| IN MEMORIAM DR. JÁNOS BÉRCZI |

LIESEGANG LAYERED CLINOPYROXENE MEGACRYST INCLUSION FROM SZENTBÉKKÁLLA, HUNGARY

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ABSTRACT

Fourteen parallel bands composed from submillimeter sized olivine grains scattered along planes form an almost uniformly spaced (7—9 mm) layered structure in the large, decimeter sized single crystal of clinopyroxene of our Spx—3 inclusion. A medium range of uniformly enriched REE abundances, the spinel-olivine components and a small (1 cm) grain of lherzolite xenolith inclusion in the Al-augite megacryst suggest a complex history of the Spx—3 specimen. Measurements carried out by optical microscopy, X-ray and electron diffraction, INAA and electron microprobe analyses have revealed textural, chemical and REE characteristics of the Spx—3 specimen's mineral components from which its formation can be reconstructed. They indicate its origin in an approximately 15% partial melting of the primary lherzolithic mantle, subsequent lherzolite inclusion transfer to the place of crystallization of this melt in the deep crust or upper mantle region where a cooling process in a concentration gradient field resulted in Liesegang layering of olivine (and partly spinel) in the megacryst producing plutonic massivum, to the final stage of uplift from the upper mantle — lower crust region by volcanic eruption which delivered the specimen to the surface and embedded it in the alkali basalt tuff of Szentbékkálla about 5 million years ago.

INTRODUCTION

REE abundance and corresponding petrological and mineralogical investigations on the Szentbékkálla series of peridotite inclusions have revealed genetic relationships between different upper mantle and lower crust inclusions and host alkali basalts (Sz. Bérczi, J. Bérczi, 1986). During this work a unique specimen was found at Szentbékkálla by one of our students on mathematics and technology, L. Kőszegi in 1984. Initial inspection showed that the specimen was the size of a fist clinopyroxene megacryst nodule broken into four pieces. The interesting and puzzling aspect of its otherwise megacryst appearance was about dozen thin, parallel ribs protruding from its smooth rounded surface. The ribs were visible in low angle incident light. However, a centimeter sized peridotite inclusion was embedded in the speciment together with many smaller and larger spinel crystals. The Spx—3 speci-

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men seemed so unique and promising that the four fragments were given to colleagues working in different Departments of the Eötvös Loránd University and the Budapest Technical University who had different measuring facilities. The results of our investigations are summarized on the following pages.

DISCOVERY SITE AND SPECIMEN DESCRIPTION

The Spx—3 specimen was found on the Church hill of Szentbékkálla, North edge of Káli-Basin, Nort-West Balaton Region, Hungary. The specimen was an inclusion protruding from the basalt tuff in a cart-track and hence free of grass vegetation *(Fig. 1.)*, The specimen was extracted in four pieces as shown in *Fig. 2*. Preli-
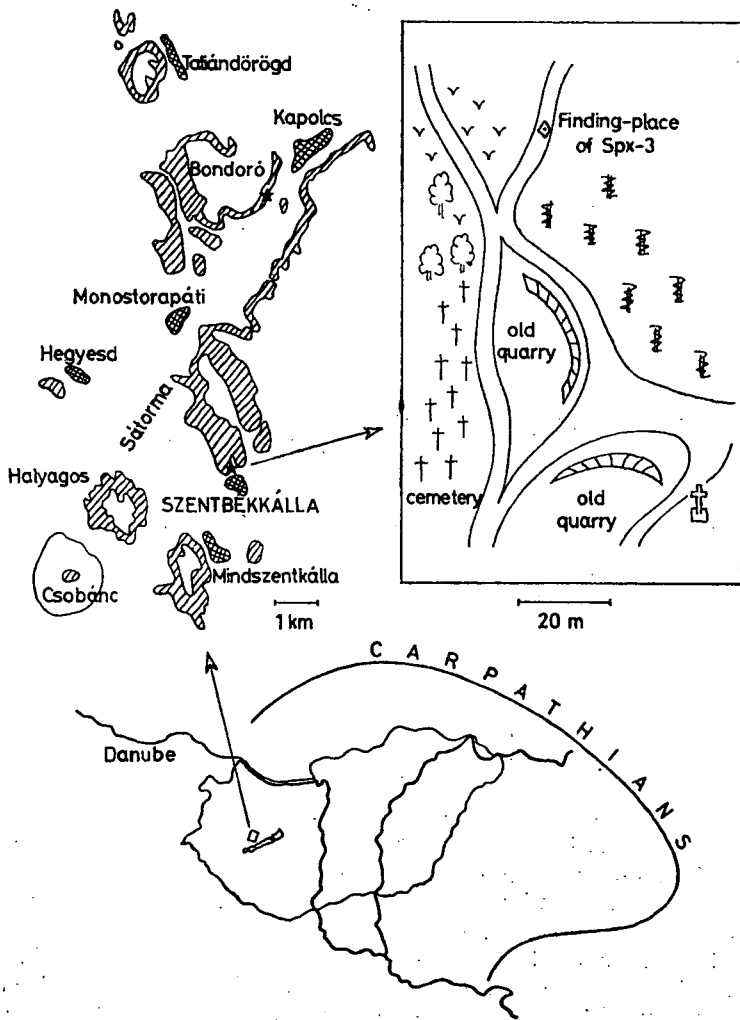

