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## SPHERICAL ACCESSORIES ("SPHERULES") IN GEMERIC GRANITES (WEST CARPATHIANS — CZECHOSLOVAKIA)

(22 Figs., 4 Tabs.)



**Abstract:** Separation of zircons from the Gemic Granites resulted in the finding of spherules of various shape, colour, size and composition in the heavy fraction sized 0.12 to 0.09 mm. Most of the spherules are gray, with polygonal surface disintegration and their composition is likely to correspond to some of Fe—Si phases. Other spherules are black, metallic and/or spinelid (Fe, Ti, Al), and exceptionally also silicate ones (Fe, Si, Al, Ti,  $\pm$  Ca, Mg, P) have been found. The authors describe their characteristics and discuss their meteoric or terrestrial origin.

**Резюме:** При сортировании цирконов из гемеридных гранитов Западных Карпат были найдены в тяжелой фракции 0,12—0,09 мм сферулиты различного цвета, формы, размера и состава. Большинство этих сферулитов является серыми и имеет полигональный распад поверхности и по составу они соответствуют наиболее правдоподобно некоторой Fe—Si фазе. Другие сферулиты являются черными, металлическими, или шпинелидными (Fe, Ti, Al) и спорадически были найдены тоже силикатовые (Fe, Si, Al, Ti  $\pm$  Ca, Mg, P) в статье описываются их свойства и дискутируется о их космическом или наземном происхождении.

### Introduction

In the southernmost West Carpathian tectonic unit — Gemicum — there are known small granite massifs near Hnilec, Súľová, Betliar, Poproč, Zlatá Idka and in some other places (Fig. 1). Because of the territory of their occurrence but mainly due to the fact that their properties are different from granites in the other West Carpathian units, these granites have been designated as "Gemic". Geophysical data suggest that their extent in the depth is much larger than on the surface (Fig. 1). The Gemic Granites are acid, potassium-rich, tourmalinic and relatively little tectonically fractured and faulted. In places (e. g. Hnilec) the granites are greisenized and tin-bearing, but they are mostly biotite or muscovite-biotite granites with tendency to porphyric development. From the petrographic viewpoint they have been designated as palingenetic and their petrographic characteristics were given mainly by Ončáková (1954), J. Kamenický — L. Kamenický (1955). The petrologic and metallogenic properties of the granites were studied by Baran et al. (1970), Varga (1975), Tauson et al. (1983). The accessory minerals — predominantly tourmaline, zircon, apatite, fluorite, topaz, monazite,

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garnet and numerous ore minerals including cassiterite and native tin were described by Veselský et al. (1983). The Gemic Granites intrude weakly metamorphosed Lower Paleozoic (Ordovician—Devonian) sedimentary and volcano-sedimentary rocks of the Gemicum. Various cleavage planes of the Lower Paleozoic rocks are disturbed near the contact with the granites suggesting their relatively young geological age: Alpine—Upper Cretaceous (Rozložník, 1976). Their isotopic ages, however, are controversial: Permian—Triassic—Jurassic—Cretaceous (Cambel et al., 1980).

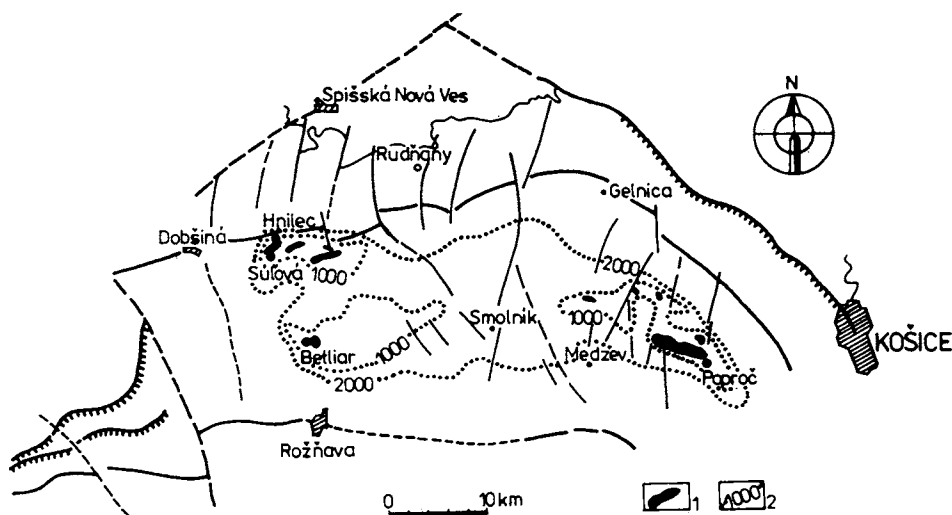


Fig. 1. Schematic map of Gemic Granite occurrences.

*Explanations:* 1 — granite outcrops, the most important localities are marked; 2 — geophysically determined isohyphs of granite extent at depth (Šefara—Plančár et al. 1978 — simplified and supplemented by L. Rozložník, 1987).

Our researches have been aimed at the determination of the morphometric properties of the zircons, which enabled us to establish petrological properties of the Gemic Granites (Jakabská — Rozložník, 1988).

During separation and identification of the zircons, small spherical remarkable accessories have been found.

