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## META-TORBERNITE

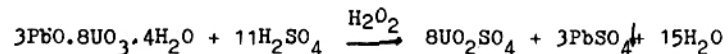
R.F.C.Vochten, G.A.Remaut, Antwerpen, Belgium

## Transformation of curite into meta-torbernite.

As torbernite is often found in association with curite, as it is the case at Shinkolobwe in Zaire, it is our intention to investigate the transformation of curite into torbernite.

Direct transformation of curite into torbernite results in an inhomogeneous material.

By treating curite with  $H_2SO_4$  2N in the presence of small quantities of  $H_2O_2$  at  $70^\circ C$ , the following reaction takes place:



$H_3PO_4$  10% is neutralized with solid  $CuCO_3$  and added to the acid  $UO_2SO_4$  solution; after several days at  $70^\circ C$ , one obtains spherical aggregates of meta-torbernite.

Solutions which contain free  $H_3PO_4$  delay the crystallization, resulting in badly crystallized specimens.

X-ray powder diffraction, optical data and D.T.A. identify the crystals as meta-torbernite.

Some photographs of the grown crystals are added to the abstract. Scanning electromicrographs show that the spherical aggregates consist of intergrown plates.

## Zeta-potential of meta-torbernite in aqueous suspension.

The zeta-potential of meta-torbernite is measured as function of the pH of the suspension, at different temperatures.

The effect of the addition of  $H_3PO_4$ ,  $H_2SO_4$  and HCl on the surface-charge of meta-torbernite has been investigated.

The zeta-potential is calculated from the electrophoretic mobility of the particles.

Taking into account the structure of meta-torbernite consisting of negative charged  $(UO_2PO_4)_n^{n-}$  sheets, on which copper is bounded, one would expect the zeta-potential to be negative.

However, our measurements indicate a positive value for the zeta-potential for temperatures from 3°C to 40°C and pH-values from 2 to 9.

The zeta-potential decreases with decreasing temperature and decreasing pH-values, but the point of zero-charge is not obtained. BaSO<sub>4</sub>-crystals have a positive zeta-potential, caused by a release of SO<sub>4</sub><sup>2-</sup>-ions at the surface. In analogue, one can explain the positivity of the zeta-potential of meta-torbernite by a release of PO<sub>4</sub><sup>3-</sup>-ions from the (UO<sub>2</sub>PO<sub>4</sub>)<sub>n</sub><sup>-</sup>-sheets.

For pH-values above 7, the UO<sub>2</sub><sup>2+</sup>-ions are surrounded by OH<sup>-</sup>-ions, which reduce the interaction between the PO<sub>4</sub><sup>3-</sup>-ions and the UO<sub>2</sub><sup>2+</sup>-ions, allowing a release of the PO<sub>4</sub><sup>3-</sup>-ions from the crystal lattice. As a consequence, the zeta-potential becomes more positive. An increase of temperature results also in a higher positive value of the zeta-potential.

For decreasing pH-values, a decrease of the zeta-potential can not be explained by protonation of the PO<sub>4</sub><sup>3-</sup>-ions in the crystal lattice. This result is probably caused by an equilibrium between PO<sub>4</sub><sup>3-</sup>-ions in solution and the PO<sub>4</sub><sup>3-</sup>-vacancies in the lattice. Our measurements show that the zeta-potential is influenced by the presence of the following ions: PO<sub>4</sub><sup>3-</sup>, SO<sub>4</sub><sup>2-</sup>, and Cl<sup>-</sup>.

This is related to the probability of these ions for entering in the vacancies at the surface of the lattice.

The surface conductivity is being calculated from the zeta-potential in acid suspension.

## MULTIPLICITY OF PYROXENE-AMPHIBOLE REACTION MECHANISMS

D.R. Veblen, P.R. Busek, Arizona, USA

Recent results from high-resolution transmission electron microscopy and single crystal X-ray studies indicate that there are several mechanisms by which transformation reactions occur among the pyroxenes, amphiboles, and micas (including talc).

At Chester, Vermont, USA, anthophyllite and cummingtonite in a metamorphosed ultramafic body have partially reacted to talc [(100)<sub>An</sub>//(001)<sub>Tc</sub>], first by transforming to chesterite and its monoclinic analog [(Mg,Fe)<sub>17</sub>Si<sub>20</sub>O<sub>54</sub>(OH)<sub>6</sub>], which contain rigorously alternating double and triple chains, and then to jimthompsonite and clinojimthompsonite [(Mg,Fe)<sub>10</sub>Si<sub>12</sub>O<sub>32</sub>(OH)<sub>4</sub>], which have only triple chains. These reactions take place by growth of structurally disordered intermediates having a variety of chain width; these disordered chain silicates reorder by the passage through the structure in the c direction of stacking faults having displacement vectors of ~4.5 Å multiples in either the a\* or b\* directions. Another important anthophyllite → talc mechanism in this occurrence involves simple replacement with (210)<sub>An</sub>//(001)<sub>Tc</sub>; in this case, talc apparently grows by migration of ledges having the height of one or more T-O-T sheets.

Pyroxene amphibole replacement (uralitization) reactions take place either by growth of double, triple, and wider chains which later reorder to amphibole, or by oriented bulk replacement of pyroxene by amphibole along a reaction front of undetermined structure. The amphibole thus produced may further react toward mica composition by either of the two mechanisms noted above for anthophyllite talc. In places, pyroxene may also react directly to mica structure, or to mica via a triple-chain intermediate.

Planar amphibole lamellae parallel to (010) are common in pyroxenes formed in high pressure environments. These may arise by hydration reaction. Alternatively, they may form by exsolution of metastable oxyamphibole from pyroxenes rich in M-site vacancies or by a combination of exsolution and hydration.

INVESTIGATION OF ION EXCHANGE OF ZEOLITIC MINERALS IN  
THERMOGRADIENT FIELD

L. Filizova, G.N. Kirov, V. Dragostinova, Sofia, Bulgaria

There has been investigated the distribution of alkaline and alkaline-earth elements in columns of zeolitic minerals under thermal gradient, exchanging in water solutions of nitrates. The relationship zeolite : solution = 1.3:1, and the alternation of temperature is about 100 °C.

The distribution of exchanged cations in some zeolitic structures is ascertained to proceed in different ways. For instance, the contents of Cs in a cold zone of column of clinoptilolite is 8 times more than in a hot one. And in a column of mordenite-it is 3 time more in a hot zone.

The obtained results are explained in connection with the ion solvational properties and the structural peculiarities of zeolitic minerals.

Their comparison with natural zeolitic deposits tufogeno-sedimentary origin permits to give some analogues. The role of these factors is discussed for the practical use of ion-exchanged properties of zeolitic minerals.

COSMIC MINERALOGY

## MINERALS OF METEORITES - INDICATORS OF COSMIC EVENTS

A.K. Lavrukhina, G.V. Baryshnikova, Moscow, USSR

Evidences of the shock effects and metamorphism in minerals of eight ordinary chondrites: Krymka (L 3), Saratov (L 3, 4), Elenovka (L 5), Kyushu (L 6), Tieschitz (H 3), Ochansk (H 4), Pultusk (H 5) and Doroninsk (H 6) are considered in the report. In the investigated chondrites the following evidences of shock metamorphism have been revealed: fracturing (kataclas) of silicate grains (Saratov, Elenovka, Tieschitz), kataclas and undulatory extinction in grains of olivine and pyroxene (Krymka, Ochansk), mosaic structure of olivine grains (Ochansk), strong fracturing planar structure in olivine, abundance of maskelinite (Kyushu). On the basis of criteria established by Carter et al., (1968) these meteorites have been subject to shock deformation from 150 to 400 kbar.

Chondrules in the Elenovka (L5) chondrite have been found in which fractures are observed not being continued in the matrix. This indicates that the given chondrules had undergone shock effect before accretion. In the nonequilibrium chondrites Krymka (L3) and Tieschitz (H3) together with prevailing clinopyroxene the presence of a little amount of orthopyroxene has been revealed mainly in the so-called irregular microporphyritic and granular chondrules. Such orthopyroxene is in nonequilibrium ordinary chondrites an evidence of preaccretion recrystallization of these chondrules.

Besides that in the chondrite Elenovka the presence of martensite-like structures and of a cloudy taenite in taenite-pleistite particles has been revealed, what evidences on the absence of a high-temperature metamorphism of this meteorite after accretion.

EFFECTS OF IRRADIATION ON METEORITE AND LUNAR SOIL MATTERS  
BY COSMIC RAYS ACCORDING TO TRACK INVESTIGATION DATA

N.N. Korotkova, L.L. Kashkarov, L.I. Genaeva, A.K. Lavrukhina  
Moscow, USSR

In this report are given the results of detailed investigations of characteristics of tracks caused by irradiation of individual crystals of olivine, pyroxene, feldspar from cores of lunar soil (Luna-16, Luna-20, Luna-24) and of olivines from the carbonaceous meteorite Allende by nuclei of elements of the iron group of low-energetic ( $1 \lesssim E \lesssim 100$  MeV/nucleon) cosmic rays. The obtained characteristics of tracks in crystals of lunar soil (values of track density, regularities in track distribution a.o.) allowed to find out some parameters of irradiation of the Solar system bodies by low-energetic cosmic rays at a distance of 1 a.u. from the Sun during various time epochs, from the present up to the being remote from it by billions of years. Comparison of track characteristic in lunar minerals with corresponding characteristics for crystals from meteorites enriched in light inert gases (for example, Weston, Kapoeta, Fayetteville) allowed to interpret the meteorite data from the point of view of irradiation of individual grains of minerals on the surface of protoplanets having existed in the early solar system. Parameters of preaccretion irradiation by low-energetic cosmic rays being similar for crystals of meteorites rich in gases and of the lunar surface noticeably differ from those for carbonaceous meteorites which moreover have lower contents of inert gases. Olivines and pyroxenes from the carbonaceous meteorites Orgueil (type C1), Murray, Murchison, Mighei, Nogoya (type C2), Allende (type C3) our data have lower irradiation doses ( $10^7$ - $10^8$  tr/cm<sup>2</sup>) a lesser amount of irradiated grains in the sample (1-10%); little irradiation duration and other signs indicating substantial different irradiation conditions than the conditions on the surface of the planetary body being similar to the lunar one. Discovered by us on the ground of track characteristics in olivine crystals from meteorite Allende the form alteration of the energetic spectrum in comparison with the modern spectrum may witness about the irradiation of mineral grains in the expansive shell of the Supernova.

ASSOCIATION OF PLESSITE, TROILITE, NATIVE COPPER IN STONE  
METEORITES, AND ITS USE FOR THE EVALUATION OF THE TEMPERATURE  
OF FORMATION (ACCORDING TO EXPERIMENTAL DATA-

E.G. Osadchiy, Chernogolovka, USSR

Topological analysis of the Fe-Cu-S system and the available experimental data allow admitting that the Fe, FeS, Cu association can exist up to 694°C. Above this temperature bornite will be stable in the equilibrium with Fe and FeS. The Fe, FeS, Cu, Cu<sub>5</sub>FeS<sub>4</sub> association at 694°C and 1 atm, will be nonvariant in coordinates  $f_{S_2}^4$ -T.

However, the application of the calculated data is not quite correct, since chondrites always contain nickel which forms with iron the kamasite and taenite phases and is isomorphously contained in troilite. For determining the possibility of the existence of equilibrium associations of plessite and troilite with copper or bornite and for determining the temperature of copper replacement with bornite it is necessary to have experimental data.

An experiment was conducted in evacuated quartz ampules by the method of the appearance of phase. The reagents used were synthetic troilite, metallic copper and fine shavings of the Sikhote-Alin iron meteorite. The phase composition was determined by X-ray analysis, while the composition of phases by microprobe analysis.

The temperature of transition copper = bornite is 680±5°C. Thus the iron-sulphide component of chondrites has not registered any temperature above 680±5°C. The composition of bornite differs from the theoretical one by a higher content of Fe, S and the presence of Ni; Cu - 61.0; Fe - 12.5; Ni - 1.1; S - 26.1; the sum being 99.7. This points to the existence of a solid bornite-troilite solution and the isomorphous content of nickel in the bornite. The closeness of experimental and calculated data is explained by the negligible difference between solid solutions containing nickel and their ideal.

