2, comp: 1, 2). Moreover, Na and H_3O^+ compete for the same structural position (see general formula of EGM) and in case of aqualite end-member, the Na count will become invariably low. We attribute the lower Na content in our analyses due to alterations as well as presence of significant aqualite component.

In terms of the calculated site occupancy (Table 3) the Sushina eudialytes are characterized by the very high Mn_{M2} (> 2.37 afu.) and where the Mn exceeds 3 afu the $[M(2)]$ site is fully occupied by the Mn (Table 3 comp: 6-P3) (Fig. 4). This is also observed for Nb_{M3} which is exceptionally high for Sushina eudialytes (Fig. 4). In terms of Mn_{M2} and Nb_{M3} , such niobo-manganoan eudialyte is rare and only reported from the Pilansberg complex of South Africa (Fig. 4) while in all other localities the Mn_{M2} - Nb_{M3} is very limited and indicates a narrow compositional range. Although, some of the North Qôroq eudialytes are comparable Nb_{M3} to the Sushina and Pilansberg eudialytes, they are exceptionally poor in Mn_{M2} (Fig. 4). The late-magmatic eudialytes show a wide range of compositional variability in terms of the Mn_{M2} and Nb_{M3} (Fig.4). The same can not be said for hydrothermal eudialytes owing to limited data. The calculated site occupancy of the late-magmatic Sushina eudialyte makes them analogous to the solid-solution series between kentbrooksite-taseqite-aqualite. In contrast, the hydrothermal eudialytes are characterized by the Sr and Ce in the N site and this variety of eudialyte can be representative of the solid-solution series between kentbrooksite-taseqite-Ce-zirsilite (Mitchell and Chakrabarty, 2011).

