# NOTES ON MINERAL GROWTH FROM THE ARCHIVE OF
# V.I. STEPANOV (1924 - 1988)

by

## V.I. STEPANOV

## ABSTRACT

This paper comprises three separate texts, compiled from the unpublished archive of the late V.I. Stepanov. The texts all discuss the examination of both structure and texture during the study of mineral aggregates. The first text demonstrates how the Curie Universal Principle of Symmetry can be used in the study of mineral aggregate structures and textures. By this means some mineralogical problems were solved, for example: so-called "collomorphic" aggregates of lussasite and chalcedony crystallise from real solutions, not gels. During this study, it was found that the structure of an aggregate is determined by the position of the growth centres of individuals and by the kinetics of the crystallisation process, not by the phase state of the crystallisation medium. A study of metasomatic substitution by todorokite in marine volcanic tuffs confirmed this. It was also found that different structures may form during synchronous crystallisation of a homogeneous aggregate. Application of the Curie Principle to the whole crystallisation space revealed a new and more exact definition of texture. The second text examines the factors affecting the morphology of cave mineral aggregates. These are: crystallisation kinetics, method of transfer of the feeding solution, and mass-transfer symmetry in the medium of crystallisation. The typical forms of subaerial aggregates and ensembles are described. Some paragenetic ensembles have such distortions of structure, texture and morphology that they are best described as hybrid textures; names for these textures are suggested. A new term "kora" is proposed for an association of aggregates generated by the same crystallisation environment in all the space of synchronous crystallisation. The third text proposes that the term "typomorphism" should be returned to its original meaning: occasional and individual features in the morphology of minerals. An example is given of the very varied and individual corallite textures that grow in the subaerial conditions of karst caves.

## FOREWORD

Several years ago Russian mineralogy lost one of its most brilliant researchers, V.I. Stepanov. Victor Ivanovitch was perhaps the best diagnostician of minerals that Russia has ever known. If someone came to a Moscow museum with a strange mineral sample, he would be told, "Visit Stepanov. If anyone can give an answer, he can." And Stepanov always gave his time, even for children.

Professionally, Stepanov worked at the Fersman Mineralogy Museum as a chief of department, though he was (informally) regarded as second only to the Museum Director, Academician Godovikov. He was a key member of a team that studied mineralogy not as an independent subject, but in connection with other disciplines. He was also a bridge between the professionals and all mineralogy-related amateur societies in Moscow. Once or twice a year, he gave lectures at Moscow State University which were highly popular, both with geologists and with ordinary cavers. Though he was purely a scientist, cavers held him in high regard and any team was happy to have him join their expedition.

Unfortunately, Victor Ivanovitch was one of those scientists who are always willing to share their knowledge by word of mouth, but never publish their works until they have been completely perfected. As a result of this he left only about ten printed articles along with a huge archive of samples, jottings, notes, drawings and photographs, now stored in the Fersman Mineralogy Museum of the Russian Academy of Sciences. A significant part of the archive is on the subject of crystallisation in free space, basically on material from karst caves.

In 1994, an attempt was made to systematise the part of the archive relating to crystallisation in karst caves. At the instigation of Paolo Forti, the Italian Institute of Speleology allotted a grant and two members of the museum staff, T.I. Matrosova and A.B. Nikiforov carried out the work. They found two variations of notes for a dissertation on crystallisation in free space, which Stepanov obviously intended to present for a Doctor of Science degree (a rare and very high honour in the Soviet Union, representing a lifetime's work). They also found several outlines of his famous lectures for mineralogists and cavers. (However, these were written very concisely, so it has been necessary to supplement them from student notes).

This paper was compiled by V.A. Maltsev from some of these fragments, dated between 1969 and 1985. Some details, certainly, have become obsolete. Some terms that were not published at the time have been partially replaced by alternatives, or have been given another interpretation. However, the completeness of his work and his system of approach may still be appreciated as of the highest standard in research of this kind.

The ordinary caver may possibly be surprised that this work begins with a treatise on ore bodies, not caves. But one of the great values of Stepanov's work is that he did not separate common mineralogy from cave mineralogy. He regarded caves as an excellent testing area for research into rare mineralogical situations. If the names of the minerals in the first part of this work are changed to those which are normal for caves, for example calcite instead of chalcedony in the discussion on crusts, a typical cave set-up will be seen. Also, this fragment is the only text that has been found which clearly shows the methods and conceptual basis of Stepanov's approach to mineral ontogeny. Without understanding this, all the "purely speleological" applications cannot be understood.

V.A. Maltsev, T.I. Matrosova, A.B. Nikiforov, P. Forti.


EDITORIAL NOTE

As this paper has been compiled from preliminary drafts of text and from notes, it is not a "finished product" and cannot be treated as such. If the author were still living, it would be possible to ask him to expand some sections or to rewrite them for greater clarity. Instead this paper has been treated as a historical document and the text has been supplemented with footnotes and a glossary, written by V.A. Maltsev and C.A. Self. The main text remains true to the author.

The original compilation in the Russian language, with footnotes by V.A. Maltsev, has been awaiting publication in Moscow for some time. The main differences between that text and this is the addition of further footnotes and a glossary to the English version. This translation was originally made in a very "strict" form, with a style and grammar typical of Russian scientific writing. C.A. Self and G.I. Self worked with the translator to produce the present version. K. Ray transcribed the text for the two figures. Within the text, italics are used to denote quotations and bold to denote the Author's own emphasis.

This paper was given to C.A. Self by V.A. Maltsev as part of a collaborative project to bring the Russian view of genetic mineralogy, in particular mineral ontogeny, to a western audience. This has now become one of the main projects of the newly formed Mineralogical Commission of the Union Internationale de Speleologie and further translations from Russian cave science are planned.

