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ORIGINAL PAPER

Fractal Dimensions of Simulated and Real Fat Crystal Networks in 3D Space

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Abstract The microstructure of fat crystal networks is closely related to rheological properties of fat products, and thus is of particular interest to food scientists. The fractal dimensions of fat crystal networks calculated by microscopy methods such as box-counting, D_b, particle-counting, D_f, and mass fractal dimension, D_m, have been extensively employed to quantify the microstructure of fats. This work revealed the microstructural basis of D_b, D_f, and D_m in 3D space through both computer simulation and experiments on the high melting fraction of milk fat crystal networks. Similar to our previous simulation study on the fractal dimensions of fat crystal networks in 2D space, D_b is sensitive to crystal size, area fraction, and not sensitive to distribution orderliness of crystals, which is the percentage of evenly distributed fat crystals within the simulated fat crystal networks. D_f and D_m were not affected by any of the microstructural factors studied in the simulation.

Keywords Simulation · Fractal dimensions · Fat crystal networks

Introduction

For most semisolid fat products, textural properties are determined by the solid component of the system, which normally exists as a 3D colloidal fat crystal network. A variety of microscopy methods such as Polarized Light Microscopy (PLM), Scanning Electron Microscopy (SEM), Confocal Scanning Light Microscopy (CSLM), Freeze Fracture Electron Microscopy (FFEM) [1–6] and many others have been employed to visualize the microstructure of fat crystal networks. Nowadays, as image analysis software becomes widely available, food scientists are not content with only the qualitative description of the microstructure of fats. Attempts [1, 5, 7] have been made to quantify the structure and spatial distribution of fat crystals within fat crystal networks, among which fractal analysis has gained popularity.

The concept of a fractal dimension was first introduced by Benoit Mandelbrot [8]. The idea is to use a fractal number to quantitatively describe the self-similar or self-affine character of some objects. Fractal geometry provides a new paradigm to understand many physical phenomena. The fractal nature of fat crystal networks was first recognized by Vreeker et al. [9] and further explored by our research group [1, 2, 10]. Fractal dimensions calculated by microscopy methods such as box-counting, particle-counting, and Fourier Transforms of polarized light micrographs of fats (termed D_b, D_f, and D_{FT} separately) can be used to quantify the microstructure of fat crystal networks. The fractal dimensions describe the combined effects of morphology and spatial distribution patterns of the crystal clusters in the fat crystal networks.

According to the fractal model developed for fat crystal networks by our group [1, 2, 11], the microstructure of the fat crystal networks is related to their rheological properties through the fractal dimension (D), as:

$$G' = \lambda \Phi^{\frac{1}{3-D}} \quad (1)$$

where G′ is the shear storage modulus of the fat samples, Φ is the volume fraction of solids (SFC/100) of the samples, D is the fractal dimension of the fat crystal networks, and λ is the pre-exponential factor, which depends on the size of

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the primary microstructural elements within the network and the nature of intermolecular forces.

When we determine the microscopy fractal dimensions of fat crystal networks from their polarized light micrographs, the change in the fractal dimensions normally results from the combined effects of several structural factors and it is difficult to predict how each individual structural factor affects the value of the fractal dimensions. Tang and Marangoni [12] studied how each microstructural factor individually affected D_b, D_f, and D_{FT} separately in 2D space through computer simulation. In this paper, we built on our previous simulation study and used computer simulation to show how each of the microstructural factors, such as crystal size, area fraction (AF) and distribution orderliness (randomly distribution or even distribution) of fat crystals affect the fractal dimensions of the fat crystal networks in 3D space. Here we also included an experimental study on fat crystal networks of a mixture of the High Melting Fraction of milk fat (HMF) and canola oil used as model fractal objects.

