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TYPOMORPHIC SPECIFICS OF ZONAL GARNETS IN CALCAREOUS SKARNS

(Figs. 9, Tabs. 2)

Abstract: The results of a microprobe analysis of the chemical zonation of calcic skarn garnets from different ore skarn deposits of the U.S.S.R. and Mongolia have been systematized. Three types of zonation depending on isomorphism have been revealed: 1) Al^{3+} and Fe^{3+} (in grossular-andradite); 2) $Ca \rightleftharpoons Mn$ and $Ca \rightleftharpoons Fe^{2+}$ (in grossular garnets with almandine-spessartite content > 10 mol. %); 3) $Mg \rightleftharpoons Fe^{2+}$ and $Al^{3+} \rightleftharpoons Fe^{3+}$ in the garnets of apomagnesian calcic skarn. Possible causes of different types of the zonal garnet formation are being discussed: i. e. migration of mineral-forming components, alteration in the temperature, acidity-basicity, oxidizing-reducing conditions, etc.

Резюме: Систематизированы результаты микрозондового изучения химической зональности гранатов известковых скарнов на примере различных скарново-рудных месторождений Советского Союза и МНР. Выделены три типа зональности, обусловленной изоморфизмом: 1) Al^{3+} и Fe^{3+} (в гроссуляр-андрадитах); 2) $Ca \rightleftharpoons Mn$ и $Ca \rightleftharpoons Fe^{2+}$ (в гроссуляровых гранатах, с содержанием альмандина + спессартина > 10 мол. %); 3) $Mg \rightleftharpoons Fe^{2+}$ и $Al^{3+} \rightleftharpoons Fe^{3+}$ в гранатах апомагнезиальных известковых скарнов. Обсуждаются возможные причины образования различных типов зональных гранатов: миграционная способность минералообразующих компонентов, изменение температуры, кислотности-основности, окислительно-восстановительных условий и др.

We have studied zoning in garnets of certain skarn-ore deposits in the U.S.S.R. and Mongolia by microprobe analyzer. As a result we have identified: 1) different types of zoning and considered the possible causes behind its formation; 2) specified and partly revised the earlier concepts concerning typomorphism in garnets of skarn deposits.

Types of zoning

An analysis of accumulated data enables us to identify in garnets of calcareous skarns the primary zonation which had arisen in garnets of the earlier generations of both the calcareous skarn proper and their apomagnesial varieties and the secondary zonation which usually appears at the stage of acid leaching and ore mineralization.

According to the distribution specifics of the contents of the constituents in skarns garnet crystals there are distinguished at least three types of zoning (Tab. 1). In each of these types there are, moreover, discerned the normal

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Table 1

Types of zoning in skarn crystals

Zoning: kind \ type		Primary I	Primary II	Primary III	Secondary I	Secondary II	Secondary III
Distribution of the contents of principal components from the centre of the crystal towards its margin	normal (direct)	Increasing: Fe_2O_3	FeO, MnO	—	Fe_2O_3	—	—
		Decreasing: Al_2O_3	CaO	—	Al_2O_3	—	—
	reverse	Increasing: Al_2O_3	CaO	CaO, Fe_2O_3	not observed		
		Decreasing: Fe_2O_3	FeO, MnO	MgO, Al_2O_3			

(direct) and reverse zoning. The normal zoning is characterized by a higher iron content (andratite, almandine content) and corresponds to the natural progress of the skarn-forming process and the reverse one, by a respective decrease of the iron content or an increase of the alumina content (grossular content) from the core towards rim zones of garnet crystals.

First-type zoning, the particularly widespread one in the skarn garnets of the grossular-andradite series, is determined by a variation of the andradite and grossular contents within the garnet crystal. The pyralspite constituents are contained in garnet in an insignificant quantity (not more than 10 mol. %) and fail to have any effect on zonation.

Regarding the distribution of the Fe^{+3} content in garnet the zoning can be both direct and reverse. The zoning of the first type is particularly characteristic of the garnets of the calcareous skarns in the magnetite, copper, and polymetallic deposits; it is less abundant in the garnets of the scheelite and other skarn-rare metal deposits. Fig. 1a shows the compositions of the garnets with the first-type zoning from the skarn-iron ore deposits Auerbakh, Dashkesan (U.S.S.R.) and Tomur-Te (Mongolia). Arrows in the figures indicate the variability trends of the garnet composition from the core towards the rim of the garnet; isolated points are the garnet composition calculated from the total chemical analyses.

