Investigation of Intermingled Fractal Model for Organic-Rich Shale

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ABSTRACT: Organic-rich shales have abundant micronanopores and slits. Most micronanopores exist in organic matter and pyrites. Pore size distribution significantly influences shale gas flow. In micronanopores of organic matter and pyrites, pore size distribution has a self-imitating or fractal property. This property is helpful in upscaling calculation. In this work, intermingled fractal model for organic-rich shale is investigated. First, on the basis of scanning electron microscopy (SEM) images of shale samples in Longmaxi shale formation, fractal units are built in representative elementary surfaces of organic matter and pyrites. Second, intermingled fractal models (IFMs) are then built considering the organic matter fractal pore units, the pyrite fractal pore units, and slits. Influences of fractal parameters on apparent gas permeability of IFMs are investigated. Third, an upscaled shale model is successfully built using the mirror image method. This upscaling method is verified by comparing the apparent gas permeability between upscaled models and experimental results. The upscaling method based on fractal theory can significantly shorten the calculation time and provide a promising way for apparent gas permeability calculation in the upscaled model.

1. INTRODUCTION

A shale play, where most hydrocarbon is stored in tight matrix blocks, contains assemblage rock, reservoir, and caprock. This reservoir has ultralow permeability and porosity with abundant micronanopores, especially organic pores. Many problems occur in the developing process. The gas flow mechanism of shale is an especially complex problem. Investigating the flow mechanism of shale is important for gas production, hydraulic fracturing, and production forecasting of shale.

Shale has relatively strong heterogeneity with complex pore space and various pore types. Pore space is not easy to present precisely and effectively in a permeability calculation model. Many studies have adopted the rebuilding method for pore spaces, which can be generally classified via two ways. One is building a theoretical pore model based on statistical data, and the other is refactoring pore space based on core scanning data. Since the early 20th century, theoretical pore space has developed from the parallel tube bundle model to the network model. Fate† first combined parallel tube bundles based on actual pore size distribution with the network model and studied the relationship between pore space and apparent permeability. Fischer et al.‡ introduced a capillary pressure curve in the pore network model and studied the relationship between confine pressure and relative permeability. Bryant et al.§ introduced stress deformation into the pore network model to study the effect of confine pressure on permeability. Song et al.¶ investigated non-Darcy shale permeability under different stress conditions, taking both organic and inorganic matter into consideration. Jiang et al.‖ studied gas transport in pore space based on a high-precision pore network extraction algorithm. Meng et al.¶,‖ studied the influence of autoremoval mechanism on shale permeability. Yuan et al.¶,‖ examined nanofluid adsorption and liquid damage on permeability in nanoscale pore space. Peng et al.¶,‖ used thin-section image analysis to express pores to estimate permeability and investigated multiphase fluid flow in shale. Shabro et al.¶ established an apparent gas permeability model of shale, including diffusion, slip, and adsorption—desorption effect in kerogen. These factors significantly affect gas flow rate.

Upscaling is essential. In recent years, computer-scanning-based methods have considerably improved with the development of computer science. Traditional core experiment data and nanoscale computer simulation data must be combined. Numerous researchers have studied upscaling methods from the microscopic to the mesoscopic or macroscopic scale. Based on the network model, Ahmadi et al.¶† used volume-average upscaling method to investigate the diffusion coefficient of nonwetting phase in porous media. Frykman et al.¶‡ studied an upscaling method based on statistical information on geology. Gauiter et al.¶§ combined Bayesian theory with an upscaling method to study the physical parameters of rocks.

Pore space in cores have fractal properties on a specific scale, so does shale, thereby suggesting that such space can be rebuilt as fractal models based on their self-imitating properties. Fractal models provide an efficient way to express pore size distribution to calculate permeability. Many permeability models have been developed, based on fractal theory. Yu et al.¶¶ first developed a fractal in-plane permeability model for various fabrics. Xu and Yu¶¶ developed a permeability-prediction model for homogeneous porous media by introducing cross-section parameters. Cai et al.¶¶,¶§ established an effective permeability model based on spontaneous imbition by using the self-imitating properties of porous media. Zheng and Yu investigated gas flow and diffusivity in fractal porous media and derived related analytical models.¶¶,¶§ Shou et al.¶¶ developed a difference—fractal model to predict the permeability of fibrous porous media. Cai¶ introduced low-velocity non-Darcy terms in a low-permeability porous medium. Zhang¶ introduced the Knudsen diffusion term in the gas permeation equation of the fractal model. Pia and Sanna¶ first built intermingled fractal units.
(IFUs), which made pores easier to express. In following years, they developed the IFU model, which makes it easier to use in the calculation of conduction properties, such as permeability and imbibition.\textsuperscript{40–42} Innovation of the IFU model greatly simplifies the expression of pore size distribution.

