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LETTER

A natural scandian garnet

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ABSTRACT

Garnet from an aposkarn achtarandite-bearing rodingite-like rock in Sakha-Yakutia, Russia, has a Sc content close to 6 wt% Sc_2O_3 (~0.45 apfu). The scandian garnet is a relict mineral from a high-temperature, shallow-level melilite skarn. Structural and electron microprobe data for a crystal of the scandian garnet with cell parameter a = 12.331(1) Å, $Ia\bar{3}d$ allows refinement of the structural formula $(Ca_{2.97}Mg_{0.02}Y_{0.01})_{\sum 3}(Fe^{3+}_{0.663}Zr_{0.584}Ti^{4+}_{0.294}Sc_{0.153}Cr_{0.152}Mg_{0.094}Fe^{2+}_{0.04}Hf_{0.008}V_{0.003})_{\sum 2}(Si_{1.898}Al_{0.420}Ti^{4+}_{0.359}Fe^{3+}_{0.323})_{\sum 3}O_{12}$. Investigation of the composition of many of the scandian garnets reveals the existence of a solid-solution between kimzeyite-schorlomite $Ca_3(Zr,Ti)_2(Al,Fe)_2SiO_{12}$ and the scandium analog of andradite $Ca_3Sc_2Si_3O_{12}$. This is the first report of a natural scandian garnet.

INTRODUCTION

Unlike synthetic analogues, natural garnets (general crystal-chemical formula $X_3Y_2Z_3O_{12}$) are characterized by a limited number of mineral-forming cations: X = Ca, Mg, Mn, Fe; Y = Al, Fe, Cr, Ti, Zr, Mn, V; Z = Si, which can be partially substituted by Fe and Al (Mandarino and Back 2004).

Achtarandite-bearing, rodingite-like aposkarn rocks from the Wiluy River, Sakha-Yakutia (Russia), contain Ti-Zr garnets with scandium contents above 6 wt% Sc_2O_3, and zirconium contents approaching 25–30 wt% ZrO_2. Up to now, maximum Sc_2O_3 contents ~0.3 wt% Sc_2O_3 have been reported for spessartine from a pegmatite with scandium mineralization in Norway (Raade et al. 2002). Kimzeyite was first described by Milton et al. (1961) in carbonatites at Magnet Cove, Arkansas, USA. Subsequently, kimzeyite with a high zirconium content (ZrO_2 > 25 wt%) was discovered in volcanic rocks from both Anguillara Sabazia and Stromboli, Italy (Munno et al. 1980, Schingaro et al. 2001), and in skarns from Chihuahua, Mexico (Mauger 2001).

There are no scandium ore deposits and only 10 scandium minerals are known even though the crustal abundance of scandium is 21.9 ppm (Rudnick and Gao 2003). This reflects the wide dispersion of Sc in rock-forming and accessory minerals (Mellini et al. 1982; Orlandi et al. 1998; Bernhard et al. 1998; Liferovich et al. 1998; Raade et al. 2002, 2004; Moëlo et al. 2002; Gramaccioli et al. 2004). Scandium, with a 3^+ charge and an ionic radius of 0.745 Å (6-coordination), can substitute in various ways for different cations: Mg, Al, $Fe^{2+,3+}$, Ti^{4+}, Mn^{2+}, Zn^{2+}, Zr^{4+}, Sn^{4+}, Ta^{5+}, Nb^{5+}, Y^{3+}, etc. (Shannon 1976; Raade et al. 2002).

Investigations on synthetic scandian pyrope and grossular show that scandium occupies octahedral positions in grossular whereas it occupies the eightfold coordinated position in pyrope (Quartieri et al. 2004). Preliminary results on the solid-solution series between andradite $Ca_3Fe^{3+}_2Si_3O_{12}$ and its scandium analog $Ca_3Sc_2Si_3O_{12}$ (Ito and Frondel 1968) suggest little deviation from ideal mixing behavior (Woodland and Angel 1996).

