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Fe^{3+} - RICH KATOITE FROM PERMIAN BASALTS (HRONICUM, WESTERN CARPATHIANS, SLOVAKIA): COMPOSITION AND ORIGIN

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Abstract: The study of Permian basalts (rift-related continental tholeiites) from the Nízke Tatry Mts., Hronicum Superunit, Western Carpathians (Slovakia) has shown the presence of an assemblage containing hydrated andradite-grossular garnet. The primary minerals of basalts in this assemblage are plagioclase, clinopyroxene, magnesio-hornblende, biotite and fine-grained matrix. This association is partly altered and hydrated garnets together with chlorites formed by replacement of primary clinopyroxenes. Other secondary minerals are albite, actinolite and rarely pumpellyite-prehnite. The hydrated garnet is characteristic for high contents of Ca, Fe and Al, increased contents of Ti and very low contents of Mn, and Mg. The presence of OH in the garnet was confirmed in Raman spectroscopy. According to the new nomenclature of garnets (IMA), this garnet corresponds to kenogarnet (katoite).

Key words: kenogarnet, katoite, basalt, Nízke Tatry Mts., Hronicum, Permian

1. INTRODUCTION

The microscopic study of dolerite dykes belonging to the Hronicum Permian basalts (Nízke Tatry Mts., Fig. 1) revealed a mineral formed by a metasomatic alteration of clinopyroxene and/or plagioclase. The electron microprobe analysis of the mineral proved it is hydrated garnet.

The term "hydrogrossular" was introduced to the literature by Hutton (1943). In the case of hydrated Ca-garnets, it is an isomorphous series of grossular-katoite. The term "hibschite" is used for members with a substitution of (SiO_4)-tetrahedra < 50% by (OH)-group and formula $Ca_3Al_2(SiO_4)_{3-x}(OH)_{4x}$ and x within range 0<x<1,5. Members with a substitution of 50 % to 100 % are named as katoite (Pasaglia & Rinaldi 1984). Modern physical and chemical investigation of natural hydrogrossulars confirms substitution of SiO_4 by OH (Pertlik 2003). In the past, also the terms "hydrograndite" (Huckenholz et al., 1981) or "hydrougrandite" (Zabinski 1966) were used for hydrogarnets of andradite grossular composition. The new nomenclature of the garnet supergroup approved by the Commission on New Minerals and Mineral Names (Grew et al., 2013) regards many of formerly used names obsolete.

The main objective of our research was to examine the relationship between clinopyroxene and garnet and find out about the conditions under which they originated. We studied the composition of clinopyroxene, hydrated garnet and chlorite by microprobe and OH content in the structure of garnets by Raman spectroscopy.

2. ANALYTICAL METHOD

The thin sections were studied with a NIKON Eclipse LV 100N microscope. The selected minerals were quantitatively studied on Fieldemmision microprobe JEOL JXA-8530F (Earth Science Institute of the Slovak Academy of Sciences, Banská Bystrica, Slovakia) at following condition: 15kV, 20nA, spot diameter 2-4 μm, counting time 10s–peak and 5s for background, ZAF correction. Used standards: diopside (CaKα PETL, SiKα TAP), rhodonite (MnKα LIFL), albite (NaKα, AlKα TAP), fluorite (FKα LDE1), olivine (MgKα TAP), rutile (TiKα PETH), hematite (FeKα, LIFH), orthoclase (KKα PETJ.