2. INTERMINGLED FRACTAL UNITS AND PERMEABILITY CALCULATION

2.1. Introduction of IFU Model. Pia et al.\textsuperscript{39} utilized an approach based on the description of the microstructure of voids using the fractal method called the IFU model. This model consists of several fractal unit types based on different iteration parameters. Each fractal type is revised as Sierpinski carpet. The volume (area in the plane) of the IFU model can be expressed as

$$V_{n} = n_{A}V_{A} + n_{B}V_{B} + n_{C}V_{C}$$

where $V_{n}$ is the volume (area of the IFU model; $n_{A}$, $n_{B}$, and $n_{C}$ represent the number of each unit; and $V_{A}$, $V_{B}$, and $V_{C}$ represent the volume (area) of each unit.

To make the model understandable, assuming that the void space of the IFU model is combined with units A, B, and C, each unit has different iteration parameters. Figures 1(a) to (c) and (d) to (f) are two iteration examples to illustrate basic structure of units with different iteration parameters. The white area is void space, the black area is the iteration area, and the shaded area is a non-iteration space. The iteration factor $F$ is the number of generated squares on each side in one iteration process. In each iteration, the sides of a square are divided by three ($F = 3$), thus obtaining nine subsquares with some squares removed (for example, in Figure 1a, three squares are removed). In the next iteration, the remaining subsquares, each one is regarded as a new square and divided into nine subsquares; some subsquares will be removed. Particularly, if non-iteration space exits, the space does not take part in the iteration (see Figures 1d and 1e). $N_{p}$ represents the number of squares removed in each iteration. $N_{\text{solid}}$ represents the number of squares that always remained solid in each iteration. $\lambda_{i}$ represents the edge length of void area in the $i$th iteration.

According to fractal theory, the edge length of the void area in the $i$th iteration ($\lambda_{i}$) is

$$\lambda_{i} = \frac{\lambda_{i-1}}{F_{A}^{N_{p}}}$$

with the number of void areas generated in the $i$th iteration ($N_{i}$) being given as

$$N_{i} = (F_{A}^{2} - N_{\text{solid}} - N_{\text{non}}) \times 1 \times N_{p}$$

where $N_{\text{solid}}$ is the number of squares that always remain solid. The volume/area of void area with edge length $\lambda_{i}$ is

$$V_{i} = \lambda_{i}^{2}N_{i}$$

The total volume/area of void area in unit A ($V_{A}$) is given by

$$V_{A} = V_{A1} + V_{A2} + \cdots + V_{Ak} + \cdots + V_{Ai} = \sum_{k=1}^{i} V_{Ak}$$

where the total volume/area of void space before the $k$th iteration is

$$V_{A,k\text{accumulate}} = V_{A1} + V_{A2} + \cdots + V_{Ak}$$

The apparent gas permeability calculation method based on intermingle fractal distribution is discussed. Non-Darcy effects are negligible for gas flow in nanoscale pores. For a single capillary, Javadpour et al.\textsuperscript{43} provided a gas flow equation considering non-Darcy effects, such as slip and Knudsen diffusion effect, as

$$q_{\text{square}} = \frac{\lambda_{A} \mu M}{3 \times 10^{6} RT \rho_{\text{avg}}^{0.5}} \left(\frac{8RT}{\pi M}\right)^{0.5} \lambda^{3} \left[1 + \left(\frac{8RT}{\pi \rho_{\text{avg}}^{0.5} \lambda^{2}} \frac{2 \mu}{\lambda} \left(\frac{2}{3} - 1\right)\right)\right] A \left(P_{i} - P_{out}\right) \frac{P_{in}}{L}$$

where $\lambda_{A}$ is the equivalent diameter (which, in a square tube, is the edge length); $M$ is the molar mass of natural gas (kg/kmol); $\mu$ is the viscosity of natural gas (Pa s); $R$ is the gas constant ($R = 8.314$ J/(mol K)); $T$ is the temperature (K); $\rho_{\text{avg}}$ is the average density of natural gas (kg/m$^{3}$); $P_{in}$ and $P_{out}$ are the inlet pressure and outlet pressure.
pressures, respectively (Pa); \( p_{\text{avg}} \) is the average gas pressure (Pa); and \( \alpha \) is the tangential momentum accommodation coefficient. The viscosity and density of natural gas change greatly with average pressure, and the equation of viscosity and density of natural gas have been tested by Lee et al.\(^{44} \) in laboratory experiments. \( A \) is the cross-sectional area (in a single tube, \( A = \lambda_i^2 \)). \( L \) is the length of tube, the tortuosity can be expressed by fractal dimension in length as eq 8:\(^{40} \)

\[
L = L_0 D_T^{-1} \lambda_i^{-1-D_T} \quad (8)
\]

where \( L_0 \) is the straight length between two sides and \( D_T \) is the fractal dimension in length. \( D_T \) can be calculated by eq 9:\(^{45} \)

\[
D_T = 1 + \frac{\ln \tau}{\ln(L_0/\lambda)} \quad (9)
\]

