# Grain Growth in Three Dimensions Depends on Grain Topology

James A. Glazier[a]

*Research Institute for Electrical Communications, Tohoku University, 2-1-1 Katahira, Sendai 980, Japan*

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While the growth rate of the average volume in a three-dimensional polycrystal is well understood, the growth rates of individual grains (which determine material properties through the topological and volume distributions) are not known. Using a three-dimensional $Q$-state Potts model simulation, I find that the average canonical growth rate of a grain depends linearly on its number of faces and is independent of its volume, a surprisingly simple and unexplained result.

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What is the rate of growth (or shrinkage) of a single grain in an ideal three-dimensional froth or polycrystal during annealing? On what variables does it depend? The *growth exponent* $\alpha$ of the *average grain volume* $\langle v\rangle \propto t^{\alpha}$, $\alpha=1.5$, is well understood, but says little about the dynamics of individual grains [1–3]. Static grain shape distributions are known in some special cases in metals, in froths, and elsewhere [1,2,4], but depend on details of the experiment. Such measurements cannot predict the evolution of the volume or topological distributions, which determine many properties of materials. Thus understanding how the topology and volume of a grain affect its growth is an important open problem.

A physically based theory for the growth of individual grains, which is a fundamental process in the evolution of cellular structures, is lacking [5]. Rivier has made the interesting phenomonological proposal that $[dv_f/dt] \propto f-\langle f\rangle$ [5,6], where $v_f$ is the volume of a grain with $f$ faces and $\langle f\rangle$ the average number of faces in the froth. However, because of the normalization, his growth rate predicts $\alpha=1$, disagreeing with experiment. Fortes has also studied the problem [5].

In two dimensions, on the other hand, von Neumann's law suggests that the growth rate of a grain depends only on its number of sides $n$ [7]. The simplicity of the law has led to a host of theoretical and experimental studies of quasi-two-dimensional grain growth, in metal foils, soap froth, lipid monolayers, and elsewhere [8], and to practical results in integrated circuit fabrication [9].

In this Letter I use a $Q$-state Potts model simulation to show that even in three dimensions the properly normalized average growth rate of $f$-faced grains is a linear function of $f$.

*von Neumann's law.*— In an ideal soap froth, minimization of surface energy results in grain boundaries that are minimal surfaces with constant mean curvature, $K \equiv 1/r_1+1/r_2$, where $r_1$ and $r_2$ are the two independent radii of curvature. The mean curvature determines the effective pressure difference across the grain wall, $\Delta P$, via the Young-Laplace law

$$\Delta P=\kappa_0(1/r_1+1/r_2)\,, \tag{1}$$

where $\kappa_0$ is a diffusion constant [10]. In three dimensions, edges meet in fourfold vertices at tetrahedral angles [$\theta\equiv\arccos(-1/3)$] [5]. In two dimensions, edges meet in threefold vertices at 120°. The rate of transfer of volume across the grain boundary is proportional to the boundary area times $\Delta P$. von Neumann observed that in two dimensions the sum over all sides of this product depends only on the grain's $n$, and hence [7]

$$da_n/dt=\kappa(n-6)\,, \tag{2}$$

where $\kappa$ is a diffusion constant and $a_n$ the area of the grain. Thus the growth rate for all $n$-sided grains is identical and area independent. Grain growth in metals requires a slightly different derivation but leads to the same result [8]. Avron and Levine have extended the law to two dimensional curved surfaces [11].

In three dimensions the derivation fails because the topology does not determine the mean curvature, but the Gaussian curvature $G\equiv\langle 1/r_1r_2\rangle$ [11]. In two dimensions the two curvatures are equivalent, which leads to von Neumann's law. In three dimensions they are independent.

In real soap froth the growth rates for individual bubbles fluctuate due to the finite fluid fraction [though in the limit of an ideal dry froth Eq. (2) is exact] [8,12–15]. However, an averaged von Neumann's law holds:

$$[da_n/dt]=\kappa(n-6)\,. \tag{3}$$

Note that $[da_n/dt]\neq d\langle a_n\rangle/dt$, since changes in grain topology affect the right-hand side (rhs). When the distributions of the topologies and normalized volumes are constant, i.e., in the *scaling regime*, Eq. (3) implies that the left-hand side is negative for $n<6$, while the rhs is positive for all $n$, since the average area of all grains $\langle a\rangle$ increases with time, and $\langle a_n\rangle$ must be proportional to $\langle a\rangle$ in a scaling state.