#### *Morphological and physical properties of the spherical accessories*

The spherical accessories (spherules) have been found in all investigated samples of the Gemic Granites from the localities: Betliar, Súľová and Hnilec (Fig. 1), except for the village of Poproč. The spherules came along with

the zircons in the heavy fraction sized 0.12—0.09 mm. After the spherules had been found among separated accessory minerals, they were looked for also in thin-sections where they were eventually discovered (Fig. 2). The spherules in thin-sections can easily be confused with air-bubbles and thus overlooked. Moreover, in the course of polishing most spherules may have fallen out of the thin-sections. Under ordinary binocular microscope, several types of spherules can be recognized. The most abundant are gray spheroidal bodies, followed by black — blue-black ferruginous and "titanium" ones. Brown-black (alumoferrosilicium) and dark-gray ("aluminium") spherules occur exceptionally. The black and black-brown spherules form almost perfect spheres, whereas the gray ones are of very different shapes designated as spheroidal.

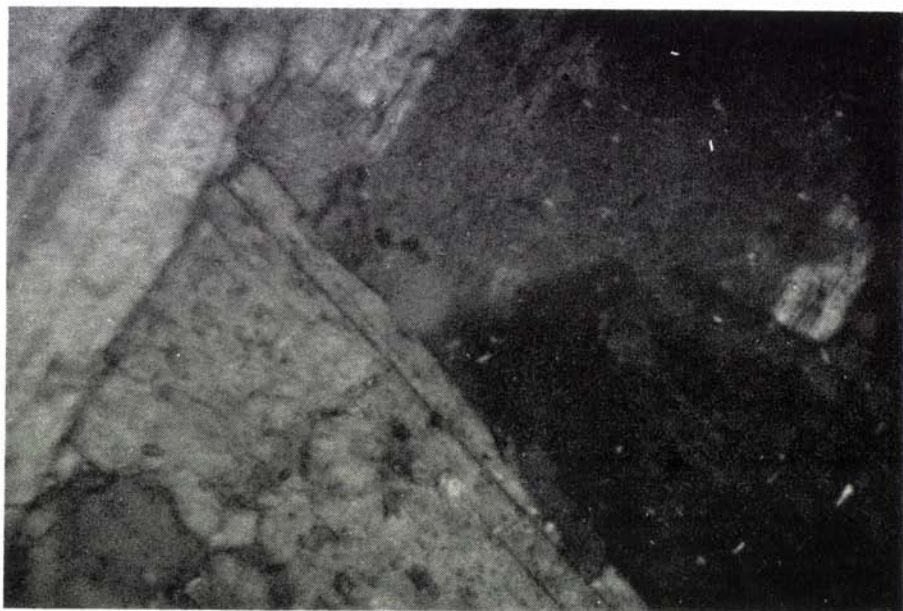


Fig. 2. Section of a gray spherule in Betliar Granite, in the central part nicols  $\parallel$ , magn. 66  $\times$ .

The gray bodies — as observed under the scanning electron microscope (SEM) — are sometimes spherical (Fig. 3). On their surface, however, there are often small conical or tumour-shaped protrusions or holes (Fig. 4), which are frequently intergrown (Figs. 5, 6). Potato- (Fig. 4) and pear-shapes (Fig. 7), even with some sort of "branches" (Figs. 8, 9), are common. When magnified several thousand times, the crust of the gray spherules exhibits polygonal disintegration (Fig. 10). Such a type of crust usually covers materials which undergo rapid cooling and shrinking — e.g. meteorites or amorphous alloys.

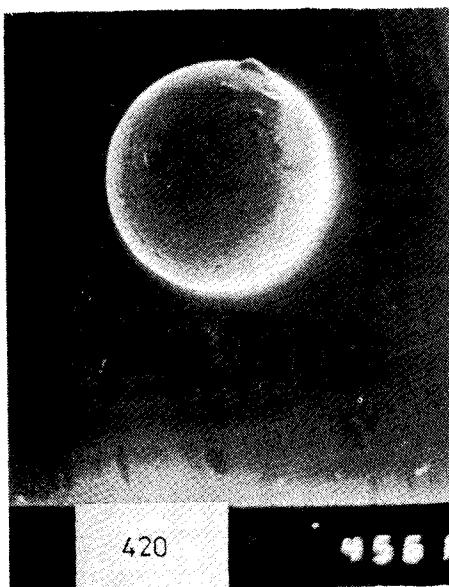


Fig. 3. Spherical shape of a gray spherule, Betliar Granite, magn. 420  $\times$ .

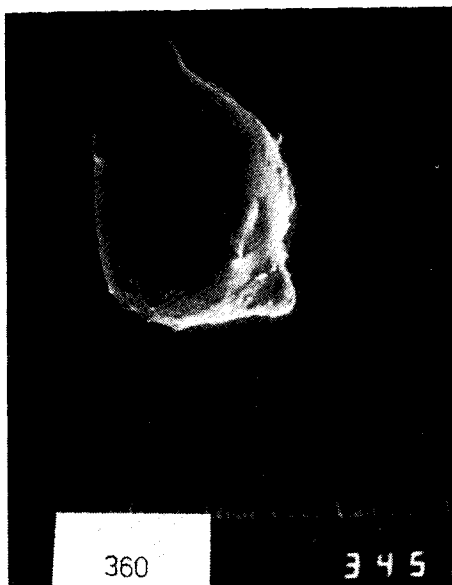


Fig. 4. Potato-shaped gray spherule with conical protuberances, Hnilec Granite, magn. 360  $\times$ .



Fig. 5. Gray spherule intergrowth, Hnilec Granite, magn. 360  $\times$ .