Materials and Methods

3D Box-Counting Fractal Dimension, D_b

To calculate the box-counting fractal dimension, D_b, in 3D space, a 3D image of a fat crystal network was divided into cubes with side length l_i. Any cube containing pixels belonging to the object was considered to be occupied. The number of occupied cubes, N_i, for side length, l_i, is counted. This process is repeated for cubes with different side lengths. The 3D box-counting fractal dimension, D_b, in 3D space, is calculated as the negative of the slope of the linear regression curve of the log–log plot of the number of occupied cubes, N_b, with the side length l_b:

$$D_b = -\frac{\Delta \ln N_b}{\Delta \ln l_b}. \quad (2)$$

3D Particle-Counting Fractal Dimension, D_f, and Mass Fractal Dimension, D_m

The concept of the particle-counting fractal dimension, D_f, is derived from the mass fractal dimension, D_m, where D_m relates the mass of a fractal object, M, to its linear size, R, as:

$$M = cR^{D_m}. \quad (3)$$

where c is a constant. If the particles making up the fat crystal networks can be assumed to be spheres, M is related to the average diameter, σ, of the particles as:

$$M = \frac{N\pi\sigma^3}{6}. \quad (4)$$

where N is the number of particles within the network. For fat crystal networks, if the average diameter of the particles of fat crystal networks can be assumed to remain constant [13], then the number of particles, N, within a fat crystal network is related to the linear size of the network, R, as:

$$N \sim R^{D_f}. \quad (5)$$

where D_f is the particle-counting fractal dimension. To calculate the value of D_f, in 3D space, the logarithm of the number of particles, log(N), in a sphere, is plotted against the radius of the sphere, log$R$, for varying values of R. The slope of the linear regression curve is the particle-counting fractal dimension, D_f. Similarly, the mass fractal dimension, D_m, is calculated from the slope of the linear regression curve between log(M) and log(R) for varying values of R.

To determine the fractal dimensions of 3D fat crystal networks, a new software, 3D-FD, was developed. Three fractal dimensions can be calculated using 3D-FD: Box-counting fractal dimension, D_b, particle-counting fractal dimension, D_f, and mass fractal dimension, D_m. The input of 3D-FD must be binary images.

Generation of 3D Images of Fat Crystal Networks

Each image of the simulated 3D fat crystal networks was stored in a 512×512×200 array. The size of the simulated fat crystal networks in X, Y, and Z direction is 512, 512, and 200 pixels separately. To generate a 3D fat crystal network with randomly distributed fat crystals, firstly, a 3D array containing 512 by 512 by 200 pixels was generated. Secondly, a 2D binary image with 512 by 512 pixels having randomly distributed fat crystals of radius r_i and area fraction AF_i was generated and copied into the 3D array. The same image was continuously copied into that 3D array for r_i times to create randomly distributed 3D fat crystals with radius of r_i on x–y direction and length of r_i on z direction. This process was repeated until the 3D array is full. The randomly distributed 3D fat crystals were generated again each time and the newly generated 3D fat crystals are separated from the previously fat crystals by a blank 2D plane. The simulated fat crystal networks with evenly distributed fat crystals were generated in the similar method except the 3D fat crystals are evenly distributed. Some images used in the 3D simulation images of fat crystal networks are shown in Fig. 1.

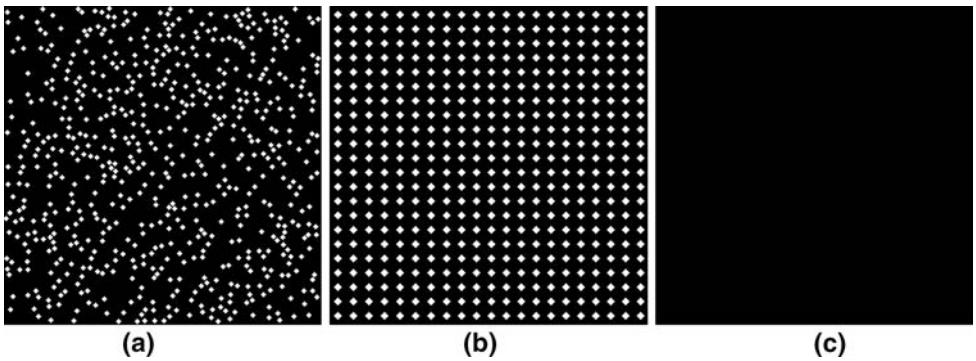


Fig. 1 **a** The effects of AF on D_b for different crystal sizes. r is the radius of fat crystals. **b** The effects of crystal size on D_b of fat crystal networks with evenly distributed crystals. AF is area fraction of crystals

