Original Article
ISSN (Online): 2454-1907

International Journal of Engineering Technologies and Management Research
May 2025 12(5), 11–19

THE CRISTAL WITH PARTICIPATE OF THE XENOMORPHIC SURFACE AND THE VARIANCE OF FACE SYSTEM

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Received 09 March 2025
Accepted 08 April 2025
Published 14 May 2025

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DOI
10.29121/ijetmr.v12.i5.2025.1551

Funding: This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

ABSTRACT

Geometric crystallography studies the external form of a crystal, formed by a combination of geometric planes - reticular faces. This form of natural crystals can vary from simple forms, with the smallest number of faces, to polyhedral, with a certain number of faces. Such crystals are called idiomorphic, since they are geometric crystalline polyhedra. Faces that differ in reticular densities are called reticular, and faces multiplied by symmetry elements are called identical. The set of these faces is called a faceted form. By symmetry operations, the faceted form is transformed into a form consisting of only reticular faces. Such a form is called a faceted system. However, natural crystals are often subject to mechanical and (or) chemical dislocations and a xenomorphic surface appears in the faceted form. The faces can be partially or completely destroyed, and the crystalline polyhedron becomes unclosed, a greater or lesser part of the surface of its previously euhedral crystal becomes xenomorphic, and the face system consisting of the preserved reticular faces and the xenomorphic surface is combined; in it, the xenomorphic surface closes the crystalline polyhedron. Such crystals are hypidiomorphic. Xenomorphization, as a process of destruction of reticular faces, changes the thermodynamic state of the crystal. However, in geometric crystallography this surface is considered as a crystal defect, and the hypidiomorphic crystal is still studied as euhedral, its equilibrium is estimated, which is unjustified, since the xenomorphic surface is characterized by its own surface tension and, in essence, plays the role of an independent faceted surface. The role of the xenomorphic surface in the variability of such crystals was studied for the first time. Variance functions are obtained for different states of a face system involving a xenomorphic surface.

Keywords: Xenomorphic Surface, Geometric Crystallography Studies, Reticular Faces

1. INTRODUCTION

A crystal is a homogeneous crystalline body, limited by a regular set of planes - faces, which can differ in crystal structure. Structurally homogeneous faces are called reticular Bravais faces Brave (1974). The number of reticular faces in a crystal can vary widely. Faces with the same crystal network were called identical by R. J. Haüy Haüy (1962). These are samples of the same generating reticular face, multiplied by elements of symmetry.

How to cite this article (APA): A, A. L. (2025). The Cristal with Participate of the Xenomorphic Surface and the Variance of Face System. *International Journal of Engineering Technologies and Management Research, 12*(5), 11–19. doi: 10.29121/ijetmr.v12.i5.2025.1551

The set of all faces of a crystal is called a facet form Shafranovsky (1968). Within the framework of geometric crystallography, attempts have been made repeatedly to systematize faces based on various features: the degree of smoothness of the facet surface Stranski (1928), Honigman (1961), on the idea of "chains of strong bonds" Hartman and Perdok (1955). At the same time, natural crystals during growth are often subject to mechanical and chemical dislocations and the faceted shape can be complicated to a greater or lesser extent by the appearance of a xenomorphic surface and the previously euhedral crystalline polyhedron becomes incompletely closed by faces. Its shape turns into a combination of faces and a xenomorphic surface. Such crystals are called hypidiomorphic Admakin and Trubachev (2012). In this case, the thermodynamic state of the crystal is disrupted, its extensive and intensive parameters change. If the euhedral crystal was previously in equilibrium, then with the appearance of a xenomorphic surface it becomes nonequilibrium. The hypidiomorphic crystal turns into a carrier of additional properties relative to the euhedral crystal. From the point of view of geometric principles, such material properties are not revealed, and their study is relevant in geometric crystallography.