The diagram (Fig. 1b) shows the compositions of the zonal garnets of the first type from calcareous skarns with molybdenite-scheelite mineralization (Chorukh-Dairon, Tyrnyauz,), from the Ak-Dzhilga rare-metal deposit and the Beise deposit (Mongolia). Here are also given garnets from the sulphide-tungsten-molybdenum deposits in the Altai-Sayan area (using total chemical analyses borrowed from Индукаев, 1980). As seen in the diagram, at the

Altai-Sayan deposits these are garnets with the first-type zoning. Zonal garnets from calcareous skarns in the Strawberry deposit, Central Sierra Nevada (U.S.A.), as noted by Nockleberg (1981), and from the King Island deposit (Australia), as noted by Kwak (1978), revealed a reverse zonation of the first type.

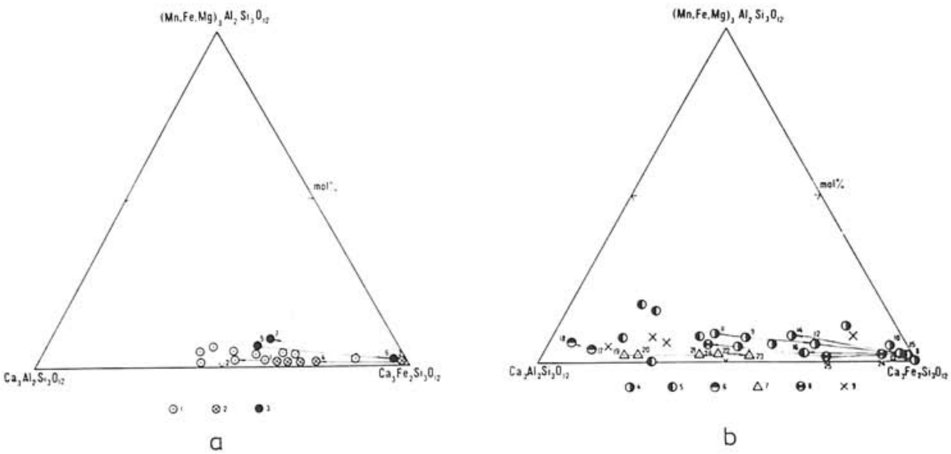

Fig. 1. The component composition of calcareous skarn garnets with the first-type zonation.
Explanations: a — skarn-iron ore; b — skarn-scheelite and other rare-metal deposits. *Designations:* 1 — Auerbakh; 2 — Dashkesan; 3 — Tomur-Te; 4 — Chorukh-Dairon (Власова и др., 1975); — Tyrnyauz; 6 — Beise; 7 — Ak-Dzhilga; 8 — King Island (Kwak, 1978); 9 — Teresia, Darinsk etc. (Индукаев, 1980). Arrows indicate composition variation in zonal garnets from centre towards margin.

Second-type zoning occurs in the garnets of calcareous skarns in the scheelite deposits, but in this case the skarn garnets are characterized by the grossular-rich composition (the andradite content being below 20—25 mol.%) and are distingushed by a significant content of pyralspite constituents, predominantly almandine and spessartine (totalling from 20 to 65 mol.%). Zonation may be direct and reverse with an increase (decrease) of the almandine and particularly spessartine content and the corresponding decrease (increase) of grossular from the core of the garnet grain towards its rim.

The diagram (Fig. 2a) shows the compositions of the zonal garnets from the skarn-scheelite deposits in Central Asia (U.S.S.R.): Ingichke Lyangar, Maikhura. The distribution of points on the diagram shows perceptible deviations of the compositions of zonal garnets from the total chemical determinations towards a higher spessartine content. A similar tendency in the distribution of pyralspite constituents has been noted by Simazaki (1977) and Sato (1980) garnets (1.9—12.6 mol. % of andradite) from Japanese skarn-scheelite deposits as shown in iFg. 2b. As can be seen on the diagram, zonality in the grossular garnets (1.9—12.6 mol.% of andradite) from Japanese skarn-scheelite deposits is a direct one and it is determined by a variation (increase) of the almandine and spessartine content and a decrease of the grossular content from the core of the garnet grain towards its rims.

Third-type zoning occurs exclusively in garnets of the apomagnesial varieties of calcareous skarns. The garnets are the grossular and andradite--grossular ones, with the pyrope content from 4 to 25 mol.%; in a maximal case