3D Imaging of the High Melting Fraction of Milk Fat Samples

Mixtures of HMF and canola oil with a blending ratio ranged from 5 to 9% HMF (w/w) were prepared and their 3D images were taken as described in a study on the 3D fractal dimension of fat crystal networks [14]. 3D polarized light micrographs were taken using a light microscope (DMRXA2, Leica Microsystem, Wetzlar, Germany). A 3D polarized light micrograph of a fat crystal network was composed of 60 2D polarized light micrographs taken at different depths of the thick fat sample. The distance between two neighboring micrographs was 2 μm.

Calculation of 3D D_b, D_f, and D_m

D_b, D_f, and D_m of the simulated and HMF fat crystal networks are calculated using 3D-FD (TruSoft Int'l Inc, St Petersburg, FL, USA). The settings of the 3D-FD for the calculation of D_b, D_f, and D_m are:

1. For the calculation of Box-counting fractal dimension, D_b, the angle increment is 15°, the start cell size is 100 pixels, and the cell size decrease rate is 1.3.
2. For the calculation of particle-counting fractal dimension, D_f, the coordinates of the origin is (256, 256, 100), the starting radius is 256, the decrease ratio is 1.2, and the counting of the particles is based on 12 neighbors.
3. For the calculation of mass fractal dimension, D_m, all the settings are the same as that for the calculation of D_f.

Results and Discussion

Figure 2 shows the D_b of the simulated fat crystal networks with different AF and crystal sizes. The D_b of 3D fat crystal networks increased with AF and crystal size. The statistical analysis using SAS (SAS Institute Inc, Cary, NC, USA) showed that there is a significant interaction between AF and crystal size (data are not shown). The variance ir D_b became smaller at larger AF and/or large crystal sizes. In Fig. 2a we can appreciate that D_b increases with increasing AF, and that the curve of D_b becomes more and more flat at larger crystal sizes. In Fig. 2b, the slope of D_b curve as a function of crystal radius also became smaller at larger AF. This implies that there is an interaction between AF and crystal size: the effects of crystal size became less significant at large AF and the effects of AF also became less significant at larger crystal sizes. The effect of distribution order on D_b was studied by comparing the D_b values of fat crystal networks with evenly