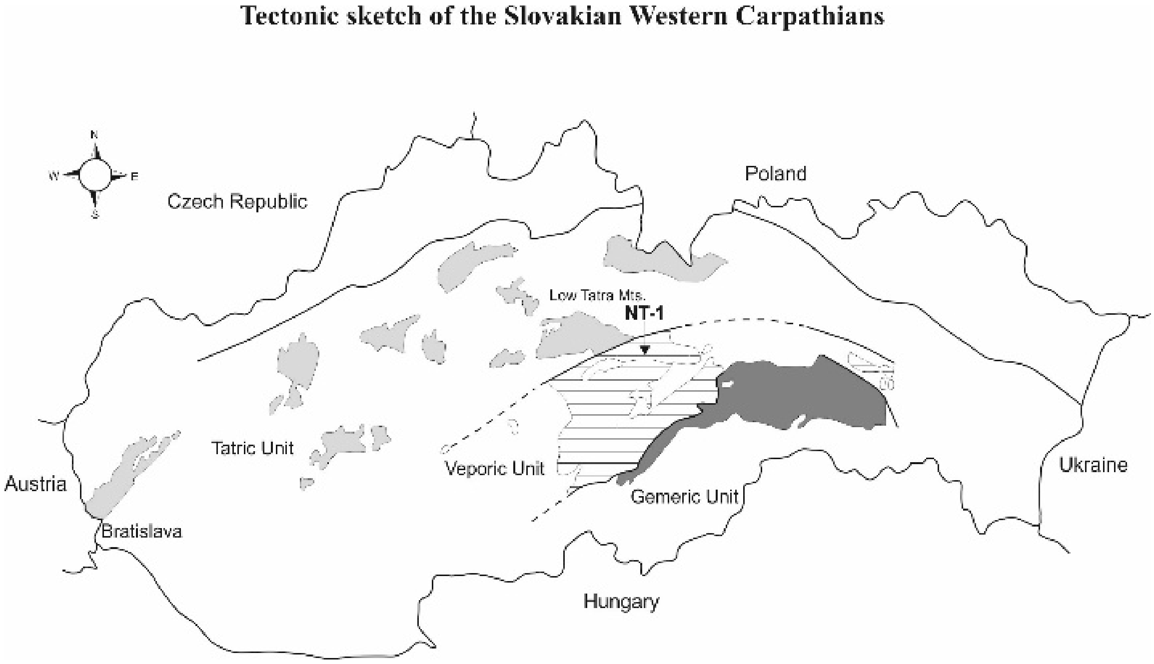

Figure 1. Tectonic sketch of the Western Carpathians with the studied locality

Raman spectra of hydrated garnets in thin sections were acquired on microspectrometer LabramHR (Horiba Jobin-Yvon, Earth Science Institute, Slovak Academy of Sciences, Banská Bystrica, analyst Stanislava Milovská), based on an Olympus BX41 microscope with confocally coupled Czerny-Turner type monochromator (focal length 800 mm). A polarized laser emission at λ=532 nm was used for excitation (frequency-doubled Nd:YAG laser) with approx. 200 mW power on sample. Scattered light was collected through a 100x objective lens with numerical aperture 0.8 and dispersed by diffraction grating with density 1800 gr/mm onto a cooled CCD detector, exposition time was 200 s. Possible artifacts and photoluminescence peaks were excluded by observation with He-Ne laser excitation (λ=633 nm).

3. GEOLOGY

Hronicum, as a rootless multi-nappe unit in the tectonic structure of the Inner Western Carpathians, is characterized by dominant Carboniferous/ Permian volcanic-sedimentary sequences and is defined as Ipoltica Group by Vozárová & Vozár (1981, 1988). The lithological and lithofacial characteristics, mineral composition of detritic material, as well as the type synsedimentary volcanism (Vozárová 1981, Vozárová & Vozár 1981, 1988) permits to interpret the original basin as a consequence of continental rifting in the post-collisional stage of the Variscan orogeny (Vozárová 1996). All the mentioned data enable to presuppose a sedimentary basin 450 to 550 kms of total length and considerable width of several km to tens of km. The total 2200 to 2800 m present thickness of the Ipoltica Group also corresponds to it.The 310-340 Ma age of the supposed source area was indicated by the $^{40}Ar/^{39}Ar$ dating of detrital mica from sandstones (Vozárová et al., 2005), which points to an age of the Ipoltica Group sedimentary basin younger than 310 Ma. An essentially more intensive Permian andesite-basalt volcanism is concentrated into two distinct eruption phases. The presence of volcanic products is already evident in the upper horizons of the Nižná Boca Formation (sporadical dacite effusions and its volcaniclastics). Compositionally, the basic rocks were described as rift-related continental tholeiites (Dostál et al., 2003, Vozár et al., 2015).