*Scaling.*— Experimentally, Durian *et al.* found that $\alpha=1.5$ in three-dimensional shaving cream [16], and $\alpha=1.5$ in ideal metals and Potts model simulations [1,2,8]. However, any growth law which obeys Eq. (1) and which reaches a scaling state has $\alpha=1.5$, since Eq. (1) implies, for each grain,

$$dv/dt=\kappa_1 a\langle 1/r_1+1/r_2\rangle\,, \tag{4}$$



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where $a$ is the surface area of the grain, $\kappa_1$ a diffusion constant, and the average is over the grain surface. For two grains, $x$ and $y$, of the same shape, with volumes $v_x$ and $v_y$, $v_x^{1/3}/r_{1x} = v_y^{1/3}/r_{1y}$ and $v_x^{1/3}/r_{2x} = v_y^{1/3}/r_{2y}$. Therefore

$$v_x^{-1/3} dv_x/dt = v_y^{-1/3} dv_y/dt = \mathcal{C}(\text{shape}) . \quad (5)$$

The constant $\mathcal{C}$ (units of [area/time]) is the *normalized growth rate* of the grains. In the scaling regime, in which the grain shape distributions are independent of time, dimensional arguments yield $\alpha = 1.5$.

Thus $\alpha$ gives no information about the growth rates for individual grains, which could depend in a complicated way on $v$ or other parameters. A strict three-dimensional analog to the averaged von Neumann's law exists if the average of $\mathcal{C}(\text{shape})$ is a function only of $f$, i.e.,

$$[\mathcal{C}(\text{shape})] = C(f) \equiv [v_f^{-1/3} dv_f/dt] , \quad (6)$$

and $C(f)$ is linear in $f$.

*The Potts model.*—In both two and three dimensions, the $Q$-state Potts model reliably reproduces experimentally observed grain growth, with certain specific exceptions which I will discuss [2,16]. The Hamiltonian is

$$\mathcal{H} = \sum_{(i,j,k)(i',j',k') \text{ neighbors}} 1 - \delta_{\sigma(i,j,k),\sigma(i',j',k')} , \quad (7)$$

where $N$ degenerate spins, $\sigma(i,j,k) = 1,2,\ldots,N$, label $N$ grains, and $(i,j,k)$ identifies a lattice site. Each grain has a separate spin to prevent grain coalescence. The volume $v$ of a grain $\sigma$ is the number of lattice sites of spin $\sigma$, the surface area $a$ is the number of lattice sites of spin $\sigma$, which neighbor sites of different spin, and the number of faces $f$ is the total number of different grains neighboring all sites of spin $\sigma$.

Each lattice site has 26 neighbors, surrounding it in a cube of side three. The third-neighbor interaction reduces the orientational anisotropy of wall energies [17]. However, grain boundaries still align preferentially along low-energy directions, which increases fluctuations in the growth rates.

The size of the lattice is $100 \times 100 \times 100$, with periodic boundary conditions. At each time step I select a lattice site at random and change its spin from $\sigma$ to $\sigma'$ with probability $P(\sigma(i,j) \rightarrow \sigma'(i,j)) = \{0$: $\Delta\mathcal{H} > 0$; 0.5: $\Delta\mathcal{H} = 0$; 1: $\Delta\mathcal{H} < 0\}$, where $\Delta\mathcal{H}$ is the energy gain produced by the change, i.e., the simulation temperature is 0. One Monte Carlo step (MCS) corresponds to as many trials as there are lattice sites.

To eliminate any spurious symmetries I use an extended Potts model to generate an initial pattern consisting of about 20000 grains whose $v$'s are randomly chosen to be either 15 or 25 lattice sites [18]. To reach the scaling regime, I allow the simulation to coarsen for 160 MCS, when it has about 4000 grains. Then, after each MCS, I check for all the grains which have not changed $f$ from the previous time step (the probability that a grain will change $f$ several times within an MCS and be counted erroneously is negligible). I then record the grain's volume at the middle of the MCS, $v'$, $f$, $a$, and $c(v',f) \equiv v'^{-1/3} dv/dt$. The simulation ends when there are about 400 grains left.