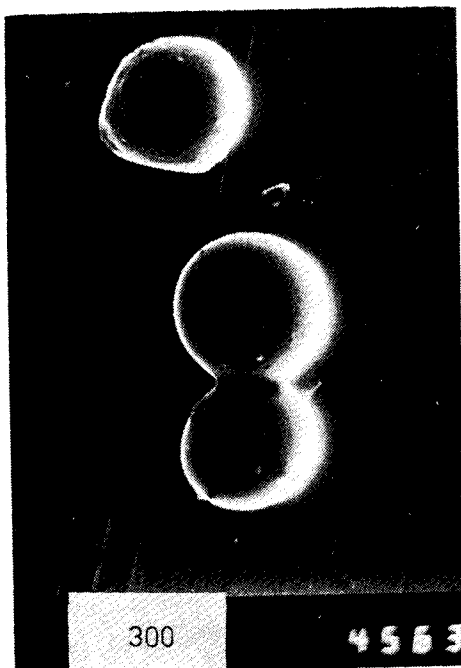
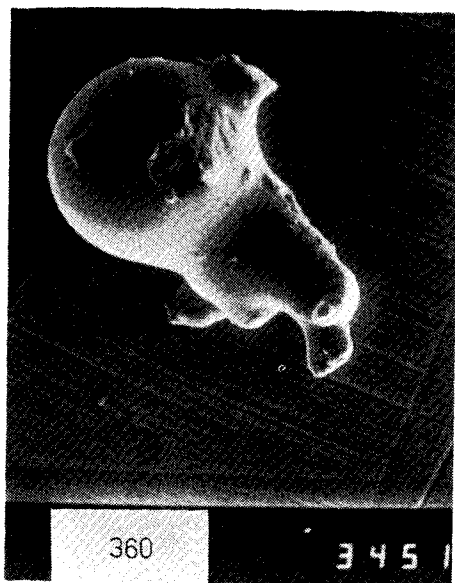
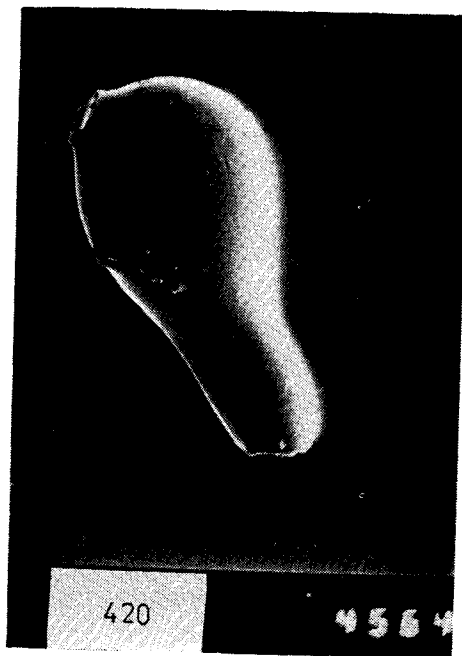


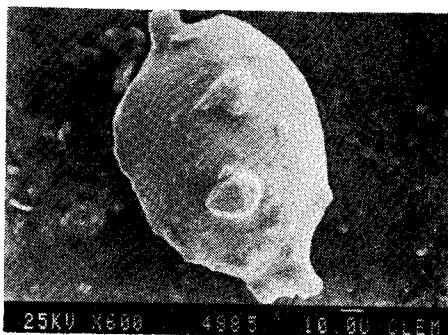
Fig. 6. Gray spherule intergrowth, Betliar Granite, magn. 300  $\times$ .

Fig. 7. Gray pear-shaped spherule.

*Explanations:* Depressions probably represent traces after broken off protuberances or traces after contacts with other spherules, Betliar Granite, magn. 420  $\times$ .



a)



b)

Fig. 8. Gray pear-shaped spherule with "branches" and traces after broken off protuberances or contacts with other spherules.

*Explanations:* a) Hnilec Granite, magn. 360  $\times$ ; b) Súľová Granite, magn. 600  $\times$ .

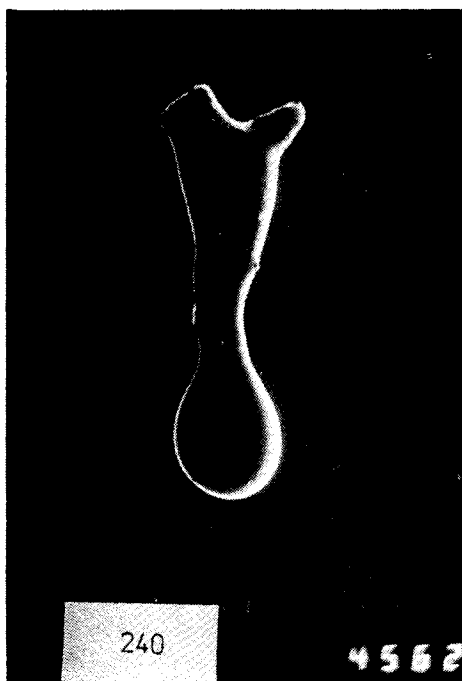


Fig. 9. Irregular gray spherule, Betliar Granite, magn. 240  $\times$ .