# STRUCTURES AND TEXTURES OF MINERAL AGGREGATES FORMED IN THE FREE SPACE OF VOIDS[1]

*Only a combined examination of data at different scales*
*can give positive results, when research of*
*ore body structures... is co-ordinated with the systematic*
*study of ore textures. By this way one can avoid an incorrect*
*approach to the study of ores which leads into a blind*
*alley for many investigators of ore formations.*

A.G. Betekhtin

The beginning of this work dates from the 1950's, when it was inspired by the publications of D.P. Grigoriev and G.G. Lemmlein (Grigoriev, 1946, 1948, 1949a, 1949b, 1951, 1961; Lemmlein, 1945, 1946, 1948). These publications expressively and clearly displayed the real structure and the geometrical elements of growth of mineral individuals and some of their aggregates. It seemed that the methods worked out by these authors for the products of crystallisation in voids would be suitable for reconstructing the geometry of mineral aggregate growth in veins of wolfram formations which at the time were being studied by the author. Even now, publications on the subject affirm that massive vein fillings appear in gaping fault fissures because of druse growth of vein minerals on the crack walls.

Observations of the structure of the veins were carried out by the author on many tens of cut samples and hundreds of thin sections, using all criteria known at the time of combined and consecutive growth of mineral grains. This led to a completely unexpected result. All of the volume of a vein is a whole homogeneous body, and all the structural details, without exception, form one system. This can apply only if the following version of their crystallisation is correct: origination under conditions of constant collecting recrystallisation all through the volume of the vein, synchronised with a pulsation supply of the material by constantly renewed capillary cracks which are being widened by the supplied material and, possibly, by crystallisation pressure. If so, then gaping cracks never existed, but crystallisation continued all through the solid volume of the vein.

This conclusion so contradicted the generally accepted opinion that the author did not have enough courage to come out with his results. Moreover he was still in some doubt - as only the simplest types of mineral aggregates, which crustified voids, had been investigated at the time and general mechanisms of such crystallisation were not at all clear.

Thus emerged a dilemma: solid-phase crystallisation, or growth on a hard matrix in free space. To solve the puzzle, the author carried out a series of experiments. The study of the geometric pattern of growth of mineral aggregates formed in the free space of voids is methodically much easier to do than that of any other situation, because their crustification structure is usually clearly displayed. That is why clearing up the general mechanisms of this type of crystallisation seemed an easier part of the task. However, there was a significant obstacle. Many guess that so-called "collomorphic" structures of mineral aggregates are secondary and appear due to

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[1] In simple terms, structure describes from which individuals an aggregate is built and the manner of their bonding together. Texture describes the general idea of construction. To take as an example: a brick wall. The geometry of the individual bricks and the distance between bricks, this is the structure of the wall and may be different for different walls. The manner of how the bricks are oriented is the texture and from this comes how strong the wall will be, is it vertical, etc.

the recrystallisation of primary colloid depositions (gels)[2]. Detailed investigation of the structure of these aggregates, first made by Yu.M.Dymkov (1957, 1960, 1962, 1966a, 1966b,) and later by other authors, pointed at their crystallisation structure. These works initiated controversy between the supporters of opposing points of view. The cardinal question, which lacked an answer at that time, was of the possibility in principle of the formation of spherulite crusts from the recrystallisation of colloid depositions. The author demonstrated that this was definitely not the case (Stepanov, 1970).

The classic examples of "collomorphic" aggregates - smooth spherulitic crusts of lussasite ($\beta 2$-cristobalite) and chalcedony ("glass heads") were the subject of investigation. The structure and texture of such crusts were seen in the light of contemporary facts of physics concerning crystallisation mechanisms. Three aspects, proven by extensive experimental findings, are essential:

1.       The crystal nuclei (the centres of growth of future grains and spherulites) appear in conditions of nucleation in supersaturated solutions. The probability of their appearance on a hard substrate bounding the solution, or on hard particles (micellae) suspended in it, is many orders greater than for a clear solution. The reason is a reduction of the threshold activation energy of crystallisation, due to epitaxy and adsorption on a solid body. In the limiting case, the growth speed is determined by the diffusion speed.

2.       The speed of diffusion of dissolved materials in real solutions[3], sols and gels is practically the same. This fact is easy to understand, taking into account that the dispersed fluid of a colloid constitutes a real solution in dispersed phase.

3.       Data on the viscosity of real solutions, brines, hydrosols, jellies[4] and melts have been used here. The rate of sinking of quartz particles in viscous solutions (for example glycerine with viscosity 20 poise - 2000 times more than a real aqueous solution) is calculated by Stokes' Law. It is shown that the speed of nucleus growth is at least commensurate with their sinking speed. In more viscous fluids, such as jellies, gels and silicate melts, the crystals grow to a significant size while staying in suspension.

Examination of the simple growth model revealed that in real solutions and sols there is normal growth of the spherulite crust. In viscous gels, jellies or melts, the nuclei grow through all the crystallisation space with formation of massive granular or spherulitic bodies. A contrary model was also considered, where it is supposed that the above arguments have defects and the spherulite crust grows from the gel material – the jelly. The impossibility of continuation of such growth was shown.

Finally, a third independent growth model was examined on the basis of application of the Curie Universal Principle of Symmetry. The general formulation of this principle says that *if any cause leads to any effect, then the symmetry of cause must be reflected in the symmetry of effect*[5]. An auxiliary concept, facilitating the application of this principle, is the one of

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[2]       It is possible to meet this understanding of "collomorphic" aggregates of chalcedony even now, for example in Turchinov (1993).

[3]       For these and other terms, see Glossary

[4]       Stepanov uses "jelly" as if it were a separate term, but it is simply a variety of "gel".

[5]       The precise form of words used for the Curie Principle of Universal Symmetry can differ slightly between authors. The original source (P. Curie, 1908, reprinted from 1894) does not express this Principle in such clear language as Stepanov uses.

characteristic symmetry: *the characteristic symmetry of a phenomenon is the maximum symmetry compatible with its existence[6]*.