*Results.*—I present $C(f)$ (units [lattice sites$^{2/3}$/MCS]) averaged for two independent simulations in Fig. 1. For $f \geq 10$, $C(f)$ is roughly linear in $f$, with a zero intercept of $f = 15.8 \pm 0.1$, substantially different from $\langle f \rangle = 14.08 \pm 0.02$. To reduce the fluctuations due to the Monte Carlo simulation, if a grain does not change $f$ over several time steps, I average $c(v,f)$ over the entire time and treat the result, $c'(v,f)$, as a single growth rate. However, treating each $c(v,f)$ independently increases the standard deviation by only about 15%, suggesting that $c(v,f)$ fluctuates very little between time steps. When I calculate $v$ dependent quantities, I use $c(v,f)$ rather than $c'(v,f)$.

The nonlinearity visible in Fig. 1(a) for $7 \leq f \leq 9$ results from the lattice discretization. In deriving Eq. (6) I assumed that the ratio, $R(v,f) \equiv v^{2/3}/a$, was independent of $v$ and $f$. For regular polyhedra with flat faces, $R(v,f)$

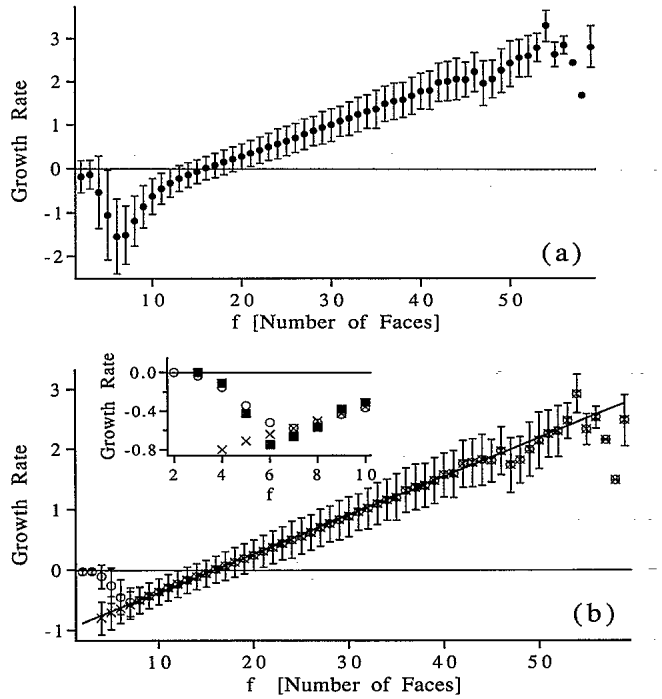


FIG. 1. Growth rates as a function of number of faces. (a) Raw normalized growth rate $C(f)$. Error bars are 1 standard deviation. (b) Growth rates corrected for the volume dependence of the ratio $R$ (see Fig. 2). Open circles are the corrected growth rate $C'(f)$, calculated for all volumes. ×'s are the canonical growth rate $C_0(f)$, calculated for volumes $\geq 8$. For $f > 6$, $C'(f) = C_0(f)$. Error bars are 1 standard deviation. Solid line is a linear fit to the canonical growth rate between $f = 4$ and $f = 45$. Inset shows growth rates for $f \leq 10$: Corrected growth rate for one MCS time step (circles), corrected growth rate for MCS/4 time step (solid squares), and canonical growth rate for one MCS time step (×).

depends on $f$ only [e.g., for $R(v,4)=0.114$, $R(v,6)=0.167$, and for a sphere, $R(v,\infty)\equiv R_{\rm sph}=0.207$]. However, in the Potts model, $R(v,f)$ depends on $v$. For any grain of $1\le v\le 26$, all lattice sites are surface sites, so $a=v\to R(v,f)=v^{-1/3}$. Empirically, in the scaling regime, for $v>26$, $R(v,f)=0.138+0.138e^{-0.0015v}+0.226e^{-0.021v}\pm 3\%$. So $R(v,f)$ is essentially independent of $f$. I define $\lim_{v\to\infty}R(v,f)\equiv R_\infty\approx 0.138\pm 0.003$, and $R(v)\equiv\langle R(v,f)\rangle$ averaged over $f$.

$R(v)$ establishes $v\approx 100$ (corresponding to $f\approx 10$) as the minimum above which I except the uncorrected Potts model to agree quantitatively with continuum models and experiments. I therefore define the *corrected growth rate as a function of f*:

$$C'(f)\equiv\left[\frac{R_\infty}{R(v)v_f^{1/3}}\frac{dv_f}{dt}\right]. \tag{8}$$

I plot $C'(f)$ in Fig. 1(b). It is linear for $f\ge 7$ [$C'(f)=-1.014+0.064f$ with $\chi^2=0.02$].

