

CHAPTER 4

DISCUSSIONS AND CONCLUSION

4.1 Gemological properties

The gemological properties of green tourmalines examined for this study, and comparable reports from other localities are summarized in Table 4.1.

Table 4.1 Gemological properties of green tourmaline samples used in this study compared with reports from other localities

Localities	Colors	S.G.	R.I.	Birefringence
Madagascar (this study)	green, bluish green, yellowish green to brownish green and watermelon tourmaline	2.99-3.11	$N_o = 1.640-1.645$ $N_e = 1.620-1.625$	0.017-0.021
Mozambique (this study)	greenish blue, blue, green and bi-color tourmaline	2.99-3.12	$N_o = 1.640-1.643$ $N_e = 1.620-1.625$	0.017-0.021
Tanzania (this study)	yellowish green, yellow-green and green	2.98-3.15	$N_o = 1.640-1.644$ $N_e = 1.621-1.625$	0.017-0.021
Brazil ¹	bluish green, greenish blue	3.09	$N_o = 1.640-1.641$ $N_e = 1.619-1.620$	0.021
Africa ¹	Green	3.09	$N_o = 1.642$ $N_e = 1.622$	0.02
Afghanistan ₁	bluish green, greenish blue	3.05-3.07	$N_o = 1.638-1.639$ $N_e = 1.619-1.620$	0.019-0.020
Namibia ¹	greenish blue	3.09	$N_o = 1.639$ $N_e = 1.620$	0.019
Nigeria ¹	green, blue	3.06	$N_o = 1.638$ $N_e = 1.619$	0.019
USA ²	yellowish green, greenish blue	3.04-3.08	$N_o = 1.638-1.640$ $N_e = 1.619-1.622$	0.018-0.020
Rwanda ³	yellowish green, bluish green and green	3.04-3.07	$N_o = 1.639-1.641$ $N_e = 1.619-1.621$	0.020

¹Merkel and Breeding, 2009; ²Simmons and others, 2005; ³Henn and Schmitz, 2014

The S.G. values of samples in this study presented a wide range than the values for green tourmaline from other localities reported by Merkel and Breeding (2009); Simmons and others (2005), with lower specific gravity values for some samples from Tanzania, while the specific gravity of samples from Madagascar and Mozambique have higher S.G. values and similar to green tourmaline from other localities. The specific gravity, refractive indices and birefringence of tourmaline increases with contents of transition metals e.g. Fe, Mn, Ti (Deer and others, 1997) like as green tourmaline samples from Madagascar and Mozambique.

Microscopic examination revealed internal features that are typical of inclusion in tourmaline such as growth tubes, hollow tubes, healed fractures, fluid inclusions, two-phase (liquid-gas) inclusions and small fractures (Webster, 1994; Dietrich, 1985; Gubelin and Koivula, 2005). All specimens exhibited a wide variety of internal features. Most samples were presented healed fractures containing fluid- and/or two-phase (liquid-gas) inclusions (Abduriyim and others, 2006). The samples occurred as trichite, are fluid-filled cavities linked by networks of very thin capillaries (Lauris and others, 2008; Simmons and others, 2005; Dirlam and others, 2002). Few samples showed flat fluid inclusion (Lauris and others, 2008). Hollow tubes and growth tubes oriented parallel to the C-axis, were found common (Henn and Schmitz, 2014; Lauris and others, 2008). Small fractures were observed in most samples, and are filled with ion stains.

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4.2 UV-Vis-NIR absorption spectroscopy

UV-Vis-NIR absorption spectra were measured in all green tourmaline samples to determine the causes of coloration.

The absorption spectrum of green tourmaline from Madagascar and Mozambique were presented the similar spectral range. The narrow band near 332, 385, 400 and 415 nm are assigned to Mn^{2+} from d-d transitions at the octahedral site (Castaneda and others, 2006; Reinitz and Rossman, 1988; Rossman and Mattson, 1986). A low intensity Mn^{3+} band was observed in a few samples from Madagascar near 510, 517 and 520 nm (Reinitz and Rossman, 1988). Reddy and others (2007) reported that the band at 520 nm due to Mn^{3+} ions in the high spin state, distorted octahedral sites. A strong band centred at 720 nm related to Fe^{2+} - Fe^{3+} intervalence charge transfer (IVCT) and the spectrum ordinary ray (90°) had a higher concentration of iron (Reddy and others, 2007; Mattson and Rossman, 1987). There are reported that assigned the band to Fe^{2+} octahedral in Y site (Liu and others, 2011; Da Fonseca-Zang and others, 2008; Castaneda and others, 2006), while De Camargo and Isotani (1988) reported that the band presence of Fe^{2+} at Z site. The strong bands between 918 to 1220 nm are related to Fe^{2+} octahedral in Y site (Liu and others, 2011). The sharp absorption bands in the 1400-1500 nm are attributed to hydroxyl groups, noticeably parallel to the c-axis (Rossman, 2011).

