

Cluster Formation of Cr-Spinel during Magmatic Differentiation in the Upper Mantle

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Abstract. Patterns of distribution of Cr-spinel in ultramafic cumulates and residues were examined by means of two dimensional center-center correlation and mass correlation functions. Both the center-center correlation and the mass correlation functions show a power law with various power exponents. The power exponents of center-center correlation and mass correlation range from 0.0 to 0.9 and 0.0 to 0.5, respectively, with development of clustering of Cr-spinel in the upper mantle peridotites. The patterns of distribution of Cr-spinel can be characterized by fractal dimension which regards the grain size distribution of cluster and matrix. Two distinctive patterns can be recognized in cumulate and residue peridotites: one is the homogeneous grain size distribution function and another is inhomogeneous size distribution function incorporated in the cluster formation, suggesting clusters sinking fractionation in the turbulent magma chamber.

1. Introduction

Dunite that formed accumulation process of olivine grains due to gravitational sinking at the bottom of magmatic chamber contains various amounts of chromite which grew simultaneously with olivine. Because of large discrepancy of density and growth velocity between olivine and chromite, the particle trajectories of them are quite different in the magma chamber. Considering that the magma shows turbulent motion in the magma chamber and olivine and chromite grains behave as a dense suspension particle in magma, they should collide frequently and stick with each other. Then, in the cumulate dunite, chromite grains occurs in the various

manner of aggregate: they are dispersive type, nodular, occluded, layer and podiform (Cassard *et al.*, 1981; Nicolas, 1989).

The distribution patterns of minerals that formed during gravitational sinking and nucleation and growth of mineral grains in magma chamber should be strongly governed by motion of magmatic fluid and mineral grains. In the case of very low particle density, falling velocity of grains is followed by the Stokes law, being independent of the particle density. In contrast, the falling mineral grains affect their motion with each other through surrounding fluid flow and grain collision in the relatively dense dispersion system (Batchelor, 1982). This effect results in clustering of like grains during sedimentation (Batchelor and Janse Van Rensburg, 1986).

Motion of magmatic fluid is a controlling factor of crystallization differentiation of the magma and mineral distribution in the magmatic chamber. Irvine (1980), Huppert and Sparks (1980), and Turner and Campbell (1986) suggested that the convective fractionation which is defined by the crystallization differentiation at the top of cumulus layer (bottom of the convective magma chamber) and at the side walls is essential in differentiation in the magma chamber. On the other hand, Martin and Nokes (1989) stressed the role of differentiation due to gravitational sinking of mineral grains in the turbulent magma chamber. As far as both processes should operate simultaneously, the ratio between crystallization rate at the bottom of the magma chamber and the settling rate of the grains in the turbulent magma chamber is very important to study dynamics of differentiation.

The simple clustering of grains has been modelled by Smoluchowski type coagulation (Friedlander, 1977; Ziff, 1984; Ernst, 1986). Ziff (1984) obtained the long-time behavior of the equation, and suggested that the size distribution of clusters after long times has a scale invariant law. Simulation of Smoluchowski type coagulation also shows a universal distribution pattern of grains that is measured by fractal dimension. The distribution patterns obtained from the Smoluchowski coagulation is just similar to those formed by simulation of the diffusion-limited-aggregation (so-called DLA) in the viewpoint of scale-invariance of the grain distribution. The fractal dimensions of the distribution patterns formed by Smoluchowski coagulation are controlled by mode of particle migration (ballistic or Brownian) and aggregation (cluster-cluster or particle-cluster) (Meakin, 1988). Thus, it is expected that the patterns of like grain distribution in cumulus peridotites are potential indicator of the dynamic state of the turbulent magma chamber.

2. Experimental Procedures

The distribution patterns of Cr-spinel grains in cumulates and residues were obtained by measurements of positions of significant amounts of grains under photographs of thin sections, which are normal to foliation plane and parallel to the lineation if present. The position data of Cr-spinel grains in a rock were measured by means of a digitizer connected with a microcomputer (Toriumi, 1986). Positions of four crests of ellipsoid-approximated grains of Cr-spinel were acquired and