The nonlinearity present in Fig. 1(b) for $f\le 6$ also depends on the time discretization. For grains with $f\le 6$, the expected shrinkage per grain per MCS, extrapolating $C'(f)$ linearly, is about 1.5 lattice sites, while the typical $v$'s are $\langle v_6\rangle\approx 26$, $\langle v_5\rangle\approx 10$, $\langle v_4\rangle\approx 2.5$, and $\langle v_3\rangle\approx\langle v_2\rangle\approx 1$. Hence, many grains in these classes will either disappear or change $f$ within a single MCS, excluding them from the average used to calculate $C'(f)$. Only those grains which, for statistical reasons, shrink more slowly, are included. This selection bias forces $C'(f)$ towards zero as $\langle v_f\rangle$ approaches zero.

I can correct for this bias by considering only grains with $v\gg -v^{1/3}C'(f)$. In Fig. 1(b) I display $C_0(f)$, the corrected growth rate calculated only for grains with $v\ge 8$. No grains with $f=2,3$ have $v\ge 8$, however, grains with $f=2$ and 3 do not occur spontaneously in materials, but are an artifact of the discrete simulation. $C_0(f)$ is linear for all $f>3$ [$C_0(f)=-1.028+0.064f$ with $\chi^2=0.02$], and for $f>6$, $C_0(f)=C'(f)$. I therefore call $C_0(f)$, the *canonical growth rate.*

Reducing the time step also reduces the bias, but at the cost of larger fluctuations and more computer time. The inset to Fig. 1(b) compares the corrected growth rate obtained in a short duration simulation using a time step of MCS/4, to $C'(f)$ and $C_0(f)$. The linearity is better than for $C'(f)$, but worse than for $C_0(f)$.

*Discussion.*—Since $\langle v_f\rangle\propto f^3$ and the corrected growth rate as a function of volume, $C'(v)$ (Fig. 2), increases with $v$, the increase of $C_0(f)$ with $f$ might result from the correlation between $f$ and $\langle v_f\rangle$ [2]. Consider $C'_f(v)$, the corrected growth rate for grains with a fixed $f$, as a function of $v$, calculated using a logarithmic $v$ bin. If the corrected growth rate depends primarily on $v$ and not on $f$, $C'_f(v)$ should be essentially identical to $C'(v)$ for all $f$. On the other hand, if the corrected growth rate depends primarily on $f$, then $C'_f(v)$ should be essentially independent of $v$, but a strong function of $f$.

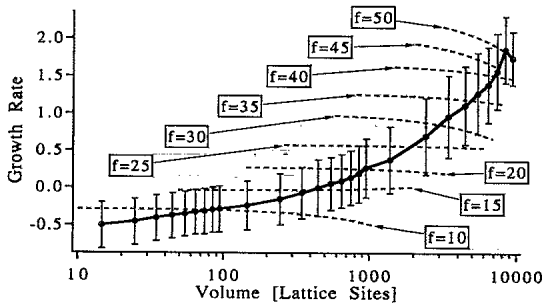


FIG. 2. Corrected growth rates as a function of volume. Dashed lines show linear fits to the corrected growth rates for representative topological classes (as labeled) as a function of volume $C'_f(v)$. Solid line shows the total corrected growth rate as a function of volume $C'(v)$. Error bars are 1 standard deviation. All calculations use logarithmic volume binning.

In Fig. 2, I display $C'_f(v)$, for every fifth topological class, from $f=10$ to $f=50$ [19]. The typical slope $dC'_f(v)/dv=-2\times 10^{-5}(+2.5\times 10^{-5}/-7\times 10^{-5})$, i.e., it is essentially 0. The slope $dC'_f(v)/dv$ decreases monotonically from $1.7\times 10^{-3}$ at $v=50$, to $5.3\times 10^{-4}$ at $v=500$, to $1.8\times 10^{-4}$ at $v=5000$, but is always at least 40 times larger than the largest positive slope for any of the $C'_f(v)$. [The apparent increase in the slope of $C'(v)$ is due to the logarithmic $v$ scale.] Thus the $v$ dependence of the corrected growth rate is a consequence and not a cause of the $f$ dependence.