Fig. 1. The site of discovery of Spx—3 sample and that of samples chosen for comparison from Szentbékkálla and Kapolcs, NW Balaton region, Hungary. The upper left part shows piroclastic rock from the geologic map of Lóczy (1920)

minary observations in low angle incident light revealed the almost equidistant rib system of protruding mineral layers running parallel to each other. One of the fractures which divided the sample in to four parts occurred along one of these layers (that separating A +B and C +D pairs in *Fig. 2.*). On the surface of this fracture could be seen olivine crystals 0.1—0.5 mm in size which populated and so formed the layers. As well as these parallel layers, black spinels arranged along the layers also protruded from the nodule although not so densely as the olivines. Moreover a rounded, 1 centimeter sized lherzolite inclusion was embedded in the specimen (fragment C, *Fig. 2.*).
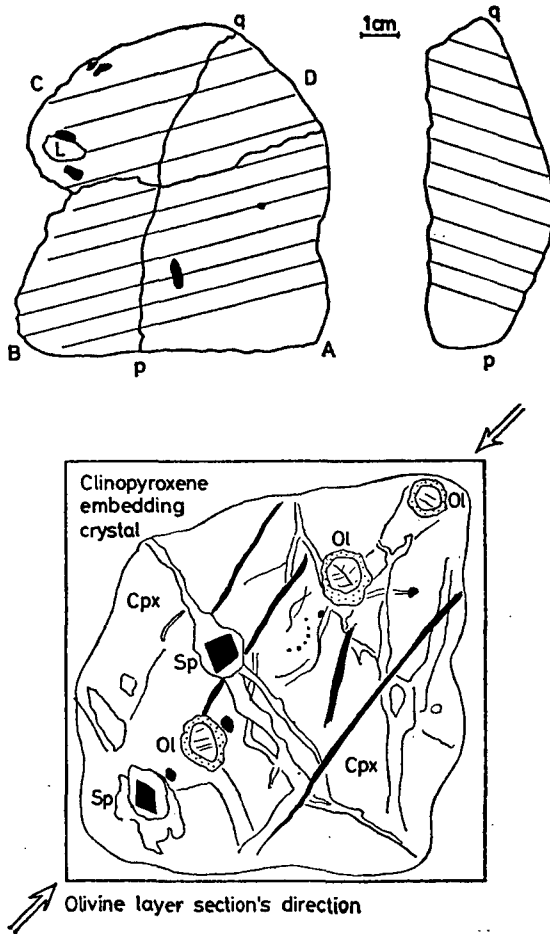

Fig. 2. The fist sized Spx—3 specimen. a The 14 parallel bands of scattered olivine grains arranged along planes protruding from the surface of the sample. The protruding lherzolite (L) grain in C fragment and some of the large spinels in A and C fragments are also drawn in. b The cross section of the Spx—3 sample at the p-q breaking-line shows the position of olivine layers. c Sketch of a cut layer region enlarged from a thin section. Arrows indicate the position of the olivine layer and show that olivine grains rarely populate the planes and that spinels accumulated along these planes.

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The mineral phases which constitute the sample have a very strange arrangement. The host clinopyroxene exhibits no optical zoning and twinning, but effects of mechanical strain. The whole large megacryst of Spx—3 seems to be homogeneous, and according to its optical characteristics it is augite.