2. RETICULAR FACETS AND XENOMORPHIC SURFACE OF A CRYSTAL

A facet system is material and, from an energetic point of view, identical to a phase system; both are characterized by crystalline components. However, with respect to internal intensive and extensive parameters, they differ fundamentally. A phase system is a set of homogeneous bodies of different chemical composition and crystalline structure. As a physicochemical phase system, it is characterized by certain thermodynamically established relationships between phases, different from mechanical mixtures. A crystal is a set of reticular faces that form a closed or open crystalline polyhedron, including a xenomorphic surface. The facet system is thus only the external part of a crystal, which is formed by an elementary layer of molecular thickness Gibbs (1982). The theory of equilibrium of the face system was based on the capillary theory, in which the sets of reticular crystal faces are taken as homogeneous components in terms of their behavior style during crystal growth. The failure to satisfy the equilibrium condition for the crystal derived by J.V. Gibbs Gibbs (1982) was noticed only 145 years later, but the reasons for this phenomenon were not established Sheftal (1977). With the recent identification of two groups with opposite development styles, the phenomenon of polarity of the reticular crystal faces was discovered, which is of fundamental importance in the theory of equilibrium of the face system, and the Gibbs method lost its significance in crystallography. The systematics and thermodynamic characteristics of the reticular crystal faces became relevant in the study of the state of crystals. The face system of a crystal is a thermodynamic concept Gibbs (1982). Its reticular faces are characterized by their own reticular densities, and, accordingly, by their own values of the surface partial free energy. They participate in chemical reactions during crystal growth, accompanied by their own thermal effects, and in this respect, they behave similarly to phases of a phase system. The surface free energy of a reticular face is the potential of the face system and is created by unsaturated coordination bonds. Therefore, in a face system there are as many partial potentials as there are reticular faces. Each reticular face is a carrier of an intensive parameter, and its area is an extensive parameter and face entropy. We emphasize that identical faces are not thermodynamic parameters; they are reduced to the corresponding generating reticular faces by symmetry. However, if a binary transformation of a cube into an octahedron exists, then such a crystal is polar, and in the cube → octahedron transition, the cube performs the basic function with increasing temperature, and the octahedron performs an additional function, and in the octahedron → cube transition, with decreasing temperature, the functions of simple forms change to the opposite.

Basic reticular faces, or b-faces. Their Miller indices are identical to the indices of the corresponding faces of the unit cell {100}. The number of basic reticular faces is determined by the syngony and varies within 1–3. Simple forms of cubic crystals are characterized by one reticular face, in crystals of the middle category there are two, in crystals of the lower category there are three.

Basic faces are fundamental elements of the basic face system and crystal, systematic in any crystal, i.e. they are obligatory crystallographic elements. They are characterized by the highest reticular densities, respectively, the lowest values of partial free energy, can grow by the translational/transfer mechanism, or be in equilibrium, have the lowest growth rates.

The basic face system is conservative, stable within the boundaries of crystal stability, invariant, therefore basic crystals are found in various types of igneous rocks. In relation to this face system, and only it, the Gibbs equilibrium condition and Euler's theorem are fulfilled.

Additional reticular faces, or s-faces. Miller indices {hk0} and {hkl}, face planes pass through systems of parallel nodal rows that form edges, as elements of the Haüy retreat, or through sets of vertices of unit cells. Therefore, such reticular faces are called edge and vertex faces in geometric crystallography Shafranovsky (1961), respectively. In the systematics of P. Hartman and W.J. Perdock Hartman and Perdok (1955) they correspond to K- and S-faces.

Additional faces of any crystal are homogeneous. They are associated with the basic faces. Nodal rows of the s-face form their own crystal networks, which do not lie in the planes of the faces of the unit cells, i.e. are oriented obliquely relative to at least one coordinate axis. These faces are characterized by the lowest reticular densities relative to the basic reticular faces, respectively, the highest values of the surface partial free energy and the moduli of the rates of chemical reactions of transformation. Therefore, additional reticular faces spontaneously and irreversibly contract and disappear during crystal growth in the sequence of decreasing surface partial free energy, in accordance with Berthelot's rule; they are, therefore, variable elements of the face system, in contrast to the systematic basic reticular faces. All the variety of external forms of crystals is due to the presence of additional reticular faces, among which the base faces act as windows; they cause a decrease in the symmetry of the crystal. It can be said that, thanks to them, crystals are rich in the variety of their external forms.