In this letter, we provide data on the morphology, composition, and structure of a scandian garnet from Sakha-Yakutia and, in addition, we discuss the genesis of the garnet and the source of the scandium.

ANALYTICAL METHODS

The garnet morphology and composition were investigated using a Philips/FEI XL30/EDAX scanning electron microscope and a CAMECA SX100 electron microprobe analyzer. Compositions were measured at 15 kV and 30 nA using natural and synthetic standards. X-ray maps were recordered at 15 kV, 40 nA. Structural data for a single crystal of scandian garnet were collected using an Enraf-Nonius CAD4 diffractometer (exposure time = 120 s). Cell dimensions were determined from reflections > 23.56° theta. The Raman spectra of the scandian garnet were recorded using a Raman microprobe T-64000 (Jobin-Yvone) with a 514.5 nm Ar ion laser. The absence of hydroxyl groups was confirmed by Raman microprobe and FTIR (BioRad, KBr) investigations.

RESULTS AND DISCUSSION

The scandian garnets occur in unusual achtarandite rodingite-like rocks comprising vesuvianite–Si-deficient vesuvianite, grossular–hydrogrossular, chlorite, and serpentine (Fig. 1) with rare relict crystals of wiluite, Al-Ti-Fe^{3+}-diopside, and perovskite (Galuskin et al. 2003, Galuskin 2005). The Wiluy deposit is the type locality for grossular, wiluite, and also achtarandite—an unusual tetrahedral pseudomorph of hibschite (hydrogrossular) after wadalite (Lyakhovich 1954; Groat et al. 1998; Galuskina et

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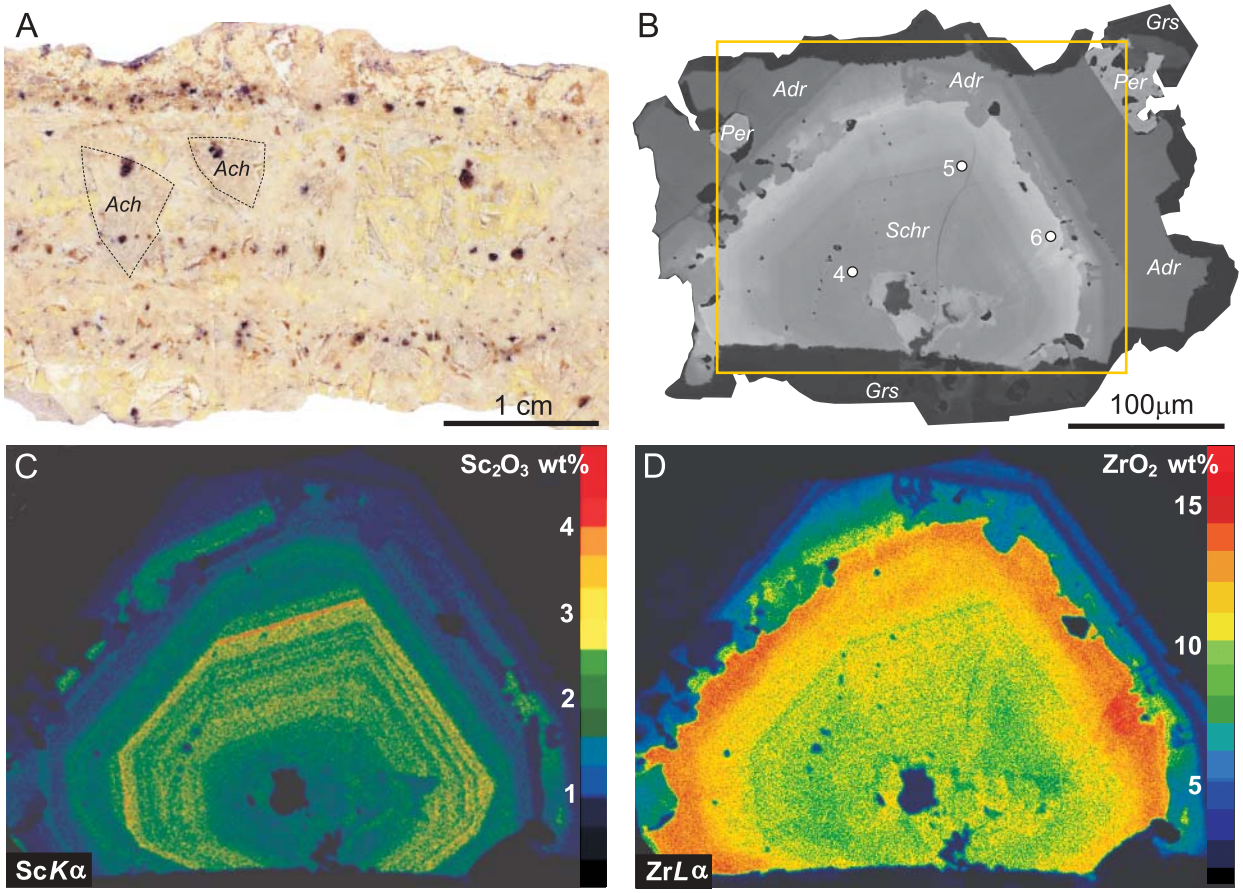