Let us consider the characteristic symmetry of mass transfer of material in the example of crystallisation in a viscous fluid, and the symmetry of structure and texture of the emerging aggregate. In consequence of the above, the nuclei, which have grown to a significant size in suspended condition, cannot stay unaffected by the pull of gravity. Gravity restricts the symmetry of the fluid and thereby causes dissymmetry (the Curie Principle defines dissymmetry as an incomplete symmetry in comparison to the maximum possible for the phenomenon). In this case it is shown by the formation of aggregates in the lower part of cavities bounded above by the horizontal surface of the viscous solution (gel, jelly, melt); that is to say it has a gravity character. The typical examples of such aggregates are agates of so-called "Uruguayan type", showing a well-defined horizontal zonality in the layers of deposits of silica gel, quickly hardening (jellying) and then turning into a spherulitic aggregate of chalcedony in the way of crystallisation described above. The characteristic symmetry of this texture is a cylinder, with axis vertical relative to the horizon. In accordance with the Curie Principle, *when actions reveal some dissymmetry, this dissymmetry must also be found in the causes which generated them.* Obviously, the cause is the dissymmetry of the Earth's gravity field, having the same characteristic symmetry of a cylinder in any point on the Earth's surface.

The structure of a "collomorphic" spherulitic crust differs by its disorderly distribution of spherulites of the same size on the surface of growth, with a clearly expressed geometric selection (the growth optotropism) between them, like that of usual crystal druses. Characteristically, the individual spherulites of crust are externally bounded by a proper spheroid surface and have a concentric zonality, with the same thickness of appropriate zones over all the surface of the aggregate. It is possible to define the texture of the homogeneous aggregate only by observing the variations in structure (or the absence of these) through all the volume of synchronous crystallisation. In this case, it is characterised by identical structure of the "collomorphic" crust on the lower, upper and side walls of the cavity, i.e. the formation process is not affected by the pull of gravity. As seen from the description, the characteristic symmetry of the crust structure is that of a cylinder, i.e. with a clearly defined dissymmetry relative to the surface of growth caused by the dissymmetry of the forces of adsorption, having the same characteristic symmetry relative to the liquid-solid interface[7]. The characteristic symmetry of the crust texture reflects an identical structure over all the surface of the cavity, i.e. it corresponds to the symmetry of a sphere, pointing to the diffusion mass transfer of material by crystallisation.

That is how the structure of a meta-colloid aggregate, forming by the crystallisation of jelly (or gel), has the characteristic symmetry of a sphere but its texture has the symmetry of a cylinder[8]. The structure of the "collomorphic" spherulitic crust has the characteristic symmetry of a cylinder but the texture has the symmetry of a sphere. Consequently, these two aggregate types are inversely symmetrical. In accordance with the Curie Universal Principle of Symmetry, their crystallisation was affected by different causes, i.e. by crystallisation in gels, the forces able to create the structure and texture of a spherulitic crust could not occur. The above shows that the three independent models of growth of spherulitic crusts result in the same:

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[6]      This definition uses the exact words of P. Curie (1908).

[7]      The last part of this sentence is not at all clear. The crust structure has cylindrical symmetry, which is always perpendicular to the surface of growth. This feature is identical at any point, but the axes are oriented differently in different parts of the cavity. This reflects the cylindrical symmetry of the forces of adsorbtion on the phase boundary, but spherical symmetry of the diffuse mass-transportation in the cavity: two levels of control, one for structure, the other for texture.

[8]      Very similar textures also appear by crystallisation from heterogeneous fluids (Dymkov and Sletov 1981), including in hydrothermal karst caves.

collomorphic aggregates cannot be the products of crystallisation or recrystallisation of gels, but are the products of spherulitic crystallisation from real solutions.

Some general conclusions follow from the above data. It seems without doubt that the position of growth centres of the individuals composing the aggregate are the deciding influence on the origination of one or another structure of aggregates, not the phase state of the crystallisation fluid. It is evident that, in the general case, it is impossible to form an opinion on the phase state of a crystallisation fluid from the structure of aggregates.

In order to check this version, the author carried out a study on druse structures of spherulitic crusts and spheroidalites (elliptical spherulites) of the manganese oxide mineral todorokite. The structures have grown by way of metasomatic substitution of stratified marine deposits of volcanic tuffs in manganese sites of Eastern Cuba (Sokolova, Stepanov, Brito and Kowtin, 1971). It has been found that three types of todorokite aggregates are being formed, depending on the correspondence between crystallisation speed, nucleation kinetics (the quantity of formed centres of growth of individuals) and addition of material to the growing mineral:

1.        Massive layered ores, fully following the texture pattern of the surrounding rock. Their heterogeneous structure is represented by fine-grained aggregates of todorokite, which replaced the finely dispersed part of the rock, cementing bigger fragments and porphiric disseminations of olivine, pyroxene and feldspar (the cemented structure of pseudomorphism).

2.        Crusts of kidney-shaped concretions with cemented structure similar to the first type of aggregates. The aggregate outlines are sharp, gradual changes are visible from kidney-shaped ("curly") concretions to roughly branching dendrites of the same structure.

3.        Spherulitic and spheroidalitic coarse-fibred crusts of a different degree of complexity which fully replace the rock, having a sharp external contact with the surrounding volcanic tuff. (It is proposed to give the name "spheroidalites" to such types of spherulites, which have an elliptical structure but not the usual spherical concentric zonality of growth. In spherulites, the fibres diverging from the centre are rectilinear, but in spheroidalites they are bent outwards from the axis of symmetry. Spheroidalites, as well as spherulites, may be regarded as individuals able to form various aggregates).

The spherulitic crusts of todorokite have a typical druse structure with geometric selection between separate spherulites and do not differ from the classic crusts of chalcedony. At thickenings of the separate layers of crusts, a gradual change of the spherical restrictions of separate spherulite concentres into elliptical ones is visible on chips of rock. This is expressed first of all by flattening of the top and by an increasing steepness of angle of the concentre surfaces in the side parts of the spherulite, relative to the crust base. In this way, spheroidalite druses are formed with a flattened kidney-shaped surface and very clearly expressed geometric selection of spheroidalites. Such aggregates, but grown in voids, are characteristic of malachite. On chips of the massive spheroidalites of todorokite, there is often a specific glimmering shine testifying to the axial twisting of fibres, like that of chalcedony. Gradual passage of massive spheroidalitic crusts into roughly branching dendrites was observed. Sometimes branches of fine-grained todorokite grow on kidney-shaped caps of such dendrites, with a cement structure inheriting the form of the dendrite as a whole. This fact is a supplementary argument for the metasomatic growth of spherulitic aggregates of todorokite.