CHEMISTRY AND MINERALOGY OF THE Pt-METAL-BEARING FREMDLINGE  
IN CARBONACEOUS CHONDRITES

A. El Goresy, Heidelberg, FRG

C2 and C3 carbonaceous chondrites (Allende, Leoville, Murchison) contain white inclusions extremely enriched in the refractory elements Ca, Al and Ti. Other refractory lithophile and siderophile elements (Zr, Hf, Nb, Ta, U, Th, REE, W, Mo, Ru, Rh, Re, Os, Ir, Pt ...etc.) were found to be enriched compared to the abundances of these elements in C1 chondrites. In an attempt to disclose the mineralogy of the refractory siderophile elements and to understand the formational history of the white inclusions numerous inclusions in Allende and Leoville were studied with the electron microprobe and with the scanning electron microscope. Refractory elements were encountered in two distinct types of objects. 1. Objects of very complex mineralogy (Fremdlinge) consisting of silicates, phosphates, sulphides, oxides, FeNi alloys and countless grains of refractory siderophile elements (Ru, Rh, Re, Os, Ir, Pt). Fremdlinge contain also Mo and W as molybdenite and tungstenite. Along with the siderophile refractories volatile elements like Zn, Ga, Ge, As, Sn were also found either in elemental metallic form alloyed with FeNi, or with each other. All phases encountered are present as very fine-grained aggregates indicative of accretion at very low temperatures. The striking feature is the oxidized state of some refractories e.g. Ta, Nb, V, ... etc. suggesting much more oxidizing conditions than accepted for formation during condensation from a primitive solar nebula. 2. Individual metal beads of siderophile refractories (Ru, Rh, Re, Os, Ir, Pt) almost in their cosmic abundance ratios. Surprisingly, these beads in contrast to Fremdlinge also contain Nb, Ta and sometimes Zr in the metallic form. This is indicative of the siderophile behaviour of these elements in some carbonaceous chondrites. Fremdlinge and metal beads do not occur together in the same white inclusion. These features indicate that Fremdlinge and metal beads originated in two different sources.

FORMS OF THE PRESENCE OF PLATINUM GROUP ELEMENTS, GOLD AND SILVER IN THE SARATOV CHONDRITE AS IN A PROBABLE MODEL OF THE INITIAL SUBSTANCE OF THE MANTLE PLATINUM-BEARING ORE-GENERATING SOURCE

L.V. Razin, G.A. Khomenko, Magadan, USSR

In the silicate fraction of the olivine-bronzite chondrite Saratov, accepted in the experiments of A.P. Vinogradov (1961) as a model of the non-differentiated substance of the Earth's mantle, there occur as impurities elements of the platinum group (EPG), gold and silver. They are probably chemical components of pyroxenes, olivines ( $Fe_{15-25}$ ) and brown varieties of glass. These silicates, believed to be formed from melts, accumulate the EPG and Au impurities in minimum amounts ( $0.01 \cdot 10^{-4}$  -  $1 \cdot 10^{-4}$  weight %), while silver - to the greatest degree (up to  $36 \cdot 10^{-4}$ %) in comparison with other minerals of meteorites.

The maximum concentrations of EPG impurities ( $n \cdot 10^{-4}$ %; Pt-16, Pd - 10, Ir - 3.6, Rh - 5; Os - 3.7, Ru - 3.3) and Au ( $8.8 \cdot 10^{-4}$ %) were found in the metallic fraction included by A.P. Vinogradov (1971) among the products of condensation from the steam phase of a protoplanet cloud.

In the ore-silicate chondrules, regarded as a possible model of the initial substance of the mantle platinum-bearing ore-generating source, there were found two groups of the forms of occurrence of Pt, Pd and Au: an absolutely prevailing ( $>99.5$  relative %) impurity and distinctly minor ( $< 0.5$  relative %) mineral proper. The impurity forms of EPG and Au were found in the silicate fraction (brown glass which constitutes 65-80% by volume of chondrules, 5-25% of orthopyroxene, 3-14% of olivine  $Fe_{20-25}$ ), as well as in the metallic fraction (which constitutes 3-32% by volume of the chondrule mass) and in accessory troilite (0.5-3% by volume of chondrules).

For silicate-ore chondrules (consisting up to 65-70% of Fe-Ni alloys) of close minerals composition there has been recognized inverse relation between the content of EPG and Au in them and the size of these occurrences, this reflecting the process of dispersion of precious metals during agglomeration and simultaneous cooling of enlarging chondrules.



## SHOCK METAMORPHISM OF ROCK-FORMING MINERALS

(In the instance of Yanisjarvi astrobleme)

V.I. Feldman, L.B. Granovsky, N.N. Kikishina, Moscow, USSR

The Yanisjarvi astrobleme (Southern Karelia) is about 10 km in diameter and was formed about 700 mln years ago when a meteorite hit the Lower Proterozoic metapelites. Diaplect transformation in quartz, biotite, staurolite and garnet show different behaviour of these minerals under the attack of a shock wave. For their characteristics there have been studied their densities, optical properties, diffraction patterns, infrared spectrums and absorption spectrums in the range of visibility (the latter have been studied in diaplect minerals for the first time).

In quartz there were recognized all the transformations described in literature, this having allowed the use of this mineral for the determination of impact load on coloured silicates paragenetic with it.

A comparison of the behaviour of the above minerals allows one to note the following stages of their transformation under the effect of shock waves: 1) crushing of minerals into blocks (accompanied by a decrease of the intensity of absorption bands in the optical and IR spectrums, peaks in diffraction patterns); 2) decrease of the remote order of structure (manifested in the displacement of the absorption bands in spectrums and of a part of peaks in diffraction patterns); 3) gradual destruction of structure, strain and rupture of a number of bonds, change of energy states of atomic groups in the structure (manifested in the degeneration of the absorption bands in opticals and IR spectrums, change of colouring and pleochroism, disappearance of most of the peaks in diffraction patterns); 4) vitrification of the mineral (up to an X-ray amorphous state) or its dissociation into new phases (biotite).

## SHOCK METAMORPHISM OF CRYSTALLINE BASEMENT ROCKS FROM THE RIES CRATER, FRG

V. Stähle, Heidelberg, FRG

The Ries Crater in the southern part of Germany is a shallow, circular depression of about 24 km in diameter and a depth of 800 m. The crater was excavated by the impact of a stony meteorite in a 500-650 m thick layer of Mesozoic sediments. The sedimentary rocks are underlain by Paläozoic metamorphic or magmatic rocks, building up the bottom of the meteorite crater.

Shock-metamorphosed rocks occur in various proportions and shock degree within the Bunte Trümmersmassen and the Suevite, which form the two ejecta units of the Ries Crater. The complete series of moderately to strongly shocked crystalline rocks are only present in the glass-bearing Suevite breccia. Shocked crystalline rocks of different type were taken from 10 suevite quarries around the Ries Crater. In an attempt to disclose the distribution of the various crystalline rock types around the crater and their degree of shock metamorphism numerous Suevite samples were studied.

The main rock types, known from the crystalline schollen of the Ries Crater were also found in the Suevite. In the present systematic complete series of these rocks from an unshocked to a strongly shocked stage were described: amphibolite, calc-silicate-gneiss, garnet-cordierite-sillimanite-gneiss, hornblende-gneiss, blastomylonite, granite, granodiorite, tonalite, hornblende-flecken-diorite, diorite, redwitzite and hornblende-kersantite.

Based on petrographic evidence, all seven stages of shock metamorphism were documented. In the highest class 7, for example, the strongly shocked rocks consists of incompletely mixtures of mineral melts, except the refractory minerals like sillimanite or apatite.

SYSTEM OF MINERALOGICAL INDICATORS OF FACTORS OF SHOCK  
METAMORPHISM IN GRANITOID ROCKS

A.A. Val'ter, E.P. Gurov, Kiev, USSR

It is suggested to distinguish two principal factors of shock metamorphism of granitoids: the degree of shock compression determining the pulse pressure, the shock and residual temperature and (secondary) the duration of pulse. In the first approximation, the duration of pulse is determined by the time of the passage of shock wave to the back surface of a meteorite, i.e. by the dimensions of the meteorite and its velocity.

The influence of the pulse duration on the degree of the strain of grain was studied in the instance of quartz.  $\omega$ ,  $\tau$  and other systems of planar elements were studied as typical strains.

There has been recognized a non-uniform degree of strain in different minerals (quartz, garnet, feldspars, biotite) within consolidated fragments of granitoids. Investigations of shock fragmentation of grains were conducted by the Herz and Kwide method. In all cases there has been recognized a lower degree of the strain of biotite as compared to that of tectosilicate; this should be explained by higher compressibility of biotite. The result obtained could be partly explained by the form of the dependence of biotite strain on the duration of pulse differing from that of tectosilicates.

The modes of thermal decomposition and shock fusion of a number of minerals were compared using the data of the studies of shock metamorphism and shock fusion of the granitoid targets of Ukrainian craters.

IR- and  $\gamma$ -resonance spectra of massive impactites were obtained. It has been concluded that the value of  $P_{O_2}$  at the formation of impactites is low (less than  $10^{-5}$  atm) and that the tetrahedral cations (Al, Si) are more ordered in glass than  $Fe^{2+}$ .

A new indicator of high residual temperatures has been recognized - a cubic form of  $ZrO_2$  stabilized by impurities.

THE NEXT OBJECTIVE OF SPACE MINERALOGY - CONSTRUCTION  
OF BODY MODELS OF CRYSTAL GROWTH  
(Introductory address)

D.P. Grigoriev, Leningrad, USSR

Minerals in space objects and on Earth, as a reality, are always crystals, i.e. physical bodies with the composition of their own, physical and chemical properties.

The genesis of minerals under any conditions is the process of their formation as crystals, including nucleation, growth stages and phenomena of physical and chemical change in the course of natural development of crystals or under the influence of external forces.

The picture of mineral genesis can be drawn starting with the act of nucleation and up to the completion of crystal formation, just the same as it is at building construction shown in drawings starting with brick laying and up to the driving in of the last nail.

The body models of crystal growth are suggested for skeletal olivine from lunar soil, for eccentric pyroxene chondrules in stony meteorites and for kamasite in Widmanstaettian structure of iron meteorites.

The representation of processes as bodies is the final result of deciphering of the genesis of real minerals, while a record of mineral formation in the form of chemical and phase reactions shows only chemical or physico-chemical aspects of evolution of mineral bodies.

## MINERALOGY OF LUNAR MATTER

V.I. Barsukov, M.A. Nazarov, L.S. Tarasov, Moscow, USSR

The mineral composition of lunar matter is essentially limited, there having been recognized about 50 mineral species and over 40 insufficiently characterized mineral phases. Nevertheless, lunar mineralogy does not constitute, in principle, any new mineral system. It is close to the mineralogy of terrestrial ultrabasic, basic rocks and meteorites. The limited number of lunar mineral species once more emphasizes the importance of water and volatile components in the processes of mineral formation.

The mineralogy of continental and marine areas of the lunar crust is correlated as an early to a late one. The mafic and ore minerals of continental rocks are characterized by excessive magnesianness, while plagioclases contain more anorthite molecules. The minerals of marine rocks are more variable in composition. The variations of the composition of continental minerals are limited; they are close to simple compounds and contain less impurity components. The mineralogical features of continental and marine rocks are summed up in regolith, its complex of minerals characterizing the mineralogy of the outermost active layer of the lunar crust which is several meters thick.

The processes of lunar mineral formation (melting and crystallization of melts, solid-phase transformations, evaporation and condensation) are due to endogenic and exogenic sources of energy. The former are determined by the accumulated energy of Moon accretion and radiogenic heat. The latter are associated with the meteoritic bombardment of the Moon surface, as a principal factor, and with solar insolation.

The formation of lunar anorthosite crust cannot be associated with the phenomena of selective melting and is explained by the global processes of gravitational differentiation, this implying complete melting of the outer zones of the Moon down to the depth of the order of 200 km. The flotation of plagioclase re-

sults in the localization of fusible material in subcrustal zones, this predetermining the possibility of basaltic volcanism and the character of further evolution of the mineral composition of lunar crust in combination with the process of shock-blast reworking of the lunar surface.

## GAS INCLUSION IN GLASSES AND MINERALS FROM THE EXTRATERRESTRIAL SAMPLES

Ju.A. Dolgov, S.A. Vishnevsky, Novosibirsk, USSR

Quantitative gas analyses of the inclusions from minerals of meteorites show appreciable amounts of  $\text{CO}_2$ , CO,  $\text{H}_2$ ,  $\text{N}_2$ , hydrocarbons and rare gases, inclusions in enstatite from Northon County's meteorite homogenize at the temperature range from 1400-1420°C. In accordance with the experimental data those temperatures correspond to real temperatures of crystallization.

Gas inclusions from tectites are divided into 2 groups: inclusions containing gases of mixed (terrestrial-comet) atmosphere, which are discovered in moldavites; inclusions of pure  $\text{CO}_2$  composition similar to Venus atmosphere. Gas pressure in tectite inclusions is 5,000 times smaller than terrestrial atmospheric pressure. The gas composition of tectite inclusions significantly differs from terrestrial atmosphere and is not affected by diffusion during long geological epochs up to 15 m.y. and more

Various composition of gas mixtures was found in inclusions from glass sphaerules and fragments of lunar soil, which were brought to the Earth by Soviet automatic station "Luna-16". Hydrogen is present in all the inclusions. Most of them are vacuum in relation to terrestrial atmospheric pressure and only 2 inclusions were found to have the gas pressure more than 2 times higher than terrestrial atmospheric pressure.

