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Correlations and characterization of porous solids by fractal dimension and porosity

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Abstract

The fractal dimensions of zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A carbon sieve have been obtained by physical adsorption of different-sized adsorbates. The porosity of the solids is obtained from literature. Two simple equations with two integral variables can be employed to characterize a porous solid with finite fractal dimension and porosity. These two equations also illustrate the correlation between fractal dimension and porosity of the porous solid and are also helpful to construct the fractal structure of the porous solids. Two integral variables are the number of divisions (cuts) in each dimension, and the number of the d-dimensional objects (d can be 1, 2 or 3) that must be taken for iteration. Seven examples of porous solids are employed to characterize and illustrate the applicability of the two equations.

Keywords: Fractal correlations; Fractal dimension; Porosity; Porous solid; Visualized fractal structure

1. Introduction

In general, fractal geometry has been developed and has progressed rapidly in the past few years, Mandelbrot [1], Falconer [2], Ross [3] and Schroeder [4] to name a few. Fractals have two key elements of self-similarity and fractal dimension. The self-similarity has a reducing or enlarging factor called s. The universal relationship between s, D_f (fractal dimension), and N (the number of fractal units) is $N = 1/s^{D_f}$. Louis and Pereira [5] suggested the application of the concept of the Menger sponge to the porous catalysts which might be characterized as the structure effects of the catalysts. At the same time, the sponge or the swiss cheese possesses the void portion

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or porosity as well. Radoev and Tenchov [6] discussed the fractal effect on the adsorption rate of porous solids. Skeituch and Brandon [7] related the diffusion and the reaction to the Thiele modulus of the catalysts. The fractal structure of porous solids could be characterized by the physical adsorption (Sze and Lee [8], Pfeifer and Avnir [9] and Avnir [10]). Adsorbents and catalysts are mostly porous and possess a finite fractal structure and porosity. The influences of the fractal structure on adsorption, diffusion and catalytic reaction rate have been discussed by Radoev and Tenchor [6], and Sheituch and Brandon [7], etc. Nekka and Hildgen etc. [11] reported a correlation between the fractal dimension and the porosity of heterogeneous solids.

The fractal structure of a porous solid should possess a definite fractal dimension and porosity. If one assumes that the porous solids could be characterized by the fractal structure, then the physical effects on mass transfer, adsorption and catalytic reactions might be studied by computer simulations and would be as realistic as possible. In this study, adsorption and porosity data from literature on zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A were collected and correlated to characterize the fractal dimensions and porosity of these porous solids. Subsequently, two equations, capacity dimension and porosity equation, for construction of the fractal structure having the desired fractal dimension and porosity were employed and their legitimate applicability to the porous solids illustrated. Results of the porous solids were visualized and plotted out in illustrative graphs.

2. Theoretical background

2.1. A capacity dimension equation for fractal structure

In order to construct a porous fractal solid from a solid Euclidean solid, the solid is divided into N equal parts in each direction to form N^d small solids and then N_1 small solid objects are dug out. Next, the same operation of digging out will be iterated at each of the remaining objects. Repeating this procedure, a porous fractal solid with a desired fractal dimension can be finally obtained. Hence, the capacity dimension equation (Mandelbrot [1]) with two integral variables N and N_1 is represented in the following form,

$$D_f = \frac{\ln(N^d - N_1)}{\ln N}\,, \tag{1}$$

where N is the number of one-dimensional objects and N_1 can be the number of one ($d=1$), two ($d=2$) or three ($d=3$) dimensional objects corresponding to lines, areas or solids. The D_f is the resulting final fractal dimension of lines, areas or solids. For one-dimensional line, the example of a Cantor set [12] is considered. By taking out the middle one third of a line and iterating the same operation by taking $\frac{1}{3}$ on the remaining segments infinitely can create the Cantor set. Finally we end up with a dust of Cantor set points, their fractal dimension being 0.631 which is the same as the D_f calculated from Eq. (1) with $N=3$, $d=1$, and $N_1=1$. For two-dimensional area,

the well known Sierpinski carpet (Sierpinski [13,14]) of fractal dimension 1.893 was obtained by the Eq. (1) with $N=3$, $d=2$, and $N_1=1$. And finally for three-dimensional objects, employing Eq. (1) with $N=3$, $d=3$, and $N_1=7$ we can obtain the fractal dimension, 2.727, of the Menger sponge [15]. One should be aware of the fact that N_1 was the number of line segments, squares, cubes or rectangular columns, which was taken out.