The structure of the aggregates described above is identical in every detail to that of typical crustification crusts grown in voids. Examples of druse spherulitic structures which grew in the metasomatic way were known previously, for example such aggregates of hedenbergite and wollastonite from the Dalnegorsk polymetal mines. This data testifies to the validity of the thesis mentioned above; mineral aggregate structures are not determined by the phase state of the crystallisation fluid, but represent exclusively the kinetics of the crystallisation process. This conclusion does not mean there is no dependence between the aggregate structures and the phase state of the crystallisation space, but they do not have an absolute character. The examples given demonstrate this: druse structures formed in the free space of crystallisation as well as inside hard body volumes are identical, but they cannot appear in viscous fluids. The granular structures (spherulitic and usual) cannot appear by crystallisation in free space, but they are identical for solid and viscous mediums of crystallisation. The truth of this thesis also follows from the physically strict definition: **the structure of an aggregate is a geometric picture formed by the dividing surfaces of the separate mineral individuals which form the mineral aggregate**.

It is known that the shape and position of the individuals defining the main characteristics of an aggregate structure in space depend only on the numerous kinetic effects during crystallisation. These include the changing regime of the supersaturation gradient of the solution, the symmetry of supply, the speed and anisotropy of growth, surface effects on the growing faces, epitaxy, the energy of grain boundaries, the presence of stress tension etc.

This established law is somewhat disappointing in the light of the task in hand. Geologists have intuitively regarded the structure of mineral aggregates as having a larger information content. Fortunately, this is compensated by the "second side of the coin" of mineral aggregates, the texture. The concept of "texture" was proposed by Groubenman (1904, 1912) as *the spatial features of the composition of aggregates*, but it is possible to apply this definition to the concept of "structure" as well. As a result there was confusion. Moreover, many later authors did not include any boundary criteria of the two concepts in their varied definitions. Even bodies with compound parts from different origins ("brecciated texture", "streaky texture" etc.)[9] were placed under the heading "texture". The author, however, found strict divisions between these concepts (Stepanov, 1970, 1971). Firstly, it was found that **they can only be applied to homogeneous aggregates, that is to the products of synchronous crystallisation**. Observation reveals that in these conditions different structures may appear, which are interchangeable. Just these effects forced the author to consider it necessary to observe crystallisation products in the whole space of synchronous crystallisation. At the beginning of this project no thought had been given to the necessity of introducing a concept of texture. The main task was the reconstruction of the development of structure as a sequential change of purely geometric patterns. However, application of the Curie Universal Principle of Symmetry to the results showed that different parts of the crystallisation space are described as a whole reflecting the *spatial features of the composition of an aggregate* (see Grouberman's definition of texture). From this there follows a more precise definition of texture: **texture reflects the geometric characteristics (or the absence of such) of variations of the structures of mineral aggregates in the whole space of synchronous crystallisation**.

From this definition one can see that at first the author did not overstep the limits of purely geometric characteristics of mineral aggregates. Consideration of the geometric patterns of structures and their variations from the point of view of the Curie Universal Principle of

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[9] At this point there is a contradiction between the concept of mineral aggregate texture and the concept of rock texture. These are not the same.

Symmetry required them to be described with concepts of symmetry. Therewith, completely unexpected by the author, the geometric and symmetrical characteristics of structures and textures appeared as information systems, not only containing information about geometry of growth, but also giving quantitative parameters of the specific physical forces affecting crystallisation. One may imagine how an observer, not acquainted with the general theory of systems, would respond to this. Yet a superficial knowledge of this field of information theory reveals that any natural system contains excessive (emergent) information on the properties of the whole system. From this point of view **texture represents the emergent property of a system of homogeneous structures in all the synchronous crystallisation space.**


## MAIN PRINCIPLES OF RECONSTRUCTION OF CRYSTALLISATION PROCESSES FOR MINERAL AGGREGATES IN CAVES

### 1. General situation.

Despite a great number of works published on this subject, there is an almost complete absence of systematic research; until now the only general paper on the subject is that by Prinz (1908)[10]. At the same time crystallisation in karst caves is extraordinarily revealing, especially in the light of concepts described in the preceding part of this work. Indeed, in the case of karst caves the space of synchronous crystallisation usually coincides with the space observed, and structures and textures of aggregates are rarely affected by later external influences.

The mineral aggregates of karst caves are very varied and are to be found as both single forms and as paragenetic ensembles,[11] of which the simplest examples are pairs of stalactite-drapery, tubular stalactite-helictite and others (see figure 1). The aggregates joined into paragenetic ensembles usually have distortions of structure, texture and morphology in comparison with stand-alone forms of the same aggregates. In many cases the distortions reach such an extent that it is impossible to speak about paragenetic ensembles, but about hybrid textures (for example, instead of a stalactite-corallite appears a corlactite).

In many cases paragenetic ensembles display individuality in their morphology, a property hitherto unknown in mineral bodies and always considered an attribute of living organisms[12].

### 2. Principles of reconstruction of crystallisation processes.

There are actually very few factors affecting the morphology of cave mineral aggregates. These are:

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[10]      This paper has been reprinted twice in English, as a part work in *Cave Science* Vol. 2 Nos 9, 10, 11 &13, 1949/50 and in *Cave Geology* Vol 1 No. 7, 1980. Both texts are identical, the translator was Sidney Melmore.
[11]      Nowadays the concept of "ensemble" is transferred to a higher level in the hierarchy of minor mineral bodies (Maltsev 1993, 1996a). The term ensemble in this context approximately corresponds in contemporary classification to the term multi-aggregate.
[12]      In Sletov (1985) this property is also noted in aggregates of special type (helictite) and is called "behaviour". Maltsev (1996a) proposed that aggregates and multi-aggregates with "behaviour" were called inter-active, because the "behaviour" occurs as a result of the feedback between the kinetics of mass-transfer in the crystallisation fluid and the growth dynamics of the aggregate (or multi-aggregate).