THE SELECTIVE EVAPORATION AS A FACTOR OF THE FORMATION  
OF THE LUNAR SAMPLE COMPOSITIONS

C.P. Florensky, A.V. Ivanov, M.A. Nazarov, O.D. Rode,  
O.I. Yakovlev, Moscow, USSR

The permanent meteoritic impact events on the lunar surface are resulted in the intensive transformation of the lunar matter in relation with the processes of fragmentation, fusion and selective evaporation. These processes are assumed to determine the changes in chemical composition and physical properties of lunar rocks.

The intensity of evaporation of certain components in the impact melts mainly is related with the bulk chemical composition of the melt. The alkalis are found to represent the most volatile elements of the lunar basalts whereas iron and silicon belong to the moderately volatile elements.

The investigation of the lunar samples lead to a conclusion on the vast abundance of selective evaporation processes on the Moon. It was found that in the individual regolith particles the losses of alkalis are up to 90% of the initial content, the same value of silicon is about 30%. In some cases the condensation processes are resulted in the enrichment of the uppermost layers of the surface of the particles by the same elements.

At the recent stage of the lunar history, the changes in the chemical composition as a result of selective evaporation are distinguished only within the uppermost layer of the lunar surface, i.e. regolith. But the meteoritic impact events at the initial stages of lunar geological history are assumed to be more intensive. Apparently the separation of lunar matter in the evaporation process could serve as a significant factor of evolution of chemical and correspondingly mineralogical composition of the exterior parts of the Moon.

MINERALS OF LUNAR SOIL BROUGHT BY "LUNA-24"  
AUTOMATIC STATION

E.V. Sveshnikova, O.A. Bogatkov, D.I. Frikh-Khar,  
N.A. Ashikhmina, T.S. Magidovich, I.P. Laputina,  
M.K. Sukhanov, O.V. Karpova, A.D. Genkin,  
N.G. Udovkina, E.E. Laz'ko, Moscow, USSR

There have been studied +0.20-0.37 fractions of the specimens of lunar soil brought by the automatic "Luna-24" station from depths of 1824 and 924 m in Mare Crisium. The amount of mineral formations that developed or were found only on the Moon's surface (agglutinates, glasses) decreases with depth. The minerals of lunar regolith as compared to the terrestrial ones, are characterized by an abundance of varieties, inclusions of ore minerals, fused surfaces, vitrification, jointing, availability of microcraters on the surface.

Pyroxenes (over 50% of the soil) are mainly monoclinic: pigeonites, augites, ferroaugites, ferrohedenbergites, notable for their considerable heterogeneity, usually of sectoral-block character. Plagioclases (about 20% of the soil) are anorthites with a marked content of iron oxide. A characteristic feature is the non-uniformity of potassium content. Olivines vary widely in composition. Most widespread are olivines that are 80-95% ferruginous. The most frequently occurring ore minerals are chromite, ulvospinel and ilmenite. Chromite is represented by a magnesium-containing variety with varying content: of titanium and aluminium oxides. Ulvospinel, in the form of individual crystals or intergrown with ilmenite, occurs in two varieties: a common one and one enriched in oxides of chromium, aluminium, vanadium. Ilmenite is uniform in composition, in distinction from that brought by "Luna-20" it is impoverished in magnesium, aluminium and vanadium. Chromomagnetite, for the first time in lunar soils, has been found in paragenesis with bronzite and occurred in the form of inclusion in magnesian olivine. There were also examined troilite, pyrrhotite (the first find), cubanite (?), schreibersite and kamasite.

The recognized composition, features of the minerals and rocks of the studied lunar soil witness to being in their majority, associated with basic magma, having been formed in the course of intrusive and effusive processes that were accompanied by a differentiation of melt. An active role in the formation of regolith was played by impact processes and the action of solar wind.

#### ON THE MATERIAL COMPOSITION AND STRUCTURE OF THE ALISKEROVO IRON METEORITE

S.G. Zhelnin, A.A. Pliashkevich, L.N. Pliashkevich,  
S.M. Sandomirskaya, Magadan, Moscow, USSR

The Aliskerovo meteorite was found on 7 July 1977 in Bilibinsk area of Chukotsky national district of the Magadan region, in the valley of the Maingy-Pauktuvaam river, at the boundary between the alluvial depositions and bedrock, at the depth of 7-8m from the surface.

The meteorite has a streamlined wedge-like shape, 440 x 260 mm with a maximum thickness of 110 mm. The regmaglyptic surface relief is rather faint; small projections are smoothed down and distinct traces of rounding are visible, showing that the meteorite was once in a stream channel. The meteorite weighs 58.4kg.

The principal component of the meteorite is nickel-bearing iron represented by 68% of kamasite, 5% taenite and 25% plessite. The recognized impurities were schreibersite and rhabdite - 1-2%, daubreelite, limonite; trevorite and maghamite were not recognized with absolute certainty, troilite was assumed to be present. Very small quantities were found of two unrecognized minerals with low reflectivity.

Composition of the principal minerals is as follows: kamasite - Fe 93.62; Ni 6.49; Co 0.58; total 100.69%; taenite is highly heterogeneous and was located at two different areas - Fe 45.62 65.78; Ni 54.91; 33.06; Co 0.14; 0.58; total 100.67 and 99.42. The schreibersite formula, based on the results obtained from two analyses, appears simplified as Fe, Ni, Co<sub>3</sub>, P<sub>2</sub>.

Chemical composition of the meteorite is as follows: Fe 86.84; Ni 8.98; Co 0.70; P 0.31; SOO.023; Cu 0.02; C 0.78; Bi 0.01; SiO<sub>2</sub> 0.42; H<sub>2</sub>O ± 2.00; total 100.07%. The density of nickeliferous iron is 7.28 g/cm<sup>3</sup>.

On the basis of the above data, taking into account the width of kamasite rods (1-3 mm), the Aliskerovo meteorite can be classified as an octahedrite with a structure transitional from coarse to medium, and according to new classification (Scott and Wasson, 1975) as belonging to the SHV group.

#### STRUCTURES AND CHEMICAL COMPOSITION OF LUNAR GLASSES

A. Tsimbalnikova, K. Yurek, M. Palivtsova, Prague, CSSR

At the instance of selected fragments of lunar glasses and vitreous spherical particles separated from "Luna-16" regolith there have been studied their surface morphology, devitrified structures, and the possibilities of the determination with an electronic probe of chemical composition of individual phases and average composition of the studied glasses.

PARTITIONING OF Cr, Ti AND Al BETWEEN OLIVINE, SPINEL AND  
LIQUID UNDER CONTROLLED  $f_{O_2}$  CONDITIONS

J. Akella, Livermore, USA

The distribution of chromium amongst coexisting olivines, spinels and silicate liquids has been investigated as a function of oxygen fugacity, temperature and cooling rate between 1300 and 1175°C and redox states near the Fe-FeO buffer. The bulk composition in this study was a pyroxene,  $En_{68.5}Fs_{16.5}Wo_{15}$  to which 10 mole %  $CaTiAl_2O_6$  and 2 mole %  $CaCrAlSiO_6$  had been added.

The measured  $D_{Cr}$  (weight ratio of  $Cr_2O_3$  in olivine to that in the liquid) values tend to increase with decreasing  $f_{O_2}$  and decrease with decreasing temperature. However, the data possess no statistically significant structure in terms of these variables. The value of  $0.85 \pm 0.15$  for  $D_{Cr}$  reported by other workers is consistent with our data.

The failure of  $D_{Cr}$  to strongly correlate with a varying redox state leads us also to conclude that the valence state of Cr is not a primary factor responsible for high  $Cr_2O_3$  contents in lunar rocks and minerals, and that these high  $Cr_2O_3$  contents are a reflection of originally high  $Cr_2O_3$  concentrations in the source region of these materials. This observation is in agreement with that of Schreiber and Haskin (1976).

The relative insensitivity of  $D_{Cr}$  to  $f_{O_2}$  results from the fact that under reducing conditions,  $Cr^{2+}$  is the dominant chromium species in the silicate liquid; consequently, large changes in  $f_{O_2}$  are needed to significantly affect the concentration of  $Cr^{2+}$  in the system.

Controlled cooling rate experiments suggest that  $Cr_2O_3$  does not re-equilibrate during cooling, at least between olivines and liquids. Consequently,  $D_{Cr}$  and measured  $Cr_2O_3$  contents of olivines can be used to compute the  $Cr_2O_3$  content of the liquid from which the olivines formed. From the present study, it is surmised that pressure may not significantly alter the trends

of distribution of  $Cr_2O_3$  between the coexisting phases. High chrome diopsides and garnets, found in the ultramafic nodules from some kimberlites, could have equilibrated in a source region which is locally rich in  $Cr_2O_3$  in the upper mantle.

DYNAMIC CRYSTALLIZATION OF LUNAR OLIVINE-NORMATIVE BASALTS

A. Taylor and P. Gamble, Knoxville, USA

Controlled cooling-rate experimentation - a la G. Lofgren and D. Walker - of olivine-normative basalts (e.g. 15555) was conducted by the Pt-loop technique. In particular, the olivine and pyroxene growth rates, morphology, and chemistry were detailed. The diffusion rates of major elements in melt and the crystal/liquid partitioning of major and minor elements as a function of cooling rate were determined. Application of "equilibrium" partitioning data without regard for the kinetics of a dynamic system can lead to erroneous results. Various of the data obtained during the course of this study have been used to refine the olivine cooling speedometer (Taylor et al., 1977) as applied to lunar samples, including those collected by the USSR at the Luna 24 site in Mare Crisium. These investigations have allowed the estimations of cooling rates of lunar rocks based on 1) kinetic modelling of diffusive re-equilibration in olivine and 2) dynamic crystallization studies in lunar rock systems.

GENETIC IMPLICATIONS OF CHEMICAL AND TEXTURAL PROPERTIES  
OF SOME FRA MAURO BRECCIAS (APOLLO 14)

H.D. Knöll and D. Stöffler, Münster, FRG

The Fra Mauro Formation, which is generally believed to belong to the ejecta-blanket of the Imbrium crater, should bear information on the Pre-Imbrian lunar crust and the Imbrium-event itself. They can only be obtained by investigations of polymict breccias, because the primordial crustal rocks are thoroughly brecciated and mixed by impact events and embedded in polymict breccias or the lunar regolith.

Studies, which included 6072 microprobe analyses of plagioclase, pyroxene and olivine taken from the literature, indicated a difference in the main- and minor-chemistry of these minerals depending on their source rock.

We propose the following model for the formation of the Fra Mauro crystalline matrix breccias: (a) Formation of impact melt in one or more local, pre-Imbrian craters and mixing of melt and less shocked fragmented rock material during crater excavation with initial melt temperatures of at least 1700°C, (b) Formation of a series of breccia types ranging from coherent fragment-laden melt rocks to suevite-type detrital breccias rich in discrete clods of melt, (c) Concentration of cold clastic material in schlieren-like bodies by flotation effects to form the parent of the light matrix in a breccia like 14011 or formation of an intimate mixture of impact detritus with independent clods of melt to form the parents of light and dark matrix in a second breccia type (14066 and 14320 are transitional to the latter type), (d) First crystallization of melt in the clast-rich areas (light matrix) at high temperatures with clasts acting as nucleation centers: formation of a relatively coarse grained texture with pigeonitic pyroxene and An-rich plagioclase, (e) Subsequent crystallization of melt in clast-poor areas (dark matrix) at lower temperatures (supercooled liquid) with simultaneous formation of a large number of nuclei; formation of a mosaic-like texture with low-Ca-pyroxene; further textural variation in the matrix most probably resulting from variation of the bulk chemistry due to different degrees of

clast melting, (f) Breccia material now residing in a more or less thick impact formation with slow cooling from temperatures of at least 600°C as indicated by the preservation of highly siliceous glass.

EXPERIMENTAL AND PETROLOGIC STUDY OF THE SHERGOTTITE  
ACHONDRITIC METEORITES

H.Y. McSween, Jr., E. Stolper, Knoxville, Cambridge, USA

Melting experiments conducted on Shergotty and Zagami indicate the following order of phase crystallization: pigeonite, augite, plagioclase. However, the first pigeonites to crystallize are more magnesian than the most magnesian pigeonites in the natural samples. We interpret this as evidence that both meteorites contain cumulus pyroxene. Petrographic observations indicate cumulus augite is also present, but little or no cumulus plagioclase. The experimental results indicate that Zagami consists of approximately 55% intercumulus liquid and 45% cumulus pyroxene, half of which is pigeonite ( $Wo_{11}En_{57}$ ) and half of which is augite ( $Wo_{33}En_{48}$ ). Shergotty consists of 70% intercumulus liquid of the same composition as that in Zagami, and 20% cumulus pigeonite and 10% augite of the same compositions as cumulus pyroxenes in Zagami. Both meteorites crystallized under  $fO_2$  conditions approximating the QEM buffer assemblage, as determined from analyses of coexisting titanomagnetite and ilmenite. Late-stage differentiation of intercumulus liquid produced intergrowths of feldspar-like glass and silica with white-lockite, sulfides and oxides.