2.2. Porosity equation for fractal structure

The following equation generally is used to define the porosity of the porous solid

$$\varepsilon_f = \frac{V_o}{V_T}\,, \tag{2}$$

where ε_f is the porosity, V_o is the pore volume of the porous solid and V_T is the total volume of the solid. The volume of the pores in fractal structure can be represented as

$$V_o = \left(1 - \left(\frac{N^d - N_1}{N^d}\right)^i\right) V_T\,, \tag{3}$$

where i represents the number of iteration. Therefore, the porosity of the fractal structure can be written as

$$\varepsilon_f = 1 - \left(\frac{N^d - N_1}{N^d}\right)^i\,. \tag{4}$$

It should be mentioned that N and N_1 in Eqs. (1) and (4) are the same and i, the number of iterations, has to be finite. The iteration has to be stopped at some point where i is an intrinsic property of the porous solids. Consequently, the fractal structure of a porous solid is not really a true fractal in the strict sense of fractal geometry. We may call it pseudo-fractal or precursor of fractal. In this study we do not consider i as a parameter but as a unique property belonging to the specified porous solid. Furthermore, Eq. (1) may also be considered as implicitly containing i, a pseudo-fractal cut out at the ith generation. Otherwise, as i tends to infinity, the solid becomes a fractal dust and there is no counter part in the real world.

By eliminating N_1 from Eqs. (1) and (4), we obtain

$$1 - \varepsilon_f = N^{(D_f - d)i}\,. \tag{5}$$

The Eq. (5) illustrates the correlation between the fractal dimension and the porosity of a porous solid. Rearranging Eq. (5), we can obtain

$$\log(1 - \varepsilon_f) = i(D_f - d)\log N\,, \tag{6}$$

which is the same equation obtained in the paper of Hildgen et al. [11], where $N^{-i} = \varepsilon$ (Hildgen's) and $1 - \varepsilon_f = p$ (Hildgen's); the only difference is i which is implicit in their iteration statement. Porosity and fractal dimension are the essential elements to characterize the fractal structure of the solid. Hence, if the porosity and the fractal dimension of the porous solid are obtained experimentally, Eqs. (1) and (4) can be

employed to construct the fractal structure of the porous solid by the determination of N and N_1.

2.3. Determination of N and N_1

In this study, we determine N and N_1 to construct the fractal structure of the porous solid with the aid of the above equations. The following is the procedure of determining N and N_1.

(i) Specifying i and employing Eq. (5) with the desired fractal dimension and porosity to roughly estimate N.

(ii) Given N, employing Eq. (1) with fractal dimension to roughly estimate N_1.

(iii) Given N and N_1, constructing the fractal structure, and judging whether the fractal structure is reasonable for the characterization of the porous solid. If the fractal structure with N and N_1 can neither be constructed nor be reasonable for the characterization of the solid structure, the procedure might return to step (i), (ii) or (iii) by slightly adjusting i, N, or N_1 until the appropriate fractal structure is obtained.

In the following section, seven examples of zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A carbon sieve will illustrate the legitimate applicability of the two equations.

3. Results and discussion

The fractal dimension of the porous solids can be characterized by the physical adsorption of the adsorbates with different sizes of sorbates, as reported by Sze and Lee [8] and others [9,10]. The fractal dimensions of zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A carbon sieve were obtained. Figs. 1–7 display the plots of the log (adsorbed amount) vs. log (molar volume) of the seven porous objects, zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A carbon sieve, respectively. The correlation coefficients range from 90% to 96%. And the fractal dimensions of zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, ZSM-5 and MSC-5A carbon sieve are 2.57, 2.37, 2.92, 2.86, 2.45, 2.60 and 2.38, respectively. The data sources used for the correlation are also listed under the caption of the figures. The above procedure can be employed to construct the fractal structure of the six porous solids: zeolite A, zeolite X, Dowex MSC-1, Mordenite, zeolite Y, and ZSM-5. Fig. 8 displays the visualized fractal structure of zeolite A. The cube is first divided into five equal parts in each direction. By taking out the middle slab in each direction, the fractal structure of zeolite A can be obtained, as demonstrated with $N = 5$ and $N_1 = 61$ in Fig. 8. A fractal dimension of 2.58 is obtained. The porosity of the fractal structure with one iteration from Eq. (4) is 0.488. The errors of the fractal dimension and the porosity are 0.39% and 3.6%, respectively. Fig. 9 displays the visualized fractal structure of zeolite X with one iteration. The fractal structure can be obtained by taking out the middle $\frac{1}{4}$ unit in each