4. RESULT AND DISCUSSION

Individual sills in the Nižná Boca Formation vary in thickness, from a few to more than 50 metres. There are also infrequent occurrences of subvertical dykes. The central parts of the sills and dikes are medium-grained and show doleritic, less commonly ophitic textures. The peripheral parts are fine-grained and have principally a microdoleritic texture.

The mineral composition of dolerites consists of plagioclase (45%), clinopyroxene (25%), magnesio-hornblende (2%), altered biotites, fine-grained dark matrix, and opaque minerals (ilmenite, magnetite, titanomagnetite, and rutile) (Fig. 2). The detected secondary minerals are albite, chlorite, actinolite, hydrated garnets and occasionally pumpellyite-prehnite (Vrána & Vozár 1969).

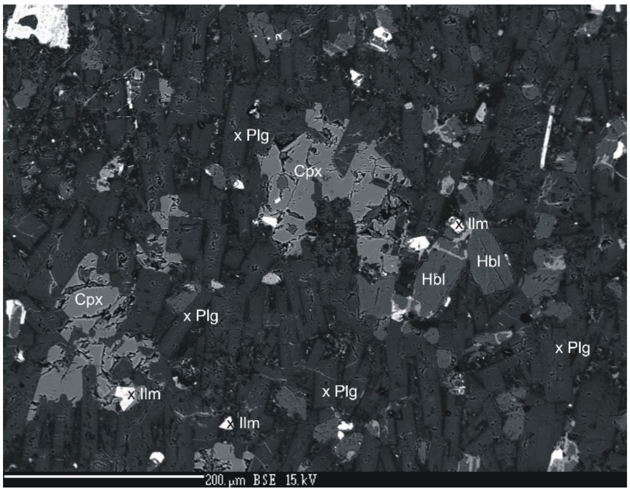

Figure 2. Back-scattered electron images of texture of the studied dolerites (Cpx – clinopyroxene, Plg – plagioclase, Hbl – hornblende, Ilm – ilmenite)

Clinopyroxene phenocrysts in the less altered part of the dolerites are generally subhedral to euhedral and may occur as discrete crystals as well as aggregates. Cracks and fractures in clinopyroxenes may be filled with secondary minerals, mainly chlorite, fibrous actinolite and hydrated garnets. Based on microprobe analyses (Table 1) and following the IMA classification (Morimoto et al., 1988), clinopyroxenes correspond to augite (Fig. 3).

Amphiboles may be altered mainly to chlorite and fibrous actinolite. Plagioclases are dominant phenocrysts (Fig. 2) in the studied doleritic rocks. They form idiomorphic prismatic crystals in the doleritic texture. The chemical composition of plagioclases corresponds to An_{35} - An_{59} (Vozár et al., 2015).

The composition of clinopyroxenes (Cpx) enclosed in minerals (mainly amphiboles) is similar to the composition of Cpx occurring separately.

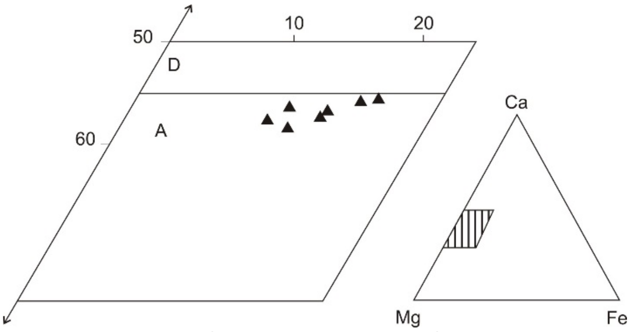

Figure 3. Classification diagram of clinopyroxenes (Morimoto et al. 1988), D-diopside, A – augite