Schmetzer (1978) noted that chromium and vanadium both cause absorption bands at the same position in the UV-Vis-NIR spectra. The green tourmaline from Tanzania showed absorption spectra of the main transition metal at 307, 390, 432, 448, 608 and 628 nm are attributed to V^{3+} on octahedral coordinated aluminium-sites (Z-sites) (Schmetzer, 1978; 1982). A small peak at 417 nm is related to Cr^{3+} , which are in the same spectral range of vanadium (Ertl and others, 2008). Schmetzer and others (2007) reported that the spectral of V^{3+} and Cr^{3+} causes the green coloration in uvite samples. The ratio of V:Cr to determine by chemical analyses presented that have higher vanadium than chromium contents and the UV-Vis-NIR spectra showed characterized by the two broad bands related to V^{3+} (Schmetzer and Bank, 1979;

Schmetzer, 1978). Absorption band at 1425 nm is due to hydroxyl groups. The UV-Vis-NIR absorption spectra comparisons of green tourmaline samples from Madagascar, Mozambique and Tanzania were displayed in Figure 4.1.

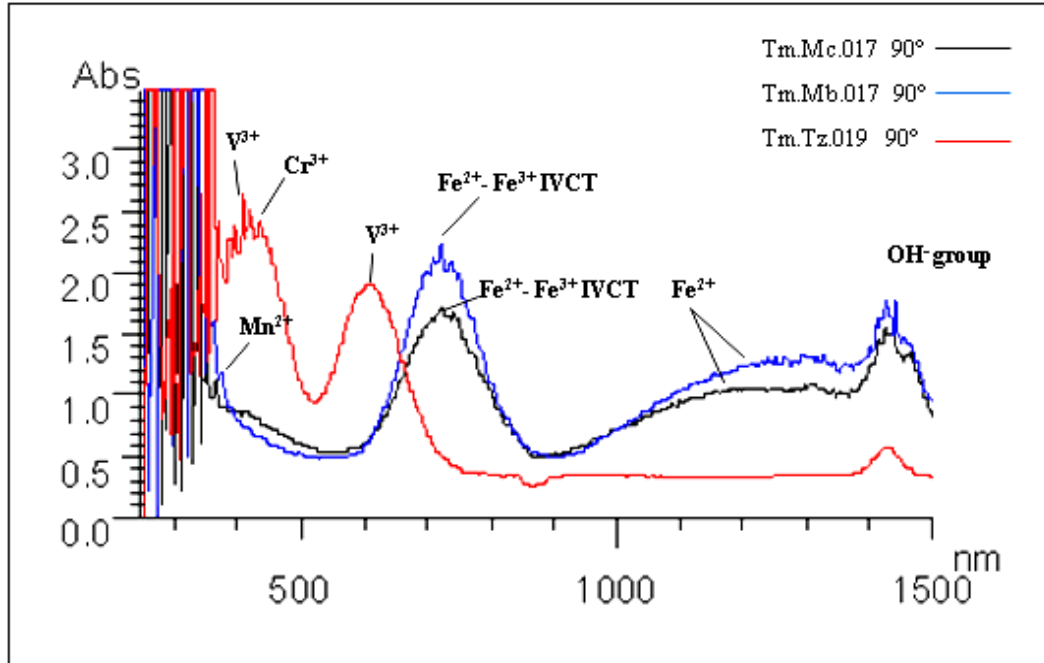


Figure 4.1 UV-Vis-NIR absorption spectra comparisons of green tourmaline samples from Madagascar, Mozambique and Tanzania.