Edge-faces. Their crystal networks are formed by a system of parallel nodal rows lying in one plane. For example, pyramidal edges associated with the law of retreat of Haüy create pyramidal faces, and, accordingly, prismatic edges - prismatic faces. Rib faces are characterized by reticular densities, which in magnitude follow the basic reticular faces. The rates of contraction of these faces during crystal growth are somewhat greater than the growth rates of the basic faces, therefore, the rib faces disappear from among the additional reticular faces last, leaving behind a stable basic crystal.

Vertex-faces are formed by systems of apical angles lying in the corresponding planes; therefore, these faces have a pyramidal orientation, which is well illustrated in the monograph of B. Honigman Honigman (1961). They are characterized by the lowest reticular densities and accordingly have the highest partial surface free energy values among additional reticular faces, and accordingly, the highest rates of contraction and disappearance. During crystal growth, the apical faces disappear first Admakin and Admakin (2023).

Note that the basal faces in crystals of the middle and lower categories, although they differ in the values of the partial surface free energy, however, as systematic ones in the crystal, they grow simultaneously. Therefore, Berthelot's rule does not apply to them.

Xenomorphic surface in a crystal. Crystals can be subjected to mechanical and chemical dislocations, as a result, of which a xenomorphic surface appears in the face system. It is characterized by an arbitrary exposure of sets of faces of elementary cells. The xenomorphic surface is structureless, thermodynamically homogeneous, and, regardless of the position on the crystal surface, is characterized by its own surface tension. Such a surface can arise at different stages of crystal growth, but for thermodynamic analysis only such a xenomorphic surface is significant, which is formed during the crystal growth process, i.e. syngenetic.

A xenomorphic surface disrupts the continuity of the crystal lattices, makes an open euhedral polyhedron, which is closed by this surface. It can partially or completely replace one or several faces and finally transform the entire crystal faceting into a xenomorphic surface. As a result, the set of s-faces disappears, the crystal turns into a xenomorphic body, or phase. G.B. Bokiy Bokiy (1971) called such bodies a crystalline substance. Sometimes the surface of these bodies retains the shape of the previous euhedral crystal, which, however, is a relict and has no relation to the face system.

The xenomorphic surface is characterized by the highest value of surface free energy, or surface tension, which determines the super-nonequilibrium of the xenomorphic crystalline body and its tendency to spontaneous, irreversible regeneration of faces Ansheles (1952). A crystal with partial xenomorphization of faces is called hypidiomorphic Admakin and Trubachev (2012). Such a crystal is nonequilibrium, its xenomorphic surface is prone to spontaneous, irreversible regeneration into reticular facets. For a face system involving a xenomorphic surface, the ratio of the contraction rates during crystal growth is written vx> vsp> vsa> vb, or vsp/vb> vsa/vb> 1, where vx, vsp, vsa, vb are the contraction rates of the xenomorphic surface, edge and apex faces, respectively, growth of the basal face system.

Bs-faced system with participation of a xenomorphic surface. Structurally combines basic, additional reticular faces, which is the general form of the face system, and an xenomorphic surface. If in the b-faced system the number of reticular faces is limited to 1-3 faces, then in the bs-faced system this number varies depending on the syngony within wide limits, forming successive geometric polyhedra during crystal growth. The limiting polyhedron is the one with the maximum number of faces. If such a polyhedron is represented as a primitive Wigner-Seitz cell, it can be divided into a series of polyhedra, including simple ones, characterized by their own shape. In simple forms of crystals of the cubic syngony there are no additional faces; the face system consists of basic faces only; in combined forms the number of s-faces is 1, for example, in a cube the blunting of the vertices gives a cuboctahedron. In crystals of the lowest category the number of reticular faces is the greatest, in crystals of the middle category it is intermediate. The combined form evolves in such a way that the basic faces can only grow or be in equilibrium, and the additional ones, on the contrary, can shrink and disappear; the face system becomes basic. The most stable face system is the one with the lowest surface free energy Admakin and Admakin (2023); any process that occurs with the reduction of the surface of the combined face system is spontaneous and irreversible. For the bs-face system, the Gibbs equilibrium condition is not satisfied.