The olivine and spinel occur as poikilitically embedded minerals arranged near to the direction of cleavage planes of the host clinopyroxene megacryst *(Fig. 2.)* Sometimes these accompanying minerals form aggregates along the planes. The olivine grains are between 0.1—0.5 mm in size and are isometric and euhedral according to their appearance in thin sections. No mechanical twinning or other deformations can be seen in the olivines, except in one larger grain 2 mm-s in diameter. The cleavage planes are clearly visible and there is no sign of their metamorphic transformation.

The spinel grains are generally 0.2—0.9 mm in size but exceptionally crystals of spinel larger than 5 mm-s occur in the Spx—3 sample. On the basis of their triangular or square sections the spinel grains are euhedral and on the basis of their uniform dark-grey color they seem to be homogeneous.

Textural types of Hungarian basic-ultrabasic xenoliths have been reviewed by EMBEY—ISZTIN (1984). The Spx—3 sample belongs to the black pyroxene (Al, Ti-augite) group of xenoliths but does not belong strictly to any of the textural types classified there because of its unique layered texture and its almost decimeter sized single crystal structure. Our sample can, however, be related (interpolated) to two textural types in this classification: clinopyroxene megacryst xenoliths and composite xenoliths.

The general assertions on black clinopyroxene group xenoliths about their almost monomineralic composition, the Al-augite composition of clinopyroxene and the presence of scattered olivine and spinel accessory accompanying minerals are valid for the Spx—3 sample. The poikilitical enclosing of the more frequent smaller, rounded or subhedral olivine and the rare but larger, mostly euhedral spinels by the megacryst clinopyroxene in Spx—3 shows a close relation to the Szg—3005 Embey—Isztin's sample. However in Spx—3 the olivines are evenly distributed along the parallel planes and spinels are also scattered rather uniformly, frequently accumulating along the olivine layers contrary to the unordered texture of Szg—3005 sample.

The embedded lherzolite nodule also relates Spx—3 to the composite xenolith EMBEY—ISZTIN's group, but the extraordinary characteristic of Spx—3 specimen is the ordered texture with the strictly parallel olivine grain populated planes, which might have been formed by a resonance effect (standing weaves) or by special causes during the very quiet, large single crystal growing crystallization process. We have found only one (probable) similar sample from the literature: SKEWES and STERN (1979) reported what we assume to be similarly textured clinopyroxene megacrysts from the Palei-Aike lavas in Chile. These "megacrysts contain opaque inclusions *regularly distributed along curved planes*". We propose forming a distinct subgroup of such black clinopyroxenes with an ordered poikilitical texture and enlarge with this subgroup EMBEY—ISZTIN's classification of Hungarian xenoliths, of which such types with unusual texture may preserve the detailed structure of the process of magmatic events of melts on their way from the mantle to the surface.

The estimated mineral composition of Spx—3 is as follows: clinopyroxene — 94%, spinel — 3%, olivine — 3%. The compositional data of the microprobe analysis are shown on Table 1. Compositional zoning of the main elements — which could have been reflected by the supposed Liesegang layering in Spx—3. — has not been observed for any of the mineral components of Spx—3, in accordance with the optical characteristics of these minerals.

TABLE 1.

Comparison of maior element composition of spinel and clinopyroxene components of two Spx and one Cpx samples (Microprobe analysis by K. G. SOLYMOS and CS. SZABÓ)

	Spx—A		Spx—3		Cpx—Kap
	cpx	spinel	cpx	spinel	cpx
SiO_2	49.22	n.a.	48.4	n.a.	48.84
TiO_2	1.17	0.49	0.99	0.40	0.97
Al_2O_3	7.87	61.89	9.25	60.90	7.63
FeO_t	6.24	16.50	5.81	16.80	5.82
MnO	0.18	0.06	n.a.	n.a.	0.10
MgO	15.10	21.41	15.50	20.90	16.12
CaO	18.64	n.a.	18.85	n.a.	18.94
Na_2O	1.24	n.a.	1.48	n.a.	1.17
Cr_2O_3	n.d.	n.a.	n.d.	n.a.	0.12
Sum	99.66	100.44	99.93	99.0	99.71
$\dfrac{10 \ Mg}{Mg+Fe}$	81.2	69.8	82.6	68.9	83.2

n.a. = non analysed, n.d. = non detected.

