

## THE GROWTH HABITS OF OLIVINE — A STRUCTURAL INTERPRETATION

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### ABSTRACT

Analysis of the crystal structure of olivine, in terms of isolated  $\text{SiO}_4$  tetrahedra and  $M$ -site cations, provides a qualitative explanation for the relative importance of the forms {010}, {021}, {110}, {210}, {101} and {001}, for the unit cell with  $b > c > a$ . Factors contributing to the dominance of {010} in skeletal olivine are: (i) the uneven distribution of the compositional units in planes parallel to this form results in faces of low surface energy, (ii) the stereochemistry of the coordination polyhedra of the  $M(2)$  protosites is unfavourable for the nucleation of growth layers and (iii) the faces of {010} are parallel to directions of easy growth.

### INTRODUCTION

Recent petrological investigations into lunar basalts, submarine pillow lavas and spinifex-textured peridotites have focused attention on the variety of growth habits exhibited by olivine and pyroxene in igneous rocks. Although cooling rate and chemical environment seem to be the principal factors controlling these variations, growth habits must reflect the constraints imposed by crystal structure and this correlation is investigated in the present paper on olivine and in a subsequent one on clinopyroxene. Particular attention is given to habits associated with relatively high cooling rates, especially to skeletal habits in olivine and clinopyroxene and to sector zoning in clinopyroxene.

### GENERAL CONSIDERATIONS

For simple structures the law of Bravais-Friedel (Friedel 1907) and its later developments, the law of Donnay-Harker (Donnay & Harker 1937), have been relatively successful in predicting equilibrium growth forms. The basic observation is that the faces of the dominant crystal forms have the largest interplanar spacings ( $d_{hkl}$ ) or, in other words, the highest reticular density. These faces incorporate a high percentage of the strong bonds of a given structure and consequently are characterized by minimal surface energies (Hartman & Perdok 1955a,b). These authors have extended this principle to more complex and less symmetrical structures with

the concept of the periodic bond chain. Possible crystal faces are more likely to develop parallel to slices of the crystal structure,  $d_{hkl}$  in thickness, which contain two or more periodic bond chains. In principle, the attachment energy of each of these slices (growth layers) may be calculated and used as a measure of growth rate. This has been attempted for K-feldspar (Woensdregt 1974). However, there are practical limitations to the calculation of attachment energies and the frequent variation in the development of the equilibrium forms for crystals from the same environment does not appear to warrant a wholly quantitative solution to this problem. Furthermore, for crystals grown under a high cooling rate from undercooled and supercooled solutions, kinetic factors tend to become dominant and the resulting habit may have little resemblance to that grown under equilibrium conditions.

Crystallization under near-equilibrium conditions will tend to produce equant crystals as this habit represents a minimization of specific surface area. With progressively higher cooling rates, a given phase initially will elongate or flatten parallel to directions of easy growth and then develop dendritic and spherulitic habits. Lofgren (1974) has demonstrated this behaviour for plagioclase and pyroxene crystallized from mafic liquids. When the growth rate ( $G$ ) of a crystal exceeds the diffusion rate ( $D$ ) in the liquid, a thin layer of impurity or low-temperature component builds up in the liquid at the solid-liquid interface. Growth perturbations on the crystal surface tend to pierce this impurity layer. At some point, for  $G \gg D$ , the increase in supercooling away from the crystal coupled with the retardation of crystallization in the areas between the growth perturbations, due to the accumulation of the low-temperature component, may cause spontaneous filament growth (Keith & Padden 1963; Lofgren 1974). The separation of the filaments is proportional to  $D$ . Growth perturbations on the sides of the filaments, or branching at their tips, lead to filament growth in a second direction. At lower cooling rates, crystallographic continuity is maintained and skeletal and dendritic structures are produced. At higher cooling rates, crystallo-

graphic continuity is not maintained and the filaments grow in the sub-parallel, radiating clusters characteristic of spherulites.

At temperatures near the melting point, the structure of a liquid is thought to have many similarities to that of the corresponding solid phase. In particular, the nearest-neighbour coordination polyhedra are similar to, although more irregular than, those in the solid. Also, in a multicomponent silicate liquid, Si, being a complex-forming metal, exists in Si-O tetrahedral complexes which are bridged as required by the stoichiometry of the liquid and the availability of complex modifying metals. The metals which occupy the *M*-sites of silicate minerals exist as free cations in irregular coordination polyhedra, the site occupancies being determined by the availability of suitable sites as well as size and other crystal-chemical considerations.