Thus, any bs-face system is a nonequilibrium system; it spontaneously and irreversibly transforms into a b-face system during crystal growth. However, this process is such that it is always carried out with an increase in the total surface of the face system, in other words, the surface of the additional faces is a potential resource of area for the growth of the basic faces. vx> vs>vb, or vx/vb> vs/vb> 1, which serves as a criterion for the nonequilibrium of the bs-faced system, in which additional reticular faces and a xenomorphic surface are present.

3. VARIANCE OF A FACET SYSTEM INVOLVING A XENOMORPHIC SURFACE

A facet system exhibits an analogy with a phase system in the thermodynamic and chemical sense, but it is a specific system with its own intensive and extensive parameters. Consequently, the principles of chemical thermodynamics are applicable to a facet system. Variability characterizes the variability of the number of variable internal and external intensive parameters and is determined by the relationship Admakin and Trubachev (2012)

$$\nu = \psi + 1 - \chi, \quad (1)$$

Where ν is the variability of a facet system; ψ is the maximum number of reticular faces of a facet system, the crystal constant; 1 is the external intensive parameter, the temperature; χ is the number of actual reticular faces of a facet system.

The reticular faces of the real face system give the number of internal intensive parameters of the crystal - basic and additional $\chi = \chi b + \chi s$. Under the condition T = const,

$$\nu = \psi - \chi, \quad (2)$$

Function (1) and its particular form (2) determine the variability of the heterogeneous face system as a whole, without dividing the reticular faces into types, according to the style of behavior during crystal growth. However, in a crystal, basic and combined face systems are structurally possible, with different structural composition and style of development during crystal growth. However, this specificity is not reflected in the thermodynamic parameters of functions (1) and (2), which makes these functions uninformative in thermodynamic analysis. It is advisable to decompose these functions into types of reticular facets

$$\nu = (\psi b + \psi s) + 1 - (\chi b + \chi s), \quad (3)$$

Where ψb and ψs are the maximum number of basic and additional reticular facets, respectively; χb and χs are the number of basic and additional reticular facets in the real facet system, and $\chi = \chi b + \chi s$.

When T = const,

$$\nu = (\psi b + \psi s) - (\chi b + \chi s), \quad (4)$$

Relations (3) and (4) carry rich information. The maximum number of basic faces in the face system can vary within 1 – 3. The maximum number of additional reticular faces is a crystal constant, depending on the structure of the crystal lattice, and is determined empirically. Therefore, the sum ($\psi b + \psi s$) is also a crystal constant. Let us introduce a xenomorphic surface into the face system with index x. This surface, as noted above, can occupy different positions in the face system. There are three extreme cases of the position of a xenomorphic surface in the face system:

1) Let a xenomorphic surface partially affect one or several real reticular faces, but their number is preserved. Consequently, the parameter χ does not change, since only the areas of the reticular faces have decreased due to xenomorphization. A xenomorphic surface, regardless of the reticular faces, is homogeneous and is characterized by its own value of partial free energy - surface tension. Consequently, a xenomorphic surface, regardless of its position on the crystal surface, increases by one, on the one hand, the maximum number of surfaces (faces + xenomorphic surface), on the other hand, also by one, the number of real surfaces; it closes the surface of the polyhedron, however, leaving it open. In this case, the variance of the face system is written