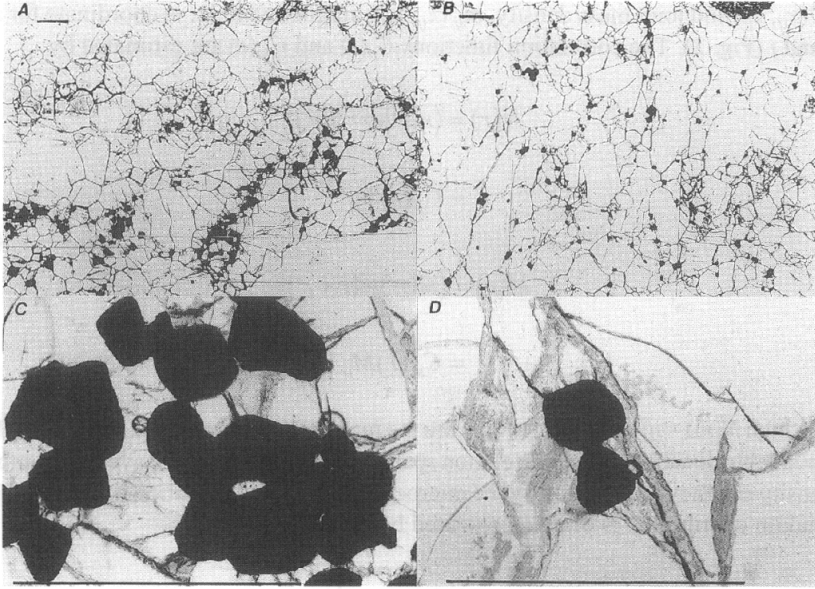


Fig. 1. Microstructures of Cr-spinel distribution in peridotite xenoliths and massive peridotites. The thin section is on the plane perpendicular to foliation/banding. Bar represents 1 mm in length. A; slightly clustered distribution in Takashima dunite, B; random distribution in Takashima dunite, C; spinel cluster in Takashima dunite, D; aggregated grains stuck by neck growth of spinel from magma.

stored in a floppy disk to obtain the center position, grain size, aspect ratio and orientation angle. Distribution patterns of Cr-spinel grains in a thin section were shown by the pair-correlation of the density function as follows; the center-to-center correlation was calculated from

$$x_{ij} = x_i - x_j \quad (1)$$

$$y_{ij} = y_i - y_j$$

in which (x_i, y_i) is the center position of i -th grain of Cr-spinel in a thin section coordinate. The mass correlation was calculated from

$$m_{ij} = (d_i d_j)^2 \quad (2)$$

where d_i is the grain radius of the i -th spinel. In the correlation functions, the position

of (x_{ij}, y_{ij}) and joint mass density, $m_{ij}(x_{ij}, y_{ij})$, are plotted in (x, y) coordinate for all i and j (Fig. 2). The correlation functions $C_p(r)$ and $C_m(r)$ are estimated by

$$\begin{aligned} N(r) &= \langle n(r')n(r+r') \rangle \\ &= C_p(r)N_o \end{aligned} \tag{3}$$

$$\begin{aligned} M(r) &= \langle m(r')m(r+r') \rangle \\ &= C_m(r)M_o \end{aligned}$$

in which $N(r)$ ($= n^2$) and $M(r)$ ($= m^2$) are the number density at r (r is normalized by a available distance) in the correlation space and N_o and M_o are the average number density and they are counted by the microcomputer. The correlation function of the random distribution is often represented by power law:

$$C_p(r) = gr_p^{-f} \tag{4}$$

or exponential form:

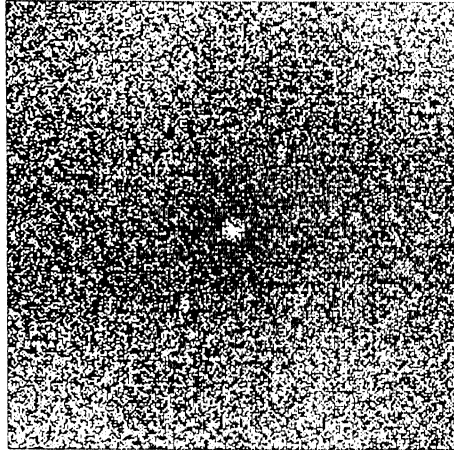


Fig. 2. The center-to-center correlation map (Fry's map) of Cr-spinel in Stillwater cumulates (444 grains).

$$C_p(r) = h \cdot \exp(-d_p \cdot r) \quad (5)$$

In the case of the power form, the homogeneous distribution is shown by $f_p = 0$ and in the case of the exponential form by $d_p = 0$. In Eq. (3), the power exponent, f_p is

$$f_p = d - D \quad (6)$$

in which d is the dimension of space studied and D is the fractal dimension (Mandelbrot, 1982, 1986).

To compare the distribution patterns of Cr-spinel clusters with simulated random fractal clusters, the center-to-center correlation functions of Levy's dis-

