

1 Nanopore size distribution heterogeneity of organic-rich shale reservoirs using multifractal
2 analysis and its influence on porosity-permeability variation

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13
14 **Abstract:** The shale nano-pore size (diameter <100nm) distribution heterogeneity
15 (SNDH) is one of the important factors affecting gas production. However, quantitative
16 analysis of the SNDH and the applicability of single and multi-fractal model needs to
17 be further studied. Here, based on low temperature liquid nitrogen and carbon dioxide
18 tests of organic rich shale in Qinshui Basin, multifractal dimension variation of micro-
19 pores (< 2 nm) and meso-pores (2–100 nm) are studied, and the multifractal factors that
20 affect the distribution of nano-pores are determined. Additionally, the differences
21 between single fractal and multifractal results are compared. Based on this, dynamic
22 variation of porosity and permeability under the constraints of nano-pore structure is
23 discussed from the perspective of multifractal variation. The results of this study are as
24 follows: 1) pore size distribution of micro-pores and meso-macro-pores in shale
25 samples exhibit typical multifractal behavior. The overall distribution heterogeneity of
26 meso-macro-pores is mainly affected by the distribution of pores in the low value area
27 of pore volume (LAPV), while the overall distribution heterogeneity of micro-pores is
28 affected by the distribution of high value area of pore volume. The multifractal
29 parameters and influencing factors of micro-pores and meso-macro-pores are clearly
30 different. 2) The single fractal dimension D_2 calculated using the Frenkel-Halsey-Hill

1 model has a negative correlation with the multifractal parameters, implying that the
2 distribution heterogeneity of the LAPV gradually decreases with the increase of the D_2
3 value, indicating that the physical meaning of the two models is obviously different. 3)
4 The pore distribution heterogeneity affects permeability variation and diffusion process
5 of shale reservoir. With the increase of the multifractal dimension of meso-macro-pores,
6 the damage effect of stress on permeability is stronger. The more heterogeneous the
7 micro-pore size distribution is, the smaller the "modification effect" of stress on the
8 diffusion coefficient.

9 **Key words:** shale reservoirs; pore structure heterogeneity; permeability variation;
10 diffusion; fractal dimension

11 **0 Introduction**

12 As an unconventional natural gas reservoir, nano-pores are widely developed in
13 shale reservoir. The adsorption pore surface adsorbs methane molecules in the form of
14 physical adsorption, which controls adsorption characteristics and affects the gas
15 bearing content of the shale reservoir. Accordingly, pore structure has become an
16 important factor in determining the production potential of shale gas ^[1-3].

17 At present, experimental techniques such as photoelectric observation, gas
18 adsorption and fluid intrusion, as well as three-dimensional structure reconstruction
19 methods, are widely used in the study of unconventional reservoir pore-fracture
20 structure ^[4-9]. Among them, fluid injection has become the most common technique.
21 Along with high pressure mercury injection (HPMI) tests, low pressure nitrogen
22 adsorption (LPN₂ GA) tests have become an effective technology to characterize shale
23 nano-pore structure ^[10-11]. However, given the large molecular diameter (0.36 nm) and
24 the accuracy of the instrument, it is difficult to accurately characterize the microporous
25 structure ^[10, 12-17].

26 In contrast to nitrogen, carbon dioxide has a low molecular diameter and strong
27 adsorption capacity and it can enter into pores of diameter 0.3–1.5 nm at 273.15 K.
28 Therefore, comprehensive utilization of both liquid nitrogen and carbon dioxide
29 adsorption tests can help realize the full-scale pore structure of shale reservoirs ^[12-13].
30 In addition, since the storage and migration of shale methane is closely related to the

1 pores and surfaces of unconventional reservoirs with self-similarity, the fractal
2 dimension value obtained from LPN₂ GA test has become an effective physical
3 parameter to quantitatively describe the nano-pore structure heterogeneity of shale
4 reservoirs [14-17]. This combination of tests and analysis therefore provides a basis for
5 quantitative description of shale pore structure heterogeneity.