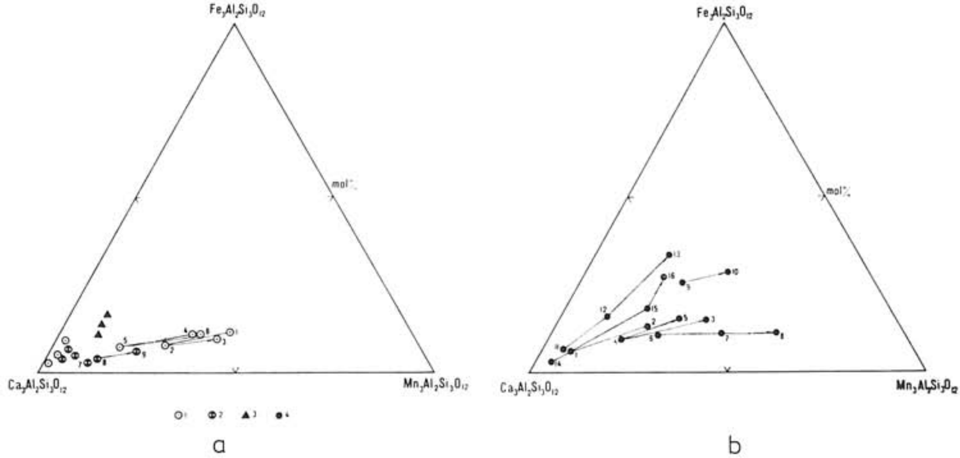

Fig. 2. The component composition of calcareous skarn garnets with the second--type zonation.
Explanations: a — the skarn-scheelite deposits in the U.S.S.R.; b — the skarn-scheelite deposits in Japan. *Designations:* Central Asia, U.S.S.R.: 1 — Ingichke; 2 — Lyangar; 3 — Maikhura; 4 — Japanese deposits (Shimazaki, 1977).

there was recorded the 45.8 mol.% content (King Island). Zonation is a reverse one and it is determined by an increase of grossular and, correspondingly, a decrease of pyrope from the core towards the rim zones of the garnet grains. Such type of zoning is usually identified with difficulty, since with the further growth of the crystals and the variations of the physico-chemical conditions of garnet crystallization this zonation, as a rule, is replaced with a secondary zoning which is due to an increasing content of andradite and a declining content of grossular from the inner towards the outer zones of the garnet.

This type of zonation was studied on the example of garnets from the apomagnesial calcareous skarns of Kuru-Tegerek (Central Asia). Variations of the chemical composition of these garnets is shown on the diagram (Fig. 3). Rather interesting is the behaviour of pyrope minal in the garnet: its high content in the central part (up to 25 mol. %), a redistribution inside the crystal and its declining content towards exterior zones. In this case in the central and interior zones of the crystal the garnet composition is principally determined by the content of grossular and pyrope minals. The andradite content varies within 15—33 mol.%; closer to the rims there is observed a discrete variation of the composition from the essentially grossular-rich towards the essentially andradite-rich garnet, the pyrope content declining down to zero. Points on the diagram (Fig. 3) indicate, moreover, the results of the total chemical analyses of garnets from Kuru-Tegerek. Only in two cases the position of points corresponds to the compositions of the zones in a zonal garnet, the rest of the points are lying in the corner of the diagram, corresponding to nearly pure andradites, with the pyrope content from 3 to 7 mol. % and belong to wall-rock

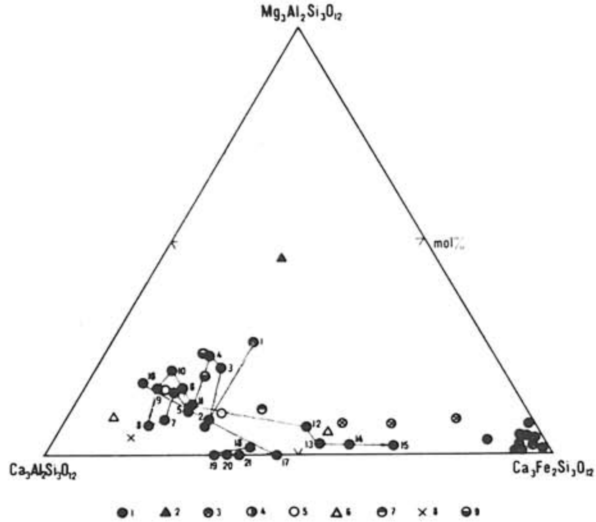

Fig. 3. The component composition of garnets of apomagnesial calcareous skarns with the third-type zonation.
Designations: 1 — Kuru-Tegerek; 2 — King Island (Kwak, 1978); 3 — Northern near-Ladoga (Иватшенко и др., 1980); 4 — Maikhura; 5 — Kharangon (Блохина, 1978); 6 — Gava-sai (Дженхураева и др., 1973); 7 — Nadezhdo--Kommercheskoe (Овчинников, 1960); 8 — Lesopromkhoznoe (Синяков, 1967); 9 — Baita Bihor (Stoici, 1974).

garnets. The diagram also comprises apomagnesian garnets total chemical analyses from tin and tungsten deposits of Middle Asia, the Urals and Siberia. The garnets from those deposits are related to andradites-grossularites containing from 6 to 23 mol. % of pyrope. Among them we can also expect the formation of garnets with the third-type zonal structure.

Secondary zoning appears as a result of andraditization of skarn garnets. Skarn garnet is often substituted by nearly pure andradite (up to 94—100 mol. %). An increase of the iron content is associated with a sharp increase of acidity and a variation of the oxidation-reduction conditions at the terminal stage of acid leaching and deposition of ore mineralization against the background of falling temperatures. Zonation is manifested in the replacement of colourless or weakly coloured zones (at the centre) with the light and dark grey ones in the external parts of the grains (Fig. 4), in the alternation of anisotropic and isotropic zones.