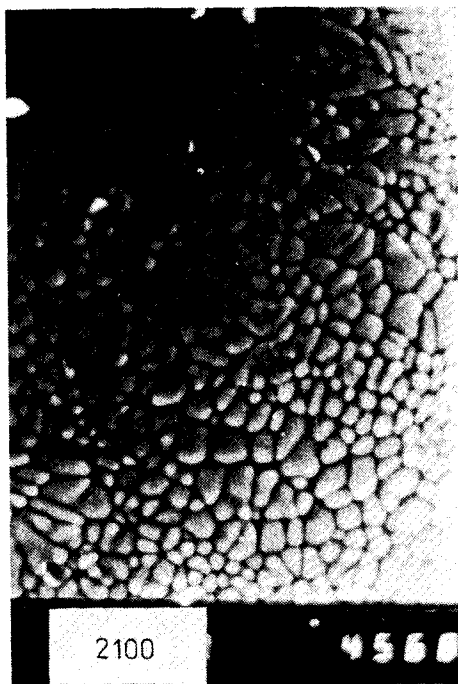


Fig. 10. Polygonally disintegrating crust of a gray spherule, Betliar Granite, magn. 2100  $\times$ .

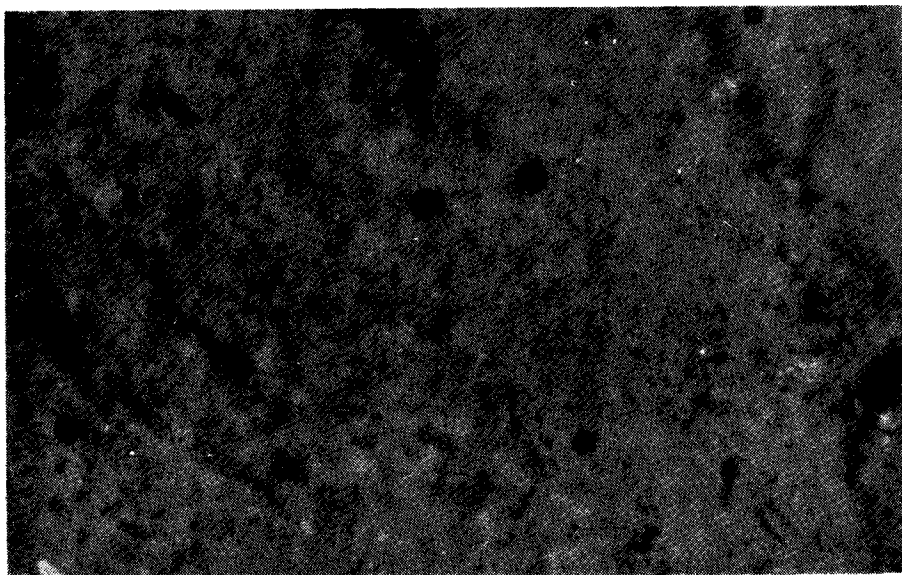


Fig. 11. Cross-sections of black spherules in a thin-section, Hnilec Granite, magn. 28  $\times$ .



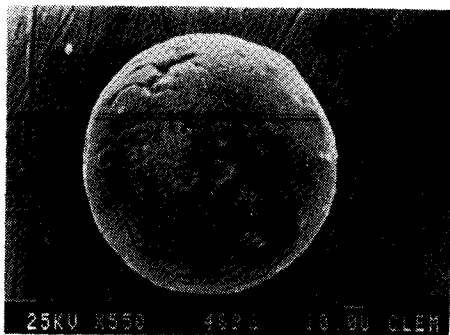


Fig. 12. Black ferruginous spherule, Hnilec Granite, magn. 550  $\times$ .

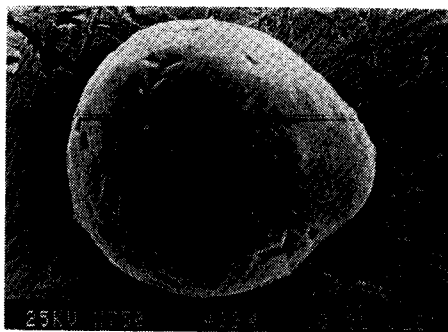


Fig. 13. Black ferruginous spherule, Hnilec Granite, magn. 750  $\times$ .

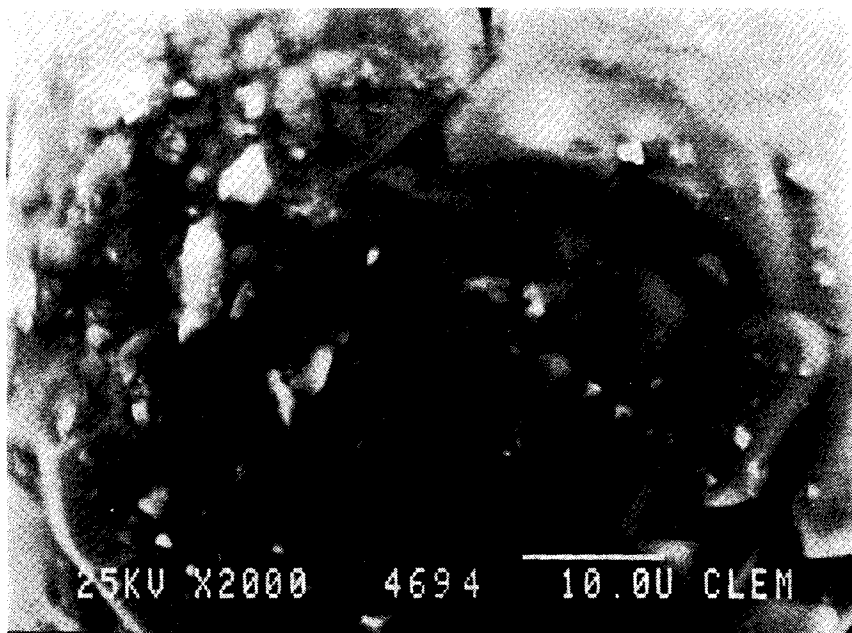


Fig. 14. Surface of a black ferruginous spherule with a visible crystalline texture, Hnilec Granite, magn. 2000  $\times$ .