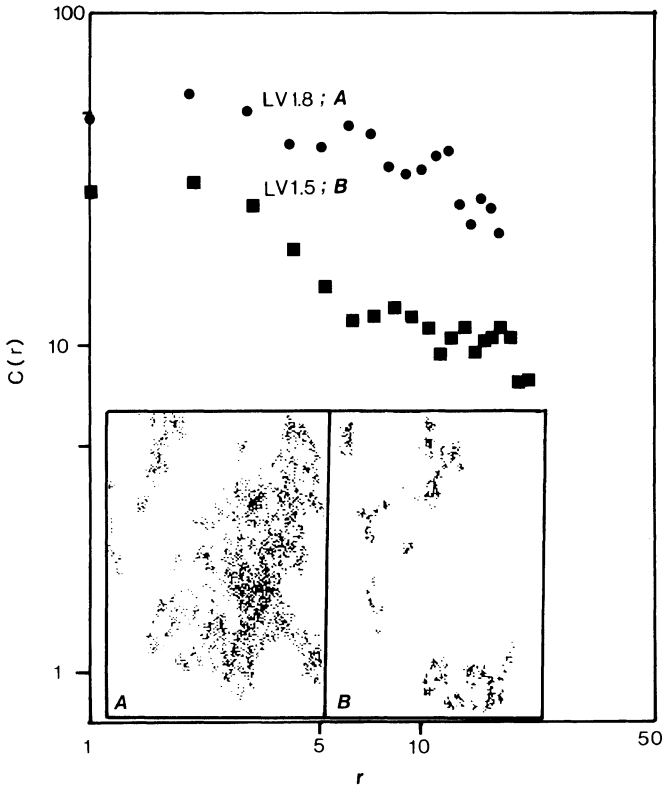


Fig. 3. The random fractal point distribution simulated by Levy's flight and their correlation functions with dimensions of 1.5 and 1.8, suggesting that 200 points statistics is safe for rough estimation of fractal dimension.

tribution (e.g. Takayasu, 1986) were investigated. The simulated results obtained for fractal dimensions of $D = 1.5$ and 1.8 are shown in Fig. 3. The correlation functions for Levy's distributions were made on 200 points to investigate stability of statistics. The fractal dimensions estimated from 200 points correlation functions are nearly equal to those prescribed at the simulation of Levy's distribution, suggesting that the 200 points correlation seems to give a stable statistics in the observed scales for random fractal structure.

Samples of upper mantle peridotites studied here were chosen from xenoliths in alkali basaltic rocks of Takashima and Kurose of northern Kyushu, Salt Lake, Hawaii, and from Miyamori and Iwanai massive peridotites, NE Japan and Hokkaido, respectively, and Stillwater peridotite complex, Montana. Peridotites studied here are olivine chromitite and Cr-spinel dunite, and Cr-spinel harzburgite and lherzolite (Fig. 1) because two end cases of high grain density and low density should be investigated. Dunite and chromitite of above localities are all cumulates and often have various sized clusters and layers of Cr-spinel, while lherzolite and harzburgite are residues. In many cases, cumulates have been formed by gravitational sedimentation of grains in the magmatic chamber having some extent, and residues by outflow of the partially molten magma in the upper mantle. Therefore, dynamic processes of formation of cumulates and residues are nearly inverse with each other. The formation of the cumulate peridotites includes the process of outflowing of trapped magma, and inversely, the formation of residue peridotites does small-scale sedimentation and compaction of several kinds of mineral grains during magma drainage. Thus, the distribution patterns of mineral grains were investigated to compare the structure formation and modification in the sedimentary and compaction processes.

3. Results

The center-to-center correlation maps of the studied Cr-spinel distribution in cumulus chromitite and dunite are shown in Fig. 4, and the correlation functions along x-axis are in Figs. 5 to 7. It is trivial from Eq. (1) that there is a centro-symmetric in the correlation map. The distribution patterns change obviously from homogeneous to clustered one. The correlation map indicates an average cluster shape in the clustered samples. In many cases, the average shapes of the cluster display the ellipsoidal outline elongated along the foliation and lineation. In the central part of the correlation map, the vacant ellipse means the average shape of chromite grains in cumulates. Even if grain density is very low, the vacant ellipse can be seen in the center, suggesting that the cluster center is slightly compact rather than in outer region of the cluster. It is clear that the average shape of the chromite grains in chromitites are a parallelogram and their flat plane is nearly parallel to the foliation plane (banding plane).

In dunites of Takashima, Stillwater, and Miyamori, the average clusters of Cr-spinel show a slight deformation from clear ellipsoids at the center (Fig. 4). It has