THE THERMAL HISTORY AND GENESIS OF LUNAR PYROXENES ON THE BASIS OF THEIR STRUCTURAL AND PHASE STATE DATA

N.R. Khisina, E.S. Makarov, Moscow, USSR

The phase and structural behaviour of pyroxene solid solutions, which is characterized by the exsolution phenomena as well as the crystalline cation ordering, is a function of cooling rates under other equal conditions. This permits to consider the phase and structural state of pyroxenes as an indicator of their cooling history, and on the basis of an analysis of the thermodynamic and kinetic factors of these solid state transformations in pyroxenes the mode for calculating the cooling rate is suggested.

The following parameters of the phase and structural state of pyroxenes are proposed for the calculation of the cooling rates:

$\Delta W_0, \%$  - the difference in  $W_0$  content of the two phases;

$\lambda$  - the scale of exsolution textures;

$T_s$  and  $T_g$  - the corresponding critical temperatures of solvus and spinodal for the given value of  $\Delta W_0$ ;

$\sigma, \%$  - the index of a structural order that means the equilibrium extent in the process of ordering in orthopyroxenes.

The investigation of phase and structural state of single grains of pyroxenes from Luna 16, Luna 20, and Luna 24 soils was carried out using the X-ray single crystal diffraction method, the transmission electron microscopy, the electron probe microanalysis and the X-ray structural analysis. On the basis of the obtained data the evaluation of cooling rates was carried out. The order of magnitude of the determined cooling rates allows to distinguish the pyroxenes, originated from basalts (the cooling rates are about  $n \cdot 10^0 / R$ ) from pyroxenes, originated from gabbro (the cooling rates are about  $n \cdot 10^{-10} / R$ ) and the pyroxenes originated from large intrusions (the cooling rates are about  $n \cdot 10^{-10} / Y$ ). The parameters of phase and structural state of pyroxenes from basalts:  $\Delta W_0$  25-35%,  $\lambda n \cdot 10^2 \text{ \AA}$ ;  $\sigma < 100\%$ , from gabbro:  $\Delta W_0 < 35\%$ ,  $\lambda n \cdot 10^2 \text{ \AA} - n \cdot 10^3 \text{ \AA}$ ;  $\sigma = 100\%$ , from plutons:  $\Delta W_0$  40-45%,  $\lambda n \cdot 10^{-1} - n \cdot 10^1 M$ ;  $\sigma = 100\%$ .

THE SYSTEM  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaFe}^{3+}\text{AlSiO}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  AND ITS BEARING ON FASSAITIC PYROXENES

K. Onuma, M. Akasaka and K. Yagi, Sapporo, Japan

The system was studied at 1 atm by ordinary quenching method. In the join  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ , forsterite, anorthite, spinel, perovskite, and clinopyroxene (ss) crystallize as the primary phases. Spinel has the widest liquidus field. Perovskite has a small liquidus field, but it is present at the subliquidus and subsolidus temperatures. Forsterite is consumed by the reaction with liquid at about  $1250^\circ\text{C}$ . Two clinopyroxenes (ss) were found in the join: one is Ti-bearing diopside(ss) and the other is Ti-pyroxene (ss) containing  $\text{TiO}_2$  up to 8.7%. The liquidus temperature of Ti-pyroxene(ss) ranges from  $1234^\circ\text{C}$  to  $1250^\circ\text{C}$  and its liquidus fields is bounded by that of forsterite diopside(ss), perovskite, spinel and anorthite. Four univariant assemblages are encountered; spinel+anorthite+mellilite+Ti-pyroxene(ss)+liquid, spinel+anorthite+mellilite+perovskite+liquid, anorthite + mellilite + perovskite + Ti-pyroxene(ss)+liquid, and spinel+anorthite+mellilite+diopside(ss)+liquid. An invariant point spinel+anorthite+mellilite+perovskite+Ti-pyroxene(ss)+liquid was confirmed at  $1230^\circ\text{C}$ . The chemical composition of the Ti-pyroxene (ss) is very close to that of clinopyroxene found in Ca-Al-rich inclusions of the Allende meteorite. This clinopyroxene coexists with spinel, perovskite, mellilite and anorthite and this assemblage is the same as that of the invariant point confirmed in the present study.

In the join  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaFe}^{3+}\text{AlSiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ , clinopyroxene (ss) forsterite, perovskite, magnetite(ss), spine(ss), anorthite mellilite, unknown phase X and hibonite were encountered. At subsolidus temperatures clinopyroxene(ss) single phase field extends up to 18 wt.%  $\text{CaTiAl}_2\text{O}_6$ , showing that the content of  $\text{TiO}_2$  in clinopyroxene (ss) increase with increasing  $\text{CaFe}^{3+}\text{AlSiO}_6$ . Following phase assemblages were confirmed in the subsolidus region: clinopyroxene(ss)+perovskite, clinopyroxene(ss) + perovskite+spinel(ss), clinopyroxene(ss)+perovskite+mellilite (+anorthite), clinopyroxene(ss)+perovskite+spinel(ss)+mellilite (+anorthite), clinopyroxene(ss)+perovskite+anorthite+spinel(ss),



and clinopyroxene(ss)+perovskite+hibonite+anorthite. Even in the field of clinopyroxene(ss)+perovskite, TiO<sub>2</sub> content in clinopyroxene(ss) continues to increase and attains up to 9.20% TiO<sub>2</sub> and 18.57% Al<sub>2</sub>O<sub>3</sub>. An implication of the join to Ti-rich fassaitic pyroxene from the alkalic rocks will be discussed.

#### THE STRUCTURE, COMPOSITION AND ORIGIN OF COHENITE FROM LUNA-20 SAMPLE

M.A. Nazarov, L.S. Tarasov, A.V. Ivanov, K.I. Tobelko,  
Moscow, USSR

The cohenite is found in the sample of metamorphosed highland breccia from Luna 20; the mineral is associated with the peripheral zone of the large grain of the metallic iron, represented by the kamacite. The cohenite and associated kamacite and taenite are analysed on the X-ray microprobe. In the chemical composition of cohenite were found (in wt %) Fe 89,5, Ni 2.0, Co 0,2. At the first time were carried out the X-ray identification and the unit cell parameters ( $a_0 = 4.501 \pm 0.008$ ,  $b_0 = 5.090 \pm 0.013$ ,  $c_0 = 6.743 \pm 0.014$  Å). The distinct asterism of the diffraction lines could be due to the impact process. The domain structure and its transformation in the magnetic field was studied using the magnetic powder method. The chemical composition, the unit cell parameters and the domain structure of the carbide under the investigation are similar to known cohenites of terrestrial and meteoritic origin. The formation of this mineral is related to the subsolidus exsolution of the  $\gamma$ -phase of metallic iron during the moderately slow cooling within the layer of hot crater ejects.

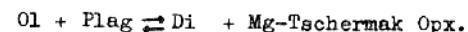
#### EXPERIMENTAL INVESTIGATIONS ON THE SPINEL CATACLASITES;

A.E. Bence, T.L. Grove and D.H. Lindsley, New York, USA

A very small number of highland lithic fragments in the Apollo 17 2-4 mm soil fraction contain the assemblage forsteritic olivine-aluminous enstatite-anorthite-pleonaste spinel. Bulk compositions, calculated from phase chemistries and modal mineralogies, indicate that phase relationships in the system MgO-Al<sub>2</sub>O<sub>3</sub>-CaO-SiO<sub>2</sub> may be used to understand these lunar lithologies. Subsolidus relationships for two compositions in the system were studied by Kushiro and Yoder (1966) who recognized the important pressure-sensitive reaction  $Fo + An \rightleftharpoons Di_{ss} + En_{ss} + Sp$ . Bence and McGee (1975), utilizing these observations, the solubility of Al<sub>2</sub>O<sub>3</sub> in orthopyroxene (Obata, 1976), the absence of calcic clinopyroxene, the rare occurrence of cordierite, and the olivine-spinel geothermometer, estimated T-P conditions of equilibration of  $\sim 1200^\circ\text{C}$  and 2-3 kb. These estimates have large uncertainties, consequently we have conducted high-pressure experiments on a composition approximating the lunar crustal assemblage.

Two crystalline starting assemblages prepared from oxide mixes were synthesized in iron capsules at  $1200^\circ\text{C}$  for 5 days in an evacuated silica tube and at 10 kb,  $1275^\circ\text{C}$  for 7 hours in a piston-cylinder apparatus. The products from the piston cylinder experiment were then rerun in iron capsules at 8 kb,  $1280^\circ\text{C}$  and 13 kb  $1275^\circ\text{C}$  for 3 days.

The evacuated silica tube synthesis experiment yielded an Opx with low Wo and Al contents coexisting with spinel, plagioclase and olivine. In the 8 kb experiment plagioclase and olivine persist with spinel and a more Ca- and Al-rich Opx. The compositional changes in Opx are consistent with the reaction:



With a further increase in pressure the olivine is consumed and the assemblage low-Ca Opx, high-Ca Opx, plagioclase and spinel is generated. The abundances of spinel and plagioclase have increased and decreased, respectively.

The evacuated silica tube experiment is a synthesis and the 8 kb and 13 kb experiments are only "reversed" in the sense that the orthopyroxene composition changes and olivine or high-Ca Opx respond in a consistent manner. If the thermochemical calculations of Obata (1976) are applicable, we can place limits on conditions of lunar spinel cataclasite equilibration. Obata's (1976) calculations suggest that the olivine + plagioclase consuming reaction is pressure-dependent. Our 8 kb and evacuated silica tube experiments provide two calibration points for Al isopleths in orthopyroxene in the stability field of olivine + plagioclase. By comparing the average percentages of octahedral aluminium in the experimental and natural orthopyroxenes, we make a preliminary preliminary pressure estimate of  $3 \pm 1$  kb , which is equivalent to a depth of  $60 \pm 20$  km.

## CRYSTAL GROWTH

## MORPHOLOGY OF GROWTH SPIRALS ON NATURAL MINERALS

I. Sunagawa, Sendai, Japan

Morphology of growth spirals so far observed on mineral crystals of a wide spectrum of origins is critically analysed based on the recent theoretical developments and computer simulations on spiral morphology. Based on these analyses, characteristics of crystallization taking place in geological environments are discussed. The following growth environments are discussed. The following growth environments will be treated.

1. Vapour phase; druse minerals, pegmatite minerals
2. Aqueous solution phase; hydrothermal minerals, hydrothermal metasomatic minerals
3. High temperature solution phase; minerals of magmatic origin
4. Metamorphic minerals.

## NUCLEATION AND BUILDING UP OF CRYSTALLINE STRUCTURES DURING GROWTH OF MINERAL CRYSTALS

S.A. Stroitelev, Novosibirsk, USSR

On the basis of crystallochemical regularities crystallization and growth of mineral crystals is viewed as a process of nucleation and subsequent building up of crystalline structures. The peculiarities of crystallization and growth of mineral crystals are established depending on the type of crystal structure.

For each crystal type the particular crystallochemical pattern of habitus transformations is established and compared with the experimentally known growth forms. The crystallochemical model is proposed for the formation of grown-in structural defects and impurity distribution.

## STRUCTURAL MORPHOLOGY OF RUTILE AND TRIRUTILE TYPE CRYSTALS

R.O. Felius, Leiden, Netherlands

The PBC-theory (see e.g. Hartman, 1973) is applied to the rutile structure:  $P4_2/mmm$ ,  $a = 4.594 \text{ \AA}$ ,  $c = 2.959 \text{ \AA}$  and  $Z = 2$ , and to the trirutile structure: ditto, except for  $c = 8.877 \text{ \AA}$ , because of the ordered replacement  $6 \text{ Ti}^{4+} \rightarrow 2 \text{ A}^{2+} + 4 \text{ B}^{5+}$ . The F-forms of rutile are:  $\{110\}$  and  $\{101\}$ , those of trirutile  $\{101\}$ ,  $\{103\}$ ,  $\{001\}$ ,  $\{110\}$ ,  $\{112\}$  and (as could only be estimated by applying energy calculations)  $\{111\}$ .

Starting from an electrostatic point charge model of both structures, the electrostatic attachment energies are calculated with respect to the F-faces and some S- and K-faces, neglecting the Born repulsion and the partly covalent character of the bonds.

In a broken bond model of both structures, in which all cation-anion bonds are taken equal, the numbers of broken bonds are counted with respect to the same faces.

By application of the Wulff plot method the "ionic" growth forms of rutile and trirutile are drawn from the attachment energy values and the "covalent" growth forms from the numbers of broken bonds.

The structural growth form of rutile thus estimated consists of the forms  $\{110\}$  and  $\{101\}$ . The "ionic" structural growth form of trirutile contains the forms  $\{110\}$ ,  $\{101\}$ ,  $\{103\}$  and  $\{001\}$ , its "covalent" structural growth form the forms  $\{110\}$ ,  $\{101\}$  and  $\{001\}$ .