The gray spherules in thin-sections (Figs. 2, 11) were always present as inclusions in feldspars (in perthitic orthoclase or albite—oligoclase). Under a polarization microscope, cross-sections of spherules show frequent occurrences of tumours and cavities on their surface (Fig. 11). They are not transparent, in reflected light with bright orange reflection. Under binocular microscope their surface is lustrous.

The black — blue-black and brown-black spherules are most often of regular spherical (Fig. 12) or spheroidal oval shape (Fig. 13). Their surface is very lustrous with blue-black shades. The superficial texture of some spherules reveals their crystalline structure (Fig. 14). The brown-black and dark gray spherules of larger size, "aluminium" or "aluminium—ferrosilicate" are sporadic.

Because of their small number and small size, which is only several hundredths of millimetre, the chemical analysis of the spherules is difficult. Properties of the gray spherules have been determined more accurately, because they are relatively more abundant and therefore also X-ray diffraction analysis could have been carried out.

#### *Chemical composition and texture of gray spherules*

The chemical composition of the gray spherules determined by means of electron microprobe analysis carried out on the apparatus JEOL - Super Probe 733 supplemented by a microparticle analyser and the control computer PDP - 11/04 (Geological Institute of Dionýz Štúr, Bratislava) is given in Tabs. 1, 2.

Table 1

Microprobe analyses of gray spherules in a section from Hnilec Granite

Points Elements <sup>0/0</sup>	1	2	3	4	5	6
Fe	82.9586	81.1299	81.2779	82.5038	82.8449	81.5273
Cr	0.2406	0.2130	0.2165	0.1104	0.2086	0.2623
Mn	0.3811	0.4385	0.3896	0.3261	0.3439	0.4029
Al	0.0582	0.0777	0.1240	—	—	0.1063
Si	6.8245	6.6625	7.0283	7.3920	7.0569	7.0076
Σ	90.463	88.5216	89.0363	90.3323	90.4543	88.9035

1, 2, 3, 4, 5, 6 — points of records in sections.

Microprobe analyses carried out at Geological Institute of D. Štúr, Bratislava.

In the gray spherules, Fe and Si are by far the most predominant elements. Other elements — Cr, Mn and Al are much less abundant. The analyses carried out do not exclude eventual occurrence of oxygen and carbon. This assumption is justified by the fact that the sum of analyses given in Tab. 1. is only 88—90 %. However, the stoichiometry of ferrosilite or fayalite requires more than 10 % oxygen and less Fe. Or is the residual 10—12 % represented by carbon and the composition corresponds to a phase between moissanite and cohenite?



Table 2

Semiquantitative analysis of gray spherules

Locality Elements (%)	Hnilec	Hnilec	Súľová
Fe	77.955	75.371	81.50
Cr	0.479	0.483	—
Mn	0.323	0.356	0.34
Si	21.020	23.389	17.55
Cu	0.222	0.401	0.10
$\Sigma$	99.999	100.00	99.49

Copper content may be due to reflection from copper base. Semiquantitative analyses carried out on an energy dispersion analytic unit attached to REM-JEOL-JSM-35CF (Department of Material Science, Metallurgical Faculty, Institute of Technology, Košice).

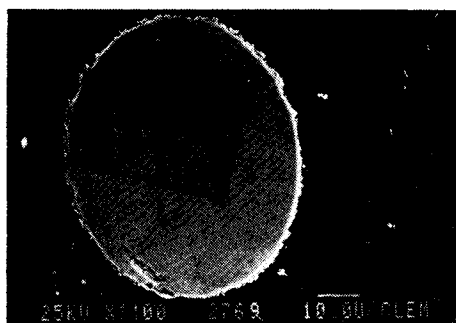


Fig. 15a. Cross-section of a gray spherule from Hnilec Granite, magn. 1100  $\times$ .

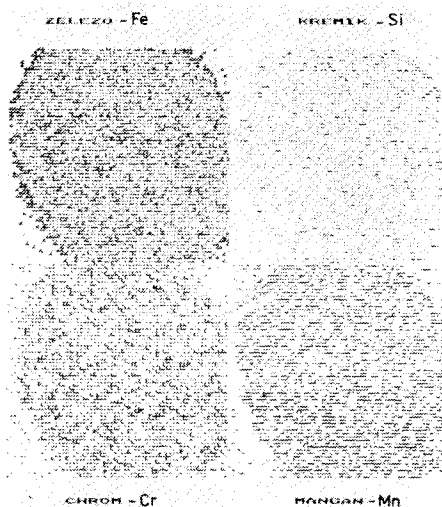


Fig. 15b. Areal Fe, Si, Mn and Cr distributions in a gray spherule from Hnilec Granite.

The chemical composition of the gray spheroidal bodies is similar to that of ferrosilicium. Ferrosilicium produced by the firm Knapsack has the following composition: 66.35 % Fe and 33.35 % Si. If our gray spherules are of Fe — Si character, then their Fe/Si ratio is higher than that corresponding to ferrosilicium. The Fe/Si ratios given mainly in Tab. 2 best correspond to the formula

Fe<sub>2</sub>Si. Ferrosilicium with the formula Fe<sub>2</sub>Si contains 79.91 % Fe and 20.09 % Si. Analyses in Tab. 2 show that our spherules contain 75—81 % Fe and 17—23 % Si. Nevertheless, Tab. 1 does not exclude also other compounds, mainly with carbon and oxygen. Distribution of Fe, Si, Mn and Cr in the gray spherules observed in the section in Fig. 15a under the scanning electron microscope by means of EDAX - JEOL - JSM - 35 CF (Department of the Science on Metals, Metallurgical Faculty, Institute of Technology, Košice) indicates a homogenous material with no zonation and inclusions (Fig. 15b).