CHEMICAL COMPOSITION OF THE NZS

EGM are often found to be associated with other

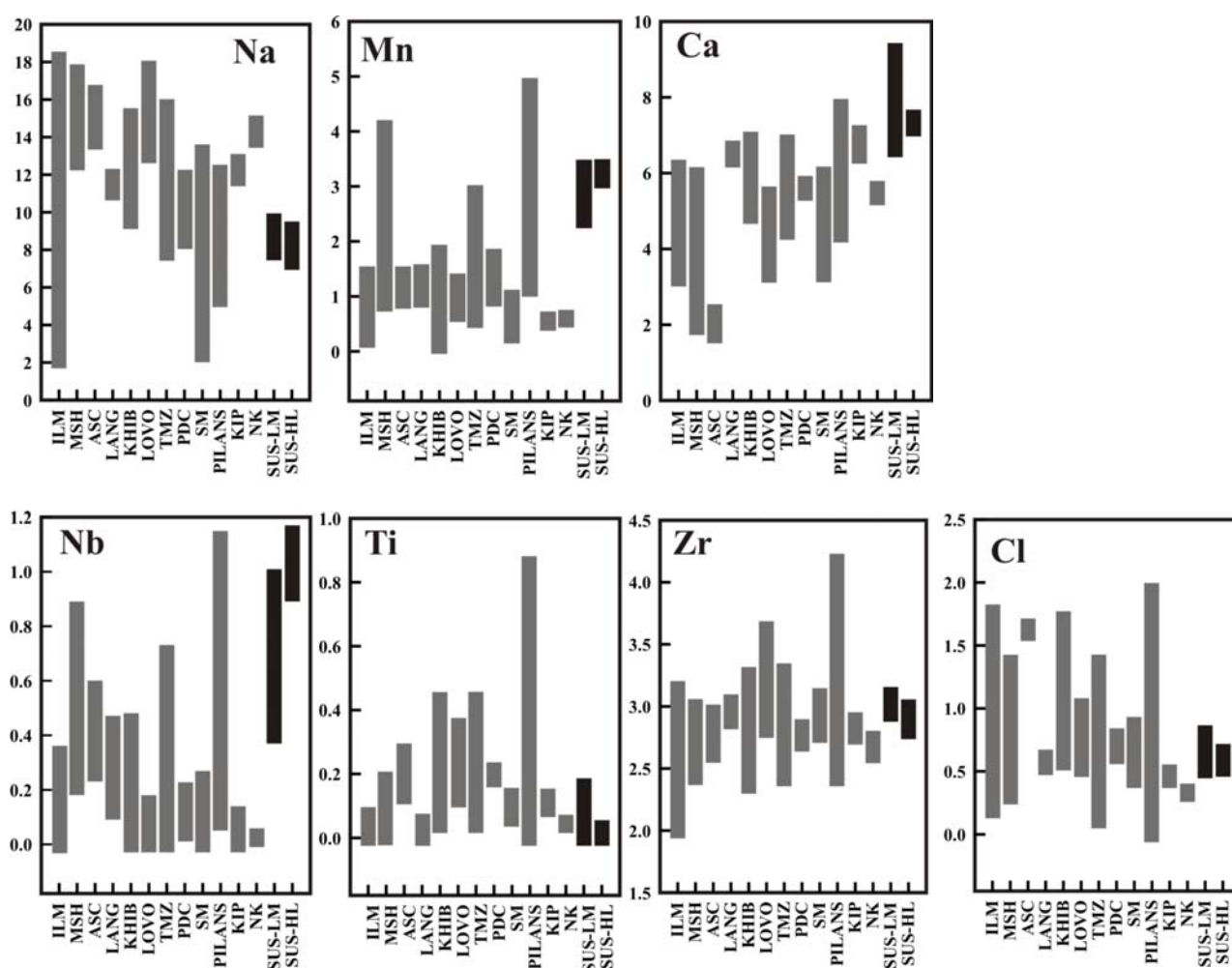


Fig.3. Comparative plots showing the entire range of variation for Na, Mn, Ca, Nb, Ti, Zr and Cl in EGM of the Sushina region and all other EGM occurrences. Note that the lower limit of the Mn-Nb-Ca of the Sushina EGM is very high while the Na is much lower compared to all the EGM. (Abbreviations are same as in Fig. 2)

NZS like catapleiite ($\text{Na}_2\text{ZrSi}_3\text{O}_9 \cdot 2\text{H}_2\text{O}$), elpidite ($\text{Na}_2\text{ZrSi}_6\text{O}_{15} \cdot 3\text{H}_2\text{O}$), parakeldyshite ($\text{Na}_2\text{ZrSi}_2\text{O}_7$), vlasovite ($\text{Na}_2\text{ZrSi}_4\text{O}_{11}$) etc. (Birkett et al. 1992; Marks et al. 2011). However, elpidite is the more common magmatic phase in peralkaline granite e.g. in Ilímaussaq, Strange Lake etc. and catapleiite is more common in the agpaitic syenites e.g. in Pilansberg, Mont-Saint Hilaire, Khibina and Lovozero (Marks et al. 2011 and references therein). It is now well established that both the magmatic eudialytes and catapleiites are not mutually exclusive to each other i.e. where catapleiite is associated with eudialyte, the former is generally present as a secondary phase replacing eudialyte (Marks et al. 2011). A similar observation is made in this study with the late-magmatic eudialytes frequently found replaced by unknown NZS (Table 4, Fig. 1g). These NZS are compositionally different from the above mentioned Na-Zr silicates commonly found in association with the eudialytes.

Compositionally they can be tentatively represented as $\text{Na}_2\text{Zr}_2\text{S}_6\text{O}_{17}$. Replacement of the eudialytes by NZS indicates that the NZS are of later origin. These NZS are extremely Zr rich (31.9-36.7 wt.% ZrO_2) and poor in Nb, Mn and Ca compared to the associated eudialytes (Fig. 5). A similar unknown NZS is also reported from the Pilansberg complex (Mitchell and Liferovich, 2006) but these NZS are compositionally different from the other NZS reported so far from the peralkaline complexes being enriched in Si (> 50% wt.% SiO_2) and poorer in Na (5.04-7.93 wt.% Na_2O).

DISCUSSION

Eudialyte composition can be used successfully for monitoring the pH (alkalinity) changes of the melt during their entire course of evolution starting from the ortho-magmatic to post-magmatic stages as seen in many localities

Table 3. Calculated site occupancies of the studied eudialyte. #1, 3, 6, 8: Late-magmatic eudialyte (this work) and P1-P3 are the hydrothermal eudialyte of Chakrabarty et al. (2011). The site occupancies are calculated according to the procedure given by Johnsen and Grice (1999).