Fig. 4. A skarn garnet with a secondary zonation forming fringes at the grain edges (dark in this Fig.). The Lebedskyay skarn zone, U.S.S.R. Photograph by J. S. Cemenova.

Causes behind chemical zonation. Typomorphism

The geological conditions of formation and the characteristics of the mineral composition of calcareous skarns with different types of ore mineralization where zonal garnets have been encountered and studied reveal that calcareous skarns of the deposits concerned are distinguished by a similar geological and genetic position and belong to the same metasomatic formation.

The mineral composition, parageneses, construction of zoning, the composition of the minerals in solid solutions are dependent on such factors as the differential mobility of components, acidity and oxidizing-reducing conditions, temperature, pressure, mechanism of formation (diffusion, infiltration), etc. We shall now consider from these angles certain factors affecting the formation of garnets with different types of zoning.

Behaviour of principal mineral-forming components. First-type zonality occurs in the skarn garnets of the grossular-andradite composition with the pyralspite content not above 10 mol. %. Areal scanning of a grain of this type of garnet has revealed that zonation is determined by exchange diffusion of the isomorphic components Al^{+3} and Fe^{+3} (Fig. 5). There is a corresponding change in the distribution of the contents (activities) and migration rate of these components in the garnet crystal. In the case of a direct zonation the activity and diffusion rate of Fe^{+3} is higher than in the case of Al^{+3}, whilst in the case of a reverse zonation the opposite takes place, but in

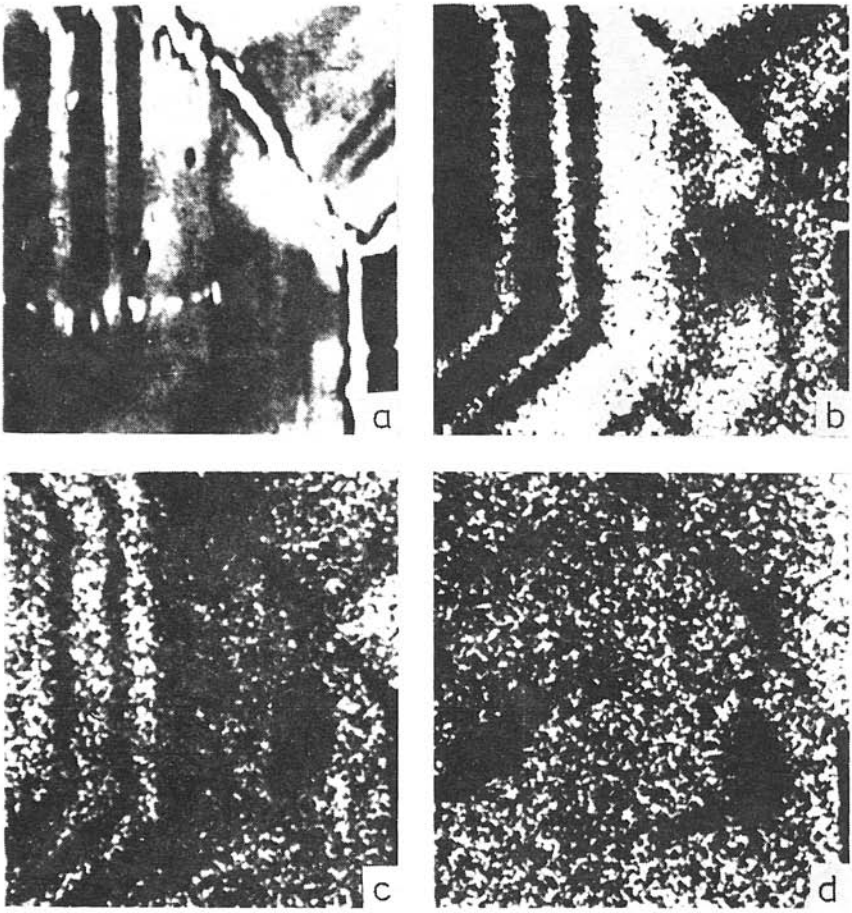

Fig. 5. Areal scanning (200×200 μm) of a skarn garnet grain with first-type zoning. Churkh-Dairon, U.S.S.R.
Explanations: a — e; b — K_{α}Al; c — K_{α}Fe; d — K_{α}Ca.

either case the migrational capacity of Fe^{+3} and Al^{+3} is superior to that of lime and silica.

The second type of chemical zonation occurs in the essentially grossular-rich garnet with a significant admixture of pyralspites (the andradite content being not above 10 mol.%).

