Fractal Structure Analysis of Some Food Materials

Toru Suzuki and Toshimasa Yano*

*Department of Food Technology and Engineering, Tokyo University of Fisheries,
Konan 4, Minato-ku, Tokyo 108, Japan
*Department of Agriculture Chemistry, Faculty of Agriculture,
University of Tokyo, Yayoi 1, Bunkyo-ku, Tokyo 113, Japan*

By applying the fractal concept, the relationship between particle diameter d_p and specific surface area S_w measured through an N_2 adsorption isotherm was investigated by using the equation $S_w \propto d_p^{D_s - 3}$, where D_s is the fractal dimension to be in the range between 2 and 3. The surfaces of porous bodies of rice flour, wheat flour, corn fiber and milk powders were fractal, with D_s values between 2.26 and 3.0. The effect of defatting was also investigated.

The physical properties of foods are a reflection of both their chemical composition and physical structure, but the relationship between the physical properties and the structure has remained obscure until now because of the lack of a quantitative method to evaluate the irregular structure of food materials. Recently, however, the quantitative evaluation of irregular forms and irregular phenomena has become possible by using the fractal concept proposed by Mandelbrot.[1] The fractal concept is based on 'self-similarity' and introduces a noninteger dimension, called the fractal dimension, to expand on the integer dimension. It has already been recognized that the forms of a tree, the branching structures of a vein,[1] the conformation of proteins,[2] *etc.*, are fractal.

By applying the fractal concept, Pfeifer *et al.*[3−6] have generalized the dependence of specific surface area S_w on particle diameter d_p and on cross-sectional area σ of the molecules to be adsorbed on the surface as follows:

$$S_w \propto d_p^{D_s - 3} \tag{1}$$

$$S_w \propto \sigma^{(2 - D_\sigma)/2} \tag{2}$$

where D_s and D_σ are the fractal dimensions defined from the viewpoints of the measure and coarse graining,[1,7] respectively.

For two-dimensional surfaces such as smooth spheres and cubes, $D_s = D_\sigma = 2.0$, agreeing with the commonly accepted relationships of $S_w \propto d_p^{-1}$ and $S_w \propto \sigma^0$. According to the fractal concept, however, D_s and D_σ may take any noninteger real value between 2.0 and 3.0,[3] for the surface in an irregular porous body, and for an irregularly structured surface.

For food materials, Peleg *et al.*[8] have characterized the degree of attrition of instant coffee particles by using scanning electron microscopy. They claimed that the ruggedness of the outer curve of particle silhouettes was fractal with a fractal dimension of 1.05 to 1.2, which is greater than that of common line 1.0. Since the irregular structure of food materials has not been studied quantitatively so far, a fractal structure analysis is attempted in this paper mainly to examine the applicability of Eq. (1). In this connection, in the early stage of this study, the authors noticed that the three well-known conflicting laws of the energy requirement for the comminution of solids —known as Rittinger's, Kick's, and Bond's equations— could be interpreted without conflict, in that the energy requirement was proportional to the increment of surface area if the surface structure of the solid particles was fractal with a D_s value of between 2.0 and 3.0.[9] A fractal analysis has also been attempted on dried potato starch gels by one of the present authors.[10]

In this paper, the fractal analysis is applied to rice flour, wheat flour, corn fiber, and milk powder. The effect of defatting is also studied.

Materials and Methods

Materials. Commercially available and polished "uruchi" rice grains were crushed and sifted into seven fractions smaller than 1 mm in diameter. During crushing, the materials were cooled to a temperature below about 40°C to avoid starch gelatinization by the heat evolved. A strong wheat flour from Nisshin Flour Milling Co., corn fiber from Nihon Shokuhin Kako Co. (60% hemicellulose and 25% cellulose), and four kinds of commercially available milk powders were sifted into six fractions.

The mean particle diameter of each fraction was taken to be the median between the two mesh sizes of JIS criteria.

Defatting. To defat the rice flour, half of each fraction was mixed with ethyl ether of 3-fold weight at room temperature, and the mixture stirred for 3 min and filtered. This procedure was repeated three times, and the defatted flour was dried at room temperature. To defat the wheat flour and corn fiber, a mixture of methanol–chloroform (1 : 2) was used instead of ethyl ether, the other procedures being the same as for the rice flour.