\( \tau \) is the tortuosity of the tube, which can be calculated using eq 10:\(^{45} \)

\[
\tau = 1 + a \ln \left( \frac{1}{\epsilon} \right) \quad (10)
\]

where \( \epsilon \) is the porosity and \( a \) is a shape-related parameter. To simplify eq 7, the original equation can be expressed in polynomial form:

\[
q_{\text{square}} = (C Ca_1 + C Ca_2 + C Ca_3) \lambda_i^2 (p_{\text{in}} - p_{\text{out}}) \quad (11)
\]

\( A \) is the cross-sectional area (\( A = \lambda_i^2 \)), and parameters \( C_a, C_b, \) and \( C_c \) are given as

\[
Q_A = \left[ C_1 \sum_{i} \left( N_{\lambda_i^3} \lambda_i^{3+D_T} \right) + C_2 \sum_{i} \left( N_{\lambda_i^4} \lambda_i^{4+D_T} \right) + C_3 \sum_{i} \left( N_{\lambda_i^5} \lambda_i^{5+D_T} \right) \right] \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right)
\]

\[
= \left[ N_{\lambda_i^3} C_1 \frac{1 - ((F_T^2 - N_{\lambda_i^3})/F_A^{2+D_T})^{1/3}}{1 - ((F_T^2 - N_{\lambda_i^3})/F_A^{2+D_T})^{1+D_T}} + N_{\lambda_i^4} C_2 \frac{1 - ((F_T^2 - N_{\lambda_i^4})/F_A^{2+D_T})^{1/4}}{1 - ((F_T^2 - N_{\lambda_i^4})/F_A^{2+D_T})^{1+D_T}} + N_{\lambda_i^5} C_3 \frac{1 - ((F_T^2 - N_{\lambda_i^5})/F_A^{2+D_T})^{1/5}}{1 - ((F_T^2 - N_{\lambda_i^5})/F_A^{2+D_T})^{1+D_T}} \right] \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right)
\]

\[
= \left[ CA_1 \lambda_i^{2+D_T} + CA_2 \lambda_i^{3+D_T} + CA_3 \lambda_i^{4+D_T} \right] \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right)
\]

where \( CA \) indicates Knudsen diffusion item,

\[
CA_1 = N_{\lambda_i^3} C_1 \frac{1 - ((F_T^2 - N_{\lambda_i^3})/F_A^{2+D_T})^{1/3}}{1 - ((F_T^2 - N_{\lambda_i^3})/F_A^{2+D_T})^{1+D_T}}
\]

\( CA_2 \) indicates the Hagen–Poiseuille item,

\[
CA_2 = N_{\lambda_i^4} C_2 \frac{1 - ((F_T^2 - N_{\lambda_i^4})/F_A^{2+D_T})^{1/4}}{1 - ((F_T^2 - N_{\lambda_i^4})/F_A^{2+D_T})^{1+D_T}}
\]

\( CA_3 \) indicates the slip item,

\[
CA_3 = N_{\lambda_i^5} C_3 \frac{1 - ((F_T^2 - N_{\lambda_i^5})/F_A^{2+D_T})^{1/5}}{1 - ((F_T^2 - N_{\lambda_i^5})/F_A^{2+D_T})^{1+D_T}}
\]

This expression makes the flow equation more explicit. \( Q_b \) and \( Q_c \) can be expressed in the same manner:

\[
Q_b \quad [CB_1 \lambda_i^{2+D_T} + CB_2 \lambda_i^{3+D_T} + CB_3 \lambda_i^{4+D_T}] \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right)
\]

\[
Q_c \quad [CC_1 \lambda_i^{2+D_T} + CC_2 \lambda_i^{3+D_T} + CC_3 \lambda_i^{4+D_T}] \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right)
\]

Based on the theory of Javadpour et al.\(^{43} \), the volumetric gas flux is determined using three parts: Knudsen diffusion, Darcy flow, and increment of flow caused by slip effect. In eq 11, \( C_1 \) represents the Knudsen diffusion part, \( C_2 \) represents the Darcy flow part, and \( C_3 \) represents the increment part of flow caused by the slip effect. This expression makes the flow equation more explicit.

In basic fractal units, the apparent gas permeability model can be expressed as a bundle of square tubes. The gas flow rate can be expressed as eq 12 (taking Unit A for example):

\[
Q_A = \sum_i \left[ N_{\lambda_i^3} (C_{a1}^3 + C_{a2}^4 + C_{a3}^5) \left( \frac{p_{\text{in}} - p_{\text{out}}}{\mu L_0 D_T} \right) \right]
\]

where \( N_{\lambda_i} \) is the number of tubes in stage \( i \), determined by eq 3; by plugging eqs 2–5 into eq 12, the gas flow rate of a single fractal unit (e.g., Unit A) can be expressed as eq 13:
contain abundant organic matter, especially graptolites. The Longmaxi Formation of the Sichuan Basin. These shales core samples from a well drilled at a depth of 2357 m in the Lower Silurian in the Sichuan Basin. Samples 3 and 4 are fresh from a fresh outcrop of the Longmaxi Marine Shale Formation of the Lower Silurian in the Sichuan Basin. Shales come from the Longmaxi Marine Shale Formation of the Lower Silurian in the Sichuan Basin. Samples 3 and 4 are fresh core samples from a well drilled at a depth of 2357–2406 m in the Longmaxi Formation of the Sichuan Basin. These shales contain abundant organic matter, especially graptolites.