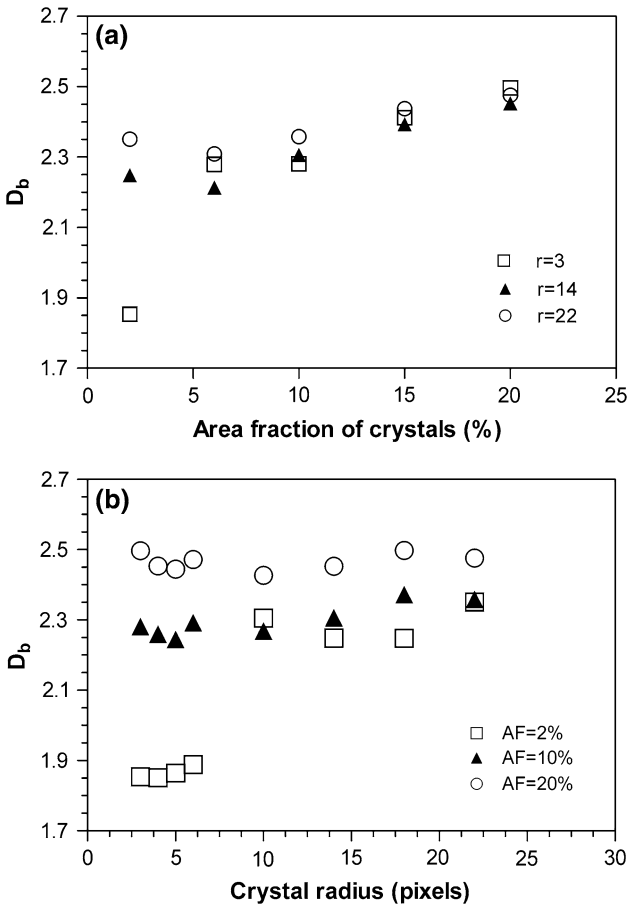


Fig. 2 Images used in the 3D simulation image of fat crystal networks; **a** area fraction = 10%, radius = 4 pixels, completely randomly distributed crystals; **b** area fraction = 15%, radius = 6 pixels, completely evenly distributed crystals; **c** a blank image used to separate different layers of fat crystals in the z dimension

Table 1 D_f values of simulated 3D fat crystal networks with a diamond shape and evenly distributed fat crystals, but different crystal AF and sizes

Radius of crystals (pixel)	AF = 2%	AF = 6%	AF = 10%	AF = 15%	AF = 20%
3	3.121	2.858	2.841	2.887	2.877
4	2.704	2.835	2.851	2.833	2.848
5	2.641	2.770	2.356	2.856	2.919
6	3.100	2.685	2.796	2.862	2.618
10	2.722	2.726	2.089	2.936	2.921
14	3.175	2.924	2.806	2.712	2.967
18	3.037	2.215	2.903	2.725	2.480

and randomly distributed crystals. No significant difference ($P < 0.05$) between the D_b values of these two types of fat crystal networks was found, thus the effect of distribution order on D_b was not significant. This suggests that D_b may not be suitable to quantify the microstructure of fat crystal networks at high SFCs since the value of D_b has been shown to not vary much at high AF in the simulated images.

The same study on the effects of AF, crystal size, and distribution order was carried out on D_f. AF and crystal size had significant effects on D_f ($P < 0.01$), but no clear trend was found for those two microstructural features. Distribution order had no significant effect ($P < 0.05$) on D_f. The results for D_f of evenly distributed fat crystal networks are shown in Table 1. Results for D_m showed the same trends as for D_f. In the simulation study in 2D [12], D_f (and thus D_m) was shown to be sensitive to the radial distribution pattern of fat crystals within fat crystal networks. It is much harder to generate fat crystal networks with different radial distribution patterns in 3D than that in 2D, and thus the effects of radial distribution pattern were not studied in this 3D simulation. But from their calculation algorithms, we predict that D_f and D_m in 3D must also be sensitive to the radial distribution pattern of crystalline mass.

Although it is difficult to show the effects of each individual microstructural factor on their 3D fractal dimensions, the effects of SFC on 3D fractal dimensions of HMF samples are shown in Table 2. Table 2 shows that as SFC increases from 5 to 9%, the 3D D_b of HMF samples increased from 1.87 to 2.12; while 3D D_f and D_m of HMF samples remain much more constant among different SFCs. This is in consistent with results from the computer simulation study, which showed that D_b increased with the area fraction of fat crystals, and D_f and D_m did not change much as a function of area fraction of fat crystals.

Table 2 3D box-counting fractal dimension (D_b), particle-counting fractal dimension (D_f), and mass fractal dimension (D_m) of HMF fat samples

SFC (%)	D_b in 3D[a]	D_f in 3D[a]	D_m in 3D[a]
5	1.87 (SD = 0.26)	2.71 (SD = 0.19)	2.61 (SD = 0.08)
6	2.09 (SD = 0.24)	2.62 (SD = 0.002)	2.99 (SD = 0.02)
7	2.07 (SD = 0.1)	2.83 (SD = 0.22)	2.82 (SD = 0.17)
8	2.14 (SD = 0.06)	2.78 (SD = 0.06)	2.68 (SD = 0.14)
9	2.12 (SD = 0.08)	2.72 (SD = 0.24)	2.92 (SD = 0.08)
Average	2.05	2.73	2.80