#### *X-ray diffraction analysis of gray spherules*

The X-ray diffraction analysis has been carried out by Ž d i m e r a (Faculty of Natural Sciences, Charles University, Prague) by means of the Debye-Scherrer method with diffracted radiation registered on a flat film. The analysis has been carried out under the following conditions: radiation Co/without filters, exp. 92.30 hrs, chamber Ø 114 — MÜLLER MIKRO. The results can be seen in Tab. 3. The same sample has been investigated by means of X-ray diffraction analysis Derco (ATNS Košice). He used the Bragg-Brentano method on the apparatus Mikrometa II under these conditions: radiation FeK  $\alpha$ ; filter Mn, voltage 25 kV, current 12 mA, displacement of the arm of the goniometre 2°/min, movement of paper 600 mm/hr, subduing T/10, screen 10' — 5', sensitivity  $3 \times 10^2$ .

The X-ray diffraction analysis (Fig. 21) suggests a mineral phase with lower degree of crystallinity. The main plane (100) is developed very well, whereas the other secondary ones are developed only very slightly. This fact confirms the presence of a crystalline mineral phase of the following types: FeSi, Fe<sub>2</sub>Si, FeSi<sub>2</sub>, Fe<sub>3</sub>Si, with the best proximity to the phase Fe<sub>2</sub>Si.

The assumption that our gray spherules are related to some phase in the Fe — Si system is confirmed also by an X-ray diffraction pattern (Fig. 21) made in the same way as the record in Fig. 22. The main planes of both these materials are very close.

#### *Characteristics of the other spherules*

As has already been mentioned, the most abundant gray spherules are sporadically accompanied by black, and/or brown-black and dark gray ones. These could not have been studied by X-ray techniques because of their insufficient amount. Semiquantitative analyses have been carried out by means of electron microanalysis by the method of energetic dispersion from the surface of the spherules coming from the Gemeric Granites from the localities Hnilec, Súľová and Betliar. The following types are present (ranked according to their abundance):

Group of *ferruginous spherules* — on the basis of 11 measurements they average 98—99 % Fe, 0.2—0.9 % Mn, 0.5 % Ti (only traces of other elements).

Group of *titanium spherules* is characterized by very small dimensions and following composition: 55 % Ti, 19 % Fe, 10 % Mn, (1—2 % Al, K, Ca).

Group of *aluminium spherules*: 91.98 % Al, 5.56 % Fe, 0.97 % Ti, 0.42 % Mn.

Group of Fe—Al—Ca—Si spherules has a very variable composition,



Table 4

Semiquantitative analysis of silicate spherules from Súľová Granite

Elements (%)	1	2	3
Al	43.52	7.99	1.27
Fe	23.20	34.16	52.36
Si	23.29	34.29	10.95
Ti	8.56	6.72	5.27
Mn	—	0.80	0.29
Ca	0.79	9.70	25.09
K	0.49	0.34	0.19
Mg	—	6.00	0.34
P	—	—	4.24
$\Sigma$	99.76	100.00	100.00

Semiquantitative analyses carried out on an energy dispersion analytic unit attached to REM-JEOL-JSM-35 CF (Department of Material Science, Metallurgical Faculty, Institute of Technology, Košice).

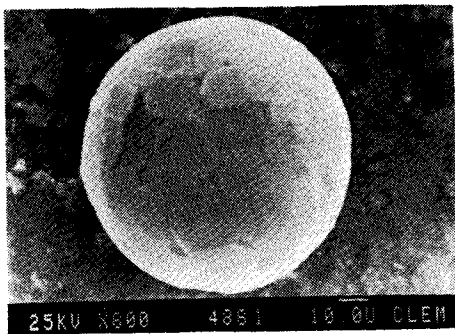


Fig. 16. Fe-spherule with an even surface from Súľová Granite, magn. 600  $\times$ .

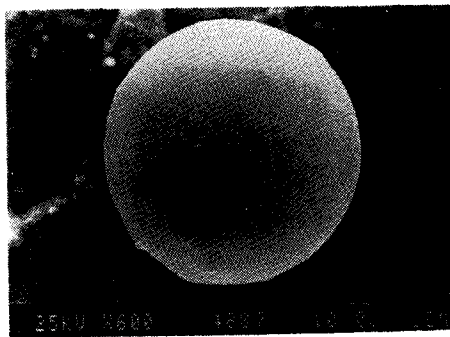


Fig. 17. Fe-spherule with disintegrated crust similar to tortoise shell or socker ball from Súľová Granite, magn. 800  $\times$ .

as is shown by Tab. 4. A high Ti content is very surprising in this "silicate" composition.