**Figure 1:** *The system of links between physical factors of crystallisation and the morphology of mineral aggregates in karst caves.*

Decrease of summary density and hydrostatic pressure in the phase system of the crystallisation medium.

| | Situations → | 1 | 2 | 3 | 4 | 5-6 | 7 | 8 | 9 | 10 | 11-12 | 13-14 | 15 | 16 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **I** | phase state of crystallisation medium | liquid supersaturated solution | | | | | | | | | | | | supersaturated vapour |
| **II** | phase system of crystallisation medium | subaqual solution-vapour | | | vapour-solution (subaerial) | | supersaturated vapour-solution | | | subaerial saturated vapour-solution | | | subaerial unsaturated vapour-solution | aerial (vapour) |
| **III** | state of mass-carrier solution | gravitational | | | | | capillary-gravitational (thin film) | | | capillary-gravitational (tubular) | capillary a) of individual capillars b) capillary cracks | capillary-gravitational a) of a bulky porous body b) capillary cracks | capillary a) of a bulky porous body b) capillary cracks | aerodynamic |
| **IV** | dynamics of mass-carrier solution | "stationary" | | | gravitational jets / 6. pressure jets | | moving / capillary uplift and wetting / gravitational flow of films | | | | moving capillary uplift and wetting | | | moving aerodynamic and convective jets |
| **V** | system of mass-exchange between phases during crystallisation | ↑CO₂ liquid solution A↓H₂O phase boundary (solid) | ↑CO₂ phase boundary (liquid) B↑H₂O A↑ liquid solution | ↑CO₂ B↑H₂O A↓ | ↑CO₂ ↓A | ↑CO₂ H₂O↓B ↑A | ↑CO₂ liquid solution ↓A phase boundary (solid) | ↑CO₂ H₂O↓B ↑A | ↑CO₂ ↑A | ↓CO₂ ↑CO₂ A↑H₂O | ↑CO₂ 11↑ 12↑ a porous solid body | ↑CO₂ ↑CO₂ phase boundary (solid) 13↑ 14↑ A↓ | ↑CO₂ ↑H₂O B↑H₂O | B ↕ H₂O phase boundary (solid) |
| **VI** | factors of supersaturation of the solution | B) phase change at 0°C | | | A) degassification / B) phase change at 0°C | | B) phase change at 0°C | | | | | | A) evaporation B) phase change at 0°C | B) sublimation |
| **VII** | the most active factors of morphogenesis | - | - | Symmetry of mass-transfer of the material to aggregates during their growth / variations of solution level | shape and dynamics of jets | | A. The speed of degassification and aerodynamics of CO₂ mass-transfer / variations of thickness of films | | | Symmetry of mass-transfer of the material to aggregates during their growth / strength of capillary pressure | | | aerodynamics of H₂O mass transfer / shape of aerodynamic jets |
| **VIII** | nucleation (appearance of crystal nuclei) on phase boundaries | liquid - solid | vapour - liquid | liquid - solid | | | | | | | | | vapour-solid |
| **IX** | the most active factors of morphogenesis | shape of cavity; contour, structure and composition of matrix | - | shape of cavity; contour, structure and composition of matrix | influence of crystallisation kinetics on the shape of individuals | | | | epitaxy | | shape of cavity; contour, structure and composition of matrix | | - | - |
| **X** | widest possible range of mineral aggregates (textures) | **A** crustification crusts, druses | **AB** films on the surface of solution | **AB** shelfstone | **A** 1) tufa dams 2) pisolites | **AB** 1) stalactite kora, curtains 2) stalagmite kora 6. geyserrules | **A** spherolitic covering kora | **AB** 1) coxlactites 2) corlagmites 3) crystlactites 4) crystlagmites | **A** 1) corallites 2) crystallicites | **A** helilactites | **A** 11 - helictites 12b - shields | **A** 13a) tuflactites 13b) moonmilk (CaCO₃ suspension) | **AB** antholites (efflorescences) | **B** crustification crusts, druses (sublimate) |
| | texture types | non-gravitational | gravitational | gravitational | non-gravitational | different | gravitational | gravitational | non-gravitational | gravitational | non-gravitational | gravitational | non-gravitational | non-gravitational |
| **XI** | presence of gradual changes between texture types | 1 3 / 1 41 | (B)-2 3 | 3 52 / 3 1 / 3 41 | 41 3 / 41 1 | 52 7 / 51 7 / 5 8 / 51 12 / 5 3 / 51 10 / 6 12 / 5 13 | 7 52 / 7 91 / 7 92 / 7 51 / 7 13 | 83 81 / 82 84 / 8 9 / 8 5 / 81 13 | 91 92 / 91 7 / 9 8 | 10 51 / 10 11a | 11 7 / 12b 51 / 12b 6 | 13 5 / 13 81 / 13 7 | - | - |
| | hybrid textures | | | (B)-3 2 | | | | | | | | | | |
| **XII** | observed paragenetic ensembles of aggregates (texture types) (without type B) | (1,3,52) - often (1,3,41,52) - less common (1,3,512,10,11) - rare (1-11) - very rare | (2) - rare (12,3) - more rare (1-11) - very rare | (1,3,52) - often (1,3,41,52) - less common (1,3,512,9) - rare (1-11) - very rare | (41,52) - often (41,2,52) - less often (1,3,41,52) - rare (1-14) - very rare | (512) - very often (1,3,512,10,11) - often (1,3,4,512,9) - often (13,4,512,9,10,11) - rare (1-14) - very rare | (7) - rare (52,7,91) - rare (1-14) - very rare | (512,83,9) - rare (512,7,8,9) - more rare (1-14) - very rare | (512,9) - often (1,3,512,9) - often (1,3,512,7,9) - rare (1-14) - very rare | (512,10,11) - often (1,3,512,10,11) - less common (1,3,512,92,10,11,12) - rare (1-14) - very rare | (512,10,11) - often (1,3,512,9,10,11) - less common (1,3,512,92,10,11,12) - rare (1-14) - very rare | (14) - rare (4,5,7,81) - rare (1-14) - very rare | (15) - rare (7,92,15) - rare (52,7,834,92,15) - very rare B. (15) - very rare | (16,5) - very often (16,5,23) - often (16,83,15) - very rare (51,83) - very rare |
| **XIII** | characteristic structure types / presence of elements of gravitational orientation | crusts of skeletal crystals, spherolitic druse crusts, spongy-porous crusts, skeletal crystals (tufa), crusts of dendritic spheroidolites | not studied (granular?) | crusts of skeletal crystals, spherolitic druse crusts, spongy-porous crusts, skeletal crystals (tufa) | Little studied | Not filled in (apparently, was considered obvious) | Not filled in (apparently, was considered obvious) | Not filled in (apparently, was considered obvious) | Not filled in (apparently, was considered obvious) | Not filled in (apparently, was considered obvious) | 11. mostly monocrystals with separate grains having elements of druse structure / 12. not studied, for the most part granular and recrystallised | 13. not studied Heterogeneous structures | granular-columnar plastically deformed bodies | spongy-porous crusts of skeletal crystals, crusts of skeletal crystals druse crusts |