FIGURE 1. (**A**) Rodingite-like rock comprising plate-like pseudomorphs of vesuvianite–Si-deficient vesuvianite (yellow) and grossular–hydrogrossular (white) after melilite. The space between the pseudomorphs is filled with serpentine (pink) where shadowy achtarandite pseudomorphs (*Ach*) are visible. The distribution of the dark garnet crystals reflects the primary sedimentary layering of the protolith—high-temperature skarn later transformed into rodingite-like rock. (**B**) Back-scattered electron image of a garnet with a core of oscillatory-zoned scandian schorlomite showing the points of microprobe analyses and the framed area from which quantitative X-ray maps of Sc and Zr distribution were obtained (**C**, **D**). The distribution of scandium highlights the garnet oscillatory zoning. Per = perovskite, Adr = andradite, Schr = schorlomite, Grs = grossular.

al. 1998). The achtarandite serpentinite and rodingite-like rocks are products of the retrograde alteration of high-temperature shallow-level melilite skarn that had previously replaced gigantic (> 200 m) xenoliths of Ordovician sedimentary rock in a large Triassic gabbro-dolerite intrusion of the Siberian Trap Formation (Galuskina et al. 1998, 2001; Galuskin 2005).

On the classification diagram (Fig. 2), garnets from the rodingite-like rocks plot in the kimzeyite-schorlomite and ugrandite fields. Stain-like relicts of Ti-Zr garnet (<100 μm) in the central parts of 1–2 mm rombododecahedral grossular-andradite crystals are enriched in scandium. Rare garnet crystals with large fragments of kimzeyite-schorlomite show oscillatory zoning emphasized by the scandium distribution (Figs. 1B–D). Scandium contents rarely exceed 0.1 wt% Sc_2O_3 in the innermost core of kimzeyite with 25–30 wt% ZrO_2 (anal. 1, Table 1), whereas the next zone, zirconian schorlomite, contains more than 6 wt% Sc_2O_3 (anal. 2–5, Table 1). In relatively late zones of kimzeyite-schorlomite, Sc_2O_3 contents do not exceed 2–2.5 wt% (Figs. 1C, 1D, anal. 6, Table 1). These latter zones are followed by a Zr-Cr-Sc-andradite-schorlomite zone, in which scandium contents may occasionally reach 3 wt% Sc_2O_3 (anal. 7, Table 1). For the relic zones of Zr-Ti garnet, a positive correlation between Sc and Si and a negative correlation between Sc and Ti + Zr + Hf are noted (Figs. 2B and 2C).