#### GROWTH HABITS

Olivine in igneous rocks shows a large variety of habits, from equant, skeletal, tabular and acicular to dendritic. Petrographic studies suggest that this variation is due to cooling rate and this is essentially confirmed by the experiments of Lofgren *et al.* (1975).

Equant crystals tend to be slightly prismatic or tabular, with least growth velocity normal to {010}, for the unit cell with  $b > c > a$ . The forms commonly developed in approximate decreasing order of importance are {010}, {{021}}, {110}, {210}, {{101}}, {001} and {111} in fayalite (Deer, Howie & Zussman 1962). Drever & Johnston (1957) have demonstrated that, in picritic minor intrusions, skeletal Mg-rich olivine crystals show a definite tendency to be tabular parallel to {010}, with maximum elongation along either [100] or [001], the skeletal habit being developed by growth parallel to the forms {010}, {021} and {110}. Sections (100)

of skeletal subequant crystals (Drever & Johnston, Fig. 8), which have a lantern-like appearance (Fig. 1), are particularly characteristic and have been observed also from such diverse rocks as submarine basalts (Bryan 1972, Fig. 1e), Apollo 12 basalts (Brett *et al.* 1971) and spinifex-textured rocks. The megacrysts (or phenocrysts) in spinifex-textured rocks are composite skeletal crystals, tabular parallel to {010} (Fleet & MacRae 1975). Their appearance in thin section varies markedly with orientation. In sections cut normal to [010], the composite character is seen to be due largely to pronounced parallel growth in which the individual platy crystals are arranged en echelon approximately parallel to one of the [103] directions; it is possible that the swallow-tailed habit (Bryan 1972, Fig. 1d) results from an analogous but more exaggerated parallel growth development. Unlike the skeletal crystals examined by Drever & Johnston, these megacrysts do not show {110}. In sections normal to the *c*-axis the individual crystals are tapered and have a tendency to be connected through steps. Out-growths extending from intersections of faces of {021} appear acicular in sections cut normal to [100] although they may, in fact, be platy parallel to {010}. The acicular chains reported by other authors (for example, Bryan 1972, and Lofgren *et al.* 1975) may be made of H-shaped units of {010} and {021} forms extending parallel to [001] (Fig. 1). However, in light of Drever & Johnston's work (1957, Fig. 19), these skeletal crystals could well be tabular, and merely give the appearance of an acicular habit in thin section.

#### DISCUSSION

An olivine crystal must grow from a liquid by the addition of isolated Si-O tetrahedral complexes and *M*-site cations ( $Mg, Fe^{2+}$ ). The representation of the olivine structure used in this study (Fig. 2) emphasizes these composi-

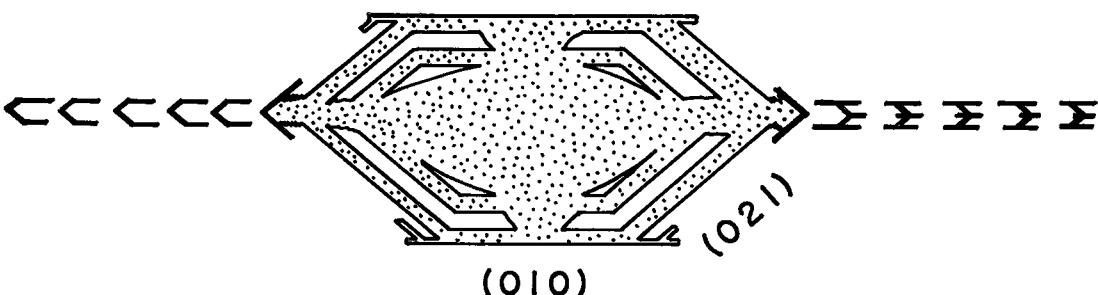


FIG. 1. Composite idealized sketch of a skeletal olivine crystal with the lantern and chain habit, in (100) section; right-hand-side chain has H-shaped units, left-hand-side chain has U-shaped units.