The true growth forms must be in between the "ionic" and "covalent" forms concerned. With respect to minerals with the trirutile structure, this can be demonstrated by using relative bond strengths (Povarennykh, 1972) instead of attachment energies and broken bonds.

## APPEARANCE OF THE ELEMENTARY PROCESSES OF GROWTH AND SOLUTION ON THE MAGNETITE CRYSTAL FACES IN CONNECTION WITH THEIR TYPOMORPHISM

R.M. Alijew, V.G. Feklichev, Moscow, USSR

The results of special investigation of the crystallization of magnetite under hydrothermal conditions are in good agreement within the phenomena observed in nature:  $o\{III\}$  and  $d\{110\}$ -forms define the habit of natural and synthesized crystals.

There were calculated the atomic densities of the elementary growth layers of 10 statistically most usual magnetite facies. This has revealed that the most close packed faces are  $o$  and  $d$  ones ( $E_o = 45,723$  and  $E_d = 28,00$ ). The analysis of the atomic structure of  $o\{III\}$  and  $d\{110\}$ -faces within the  $d_{hkl}$ -range leads to the theoretical conclusion that in magnetite structure the lauer growth mechanics is typical of  $o$  and sometimes of  $d$ -faces.

At free and metasomatic growth which are realized through layer and spiral growth mechanism, the defects concentrated on edges and in central areas of faces (i.e. dislocations, inclusions) are the centres of the generation of growth layers and dissolution.

The microrelief and types of microstructure on faces (including  $o$  and  $d$ -faces) depend on mechanism and physicochemical peculiarities of growth and solution of magnetite crystals. In all the cases the directions of edge forms of the elementary growth and solution figures on the faces under investigation are within the set theoretically possible directions which stand out in the structure of the surface layer of each face.

RELATION BETWEEN THE CRYSTAL STRUCTURE AND MORPHOLOGY OF  
ALKALI FELDSPARS

C.F. Woensdregt, Leiden, Netherlands

The influence of the crystal structure on the crystal morphology of monoclinic potassium feldspars has been investigated according to the Periodic Bond Chain Theory. The most important crystal faces are classified as either  $F_1$ -faces, parallel to at least, two PBCs having only T-O bonds, or as  $F_2$ -faces, parallel to at least two PBCs, one of which contains in addition K-O bonds.

The  $F_1$ -faces are  $\{110\}$ ,  $\{001\}$ ,  $\{010\}$ ,  $\{2\bar{0}\}$  and  $\{11\bar{1}\}$ . The  $F_2$  faces are  $\{130\}$ ,  $\{021\}$ ,  $\{22\bar{1}\}$ ,  $\{11\bar{2}\}$ ,  $\{100\}$  and  $\{10\bar{1}\}$ .

The attachment energy, which is assumed to be proportional to the growth rate, can be calculated in an electrostatic point charge model. For this ionic model the growth form consists only of  $\{001\}$ ,  $\{010\}$ ,  $\{110\}$  and  $\{021\}$ .

Calculations in a broken bond model based on the relative bond strength given by Povarennykh reveal that the growth form does not change substantially. Only  $\{021\}$  disappears.

Neither does the growth form differ, when the effective charges in the point charge model are lowered to about 50% of the normal values. So the degree of Si-Al-O polymerization in the melt and covalency of the T-O bonds have a minor influence on the crystal morphology. Assuming that Al can be orderly built in, the growth form of maximum microcline is fairly triclinic. The form  $\{110\}$  is more important than  $\{1\bar{1}0\}$ ,  $\{021\}$  is present and  $\{0\bar{2}1\}$  is lacking. Also  $\{001\}$  and  $\{201\}$  are present.

Calculations for the Na endmembers show that the substitution of K by Na does not change the growth form at all. The anorthite growth form is, however, elongated along the a-axis and slightly platy following  $\{001\}$ .

LATTICE DEFECTS IN MICRO DIAMOND

T. Nishida and Y. Takano, Tokyo, Japan

The relation between lattice defects and micro-morphology of crystal faces in micro diamonds was studied by X-ray topographic method, phase contrast microscopy and scanning electron microscopy. The feature and distribution of the defects contained in the crystal reveal the history of the crystal growth and the surface micro-morphology shows the environmental conditions of the last growth stage. Specimens are natural microdiamonds having octahedral habit from Congo with the size of 0.5mm in diameter. In spite of the belonging of the cubic system, few samples are free from birefringence. Almost all of the residual crystals were studied by X-ray section topography because of disturbance of the contrast by the strain.

The observation results indicate the existence of several types of the defect contained in the micro diamond as follows; (1) the dislocation lines radiating out to every direction from the middle of the crystal. (2) the dislocation lines parallel to  $\langle 110 \rangle$  direction. (3) the triangular plane defects parallel to  $\{111\}$  face varying in size. The second type of the dislocation line has sometimes the mutual relation to the surface morphology of the fault line observed on the  $\{111\}$  face. The direction of the fault line is comprised in the slip plane  $\{111\}$  of the stacking fault most easy to occur in the diamond structure. The existence of the fault line suggests the genesis of the twin nucleus considering the similarity in the atomic arrangement in the stacking fault and the twin boundary. Experimental results show the fault line is closely related to the origination of the twin crystal. In the first and third cases of the defect type it seems that there exists no proper correlation between inner texture of defects and surface morphology.

ZONALITY AND SECTORIALITY OF COMPOSITION IN ZIRCON CRYSTALS  
FROM PEGMATITE OF THE ILMEN MOUNTAINS BASED ON THE NEUTRONO  
GRAPHIC DATA

V.I. Popova, Miass, USSR

Zircon crystals from feldspathic and nepheline-feldspathic pegmatite of the Ilmen Mountains are characterized by the diversity of habit and occur in growth with different minerals: aeschynite, magnetite, minerals of pyrochlor group, plagioclase, orthite, apatite, ilmenite (Rubel, Simonov, 1949; Popova, Tscherbakova, 1977).

The heterogeneity of composition (zonality and sectoriality) in zircon crystals were studied by the neutronography technique (neutron activation radiography) by means of treating the preparations with the current of heat neutrons  $\Phi=3,2 \cdot 10^{10}$  and  $7,9 \cdot 10^{10}$  n/cm<sup>2</sup>.sec, 5 and 3 min respectively, at power  $W=10$  MW and their successive radiography at the photoplates. Similar investigations under different regimes of irradiation are known for garnet (Banno et al., 1970).

In 350 zircon crystals from 29 pegmatitic veins of the Ilmen Mountains studied by neutronography technique were revealed the distinction of composition in the growth zones and pyramids of different forms, the change of crystal forms in the process of growth, the alteration of relative growth rates of different faces, the regeneration of debris, mineral inclusions and other peculiarities. It was determined that the growth pyramids of prisms  $\langle 010 \rangle$  and  $\langle 110 \rangle$  are characterized by the increasing content of hafnium and rare earths (it is confirmed by gamma scanning and the data of X-ray microanalyses). In different growth zones of crystals in the growth pyramids  $\langle III \rangle$ ,  $\langle 010 \rangle$  and  $\langle 110 \rangle$  were revealed the content variations of hafnium, yttrium, thorium, uranium and other elements.

The revealed zonality and sectoriality of composition in zircon crystals from pegmatite of the Ilmen Mountains give the possi -

bility to compare the change in the form and the composition of zircon crystals in the different pegmatitic veins and to reveal veins with synchronously crystallized zircon.

The neutron activation radiography should find a broad usage in the investigation of natural and technical mineralgenesis as an effective and perspective method for studying the heterogeneities of composition and structure of different crystals (Boltov et al., 1976). The method is suitable for the investigation of any chemical combinations including the organic ones.

CRYSTAL GROWTH FROM SOLUTION STUDIED BY HOLOGRAPHIC INTERFEROMETRY TECHNIQUE

F. Bedarida and L. Zefiro, Genova, Italy

Transmission holographic interferometry is applied to crystal growth from solution. It is possible to control the mechanism of growth of the crystal, checking also the concentration gradients and the influence of the convective movements of the liquid.

## LAYER AND SPIRAL GROWTH OF KAOLINITE CRYSTALS

N.D. Samotoin, S.S. Chekin, Moscow, USSR

Growth hillocks on 001 kaolinite faces were observed during vacuum decoration microscopic studies of these crystals formed of the weathered and hydrothermally changed framework and layer aluminosilicates.

The hillocks were formed by elementary layers with the steps distributed in the form of disordered hexagonal spirals and(or) loops corresponding to the triclinic kaolinite structure.

One can distinguish the hillocks formed 1) as a result of layer growth (rare and only in the first stages) and 2) as a result of a layer and spiral growth (usually), where the screw dislocation density is in the order of  $10^4$  to  $10^9$   $\text{cm}^{-2}$ .

Depending on the sign and the density of dislocations one can specify several types of growth spirals: a) spirals formed by a positive or a negative screw dislocation with single Burgers vector; b) spiral groups formed with two or more screw dislocations of the same sign; c) spirals and loops formed by a growth analogue of the Frank-Reed dislocations or by a group of them.

The screw dislocations are not caused by any superposition on any of the layer regions. They result from a rupture and a vertical shift of the ruptured parts occurred during the growth of the crystal, following the major crystallographic directions. The crystal stops growing when the density of dislocations becomes  $10^9$   $\text{cm}^{-2}$ .

The particular features of the hillocks witness for a low super saturation of the solutions where the kaolinite crystals were formed.

The results of this study have shown that the layer and spiral growth is the major mechanism of the mineral growth. It brings to a new light the results of the kaolinite investigations made by different analytical and instrumental methods.

## THE GROWTH OF GALENA CRYSTALS

I. Bonev, Sofia, Bulgaria

Galena crystals of various morphology are widespread in the lead-zinc ore deposits of the Rhodope massif, Bulgaria. They are bounded by cubic and octahedron faces only, and often reach a considerable size. The most characteristic feature of their surface sculpture are nearly polygonal ( $\sphericalangle[110]$  and  $[100]$ ) growth steps of a macroscopic height.

The main part of the crystals is formed through layer growth at a comparatively low supersaturation. Generally, the reentrant angles at the mosaic subgrain boundaries, and particularly, the points where the height differences between the adjacent subgrain surfaces are maximal, act as permanent layer sources. Peculiar spiral step systems are formed by the interaction of the thick layers belonging to the neighbouring subgrains. However, the layers in these systems are plane-parallel, not helicoidal. There is a close connection between the macroscopic heights of the growth layers and of the corresponding step sources. The radial-mosaic (lineage) inner structure of the crystals as well as their zonality indicate that these layer sources were active almost during the whole period of the crystal growth. In the presence of self-perpetuating rough surface defects, the screw-dislocation growth mechanism becomes inefficient.

In case of hindered diffusion and increased and non-uniform supersaturation, skeletal crystals of hopper-like and macro-spiral shape as well as porous aggregates are formed. Changes in the growth conditions lead to the formation of platy skeletons, 100 single-crystal plates and linear or kinked whiskers.

The growth features of galena from other important deposits are similar to those described here.

HYDROTHERMAL CRYSTALLIZATION AND TYPOMORPHIC FEATURES  
OF MERCURY AND ANTIMONY SULPHIDES

N.A. Ozerova, V.S. Balitsky, V.V. Komova, Moscow, USSR

To learn dependence of constitution and properties of mercury and antimony sulphides on crystallization conditions we attempted a series of experiments involving these particular sulphides growing in hydrothermal solutions of various compositions, pH and Eh in a wide temperature and pressure range, study of grown crystals morphology and distribution of selenium isomorphic impurity in them and determination of certain physical properties. Change in crystallization conditions is mostly reflected in cinnabar crystals outward morphology. Depending on concentration of alkaline-sulphide media isometric, columnar, and rhombohedral crystals are formed; tabular crystals of hexagonal habit which become thicktabular with acidity decrease are formed in muriatic media. Change in crystal morphology from isometric to columnar occurs in sulphide-sodium solutions with temperature increase. In acid solutions under antimonite recrystallization prismatic crystals are mainly formed. In pure water, sulphide-sodium and nearly neutral and subalkaline chloride solutions crystals have fine-acicular habit. Relation between crystallization conditions and selenium isomorphic impurity concentration in mercury and antimony sulphides have been also estimated. Ratio  $\sum \text{Se} : \sum \text{S}$  in solution of Eh system has the most effect on structural trapping of selenium, acidity-alkalinity of solutions has the lower one. The distribution manner of selenium in crystals is determined by ratio of selenium to sulphur as well as by temperature. Selenium doping to mercury and antimony sulphides results in regular change in crystal lattice parameters as well as in optical and other physical properties of grown crystals.