As mentioned above, some clinopyroxenes were subjected to a specific type of alteration ultimately producing hydrated garnets (Fig. 4a, b). The increased contents of TiO_2 a K_2O in some chlorites point to the formation of these chlorites by biotite alteration (Table 2). Hydrated garnet occurs in irregular shapes prolonged in the direction of the original cleavage of clinopyroxene (Fig. 4a), and/or it forms irregular oval shapes with diffuse (frayed) rims. Hydrated garnets always associate with chlorite.

Locally, beside hydrated garnets, these oval shapes contain also titanite and sphalerite (Fig 5b). In spite of a rather inhomogeneous surface the composition of hydrated garnets is rather stable. The studied hydrated garnets have high Ca, Fe and Al contents, elevated Ti content and low Mg and Mn content (Table 3).

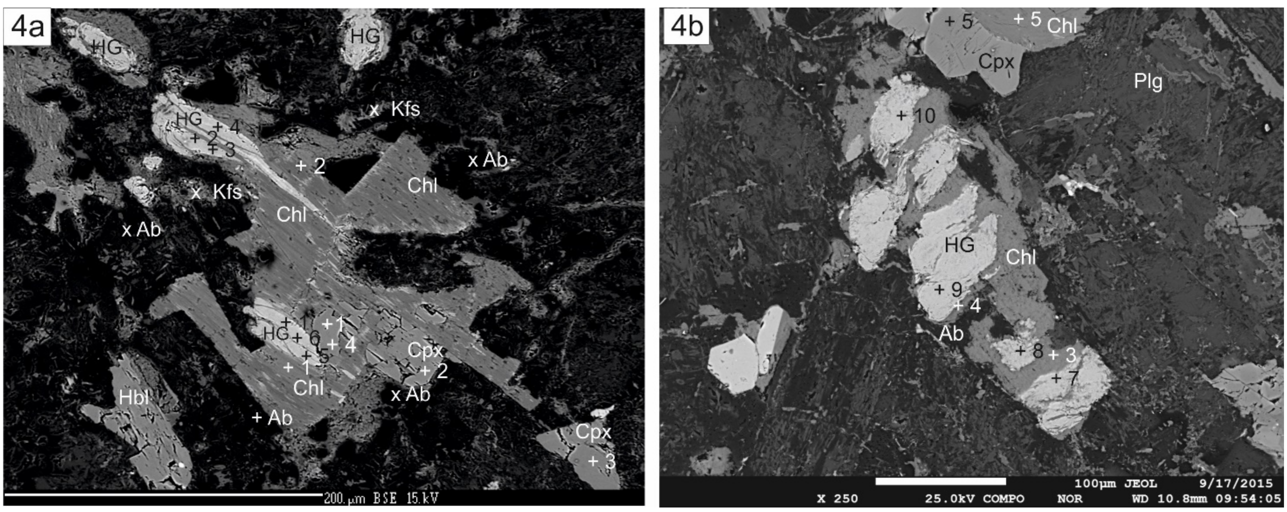

Figure 4a, b. Back-scattered electron images of altered clinopyroxenes (HG – hydrated garnet, Cpx – clinopyroxene, Ab – albite, Kfs – K-feldspar, Hbl – hornblende, Chl – chlorite); the numbers in figures correspond to those in Tables.