$$\nu = (\psi + 1) + 1 - (\chi + 1) = \psi + 1 - \chi, \tag{3}$$

at T = const,

$$\nu = \psi - \chi. \tag{4}$$

Thus, the xenomorphic surface, partially affecting the reticular faces, does not change the variance of the face system. A hypidiomorphic crystal arises, characterized by the same variance as an euhedral crystal. Let us emphasize that the following rule follows from the structure of function (1): if any parameter is included in ψ and χ, it does not affect the variance of the crystal. This follows from the fact that the parameter ψ is an intensive parameter, a constant of the crystalline substance. The value χ is the number of actual reticular faces, i.e. extensive internal parameters of the face system. The appearance of a xenomorphic surface increases the internal intensive parameters by one, since this surface is characterized by its own partial free energy. The value of the xenomorphic surface can vary, provided that the number of reticular faces is preserved. The variability of the crystal remains constant, and its function is similar to the variability of the idiomorphic face system.

2) The xenomorphic surface completely affects one or more reticular faces. If the complete xenomorphization of some faces is also accompanied by partial damage to other faces, then the maximum number of intensive parameters remains equal to ψ + 1. The number of internal intensive parameters of real reticular faces is determined as χ + 1 – x, where x = 1, 2, … χ < ψ. Then,

$$\nu = (\psi + 1) + 1 - (\chi + 1 - x) = (\psi + 1) - (\chi - x). \tag{5}$$

Expression (5) is the variance function of the face system under the condition of complete xenomorphization of part (x) of the reticular faces. For example, if only one reticular facet is completely affected, then, at T = const,

$$\nu = \psi + 2 - \chi.$$

The similarity of function (6) with the Gibbs phase rule is formal and is only one of the special cases of the variability of the facet system.

3) The variant of complete xenomorphization of reticular faces is interesting. In this case, x = χ - 1 and

$$\nu = \psi. \tag{6}$$

Let us explain condition (6). The parameter ψ is the potentially possible number of reticular faces in the face system and their number cannot be greater than ψ + 1 with a variable parameter T, provided that T = const, not greater than ψ. It should be remembered that ψ is a potential number, a constant of the crystalline substance, or the number of internal intensive parameters. The parameter χ is the actual number of reticular faces in a particular crystal. Consequently, in this case, out of the potentially maximum number ψ, in the face system of real reticular faces there are only χ, the remaining ψ – χ are only potential. Consequently, ψ is a potential number of possible faces, but not an actual one. This potential number is a constant of the crystalline substance. Condition (6). ψ – χi shows how many free potential possibilities for the emergence of actual reticular faces remain in the face system. This number is the variance of the face system. At T=const, the number of actual reticular faces can vary from zero, at χi = ψ, to ψ, at χi = 0, respectively, the variance will vary within ν = 0 ÷ ψ, depending on χi. A change in variance is possible only as a result of regeneration of reticular faces, or with the disappearance of additional reticular faces. Now it is obvious that it is not justified to consider a crystal as a phase, or, conversely, as a xenomorphic homogeneous crystalline body. A crystal is a special state of a crystalline substance, with its own value of the surface free energy of its reticular faces. However, K. Hurlbut and K. Klein Hurlbut and Klein (1982), following the tradition of materials scientists, define a crystal broadly, "as a homogeneous solid body with a three-dimensional long-range internal order" (p. 18). Any crystalline homogeneous xenomorphic body of any arbitrary shape falls into the category of crystals. If we use the terminology of igneous rock structures Rosenbusch (1934), then such a crystalline homogeneous xenomorphic body can be called an allotriomorphic crystal. However, in chemical thermodynamics, such a body is considered a phase Sheftal (1977), and its variance is determined according to the Gibbs phase rule. For example, in a single-component composition, at T = const,

$$f = k - r = 1 - 1 = 0,$$

where k = 1 is the number of components; r = 1 is the number of xenomorphic surfaces. However, a single-component phase is an equilibrium potential with respect to the regeneration of the faceted system, since the faceted system is characterized by the lowest surface free energy. Therefore, the xenomorphic surface is regenerated into a faceted system, and this process is spontaneous, irreversible, i.e. natural. Regeneration ends with the formation of a bs-face system, which is more stable than the xenomorphic surface. The bs-crystal is non-equilibrium and occupies an intermediate position in the transformation series:

phase → bs-crystal → b-crystal.