The clinopyroxene of Spx—3 appears to be very near in composition to the two earlier spinel-pyroxenite and clinopyroxene samples (BÉRCZI, BÉRCZI 1986) in the Ca—Mg—Fe diagram *(Fig. 3)*. These three clinopyroxenes with their average $Ca_{42}Mg_{48}Fe_{10}$ metal composition seem to be richer in Mg and poorer in Ca than the Australian, French and Nógrád—Gömör (Medves) Hungarian black clinopyroxene megacrysts which have been investigated in more detail earlier (WILKINSON 1975, WASS 1979, LIOTARD et al. 1988, DIENES 1979) so our NW Balaton regional samples can be found in the endiopside region of the Ca—Mg—Fe pyroxene triangle.

The high Al_2O_3 and Na_2O content of clinopyroxene suggests a considerable amount of jadeite (10.2 percent) and $CaAlAlSiO_6$ (6.4 percent) end members. Clinopyroxenes characterized by a high Mg value (about 80) a greater proportion of jadeite and $CaAlAlSiO_6$ components and a low Cr content are widespread in alkali basalts (WHITE 1966, BINNS et al. 1970, IRVING 1974.) and such clinopyroxenes are called Al-augite after WILSHIRE and SHERVAIS (1975).

Most of the authors (for example AOKI and KUSHIRO 1968, AOKI 1971, SKEWES and STERN 1979, WASS 1979, IRVING and FREY 1984,) have considered clinopyroxene inclusions embedded in the host magmatite as cognate precipitates in equilibrium with the host basalt under high pressure and temperature conditions. Using to this model we could estimate the two thermodynamic parameters of the source region of our three samples. In the AOKI—KUSHIRO diagram of Al^{IV}/Al^{VI} ratios *(Fig. 4)* the
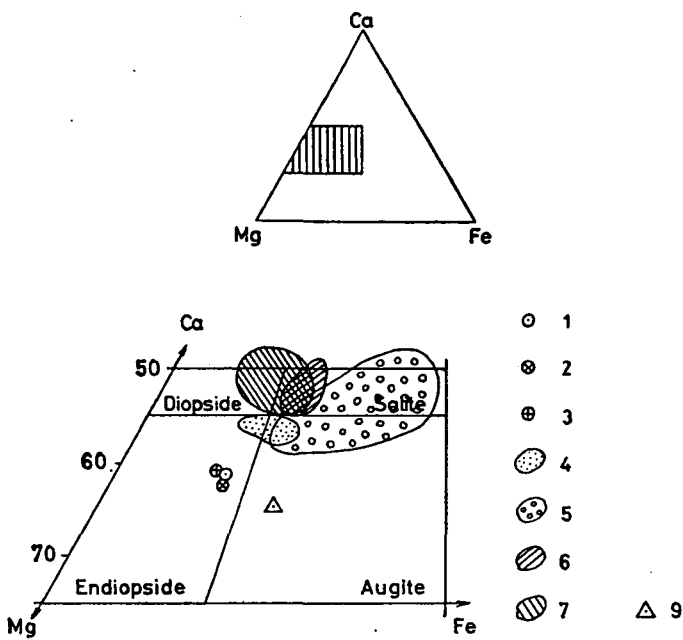
Fig. 3. The position of composition of clinopyroxenes from the Spx—3 sample, from two earlier NW Balaton region samples and the compositional regions of the Australian, French and Chilean clinopyroxenes used for comparison in the Ca—Mg—Fe diagram. (**1.** — cpx from Spx—3. **2.** — cpx from Spx—A. **3.** — cpx of Cpx—Kap. (2.—3. from BÉRCZI, BÉRCZI 1986), **4.** black cpx megacrysts (WILKINSON, 1979.). **5.** French cpx megacrysts (LIOTARD *et al.* 1988), **6.** French cpx megacrysts, **7.** Australian cpx megacrysts (6.—7. from WASS, 1979), **9.** — Chilean cpx (SKEWES, STERN 1979.).)
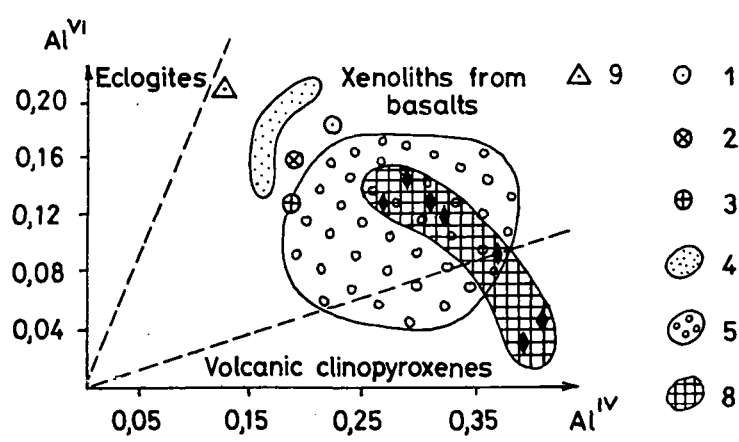