MICROMORPHOLOGY OF CRYSTALS SYNTHETIC HYDROTHERMAL EMERALD

V.S. Shatsky, A.S. Lebedev, V.A. Kljakhin, Novosibirsk, USSR

Micromorphology of emerald crystals grown by hydrothermal conditions was investigated. The emerald crystals are faceted by the following facets:  $\{0001\}$ ,  $\{10\bar{1}0\}$ ,  $\{11\bar{2}0\}$ ,  $\{11\bar{2}1\}$ ,  $\{10\bar{1}1\}$ .

The flaky-spiral mechanism of growth is characteristic of all the faces in the crystals studied that is in agreement with the data of microcrystalomorphological investigations. The centres of spiral generation is shown to be microcrystals of beryl or foreign particles, adhered to a face.

The investigation of the crystals grown in the solutions with a different cation-anion ratio under different P-T conditions, bringing the conclusion that the main factor determining micromorphology of faces is their growth rate.

Modification of spiral forms depending on the growth face is considered. At the face  $\{10\bar{1}0\}$  with increasing of the facet's growth rate, the form of spirals changes as follows: quadrangular pyramids, pyramids, formed by the layers, having the form of razor blade, hexagonal pyramids, spirals of growth in the form of cone.

The polygonized spiral layers are changed by round ones for faces of dipyramids  $\{11\bar{2}1\}$ ,  $\{10\bar{1}1\}$  with increasing the growth rate.

The comparison of the face micromorphology of natural and synthetic beryls is given. It is noted that there is no principal difference in the mechanism growth of natural and synthetic beryl crystals. The possibility of using the micromorphologic faces of beryl crystals as typomorphic feature is discussed.



SURFACE MICROTOPOGRAPHY OF NATURAL DIAMONDS FROM  
VARIOUS SOURCES

H. Komatsu, Sendai, Japan

It is interesting to examine the surface features of diamonds, since the process of growth or dissolution has been reflected on the surface topography. Diamonds are ideal crystals for this kind of investigations as they are the hardest crystals and can endure severe weathering for a long period of time.

An attempt was made to compare the surface features of diamonds from various sources. They were collected from Mali, Ghana, South Africa and Siberia. After examining the percentage of habit distribution among them, the surface microtopography was studied by two-beam interferometry and phase-microscopy. Although the number of crystals is very much limited, it was possible to find conspicuous differences among them.

The diamonds from Mali (24) and Ghana (70) show dissolution forms with extremely flat and smooth surfaces having etch pits or Pandeya's rings (Pandeya and Tolansky, Proc. Phys.Soc., 78, 12:1961). It is quite obvious that they suffered weathering after they were brought up to the surface of the earth.

South African diamonds show a variety of surfaces such as dendritic over growth, regrown-fractured surfaces and trigons. They give us an impression that they have persisted in violent geological conditions while they grew. The number of crystals examined exceeds 100.

Some diamonds from Siberia have remarkably smooth surfaces with sharp edges and corners, showing no trace of dissolution. It seems that some Siberian diamonds have retained as-grown surfaces, which are suited for an analysis of growth mechanism of natural diamonds.

HYDROTHERMAL SYNTHESIS OF GERMANIUM ANALOGUES OF SILICATES

T.G. Uvarova, L.N. Demyanets, A.N. Lobachev, Moscow, USSR

Germanium being of a small clark value, it forms practically no natural minerals, though it constitutes an impurity in a number of associated compositions. At the same time, the chemical and crystallochemical similarity between Ge and Si, for example, invokes a large isomorphous miscibility of the compositions where these elements participate. It may be seen quite clearly in the experimental studies. The experimentators aimed at the physical and chemical particularities in the formation of some silicate analogues. They want to specify the particular features in the crystallization of Ge and Si compositions and to obtain the germanate type single crystals.

We have studied the hydrothermal crystallization in a quadruple system:  $\text{Na}_2\text{O} - \text{Al}_2\text{O}_3 - \text{GeO}_2 - \text{H}_2\text{O}$  with largely varied component percentage ( $\text{Al}_2\text{O}_3/\text{GeO}_2 = 1:3 - 3:1$  mol.;  $\text{H}_2\text{O}/\text{Na}_2\text{O} = 6:0,03 - 3:0,6$  mol; at temperatures of 300-500°C. The physical and chemical crystallization conditions have determined the phase fields obtained:  $\text{Na}_2\text{Al}_2\text{Ge}_2\text{O}_7 \cdot \text{H}_2\text{O}$ ;  $\text{NaAlGe}_4\text{O}_{10}$ ;  $\text{NaAlGe}_4\text{O}_9 \cdot \text{N}_2\text{O}$ ;  $\text{Na}_8\text{Al}_6\text{Ge}_6\text{O}_{24}(\text{OH})_2 \cdot \text{H}_2\text{O}$  cub;  $\text{Na}_8\text{Al}_6\text{Ge}_6\text{O}_{24}(\text{CO})_2 \cdot 2\text{H}_2\text{O}$ . All these compositions but the last, are structurally analogous to silicates (analcime, nepheline, nepheline-hydrates and sodalite). The only exception is also analogous (compositionally) to the sodalite and cancrinite minerals but it has a different crystal structure and a trigonal unit cell (P31c in Fedorov group).

The examples of alumogermanates  $\text{Na}_8\text{Al}_6\text{Ge}_6\text{O}_{24}(\text{OH})_2 \cdot \text{H}_2\text{O}$  (cub.) and  $\text{Na}_8\text{Al}_6\text{Ge}_6\text{O}_{24}(\text{CO})_2 \cdot 2\text{H}_2\text{O}$  (trigon.) demonstrate a possibility of single crystal layers and large single crystal hydrothermal epitaxy on the corresponding silicate seeding substratum.

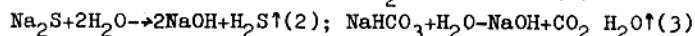
SOME PHYSICO-CHEMICAL ASPECTS OF HYDROTHERMAL CRYSTAL  
GROWTH AND NATURAL MINERAL GENESIS

I. Ganeev, Moscow, USSR

One of the information sources to develop a theory of mineral genesis is formation of crystals by means of hydrothermal method employing temperature gradients. It is supposed that mineralization occurs as a result of supersaturation in the upper low temperature zone. However, some experiments with quartz corundum and other minerals have shown that supersaturation is necessary but not sufficient conditions.

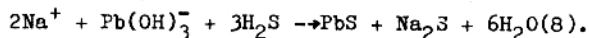
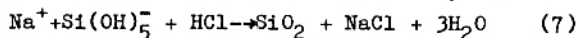
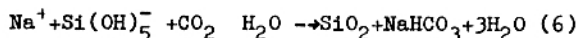
The object of our investigation was those solutions of electrolytes which were found most typical of natural hydrothermal solutions. The analysis of the results prove that if there is  $\Delta t$  in the structure field, then solutions at electrolytes undergo differentiations with the formation of acid solutions in the upper zone and alkaline ones in the lower zone. The mechanism of formation of differentiation involves hydrolysis of salts at a movable product ( $\text{CO}_2$ ,  $\text{HCl}$ ,  $\text{HF}$ ,  $\text{H}_2\text{S}$ ) less dense than a water solution accumulating in the upper zone.

The results which were obtained have permitted to propose the following process model to form minerals by hydrothermal method. In lower zone we have:  $\text{NaCl} + \text{H}_2\text{O} \rightarrow \text{NaOH} + \text{HCl} \uparrow (1)$



The alkaline solution in the lower zone reacts a charge with high solute contents formation:  $\text{NaOH} + \text{SiO}_2 + 2\text{H}_2\text{O} \rightarrow \text{Na}^+ + \text{Si}(\text{OH})_5^- (4)$   
 $\text{NaOH} + \text{PbS} + 2\text{H}_2\text{O} \rightarrow \text{Na}^+ + \text{Pb}(\text{OH})_3^- + \text{H}_2\text{S} \uparrow (5).$

The solution is transported by means of convection laws to the upper zone, where the complexes are subject to an acid hydrolysis to form a mineral, water and a salt:



The main form of migrations of substance in natural hydrothermal solutions are most probably hydrocomplexes formed in the lower root parts at the vein. Mineral formations is realized as a result of the acid hydrolyses of hydrocomplexes along with the rise of solutions to the upper horizons, enriched in  $\text{CO}_2$ ,  $\text{HF}$ ,  $\text{HCl}$ ,  $\text{H}_2\text{S}$ .

PREFERRED ORIENTATION OF ROCK-FORMING CRYSTALS EXPERIMENTALLY  
PRODUCED

P. Paulitsch, Darmstadt, FRG

In order to understand the origin of preferred orientation of rock-forming minerals in nature experiments were carried out by a piston cylinder press between 200 and 500°C and 1-15 kb to produce cordierite-fabrics, mostly garnet crystal powder was used.

Beyond transition temperature cordierite crystals ( $\phi$  0,1) appear and show a preferred orientation in a small circle around the pressure-direction.

In a similar way calcite-fabric were experimentally produced starting with an aragonite powder.

Both fabrics show a nearly homoaxial symmetry. The origin of a preferred orientation of rock-forming crystals can be caused by external rotation, by translation-gliding mechanism or by preferred crystal-growing under pressure.

Under conditions above we assume growing under pressure is the first reason for the observed preferred orientation. The investigations were supported by DFG which I gratefully acknowledge.

## PREPARATION OF STAR-RUBY CONTAINING LARGE CRYSTAL OF RUTILE

H. Takubo, Y. Kitamura, Sh. Kume and M. Koizumi, Osaka, Japan

Needle crystals in the synthetic star-rubies by the Verneuil - technique are known to be too small for identifying phases. In this study, ruby crystals containing such large needle crystals as available for microscopic and X-ray studies were prepared by flux-melt growth. Compositions of the flux-melt used were  $1.5\text{Al}_2\text{O}_3 \cdot 0.005\text{Cr}_2\text{O}_3 \cdot \text{PbF}_2 \cdot \text{PbO} \cdot 0.74\text{B}_2\text{O}_3 \cdot x\text{TiO}_2$ , where  $x=0, 0.005, 0.05, 0.10$  and  $0.20$ . The flux-melt was slowly cooled from  $1370^\circ\text{C}$  to  $800^\circ\text{C}$  keeping general rate of cooling at  $1 \sim 2^\circ\text{C/hr}$ . In addition to the slow cooling, temperature was intermittently dropped and raised by  $150^\circ\text{C} \sim 200^\circ\text{C}$  with interval of  $12 \sim 24\text{hr}$ . Ruby crystals obtained were  $3 \sim 20\text{mm}$  across,  $0.1 \sim 4\text{mm}$  thick and translucent. Zonal structure and twinning were common. The maximum value for  $\text{TiO}_2$ -concentration was  $0.75\text{wt.}\%$ . By aging at  $1300^\circ\text{C}$  in oxygen gas flow for 13 days, the as-grown ruby crystals lost their lustre and translucency. A number of needle crystals showing lath-shaped texture were observed on the  $\{0001\}$  faces and along the internal sub-boundaries of the aged crystals. The needle crystals were  $0.1 \sim 1\text{ mm}$  long,  $0.001 \sim 0.05$  wide and less than  $0.002\text{ mm}$  in thickness. They were identified as rutile by microscopic and X-ray examinations. The  $(100)$  faces of rutile were nearly parallel to the  $(0001)$  faces of ruby and the  $\langle 001 \rangle$  direction of rutile was parallel to  $\langle 10\bar{1}0 \rangle$  direction of ruby. This relation can be explained by considering the oxygen packing of both minerals. The asterism effect was not so clear as seen in commercial star-rubies. The result of preliminary experiments suggests that, if appropriate control is made on temperature fluctuation, Ti-concentration and twin configuration, it may be possible to make a synthetic star-ruby similar, in both external and internal appearances, to a Burmese star-ruby.

## HIGH TEMPERATURE CRYSTALLIZATION CONDITIONS OF ANHYDROUS BORATE CRYSTALS. THEIR COMPOSITION, HOMOGENEITY AND MORPHOLOGY

N.I. Leonjuk, T.I. Timtsenko, L.I. Alshinskaya, A.V. Pashkova, N.V. Belov, Moscow, USSR

Some 40 anhydrous refractory borates have been studied in their crystallochemistry, morphology, composition, homogeneity and crystallization conditions. The most common features have been disclosed. Crystals of some isostructural compositions are obtained and a possibility of a large isomorphism proposed. The crystal structures of some compositions being scrupulously analysed, the distortion of cation polyhedra in the isostructural series are made more evident.

The borate make it possible to study the general effect of the crystal structure and the composition at one hand and that of the crystallization conditions (temperature, supersaturation, chemistry and impurities) - at the other hand, crystal morphology. It appears that the cations not only determine the crystal structure formation, but do define critically their faceting laws. Some methods are proposed to estimate the major primitive forms of the crystals, considering their real structures and compositions. It permits now to explain satisfactorily some particular features in crystal morphology.