The rock mineralogy of the samples was analyzed by energy-dispersive spectroscopy (EDS) in an area of 409.7 μm × 409.7 μm, using a Zeiss–Merlin rock mineralogy analysis machine, with a resolution of 5 nm. Table 1 shows that the main minerals of shale samples 1, 2, and 3 are quartz, clay minerals, and feldspar. Dolomite is the dominant mineral in Sample 4.

### Table 1. Rock Mineralogy of the Samples

<table>
<thead>
<tr>
<th>mineral</th>
<th>samples 1 and 2 vt. (%)</th>
<th>sample 3 vt. (%)</th>
<th>sample 4 vt. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>quartz</td>
<td>60.9</td>
<td>18</td>
<td>6</td>
</tr>
<tr>
<td>clay minerals</td>
<td>8.9</td>
<td>38</td>
<td>3</td>
</tr>
<tr>
<td>feldspars</td>
<td>14.3</td>
<td>35</td>
<td>10</td>
</tr>
<tr>
<td>calcite</td>
<td>1.71</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>pyrite</td>
<td>1.53</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>biotite</td>
<td>4.04</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>dolomite</td>
<td>2.73</td>
<td>2</td>
<td>76</td>
</tr>
<tr>
<td>total organic content, TOC</td>
<td>3.0</td>
<td>0.97</td>
<td>2.41</td>
</tr>
<tr>
<td>other</td>
<td>2.89</td>
<td>1.73</td>
<td>0.39</td>
</tr>
</tbody>
</table>

Dry sample permeability of gas is measured by pulse-decay permeability measurement under 25 °C from three different directions. Other test conditions are shown in Table 2.

### Table 2. Test Conditions of Pulse-Decay Permeability Test

<table>
<thead>
<tr>
<th>flow parameter</th>
<th>value</th>
</tr>
</thead>
<tbody>
<tr>
<td>molecular mass of N2</td>
<td>28 g/mol</td>
</tr>
<tr>
<td>molecular mass of He</td>
<td>4 g/mol</td>
</tr>
<tr>
<td>pressure difference, ΔP</td>
<td>1.1 MPa</td>
</tr>
<tr>
<td>average temperature</td>
<td>306.24 K</td>
</tr>
<tr>
<td>average pressure</td>
<td>0.55 MPa</td>
</tr>
<tr>
<td>gas constant</td>
<td>8.314 J/(K mol)</td>
</tr>
</tbody>
</table>

The average gas permeability is calculated by the quadratic polynomial method, using the following equations:

\[
\frac{P_{1,t} - P_{2,t}}{P_{1,0} - P_{2,0}} = e^{-\alpha t}
\]

where \(P_{1,t}\) and \(P_{2,t}\) are the inlet and outlet pressure, respectively, at time \(t\), and \(P_{1,0}\) and \(P_{2,0}\) are the initial inlet and outlet pressure, respectively. Parameter \(\alpha\) can be expressed as

\[
\alpha = \frac{k(A + 2\mu L)}{2\mu L} \left(\frac{1}{V_1} + \frac{1}{V_2}\right)
\]

where \(k\) is the apparent permeability, \(A\) is the cross-sectional area, \(L\) is the length of the sample, \(\mu\) is the gas viscosity, and \(V_1\) and \(V_2\) are the inlet and outlet volume, respectively. Using sample 1 as an example, the original pulse-decay permeability measurement curve is shown in Figure 2; the apparent permeability has been tested under confining pressures of 3, 5, 7, and 9 MPa. In addition, since SEM images are tested with no confining pressure, the quadratic polynomial method is used to calculate the apparent permeability under no confining pressure to ensure that the conditions are unchanged.

As the result shows, the average gas permeability of sample 1 is 91.36 nD, as calculated by the quadratic polynomial method. The apparent permeability in other samples has been derived in the same way, the results have shown in Table 5 (presented later in this work). Nitrogen is used in this test. The viscosity, density, and compressibility factor of the gas changes with the average temperature and pressure. We use the experimental results that have been reported by Lee et al.

Scanning electron microscopy (SEM) is used to investigate the nanoscale pore shape and the spatial distribution of Longmaxi samples. The samples have ion polishing before SEM tests. The scanning area is 380 μm × 380 μm, with a maximum resolution of 2.5 nm. Total spliced images of two different areas are shown in Figures 3a and 3b. Figures 3c and 3d are magnified images in a specific area, which clearly show inorganic slits, as well as pores in organic matter and pyrites. Specifically, pores in pyrites are filled in overdeveloped pyrites.