Fig. 4. The Al^{IV}/Al^{VI} positions of clinopyroxenes from our three samples (1., 2. and 3.) and the region of the (4.) Australian (WILKINSON, 1979) and (5.) French (LIOTARD *et al.* 1988) comparison samples in the AOKI—KUSHIRO diagram. (The numbers refer to the indications used on *Fig. 3.*) According to their positions the Nógrád—Gömör samples from Medves (DIENES, 1968) have a partly low pressure origin (8.). The Chilean clinopyroxene megacryst (9.) is the only sample which may have similar ordered texture to that of Spx—3. (The 9. point from SKEWES, STERN 1979.)

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clinopyroxenes of Spx—3, Spx—A and Cpx—Kap samples fall within the 10—15 kbar pressure range. On the basis of the experimental data of THOMPSON (1974) the temperature at their source region would probably have been in the 1200—1300 °C range. (THOMPSON's data confirm the pressure estimation of 10—15 kbar from the AOKI—KUSHIRO diagram, too.)

The composition of olivine crystals of Spx—3. (Table 2) can be compared to two different earlier measurements. The olivine of Spx—3. is similar in composition to the megacrysts of alkalic basalts from the Nógrád—Gömör region (ÁRGYELÁN 1987, FORGÁČ et al. 1986.) which are always richer in Fe than the olivine phenocrysts of the basalt. This fact agrees with the results of WILKINSON (1975) who also concludes that olivine connected to Al-augite shows similarities in its chemical character to megacrysts.

TABLE 2

Comparison of the composition of olivines from Spx—3, from wehrlite
and from basalts as megacrysts
(1. and 2. K. G. SOLYMOS and Cs. SZABÓ, 3. ÁRGYELÁN (1987) average of 7 samples.
4. FORGÁČ et al. (1986) average of 4 samples; 1.—2. Szentbékkálla, 3.—4. Nógrád-Gömör)

	Spx—3	Wehrlite	Av. Megacrysts	Av. Megacrysts
SiO_2	39.4	38.31	39.6	39.5
Al_2O_3	0.10	0.07	0.03	0.04
FeO	16.0	13.90	14.7	17.8
MnO	n.a.	0.27	0.20	0.36
MgO	43.6	47.12	45.3	42.8
CaO	0.15	0.05	0.21	n.a.
Sum	99.25	99.72	100.04	100.5
Mg value	82.9	8.58		

n.a. = non analysed.

The second sample for comparison is from our earlier work (BÉRCZI, BÉRCZI 1986): a wehrlite sample. Not only the Fe enriched composition of its olivine, but also its REE abundance connects this sample more closely to the clinopyroxenite group of inclusions from the Szentbékkálla Series.

Because of its very high Al_2O_3 content, the spinel of Spx—3. is rich in the $MgAl_2O_4$ phase, and so it is the same in composition as spinels of our earlier spinel-pyroxenite samples, Spx—A and Spx—D (BÉRCZI, BÉRCZI 1986). The X-ray diffraction measurements carried out with associated salt crystal also confirm this high $MgAl_2O_4$ content: the grid-unite from the (220) lattice's reflection is 0.812 nm, from the (111) planes it is 0.813 nm and from the (311) planes' reflection it is 0.806. The average of 0.810 nm is very near to the 0.808 nm for true spinels.