We study also the effect of the crystallisation conditions on crystal composition and homogeneity. It is shown that the solvent-produced impurities are isomorphic in the crystals, and the isomorphism of the elements-impurities is stated to be both - iso- and hetero-valent.

The effect of the impurities on LASER and electrooptic properties of the rare-earth borate crystals is being also discussed.

The comparative investigations of properties obtained varieties of tourmaline also have been made (that is goniometric, optical investigations, investigations of density, microsolidity and some other properties).

OH-PHLOGOPITE MONOCRYSTALLINE GROWTH UNDER HYDROTHERMAL CONDITIONS, CHANGES OF COMPOSITION AS A FUNCTION OF GROWTH RATE

A. Baronnet, B. Velde, Marceille, Paris, France

Using the "temperature drop" technique of hydrothermal monocry-stalline growth, growth rates of the lateral faces of mica ranging from 50 to 200  $\mu\text{m}/\text{day}$  have been reached. Experimental conditions are as follows: KOH (6.5 m) aqueous solution, 500-520°C growth temperatures, temperature drops  $\Delta T$ : 5 to 18°C, solution pressure: 2 kb. Nutrient mica as well as seeds were total Fe-poor natural phlogopites. The knowledge of experimental isobare solubility curves of phlogopite as a function of temperature allows one to define the solution nominal supersaturation and, then all growth kinetic parameters are controlled.

The mica overgrowths around seeds exhibit growth sectors whose respective developments are strongly a function of supersaturation:  $\{0K1\}$  and  $\{hhl\}$  forms are always present but  $\{h0l\}$  are degenerated ones,  $\{h\ 3h\ 1\}$  development being very sensitive to supersaturation.

Within a given sector, growth zones are observed, directly related to changes of steady-state growth regimes. An electron microprobe investigation shows that the As-grown mica is a Mg-rich, Al-poor phlogopite. Its total-Fe and Ti content increases with the supersaturation (and growth rate) while Mg decreases, whereas K and Si are insensitive to the kinetics. Fe is mainly present as  $\text{Fe}^{3+}$  in the mica and its concentration in the solution is buffered by the spontaneous growth of titanomagnetite onto the walls of the autoclave.

SYNTHESIS AND INVESTIGATIONS OF A ROW OF TOURMALINE VARIETIES

I.E. Voskresenskaya, Moscow, USSR

A correlation of some structural peculiarities in natural and synthetic tourmalins has been made by means of infra-red spectroscopy, electron-paramagnetic-resonance methods and gamma-resonance investigations.

As a result of a series of experiments carried out in the hydrothermal laboratory of the Institute of Crystallography there have been established some peculiarities in tourmaline crystallization under following conditions: in boron-fluorine, boron-chlorine and boron-fluorine-chlorine mixed aqueous acid systems (concentration of chlorides did not exceed 2-3%); under condition of monomineral synthesis from oxides of a corresponding composition and finally in high-concentrated chloride mediums (concentration of chlorides 70-80%).

The definite regularities in succession of the formation phases accompanying tourmaline were observed on each stage of investigation. Accompanying phases of tourmaline are the main crystalline phases of system  $\text{Al}_2\text{O}_3\text{-SiO}_2\text{-FeO}$ , various fluorine-aluminates and boracites.

In this report the last stage of investigations is considered in greater detail: the experiments on synthesis and growth of tourmaline in high-concentrated chloride medium of magnesium, iron, nickel, cobalt, chrome, manganese were carried out using the installation constructed by A.A. Shternberg at  $T=750\text{-}800^\circ\text{C}$  and  $p=1000\text{-}2000$  atm by one scheme.

It was found that from spatially-divided hardly-soluble components in high-concentrated chloride medium it is possible to obtain tourmaline single crystals of different composition both colourless and also coloured in various hints, suitable for investigations of their physical properties.

Thus it was possible to obtain single crystals colourless magnesium and also ferriferous, cobaltous, nickeliferous, chromic and manganic tourmalines.

KINETICS OF THE GROWTH OF STIBIOTANTALITE GROUP SINGLE CRYSTALS

V.I. Popolitov, A.N. Lobachev, V.F. Peskin, Moscow, USSR

The communication describes the conditions of the growth of  $\text{SbNb}(\text{Ta})\text{O}_4$  single crystals with the structure of natural mineral stibiotantalite. The aim of the experiments was the determination of physical-chemical conditions of the formation of natural stibiotantalite single crystals, which are of interest as photo-, pyro-, piezo-ferroelectric crystals.

The crystals were grown in the systems  $\text{Sb}_2\text{O}_3\text{-Nb}_2(\text{Ta}_2)\text{O}_5\text{-R-H}_2\text{O}$  (R -  $\text{KF}$ ,  $\text{KHF}_2$ ,  $\text{K}_2\text{CO}_3$ ,  $\text{H}_2\text{O}_2$ ) at the various admixtures which were typical of natural mineral. The kinetics of transfer and crystal growth were found to be determined by several parameters which were connected with each other. These parameters were the following: temperature, temperature gradient, Eh-pH value, solvent concentration, molar ratio of starting components in nutrient.

The crystals of  $\text{SbNb}(\text{Ta})\text{O}_4$  with the size of 3-4  $\text{cm}^2$  were grown; the composition of the crystals was similar to that of the natural mineral.

On the basis of the experimental data the scheme of the genesis of natural stibiotantalite single crystals in aqua fluoride and carbonate solutions was proposed.

THE GROWTH AND EXAMINATION OF OPTICAL PROPERTIES OF GEM ALEXANDRITE

V.N. Matrosov, G.V. Bukin, V.P. Solntsev, E.G. Tsvetkov, A.V. Eliseev, E.I. Kharchenko, Novosibirsk, USSR

The crystals of alexandrite about 100x20x10 mm in size were grown by Czochralski method on oriented seeds from melt stoichiometric to the chrysoberyl with  $\text{V}_2\text{O}_3$ ,  $\text{Cr}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  dopings.

Even at high growth rates as high as 5 mm/hour the faces  $\{100\}$   $\{010\}$  and  $\{120\}$  at the growth in the direction  $[001]$ , the faces  $\{010\}$  at the growth in the direction  $[001]$ , the faces  $010$  at the growth in the direction  $[100]$  and the faces  $\{100\}$  at the growth in the direction  $[010]$  are exhibited. High rotation rates of the crystals growth up to 60 rev/minute lead to disappearance of the faces  $120$ .

The isomorphous admixture of  $\text{Cr}^{3+}$ -ions colours the crystals to cherry-red colour in the direction  $[001]$ , blue-green in the direction  $[010]$ , yellow-green in the direction  $[100]$ .

The content of  $\text{Cr}^{3+}$ -ions at directions of the crystal growth  $[001]$ ,  $[100]$  decrease along the crystals length from the seed within the range of 0.114-0.054% wt. More regular distributions of  $\text{Cr}^{3+}$ -ions (over 0.176-0.109% wt.) was obtained in  $[010]$ -direction of the crystal growth.

The investigation of EPR spectra in synthetic and natural alexandrites has shown that  $\text{Cr}^{3+}$ -ions substitute  $\text{Al}^{3+}$ -ions in two different structural positions: 65-80% wt of  $\text{Cr}^{3+}$ -ions substitute  $\text{Al}^{3+}(\text{II})$  (point symmetry  $C_g$ ,  $\text{Al-O}=1.938$  A) and 35-20% wt of  $\text{Cr}^{3+}$ -ions substitute  $\text{Al}^{3+}(\text{I})$  (point symmetry  $C_i$ ,  $\text{Al-O}=1.890$  A) as a function of the growth conditions.

The absorption spectra of synthetic and natural alexandrites are studied in polarised and unpolarized light and their color characteristics are calculated.

It is established that the colour shade is dependent upon the ratio of  $\text{Cr}^{3+}(I)/\text{Cr}^{3+}(II)$  and may be made close to natural alexandrite shade of colour.

#### ANALYTICAL THEORY OF MULTICOMPONENT SYSTEM CRYSTALLIZATION

T.A. Cherepanova, Riga, USSR

The description of crystal growth from melt and solution in terms of statistical mechanics is given here. The basic physical model is limited by consideration of the nearest neighbour particles interactions in a crystalline lattice. The bond energies describing these interactions correspond to the main state of the system. The entropy factors are introduced as characteristics of the excited state spectrum. The connection is obtained between the probabilities of elementary events accomplishment on the surface of growing crystal faces and thermodynamic characteristics of the metastable system. General approach worked out here makes it possible to give an analytical description of multicomponent system crystallization kinetics, both for the case of a completely mixed liquid phase, and considering diffusion processes in it. The set of equations describing the binary crystal growth is obtained in a mean field approximation and in a pair approximation of distribution function as well. The essential influence of diffusion processes on the kinetics of crystalline phase structure formation is shown. Comparison of the obtained results with the experiment has been made.

#### KINETICS OF CRYSTAL GROWTH FROM SOLUTIONS: DEVELOPMENTS IN THE APPLICATION OF CRYSTAL GROWTH THEORIES TO GROWTH FROM SOLUTION

P. Bennema, Ed Nijmegen, Netherlands

Firstly a brief survey will be given on recent developments in computer simulation of the crystal growth process. The attention will be focussed on:

- (i) rate versus  $\Delta\mu/kT$  curves
- (i) very recent developments in the concept of the roughening transition of a continuous to a layer growth mechanism.

Secondly ways will be discussed how to bridge the gap between computer simulations of the crystal growth process and experiments on crystal growth from solution.

Special attention will be focussed on the problem how Jackson's  $\omega$  factor, which in the simulation determines the surface properties of the surface, may be calculated for a real solid-saturated solution. For this calculation we use:

- (i) the crystallographic morphological theory of Hartman-Perdok
- (i) thermodynamic models for the solution.

Also ways, how, to calculate the driving force for crystallization  $\Delta\mu/kT$  will be discussed.

Thirdly calculations will especially be compared with some experimental data, which suggest the phenomenon of surface roughening.

If time permits we will discuss some observations of spirals, which under the influence of strain show a very peculiar behaviour. This can be explained with recent generalizations of the spiral theory of Cabrera and Levine.

MORPHOLOGY OF NATURE AND SYNTHETIC CRYSTAL OF FLUORITE IN CONNECTION WITH THE CONDITIONS OF CRYSTALLIZATION

A.F. Kuntz, Syktyvkar, USSR

Materials received during the research on hydrothermal synthesis of fluorite allow considering some problems of natural crystallogenesis.

The evolution of simple forms and their combinations in the process of growth of natural crystals comes to the sequence of  $\{III\} \rightarrow \{110\} \rightarrow \{100\}$ , other simple forms -  $\{211\}$ ,  $\{311\}$ , etc. are "intermediate", that usually is connected with the drop of the temperature of crystallization ( $t$ ) and the supersaturation ( $\Delta t$ ) of the mineral-forming solutions or with the change of their composition. The determination of  $t$  by inclusions does not allow to define clear boundaries of the change of crystal forms. The estimation of the influence of  $\Delta t$ , composition, concentration and pH solutions on the change of crystal morphology on the basis of only geologic observations is practically impossible.

The influence of  $\Delta t$  on the morphology of crystals has been studied in the process of the fluorite synthesis in 44% LiCl solutions ( $t=360^\circ\text{C}$ ,  $\Delta t = 0,6-1,2$  degree per cm). The obtained data do not confirm the views on the leading role of the supersaturation of solutions in the change of the crystal forms, as with the growth of  $\Delta t$  only an increase of the number of crystallization centres takes place, with the replacement of individual well-faces crystal by their aggregates and by the complication of the structure of the crystal face surface.

A study of the influence of the composition, the concentration and pH of the solution on the morphology of synthetic crystals of fluorite has been carried out with the use of solution of HCl, LiCl and others with  $\Delta t = \text{const}$ . As a result, definite regularities in the change of crystal morphology have been stated in the range from 100 to 500°C and  $P_{\text{H}_2\text{O}}$  from 50 to 1000 bars.

Thus, for example, the boundary of the form changing from  $\{100\}$

to  $\{III\}$  in  $tp$  coordinates, for the crystals grown in 4% HCl solutions (pH 0,05), lies through the following points:  $t_1=100$ ,  $p_1=150$  and  $t_2=300$ ,  $p_2=50$ ; 44% LiCl (pH 6,80) -  $t_1=230$ ,  $p_1=600$  and  $t_2=500$ ,  $p_2=300$ ; 3%  $\text{NH}_4\text{Cl}$  (pH 5,04) -  $t_1=350$ ,  $p_1=1000$  and  $t_2=500$ ,  $p_2=700$ ; 5%  $\text{NH}_4\text{Cl}$  (pH 4,90) -  $t_1=300$ ,  $p_1=1000$  and  $t_2=500$ ,  $p_2=500$ ; 7%  $\text{NH}_4\text{Cl}$  (pH 4,85) -  $t_1=250$ ,  $p_1=1000$  and  $t_2=450$ ,  $p_2=400$ .