4.3 Infrared absorption spectroscopy

Infrared absorption spectroscopy was obtained the stretching vibration of Si-O, BO₃ groups and hydroxyl groups in the structure of typical green tourmaline samples. The absorption spectrum of green tourmaline from Madagascar, Mozambique and Tanzania were presented the similar spectral range. The vibration absorption bands around 1200-820 cm⁻¹ is assigned to the stretching vibration of Si₆O₁₈ rings (Ahn and others, 2013; Reddy and others, 2007). The band at 1110 cm⁻¹ is attributed to MgOH bending modes (Reddy and others, 2007). The sharp bands at around 1350 and 1250 cm⁻¹ are identified as the stretching vibration modes of BO₃ groups (Ahn and others, 2013; Reddy and others, 2007). The stretching modes of hydroxyl groups are observed in the range from 3700 to 3400 cm⁻¹ (Hainschwang and others, 2007). Reddy and others

(2007) documented hydroxyl groups (OH1 and OH3) are distributed two different sites (Y- and Z- cation sites) and the bands are related to occupancy of octahedral site. The local vibrations OH groups associated with Al in Z sites and Fe, Mn or Mg in Y sites near 3555 cm^{-1} . Some authors reported that the bands located at 3657 cm^{-1} are assigned to OH stretching vibration at W(O1) site whereas the band at 3586 cm^{-1} related to OH vibration at V(O3) site (Ahn and others, 2013; Skogby and others, 2012; Hoang and others, 2011). The polarized FTIR absorption spectra comparisons of green tourmaline samples from Madagascar, Mozambique and Tanzania were presented in Figure 4.2 a, b and c.