	N	[M(1)] ₆	[M(2)] ₃	Zr ₃	[M(3)]	[M(4)]	Si ₂₄ O ₃₉ *	OH _{0.3} X ₂
#1	Na _{7.86} K _{0.16} Ca _{1.22} □ _{5.76}	Ca ₆	Mn _{2.37} □ _{0.63}	Zr	Si _{0.37} Nb _{0.40} Ti _{0.16} Zr _{0.06}	(Si _{0.94} Al _{0.06})	Si ₂₄ O ₇₂	(O, OH, H ₂ O) ₃ (Cl _{0.75} OH _{1.25})
#3	Na _{8.86} K _{0.37} Ca _{1.78} □ _{3.99}	Ca _{5.72} Mn _{0.28}	Mn _{2.98} Fe _{0.02}	Zr	Nb _{0.97} Ti _{0.02} Zr _{0.01}	(Si _{0.96} Al _{0.03} Zr _{0.01})	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.59} OH _{1.41})
#6	Na _{8.84} K _{0.36} Ca _{2.09} □ _{3.71}	Ca _{5.66} Mn _{0.34}	Mn ₃	Zr	Nb _{0.95} Zr _{0.05}	(Si _{0.97} Al _{0.02} Zr _{0.01})	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.61} OH _{1.39})
#8	Na _{7.92} K _{0.35} Ca _{1.55} □ _{5.18}	Ca _{5.86} Mn _{0.14}	Mn ₃	Zr	Nb _{0.97} Zr _{0.03}	(Si _{0.96} Al _{0.01} Zr _{0.03})	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.59} OH _{1.41})
P1	Na _{8.38} K _{0.37} Ca _{1.61} Sr _{0.45} Ce _{0.38} □ _{3.81}	Ca _{5.77} Mn _{0.23}	Mn ₃	Zr _{2.81} Ti _{0.02} Nb _{0.18}	Si _{0.24} Nb _{0.76}	(Si _{0.96} Al _{0.04})	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.60} OH _{1.40})
P2	Na _{8.62} K _{0.34} Ca _{1.62} Sr _{0.41} Ce _{0.38} □ _{3.63}	Ca _{5.66} Mn _{0.34}	Mn ₃	Zr _{2.81} Ti _{0.03} Nb _{0.16}	Si _{0.23} Nb _{0.77}	(Si _{0.99} Al _{0.01})	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.52} OH _{1.48})
P3	Na _{8.84} K _{0.35} Ca _{1.59} Sr _{0.41} Ce _{0.48} □ _{3.33}	Ca _{5.85} Mn _{0.15}	Mn ₃	Zr _{2.80} Ti _{0.03} Nb _{0.17}	Si _{0.07} Nb _{0.93}	(Si) ₁	Si ₂₄ O ₇₃	(O, OH, H ₂ O) ₃ (Cl _{0.60} OH _{1.40})