Table 1. Selected analyses of clinopyroxene

N. anal.	1	2	3	4	5	6	7
SiO_2	51.58	52.10	51.98	52.73	51.03	51.13	50.02
TiO_2	0.92	0.14	0.39	0.14	1.09	1.18	1.11
Al_2O_3	2.09	0.57	0.95	0.53	2.08	2.13	2.49
Cr_2O_3	0.22	0.00	0.05	0.00	0.00	0.10	0.63
FeO_{tot}	9.10	11.62	10.68	11.83	9.34	10.26	11.06
MnO	0.26	0.28	0.31	0.39	0.26	0.33	0.34
MgO	15.25	12.54	13.76	12.36	14.61	14.98	13.61
CaO	20.11	21.22	20.43	21.34	20.22	19.54	19.57
Na_2O	0.33	0.31	0.32	0.31	0.26	0.28	0.29
K_2O	0.00	0.01	0.00	0.00	0.00	0.00	0.00
Total	99.85	98.79	98.88	99.62	98.89	99.93	99.12
Formula based on 6 oxygens							
Si	1.916	1.987	1.968	1.998	1.921	1.906	1.894
Al^{IV}	0.084	0.013	0.032	0.002	0.079	0.094	0.106
Al^{VI}	0.008	0.013	0.010	0.021	0.013	0.000	0.005
Ti	0.026	0.004	0.011	0.004	0.031	0.033	0.032
Cr	0.006	0.000	0.001	0.000	0.000	0.003	0.019
Fe^{3+}	0.042	0.015	0.021	0.000	0.024	0.045	0.040
Fe^{2+}	0.241	0.356	0.317	0.375	0.270	0.275	0.310
Mn	0.008	0.009	0.010	0.013	0.008	0.010	0.011
Mg	0.845	0.713	0.777	0.698	0.820	0.833	0.768
Ca	0.801	0.867	0.829	0.866	0.815	0.781	0.794
Na	0.024	0.023	0.023	0.023	0.019	0.020	0.021
K	0.000	0.000	0.000	0.000	0.000	0.000	0.000
FeO_{tot} - total Fe as FeO,							

Low sums in microprobe analyses and anomalous optical properties (anisotropic character) point to elevated OH content in the structure of garnets. The presence of OH was also confirmed in Raman spectroscopy (Fig. 6) by stretching vibrations at 3600 cm^{-1}.

For the calculation of structural formulae and end members, we used Locock´s Excel spreadsheets (2008). Water content was estimated to be equal to the amount compensating for the deficiency of silica in calculations to 5 cations (Table 3). The calculated water content varies from 0.5 to 1.16 wt. %.

Table 2. Selected analyses of chlorites

N. anal.	1	2	3	4	5
SiO_2	30.83	28.91	29.11	30.04	30.07
TiO_2	3.20	0.74	0.04	0.04	0.06
Al_2O_3	14.66	16.55	14.07	15.66	14.39
Cr_2O_3	0.03	0.01	0.00	0.00	0.00
FeO_{tot}	21.85	23.41	24.70	25.87	25.41
MnO	0.09	0.18	0.22	0.19	0.21
MgO	15.13	16.90	16.04	17.39	17.18
CaO	1.55	0.30	0.14	0.32	0.19
Na_2O	0.03	0.01	0.01	0.01	0.01
K_2O	2.09	0.27	0.00	0.00	0.00
Total	89.45	87.27	84.33	89.52	87.52
Formula based on 28 oxygens					
Si	6.234	6.030	6.428	6.222	6.380
Al^{IV}	1.766	1.970	1.572	1.778	0.009
Al^{VI}	1.784	2.118	2.088	2.045	1.620
Ti	0.487	0.116	0.009	0.006	1.978
Fe^{3+}	0.317	0.180	0.000	0.000	0.000
Fe^{2+}	3.379	3.904	4.553	4.474	4.502
Mn	0.015	0.032	0.040	0.030	0.040
Mg	4.561	5.255	5.276	5.367	5.429
Ca	0.336	0.067	0.033	0.072	0.042
Na	0.024	0.008	0.000	0.000	0.000
K	1.078	0.147	0.000	0.000	0.000
FeO_{tot} - total Fe as FeO					