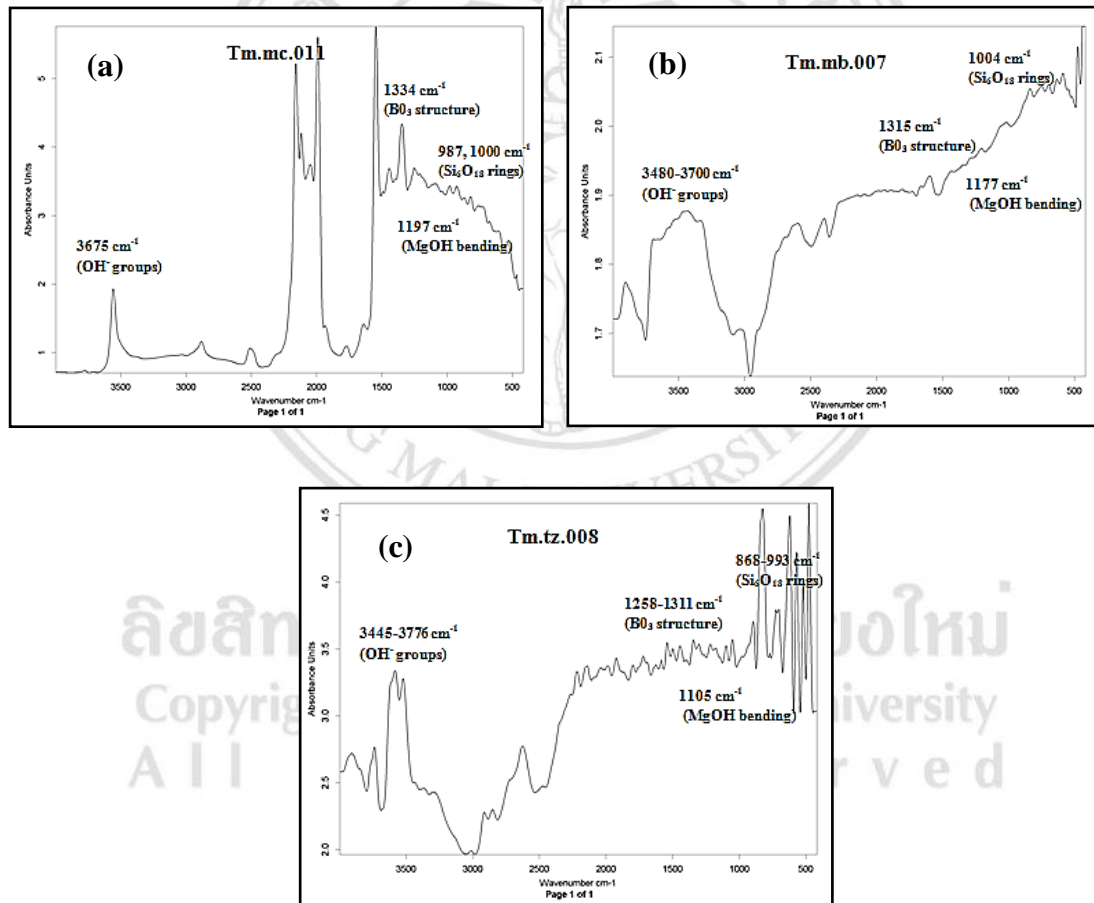


Figure 4.2 The polarized FTIR absorption spectra comparisons of sample from (a) Madagascar, (b) Mozambique and (c) Tanzania

4.4. Chemical composition

Quantitative chemical analyses in this study were obtained by electron probe micro-analyser (EPMA) and Laser Ablation-Inductively coupled plasma-mass spectroscopy (LA-ICP-MS).

Major elements of green tourmalines from Madagascar: SiO₂ (36.12 to 38.67 wt.%), Al₂O₃ (40.04 to 40.40 wt.%) and B₂O₃ (10.61 to 11.10 wt.%) are similar to natural tourmaline (Dirlam and others, 2002). Chemical analyses in this study correspond to elbaite position. The elbaite samples were analyzed Na content (1.75 to 2.36 wt.%) more than Ca content (0.17 to 0.72 wt.%). The most Ca-rich (4.15 wt.%) and Na-poor (0.96 wt.%) were liddicoatite from Madagascar (Dirlam and others, 2002). Akizuki and others (2001) were republished contained Ca content (4.69 wt.%) and Na content (0.54 wt.%). Tourmaline from Madagascar can be identified both elbaite and liddicoatite because of tourmaline solid solutions involve the occupancy of equivalent structural sites by difference ions. Moreover, it can be expected in members of any mineral group with cation sites in which substitutions possible (Dietrich, 1985). In the case of the chemical substitution this locality occurs from elbaite-liddicoatite series that is based on Na⁺: Ca²⁺ differences in X-site, which are composed of charge compensating differences between Al³⁺ and Li⁺ in the Y-sites (Superchi and others, 2006; Hawthorne and Henry, 1999). Dunn and others (1977) reported that liddicoatite cannot be different from elbaite by its optical and physical characteristics. However, quantitative chemical analyses are required to confirm the tourmaline species. The chromophoric elements Fe, Mn and Ti were typically in Madagascar tourmaline (Dirlam and others, 2002). These chromophoric affect the coloration of liddicoatite in the same way they affect elbaite (Dietrich, 1985). The highest Fe values (3.02 to 3.57 wt.%) were found in green color (Webber and others, 2002). The Mn content is a wide range in the various colors of samples. The high content of Mn was observed in green and watermelon tourmaline (spot in pink core) samples. The Ti content is very low or below detection limit in these samples. The most abundant trace elements in this study were Zn, Ga, Sn, Pb and Bi. The trace element amounts of Zn and Pb are more common

in green and blue elbaite (Dietrich, 1985). The multicolored tourmalines (pink, purple, red, olive green, yellow-brown and yellow) from the Antseny pegmatite, Madagascar have been found the trace elements Bi and Pb, whereas multicolored tourmalines from the Anjahamiary pegmatite, Madagascar have been found only Pb (Superchi and others, 2006).

Major element contents of green tourmalines from Mozambique contain SiO_2 (35.01 to 36.12 wt.%), Al_2O_3 (40.26 to 40.93 wt.%) and B_2O_3 (10.52 to 11.28 wt.%) and are similar with natural tourmaline (Laurs and others, 2008; Abduriyim and others, 2006). The analyses fell in elbaite field of alkali tourmaline. The samples were elbaite component mostly lower in Ca content (0.23 to 0.57 wt.%) and richer Na content (1.08 to 2.40 wt.%) (Simmon and others, 2005). The chromophoric elements Fe, Mn and Ti exhibited strong correlation with color. Fe is the most important trace element. The next-highest Fe values (3.18 to 3.74 wt.%) were found in green and greenish blue color tourmaline similar to tourmaline from Mt.Mica, USA (1.79 to 4.46 wt.% FeO) (Simmon and others, 2005). All samples had very low Ti contents. Mn was highest in bi-color and greenish blue. By comparison, the Mn contents of the samples from Mozambique have the same range with the samples from Madagascar (0.54 to 2.20 wt.%). Shigley and others (1986) noted that elbaite containing minor amounts of Mn are common and appear to be pegmatitic origin including green color. The most abundant trace elements Zn and Sn were detected in the samples from Mozambique.