Areal scanning of the garnet crystal with the second-type zonation (Fig. 6) reveals that zonation is determined by the exchange diffusion of isomorphous components Ca ⇄ Mn and Ca ⇄ Fe^{+2}. In the case of direct zonation the content of divalent iron and manganese, as well as diffusion rates are observed to increase, whilst the calcium content and the diffusion rate decrease from the centre of the crystal towards its outer zone. Thus, the second-type zonation

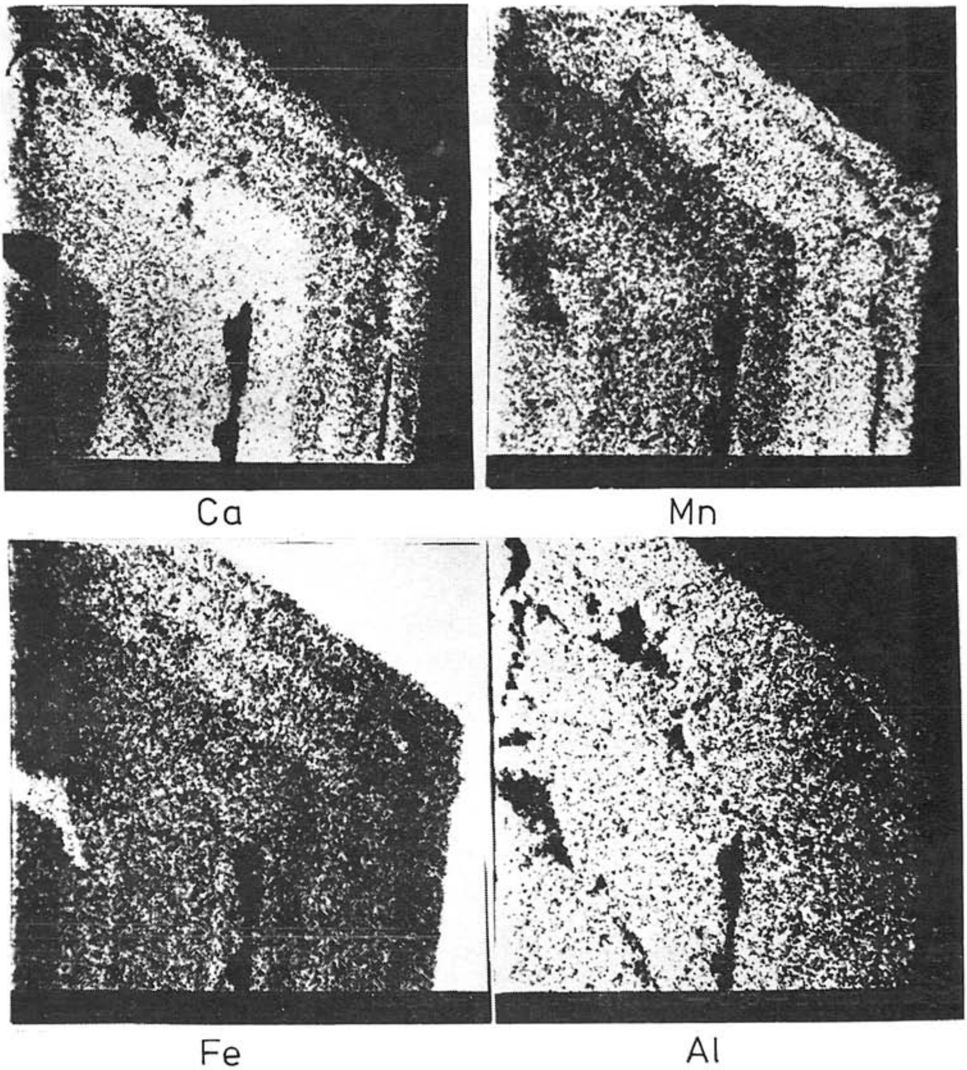

Fig. 6. Areal scanning (250×250 μm) of a skarn garnet grain with second-type zoning. Ingichke, U.S.S.R.
Explanations: a — K_{α}Ca; b — K_{α}Mn; c — K_{α}Fe; d — K_{α}Al.

is determined by the migrational capacity of manganese, divalent iron and calcium, which are correspondingly superior to those of alumina and silica.