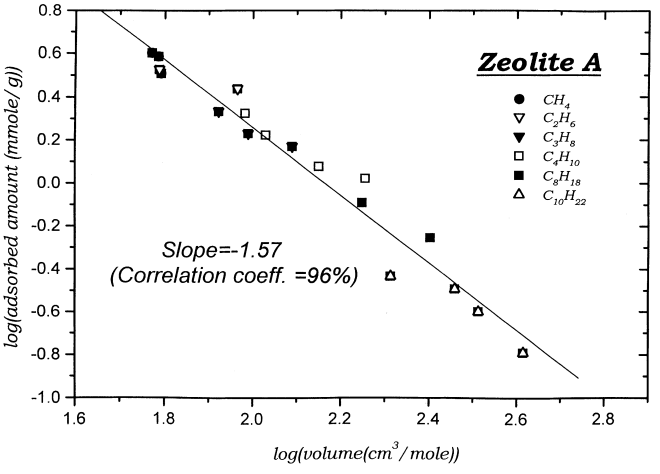

Fig. 1. Adsorption capacity vs. adsorbate molar volume: Data source: Danner and Wenzel [16], Derrah et al. [17], Haber et al. [18], Kual [19], Lederman and Williams [20], Loughlin and Ruthven [21], Miller [22], Ruthven and Loughlin [23], Ruthven [24], Sorial et al. [25], Valitis et al. [26], Verelst and Baron [27], Zuech et al. [28], Mcclellan and Harnsberger [53], and Reid et al. [54].

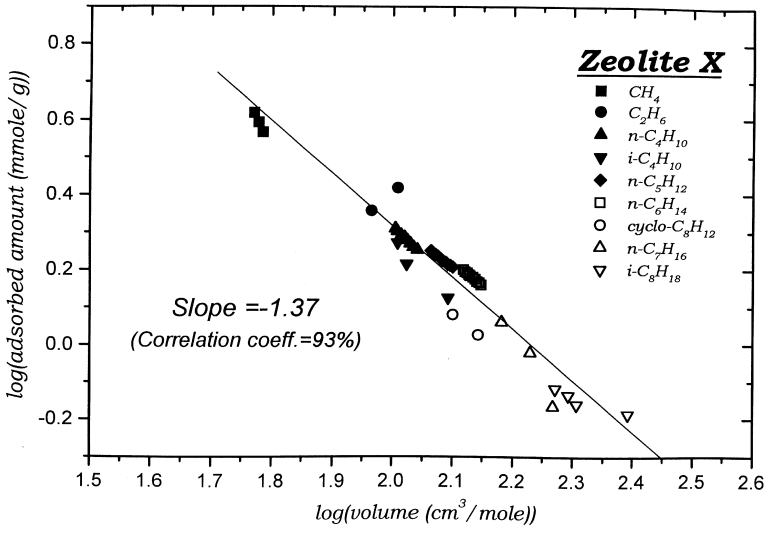

Fig. 2. Adsorption capacity vs. adsorbate molar volume: Data source: Barrer et al. [29], Danner and Choi [30], Danner et al. [31], Hyun and Danner [32], Ruthven [33], Ruthven and Francis [34], Wukasugi et al. [35], Youngquit et al. [36], Mcclellan and Harnsberger [53], and Reid et al. [54].

direction in the cube. The fractal dimension is 2.38 by employing Eq. (1) with $N = 4$ and $N_1 = 37$. The porosity of the fractal structure is 0.58 from Eq. (4). The errors of the fractal dimension and porosity are 0.42% and 13.8%, respectively. In the case of Dowex MSC-1, the cube is divided into eight equal parts in each direction and then