*yy = 66-73

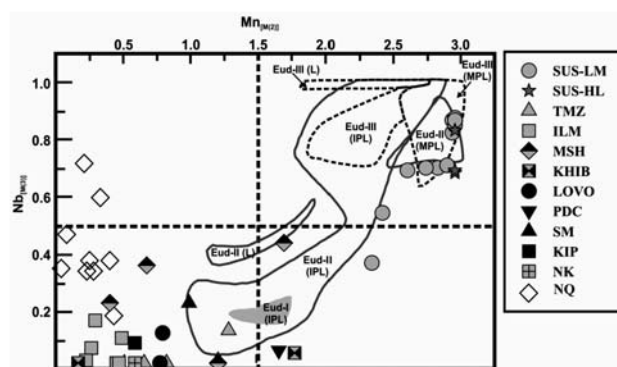


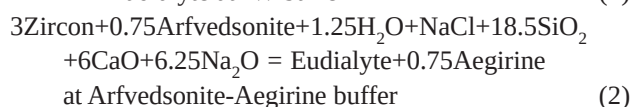
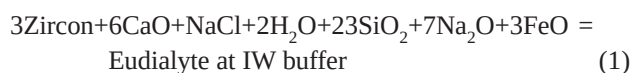
Fig.4. Comparison of compositional variations of the Sushina EGM with that of the other EGM occurrences in terms of Nb in the M(3) versus Mn in the M(2) sites, calculated following the procedure of Johnsen and Grice (1999). It is quite clear that the both types of Sushina eudialytes are more enriched in Mn and Nb compared to all other EGM. Compositionally the majority of the Sushina EGM is showing remarkable similarities with the Pilansberg late-magmatic and hydrothermal eudialyte of the melanolujavrite. Note that the field for late-magmatic and hydrothermal eudialyte of the Pilansberg complex are overlapping. Abbreviations: IPL: Inequigranular and porphyritic lujavrite; L: Lujavrite; MPL: Melanolujavrite; NQ: North Qôroq, Greenland (Coulson, 1997) (Rest of the abbreviations are same as in Fig. 1).

like Ilímaussaq (Markl and Baumgartner, 2002), Pilansberg complex (Mitchell and Liferovich, 2006) etc. Even in some metamorphosed agpaite complexes such as Norra Kärr (Sweden), Kipwa (SE Canada) eudialyte compositions are not affected by post-magmatic metamorphic events (up to amphibolite facies) and can be used to trace the magmatic evolutionary history of the unmetamorphosed parent agpaite rock (Schilling et al. 2011). In general the most essential prerequisite for the formation of agpaite nepheline syenite is the extensive differentiation of a mafic melt at low fO_2 (Marks et al. 2010) though the relative stability of the Ti-Zr rich phases does not directly depend on the fO_2 instead μNa_2O , μCaO and μK_2O are the governing factors (Marks et al. 2011). In such agpaite system the concentration of Na, Cl, F and other volatile species are generally high which raises the solubility of certain HFSE like Ti, Zr, Nb and as a result, phases like eudialyte, aenigmatite, astrophyllite etc. crystallize in such system (Marks et al. 2011). In the present work two different types of eudialytes are present with substantial difference in their chemistry particularly in terms of Zr-REE-Sr compositions. The hydrothermal eudialytes are enriched in REE and Sr, while the late-magmatic eudialytes are devoid of these elements. However, Nb-Mn enrichment is a common feature of the both types of

Table 4. Representative microprobe analysis of Na-Zr silicates (The \$ symbol indicates representative point of analysis given in the Fig. 1)

Oxide wt. %	\$1	\$2	\$3	\$4	\$5	\$6	\$7
Na ₂ O	5.25	5.04	7.93	7.34	7.65	7.29	6.70
Al ₂ O ₃	0.05	0.05	0.02	0.03	0.15	0.18	0.22
SiO ₂	51.36	52.15	51.29	50.85	52.28	51.83	51.80
K ₂ O	0.15	0.07	0.05	0.06	0.06	0.04	0.07
CaO	0.73	0.81	0.73	1.09	0.29	3.76	1.69
TiO ₂	0.05	0.04	0.01	0.12	0.00	0.01	0.04
MnO	0.15	0.17	0.21	0.16	0.09	1.58	0.28
FeO	0.18	0.17	0.07	0.15	0.32	0.00	0.00
ZrO ₂	35.68	35.95	36.15	35.93	36.66	31.91	34.26
Nb ₂ O ₅	1.60	1.35	0.27	2.09	0.27	0.38	0.44
Total	95.20	95.80	96.73	97.82	97.77	96.98	95.50
<i>Formula based on 17O</i>							
Na	1.19	1.13	1.78	1.64	1.69	1.62	1.51
Al	0.02	0.02	0.01	0.01	0.05	0.05	0.07
Si	5.99	6.03	5.93	5.84	5.97	5.95	6.01
K	0.02	0.01	0.01	0.01	0.01	0.01	0.01
Ca	0.09	0.10	0.09	0.13	0.04	0.46	0.21
Ti	0.00	0.00	0.00	0.01	0.00	0.00	0.00
Mn	0.01	0.02	0.02	0.02	0.01	0.15	0.03
Fe	0.02	0.02	0.01	0.01	0.03	0.00	0.00
Zr	2.03	2.03	2.04	2.01	2.04	1.79	1.94
Nb	0.08	0.07	0.01	0.11	0.01	0.02	0.02
O	17	17	17	17	17	17	17

eudialytes. Enrichment of Nb in the Sushina eudialyte indicates increased NaCl activity in the melt/fluid from which they crystallized. The most notable feature of the host nepheline gneiss is the complete absence of zircon compared to the other units of nepheline syenite gneiss (miarctitic) present in the near vicinity (Chakrabarty, 2009). The dominant Na-Zr phase in the studied nepheline syenite gneiss is eudialyte and some unknown NZS. This is possible when NaCl activity of the melt is sufficiently high so that zircon, if present, will go into the solution to form eudialyte. The observed mineralogical assemblage of eudialyte + aegirine ± amphibole thus can be represented by the following NaCl consuming reactions as described by Marks et al. (2011):