Similarly, if the topology alone determines the corrected growth rate $C'_v(f)$ the face dependence of $C'$ for a given $v$ range should be independent of the range of $v$ and equal to $C'(f)$. I plot $C'_v(f)$ for representative $v$ ranges in Fig. 3. For $v>10$ and $f>6$, all $C'_v(f)$ curves are equivalent to each other in their region of overlap and to the linear fit to $C_0(f)$ given in Fig. 1(b). Even for $10\le v<20$ the agreement is exact. For $v<8$, $C'_v(f)$ approaches 0 due to selection bias. There is insufficient data for the fit for $f\le 6$ and $v>10$. For $f>40$ and for larger $v$, the error bars are larger but the general agreement remains good. Thus for $10\le v<9000$ the $C_0(f)$ does not depend on $v$.

The canonical growth rate of three-dimensional grains is independent of volume and depends linearly on the number of faces, yielding the averaged three-dimensional law

$$\left[\frac{dv_f}{dt}\right]=\kappa'\frac{R_\infty}{R(v)}v^{1/3}(f-15.8), \tag{9}$$

where $\kappa'=0.064$ [lattice sites$^{2/3}$/MCS] and the average is restricted to $v\ge 8$. In the continuum limit the average is unrestricted and $R_{\rm sph}/R(f)=1$ replaces $R_\infty/R(v)$. In real materials it may also be necessary to correct for finite fluid fractions, buoyancy effects [20], and finite grain boundary widths.

This ideal linear result is surprising. First, the lack of $v$ dependence implies that in a froth or polycrystal the

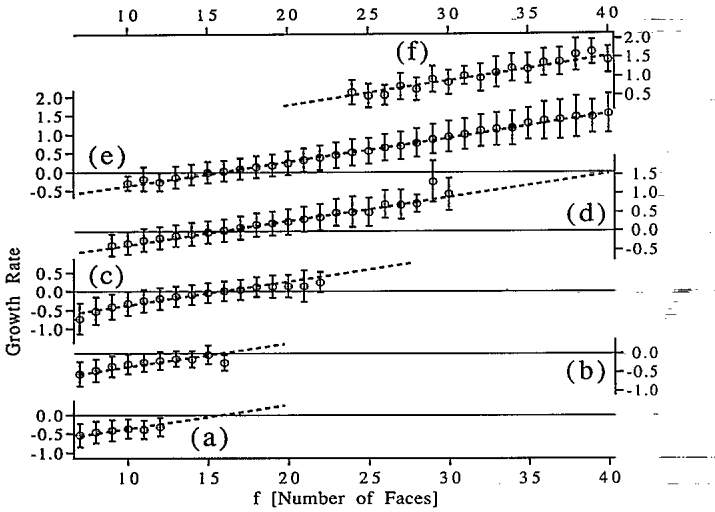


FIG. 3. Corrected growth rates as a function of number of faces $C'_v(f)$ for representative area classes. (a) $10 \leq v < 20$, left axis scale. (b) $50 \leq v < 60$, right axis scale. (c) $100 \leq v < 200$, left axis scale. (d) $500 \leq v < 600$, right axis scale. (e) $1000 \leq v < 2000$, left axis scale. (f) $5000 \leq v < 6000$, right axis scale. Error bars are 1 standard deviation. Dashed lines show best linear fit from Fig. 1(b).

physics of the pattern evolution creates a relation between the Gaussian curvature and the mean curvature, which are, in principle, independent quantities. Second, even if $C_0$ depends only on $f$, there seems no reason that the relation should be linear.

Because the Potts model is discreet and statistical, it cannot directly determine if Eq. (9) holds for individual grains in the continuum limit. Simulations on a larger lattice would help. If not, is there an exact law depending on detailed topology (the organization of faces and number of sides per face) [21]? The standard deviation of $C_0$ is nearly constant with $f$, while the number of detailed topologies increases rapidly [21], hinting that $f$ is more likely to determine the growth rate. Experimental investigations in soap froth, or continuum simulations should clarify these points.

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[a]Permanent address and address for correspondence: Department of Physics, University of Notre Dame, Notre Dame, IN 46556.

[1] There have been many experimental studies of three dimensional grain distributions in different materials. For a bibliography see J. A. Glazier, Ph.D. dissertation, University of Chicago, 1989 (unpublished).