In naming the garnets, only the prevalent cation in the octahedral position was considered (Table 1). Standard calculations based on electron microprobe analyses of garnet end-members result in a very complicated picture (Table 1). The $Ca_3Sc_2Si_3O_{12}$ component reaches 23.5% for a phase with 30.5% schorlomite, and 23.5%, 13%, and 6.5% for kimzeyite, morimotoite, and andradite, respectively (anal. 2, Table 1). The correct naming of a garnet with a similar $Ca_3Sc_2Si_3O_{12}$ content but with a higher silicon content (anal. 3, Table 1), comprising mainly schorlomite (25%), kimzeyite (18%), morimotoite (16%) and andradite (15%), is not straight forward. Calculation reveals that, in the analyses of schorlomite garnet with oscillatory zoning (Fig. 1B–D), the morimotoite or kimzeyite end-members predominate (anal. 4–6, Table 1).

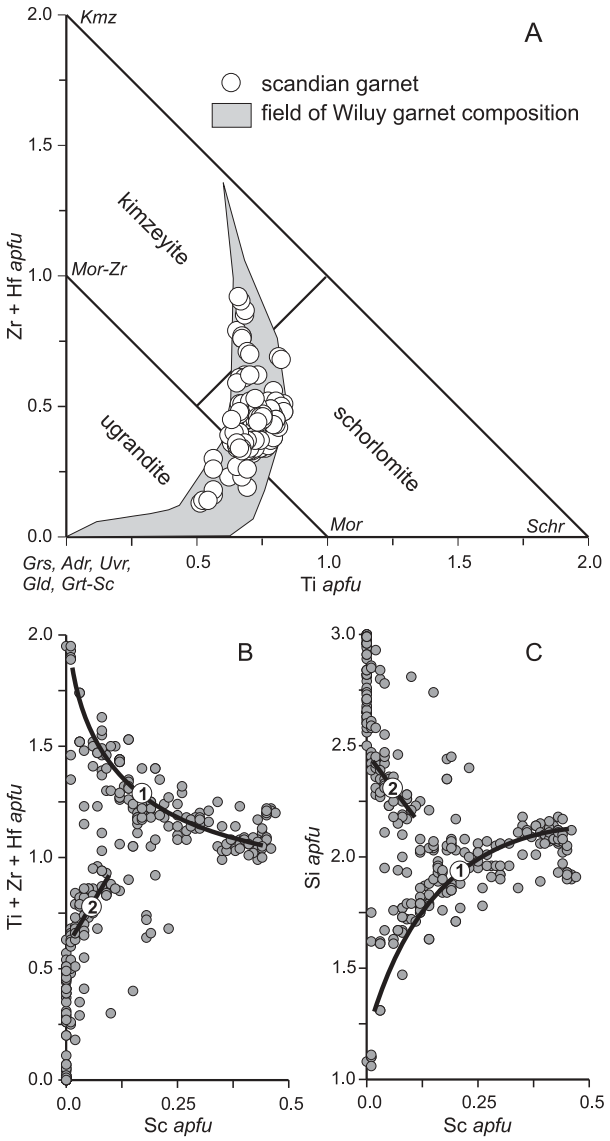

FIGURE 2. Analyses of garnet from Wiluy plotted on the classification diagram ugrandite (and related end-members)–schorlomite–kimzeyite (**A**) and on the diagrams Sc vs. Ti + Zr + Hf (**B**) and Sc vs. Si (**C**); apfu = atom per formula unit. 1 = trend of composition change of relict zones, 2 = trend of composition change of late, often two-phase, zones. For description of symbols see Table 1. Mor-Zr = hypothetical end-member zirconium morimotoite $Ca_3Zr(Mg, Fe^{2+})Si_3O_{12}$.