In organic-rich shales, organic matter (Figure 3e) has the most organic pores, and pyrites (Figure 3f) contain some organic pores. In addition, inorganic slits scatter in SEM images (Figure 3e). Micronanopores have fractal properties at a specific scale. Pores have fractal characteristics in each component. We generate IFUs for pores in organic matter and pyrites; the pore size distribution of models follows that of actual samples calculated from the SEM image. Intermingled fractal models (IFMs) are built by mixing pores in organic matter, as well as pores in pyrites and slits. Slit density is calculated from the statistical data of SEM images. This model combines well with both microscale mineral content distribution and pore size distribution of each mineral in nanoscale. Shale gas flow in IFM...
can be calculated easily and effectively considering the non-Darcy effect, such as slip and Knudsen diffusion.

3.2. Organic Matter Pores. Organic matter has abundant pores with a vast distribution. The modeling process of organic matter is shown in Figure 4 and is detailed below.

(1) The representative elementary surface (RES) is obtained from statistical regularity. The heterogeneity of organic matter distribution is relatively strong in a small area. To calculate the smallest RES, four different initial points are chosen (four corners of an image in this example). We then expand the target area starting from the source points (1 pixel on the edge) in the original SEM image and calculate the average grayscale in the subsquare. The variation of the average grayscale will diminish gradually as the pixels on the edge of the subsquare increases. By calculation, since the number of pixels on the edge is >400, the average grayscale remains stable. Therefore, an image with no fewer than 400 pixels can be regarded as RES. We choose 400 pixels on the edge as RES.

(2) After being exacted, the RES image is segmented to exact pores. In the segmentation process, the threshold value is the most important parameter. The pore fraction of RES is regarded to be the same as the experiment porosity. By adjusting the grayscale of the threshold with a certain value, the pore fraction of exacted RES is equal to the experiment result. Hence, pores in RES have been derived. Pore cumulative distribution curve (blue curve in Figure 5) is calculated from the chosen two images (Figures 3a and 3b).

(3) The basic IFU model is built, and the cumulative pore curve (red curve in Figure 5) is calculated. Fractal parameters are adjusted to narrow the differences between cumulative curves of the basic IFU model and image calculation. Afterward, certain parameters of the basic IFU model are determined, as shown in Table 3.
3.3. Pyrite Pores. Pyrites are scattered in the SEM area. First, the distribution density of pyrites (number of pyrites in this area) is observed from statistical data based on the SEM image. The pores in one pyrite then are represented as IFUs. Finally, the pores in the model are scattered in the IFM with the observed distribution density. The modeling process of the pyrite parts is shown in Figure 6.

1. All the pyrites are exacted in a relatively large scale (similar to that in Figure 3a), and the distribution density of pyrites is calculated from the statistical data of the SEM image, excluding overdeveloped pyrites. \( N \) pyrites are assumed in the area.

2. One pyrite with an average diameter is selected and magnified (see Figure 6). The image is segmented to exact pores, and the pore cumulative distribution curve (the blue curve in Figure 7) is calculated.

3. The basic IFU model is built, and the pore cumulative curve (the red curve in Figure 6) is calculated. Fractal parameters are adjusted to narrow the differences between cumulative curves of the IFU model and image calculation. Afterward, certain parameters of the basic IFU model are determined, as shown in Table 4. The result in Figure 7a is calculated from Figure 3a, while Figure 7b is calculated from Figure 3b.

4. The basic IFU model is scattered in the area of Figures 3a and 3b, and the distribution density of pyrites is calculated.

3.4. Slits. Slits or cracks also have influence on gas permeability. Distribution of slits have relatively strong heterogeneity. They should be observed from different scales. The distribution of slits should be obtained from a relatively larger scale, for the heterogeneous distribution, while the aperture of slits should be observed in larger-resolution images. We determine the statistical parameters of inorganic slits by calculating the distribution density of inorganic slits in a large area and their average length and aperture in a magnified image. The method of intermingling the IFU of the organic matter, IFU
of the pyrites, and inorganic slits will be discussed in the subsequent section.

3.5. Building IFM. The foundation process of IFMs based on SEM images is mainly divided into three steps. First, the SEM images are obtained, and the representative elementary surface (RES) is determined. Second, IFUs are built for pores in organic matter and pyrites. Third, IFM is built by IFU and slits. This section discusses the modeling process of organic matter and pyrites.

IFUs for organic matter and pyrites are presented in section 3.4. In this section, the IFM is developed by considering the pores in organic matter, as well as the pores in pyrites and slits. Based on this model, the upscaling process is tested by using the mirror image method. The accuracy and reliability of this upscale method is examined by experiment verification.