REE ABUNDANCES OF CLINOPYROXENE AND SPINEL FROM SPX—3
AND FROM OTHER SPINEL-PYROXENITE NODULES

The chondrite normalized REE abundance pattern of clinopyroxene and spinel of Spx—3, that of Spx—A and Spx—D (BÉRCZI, BÉRCZI 1986) and that of Spx—7 and Spx—Kap (this work for comparison) all have similar shapes in two distinct ranges *(Fig. 5)*. The clinopyroxenes are enriched about 10 times as compared to the

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chondritic values, the spinels are on chondritic abundances except in the light REE region, where they are enriched in La to the clinopyroxene level. The run of the REE curves of clinopyroxenes show little HREE depletion and thus a modest decreasing trend from light to heavy REE *(Fig. 5)*. The global range of spinel-pyroxenites is between the range of basalts and that of lherzolites *(Fig. 6)* in the Szentbékkálla series and it is the smoothest of them. Our REE values are in close agreement with the data and range for clinopyroxenes given by IRVING and FREY (1984), (except that our samples exhibit a little negative Eu anomaly proving the differentiated, partly exhausted nature of the source region). From the fact that the clinopyroxene/host basalt distribution coefficients at these authors fall within the range of experimentally investigated clinopyroxene/melt ratios in the case of middle and heavy REE, the conclusion that clinopyroxene megacrysts were in equilibrium with their host basalts is widely accepted (e.g. LIOTARD *e t al.* 1988). But IRVING and FREY in their paper (1984) emphasize that there only *may* be equilibrium between clinopyroxenes and host basalts and they did not exclude other kinds of genetics as well.

Contrary to the rather smooth (compared to chondritic) range of clinopyroxenes from spx-samples the corresponding contribution coefficient range *(Fig. 7)* clearly reflects only the inverse of the LREE enrichment of basalts. The experimental data do not fit those of clinopyroxenes in the LREE region alone. A smoother REE range of host melt could give a better approach to the calculated distribution coefficients:
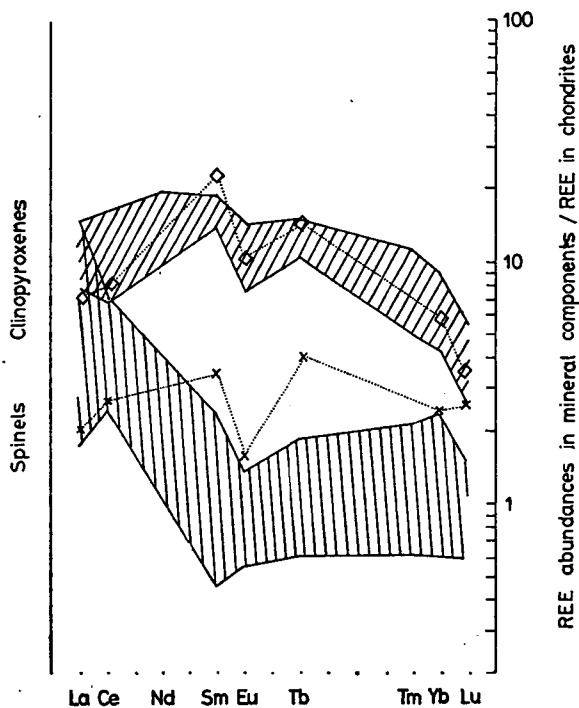

Fig. 5. Chondrite normalized REE abundances of the clinopyroxene and spinel components of Spx—3 and the REE abundance range of these two mineral fractions from the NW Balaton region Hungarian spinel-pyroxenites.) ∇ — cpx of Spx—3, × — sp of Spx—3)
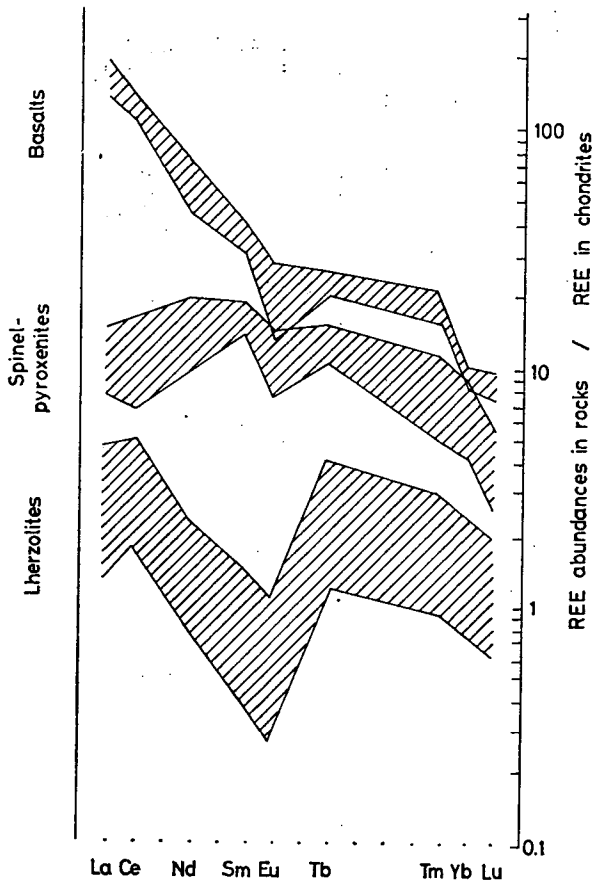
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Fig. 6. The REE abundance range of spinel-pyroxenites (this work) between the REE ranges of lherzolites (below) and alkalic basalts (upper) (BÉRCZI, BÉRCZI 1986). The spinel-pyroxenite range is represented with the range of clinopyroxenites, because of their one order of magnitude greater REE content, than that of spinels, except in the LREE region, where they shift of the range to higher values