A.        The influence of crystallisation kinetics on the morphology of individuals. An example is the case of subaerial growth of carbonates of the calcite group, that is to say the low-molecular compounds with solution supersaturation controlled by degassing.

In crystals, the morphology of individuals changes in accordance with an increase of supersaturation degree of the solution (or crystallisation speed, which is the same) from large ideal crystals (very low supersaturation degree) to smaller incomplete crystals (medium super-saturation degree) and on to acicular and skeletal crystals (high supersaturation degree), and further on to suspensions of shallow skeletal crystals[13] formed by bulk crystallisation (very high supersaturation degree).

In split crystals, in the same course of increasing supersaturation degree are found spherocrystals, crystalline split needles, spherulites and spheroidalites, dendrites of spherulites and spheroidalites, and suspensions.

B.        The influence of the method of transfer of the feeding solutions. There are three main types of feeding for the most common subaerial case: linear (axial) feeding which is produced by capillary supply as well as by gravitational supply, area feeding produced exclusively through capillary films[14], and bulk feeding produced exclusively by gravity streams. A more detailed layout is given in figure 2.

C.        The influence of structure and mass-transfer symmetry in the medium of crystallisation. In the case of coincidence of the ways of feeding solution supply, just this factor makes it possi-ble to select criteria to distinguish between crystallisation mediums during the reconstruction of processes. Each of the main crystallisation mediums has its own characteristic symmetry (Stepanov 1979, 1971). For subaqual mediums it is the symmetry of a sphere, for mediums with gravitational currents it is a cylinder, for mediums with capillary membranes it is a cone.

As shown in Stepanov (1971), crystallisation in karst caves is a cyclic process[15]. The mediums of crystallisation replace each other in a cyclic way, thereby initiating a total change of textures through all the synchronous crystallisation space. In the light of this, it makes sense to introduce the concept of "kora": an association of aggregates that appear in all the space of synchronous crystallisation, during episodes of the crystallisation cycle when the phase state of the crystallisation mediums remain constant for significant intervals of time[16]. So, in most karst caves can be found stalactite-stalagmite tufa koras, stalactite-stalagmite spherulitic koras, corallite-crystallictite koras, and antholite koras.

From the general poverty of mineral species in karst caves, and also by the "purity" and stability of physical and chemical conditions of crystallisation, it is in principle possible to

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[13]        "Moonmilk," by which the author means suspensions (see diagrams), in most cases has a more complicated structure (Hill and Forti 1986, 1997) and a biogenic origin.

[14]        Stepanov does not take into consideration here that it is characteristic for the sulphate case (though impossi-ble for the carbonate case) to have capillary, but not capillary film, ways of area and bulk feeding through porous substrates.

[15]        Cyclic deposition is well understood in sedimentology, and applies to crystallisation products as well as to sediments. In caves there is obviously an asymmetric cycle (A-B-C-D; A-B-C-D) as subaqueous products are replaced by the products of successively drier conditions, until the next general inundation of the cave passage. These cycles are not always complete, but the sequence remains the same.

[16]        Stepanov's original wording was too difficult to follow, so this definition has been paraphrased. In current usage, multi-aggregates and chemogenic sediments can be included in a kora.

theoretically deduce all the possible simple forms of mineral aggregates. (Conversely, for combined forms and ensembles such a possibility completely disappears).

Figures 1 and 2 present an attempt at a full theoretical as well as empirical systematisation of possible simple forms of cave mineral aggregates, and also of the most typical paragenetic ensembles.

*3. Examples of the most typical simple forms and ensembles for subaerial mediums of crystallisation. Definitions of the aggregates not classified earlier[17].*

## A. Gravitational forms.

A (i).      Stalactites:
Tubular stalactites (soda straws) are formed by strictly axial feeding, characterised by a constant channel diameter, and by a structure controlled by geometric selection during growth on the meniscus of a drip.

Conical stalactites are formed by combined axial and area feeding. These must not be considered simply as hollow stalactites overgrown with a spherulitic crust, as the synchronous overgrowth affects the morphology of the meniscus.

Draperies are formed by linear feeding.

Ensembles of stalactites and draperies are highly characteristic.

Tuflactites may be treated separately. These are stalactites formed in conditions of high supersaturation, consisting of calcareous tufa and thus devoid of a structure programmed by geometric selection. This division between concepts removes a contradiction between the widespread definition of hollow stalactites always being monocrystalline, and the existence of hollow tuflactites (characteristic, for example, of the Khaidarkan mines).

A (ii).     Stalagmites:
Stalagmite koras are formed by area feeding (flat gravity stream), as most often are tufa stalagmite koras. Spherulitic cover (onyx) koras are a special case, and are characteristic for the Khaidarkan caves (Kirgizia) and Kugitangtau (Turkmenia).