These reactions also indicate that sufficient supply of Na is required to form both eudialyte and aegirine. This can be achieved by the break down of Na bearing phases such as nepheline, albite and aegirine during their interaction with

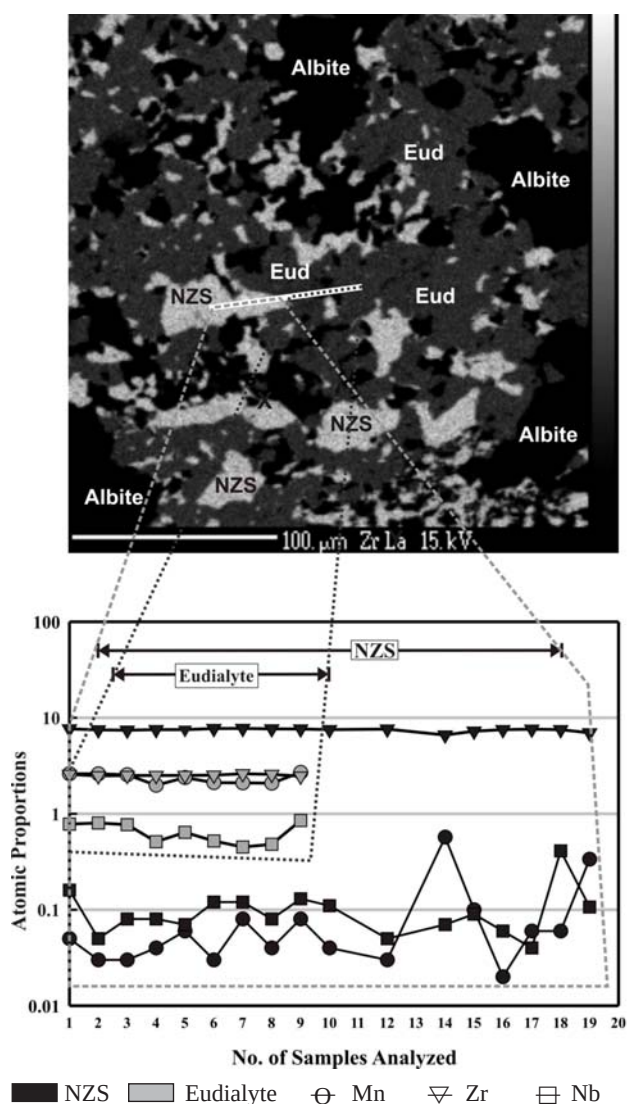


Fig.5. (a) BSE image showing the alteration of late-magmatic eudialyte by NZS (brighter zones); (b) Atomic proportions in terms of Mn-Nb-Zr are plotted for both late-magmatic eudialyte and the NZS. The NZS are rich in Zr but poorer in Nb and Mn compared to the associated eudialyte. High Mn content in one of the analyzed sample of NZS (analysis no: 14) is mainly due to the presence of serandite as an alteration product of eudialyte.

the fluid phase associated with the parent magma of the precursor nepheline syenite. However, the presence of aegirine as dominant mafic phase itself indicates that the $\mu\text{Na}_2\text{O}$ of the parent melt was high enough to promote the above mentioned reactions resulting in the formation of eudialyte in the studied unit of nepheline syenite gneiss. This is well supported by the observed Fe depletion in the eudialyte which indicate that eudialyte was stabilized either after the formation of the aegirine or consanguineous to the aegirine, as most of the Fe was utilized during the aegirine