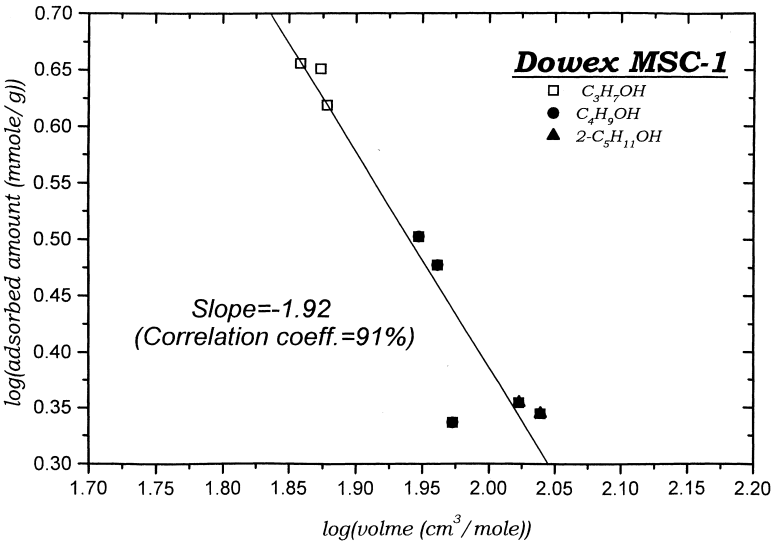

Fig. 3. Adsorption capacity vs. adsorbate molar volume: Data source: Chien [37], Mcclellan and Harnsberger [53], and Reid et al. [54].

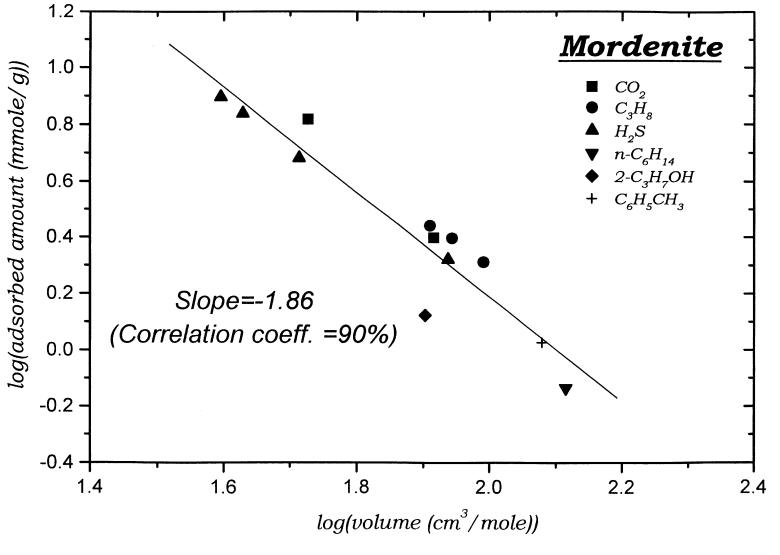

Fig. 4. Adsorption capacity vs. adsorbate molar volume: Data source: Valenzuela and Mylers [38], Talu and Zwiebel [39], Dyer and Singh [40], and Kofke et al. [41].

80 objects are taken out, as demonstrated in Fig. 10. Similarly, with $N=8$ and $N_1=80$ one can obtain the fractal dimension of 2.92 and the porosity of 0.288 in the second iteration. The errors of the fractal dimension are very small and for the porosity they are 4.2%. In the case of Mordenit it is a two-dimensional opening in two directions.

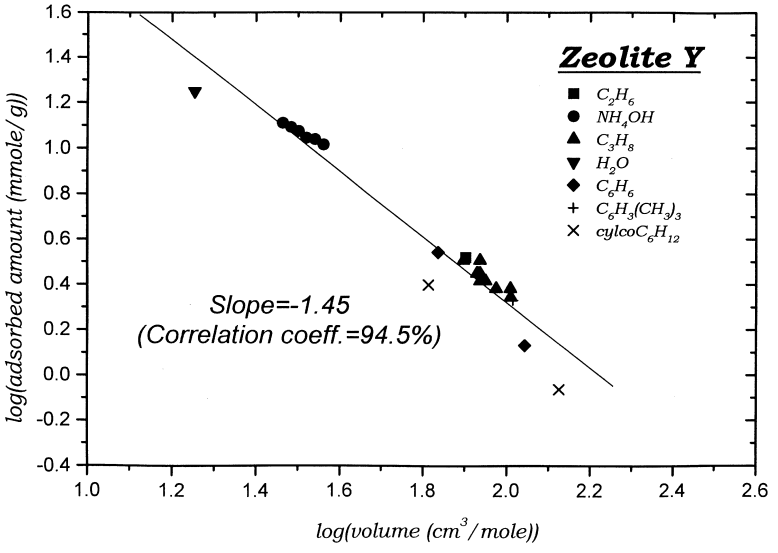

Fig. 5. Adsorption capacity vs. adsorbate molar volume: Data source: Bezus et al. [42], Shiralkar and Kulkarni [43], Dzhigit et al. [44], Chandwadkra and Kulkarni [45], and Barthomeuf and Ha [46].