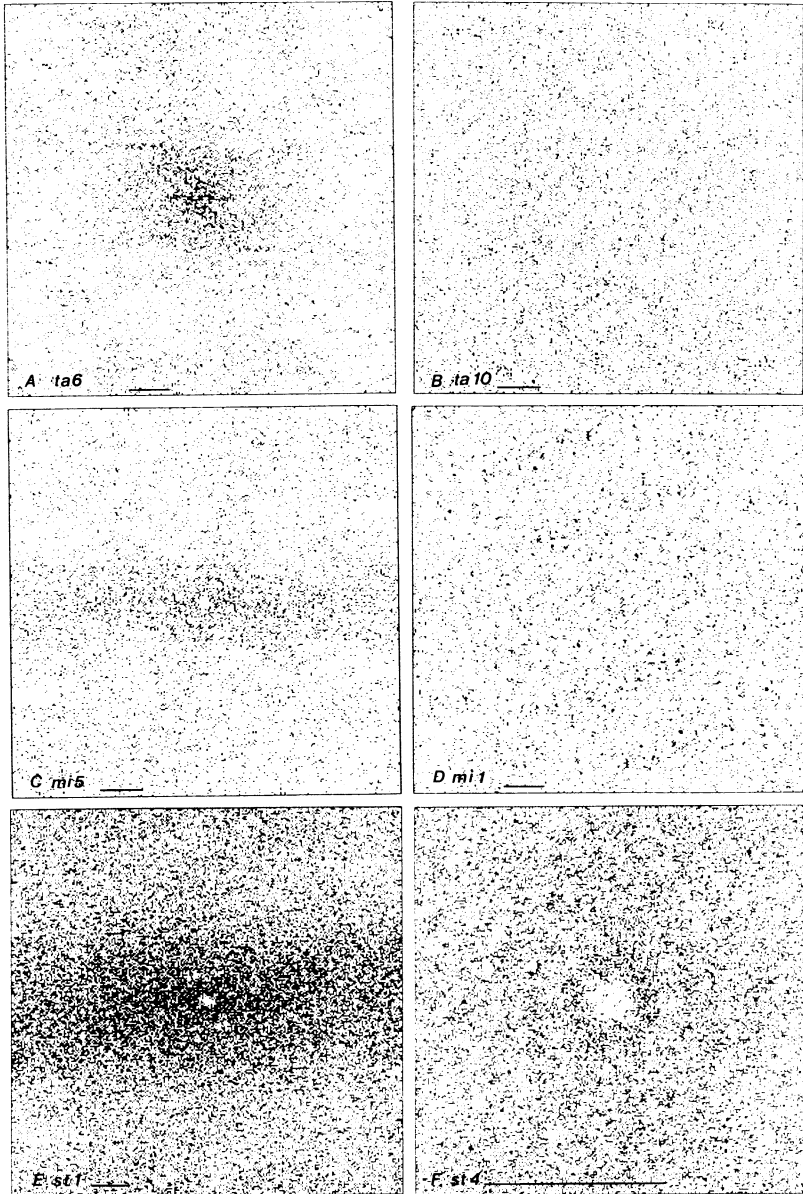


Fig. 4. The representative center-to-center correlation maps of cumulus dunite and chromitite of Takashima (A; clustered type, B; random type), Miyamori (C; clustered type, D; random type) and Stillwater (E; clustered chromitite and dunite layers, F; close-up view of the cluster center). Bar represents 1 mm in length.

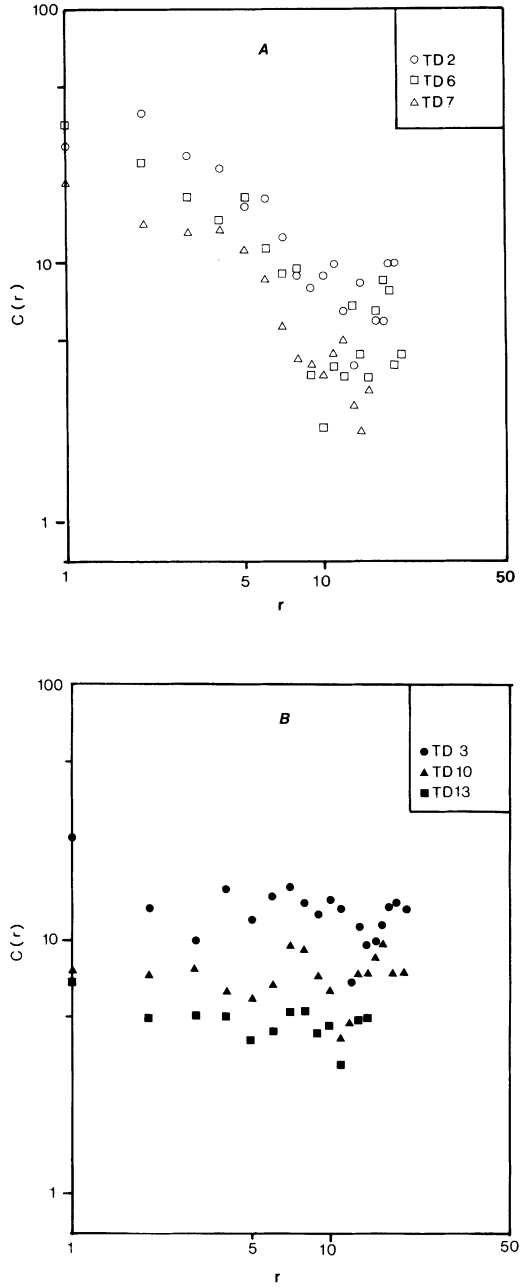


Fig. 5. The representative center-to-center correlation functions of spinel in Takashima dunite along the foliation plane in the full logarithmic diagram. A; clustered distribution, B; random distribution.

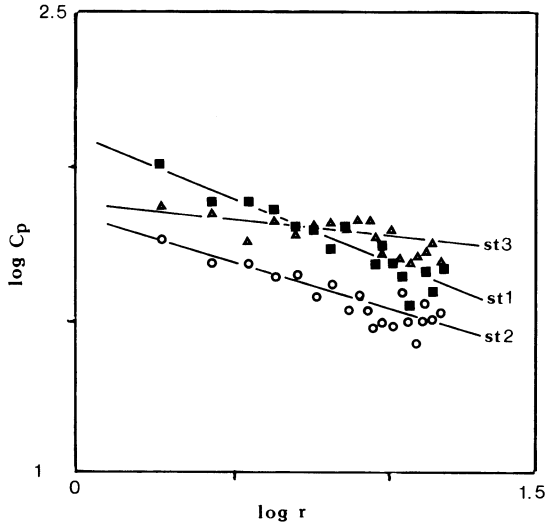


Fig. 6. The center-to-center correlation functions of spinel distribution in Stillwater cumulates(st1 and st2; chromite dunite, st3; chromitite), in the full logarithmic diagram, suggesting the power law relations.