this may be a 15—20 percent partial melt of the peridotitic mantle which should produce a more even distribution of REE because of the more extended partial melting. This model is in accordance with our earlier suggestion (BÉRCZI, BÉRCZI 1986).

LIESEGANG LAYERING OF OLIVINES IN SPX—3

The periodic precipitation phenomenon is known since the work of R. E. LIESEGANG (1896). The periodic precipitation pattern (PPP) was later commonly called Liesegang phenomenon or Liesegang-rings from its experimental reproduction in Petri dish. The PPP may be formed in the presence of concentartion gradients of two reactants in a background system (liquid or solid state). In their physical-chemical experimental studies S. C. MÜLLER, SHOICHI KAI and J. ROSS (1982) have

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TABLE 3

REE content in mineral components of Hungarian s inel-pyroxenite inclusions (data in ppm).
(+ measurements by J. BÉRCZI, X measurements by ZS. MOLNÁR)

| | Szentbékkálla | | | | | | | | Kapolcs | |
| | Spx—A+ | | Spx—D+ | | Spx—3+ | | Spx—7[x] | | Spx—Kap[x] | |
	Sp	Cpx	Sp	.Cpx	Sp	Cpx	Sp	Cpx	Sp	Cpx
La	4.3	2.9	0.5	3.2	0.6	2.1	1.8	2.50	3.28	4.85
Ce	5.7	5.8	2.0	8.9	2.1	6.6	n.a.	n.a.	n.a.	n.a.
Nd	<5.0	7.0	<2.0	5.8	n.a.	n.a.	n.a.	n.a.	b.a.	n.a.
Sm	0.40	3.34	0.09	3.88	0.7	4.7	0.45	3.2	0.51	3.1
Eu	0.10	0.98	0.04	0.04	0.12	0.75	n.a.	0.98	n.a.	0.74
Tb	0.09	0.55	0.03	0.7	0.2	0.7	n.a.	n.a.	n.a.	n.a.
Tm	0.07	0.16	0.02	0.36	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Yb	<0.5	<0.5	<0.5	<0.5	0.4	1.0	≦0.2	1.07	≦0.4	0.77
Lu	<0.05	0.17	<0.02	0.08	0.08	0.11	≦0.03	0.10	≦0.05	0.11
Cr.	240	29	430	44	91	30	760	38	1040 ·	270