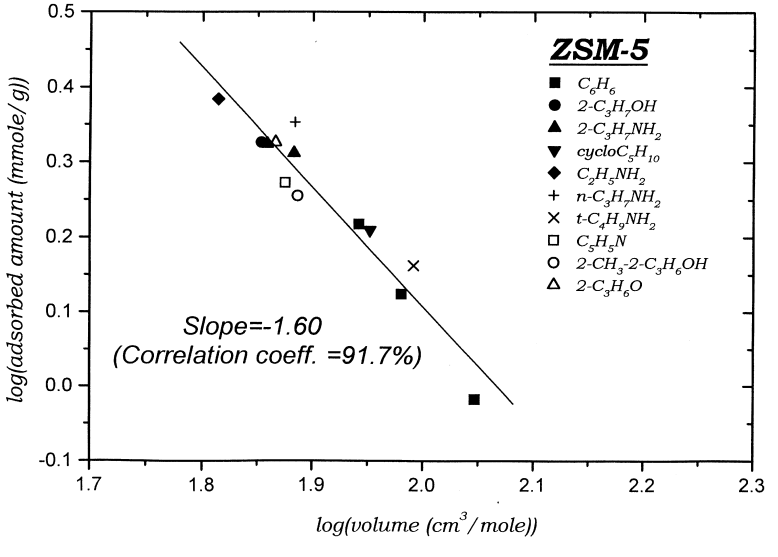

Fig. 6. Adsorption capacity vs. adsorbate molar volume: Data source: Choudhary and Srinivasan [47], Thamm et al. [48], Parrillo et al. [49], Kofke et al. [50].

The cube is divided into 10 equal parts in each direction. 240 objects are taken out on the top view of the fractal structure and 120 objects are taken out on the side view of the fractal structure. Total 296 objects are taken out since 64 objects overlap in the intersection. Employing Eq. (1) with $N = 10$ and $N_1 = 296$ and can obtain D_f of 2.85.

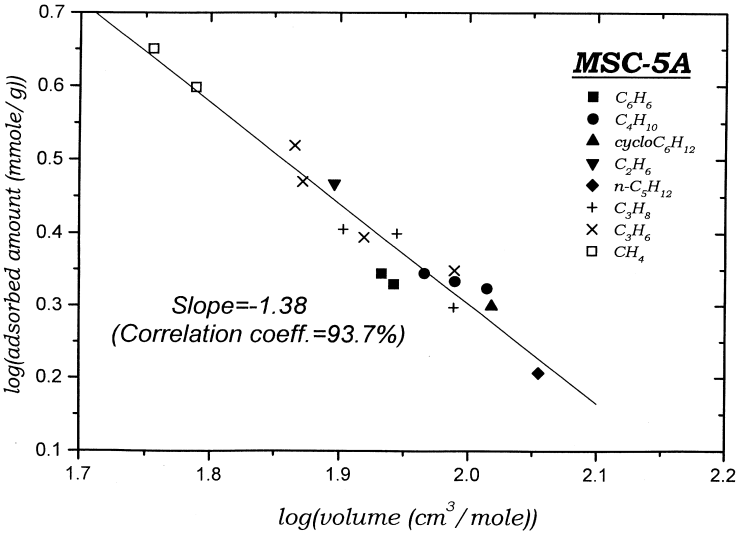

Fig. 7. Adsorption capacity vs. adsorbate molar volume: Data source: Valenzuela and Mylers [38], Nakahara et al. [51], and Nakahara et al. [52].