often satellites of spots and arms at the margin, suggesting a cluster with filaments. In residue harzburgite and lherzolite of Kurose, Salt Lake and Miyamori, the correlation maps change from homogeneous type to inhomogeneous one (clustered type) as shown in Fig. 4. However, except for Kr-22, clustering is not so strong. There are many rocks showing no obvious cluster. Strong clustering in Kr-22 is seen in the correlation map but Cr-spinel in this sample has a very wide range in size. Very coarse grains often display amoeboidal outline, suggesting that they have been formed by compaction and strong sintering after clustering. This seems to be suggested by elliptical shape flattened along the banding plane. In contrast, Miyamori and Kurose lherzolites and harzburgites have no big grains but abundant amoeboidal and meniscus shape grains whose size is not so large compared with cumulus dunites. The correlation maps of these samples show a weak to moderate clustering patterns.

To discuss quantitatively progressive clustering of Cr-spinel grains in the turbulent magma chamber, it should be tested whether the center-to-center and mass correlation functions have a power relation or an exponential relation. Figures 5, 6, 7 and 8 indicate correlation functions in the full logarithmic diagram. If the function is an exponential form, it decreases abruptly in the large r . On the other hand, in the power law regime, it should be linear. As witnessed from these figures, correlation functions of strongly clustered distribution together with slightly clustered one have a power law type.

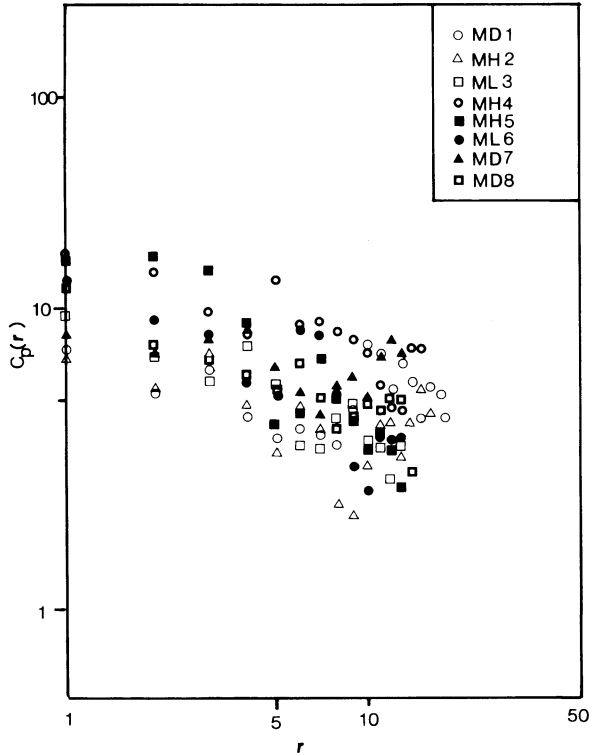


Fig. 7. The center-to-center correlation functions of spinel distribution in Miyamori cumulates(mi1, mi4, and mi7) and residues(mi2, mi3, mi5, mi6 and mi8). D, H, and L denote the dunite, harzburgite and lherzolite, respectively.

It is obvious that the systematic change of the power exponent, f_p for center-to-center correlation and f_m for mass correlation in the Takashima dunites and Miyamori residues is due to cluster development. The power exponents of the center-to-center correlation in Eq. (3) range from 0.0 to 0.8 in Takashima, and 0.0 to 0.5 in Miyamori. On the other hand, those of lherzolites of Kurose change from 0.2 to 0.4. The correlation functions of dunites and chromitites of Stillwater complex have power exponents ranging from 0.1 to 0.4. Above change is not related to the difference of grain size and grain density. Further, the power exponents, f_m , for the mass correlation are from 0.1 to 0.5 in Takashima, from 0.0 to 0.5 in Miyamori, and from 0.0 to 0.1 in Stillwater. Relations between power exponents f_m and f_p are shown in Fig. 9. It suggests that there are two types of clustering: the one shows the power exponent ranging in the region of $0.0 < f_p < 0.5$ and $0.0 < f_m < 0.3$ and the other does the range of $0.2 < f_m < 0.5$ and $0.7 < f_p < 0.9$. In addition, the

