A Fractal Model of Tensile Fracture Surfaces for Particulate-Filled Polymer Composites

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ABSTRACT: Tensile mechanical properties are important for the characterization of the utilization performances of materials. Inorganic particles are usually used to reinforce and toughen resins and to reduce costs in the polymer industry. It is, therefore, very important to quantitatively appraise the break way and mechanisms and the reinforcing or toughening effect of the fracture surface of tensile specimens. A tensile fracture surface fractal model of particulate-filled polymer composites on the basis of fractal theory is established and a corresponding equation for the tensile fracture surface fractal dimension is proposed in this article. Tensile fracture surface fractal dimensions of particulate-filled polymer composites at filler volume fractions of 2–30% were estimated by means of this equation, and the calculations were compared with the measuring values of calcium carbonate filled acrylonitrile–butadiene–styrene (ABS) resin and titanium dioxide filled ABS resin composites reported in the literature. The results show that the theoretic estimations and the measuring values were roughly close to each other. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 3763–3767, 2008

Key words: composites; fillers; fracture

INTRODUCTION

In the polymer industry, inorganic particles are usually used to reinforce and toughen resins and to reduce materials cost. On the other hand, particulate filling directly affects the processing properties, such as viscosity, die swell, and flow stability, of these polymer composites. The rheological behavior and processing properties of particulate-filled polymer composites have been extensively studied by rheologists and polymer processing engineers during the past 10 years.[1–4]

Tensile mechanical properties are an important characteristic in the utilization performances of materials. For particulate-filled polymer composites, the effects of the shape, content, and particle size of filler particles and their dispersion in the matrix on the mechanical properties and some other important behaviors, such as tensile strength, stiffness, impact toughness, and brittle–ductile transition, are quite significant, in addition to the interfacial adhesion between the matrix and the filler particles.[5–9] Furthermore, the mechanical properties of particulate-filled polymer composite depends considerably on the interfacial morphology between the inclusions and the resin matrix.[10]

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Generally, the brittle or ductile behavior of particulate-filled composites depends, to a great extent, on the interfacial structure and morphology between the inclusions and the matrix, and the strength of the materials and fracture type are closely related to the fracture surface morphology during the tensile testing of specimens. It is, therefore, very important to quantitatively appraise the tensile fracture surface of the specimens to investigate the break mechanisms and the reinforcing or toughening mechanisms more deeply. It is well known that when a piece of specimen is fractured by tensile loading, the fracture surface that is formed is rough and irregular. To quantitatively characterize the irregularity of the fracture surfaces, many statistical parameters, such as the maximum individual peak to valley, average roughness, and mean peak to valley, have been proposed.[11,12] However, the values of the parameters mentioned previously depended strongly on the sampling length, appraising length, and sampling interval of the experiment. So, it is very necessary to find a new parameter that is independent of the sampling length, appraising length, and sampling interval to characterize it. Mandelbrot[13] used fractal geometry to describe a great variety of natural structures that are irregular, rough, or fragmented by a number called the *fractal dimension*, which can range from 2, when the surface is smooth, up to 3. It was used first to characterize the irregularity of the fracture surfaces of metals in 1984.[14] Then, the fractal dimension was applied to characterize all kinds of engineering surfaces and fracture surfaces of polymer materials. At the same time, many methods used to measure and calculate the fractal dimension of different kinds of surfaces have been developed, mainly including slit island analysis,[14] the reticular cell counting method,[15] the power spectrum method,[16] the variation method,[17] the yard-stick method,[18] the structure function method,[19] and the two-dimensional variation method.[20,21] Among the first six methods, the value of the fractal dimension measured ranges from 1 to 2. One can obtain the value of the fractal dimension range from 2 to 3 only by the two-dimensional method.

Wang et al.[22] appraised the fractal characterization and established a fractal model for a machined surface by the Weierstrass–Mandelbrot fractal function, and the fractal model was used to simulate the two-dimensional profile of a machined surface. Rajendra[23] established a fractal model to describe the surface roughness by the Weierstrass–Mandelbrot function also, and the fractal dimension predicted by the fractal model ranges from 1 to 2. The fractal models mentioned previously can only describe the two-dimensional profile of a surface, so it is necessary to establish a fractal model to describe the three-dimensional morphology of a rough surface. The objective in this study was to establish a fractal model of a tensile fracture surface of a particulate-filled polymer composite on the basis of the principles of generating a fractal surface and the definition of the Hausdorff dimension of a fractal in fractal theory first, to investigate deeply the tensile fracture mechanisms and the fracture patterns of a particulate-filled polymer composite, and then to quantitatively characterize the tensile fracture surface morphology by means of the calculations of these fractal dimensions.