The group of black and brown-black spherules as a whole has an unclear chemical composition because their semiquantitative analysis does not identify the presence of oxygen and carbon. Therefore in the case of the iron spherules we do not know if they consist of magnetite or wüstite or native iron. The lustrous surface of black-blue relatively perfect spherules with some crystalline structure on the surface (Fig. 14) resembles magnetite spherules rounded in placers, alluvium or beach. Some ferruginous spherules, however, have a

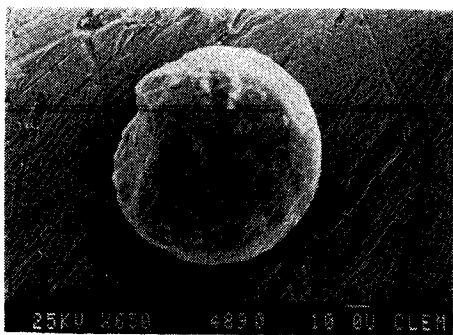


Fig. 18. Ti-spherule from Hnilec Granite.  
magn. 650  $\times$ .

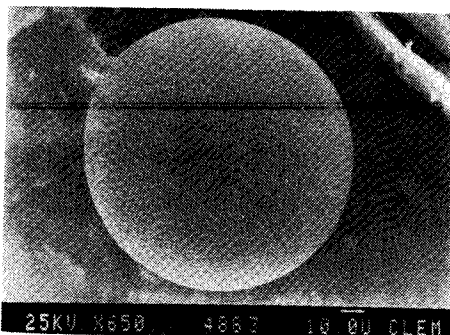


Fig. 19. Al-silicate spherule from Súľová  
Granite, magn. 650  $\times$ .

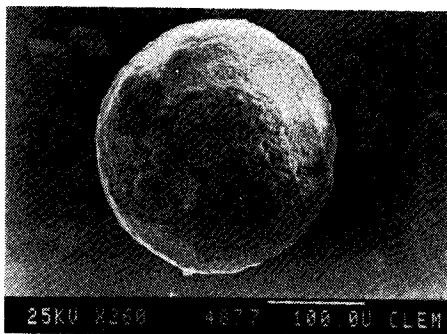


Fig. 20. Al-spherule from Súľová Granite.  
magn. 260  $\times$ .

very regular surface — either without regular texture manifestations (Fig. 16) or represented by polygonal disintegration of the spherule surface similar to the surface of a tortoise shell or football ball (Fig. 17).

Similarly, also in the case of titanium-rich spherules (Fig. 18) we may only suppose that they consist of ilmenorutile (?).

The chemical composition of the spherules rich in Al (Fig. 20) or Al—Fe—Si—Ti and/or Fe—Si—Ca—Al—Ti—Mg or Fe—Ca—Ti—P—Al (Fig. 19) can be expressed even less exactly.

The latter might have been contaminated by remnants of other minerals on their surface. It is worth noting that the Al and Al—Fe—Si—Ti spherules (Figs. 19, 20) have the largest dimensions, whereas those rich in Ti and Fe (Figs. 16, 17, 18) are smallest.

#### *Occurrences of spherical bodies in the world and their genetic interpretation*

Tiny ball-shaped (spherical, spheroidal) bodies (spherules) are abundant on the Earth surface as well as on the Moon and are dealt with in very extensive literature. Spherules found on the Earth are usually regarded as extraterrestrial, meteoric-intrastellar dust fallen onto the Earth surface. The meteoric origin of these spherules is proved by their relatively abundant occurrence on

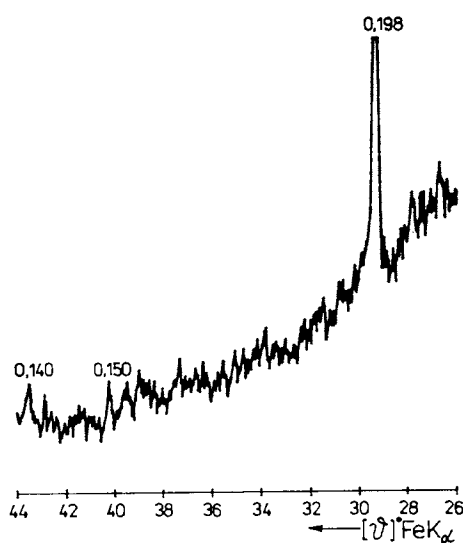


Fig. 21. X-ray diffraction record of gray spherules from Hnilec Granite.

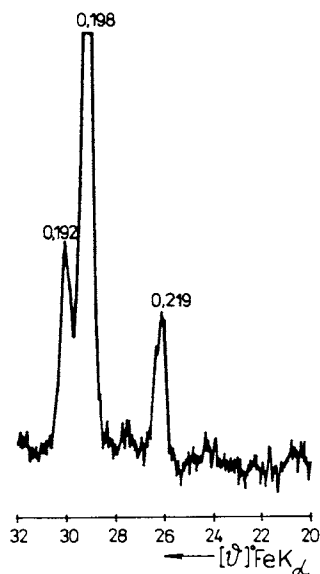


Fig. 22. X-ray diffraction record of ferro-silicium produced by the firm Knapsack. Chemical composition: 66.35 % Fe, 33.35 % Si.