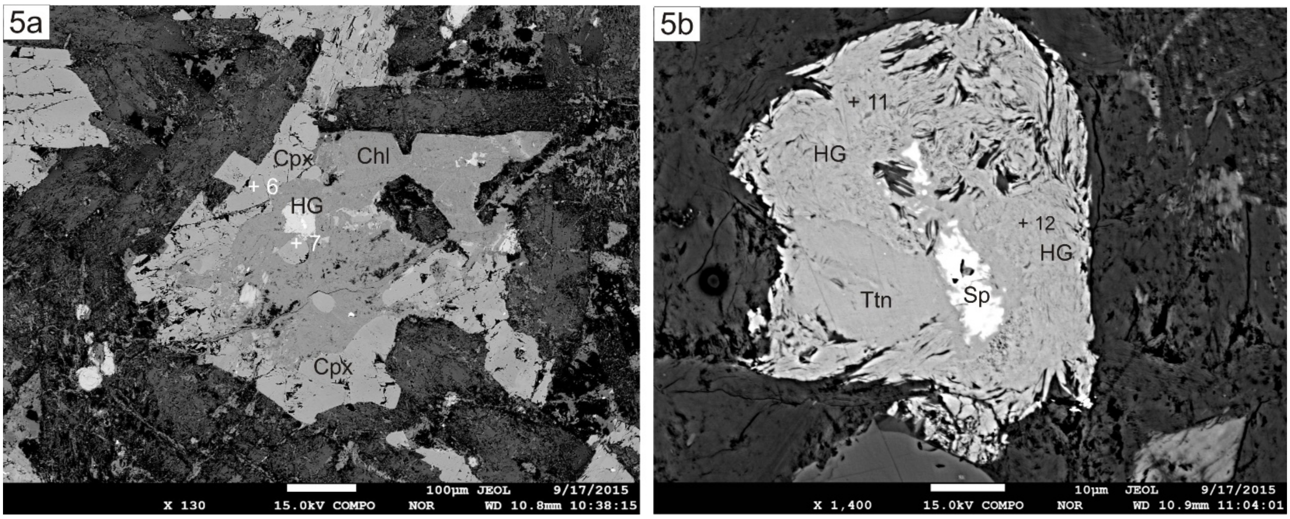

Figure 5a, b. Back-scattered electron images of altered clinopyroxenes (HG – hydrated garnet, Cpx – clinopyroxene, Chl – chlorite, Ttn – titanite, Sp – sphalerite), the numbers in figures correspond to those in Table 1