n.a. = non analysed.
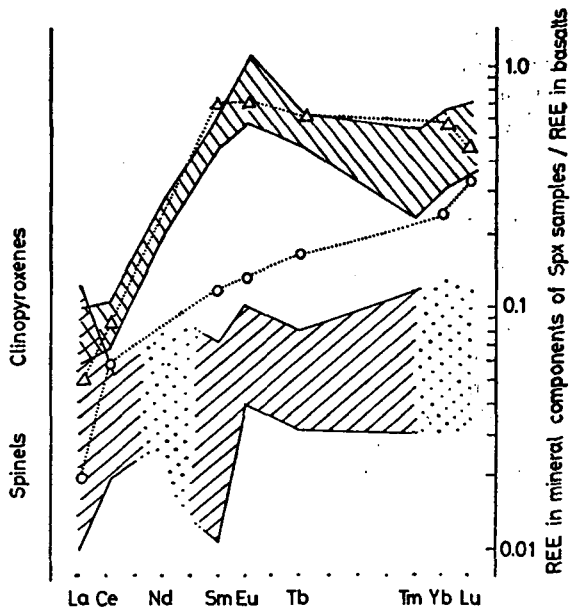
Fig. 7. Clinopyroxene/host basalt and spinel/host basalt distribution coefficients ranges for the mineral components of Hungarian spinel-pyroxenites. (△ — distribution coefficients for the clinopyroxene of Spx—3, ☉ — distribution coefficients for the spinel of Spx—3.)
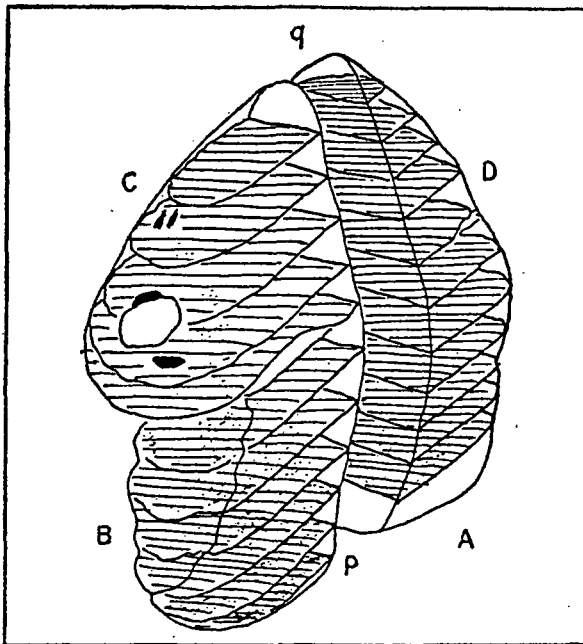

Fig. 8. Perspectivical view the Liesegang layers in the Spx—3. clinopyroxene megacryst inclusion.

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concluded the dependence of PPP on the initial concentrations of reactants. According-ing to their work the relevant parameters in the formation of PPP were the initial concentration difference of reactants and the initial ion-concentartion product of reactants. When both of these two parameters were large they have got large number of thin precipitating layers in their experiments. On the basis of the studies of S. C. MÜLLER *et al.* a preliminary inference can be given here to the PPP formation in our Spx—3 specimen *(Fig. 8)*.

In the parent liquid of Spx—3 SiO_4^{2-} and Mg^{2+} ions might have been the main actors of the PPP process. Large initial concentration difference might have been produced by large Mg^{2+} and small SiO_4^{2-} concentration (at the sides of) or gradient (through) the PPP region. The more agile Mg^{2+} ions could have moved — and con-centrated by the colloid formation and the subsequent focusing mechanism (MÜLLER *et al.* 1982.) — against the small SiO_4^{2-} gradient in the liquid to form the Mg_2SiO_4 layers in the extended clinopyroxene environment. (In a similar process Mg^{2+} and Al^{3+} ions — which might have been in excess over the average clinopyroxene back-ground — could have formed the $MgAl_2O_4$ spinels against the small oxigene gradient.) These suggestions should be studied in more details if a collection of different Liese-gang layered clinopyroxene megacryst specimens makes comparison possible.

SUMMARY

Texture, composition, chemistry and REE abundances of mineral components of Spx—3 megacryst inclusion from alkali basalt of Szentbékkálla prove its origin by crystallization from melt.

1. The primary melt nature of the liquid of Spx—3 is proved by an embedded lher-zolite xenolith in the specimen.
2. The melt might have been produced by a 15—20 percent partial melting of the parental mantle peridotite. (according to the REE concentrations)
3. During crystallization olivine and spinel might have been precipitated together with or from clinopyroxene (as shown by their equal Mg-values in Table 2 and 3).
4. Concentration gardients might have resulted in Liesegang layered precipitation arrangement of olivine (and partly the spinel) crystals.
5. According to the compositional data of clinopyroxene of Spx—3 the specimen crystallized in the 10—15 kilobar pressure and the 1200—1300 °C temperature ranges.
6. The crystallization in deep was the first stage of uplifting of the specimen from a mantle region. The second stage was the alkali basaltic eruption which embedded the specimen into the basalt tuff of Szentbékkálla.

CALL FOR COOPERATION IN LIESEGANG LAYERED MEGACRYSTS

We ask investigators of mantle and lower crust xenoliths to kindly inform us if they have measured Liesegang layered structures (PPP) in megacrysts similar to those in our Spx—3 specimen. A comprehensive comparison of these Liesegang layer-ed megacrysts may give us a new method for determination of lower crust — upper mantle conditions where these megacrysts have been formed from partial melts of mantle peridotites.

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