Third-type zonation occurs exclusively in the earliest garnets of the apomagnesial varieties of calcareous skarns. These garnets are likewise of the essentially grossular composition (with the andradite content not above 30 mol.%), but they are distinguished by an increased content of pyrope. Areal scanning

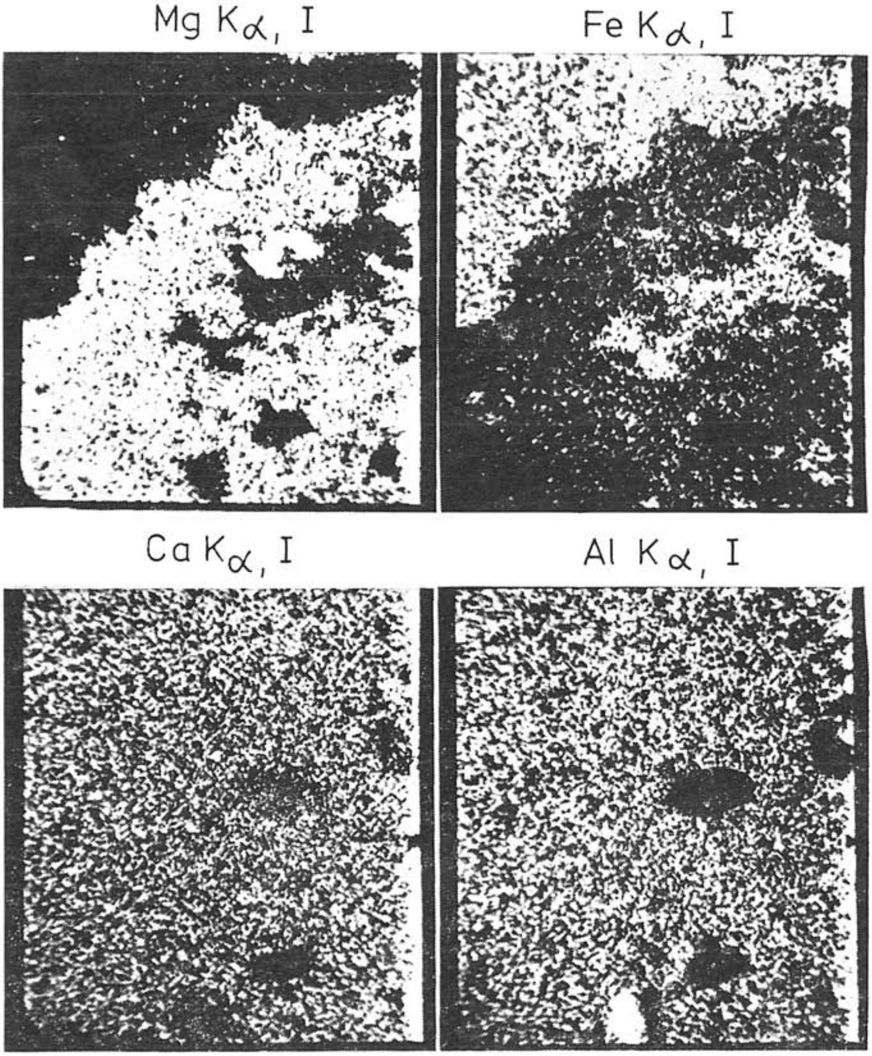

Fig. 7. A fragment of the central part of a zonal skarn garnet grain (third-type zoning). Kuru-Tegerek, U.S.S.R. Areal scanning 200×200 μm.
Explanations: a — K_αMg; b — K_αFe; c — K_αCa; d — K_αAl.

of the apomagnesial garnet crystal (Fig. 7) has revealed that the zonal structure is determined by a variation in the magnesium and iron contents. The content of calcium, alumina and silica does not change. The behaviour of principal mineral-forming components in the given case corresponds to the conventional progress of the process of the calcareous-skarn transformation of magnesial skarns: the garnets are distinguished by an increased magnesium

Table 2

Physico-chemical characteristics of the formation of skarn garnets

Zoning Type	Kind	Deposit	(%)	(kcal/mol)	K^{ox}
First	Direct	Tomur-Te	56—50	188.5—189.1	1.0—0.96
		Ak-Dzhilga	25—56	187.8—188.5	1.0
	Reverse	Auerbakh	61—54	188.5—188.3	0.98—0.99
		Dashkesan	97—75	189.3—188.9	1.0
		Chorukh-Dairon	100—72	189.2—189.1	1.0—0.98
			97—66	189.4—189.3	0.98—0.88
		Tyryauz	97—70	189.1—188.7	0.99—0.96
		Beise	12—11	189.4—187.0	1.0—0.52
		King Island	90—43	189.1—188.7	1.0
Second	Direct	Ingichke	17—18	191.2—191.4	0.24—0.17
		Lyangar	10—13	188.4—189.7	0.57—0.29
		Kuga	11—20	188.4—191.5	0.35—0.08
			19—22	190.5—193.0	0.32—0.25
		Kagata	21—37	192.1—192.8	0.54—0.12
		Fujigatani	18—34	188.1—191.8	0.78—0.20
	Reverse	Ingichke	18—15	192.1—191.8	0.15—0.12
		Fujigatani	37—33	192.0—192.8	0.15—0.13
Third	Reverse	Kuru-Tegerek	29—15	189.9—189.3	1.0

Note: y — arbitrary ionization potential; $K^{ox} = Fe^{+3}/Fe^{+3} + Fe^{+2}$ — degree of mineral oxidation.

content, the great migrational capacity of magnesium compared to that of calcium being clearly manifested.