The reticular faces of the b-crystal are invariant in the face system of any crystal. Excluding from the variability (3) the parameters related to the disappeared additional faces, we find

$$\nu b = \psi b - \chi b, \tag{7}$$

However, in the basic system of any crystal the condition is always satisfied

$$\psi b = \chi b, \tag{8}$$

relation (7) gives

$$\nu b = 0. \tag{9}$$

Equality (9) is a very important indicator of the state of the face system. It shows that in any crystal the basic reticular faces do not affect the variability of the face system. Such a face system is in equilibrium within the stability limits of the crystal and can be called indifferent.

The basic face system is characterized by the lowest free energy. There are no other faces with lower free energy, which makes the b-face system special, fundamental, which can only grow or be in reversible equilibrium. This state of the crystal is called quasi-static, and chemical equilibrium is quasi-static equilibrium Leontovich (1983). Its peculiarity lies in the fact that, although the b-face system is in a state of minimum surface free energy, the b-crystal can reversibly grow, remaining in equilibrium. Therefore, only in relation to the basic face system is L. Euler's theorem fulfilled, in other words, the b-crystal always grows by the translational/transfer mechanism. Examples are giant bipyramidal-prismatic quartz crystals Nauka (1965), the formation of which is a consequence of geometric selection under conditions of reversible crystal growth.

4. CONCLUSION

Natural crystals are often subjected to mechanical and chemical influences that destroy the crystal or transform the planes of the faces into a xenomorphic surface. Thus, combinations of reticular faces and their fragments with a xenomorphic surface arise. The previously closed crystalline polyhedron turns into an open one, in which the flat face becomes a xenomorphic surface. As a result, the thermodynamic state of the crystal changes, and if it was previously in a certain state, or in quasi-static equilibrium, then, due to disturbances, this state changes, the equilibrium crystal comes to a nonequilibrium state. In geometric crystallography, this phenomenon is not given due attention, the crystal is considered as equilibrium.

The study of the state and equilibrium of a crystal with a xenomorphic surface is no less important than that of an euhedral crystal, without which the theory of equilibrium of a faceted system cannot be considered complete. The solution to the problem of equilibrium of such crystals has become real on the basis of the properties of thermodynamic variability. In this aspect, the crystallographic and thermodynamic characteristics of the reticular faces of the facet system and the xenomorphic surface are expanded. A crystal whose surface is formed by reticular faces and a xenomorphic surface is called hypidiomorphic. The variance functions of the facet system with varying degrees of xenomorphization of the reticular faces are derived. Three states of the facet system with the participation of the xenomorphic surface are distinguished, which are characterized by their own variance functions:

1) Facet system with partial xenomorphization of one or more reticular faces; variance does not change.
2) Facet system in which one or more reticular faces are completely xenomorphized; variance of the facet system increases by the number of xenomorphized faces.
3) Facet system is completely xenomorphized; faceting has disappeared, the crystal is transformed into a xenomorphic crystalline body, or phase.

A xenomorphic surface introduces one degree of freedom into the variability of the face system. A hypidiomorphic crystal is nonequilibrium; during growth, it spontaneously and irreversibly regenerates into a euhedral bs-crystal consisting of basal and additional faces. Such a crystal is heterogeneous, nonequilibrium; its s-faces spontaneously contract and disappear, the face system turns into a basal face system, which is invariant, can grow reversibly or be in a state of equilibrium, and is stable within the stability limits of the crystal. This state of the face system is quasi-static.

A xenomorphic surface in a face system is a common phenomenon in natural crystals. It is a visual indicator of a nonequilibrium state. However, in geometric crystallography it is noted only as a defect and is not taken into account when assessing the equilibrium of a crystal. This makes such assessments invalid; a xenomorphic surface is a carrier of a thermodynamic degree of freedom.

CONFLICT OF INTERESTS

None.

ACKNOWLEDGMENTS

None.

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