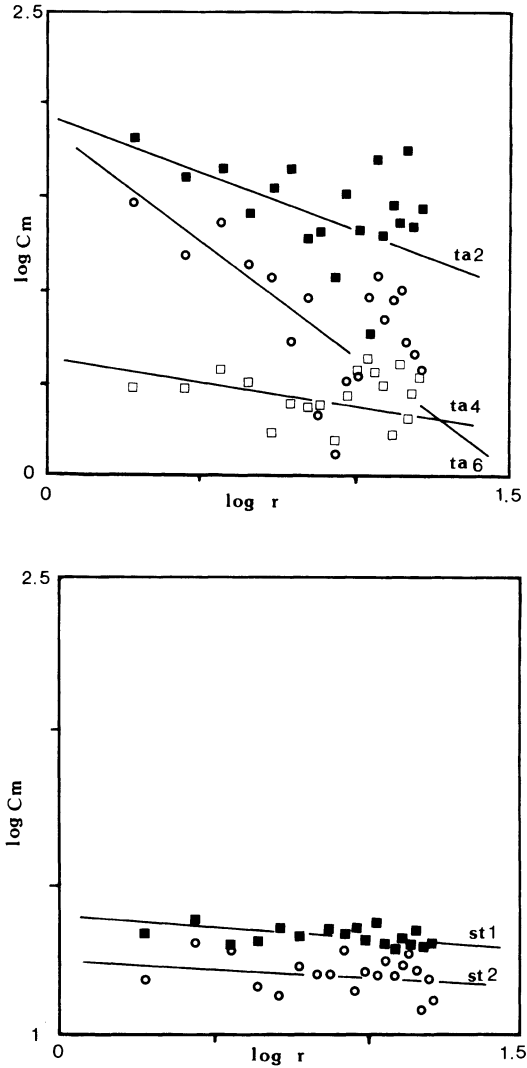


Fig. 8. The mass correlation functions of the representative peridotites of Takashima (upper) and Stillwater (bottom) in the full logarithmic diagram, suggesting the power law relations.

following results are summarized:

- 1) The number density of Cr-spinel grain does not give a significant effect on a whole cluster structure which is shown by center-to-center and mass correlations, judging from that power exponents of chromitites are in the same range as those in dunites.

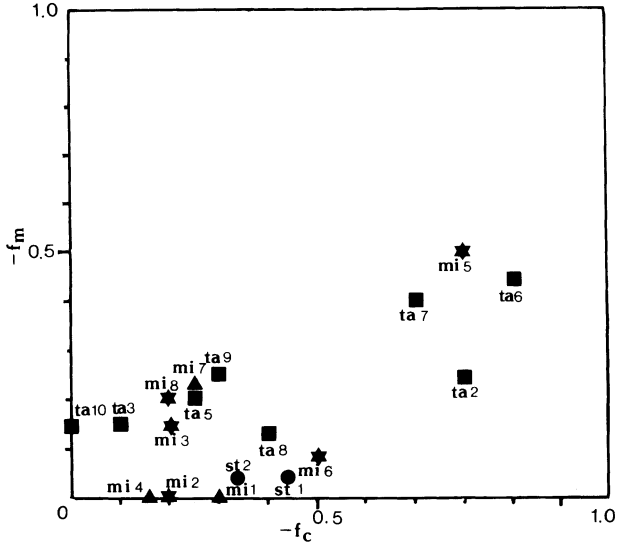


Fig. 9. The relations between power exponents of the center-to-center and the mass correlation functions of the Takashima (ta), Miyamori (mi), and Stillwater (st) peridotites cumulates (triangle and square) and residues (star).

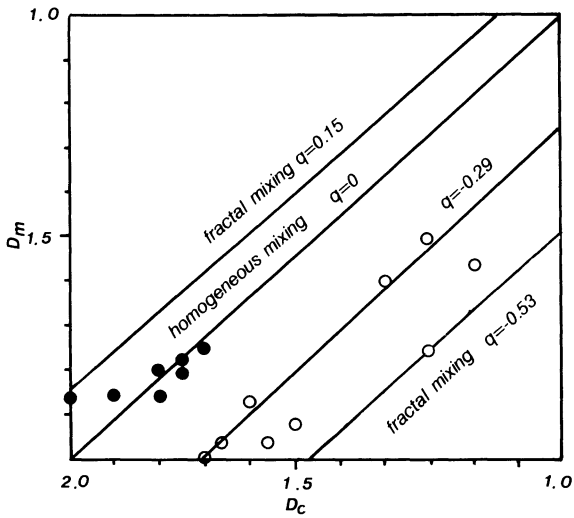


Fig. 10. Relations between fractal dimensions of center-center and mass correlation functions in the case of homogeneous and fractal mixing of spinel grains with various sizes in clusters.

2) The grain size does not give an effect significantly the center-to-center correlation function because there is no systematic relation in the power exponent and the grain size in Takashima dunites.

The grain size distribution of Cr-spinel in dunite has been also obtained simultaneously in this study. The grain size frequency of Stillwater samples display a LSW distribution (Fig. 10). Thus, the grain size distribution pattern is approximated to be one of simple Ostward type growth from magma.

4. Discussion

In this paper, it is first suggested that the distribution patterns of Cr-spinel in peridotite cumulates and residues can be divided into two types; the homogeneous type and clustered type. The center-to-center and the mass correlation functions display a power law. The power exponents in this regime represent the characteristic structure of the upper mantle peridotites formed by magmatic processes. If we consider the turbulent magma chamber, the maximum scale of eddies is to be the depth scale (Z) and the space of effective gathering of grains is thus to be $Z^3 \cdot n \cdot a^3$. Then, one can evaluate the maximum size of the cluster to be the order of $Z \cdot a \cdot n^{1/3}$, suggesting the same order of the magnitude as that in the quiescent sedimentation. On the contrary, the violent turbulent motion in magmatic fluid should breakup such clusters to form many small clusters due to various eddies. This process operates hierarchically on various clusters.