Thus the leading role in the change of the crystal morphology of fluorite is played by the  $tp$  conditions of their growth. The solution composition, their concentration and pH influence the position of the boundary where one form turns to another. The range of  $t$  and  $p$  formation of octahedral crystals is wider in an acid medium and for the cubic crystals this is true in weak-acid and subneutral conditions. The wide occurrence of fluorite crystals of cubic faces in nature, formed under comparatively low  $t$  and  $p$ , testifies to their crystallization from moderate alkaline or subneutral solutions.

**ORE MICROSCOPY**



OPTICAL ELLIPSOMETRY AND ITS IMPORTANCE FOR THE MINERALOGY  
OF TO-DAY

A.D. Rakcheev, Moscow, USSR

Ellipsometry is a new sensitive and precise polariso-optical method based on analysis of the light polarization changes when the light is met by the solid body surfaces. The last years have seen a growth of interest to this procedure in a number of branches of science and technology, including the mineralogy (France, Japan, USA and USSR). This interest is stimulated by the line production of ellipsometers in the USSR and abroad.

The optical ellipsometry is practiced in a series of applications. In Moscow University mineralogical laboratories the ellipsometric method is also used to obtain optical constants of the minerals from the major angle of incidence and from the major azimuth. It is the most convenient and precise method of obtaining parameters in the range of the visible and the nearest IR-spectra.

The ellipsometry advantages in the study of the strongly absorbing crystals are spectacular - it is possible to have 5 to 6 optical constants (refraction, absorption and reflection indexes) in common polished sections in a comparatively great range of the spectrum. The method permits us to obtain the optical constants using no standards. It is good to determine the refraction indexes of ore minerals and of such materials of high and small index values as garnets, zircon, etc.

The optical ellipsometry is utilized in mineralogy to:

- 1) diagnose the minerals;
- 2) make evident their polymorphic particularities;
- 3) determine (using formulae) pair and multiple regressions in the chemistry of minerals;
- 4) determine some energetic characteristics and predict some other physical properties of the minerals;
- 5) take part in the geological thermometry.

S.I. Lebedeva, Moscow, USSR

The study in the field of micro-hardness of the minerals is being carried out now in several new directions.

The new indentation test instruments with continuous kinematic registration (Alekhin V.P., Berlin G.S., Kaley G.N. et al. and Korneev A.I., Moiseitseva Z.K. et al.) make it possible now to have an experimental model to check the minerals of Mohs scale of hardness and some other ore minerals (galenite, pyrite and molibdenite). Conventional elastic ( $K_{el} = \frac{h_{rec}}{h_{prod}}$ ) and plastic ( $K_{plast} = \frac{h_{unrec}}{h_{prod}}$ ) indexes are estimated.  $K_{el}$  is a relation of the recovered depth of indentation ( $h_{rec}$ ) to the depth initially produced ( $h_{in}$ ), the indenter being in place.  $K_{plast}$  is a relation of the uncovered depth to the depth initially produced. The  $h$  parameters are taken from the indentation diagrams.  $K_{el}$  is found: 0,35 for talc; 0,27-0,31 for gypsum; 0,37 - for calcite; 0,39 - for feldspar; 0,42 - for fluorite; 0,34 - 0,43 - for quartz; 0,53 - for topaz and 0,21 - for galenite.

Three homologous mineral types are specified in order to enlarge the sphere of application of the anisotropy coefficient for the hardness of the 2-nd order ( $K_{h2}$ ), when it is correlated with the structural particularities of the minerals. The type I unite the minerals of cubic syngony with very small  $K_{h2}$  (1,00 - 1,12). The minerals of tetra- and hexagonal as well as mono- and triclinic syngonies form the type II group with  $K_{h2} \sim 1,12 - 1,50$ . The homological type III correspond to the rhombic syngony. These minerals have as a rule two  $K_{h2} > 1,50$ .

In the last years studies the micro-hardness data for the common and the overall distributed minerals are used as a typomorphic index when analysing the ore genetic laws. A good use of the microhardness parameters ( $H$ ,  $K_h$ , etc.) and of the newly elaborated microfragility scale may be made in the modern technology as well.

T.N. Chvileva, M.S. Bezsmertnaya, Moscow, USSR

Any observation and analysis of the optical effects in ore microscopy is based on images perceptible and estimable by our eyes. In other words, the perception is highly relative. The quantitative character of reflections makes it possible to quantitatively estimate the capacities linked to the reflections: anisotropy, colour and the optical contrasts of these effects perceptible by our eyes for the analysis.

The reflection studies we made permit us to state the following:

a) the anisotropy expression in  $R_{rel} = \frac{R_{absol}}{R_{max}}$  has a great informative value; b) the mineral colour expression in  $B$ ,  $\lambda$  and  $\rho$  parameters is highly advantageous.

A colorimetric method of description of the mineral colours was earlier proposed by H. Piller (BRD, 1966) and by V. Htein and R. Phillips (Great Britain, 1973). We have calculated the colours of 200 ore minerals. A comparative analysis using real visual colour images permits us to make the following statements: 1. The ore microscopy is a special branch of the visual precise colorimetry for the polycomponent mineral aggregates with mostly weak colours. In these highly abnormal conditions of colorimetry the calculated colour does not wholly characterize the visual image, as the image formation depends largely on colour transformations caused by the simultaneous contrast and the brightness level differences in minerals. 2. The limits of this effect were determined in calculation of colour when two sources (A and B) of white light and the colorimetric purity graphs to utilise  $B$ ,  $\lambda$  and  $\rho$  parameters were used. 3. The data obtained witness against the opinion that the colour perception in microscopy is subjective.

## OPTICAL STUDY OF ACANTHITE, BISMUTHINITE AND GALENA

M. Vendrell-Saz, S. Karup-Møller and A. Lopez-Soler,  
Barcelona, Spain; Lyngby, Denmark

The reflectance in air and in oil (or glycerin) has been measured by quantitative microphotometry at normal incidence, on oriented sections of the three minerals.

The measurements have been obtained from 250 nm to 1100 nm, each 10 nm, in order to know the dispersion curve of its optical properties from the ultraviolet to the near infrared.

With these values we have calculated the dispersion curves on  $n$  (refractive index) and  $k$  (absorption coefficient) of each mean direction of vibration.

Studying all the curves we intend to make an interpretation of the optical behaviour of these three minerals. And we can observe that acanthite and bismutinite have similar dispersion curves, because their electronic structure is also similar. They have an energy gap between the conduction and the bonding band of 1,0 eV and 1,2 eV, respectively.

The different behaviour of galena is due to its energy gap of 0,37 eV.

On the other hand, the curves of the dielectric constants are very similar in the three minerals, showing a narrow band of frequency in which the behaviour of galena is nearly metallic. This happens in the ultraviolet range of the spectrum.

## ON SYSTEM OF AUTOMATIC DIAGNOSTICS OF ORE MINERALS

N.I. Schumskaya, Leningrad, USSR

The system of automatic diagnostics for main, widespread accessory minerals (96, except platinum) of all types of ore deposits has been worked out by the group dealing with ore microscopy from VSEGEI.

The method is based on the colour parameters of minerals  $Y, y, x, \frac{X}{Y}, \frac{Z}{Y}$  calculated from the spectral reflection curves of the  $E$  source; three main curves of spectral reflection being determined for greater part of biaxial minerals.

The limits of variations of the above parameters are determined with regard to scattering of data due to the character of spectral curves as influenced by the quality of polishing, admixture elements and orientation grains.

The 65% of minerals from all included into the system are diagnosed with certainty, the others are grouped in two or rarely three minerals. Sulphoantimonides of lead are well separated from other minerals but inside the group they are distinguished only when orientation of grains is good.

The distinct feature of the given system is its complete objectivity, i.e. it is independent of the observer perception of the colour and character of surface of the grains.

The investigator puts the necessary grain under optical sound, all other operations being done mechanically including the name of mineral. All the parameters of colour are put out simultaneously with the name.

This system is very simple and it may be realized by the students of the 4-or 5-th year. The minimum size of analyzed grains is 5  $\mu\text{m}$  when we used the microspectrophotometer MPN-10 of home production.

The rate of diagnosis is determined by the rate of obtained spectral reflection curve +3,0 min in mass analyses. The system is intended in the first place for conducting express diagnostics of the ore minerals in industry.

M. Vendrell-Saz, J. Nogués-Carulla, M. Font-Altava,  
Barcelona, Spain

In this work we intend to establish a differential behaviour of the different iron minerals, from an optical point of view.

To obtain this, the reflectance in air and oil (or glycerin) has been measured from 250 nm to 1100 nm, each 10 nm. And with these values we have calculated the optical constants  $n$  (refractive index) and  $k$  (absorption coefficient), using Frehel's formula.

The measurement and calculation of these curves with well known and single structure minerals, permit us to establish a general behaviour of each group.

We observe in the native iron a typical metallic behaviour, with a large  $n$ , and a large  $k$ . This shows a high electric conductivity.

On the other hand we have the iron oxides (magnetite and hematite), whose optical behaviour is similar to the dielectric materials, with  $k$  almost zero, and  $n$  coming down to the infrared range of spectrum.

Between these two groups there are the iron sulphides (pyrite, marcasite and chalcopyrite), which have a thin energy band between the conduction and bonding bands. This is produced when the energy is enough. Some electrons can skip to the conduction band, and the material shows a more metallic behaviour in this frequency. They are semiconductors, as show their optical characteristics.

L.N. Vyal'sov, Moscow, USSR

The analysis of the theoretic formulae proposed by M. Bereck in 1937 permits us to establish a link between the anisotropy parameters and the polarised light reflection coefficient of the anisotropic crystals. It is supposed, at that, that the light incidence is normal and the ellipticity of the waves entering the crystal is not far from zero. The latter condition is practically always real for the most part of the anisotropic ore minerals.

The colour effects of anisotropy are characterised by a coefficient  $(R+)_{\max}$  which is calculated from the constants " $n$ " and " $k$ " of the mineral or may be found in an experiment of the light intensity measurement when this light is reflected from the surface of a section diagonally placed under the crossed nicols.

I propose a new formula to determine  $(R+)_{\max}$  :

$$(R+)_{\max} = \bar{R} - \bar{R}_{(\parallel)}$$

where  $\bar{R}$  is the reflection coefficient measured when the section is oriented diagonally and one nicol is used; and  $\bar{R}_{(\parallel)}$  is also the coefficient but measured when the nicols are parallel.

$(R+)_{\max}$  value changes in the spectrum as well. An analysis of the curve  $(R+)_{\max}$  reveals  $\bar{x}$ ,  $\bar{y}$  and  $\bar{z}$  coefficients that characterize the visual perception of the colour effects of anisotropy.

$(R+)_{\max}$  value for a number of anisotropic ore minerals, including sign-changing ones, have been obtained.

The new method notably contributes to the possibilities of studies of the anisotropic ore mineral optics.

DIAGNOSTICS OF ORE MINERALS BY THEIR ELECTRON  
AND VIBRATION SPECTRA

E.M. Muratov, Alma-Ata, USSR

The reflection spectra of minerals measured within the frequency interval of the visible light (from 400 to 700 nm) are widely used in the studies of ore composition under the microscope for the diagnostics of minerals. However, it is sometimes difficult to obtain unambiguous diagnostics because in this narrow zone of the wave lengths there are many minerals with similar spectral characteristics. Therefore, we have carried out the study of the nature of reflection dispersion in galenite, pyrite, chalcopyrite, tennantite and tetrahedrite in a wide spectral range (0.01-6.00 eV). Maxima of various intensity are manifested in the reflection spectra of galenite and pyrite in the region of their own absorption, with following energies: in galenite - 5.14 eV, doublet 3.53 and 3.63 eV, 1.87 eV and in pyrite - 5.14 eV, 3.92 eV, 1.76 eV. The energy position of maxima and their intensity in the reflection spectra of these minerals is connected with the values of the cross-over interzonal transitions at various points of the Brillouin zone and with the value of the density of states in both energy zones. In the infra-red region the absorption has the resonant nature, its maximum being achieved when the frequencies and wave vectors of the optical phonon and photon are similar in value or coincide. The oscillation frequencies of the crystal lattice of pyrite are within the range of 33-62 meV, the doublet structure of the reflection band with the energy of 51.6 meV and of the band - 42.7 meV, 38.7 meV and 36.0 meV. The structure of the vibrational spectrum of the cubic chalcopyrite is manifested within the range of 40-50 meV with the reflection bands of 39.8 and 44.7 meV. The region of the residual rays of the grey ores ranges between 27 and 50 meV. Within this spectral interval the reflection bands with maxima are manifested at 42.7, 38.1, 31.4 meV in tennantite and 40.0, 36.4 and 30.2 in tetrahedrite. The results obtained show that for the diagnostics of the ore minerals the electron and vibration spectra in which the inten-

sive reflection bands are manifested, are of the greatest importance. Therefore, we have suggested the method for the identification of minerals by their reflection spectra in the regions of their own absorption and residual rays.

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