Chemical analyses. The Kjeldahl method was used for total nitrogen determination. Soxhlet extraction with a methanol–chloroform (1 : 2) mixture at 70°C was used for lipid determination.

Specific surface area. The nitrogen adsorption isotherm was measured with an Accusorb 2100 (Micromeritics Co.), from which the specific surface area of a sample was calculated by the B.E.T. method.[11] For the milk powders, however, Kr was mainly used instead of N_2 to measure the adsorption isotherm, in order to increase the precision of measurement for the small amount of the samples available. The purity of nitrogen gas used was guaranteed to be 99.9995%, and that of Kr to be 99.9%.

Results

The lipid contents before and after defatting, respectively, were 0.60% and 0.04% for rice flour, 6.39% and 1.08% for wheat flour, and 2.83% and 0.53% for corn fiber. The total nitrogen contents before and after defatting, respectively, were 1.29% and 1.26% for rice flour, for 2.24% and 1.95% wheat flour, and 1.13% and 0.92% for corn fiber. The contents of lipid and total nitrogen were independent
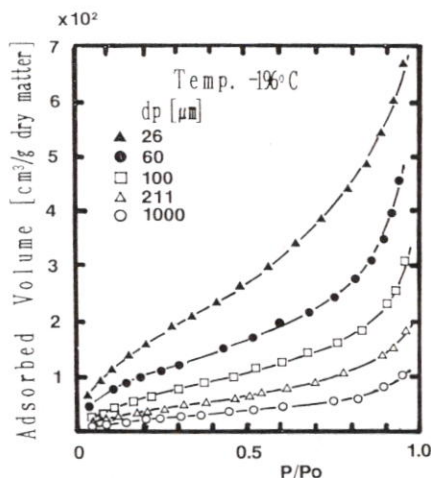

Fig. 1. Adsorption Isotherm of N_2 Gas on Rice Flour Particles.

P/P_0, relative pressure of nitrogen.

of particle size.

Figure 1 shows the N_2 adsorption isotherms of rice flour as an example, where P and P_0 on the abscissa are the nitrogen gas pressure at adsorption equilibrium and the saturation nitrogen gas pressure at the temperature of adsorption, respectively. The B.E.T. plots of the adsorption isotherms show linearity, although the figures are not shown to avoid redundancy.

Figure 2 shows log–log plots of specific surface area S_w vs. mean particle diameter d_p for rice flour, wheat flour, and corn fiber, before defatting. The dotted line in Fig. 2 shows a calculation of $D_s = 2.0$ in Eq. (1) for comparison. From the observed linear relationships in Fig. 2, the value of D_s for the rice flour and corn fiber was calculated by using the least square method to be 2.29 and 2.48, respectively. For the wheat flour, $D_s = 2.42$ for $d_p < 100\,\mu m$, and $D_s = 3.0$ for $d_p > 100\,\mu m$.

Figures 3 and 4 show log–log plots of S_w vs. d_p for three kinds of flours after defatting and for four kinds of milk powders, respectively. The fractal dimension calculated for each material is shown in Table I. In Fig. 4, the specific surface area measured with N_2 and Kr is also compared. Although the specific area recognized by N_2 and Kr molecules was very
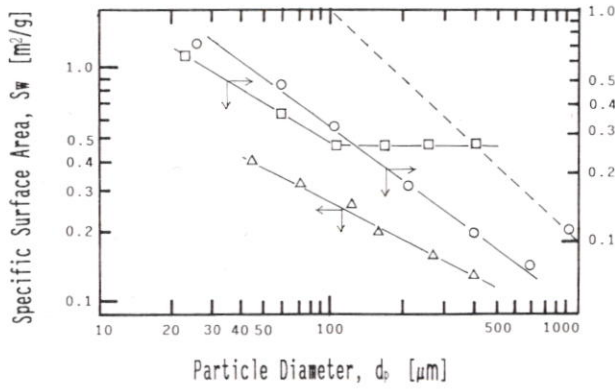
Fig. 2. Specific Surface Area *vs.* Particle Diameter.
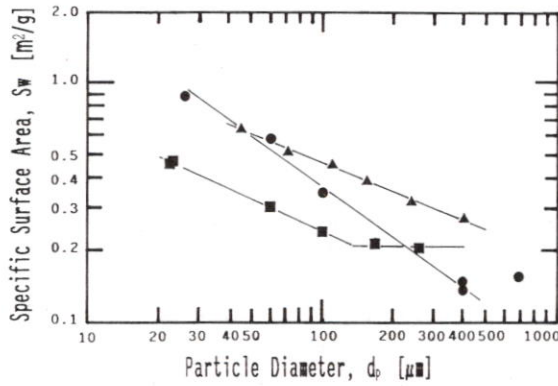○, rice flour; □, wheat flour; △, corn fiber.