crystallization. In general, the formation of eudialyte in a sodalite free system as in the case of this study indicates that eudialyte is essentially a late-magmatic phase (Marks et al. 2011). Thus we infer that the REE free eudialytes are late-magmatic in origin which is also well supported by the textural features mentioned earlier. The post-magmatic hydrothermal stage evidenced formation of hydrothermal eudialyte which is enriched in REE and Sr. This indicates that the time lag between the formations of the two compositionally different eudialytes was very small and the deuteritic fluid immediately reacted with the pre-existing minerals such as albite, aegirine, and late-magmatic eudialyte which resulted in the formation of hydrothermal eudialyte. This is in accordance with the observed textural features such as complex association of the hydrothermal eudialyte with aegirine ($\pm$ amphibole) and albite (Fig. 1j). Moreover, the enrichment of Sr and Ce in the hydrothermal eudialyte indicates that these elements behave incompatibly in the late-magmatic eudialyte.

The formation of NZS at the expense of eudialyte is an interesting phenomenon. The eudialyte formation requires certain level of NaCl activity in the melt/fluid which indicates increased alkalinity or pH during the time of eudialyte formation. In such a situation, owing to increased pH most of the Zr must go in to the solution as shown by the reactions (1) and (2). Now, if the pH decreases this would lead to the precipitation of Zr and formation of NZS like phases. Thus the replacement of eudialyte by NZS is an indication of change in alkalinity from high to low pH conditions. However, this effect also applies for other HFSE elements such as Nb and Ti as reflected by the alteration of both types of eudialyte to pyrochlore, whölerite, titanite like phases (Mitchell and Chakrabarty, 2011; Chakrabarty et al. in prep). The Mn enrichment in eudialyte can not be explained adequately and we suggest that the deuteritic fluid was itself enriched in Mn. As mentioned earlier, the late-post magmatic eudialytes are generally enriched in Mn and our observation of high Mn content for the all Sushina EGM (Schilling et al. 2011) thus indicates their highly evolved nature of origin as seen in some Mont-Saint Hilaire, Pilanesberg and Ilímaussaq eudialytes. No field evidence exists as to the extraneous fluid source in the form of veins or pegmatitic bodies of similar mineralogical assemblage associated with the studied unit of the syenite gneiss and thus we assigned this fluid as deuteritic fluid which was associated with the unmetamorphosed nepheline syenite. The replacement of eudialyte by NZS certainly indicates that the rock has experienced extensive subsolidus deuteritic alteration prior to the metamorphism.

CONCLUSIONS

Eudialyte bearing Sushina syenite gneissic complex represents unique EGM chemical compositions. Two different textural modes are observed for the Sushina eudialyte viz. a viz. late-magmatic and hydrothermal/post-magmatic eudialytes. The late-magmatic eudialytes are relatively smaller in size and at places relicts of relatively less altered grains are also present. The hydrothermal eudialytes on the other hand dominant over the late-magmatic one and frequently form complex aggregates composed of albite, nepheline and aegirine. In terms of their chemical compositions the following conclusions can be drawn:

- Sushina eudialytes are enriched in Mn-Nb-Ca. The lower limit of enrichment for these elements is much higher compared to all other EGM reported world-wide. The extremely high Mn content of the Sushina eudialytes indicates that they are highly evolved in nature.
- Owing to their high Mn-Nb content, the calculated site occupancy makes the Sushina eudialytes the second to be reported, after some Pilanesberg eudialyte, which contain significant Nb and Mn at $[M(3)]$ and $[M(2)]$ sites respectively.
- REE and Sr behave incompatibly in the agpaitic system and are incorporated preferentially in the hydrothermal eudialytes at least in this case.
- Compositional variability between the two different types of Sushina eudialytes and their replacement by NZS can be used to understand changes in alkalinity (pH) during the evolution of the magma from the late-magmatic to post-magmatic/hydrothermal stages. Eudialyte formation indicates elevated pH conditions and the formation of NZS takes place at decreasing pH conditions.
- The deuteritic fluid associated with the unmetamorphosed agpaitic syenite is suggested to be enriched in Mn, Nb, Ca, Zr and Cl.
- The present study is in agreement with observations made by Schilling et al. (2011) that the eudialyte compositions can be used in metamorphosed agpaitic rock to trace out its magmatic history as metamorphic fluids do not destabilized EGM up to the amphibolite facies of metamorphism.

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