Table 3. Selected analyses of hydrogarnets

N. anal.	1	2	3	4	5	6	7	8	9	10	11	12
SiO_2	35.36	35.94	35.67	35.93	35.80	35.91	36.17	36.07	36.19	36.06	36.18	36.10
TiO_2	1.13	0.62	0.72	0.81	0.78	0.63	0.61	0.50	0.81	0.69	0.84	0.39
Al_2O_3	9.30	9.84	9.96	10.12	9.79	9.03	9.88	9.89	9.78	9.74	9.43	9.78
Cr_2O_3	0.00	0.01	0.00	0.01	0.02	0.00	0.00	0.01	0.00	0.03	0.00	0.04
FeO^+	15.59	15.29	15.13	14.49	14.79	16.08	15.97	16.45	15.39	16.23	15.56	16.34
MnO	0.14	0.09	0.14	0.14	0.16	0.14	0.13	0.13	0.22	0.15	0.22	0.13
MgO	0.00	0.18	0.11	0.16	0.04	0.03	0.35	1.00	0.22	0.27	0.01	0.04
CaO	34.94	34.29	34.70	33.92	34.82	34.67	33.91	32.29	33.89	34.17	34.59	34.99
Na_2O	0.00	0.01	0.00	0.02	0.01	0.00	0.00	0.00	0.02	0.01	0.00	0.02
Total	96.47	96.27	96.43	95.61	96.20	96.50	97.02	96.34	96.53	97.35	96.84	97.82
Recalculated (Wt %)												
FeO	0.70	0.54	0.49	0.77	0.23	0.27	1.19	1.15	1.18	0.38	1.33	0.00
Fe_2O_3	16.55	16.39	16.26	15.25	16.18	17.57	16.42	17.01	15.80	17.62	15.82	18.16
H_2O+	1.16	0.70	0.99	0.53	0.80	0.69	0.80	0.43	0.65	0.55	0.95	0.56
Formula based on 12 O												
Si	2.837	2.902	2.862	2.926	2.888	2.904	2.896	2.921	2.920	2.892	2.900	2.881
Al	0.879	0.936	0.941	0.972	0.930	0.860	0.932	0.944	0.930	0.921	0.890	0.919
Ti	0.068	0.038	0.044	0.050	0.047	0.039	0.037	0.030	0.049	0.042	0.051	0.024
Fe^{3+}	0.999	0.996	0.982	0.935	0.982	1.069	0.989	1.037	0.959	1.064	0.954	1.091
Fe^{2+}	0.047	0.036	0.033	0.052	0.016	0.018	0.079	0.077	0.079	0.025	0.089	0.000
Mn	0.010	0.006	0.010	0.010	0.011	0.010	0.009	0.009	0.015	0.000	0.015	0.009
Mg	0.000	0.021	0.008	0.020	0.005	0.004	0.042	0.120	0.026	0.032	0.001	0.005
Ca	3.004	2.966	2.983	2.960	3.010	3.003	2.908	2.802	2.930	2.937	2.971	2.992
Na	0.000	0.002	0.000	0.003	0.001	0.000	0.000	0.001	0.003	0.002	0.001	0.003
End members (Locock 2008)												
Katoite	5.17	3.14	4.41	2.4	3.59	3.1	3.56	1.94	2.92	2.45	4.23	2.34
Morimotoite	4.69	3.28	3.31	4.49	1.55	1.84	3.68	0.92	4.93	0.6	5.08	0
Spessartine	0.32	0.21	0.32	0.33	0.36	0.33	0.29	0.3	0.49	0.34	0.51	0
Grossular	38.09	42.42	41.81	44.85	42.39	39.44	40.07	37.65	41.64	39.83	39.73	41.28
Andradite	49.97	49.8	49.09	46.72	49.12	53.44	49.47	51.83	47.95	53.18	47.7	53.81
Remainder	1.76	1.15	1.06	1.21	2.99	1.85	2.93	7.36	2.07	3.6	2.75	2.57

Following recently valid garnet classification (Grew et al., 2013), the studied garnets belong to the kenogarnets group; the content of katoite end-member varies from 2.3 to 5.1. Dominant end members are grossular (from 37.6 to 44.9) and andradite (from 46.7 to 53.4). Elevated Ti content is manifested by the presence of morimotoite end-member (from 0.6 to 4.7). According to this composition, garnet corresponds to Fe^{3+} analog of katoite. Such types of garnets have not been reported in the literature so far. The highest in the literature reported content of andradite in katoite does not exceed 35% (Armbruster, 1995), whereas in the studied garnets it attains up to 50%.

Hydrated garnets have been rare reported from metasomatically altered basaltoid rocks (Baker & Black. 1980, Buse et al., 2010, Laverne 2006), sometimes also from rodingites (Hovorka et al., 1985, Li et al., 2004, Dubinska et al., 2004, El-Shazly & El-Belushi 2004, Amthauer & Rossman 1998).