Stalagmites themselves are formed by axial feeding. The primary stage of growth of a stalagmite is represented by eggutation, that is to say the appearance of a local spherulitic crust known as "scrambled egg". With further growth geometric selection comes into effect leading, in the case of obelisk stalagmites, to monocrystalline varieties.

By analogy with the division between the concepts of stalactite and tuflactite, one can introduce the concept of tuflagmite, which in the same way is devoid of a structure programmed by geometric selection.

Note that, in contrast to stalactites, the morphology of stalagmites (but not tuflagmites) does not depend on the mineral composition. Strictly identical structures and textures appear in calcite, in gypsum, and even in ice.

Very characteristic and various are the ensembles of stalagmites and a stalagmite kora, as well as those of tuflagmites and a tufa kora.

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[17]      Unfortunately, the terminology which is introduced here was not published at the time it was written and therefore is not to be found in contemporary literature. However, for many terms there are as yet no alternatives, so the terminology presented here can still be adopted.

A (iii).    Stalactite-stalagmite koras:

A stalactite-stalagmite kora and a tufa stalactite-stalagmite kora (Stepanov 1971) both consist of aggregates of very similar structures and textures having one characteristic symmetry. At the same time the specific constituents of these koras may significantly vary in aggregates. For example, caves exist with a sharp predominance of stalagmite forms, and also in contrary. In the majority of cases, as shown by Maksimovitch (1965), the ratio between the amounts of stalactite and stalagmite forms is exclusively controlled by the amount of water in the cave.

## B. Forms due to crystallisation from evaporating capillary films.

B (i).    Corallites:

This term was introduced in Shcherban, Fiman and Koman (1961) for dendritic spheroidalites growing on substrate ledges above the level of standing or running water. In the same work was the first suggestion of their growth from evaporating capillary films. This concept is easy to prove by the Curie Principle (Stepanov 1971); the corallite texture has the characteristic symmetry of a cone with an axis directed normal to the substrate surface, corresponding to the characteristic symmetry of mass-transfer in an evaporating capillary film.

Corallites have significant variations of texture and may provisionally be classified into simple, branching and colonial. Characteristically there are gradual changes of gravitational textures into corallite ones with the film becoming thinner and thereby decreasing its ability to move under the influence of the force of gravity. Gradual changes of conical stalactites into corallites are typical.

B (ii).    Crystallictites:

These are a special case of corallites representing in structure a special kind of crystal dendrite, but not of spheroidalites as in the previous case. The texture of the aggregates is the same. Interconvertions between corallites and crystallictites are common. Crystallictites are more characteristic of minerals such as gypsum and aragonite, for which spherulites and spheroidalites are not characteristic.

Both corallites and crystallictites, in the pure case, have an area supply but sometimes produce forms intermediate between those of area supply and those of linear or axial supply. Aggregates of corallite and crystallictite textures can be considered together as a corallite kora, in the same way as for gravitational textures.

B (iii).    Intermediate forms:

As mentioned above, gradual changes into conical stalactites are characteristic for corallites. In reality, such changes also occur in the pairs "stalagmite-corallite", "conical stalactite-crystallictite" and "stalagmite-crystallictite".

If such pairs can be classified as overgrowths, on the first phases of the change, in a more developed case one should speak of hybrid textures having the properties of both compound parts at once. One can give examples such as the gypsum chandeliers of the Kugitangtau caves in Turkmenia, similar calcite ones of Kristalnaya cave in Crimea and dendritic "stalagmites" of the Muzeinaya cave in Altai. As long as there are no names for such hybrid textures, let us call them corlagmites, crystlagmites, corlactites, crystlactites.
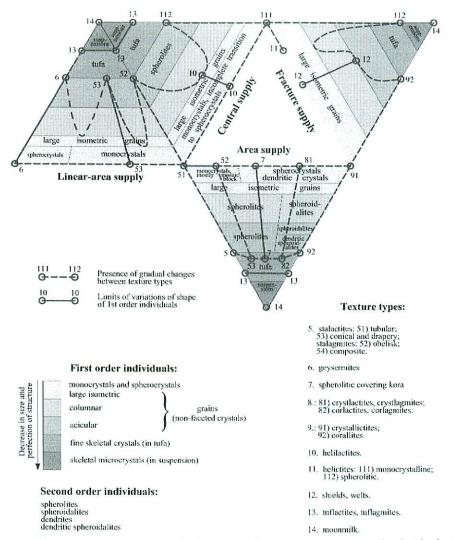
**Figure 2.** *The influence of kinetic effects of crystallisation on the shape of individuals in the subaerial conditions of karst caves (the case of carbonates of the calcite group).*[18]

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[18]      *In order to understand the crystallisation processes for aggregates, multi-level information is needed. Figure 2 attempts to do this by showing how the shape of mineral individuals relates to the mineral aggregate bodies found in caves. The written text on the figure is on the level of individuals, the numbers on the level of aggregates. Some simplification has been necessary at the apex of the triangles where the terms "tufa" and "suspensions" are used, rather than the very long names for the individuals specific for these aggregates.*

*In the list of texture types, 54 is described as "composite stalagmites" but the number 54 does not appear on the main figure. This suggests that it has been left over from an earlier version, as it does not really belong here. Also in this list of texture types, number 6 (geysermites)  may also include rings and blisters.*

*Some small differences can be observed between Figures 1 and 2, in that the sub-divisions of the main groups can have different code numbers eg. crystlactites are coded 83 in figure 1 and 81 in Figure 2. Clearly the diagrams were drawn at different times, but group 8 (crystlactites, corlactites etc) has not changed its content.*

## C. Helictites

C (i).    The classic case:

Helictites appear by crystallisation at the exit of a capillary channel, thereby being forms with an axial supply programmed by capillarity[19]. They are characterised by independence of the texture from gravitation, by changes of channel diameter and thickness of the aggregate. There are monocrystalline and polycrystalline varieties[20].