Chemical composition identified the samples from Tanzania as Ca-Mg tourmaline. Chemical analyses correspond with end-member uvite (Hainschwang and others, 2007). The Ca-rich as found in X-site ranging from 3.74 to 4.64 wt.% CaO and high Mg contents were ranging from 12.12 to 13.30 wt.% MgO. Schmetzer and others (2007) reported that the magnesium-bearing calcic tourmalines with $\text{Al}<6$ and $\text{Mg}>3$ apfu (atoms per formula unit), is consistent with composition of uvite. The specimens Al content has ranging from 5.152 to 5.170 apfu and the Mg content ranged from 2.916 to 3.234 apfu. Therefore, the samples in this study can be designated as uvite. Henry and Guidotti (1985) noted that tourmaline with $\text{Al}<6$ and $\text{Mg}>3$ apfu (uvite) originate from metacarbonates and meta-pyroxenites. The

chromophoric elements were vanadium, with a minor influence from chromium (Schmetzer and Bank, 1979). The highest V values (0.19 to 0.25 wt.%) were found in green color. The ratio of V: Cr determined by chemical analyses showed that vanadium is strong dominant (Schmetzer and Bank, 1979; Schmetzer, 1978). The trace elements of green tourmalines from Tanzania are presented high values quantities of Ti, V, Cr and Sr. Representative EPMA analyses and LA-ICP-MS chemical data of green tourmaline of various colors from Madagascar Mozambique and Tanzania are shown in Tables 4.2 and 4.3.

Table 4.2 EPMA analyses of green tourmaline of various colors from Madagascar, Mozambique and Tanzania