On the other hand, grain clustering in magmatic chamber requires for a mechanism with somewhat attractive force between two grains of Cr-spinel. As shown in Fig. 1, many Cr-spinel grains which contact with each other display the development of a neck at the contact. This neck is interpreted by two alternative ways: one is sintering after consolidation, and another is overgrowth during gravitational sedimentation. The latter can well interpret the chemical zonation and euhedral shape of Cr-spinel grains in cumulates. Thus, at the time of sedimentation and coagulation of Cr-spinel, two grains should be stucked due to overgrowth, if the contacting time is larger than the time scales for adhesion of several atomistic layers on these two grains. This process is similar to DLA (diffusion limited aggregation) mechanisms classified by Ernst (1986) and the coagulation model of garnet in solid state shear flow during synmetamorphic deformation by Toriumi (1986). In this simple physical process of grain coagulation in the turbulent magma chamber, the development of inhomogeneous distribution of Cr-spinel grains can be shown by the evolution of cluster distribution in magma chamber.

The structure formation of random clustering dynamics has been discussed by many authors from the viewpoint of coagulation dynamics in various fields of physics (Friedlander, 1977; Sander, 1984; Ziff, 1984; Ernst, 1986). There are three hierarchical structures that concern the coagulation. The first problem is the structure development of a cluster itself. The second is the frequency distribution of the cluster size. And the third is the whole structure of them. The first problem

can be generally investigated as a DLA (diffusion limited aggregation) and RLA (reaction limited aggregation) mechanisms (Sander, 1984; Jullien *et al.*, 1984; Kolb, 1984; Botet *et al.*, 1986), while the concentration of i -th clusters has been discussed by Smoluchowski's coagulation equation (Friedlander, 1977; Ziff, 1984; Ernst, 1986). Recently, the third problem has been discussed in the two dimensional experimental aggregation (Armstrong *et al.*, 1986) and shear clustering in natural phenomena (Toriumi, 1986).

The Smoluchowski's equation controlling the grain clustering dynamics is as follows;

$$dn_k / dt = (1/2) \sum_{i+j=k} K_{ij} n_i n_j - n_k \sum_j K_{kj} n_j \quad (1)$$

in which n_k is the concentration of k -th cluster composed of k grains and K_{ij} is the kernel that represents the coagulation rate between i -th and j -th cluster. The coagulation kernel is commonly assumed to be in a function of $(i+j)$ and ij . For example, shear coagulation by Ernst (1986) and Toriumi (1986) gives $K_{ij} = (i^{1/3} + j^{1/3})^2$ because two-body collisional cross section is proportional to their grain sizes. Ernst classified the coagulation into three classes by means of interaction types, that is, class I is large-large interaction dominant case, class III is large-small interaction dominant case, and class II is the intermediate case. So-called sol-gel transition occurred in the cases I and II. In the case of gravitational sedimentation of fine grains clustering dynamics seems to belong to the case I and thus it favours a sol-gel transition in which very large cluster forms likely to runaway growth. This equation has a solution of scaling form concerning the size distribution of the cluster (Ziff, 1984).

A single cluster has a scaling structure itself. Botet *et al.* (1984), Jullien *et al.* (1984) and Kolb (1984) have simulated directly above coagulation followed by Smoluchowski's equation by means of DLA model and obtain the power law type correlation functions:

$$C(r) = r^{-f} \quad (2)$$

in which C is isotropic grain density correlation function of clusters and f is about 0.6. Botet and Jullien (1985) obtained the structural change due to coagulation kernel assumed to be $K_{ij} = (ij)^w$. According to their results, it is very interesting that fractal dimensions of all clusters change asymptotically from 1.4 in the case of $w < 0$ to 1.7 in $w > 2$, suggesting the structural transition due to the change in its dynamics. In the transitional state that represents $0 < w < 2$, they obtained that the fractal dimension of large clusters is smaller than that of the small clusters, just experimentally indicated by Armstrong *et al.* (1986). They carried out two dimensional flocculation experiments and obtained that small clusters are more compact

($D = 1.7$) than large clusters ($D = 1.4$). This means a progressive change of the mean fractal dimension of many clusters due to progressive aggregation.

Next, we will investigate the change of fractal structure due to homogeneous deformation of cluster during compaction and plastic deformation after sinking of the clusters. In the case of Sierpinski carpets which are typical ordinary self-similar structure, it is trivial that homogeneous deformation does not change the fractal deformation (Mandelbrot, 1982, 1986; Lin, 1986). In contrast, when a random fractal structure such as DLA cluster deforms homogeneously, an isotropic density function changes to anisotropic function. Considering a 2D isotropic scaling density function of

$$p(r) = r^{-f} \quad (3)$$

then the scaling is $p(kr) = k^{-f}p(r)$.