The building process of IFM is shown in Figure 8. First, we calculated the RES of organic matter, as shown in the top part of Figure 8. In this scale, the IFU model of organic matter attempts to follow the pore cumulative curve in SEM images of organic matter. Fractal parameters are adjusted to narrow the difference between the curve of the model and the SEM image. Second, we calculated the distribution density and average diameter of pyrites. Overdeveloped pyrites are removed because they have no pores. Within one typical pyrite, the IFU model of the pyrite is generated in the same way. By calculating the distribution density and the average length and aperture of slits, we obtain the statistical parameters of slits, as shown in the middle part of Figure 8. Finally, the IFM in RES can be obtained by mixing these three components. Pores in the IFU model of the organic matter and the pyrites are scattered into the IFM randomly with slits. The final pore size distribution also follows the statistical data of the final SEM image.

<table>
<thead>
<tr>
<th>SEM 1</th>
<th>SEM 2</th>
<th>SEM 3</th>
<th>SEM 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unit A</td>
<td>Unit B</td>
<td>Unit C</td>
<td>Unit A</td>
</tr>
<tr>
<td>Df</td>
<td>1.89</td>
<td>1.77</td>
<td>1.46</td>
</tr>
<tr>
<td>number of units</td>
<td>1</td>
<td>5</td>
<td>230</td>
</tr>
<tr>
<td>number of pores($i=1$)</td>
<td>1</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>λ_{min} (nm)</td>
<td>7.07</td>
<td>8.89</td>
<td>8.89</td>
</tr>
<tr>
<td>number of iterations</td>
<td>4</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>λ_{max} (nm)</td>
<td>191</td>
<td>80</td>
<td>26.67</td>
</tr>
<tr>
<td>solid forever</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>total surface area ($\mu m^2$)</td>
<td>4.33</td>
<td>3.35</td>
<td>12.97</td>
</tr>
</tbody>
</table>

Figure 6. Modeling process of pyrite parts.
of 380 μm × 380 μm, we obtain the IFM, as shown in the lower right corner of Figure 8. SEM image 2 (Figure 3b) is calculated in the same way. Permeability can also be derived based on these fractal parameters. The equivalent gas permeability of organic matter, pyrites, and slits can be derived from eqs 15−17, respectively. Evidently, the apparent gas permeability of the IFM is the sum of that of the components, as shown in the bottom part of Figure 8.

3.6. Apparent Gas Permeability. Then, in organic RES (using organic RES mingled by Units A, B, and C, for example), the gas flow rate can be expressed as

\[ Q_{\text{res-org}} = n_A Q_A + n_B Q_B + n_C Q_C \]  

(14)

Denoting the area of organic RES as \( A_{\text{res}} \) and the volume fraction of TOC as \( \varphi_{\text{TOC}} \), the equivalent gas permeability of organic is given as

\[ k_{\text{app-org}} = \frac{Q_{\text{res-org}}}{A_{\text{res}}} \frac{A_{\text{sum}} \varphi_{\text{TOC}}}{A_{\text{res}}} \frac{\mu L_0}{A_{\text{sum}} (P_0 - P_{\text{out}})} \]

\[ = \frac{\varphi_{\text{TOC}}}{A_{\text{res}}} \sum_{i=A}^{C} n_i (C_i \lambda_{\text{max}}^{2+D_T} + C_i \lambda_{\text{max}}^{3+D_T} + C_i \lambda_{\text{max}}^{2+D_T}) \]  

(15)

Similarly, the equivalent gas permeability of pyrite is given as

\[ k_{\text{app-py}} = \frac{n_{\text{py}}}{A_{\text{sum}}} \sum_{i=A}^{C} n_i (C_{1,\text{py}} \lambda_{\text{max-py}}^{2+D_T} + C_{2,\text{py}} \lambda_{\text{max-py}}^{3+D_T} + C_{3,\text{py}} \lambda_{\text{max-py}}^{2+D_T}) \]  

(16)

where \( n_{\text{py}} \) is the number of pyrite in IFM.

Similarly, the equivalent gas permeability of slit is given as

\[ k_{\text{app-slit}} = C_{\text{py}} \frac{n_{\text{silt}}}{A_{\text{sum}}} (C_{1,\text{silt}} \lambda_{\text{silt}}^{2} + C_{2,\text{silt}} \lambda_{\text{silt}}^{2} + C_{3,\text{silt}} \lambda_{\text{silt}}^{2}) \]

\[ = \frac{C_{1,\text{silt}} n_{\text{silt}}}{A_{\text{sum}}} \frac{\lambda_{\text{silt}}^{2}}{L_0^{D_T-1}} \]  

(17)

where

\[ C_{1,\text{silt}} = \frac{\mu M}{3 \times 10^3 R T/\text{avg}} \left( \frac{8 R T}{\pi M} \right)^{0.5} \]

\[ C_{2,\text{silt}} = \frac{1}{32} \]

\[ C_{3,\text{silt}} = \left( \frac{8 \pi R T}{M} \right)^{0.5} \left( \frac{\mu}{16 \text{avg} (2/a - 1)} \right) \]