A relative evaluation of the acidity (basicity) regime in the formation of skarn garnets with different type of zonation was undertaken by calculating the arbitary ionization potentials (y) suggested by Жариков (1982). From the Tab. 2 and diagram (Fig. 8) it is obvious that the garnets studies are formed under a specific acidity regime: the arbitrary ionization potential of garnets with the first-type zonation lies within the interval 187.0—189.5; second type — 188.0—193.0; third type — 188.3—190.0 kcal/mol. In this case there is observed an obvious correlation between the increase (decrease) of the arbitrary ionization (acidity) potential and the increase (decrease) of the iron content (f_m) in the direct (reverse) zonation.

Furthermore, as can be seen from the calculation data, the skarn garnets with the identified types of zonation differ not only with respect to acidity properties (arbitrary ionization potentials) but also with respect to the degree of oxidation. Thus, a transition from the garnets of secondary-type zonation to those of the first- and third-type zonation is accompanied by a decrease of acidity and an increase of the degree of oxidation of the mineral. This is associated with the changing activity of oxygen, a higher content of calcium (in the andradite) and of magnesium (in the pyrope-containing) garnets or by a higher activity of strong bases contributing to an increase of alkalinity and a sharp

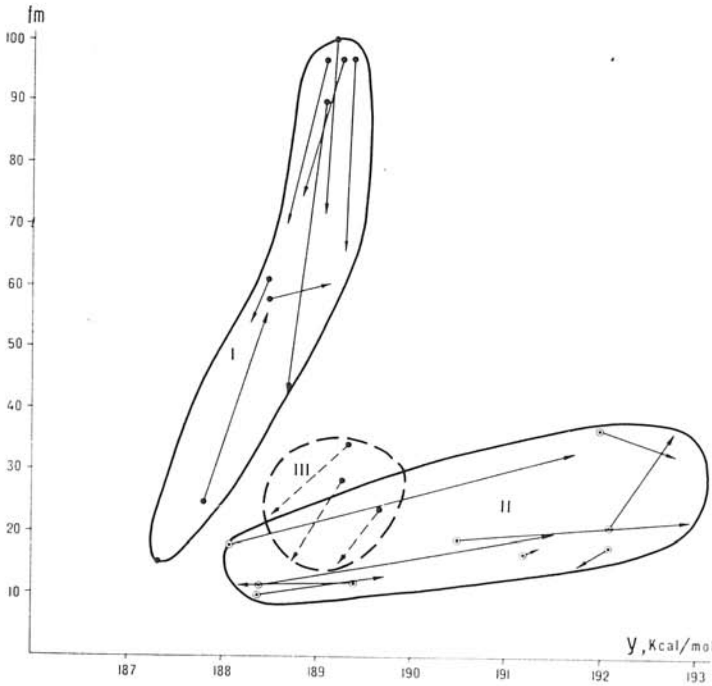

Fig. 8. Diagram in the coordinates iron content (fm) — arbitrary ionization potential (y, kcal/mol) to study calcareous skarn garnets with different types of zoning.

increase of the degree of oxidation of iron in the crystallizing garnet (high oxidation conditions). Garnets with the second-type zonation are formed under more reducing conditions, given a high activity of manganese and divalent iron and a low activity of calcium. In the case of second-type zonation a common tendency is observed to be clearly maintained in the garnets: an increase (decrease) of acidity calls forth a decrease (increase) of the degree of oxidation in the garnet.

T e m p e r a t u r e. According to the available thermometric data the temperature of formation of skarn garnets lies within 650—350 °C. Thermometric study of zoned garnets from the skarn-magmatite deposits of the U.S.S.R. (Б у х а р е в и д р., 1978; М а з у р о в, 1980) has revealed that the gaseous-liquid inclusions at the core of the zoned garnet grain become homogeneous correspondingly at 500 and 630—550 °C temperature and in the margin-zone — at 420 and 550—430 °C temperature.

Typomorphism. As regards the typomorphism of skarn garnets, the following can be noted. It was earlier assumed that pyralspite garnets are typomorphic minerals of the rocks of different metamorphic complexes formed under conditions of high temperatures (above 500—650 °C) and pressures (above 400 GPa). However, the presence in calcareous skarns of a number of deposits