[2] M. P. Anderson, G. S. Grest, and D. J. Srolovitz, Scr. Metall. **19**, 225 (1985); G. S. Grest, D. J. Srolovitz, and M. P. Anderson, Phys. Rev. B **38**, 4752 (1988); M. P. Anderson, G. S. Grest, and D. J. Srolovitz, Philos. Mag. B **59**, 293 (1989).

[3] D. J. Durian, D. A. Weitz, and D. J. Pine, J. Phys. Condens. Matter **2**, SA433 (1990); D. J. Durian, D. A. Weitz, and D. J. Pine, Science **252**, 686 (1991); D. J. Durian, D. A. Weitz, and D. J. Pine, Phys. Rev. A **44**, 7902 (1991).

[4] Simulations in three dimensions using other methods include: F. N. Rhines, K. R. Craig, and R. T. DeHoff, Metall. Trans. **5**, 413 (1974); S. K. Kurtz and F. M. A. Carpay, J. Appl. Phys. **51**, 5725 (1980); T. Nagai, S. Ohta, K. Kawasaki, and T. Okuzono, Phase Transitions **28**, 177 (1990).

[5] N. Rivier, Philos. Mag. B **47**, L45 (1983); Sir W. Thomson, Philos. Mag. **24**, 503 (1887); M. A. Fortes, J. Mat. Sci. **21**, 2509 (1986).

[6] Throughout this Letter, square brackets represent an average over a short time, restricted to those grains which do not change $f$ during that time. Angle brackets represent an unrestricted average at a fixed time.

[7] J. von Neumann, *Metal Interfaces* (American Society for Metals, Cleveland, 1952), p. 108.

[8] Recent reviews include: H. V. Atkinson, Acta Metall. **36**, 469 (1988); J. A. Glazier and D. Weaire, J. Phys. Condens. Matter **4**, 1867 (1992).

[9] D. T. Walton, H. J. Frost, and C. V. Thompson, Appl. Phys. Lett. **61**, 40 (1992).

[10] J. A. F. Plateau, *Statique Experimental et Théorique des Liquides Soumis aux Seules Forces Moléculaires* (Gauthier-Villars, Paris, 1873); J. A. F. Plateau, Mem. Acad. Roy. Belg. **16**, 1 (1843), continuing in subsequent issues through 1872.

[11] J. E. Avron and D. Levine, Phys. Rev. Lett. **69**, 208 (1992).

[12] J. A. Glazier, S. P. Gross, and J. Stavans, Phys. Rev. A **36**, 306 (1987); J. Stavans and J. A. Glazier, Phys. Rev. Lett. **62**, 1318 (1989); J. A. Glazier and J. Stavans, Phys. Rev. A **40**, 7398 (1989).

[13] F. Bolton and D. Weaire, Phys. Rev. Lett. **65**, 3449 (1990); F. Bolton and D. Weaire, Philos. Mag. B **63**, 795 (1991).

[14] F. Bolton and D. Weaire, Philos. Mag. B **65**, 473 (1992).

[15] J. Stavans, Phys. Rev. A **42**, 5049 (1990).

[16] P. S. Sahni, D. J. Srolovitz, G. S. Grest, and M. P. Anderson, Phys. Rev. B **28**, 2705 (1983); J. A. Glazier, M. Anderson, and G. Grest, Philos. Mag. B **62**, 615 (1990).

[17] E. A. Holm, J. A. Glazier, D. J. Srolovitz, and G. S. Grest, Phys. Rev. A **43**, 2662 (1991).

[18] F. Graner and J. A. Glazier, Phys. Rev. Lett. **69**, 2013 (1992); J. A. Glazier and F. Graner, Phys. Rev. E (to be published).

[19] The slope for $C'_f(v)$ for $f < 8$ is large and negative as a result of selection bias for smaller $v$. For very many faced grains, the statistics do not permit an accurate calculation of the slope. The origin of the very small negative slope for most $f$ (31 out of 40) appears to be the finite time step. I calculate the ratio $R$ in the middle of each time step. Thus for growing grains, since the second derivative of $R$ is decreasing, I multiply by a slightly larger $R$ than I would for smaller time steps. This increase in $R$ decreases for larger $v$, resulting in a slightly reduced $C'_f(v)$.

[20] Dinos Gonatas (private communication).

[21] The number of detailed topological classes for many faced polyhedra is very large. See M. A. Fortes and A. C. Ferro, Acta Metall. **33**, 1683 (1985); **33**, 1697 (1985).