\( n_{\text{silt}} \) is the number of slits in final research area, \( \lambda_{\text{silt}} \) is the equivalent diameter of slit, and \( C_{\text{silt}} \) is a shape factor, which can be determined using the work of Veltzke et al.47

If Knudsen diffusion and slip effect are ignored, \( C_1 = C_3 = 0 \), \( C_{1,\text{py}} = C_{3,\text{py}} = 0 \), \( C_{1,\text{silt}} = C_{3,\text{silt}} = 0 \). Only the Darcy item is left. The permeability, under the circumstances, is traditional Darcy permeability. The apparent Darcy gas permeability of each component is

\[ k_{\text{app-ori}} = \frac{\varphi_{\text{TOC}}}{A_{\text{sum}}} \sum_{i=A}^{C} n_i (C_i \lambda_{\text{max}}^{3+D_T}) \]  

(18)

\[ k_{\text{app-py}} = \frac{n_{\text{py}}}{A_{\text{sum}}} \sum_{i=A}^{C} n_i (C_{1,\text{py}} \lambda_{\text{max-py}}^{3+D_T}) \]  

(19)

The permeability under the circumstances is traditional Darcy permeability. The apparent Darcy gas permeability of each component is

\[ \sum_{i=A}^{C} n_i (C_i \lambda_{\text{max}}^{3+D_T}) \]

The permeability under the circumstances is traditional Darcy permeability. The apparent Darcy gas permeability of each component is

\[ \sum_{i=A}^{C} n_i (C_i \lambda_{\text{max}}^{3+D_T}) \]

\[ \sum_{i=A}^{C} n_i (C_{1,\text{py}} \lambda_{\text{max-py}}^{3+D_T}) \]
4. FRACTAL PARAMETERS ON PERMEABILITY

In the IFM, fractal parameters control the pore size distribution. To examine the relationship between pore size distribution and apparent gas permeability, the influence of fractal parameters on apparent gas permeability is investigated. The primary fractal parameters are the number of units, the iteration time \(i\), the number of removed blocks in each iteration \(N_p\), and the number of blocks that always remain solid in each iteration \(N_s\). The permeability is calculated using eq 15.

\[
k_{\text{ins, slit}} = C_{py} \frac{n_{\text{slit}}}{A_{\text{sum}}} \left( C_{2, \text{slit}} \lambda_{n, \text{slit}} \right)^{\frac{D_{p, \text{slit}}}{L_0}} \]

(20)

Figure 8. IFM procedure to calculate permeability.
To make the influence of fractal parameters more understandable, we discussed the influence of iteration time $i$, the number of removed blocks in each iteration ($N_p$), and the number of blocks that always remain solid in each iteration ($N_s$) in Unit A of organic matter component. Basic parameters are shown in Table 3.

The apparent permeability varies with pressure. The non-Darcy flow effect increases as the average flow pressure decreases during the percolation process. The apparent permeability is calculated for six different average pore pressures (see Figure 9), namely, 0.1, 0.3, 0.55, 1, 2, and 5 MPa, with the average temperature being fixed at 306.24 K. The non-Darcy flow effect emerges when the average pressure is <1 MPa, especially under 0.3 MPa. Many small pores are generated as the iteration time increases. Initially, the increase of iteration time $i$ significantly affects both Darcy and non-Darcy permeability. Meanwhile, as $i$ increases, this effect diminishes, similar to increases from 3 to 5. The diameter of pores in the fifth iteration is too small and therefore cannot significantly influence the apparent permeability of the model. Non-Darcy effects in gas flow should be considered in the analysis of gas flow in micronanopores under low pressure.

The removed number $N_p$ is the number of removed blocks in each iteration. $N_p$ generally controls the general porosity of the model. Figure 10 shows the variation of apparent permeability with pressures in different iteration numbers. The non-Darcy flow effect increases as the average flow pressure decreases during the percolation process. As the $N_p$ increases, the Darcy and non-Darcy permeabilities increase linearly. In the iteration process, as the number of removed pores in each iteration ($N_p$) increases, the cross section of the model increases linearly, thereby making the permeability of the model increase linearly at the same time.

Apparent permeability also varies with the number of solid blocks ($N_s$). In contrast to $N_p$, as $N_s$ increases, the pore number of the fractal model decreases, because some blocks will never part in the iteration process, thereby decreasing the porosity of the model. $N_s$ should be no less than zero and no greater than $F^2 - N_p$. The variation of the apparent permeability with pressures in different solid block numbers are shown in Figure 11. As $N_s$ increases, the apparent permeability decrease. While the influence of $N_s$ on permeability is not as great as that of $N_p$, because $N_s$ increases from 0 to 4, the variation of permeability is significantly smaller than $N_p$. $N_s$ slightly influences the non-Darcy permeability.