TABLE 1. Composition of scandian garnet from rodingite-like rocks, Sakha-Yakutia, Russia

	1	2	3	4	5	6	7
SiO_2 wt%	11.16	21.50	24.13	23.36	23.26	18.35	27.88
TiO_2	8.45	11.09	10.10	11.59	11.00	11.88	8.28
ZrO_2	26.98	10.64	8.57	10.47	10.26	16.93	3.21
HfO_2	0.66	0.18	0.20	0.23	0.28	0.33	0.11
Y_2O_3	<0.1	0.24	0.12	0.39	0.34	0.34	0.19
Al_2O_3	6.95	3.40	3.47	4.94	4.08	5.77	2.67
V_2O_3	0.02	0.04	0.11	0.17	0.11	0.07	0.04
Sc_2O_3	0.08	6.03	6.01	2.02	3.75	1.02	3.00
Cr_2O_3	n.d.	0.87	0.69	0.36	0.35	0.16	2.51
Fe_2O_3	15.14	12.88	12.38	11.28	12.02	11.22	17.94
FeO	0.22	0.49	0.75	1.60	1.36	1.79	1.07
MnO	0.01	n.d.	0.05	n.d.	0.01	0.03	n.d.
CaO	28.38	31.30	31.38	30.96	30.99	30.08	31.80
MgO	0.18	0.76	1.13	1.83	1.53	1.27	0.49
Total	98.23	99.42	99.09	99.20	99.34	99.24	99.19
	calculated on 8 cations and 12O						
Si apfu	1.1	1.91	2.12	2.05	2.05	1.67	2.40
Al	0.8	0.36	0.36	0.51	0.42	0.62	0.14
Fe^{3+}	1.1	0.73	0.52	0.44	0.53	0.71	0.46
Z site	3	3	3	3	3	3	3
Ti^{4+}	0.62	0.74	0.66	0.76	0.73	0.81	0.54
Zr	1.29	0.47	0.36	0.45	0.44	0.75	0.14
Hf	0.02			0.01	0.01	0.01	
Sc	0.01	0.47	0.46	0.15	0.29	0.08	0.23
Fe^{3+}	0.03	0.13	0.30	0.30	0.27	0.06	0.70
Al							0.13
V^{3+}			0.01	0.01	0.01	0.01	
Cr		0.06	0.05	0.02	0.02	0.01	0.17
Mg	0.02	0.09	0.11	0.17	0.14	0.13	0.01
Fe^{2+}	0.01	0.04	0.05	0.12	0.10	0.14	0.08
Y site	2	2	2	2	2	2	2
Ca	2.99	2.98	2.95	2.91	2.92	2.94	2.94
Y		0.01	0.01	0.02	0.02	0.02	0.01
Mg	0.01	0.01	0.04	0.07	0.06	0.04	0.05
X site	3	3	3	3	3	3	3
	end-member of garnet						
Adr	1.5	6.5	15	15.5	13.5	3	35
Grs							6.5
Uwr		3	2.5	1	1	0.5	8.5
Gld			0.5	0.5	0.5	0.5	
Grt-Sc	0.5	23.5	23	7.5	14.5	4	11.5
Kmz	39	18	18	22.5	21	30.5	7
Kmz-Fe	25.5	5.5			1	7.5	
Grt-Hf	1			0.5	0.5	0.5	
Schr	29.5	30.5	25	20.5	24	27	22.5
Schr-Al				3			
Mor	1	4	5	12	10	14	8
Mor-Mg	2	9	11	17	14	13	1

Notes: 1 = kimzeyite, 2–6 = scandian schorlomite (Fig. 1B), 7 = scandian andradite. Adr = andradite $Ca_3Fe^{3+}_2Si_3O_{12}$, Grs = grossular $Ca_3Al_2Si_3O_{12}$, Uwr = uvarovite $Ca_3Cr_2Si_3O_{12}$, Gld = goldmanite $Ca_3V^{3+}_2Si_3O_{12}$, Grt-Sc = scandium garnet $Ca_3Sc_2Si_3O_{12}$, Kmz = kimzeyite $Ca_3Zr_2Al_2SiO_{12}$, Kmz-Fe = kimzeyite-Fe^{3+} $Ca_3Zr_2Fe^{3+}_2SiO_{12}$, Grt-Hf = hafnium garnet $Ca_3Hf_2Al_2SiO_{12}$, Schr = schorlomite $Ca_3Ti^{4+}_2Fe^{3+}_2SiO_{12}$, Schr-Al = schorlomite-Al $Ca_3Ti^{4+}_2Al_2SiO_{12}$, Mor = morimotoite $Ca_3Fe^{2+}Ti^{4+}Si_3O_{12}$, Mor-Mg = morimotoite-Mg $Ca_3Mg^{2+}Ti^{4+}Si_3O_{12}$.
* FeO/Fe_2O_3 calculated on charge balance, n.d. = not detected.