Chemical composition	Madagascar				Mozambique				Tanzania			
	yellowish Green to brownish Green	Green	watermelon		Blue	greenish Blue	Green	Bi-color		Yellow-Green	yellowish Green	Green
			yG	pR				B	bG			
Oxide (wt.%)												
Al ₂ O ₃	40.04	40.35	40.10	40.40	40.70	40.93	40.26	40.33	40.61	27.18	27.15	27.19
SiO ₂	36.12	36.12	38.67	38.35	37.93	35.14	36.12	36.19	35.01	38.23	38.96	38.58
V ₂ O ₅	bdl	bdl	bdl	bdl	bdl	0.01	bdl	bdl	0.02	0.19	0.20	0.25
MnO	0.54	1.66	1.05	1.17	0.82	1.26	0.99	2.20	1.74	0.01	0.02	bdl
TiO ₂	0.02	bdl	bdl	0.01	bdl	0.01	0.01	bdl	bdl	0.12	0.12	0.20
F	0.72	0.68	0.47	0.58	0.48	0.76	0.52	0.54	0.62	0.54	0.53	0.15
B ₂ O ₃	11.10	11.02	10.61	10.73	11.28	10.52	11.14	10.84	11.23	10.57	10.42	10.85
Cr ₂ O ₃	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.02	0.08	0.07	0.09
FeO	3.26	3.57	3.25	3.02	2.51	3.18	3.74	2.22	3.39	0.03	0.01	bdl
CaO	0.72	0.36	0.17	0.45	0.37	0.27	0.23	0.57	0.39	4.64	3.74	3.76
MgO	0.05	bdl	0.09	0.10	bdl	0.06	0.01	0.03	0.03	13.36	13.20	12.12
Na ₂ O	1.99	2.36	1.84	1.75	1.08	2.40	2.17	2.08	2.37	0.88	1.24	1.16
CuO	0.01	bdl	0.02	0.02	0.01	0.03	bdl	0.03	bdl	bdl	bdl	0.02
K ₂ O	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.02	0.02	0.03	0.03	0.07
Li ₂ O calc.	1.80	1.55	1.56	1.51	1.85	1.36	1.61	1.57	1.62	0.89	1.09	1.32
H ₂ O calc.	3.41	3.45	3.59	3.55	3.62	3.34	3.52	3.49	3.45	3.47	3.51	3.65
Subtotal	99.80	101.14	101.44	101.66	101.57	99.22	100.38	100.09	100.52	100.21	100.76	99.39
-O-F	0.30	0.29	0.20	0.24	0.20	0.32	0.22	0.23	0.26	0.23	0.22	0.06
Total	99.50	100.85	101.24	101.42	101.37	98.90	100.16	99.86	100.26	99.98	100.54	99.33
Ions per 31 (O,OH,F)												
T: Si	5.775	5.746	6.076	6.018	5.918	5.694	5.758	5.790	5.601	6.161	6.216	6.217
Al	0.225	0.254	0.000	0.000	0.082	0.306	0.242	0.210	0.399	0.000	0.000	0.000
B	3.063	3.026	2.878	2.906	3.038	2.942	3.065	2.993	3.101	2.940	3.000	3.018
Z: Al	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	5.162	5.105	5.164
Mg	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.838	0.895	0.836
Cr	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Fe ³⁺	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Y: Al	1.320	1.311	1.426	1.472	1.403	1.510	1.322	1.394	1.258	0.000	0.000	0.000
Ti	0.002	0.000	0.000	0.001	0.000	0.001	0.001	0.000	0.000	0.015	0.014	0.024
V	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.003	0.025	0.026	0.032
Cr	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.010	0.009	0.011
Fe ³⁺	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mg	0.012	0.000	0.021	0.023	0.000	0.000	0.014	0.002	0.007	2.372	2.245	2.076
Mn	0.073	0.224	0.140	0.156	0.108	0.173	0.134	0.298	0.236	0.001	0.003	0.000
Fe ²⁺	0.436	0.475	0.427	0.396	0.328	0.431	0.499	0.297	0.454	0.004	0.001	0.000
Zn	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Li*	1.157	0.990	0.986	0.952	1.161	0.884	1.031	1.009	1.040	0.573	0.702	0.856
?Y	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
X: Ca	0.123	0.061	0.029	0.076	0.062	0.047	0.039	0.098	0.067	0.801	0.639	0.649
Na	0.617	0.728	0.561	0.532	0.599	0.754	0.671	0.645	0.735	0.275	0.384	0.362
K	0.004	0.004	0.004	0.004	0.004	0.002	0.004	0.004	0.004	0.006	0.006	0.014
r	0.256	0.207	0.407	0.388	0.335	0.197	0.286	0.253	0.194	0.000	0.000	0.000
OH	3.636	3.658	3.766	3.712	3.763	3.611	3.738	3.727	3.686	3.725	3.733	3.924
F	0.364	0.342	0.234	0.288	0.237	0.389	0.262	0.273	0.314	0.275	0.267	0.076
Cl	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mineral name	Elbaite	Elbaite	Elbaite	Elbaite	Elbaite	Elbaite	Elbaite	Elbaite	Elbaite	Uvite	Uvite	Uvite

Abbreviation: bdl = below detection limit

Li₂O, H₂O were calculated as mineral structural formula based on stoichiometric principles

Table 4.3 LA-ICP-MS chemical data of green tourmaline of various colors from Madagascar, Mozambique and Tanzania