SD is the standard deviation among the replicates

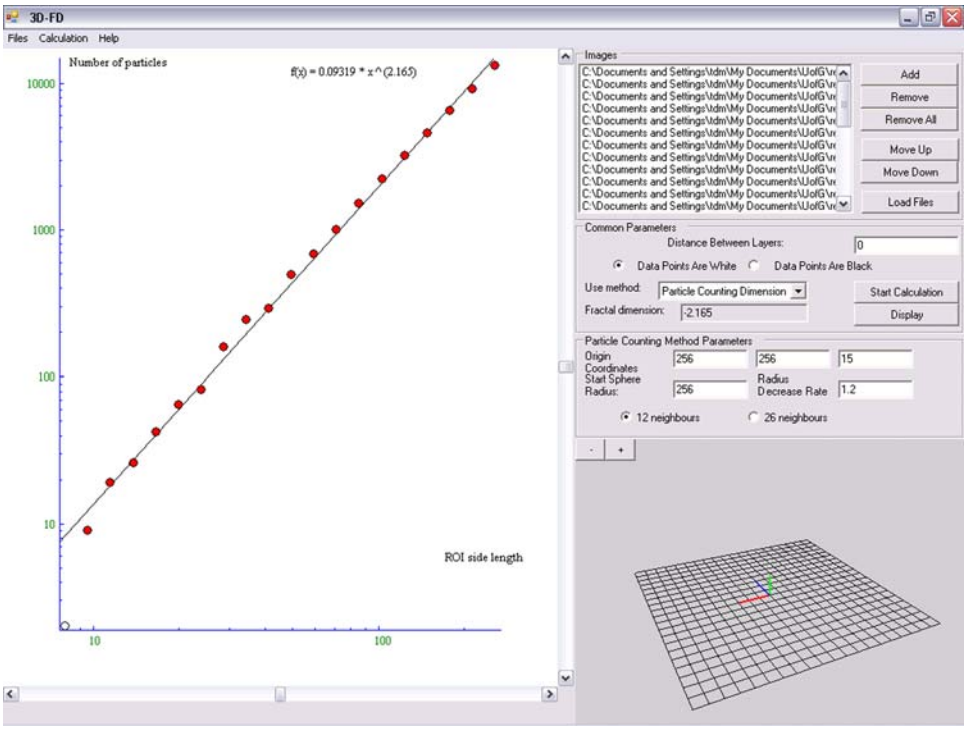
[a] The value is an average of three replicates

According to our previous study [14], it is not correct to estimate the fractal dimension of fat crystal networks in 3D space by adding one to their 2D fractal dimension. The 3D fractal dimension of a fat crystal networks used to model the rheological properties of fat samples must be obtained from image analysis of the 3D image of the fat samples. However, even doing this, there always is some doubt about the exact physical meaning of the fractal dimensions in fat crystal networks. This work demonstrated the microstructural basis of the fractal dimensions of fat crystal networks in 3D space for the first time. However, while the simulation and experimental studies revealed the microstructural basis of the fractal dimensions of fat crystal networks to some extent, those results are based on the simulated 3D images and some specific methods to calculate the fractal dimension of fat crystal networks. So, more experimental study on real fat samples, other than the high melting fraction of milk fat crystal networks, is necessary to confirm the results obtained and care must be exercised when extrapolating the results obtained to other fractal dimensions calculated by methods other than D_b, D_f, and D_m.

In conclusion, the 3D D_b, D_f, and D_m of simulated and HMF fat crystal networks showed similar results to those in our previous 2D study. D_b is sensitive to AF and crystal size, and was not affected by distribution order. There is an interaction effect of AF and crystal size on D_b. On the other hand, D_f and D_m were not affected by either AF, crystal size and the randomness in the distribution of the fat crystals.

Appendix

The fractal dimensions of fat crystal networks calculated by microscopy methods such as box-counting, D_b, particle-counting, D_f, and mass fractal dimension, D_m, have been extensively employed to quantify the microstructure of colloidal fat crystal networks. This work revealed the microstructural basis of D_b, D_f, and D_m in 3D space.



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