the surface of polar icesheets, meteorological balloons and in deep sea clays (Yabuki, 1972; Parkin et al., 1977; Bownlee, 1979; Herr et al., 1980; Lozoyaya, 1981). Spherules formed by melting of meteorites in the course of their flight through the atmosphere or impact spherules formed by the impact of large meteorites can be considered as meteoric, too. The latter might partly contain also material of terrestrial origin. The composition of the meteoric spherules is very variable. There are metallic (Fe, Fe — Ni), spinelid (magnetite, chromite, wüstite and others), glassy (Si — Ca — Fe — Mg — Al) and even also graphite spherules. The spherules were found also in volcanic rocks (Filimonova et al., 1981; Filimonova, 1982). These authors discovered tiny spherical, egg-shaped and platy bodies having dimensions of up to 0.2 mm amidst shoshonite andesites and potassium rhyolites of the Southern Sikhote—Alinja Belt. The age of the andesites is 62—52 m.y., whereas the rhyolites have been dated at 57—47 m.y. This volcanic activity was accompanied by simultaneous formation of cassiterite mineral along with Fe, Pb, Zn and Cu sulphides. The metallic spherules represent the association of cubic native  $\alpha$  — Fe, Sn, Cu and tetragonal Pb. Sometimes they have a thin crust composed of lizardite and chrysotile—asbestos and are accompanied by moissanite. Due to their shape the spherule association is regarded as a “high-pressure” one, formed at low oxidation-reduction potential under the action of carbon, which is deduced also from the presence of moissanite. The above-mentioned authors assume a genetic relationship between the metallic spherules and volcanogene hydrothermal accumulations of oxides and sulphides.

They regard the presence of the spherules in the volcanic rocks as an indication that the magma was ore-bearing — i.e. prospecting manifestation.

The origin of the terrestrial spherules is derived from the separation of two immiscible melts under subcrustal conditions. This assumption is supported by the results of experiments (M a n a k o v — S h a r a p o v, 1983). According to A s h i k h m i n a et al. (1987) spherical glassy state of the matter is a frequent natural state of magma between melt and crystals — between liquid and solid state.

Ferrosilicium alloys have been found on the Earth not only in meteorites but also in sedimentary rocks alongside with moissanite (N o v o s y o l o v a — S o k h o r, 1983). N o v g o r o d o v a et al. (1983) identified  $\text{Fe}_3\text{Si}$  (suessit) that occurred in the cavities of andesite-basalt porphyres. According to the opinion of the above authors the formation of  $\text{Fe}_3\text{Si}$  was accompanied by the formation of titanium carbide and both were created in a strongly reducing environment.

Artificial balls represent the third group of the spherules. These originate during forest fires and mainly by the combustion of coal. Their composition and overall character are the same as those in the two preceding genetic groups. That is why it is difficult to distinguish extraterrestrial, terrestrial and artificial balls from each other.

In Czechoslovakia, spherical bodies were studied by Č í l e k (1983), Č í l e k et al. (1985) who, among other things, described magnetite and silicate balls from sediments as well as crystalline of the Bohemian Massif. V e s e l s k ý et al. (1983) described spherical wüstite (?) from West Carpathian granites.

Likewise on the world-wide scale, also the genetical interpretation of the spherules from the Gemic Granite is not without problems.

#### *Discussion about character and origin of spherules in the Gemic Granites*

The most abundant — gray spherules of the Gemic Granites most probably consist of a ferrosilicium material, which is indicated by chemical analyses but mainly by X-ray analysis. If we take into account conditions under which artificial ferrosilicium balls are created then the formation of our spherules from the phase  $\text{FeSi}$  and/or  $\text{Fe}_2\text{Si}$  would require heating of the  $\text{FeSi}$  melt to a temperature of at least  $1200^\circ\text{C}$  and its rapid cooling by water. At the same time also an abrupt decrease in pressure is necessary. In nature such PT conditions may best arise by the impact of extraterrestrial bodies (and/or impacts) onto the Earth surface into water. This is indicated also by frequent occurrences of droplike shapes of our gray spherules (Figs. 7, 8, 9). And what about the other: ferruginous, titanium, aluminium and silicate spherules? Their origin can hardly be explained in this way. The black spherules rather resemble placer minerals rounded in alluvium or beach environment. Nevertheless, they might have also originated from a stellar dust fallen onto the Earth surface and then the spherules might have got, along with the impact (?) ferrosilicium ones, from sediments to the melt of the Gemic Granite where they remained preserved. Meteoric Fe-spherules, however, typically contain Fe — Ni phase, which is absent in ours. On the contrary our spherules contain a distinct Fe — Ti — Mn — Cr association.

Surprisingly, no traces of Sn — W — Mo — Nb — Ta have been found in the



spherules, although the Gemic Granites are a potential carrier of these metals. All these facts support the version that the spherules came into magma of the Gemic Granites from alluvial or beach sediments. Because of the preliminary character of the results presented and difficult distinguishing of spherules formed in various ways, we cannot exclude also the origin derived from the initial stages of the formation of the Gemic Granite magma.

Ashikhmina et al. (1987) thoroughly investigated lunar as well as terrestrial glassy balls and come to the conclusion that natural glasses formed when chemical balance of the magmatic liquid phase is disturbed. It is protocrystalline state with unfinished level of crystallinity. High-dispersion colloidal aggregation takes place by which nullvalent forms of the elements  $\text{Fe}^0$ ,  $\text{Ti}^0$ ,  $\text{Si}^0$  are formed. This state resembles the physico-chemical character of our spherules.

Regardless of their meteoric or terrestrial origin, the spherules found in the Gemic Granites are undoubtedly important from the petrological point of view.

Geochronological methods have determined that the granite near Betliar is younger than the Hnilec and Súľová Granites. Occurrences of spherules found at the localities Betliar, Hnilec and Súľová suggest that the bodies of these three massifs were formed from the same magma, more or less simultaneously.

The finding of the spherules thus confirms the results of the morphometric research of zircons (Jakabská — Rozložník, 1988) which also favours the idea of a joint mantle-crustal magma from which the small massifs of the Gemic Granites: Betliar, Poproč, Hnilec, Súľová were formed. Their different geochronological ages may be due to large and variable share of palasome by the formation of the granites.

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