A single-crystal Raman spectrum of kimzeyite from the rodingite-like rocks has a characteristic set of lines of which three strong lines near 730 cm^{-1} [$(Z\text{-}O)_{str.}$, A_1g], 500 cm^{-1} [$(Z\text{-}O)_{bend}$, A_1g], 300 cm^{-1} [$R(ZO_4)$, A_1g] are diagnostic for kimzeyite (Fig. 3, Schingaro et al. 2001). In the scandian schorlomite spectra, the lines determined by vibrations $R(ZO_4)$ and $(Z\text{-}O)_{bend}$ shift toward higher frequencies of 340–350 cm^{-1} and 507–510 cm^{-1} respectively (Fig. 3). The $(Z\text{-}O)_{str.}$ lines with frequencies above 700 cm^{-1} are weak and broadened in the spectra of kimzeyite-schorlomite compared with the analogous lines in the single-crystal Raman spectra of pyralspite and ugrandite garnets (Fig. 3, Kolesov and Geiger 1998; Schingaro et al. 2001). This can be explained by the incorporation of Al, Si, Fe^{3+}, and/or Ti^{4+} in *Z*-tetrahedra. For example, the line near 735 cm^{-1} probably corresponds to $(Fe^{3+}O_4)$ vibrations and the line near 950 cm^{-1} to (SiO_4) vibrations (Schingaro et al. 2001). The weak line near 640 cm^{-1} appears on the single-crystal Raman spectra of those garnets with high contents, 0.2–0.45 apfu, of scandium (Fig. 3). Note that coordination effects connected with different types of cations in the *Y*-site will influence the character of lines determined by *Z*-tetrahedra vibrations.

Preliminary structural data for an inhomogeneous grain (0.1

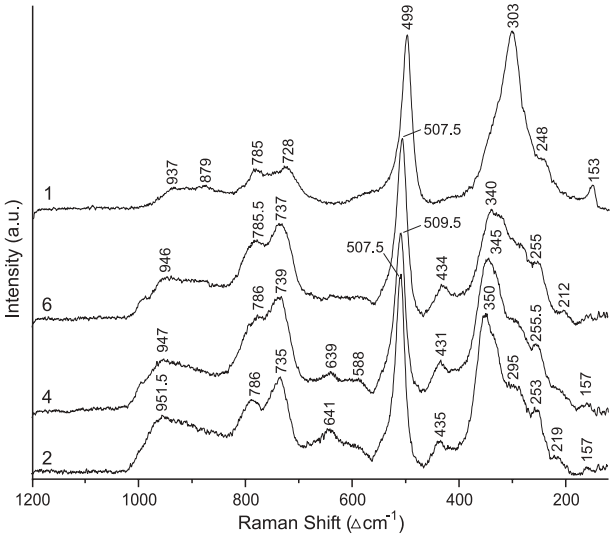

FIGURE 3. Single-crystal non-polarized Raman spectra of kimzeyite and scandian garnet. Numbers of probe analyses are the same as in the Table 1: 1 = kimzeyite, 2, 4, 6 = scandian schorlomite.