FRACTAL MODEL

Assumption conditions

Filler particles might be dispersed uniformly in a polymer matrix by means of suitable processing technologies. For example, Li et al.[1] observed the fracture surface of glass-bead (GB)-filled low-density polyethylene composites and found that the particles dispersed uniformly in the matrix, as shown in Figure 1. That is, the layer (or levels) of the inclusions in the polymer matrix may be statistically uniform. Therefore, the particles are assumed to be uniform in the polymer matrix in this article. Second, different kinds of fillers have different shapes and sizes, for example, spheroid, cube, schistose, and acicular. In general, spherical particles are a common filler in particulate-filled polymer composites. In this article, the particles are assumed to be spherical in the model for analysis simplification. This is also shown

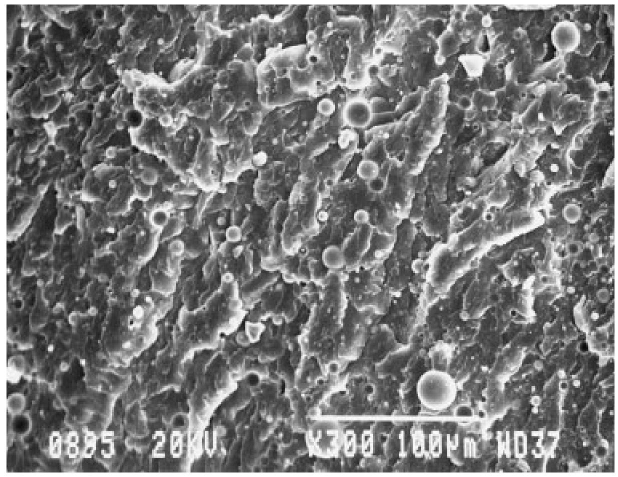

Figure 1 SEM photograph of the fracture surface of the low-density polyethylene/GB system.

in Figure 1. During the tension of the particulate-filled polymer composites, the interfacial adhesion strength between the particles and the matrix is usually somewhat lower than that of the polymer. Consequently, the first failure of particulate-filled polymer composites under a tensile load will usually happen in the interface between the inclusions and the matrix. Furthermore, the interfacial debonding between the filler and polymer matrix might develop along the equatorial circle of the particle;[24] the interfacial adhesion strength between the particle and the polymer matrix is different for different fillers and their surface treatments. Therefore, the tensile fracture surface of a particulate-filled polymer composite becomes rough, the degree of roughness of the tensile fracture surface changes with the variation of the volume fraction of the inclusions, and the morphological structure of tensile fracture surface is hierarchical (or stratiform).[5,6]

Modeling

On the basis of the assumed conditions, a new fractal model describing the fractal characterization of the tensile fracture surfaces of particulate-filled polymer composites was developed by the principles of generating a fractal surface and the definition of the Hausdorff dimension of a fractal in fractal theory[13] as described in the following text.

The first structure layer (or stratum) is constructed by the division of a square plane with a specified area of tensile fracture surface for particulate-filled polymer composites into 100 little square ones with equal area; the side length of each little square is 1 unit, as shown in Figure 2. The stratum generated in this layer is called the *original stratum.*

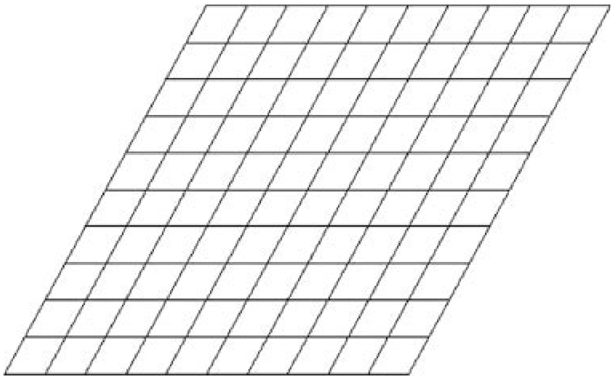

Figure 2 Original layer of the established fractal model.

The second structure layer is formed when $(2k)^2$ little squares in the middle of the original stratum become small projecting terraces (or convexes) to form $(2k)^2 + 4(2k)k$ little squares, where k represents the number of layers (or levels) of the fracture surface. Here, $0 \leq (2k)^2 \leq 100$, so $0 \leq k \leq 5$. Then, these $(2k)^2 + 4(2k)k$ little squares combine with the original stratum (not projecting) to form a closed cube. The little squares upward of the original stratum are replaced by a little square plane and a hemispheric surface whose equatorial circle cuts the little square plane according to assumption that the particle is spheroid, as shown in Figure 3.