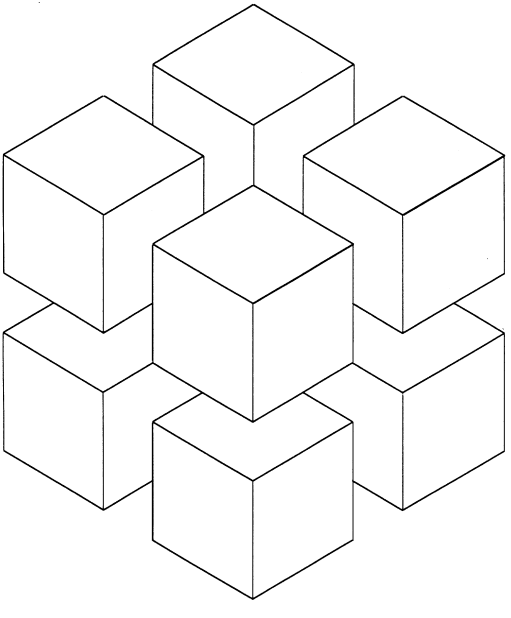

Fig. 8. Visualized zeolite A.

Employing Eq. (4) one can find out that the porosity is 0.296 at the first iteration. The errors of the fractal dimension and the porosity are 0.35% and 5.4%, respectively. Fig. 11 displays the visualized fractal structure of Mordenite herein. For zeolite Y, the cube is divided into four equal parts in each direction. Taking out 32 objects

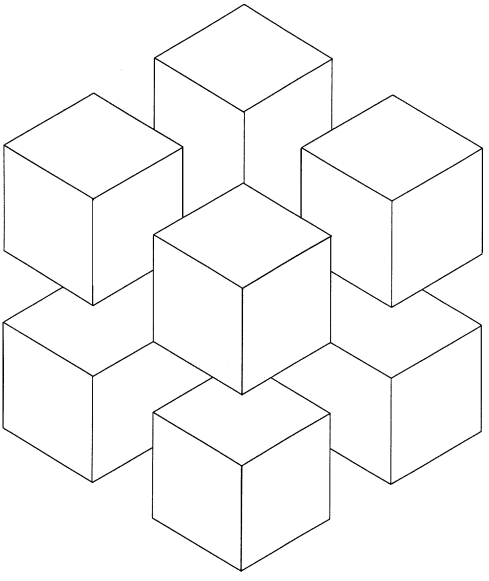

Fig. 9. Visualized zeolite X.

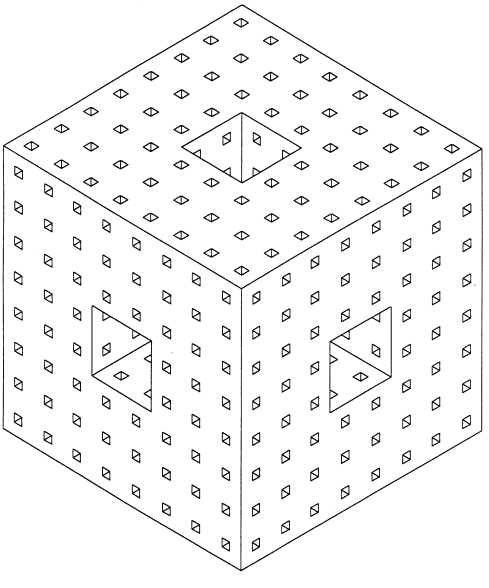

Fig. 10. Visualized Dowex MSC-1.

as displayed in Fig. 12, we can obtain the fractal dimension of 2.50 by employing Eq. (1) with $N=4$ and $N_1=32$. The porosity of fractal fracture at the first iteration is 0.5 by employing Eq. (4). The errors of fractal dimension and the porosity are 2.0% and 4.0%, respectively. Fig. 12 indicates the visualized fractal structure of zeolite Y.

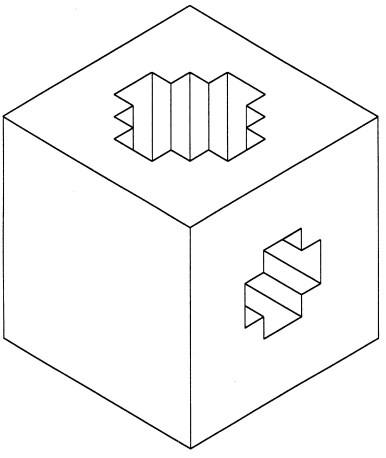

Fig. 11. Visualized mordenite.

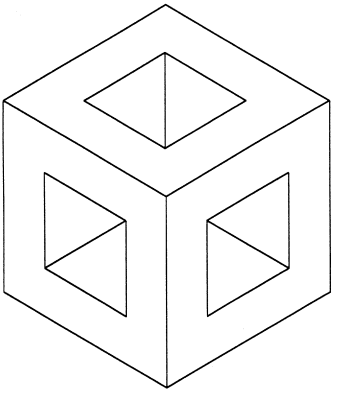

Fig. 12. Visualized Y-zeolite.