Trace element (ppm)	Madagascar				Mozambique					Tanzania		
	yellowish Green to brownish Green	Green	watermelon		Blue	greenish Blue	Green	Bi-color		Yellow-Green	yellowish Green	Green
			yG	pR				B	bG			
Li	1619.23	1424.74	1655.42	1722.44	1451.21	1304.49	1277.61	1413.09	1940.82	4.05	12.98	6.39
Be	3.52	2.61	4.06	6.95	4.66	6.74	3.42	2.98	3.23	6.13	2.34	2.26
Sc	8.74	8.42	8.11	8.07	7.01	7.54	6.36	6.23	6.51	5.40	6.19	6.11
Ti	29.05	14.96	86.57	52.25	8.85	7.92	25.11	7.20	8.17	555.03	607.27	690.10
V	0.68	0.43	0.77	1.28	0.58	0.56	0.69	0.63	0.63	170.72	552.47	645.25
Cr	7.49	5.98	11.20	16.20	9.81	6.86	7.10	8.04	7.69	18.21	249.92	492.66
Fe	46845	55462	947	283	26596	34647	39368	14608	20110	66.30	28.56	25.69
Ni	2.97	2.12	4.59	6.01	4.94	4.52	3.14	2.96	2.67	8.42	2.31	3.27
Zn	150.87	77.89	140.64	29.32	128.08	667.13	258.77	32.17	51.41	17.55	6.29	5.41
Ga	28.78	14.94	69.08	58.80	13.66	12.20	18.40	10.24	8.43	2.91	0.84	6.79
Ge	5.83	3.59	5.81	9.18	5.61	10.21	9.92	6.19	4.05	8.33	3.27	2.99
Sr	4.55	2.42	0.55	0.72	0.91	6.29	0.32	1.14	1.23	201.05	92.18	64.76
Nb	1.09	0.49	0.42	0.76	0.41	0.40	0.53	0.54	0.37	0.78	0.32	0.33
Mo	2.74	1.70	3.13	5.06	2.54	2.45	2.28	3.23	1.51	4.00	1.56	2.37
Sn	9.12	26.89	5.52	8.23	21.22	16.30	13.52	29.18	25.88	4.93	1.63	2.02
Sb	4.06	1.06	2.41	3.21	1.88	1.29	1.53	1.44	1.54	3.26	1.12	1.48
Ta	0.88	0.51	0.53	0.75	0.62	1.42	0.45	0.39	0.41	0.82	0.39	0.37
Pb	44.39	58.34	3.80	5.76	11.72	31.81	9.52	2.27	2.13	3.68	1.01	1.34
Bi	15.98	0.66	37.3	71.76	0.97	0.67	1.02	0.70	0.87	1.49	0.63	0.66

4.5. Conclusion

1. Green tourmalines from Madagascar, Mozambique and Tanzania show color in green, bluish green, yellowish green to brownish green, greenish blue and blue. Their gemological properties are typical range of natural tourmaline. Internal features are normal of inclusion in tourmaline such as growth tubes, hollow tubes, healed fractures, fluid inclusions, two-phase (liquid-gas) inclusions, crystal inclusion and small fractures.

2. The cause of coloration in green tourmaline was analyzed by UV-Vis-NIR absorption spectral. Samples from Madagascar and Mozambique present the same spectral range. The absorption bands near 332, 385, 400 and 415 nm are assigned to Mn^{2+} from d-d transitions at the octahedral site. The intensity of absorption bands at 720 and 732 nm are related to $Fe^{2+} - Fe^{3+}$ IVCT, while strong bands between 918 to 1220 nm are related to Fe^{2+} octahedral in Y site. The samples from Tanzania show absorption spectra at 307 to 628 nm are due to V^{3+} on octahedral sites (Z-sites) and a weak peak at

417 nm is related to Cr^{3+} . The green tourmalines from three localities present intense bands centred at 1425 nm are attributed to hydroxyl groups.

3. The FTIR absorption spectra in the range $4000\text{-}500\text{ cm}^{-1}$ was used to characterized the stretching vibration of Si_6O_{18} rings ($1200\text{-}820\text{ cm}^{-1}$), BO_3 (1350 to 1250 cm^{-1}) and hydroxyl groups (3700 to 3400 cm^{-1}) in the structure of tourmaline.

4. Electron probe micro-analyser (EPMA) of samples from Madagascar and Mozambique present the same chemical compositions that correspond to near end-member elbaite mostly lower in Ca content and richer Na content, whereas the samples from Tanzania identified as Ca-Mg rich tourmaline that related to end-member uvite. The chromophoric elements Fe and Mn were usually in Madagascar and Mozambique tourmaline. The Fe is the most important trace element these both localities and found in green color. The main chromophoric elements of green tourmaline from Tanzania were vanadium, with a minor influence from chromium. The green samples contained highest V concentration.

5. This study has presented that chemical fingerprinting by LA-ICP-MS technique is helpful for differentiate green tourmalines from the various localities. The Madagascar samples showed high contents of Li, Fe, Zn, Ga, Sn, Pb and Bi, whereas the Mozambique samples were enriched in Li, Fe, Zn and Sn. The green tourmalines from Tanzania contained amounts of the trace elements Ti, V, Cr and Sr. The minor and trace elements of samples from Madagascar and Mozambique are more common in green and blue elbaite and associated with characteristic occurrence of tourmaline is in granitic pegmatites, whereas samples from Tanzania are associated with occurrence in metamorphic rocks.