× 0.08 × 0.05 mm) of zirconian schorlomite (supplementary data: Tables 2 and 3)[1] with a mean Sc_2O_3 content of 2 wt%, and a = 12.331(1) Å, shows no deviation from the standard garnet symmetry $Ia\bar{3}d$. The data also suggest that Ti^{4+} partly possesses tetrahedral coordination: $(Ca_{2.97}Mg_{0.02}Y_{0.01})_{\Sigma 3}(Fe^{3+}_{0.663}Zr_{0.584}Ti^{4+}_{0.294}Sc_{0.153}Cr_{0.152}Mg_{0.094}Fe^{2+}_{0.049}Hf_{0.008}V_{0.003})_{\Sigma 2}(Si_{1.898}Al_{0.420}Ti^{4+}_{0.359}Fe^{3+}_{0.323})_{\Sigma 3}O_{12}$.

Similar ionic radii enable incorporation of Zr^{4+}, Fe^{2+}, Mg^{2+}, and Sc^{3+} in the octahedral site in the garnet structure. Varying combinations of cations, of different charge and size in the octahedral and tetrahedral positions, influence the structural stability of this complex garnet, for example: $^{[VI]}Sc^{3+[IV]}Si^{4+} \leftrightarrow {}^{[VI]}Zr^{4+[IV]}Al^{3+}$, $^{[VI]}Zr^{4+[IV]}Fe^{3+}$, $^{[VI]}Ti^{4+[IV]}Fe^{3+}$; $^{[VI]}Sc^{3+[VI]}Fe^{3+} \leftrightarrow {}^{[VI]}(Fe^{2+}, Mg)^{[VI]}Ti^{4+}$, etc.

The scandian garnets described here belong to a new type of skarn mineralization. Known silicate scandium minerals (thortveitite, bazzite, jervisite, cascandite, scandiobabingtonite, kristiansenite, scandiomilarite) and the oxide "scandioixiolite" are found only in pegmatites (Mellini et al. 1982; Orlandi et al. 1998; Bergstøl and Juve 1988; Černy et al. 2000; Raade et al. 2002, 2004; Gramaccioli et al. 2004). Scandium phosphates (kolbeckite, juonnite, pretulite) crystallize under low-temperature conditions (Bernhard et al. 1998, 2001; Liferovich et al. 1998). Recently, pretulite growing on scandian zircon and xenotime was discovered in an apatite-rich oolitic Ordovician ironstone from Saint-Aubin-des-Châteaux in France (Moëlo et al. 2002). The change of mineral composition points to the existence of a complete solid-solution between zircon and pretulite (Moëlo et al. 2002). One further type of scandium mineralization, occurring in carbonaceous chondrites, is represented by a high-titanian, Zr-free, diopside-like phase with a Sc_2O_3 content up to 16.4 wt% and a structure that is not yet investigated (Lin et al. 2003). Experiments with chondrite melt (Draper et al. 2003) have shown that the silicate phase (garnet) is enriched in scandium, whereas zirconium concentrates in the melt.

The scandian garnets from Yakutia formed at high-temperature (>800 ºC) and low pressure during isochemical skarn development in which fluids only played a minor role (Galuskina et al. 1998; Galuskin 2005). The garnet distribution in the rock reflects a primary sedimentary layering (Fig. 1A), which suggests detrital zircon and Ti-Nb-Ta oxides were the initial sources of Sc. Early crystallizing garnets were enriched in Sc before widespread Sc dispersion occurred during the subsequent formation of the main Ca-Mg-minerals.

In conclusion, a solid solution between kimzeyte-schorlomite $Ca_3(Zr,Ti)_2(Al,Fe)_2SiO_{12}$ and the scandium analog of andradite $Ca_3Sc_2Si_3O_{12}$ exists in nature. Our investigations indicate that the synthesis of new garnet solid solutions involving $Zr^{4+}Al^{3+} \leftrightarrow Sc^{3+}Si^{4+}$ isomorphism could lead to new technical and gemstone materials.

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[1] Deposit item AM-05-023, Tables 2 and 3. Deposit items are available two ways: For a paper copy contact the Business Office of the Mineralogical Society of America (see inside front cover of recent issue) for price information. For an electronic copy, visit the MSA web site at http://www.minsocam.org, go to the American Mineralogist Contents, find the table of contents for the specific volume/issue wanted, and then click on the deposit link under the article's title.

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