Fig. 13 displays the visualized fractal structure of ZSM-5. The cube is divided into five equal parts in each direction, and 45 objects in Z direction and 25 objects in X and Y directions are taken out. Totally 65 objects are taken out. The fractal dimension is 2.54. The porosity with one iteration is 0.52. The errors of the fractal dimension and the porosity are 2.4% and 3.8%, respectively. Finally, Fig. 14 illustrates the visualized fractal structure of the MSC-5A carbon sieve. Here, the cube is divided into six equal parts in three directions and only the middle four slabs are taken out in Y direction. It is a parallel layer structure. Employing Eq. (1), we can obtain the fractal dimension 2.39, and the error of the fractal dimension is 0.42%. A reliable porosity of MSC-5A is not found in the literature. However, employing Eq. (4), we can predict a porosity of 0.67 with one iteration. Table 1 summarizes the estimated results of fractal dimension

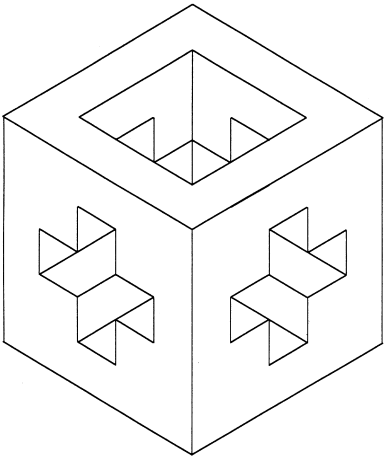

Fig. 13. Visualized ZSM-5 zeolite.

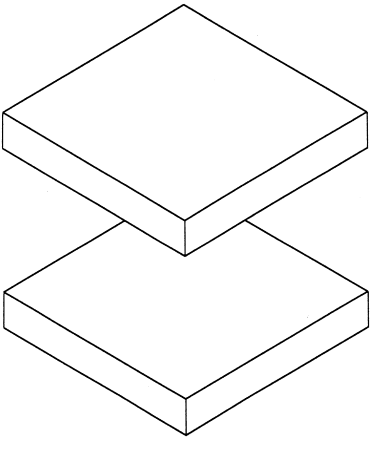

Fig. 14. Visualized MSC-5A.

and porosity of the seven porous solids. It is worth mentioning that zeolites A, X, Y have three-dimensional openings with same size of pores, but ZSM-5 is different, its opening being larger in one dimension than in the other two. Consequently, it can be seen in Fig. 13, that 9 units squares are taken out in the top plane, and 5 units squares from the side planes. As for the supercages in the channel intersections of zeolites A, X, Y, we do not have any adequate way of doing them, at present.

The visualized fractal structure of the porous solids can be characterized by Eqs. (1) and (4) as demonstrated in this study. The visualized fractal structure is

Table 1
Summary of estimation results of fractal dimension and porosity

Type of porous solid	Fractal dimension	Porosity
Zeolite A	2.58(2.57[a])	0.488(0.47[b])
Zeolite X	2.38(2.37[a])	0.58(0.5[b])
Dowex MSC-1	2.92(2.92[a])	0.288(0.3[b])
Mordenite	2.85(2.86[a])	0.296(0.28[b])
Y-zeolite	2.50(2.45[a])	0.5(0.48[b])
ZSM-5	2.54(2.60[a])	0.52(0.5[b])
MSC-5A	2.39(2.38[a])	0.67(estimated)

[a]The fractal dimension of the specified porous solid obtained from adsorption data.
[b]The porosity of the specified porous solid from literature.

useful for simulation of molecular adsorption and diffusion in the porous solids by setting up the realistic rules of the simulation process.

4. Conclusion

The Eqs. (1) and (4) can characterize the porous solid of fractal dimension and porosity. Moreover the Eq. (5) also illustrates the correlation between fractal dimension and porosity of the porous solid which also has been obtained by Hildgen et al. Since Eq. (5) involves additional N variable, it cannot be used directly to calculate fractal dimension from porosity or vice versa. Hence two independent equations are essential to construct the visualized fractal structure. It may be beneficial for simulation of molecular adsorption and diffusion in the porous solids with the fractal pictures.

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