Fig. 3. Specific Surface Area *vs.* Particle Diameter.
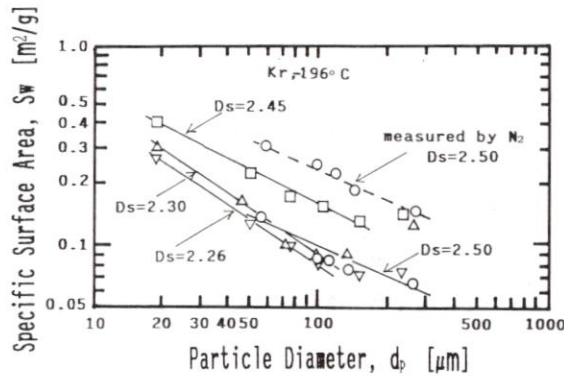●, defatted rice flour; ■, defatted wheat flour; ▲, defatted corn fiber.

Fig. 4. Specific Surface Area Measured with Kr at −196°C *vs.* Particle Diameter for Milk Powders.
○, Morinaga Milk's product; △, Snow Brand Milk's product; ▽, Wakoudo's product; □, Meiji Milk's product.

Table I. Fractal Dimensions of Some Food Materials before and after Defatting

Sample	D_s	Range of d_p [μm]
Rice flour		
raw	2.29	<700
defatted	2.29	<400
Wheat flour		
raw	2.43	<100
	3.0	>100
defatted	2.55	<130
	3.0	>130
Corn fiber		
raw	2.48	<500
defatted	2.61	<500

different, the fractal dimension of the surface was not significantly different.

The milk powder with the largest fractal dimension was the quickest to disperse in water.

Discussion

As shown in Figs. 2 through 4, all the experimental results showed that the relationship between the specific surface area and the particle diameter of the materials used was not as commonly expected to be $S_w \propto d_p^{-1}$, but agreed with Eq. (1) with a D_s value from 2.26 to 3.0. This means that the irregular porous structure of such food materials is not the same in nature as that of geometrically regular bodies such as spheres, cylinders, or cubes. The porous structure of food materials is developed within three-dimensional space with self-similarity, the degree of which could better be evaluated by fractal dimension D_s. $D_s = 3.0$ is a limiting case for the operational development of a porous structure, because the specific surface area becomes constant for any part within the particle. The porous structure of silica gel and activated carbon was also of a D_s value near 3.0.[5]

The difference of D_s values in different ranges of d_p, as shown in Figs. 2 and 3, and in Table I, suggests that the porous structure was developed by two different mechanisms in a different range of d_p.

By defatting, the specific surface area of rice flour and corn fiber was increased, but that of wheat flour was decreased. The decrease of the specific area by defatting might have been caused by a possible collapse of the pores during processing. As Fig. 3 and Table I show, the porous structures left after defatting were also fractal. This suggests that the fat was also distributed as fractal within the flour paticles and corn fiber. The D_s values for the defatted materials are not necessarily the same as those for the materials before defatting, although the difference in D_s before and after defatting was not large as shown in Table I.

It is interesting that, as shown in Fig. 4, the specific surface area was differently recognized by N_2 and Kr molecules. This is contrary to common sense, in which the surface is implicitly assumed to be smooth. From a fractal viewpoint, however, the surface may not be smooth and, for such an irregular surface, the surface area may be differently recognized by differently sized molecules. An experimental examination of Eq. (2) for such a fractal surface will be investigated in more detail in a forth coming paper.

Milk powders from different makers were differently characterized in their porous structure as shown in Fig. 4, which will be a reflection of different their production method. The relationship between the function of milk powders including their dispersion in water and the production method could be better understood if a fractal analysis was applied to the porous structure of milk powders. In general, by recognizing the fractal nature of a porous structure, the physical properties of food materials such as the adsorption equilibrium, diffusivity and texture may be better understood in future.

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