Unit number is important for permeability. To make the model understandable, we choose the organic matter part. Parameters are obtained from organic matter calculations (Table 3). Figure 12 shows the influence of the number of units. From Figure 12a, the cumulative distribution of pores can follow the SEM result with all three units; the lack of any subunits will cause deviation in the cumulative curve. As the number of subunits increase, more tiny pores will be generated, causing the permeability to increase, especially under low pressure (see Figure 12b).

### 5. UPSCALING

The next process is upscaling. The area of the IFM is the same as that in the SEM image, i.e., $\sim 0.4 \mu m \times 0.4 \mu m$, which is relatively smaller than that of the actual rock sample. Upscaling should be taken into consideration. On the basis of capillary bundle theory, IFM units are spliced with each other. On the edge of the image, some pores may not be complete. To avoid this illogical incompleteness, the mirror image method is used. Vertical neighbor images have been flipped up to down, whereas horizontal neighbor images have been flipped left to right, as shown in Figure 13.

To examine the validation of RES after upscaling, we calculated the variation of the average grayscale with pixels on the edge, as shown in Figure 14b. The original points to be enlarged are selected from four corners (Figure 14a). The number of squares on the edge of the upscaled model should be odd. Therefore, when the research area is larger than 400 pixels on the edge, the average grayscale from different directions tend to be stable, which means that the area of IFM can be regarded as an RES of the upscaled image.
During the upscaling process, the variation of apparent gas permeability should also be carefully considered. In a valid upscaling process, the apparent gas permeability should be stable with the increase of the research area. As described, the apparent gas permeability model is regarded as a capillary bundle with the cross section of IFM, similar to that in Figure 15a. Apparent gas
permeability literally represents the permeability of the model in unit area. When the restricted research area doubles or triples in size, pore distribution and porosity will remain stable. In the upscaling process, the apparent gas permeability of the model is calculated as shown in Figure 15b. The environment parameters are set the same as those of the parameters in Table 2. Figure 15b shows that the calculated apparent gas permeability remains stable as the edge length increases from 0.2 mm to 20 mm. In conclusion, the upscaling process is valid.

We conducted experimental verification to examine the validation of this model. Experimental and IFM calculation results are shown in Table 5. Model results are generally close to the experimental data. Meanwhile, some differences also exist. These differences mainly caused by two processes. First is the simulation of pore size distribution. It can be observed in Figure 5 that the simulated curve does not exactly coincide with real one. The second is fractal tortuosity calculation process. Although simplification methods based on fractal theory was used, it can significantly shorten the calculation time and provide a promising way for apparent gas permeability calculation. The method in this work also has some room for improvement for the 3D permeability calculation process and we are working on the process. For some marine shale samples, to observe some organic pores below the resolution of SEM image (2.5 nm), other observation methods, such as Brunauer—Emmett—Teller (BET) or nonlinear density functional theory (NLDFT) method in nitrogen adsorption—desorption results, may be useful. A "splice" method to combine pore size distribution result from the NLDFT method and the SEM method is innovating in future work.

6. CONCLUSIONS
In this paper, the intermingled fractal model (IFM) for organic-rich shale is successfully built, based on the SEM results of shale samples in the Longmaxi Marine Shale Formation of the Lower Silurian in the Sichuan Basin. The IFM model contains pores in organic matter, pyrites, and inorganic slits. In the IFM, the influence of fractal parameters on gas permeability is explored. The accuracy and reliability of the upscale process are examined by experiment verification. The apparent gas permeability of the model matches the experimental results. Upscaling is improved by using fractal characteristics.

Findings show that obtaining fractal parameters from SEM images is valid. The IFM model takes advantage of the detailed information at the micronanoscale and the distribution of the component in a large observation area. The calculation process for fractal parameters is efficient and easy, thus establishing a good foundation for percolation calculation. Non-Darcy effects in gas flow should be taken into account in micronanopores under low pressure. Higher iteration times (i), a higher number of removed pores ($N_{fl}$), and a lower number of solid blocks ($N_{res}$) are helpful for increasing the permeability. The upscaling process is valid and easy in IFM. With the use of an image flipping method, a small area of IFM based on the SEM image is upscaled to a larger surface. This IFM-based upscaling method can significantly simplify calculation of the apparent gas permeability for the upscaled model, which may promote the method in the future.

Table 5. Comparison between Experimental and IFM Permeability Data

<table>
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<tr>
<th>Permeability Data (nD)</th>
<th>$K_{org}$</th>
<th>$K_{py}$</th>
<th>$K_{slit}$</th>
<th>$K_{fl}$</th>
<th>$K_{py+slit}$</th>
<th>$K_{org+py+slit}$</th>
<th>error (%)</th>
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<tr>
<td>SEM 1</td>
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<tr>
<td>SEM 4</td>
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<td>1.17</td>
<td>1.24</td>
<td>8.58</td>
<td>6.24</td>
<td>37.50</td>
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Figure 15. Variation of calculated apparent gas permeability in the upscaling process.
REFERENCES