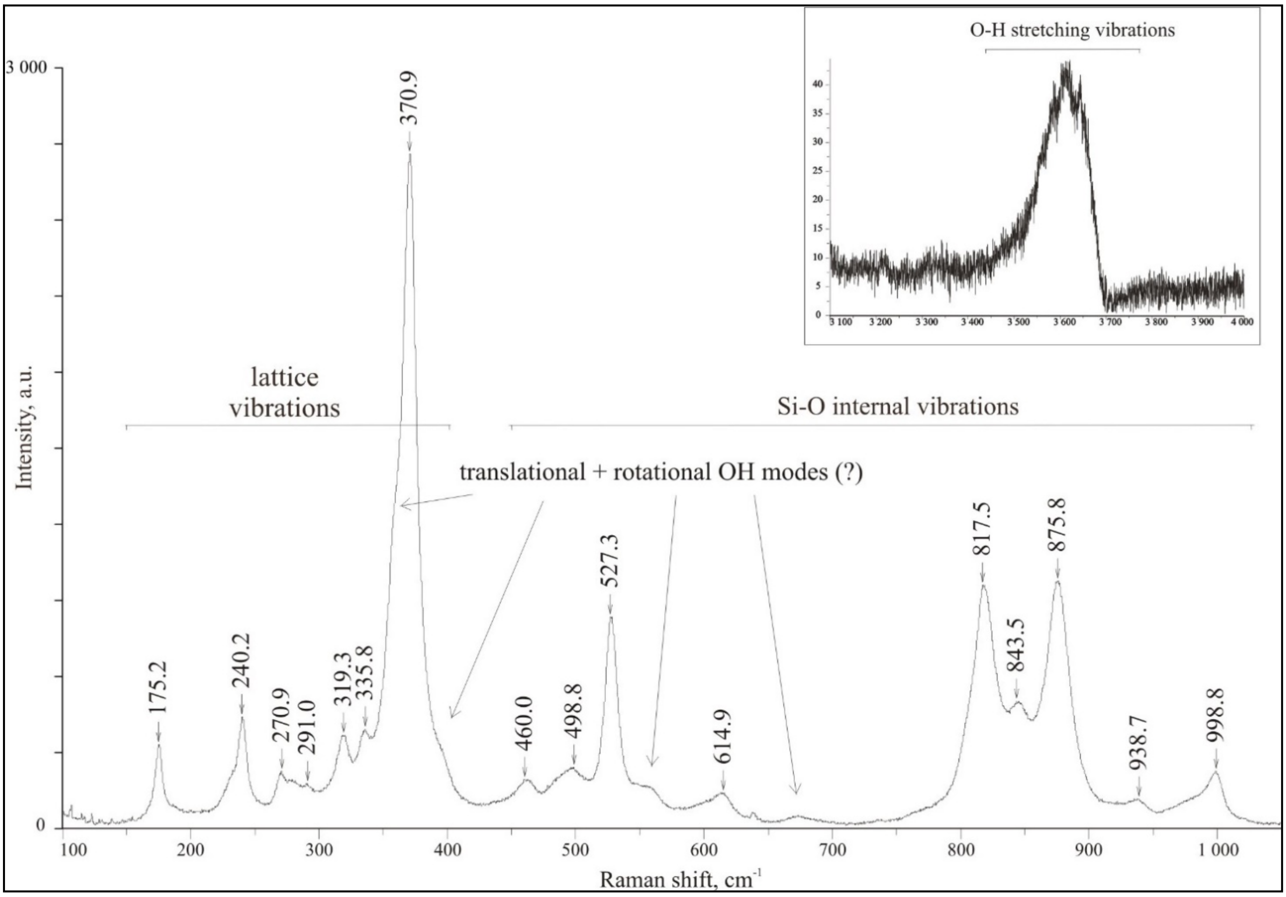

Figure 6. Raman spectra of hydrated garnets

5. CONCLUSION

In the dykes of Permian basalts specific type of postmagmatic alterations was detected. The origin of hydrated garnet is likely to represent the postmagmatic reaction in which clinopyroxene was altered to an assemblage of hydrated garnet and chlorite. The metasomatic alteration is likely to have been of low temperature character.

The composition of hydrogarnet corresponds to Fe^{3+} rich katoite. The studied hydrated garnets are characterized by high Ca, Fe and Al contents, low to zero Mn and Mg contents and elevated Ti contents.

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