The fractal dimension can be defined by

$$\int_0^R p(r) \cdot r dr \propto R^D. \quad (4)$$

If the deformation makes density function of a cluster to become

$$p(r) \rightarrow p(a \cdot r) \quad \text{with} \quad a = 1 + k \sin \theta \quad (5)$$

we obtain the following integrals

$$\begin{aligned} \int_0^R \int_0^{2\pi} p(a \cdot r) \cdot d\theta r dr &\propto g(k) \int_0^R p(r) r dr \\ &\propto g(k) \cdot R^D. \end{aligned} \quad (6)$$

In this equation, $g(k)$ is a function of k and depends on the finite strain, but not on r . Therefore, the fractal dimension of clusters is invariant against homogeneous deformation.

Another model of scale invariant cluster formation of Cr-spinel during gravitational sedimentation is considered to be as follows. A bottom current in the magma chamber makes often an inhomogeneity of spatial distribution of grains derived from momentum difference due to size and density differences. Thus, it is expected that 2D-inhomogeneity of Cr-spinel distribution on the floor of the magma chamber and successive stacking of that layer will be revealed.

5. Relation Between Center-To-Center and Mass Correlation Functions

The patterns of the Cr-spinel in cumulus and residual peridotites formed in the upper mantle can be discussed in the diagram of two fractal dimensions derived from point correlation and mass correlation functions as shown in Fig. 11. The mass correlation functions defined in the earlier section can be simply represented by two terms: average grain size d_a and correlation of number density, N at a distance, r , in the average cluster as follows;

$$M(r) = K(d_a)^2 \cdot N. \tag{7}$$

If it is the case of $d_a = k \cdot r^{-q}$, then we have

$$\begin{aligned} M(r) &= r^{-2q-f_c} \\ &= r^{-f_m}. \end{aligned} \tag{8}$$

Therefore, the relationship between the power indices of two correlation functions can be represented by

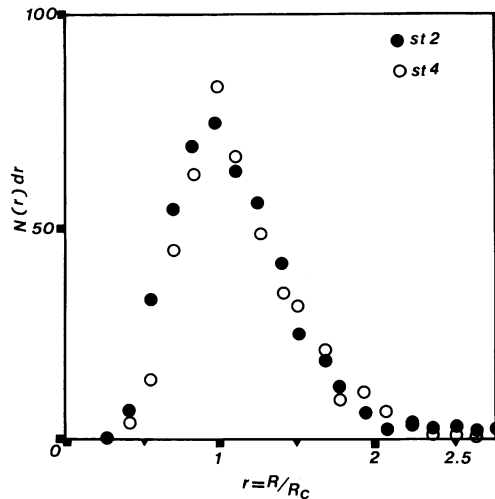


Fig. 11. Grain size distribution function of Cr-spinel for Stillwater dunite. R_c which is the grain size at the peak frequency is 450 μm for st2 and 500 μm for st4.

$$f_m = f_c + 2q. \quad (9)$$

Thus, in the case of the homogeneous mixing of various grain size, the fractal dimension of the point correlation is equal to that of the mass correlation function. As seen in Fig. 11, the natural patterns of spinel distribution can be largely divided into $q=0$ and $q=-0.3 \sim -0.5$, that is random mixing and fractal mixing of the various grains of spinel within dunites. The fractal mixing presented here is important for convective fractionation in magma chamber, because it may operate a large role for sedimentation even in the turbulent state. In this paper, the author pointed out that there are two indicators in fractal distribution of Cr-spinel in peridotites. Considering the Stokes' sinking of Cr-spinel in the turbulent magma chamber, terminal velocity (v) of Cr-spinel grain is proportional to square of their grain size. Therefore, the collisional frequency having grain size, d , is approximated to be as follows;

$$\begin{aligned} P_d &= (2d)^2 \cdot v \cdot n(d) \\ &= d^4 n(d) \\ &= d^2 \end{aligned} \quad (10)$$

for large d because of $n(d) \propto d^{-2}$ in large d .

Therefore, the large grains should be easy to collide with each other. Considering that the sticking is due to overgrow from magma, the net aggregation coefficient becomes large with increasing the size of grain. Consequently, the relations of fractal dimensions of mass and center distribution patterns of Cr-spinel in peridotites may come from the clustering dynamics during the gravitational sinking in the turbulent magma chamber.

6. Conclusion

The distribution patterns of Cr-spinel in cumulates and residues of the upper mantle peridotites of Takashima, Kurose, and Salt Lake, Miyamori and Stillwater massive peridotites, were examined by means of the center-to-center and mass correlation functions. It is suggested that the correlation functions have a scale-invariant nature, and the power exponents of the scaling law range from 0 to 0.9 for the center-to-center correlation and from 0.0 to 0.5 for the mass correlation. The relations between the above two power exponents suggests that there are two types of distribution structure of spinel: the one is the strongly clustered state and the other is the random to weakly clustered state. Thus, it is concluded that clustering dynamics during magmatic sedimentation may operate to form a fractal structure of Cr-spinel distribution in the upper mantle.

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