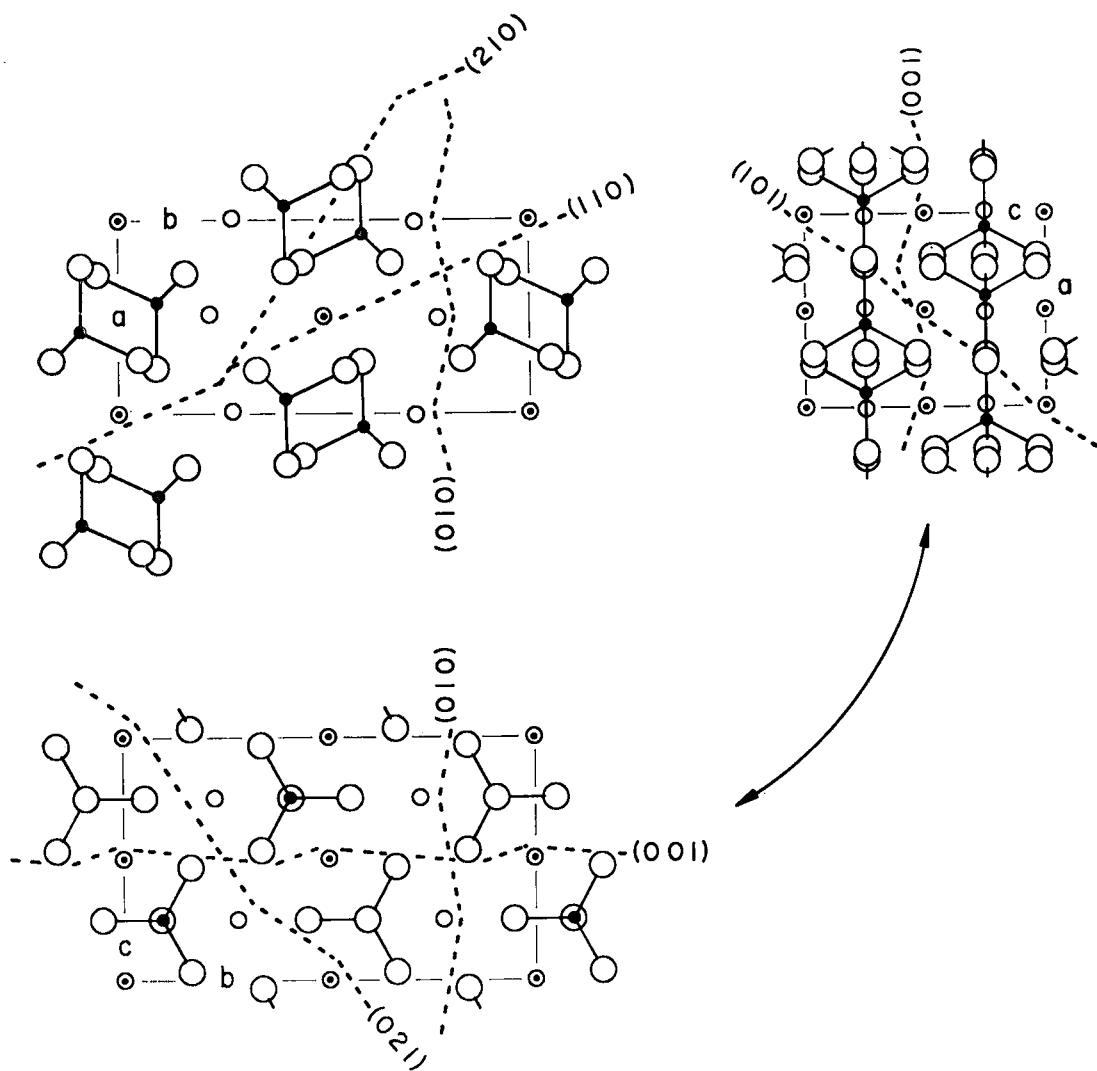


FIG. 2. The crystal structure of forsterite projected parallel to  $c$ -,  $b$ - and  $a$ -axes showing the probable compositions of faces of  $\{010\}$ ,  $\{021\}$ ,  $\{110\}$ ,  $\{210\}$ ,  $\{101\}$  and  $\{001\}$ :  $M(1)$  atoms, medium circles with dot;  $M(2)$  atoms, medium open circles; Si, small closed circles; O, large open circles; Si in upward-facing tetrahedra of  $a$ -axis projection have been omitted; data from Birle *et al.* (1968).