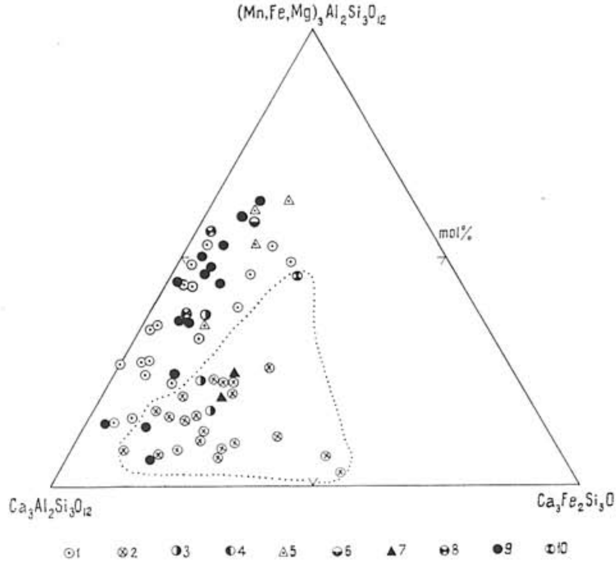

Fig. 9. The composition of garnets from skarn-scheelite deposits and apomagnesial calcareous skarns.
Designations: 1 — Ingichke, Lyangar; 2 — Maikhura; 3 — Kuru-Tegerek U.S.S.R.; 4 — Japanese deposits (Schimazaki, 1977); 5 — Victoria, Nevada (Lee, 1962); 6 — apomagnesial calcareous skarns of Central Sweden; 7 — Western Moravia, Czechoslovakia (Němec, 1967; 1971); 8 — the same; 9 — Baita Bihor, Roumania (Stoici, 1974); 10 — King Island, Australia (Kwak, 1978).

of pyralspite-grossular garnets points to their formation under conditions of lower temperatures and pressures.

The typomorphic skarn garnets were considered to be the garnets of the grossular-andradite series with the total admixture of pyralspite components not above 10 mol.%. What is more, the pyralspites in the composition of skarn garnets were not attributed any great significance.

The study of the composition of skarn garnets and their internal inhomogeneity with the use of today's methods has thrown light on the distribution in their composition of pyralspite components, the presence of which in the garnet is dependent on the type of ore mineralization of the skarns and replaced rocks.

The obtained data on the zonal garnets of the second type have enabled us, above all, to specify the contribution of spessartine and almandine in the composition of skarn garnets. The first finds of grossular-spessartine garnets were made by Lindrot (1916) and Magnusson (1940) in Swedish skarns and afterwards by Lee (1962) in the scheelite-containing endoskarns deposits Victoria with the spessartine content up to 36.3 and 42.8 mol. %, respectively. In Western Moravian skarns there have been described grossular garnets with the almandine content up to 50 mol.%. According to Němec (1967 1/2; 1971) the formation of garnets of such a composition is associated with the superposition of regional metamorphism processes on the skarns.

The diagram of Fig. 9 presents how available data on the composition of the garnets of the scheelite and other skarn deposits, from which it can be seen that these garnets can be also represented by the nearly complete isomorphous series grossular-pyralspites (the andradite content being not above 15 mol.%).

The typomorphic specifics of apomagnesial garnets from various skarn-ore deposits have been specifically examined by Шабинин (1975). On the basis of statistical treatment of total chemical analyses available in literature it was found out that: 1) apomagnesial garnets have an essentially grossular composition; and 2) the content of pyrope minal the average amounts to 3.6 mol. % (varies from 0 to 10 mol. %). In fact, the combination of obtained results in the study of the composition of zonal garnets and published data unambiguously indicates that the typomorphic specific of the given garnets of apomagnesial skarns is their essentially grossular composition and an increased magnesial content (up to 25 mol. % and higher) regardless of the composition of the ore mineralization. On the diagram (Fig. 9) these garnets are set apart in an independent field and are distinguished by a higher andradite content (above 15 mol. %).

Conclusions

1. Micro-X-ray spectral analysis study of zonal garnets of calcic skarns from different skarn ore deposits in the U.S.S.R. and Mongolia revealed three types of chemical zonation in skarn garnets.

The first type is characteristic of skarn garnets of the grossular-andradite series in iron, copper and polymetallic deposits.

This type of zonation is determined by Fe^{3+} and Al^{3+} isomorphism and formed in the conditions of a highly oxidizing environments, increased basicity of hydrothermal solutions and variation of temperatures.

The second type reveals itself in grossular garnets with spessartite and almandine components which are mostly typical accross in the skarn-rare metal deposits.

This type of zonation is determined by $Ca \rightleftharpoons Mn$ and $Ca \rightleftharpoons Fe^{2+}$ isomorphism and formed in the conditions of increased acidity and low fugacity of oxigen (reducing conditions).

The third type of zonation is characteristic of apomagnesium calcic skarn grossular garnets and it is determined by $Mg \rightleftharpoons Fe^{2+}$ and $Al^{3+} \rightleftharpoons Fe^{3+}$ isomorphism during alteration of oxidize-reducing conditions, acidity-basicity, drop of temperature, change of the mobility regime of mineral forming components of Mg, Ca, Fe, etc.

2. Study of the second type zoned garnets have also revealed almost complete miscibility between granite and puralspite in the conditions of a skarn formation.

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