The third structure layer may be structured by the division of the last $100 - (2k)^2$ little squares with the previous steps (or ways). After n steps, the original plane will become an uneven and projecting fractal surface with limited structure layers.

DETERMINATION OF THE FRACTAL DIMENSION

The fractal dimension of the fractal model of the tensile fracture of a particulate-filled polymer composites is determined by the following method:[13]

$$N = r^{-D_s} \tag{1}$$

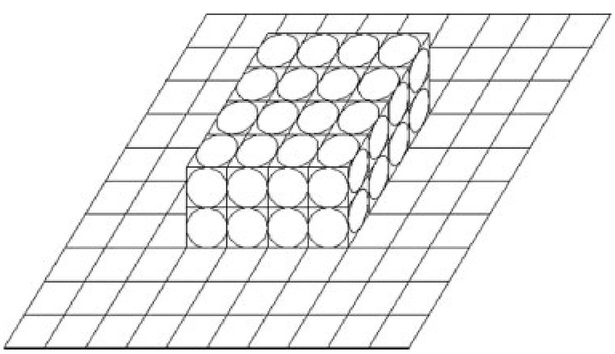

Figure 3 First layer (k = 2).

where r is the measuring size; D is the Hausdorff dimension of the fractal, which is a noninteger and varies between 2 and 3 when used to characterize the surface roughness; and N is the measuring value, which maybe a number, area, volume, or information quantity, and so on in different applications.

For the previous fractal model

$$r = \frac{1}{10} \tag{2}$$

$$N = 10^2 + 4 \times 2k \times k \tag{3}$$

where N is the number of little square planes in the second structure layer and k characterizes the fracture surface morphology and indicates the influence of the volume fraction of particles on the morphology of the tensile fracture surface (shown in Fig. 3) and $0 \leq k \leq 5$.

Then, the fractal dimension of the fractal model can be calculated with the following equation:

$$D_S = \frac{\lg N}{\lg\left(\frac{1}{r}\right)} = 2 + \lg\left(1 + \frac{2 \cdot k^2}{25}\right) \quad (0 \leq k \leq 5) \tag{4}$$

where D_s is the fractal dimension of the tensile fracture surface of particulate-filled polymer composites and ranges between 2 and 3.

An equation describing the relationship between the volume fraction (V_g) of the particles and k may be derived from Figure 3, that is

$$V_g = \frac{(2 \cdot k)^2 \cdot k \cdot \frac{4}{3} \cdot \pi \cdot \left(\frac{1}{2}\right)^3}{10^2 \cdot 5 \cdot (1)^3} = \frac{\pi \cdot k^3}{750} \quad (0 \leq k \leq 5) \tag{5}$$

Also

$$k = \sqrt[3]{\frac{750 \cdot V_g}{\pi}} \quad (0 \leq k \leq 5) \tag{6}$$

The fractal dimensions of the tensile fracture surfaces of particulate-filled polymer composites and the volume fractions of particles can be calculated by a combination of eqs. (4) and (5) and with the assumption that the value of π is 3.14159 at different k values; the results are as shown in Table I.

TABLE I
D_s and V_g Values Under Different k Values

	k					
	0	1	2	3	4	5
D_s	2.000	2.0334	2.1205	2.2355	2.3385	2.4771
V_g (%)	0.00	0.42	3.35	11.31	26.81	52.36

TABLE II
Theoretical Estimations and Measuring Values of the Fractal Dimensions of the Tensile Fracture Surfaces of the ABS/$CaCO_3$ Composites

	V_g (%)		
	2	11	18
Theoretic value	2.073	2.221	2.298
Measuring value	2.217	2.327	2.446
Relative error (%)	6	4	6

To more conveniently estimate the fractal dimension of the tensile fracture surface from the volume fraction of filler particles of a particulate-filled polymer composite, one can substitute k in eq. (6) into eq. (4) and assume that the value of π is 3.14159; then, one can get an theoretical equation describing the relationship between D_s and V_g:

$$D_s = 2 + \lg\left(1 + 3.07868\left(V_g\right)^{2/3}\right) \tag{7}$$

RESULTS AND DISCUSSION

Measurement of fractal dimensions

Liu et al.[20] measured the fractal dimensions of the tensile fracture surface of calcium carbonate ($CaCO_3$) filled acrylonitrile–butadiene–styrene (ABS) copolymer composites by a two-dimensional variation method. When the method was used to measure the fractal dimensions, SEM photos were taken of the fracture surface, and the gray photograph was taken first. Then, the gray photograph of the fracture surface was divided into many grids with size $l \times l$ (the l is defined as the length of the grid), and each grid was covered by box with size $r \times r \times h$, where r is the length and width (it is the measuring size also), the ratio of l to r is integral, and h is the height of the box proportional to the gray value of the covered part of the grid. At the same time, the number of boxes (N) with different h values was counted. In this case, the fractal dimensions of the tensile fracture surfaces of the polymer composite systems were calculated by means of eq. (4) from relevant experimental conditions. In this case, the volume fractions of the $CaCO_3$ particles were 2, 11, and 18%, respectively, and the results are shown in Table II. Similarly, Liu et al.[21] also measured the fractal dimensions of the tensile fracture surfaces of titanium dioxide (TiO_2) filled ABS resin composites by a two-dimensional variation method, and the results are shown in Table III, where the volume fractions of TiO_2 particles were 5, 10, 20, and 30%. The fractal dimensions of the tensile fracture surfaces for both the ABS/$CaCO_3$ and ABS/TiO_2 composites increased with increasing filler volume fraction. In addition, the study authors also investigated the relationship between the fractal dimension and the static friction coefficient of the composites.

Estimation of fractal dimensions

To preliminarily verify the reliability and applicability of eq. (7) in engineering, eq. (7) was used to calculate the theoretical values of the fractal dimensions of the tensile fracture surfaces for ABS/$CaCO_3$ with volume fractions of filler particles varying from 2 to 18%[20] and for ABS/TiO_2 composites with volume fractions of filler particles varying from 5 to 30%.[21] The results are also listed in Tables II and III, respectively.

Discussion

Table II indicates that the theoretical estimations of the fractal dimensions of the tensile fracture surfaces of the ABS/$CaCO_3$ composites increased with increasing filler volume fraction, and they were somewhat lower than the measuring values at the same volume fraction; the maximum relative error was 6%, and the minimum relative error was 4%. Similarly, Table III indicates that the theoretical estimations of the fractal dimensions of the tensile fracture surfaces of the ABS/TiO_2 composites increased with increasing filler volume fraction, and they were somewhat lower than the measuring values at the same volume fraction; the maximum relative error was 7%, and the minimum relative error was 4%. The relative errors may have resulted from the hypothesis that the dispersion of the inclusions in the polymer matrix was uniform. Although there were relative errors between the theoretical predictions and the measuring values, the relative error was much lower than 10%. This indicated that the fractal model and corresponding fractal dimension equation [eq. (7)] used to estimate the fractal dimensions of the tensile fracture surfaces of the particulate-filled polymer composites were reasonable, and the application in engineering was meaningful in a given range of filler concentration.

This fractal model and the corresponding fractal dimension equations of the tensile fracture surfaces for particulate-filled polymer composites were established on the basis of a limited volume fraction of filler particles. Therefore, these equations may be used in filled polymer composite systems with some reinforcing and toughening effects due to the inclusions, in

TABLE III
Theoretical Estimations and Measuring Values of the Fractal Dimensions of the Tensile Fracture Surfaces of the ABS/TiO_2 Composites

	V_g (%)			
	5	10	20	30
Theoretic value	2.128	2.207	2.314	2.354
Measuring value	2.295	2.348	2.421	2.532
Relative error (%)	7	6	4	7

addition to the previous assumptions. Liang and coworkers[5,6] investigated the effects of GBs on filled polypropylene composites; their results show that the mechanical properties increased with increasing filler volume fraction in the range from 0 to 30%.

CONCLUSIONS

A fractal model of the tensile fracture surfaces of particulate-filled polymer composites was established according to the iteration method and principles of generating a fractal surface, and a theoretical equation for describing the relationship between the fractal dimensions of the tensile fracture surfaces for particulate-filled polymer composites and the volume fraction of filler particles as well as the fracture surface morphological parameter was derived on the basis of the assumptions that the dispersion of the inclusions in the matrix was uniform and that there were some reinforcing and toughening effects for these composite systems.

The results show that the predictions of the fractal dimension of a tensile fracture surface by means of the mathematical model established in this article were roughly close to the measured values from both the ABS/TiO_2 and ABS/$CaCO_3$ composites reported in the literature, and the relative errors between them were from about 4 to 7% within a volume fraction range of filler particles from 2 to 30%. This indicates that the fractal model established in the article is suitable for application in scientific study and engineering.

The theoretical estimations of the fractal dimension of a tensile fracture surface were somewhat lower than the measuring values from the particulate-filled ABS systems, which might be attributed to the hypothesis that the dispersion of the inclusions in the polymer matrix was uniform and the interface being uneven due to debonding between the particles and the polymer matrix.

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