tional units. Because Si-O bonds are very much stronger than  $M$ -O bonds, it is unlikely that isolated or incompletely coordinated  $Si^{4+}$  atoms would be added. It has been shown that the dominant growth feature of Mg-rich olivine is the development of  $\{010\}$ . Clearly,  $[010]$  and, to a lesser extent,  $[021]^*$  are directions of slow growth whereas  $[001]$  and  $[100]$  are directions of easy growth. An explanation for this situation lies in the uneven distribution of the compositional units in planes parallel to  $\{010\}$ . Along  $[010]$  the structure is composed of layers of relatively high packing density of  $SiO_4$  tetra-

hedra and  $M(1)$ -site cations centred at  $y = 0, \frac{1}{2}$  linked by interlayers of low packing density of  $M(2)$ -site cations centred at  $y = \frac{1}{4}, \frac{3}{4}$ . The latter are certainly zones of weakness in the structure and must be responsible for the imperfect to moderate  $\{010\}$  cleavage of olivine, cleavage along one  $\{010\}$  plane requiring the breaking of only six  $M(2)$ -O bonds per unit cell. The layers of high packing density would promote the formation of  $\{010\}$  faces containing relatively few unsaturated bonds and hence of low surface energy.

On any particular  $\{010\}$  growth face the

$M(2)$  protosites (terminology after Nakamura 1973) will always tend to be occupied, and one factor controlling further growth along [010] will be the ease with which the next layer of  $\text{SiO}_4$  tetrahedra and  $M(1)$  cations may be nucleated and developed. There are two quite distinct types of  $M(2)$  protosites differing in the nature of the three oxygens their coordination polyhedra share with the growing crystal. In the  $M(2)^i$  protosite polyhedron, these three oxygens form shared coordination edges whereas in that of  $M(2)^u$  they do not. Nucleation of a further growth layer requires that the oxygens in the liquid phase which complete these coordination polyhedra be associated with isolated  $\text{SiO}_4$  tetrahedral complexes and that these complexes have the appropriate spatial orientation. The development of the  $M(2)^i$  protosite requires the organization of three independent complexes and this, in particular, must be a rate controlling step. Also, the cation on this site may tend to coordinate with two oxygens from the same complex to mimic the coordination of  $M(1)$ -site cations. Finally, because of the various distorting forces in the olivine structure (for example, Fleet 1974), the three oxygens attached to the crystal in the  $M(2)^u$  protosite polyhedron have longer O-O distances. As a result, this protosite may tend to be occupied by larger divalent cations ( $\text{Ca}^{2+}$ ), which would be a further impediment to growth.

Thus, factors contributing to [010] being a direction of slow growth are the low surface energy of the faces of {010}, and various aspects of the stereochemistry of the coordination polyhedra of the  $M(2)$  protosites which tend to inhibit nucleation of subsequent growth layers.

Further examination of the olivine structure (Fig. 2) indicates other probable (but less-pronounced) growth layers and, inasmuch as  $M(1)$  will be more efficient bridging protosites than  $M(2)$ , these provide qualitative explanation for the relative importance of the forms {021}, {110}, {210}, {101} and {001}. In particular, {100} has stepped faces and this accounts for the relative unimportance of this form. {021} and {110} are important forms because they are bounded by unstepped layers of  $\text{SiO}_4$  tetrahedra and  $M$ -site cations.

The stepped faces of {100} and the population of the most probable growth surfaces of {001} and {101} by  $M(1)$  protosites all help to promote preferential growth in the  $a, c$  plane and thus contribute to the dominance of {010}.

The olivine structure then allows a rather straight-forward explanation of why the skeletal habit of Mg-rich olivine results from preferential growth in planes parallel to {010}, {021}

and, probably, {110}. The cellulation which gives rise to the skeletal forms is instigated by the outward growth of the crystal toward the nutrient and the bridging nature of the growth parallel to {021} and {110} tends to produce the three dimensional, lantern-like habit. Curiously, in the extensions from the lantern-like habit, the development of {021}, whilst still outward, is restricted to within the H-shaped or U-shaped units (Fig. 1) which make up the chains or blades. This chain or blade-like habit may reflect a particular degree of undercooling or supercooling although the extensions usually appear to have grown later than the lanterns and may be associated also with a lower concentration of crystal-forming constituents in the liquid.

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