C (ii).    Intermediate forms and ensembles:

There are gradual changes from stalactites into helictites forming a hybrid texture in the central part of the change. Let us give to these aggregates the name helilactites. Even in the case of a full coincidence of the appearances of a helictite and of a hollow stalactite, they are easy to distinguish: a helictite has variations in the aggregate's diameter and channel thickness, which a hollow stalactite has not. Ensembles of stalactites (particularly of hollow ones) with helictites and helilactites with helictites are very widely distributed.

## D. Forms due to bulk capillary crystallisation.

These are characterised by the occurrence of moonmilk aggregate suspensions of skeletal microcrystals. Research by the author in Tvorojnaya cave (the Lagonaki plateau, Caucasus) shows the presence of intermediate forms changing into gravitational ones, as well as into forms with an orientation controlled by the wind.

## THE MEANING OF THE TERM "TYPOMORPHISM", FROM THE EXAMPLE OF MINERAL AGGREGATES IN KARST CAVES.

The original meaning of the term "typomorphism" (Fersman, 1952) suggested the presence of individual features in the morphology of minerals which depend on crystallisation conditions, i.e.. the presence of occasional single phenomena was suggested inside the general regular picture of growth of mineral individuals and aggregates.

Nowadays the term "typomorphism of minerals" is spread over all their properties, so that not only the occasional variations but the very general and strictly defined governing rules of a material's composition and properties are called typomorphic. Thus the original contrast between the typomorphic and non-typomorphic (general, regular) features of minerals has been destroyed. The concept "typomorphism" in the current meaning has lost its specific scope, and turned into a scholastic term. This has forced many researchers of mineral morphology and ontogeny to turn down the use of the term.

A detailed six-year research into the rules of growth and composition of mineral aggregates formed in open cavities (mostly in karst caves) allowed the author to find out the general mechanisms of formation of mineral aggregate textures in these conditions, and also to reveal a

[19]    Sletov (1985) also describes such helictites having a linear supply.

[20]    According to Sletov (1985) - not exactly so. There are varieties consisting of both a packet of 2-6 spherulites and a packet of 2-6 spherocrystals. Maltsev (1996a) observed the case when a packet of 3 spherocrystals was possible to distinguish from a slightly distorted monocrystal only by a detailed examination. Also, there are aggregates morphologically alike helictites but not having their essential structural features. Thus pseudohelictites (Maltsev and Self 1992; Hill and Forti, 1997) lack the capillary channel and actually are poly-mineral corallites of a special type. However, on the basis of microscopic examination, Korshunov (pers. comm.) has suggested that the solid pseudohelictite core may be permeable, in which case they may be a special type of helictite after all.

wide display of the occasional (individual) features of their composition. However, it needs to be said that although there are classes of textures where such individual features are widely displayed, in other classes they are not found. The textures of the first group exclusively appear by growth in flows, streams and capillary water films above the level of permanent or temporary reservoirs; the textures of the second group only by crystallisation in such reservoirs, or cavities entirely filled with solution. The most evident difference between these groups can be seen by the example of two classes of textures: the corallite and the druse.

Of 79 studied caves, crystallisation products were observed in 68 caves. Corallite textures were discovered in 53 caves, druse ones in 27. Both types of crystallisation were present jointly in 17 caves.

When observing druse textures, no deviations were detected from the common characteristics of their composition except those due to temperature and pressure during crystallisation and to the mineral composition of the aggregates (Grigoriev, 1961; Dymkov, 1957-1966; Stepanov, 1971). Crystallised druses and spherulite druses (spherulitic crusts) are built identically. The fine features of shape and composition of crystals and spherulites composing druses are determined by the most common rules of crystallisation kinetics and do not have an individual character.

Corallite textures (Stepanov and Lebedev, 1955; Stepanov, 1965), despite identity of their composition and structure as a whole, for each cave have their own individual set of variations of form of the individuals composing aggregates. In none of the 53 caves were such variations found to be identical; similar forms of individuals sometimes appear in different caves, but are sharply different in intensity of development and variation types. In this way, each cave is rigidly individual, relative to the individual forms.

Form variations of corallite individuals are entirely determined by the kinetics of dissociation of calcium bicarbonate in capillary films supplying the aggregate during its growth. In its turn, this dissociation is determined by the way of moving and the variations in concentration of the carbon dioxide in the cave's atmosphere in direct proximity to the growing aggregate. The aerodynamic streams carrying carbon dioxide of different concentrations are entirely determined by the shape of the cavity. As is known, the shape of cave cavities is absolutely individual. So the shape of the caves, finally, is the cause of individual fluctuations in the shape of corallite individuals.

It is impossible to apply the concept "typomorphism of minerals" even to one of these fundamentally different phenomena without solving the dilemma: what exactly is the meaning of the concept "typomorphism", common laws with strict regulations of phenomena and their relationships or individual (occasional) variations of one or another phenomena and properties. Both are determined by physical causes, which are known in the first case but still not brought to light by present-day science in the second.

By applying the term "typomorphism" to known regular phenomena and properties of minerals, we add nothing constructive to knowledge and this term inevitably will not have any specific character, as is currently the case.

By applying "typomorphism" to occasional variations of properties and phenomena, we draw attention to occasional yet unidentified phenomena and thereby create opportunities for scientific study of them.

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## APPENDIX – GLOSSARY

**Brine:** A concentrated solution of inorganic salts.

**Collomorphic:** Formed (or assumed to have formed) from colloidal gels.

**Druse:** A cavity from whose walls euhedral crystals project, or (more commonly) the crystals themselves.

**Gel:** A 'solid' colloidal solution, able to withstand some shear stress. The mechanical behaviour is defined by 'in-solution structures' rather than by viscosity.

**Melt:** A mineral or rock in liquid form, due to elevated temperature.

**Ontogeny:** The study of mineral individuals (crystals) and their aggregates (in contrast to phylogeny: the study of mineral species).

**Paragenetic:** Formed at the same time.

**Real solution:** A solution whose components can be dissociated.

**Sol:**  -  A 'liquid' colloidal solution, whose particles do not form 'in-solution structures'. Hydrosols have water as the dispersion medium.

**Spherulite:** A mineral individual, grown from a single nucleus, but divided into sub-individuals to display a spherical mass of radial fibres.

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