6 However, the above model can only describe shale nanopore size distribution
7 heterogeneity (SNDH) simply by a single fractal dimension. For shale reservoirs with
8 strong heterogeneity, the pore size distribution (PSD) curve usually fluctuates or jumps
9 randomly, and different pore size intervals may have different types of self-similarity,
10 and so it is difficult to fully characterize pore homogeneity with a single fractal
11 dimension [18-19]. To overcome this, a multifractal approach can be adopted, in which a
12 fractal body is divided into small regions with different singularities. The advantage of
13 studying the fractal characteristics of these different regions is that it enables the
14 detailed structure to be understood hierarchically and allows the multiple characteristics
15 of pore size distribution of reservoirs to be analyzed based on HPMI, LPN₂ GA and low
16 field nuclear magnetic resonance (LF-NMR) test data. In this context, Vidal-Vazquez
17 et al. [20] studied the multifractal characteristics of soil by using LPN₂ GA data, and
18 concluded that this parameter has a clear relationship with the basic properties. Liu et
19 al. [21-22] analyzed the multifractal parameters of shale samples and indicated that shale
20 nano-pores have a better multi-fractal variation. Li et al. [23] and Song et al. [24] analyzed
21 the multifractal variation of tectonic coal using HPMI tests, and showed that the
22 multifractal parameters have a clear relationship with the degree of structural
23 development. Zhang et al. [25-26] compared the variation of single and multifractal
24 parameters through LPN₂ GA and NMR T_2 spectra respectively, and concluded that the
25 multifractal model has good applicability for describing the heterogeneity of tight
26 sandstone pore size distribution.

27 While studies on multifractal features have expanded from the macroscopic to the
28 microscopic pore scale, there are still some problems that need to be addressed.
29 Compared with other types of reservoirs, there are very few studies on multifractal
30 analysis of nanopores in shale reservoirs, and the influencing factors on multifractal

1 parameters need to be clarified. Moreover, micropores are the main pores for methane
2 adsorption. There are relatively few studies on the NPSH in this pore component and
3 in particular, the variation of multifractal characteristics needs to be further studied. In
4 general, it is the above two factors that restrict the accurate quantitative characterization
5 of shale nanopore structure.

6 With this in mind, this study aim to analyze the multifractal characteristics of
7 micropores and meso-macropores of shale samples by using LPN₂/CO₂ GA of organic
8 shale in Qinshui Basin, and the factors influencing the nano-pore size distribution. The
9 differences between single fractal and multifractal results are also compared. Based on
10 this, dynamic variation of porosity and permeability under the constraints of nanopore
11 structure will be discussed from the perspective of multifractal variation. Overall, the
12 results of the study are anticipated to provide a theoretical basis for the quantitative
13 evaluation of shale reservoirs pore heterogeneity.

14 **1 Experimental sample and methods**

15 **1.1 Sample collection and basic analysis**

16 The shale samples were obtained from Well 1 in Qinshui Basin, which is located
17 at the eastern edge of the central part of the basin, with a total thickness of 162.67m. A
18 total of 12 samples were collected from black shale from top to bottom ^[16]. Basic tests
19 such as TOC, whole-rock mineral composition and $R_{o, \max}$ are performed on fresh shale
20 samples. For a detailed description of these experimental test procedures, please refer
21 to Yan et al. ^[16].

22 **1.2 Experimental methods**

23 After measuring TOC, field emission scanning electron microscopy (FE-SEM)
24 and LPN₂/CO₂ GA tests were performed on the samples in sequence (the reader is
25 referred to Yan et al. ^[16] for detailed information), and the overburden porosity and
26 diffusion coefficient tests were performed on a selection of four representative samples.
27 Information on the sample tests is shown in Table 1.

28 Different permeability was measured by pressure-sensitive testing using an AP-
29 608 Automated Permeameter-Porosimeter ^[25]. The experimental method is based on
30 the non-steady state pressure drop method and the experimental gas comprised high-

1 purity nitrogen. During the measurement process, the gas pressure difference between
 2 the two ends of the core holder is set to 0.7 MPa to form an initial attenuation pressure
 3 pulse between V_1 volume and V_2 volume. The instrument automatically tests the
 4 pressure attenuation and calculates the pulse attenuation permeability values of
 5 different confine pressure and pore pressure points. The effective stress was
 6 continuously increased to 5, 10, 15 and 20 MPa by changing the confine pressure (the
 7 gas pressure was always maintained at 1 MPa). Each pressure point was tested for 30
 8 min to remove the effects of pressurization time and total volume change, and the two
 9 pressure points were maintained at intervals of 30 seconds.

10 **1.3 Multi-fractal theories**

11 The multifractal characteristics of adsorption data (reflecting pore size distribution
 12 heterogeneity) under different test methods can be studied using the box counting
 13 method. When analyzing the volume probability of T_2 spectrum in the interval [a, b],
 14 the scale and measurement need to be determined.

15 The scale and measure expressions are, respectively

$$16 \quad \varepsilon = 2^{-k} L \quad (1)$$

$$17 \quad \overline{\Delta n_i} = \frac{\Delta n_i - \Delta n_{\min}}{\sum_{i=1}^N (\Delta n_i - \Delta n_{\min})} \quad (2)$$

18 where the relative pressure is divided into several boxes of equal length for gas
 19 adsorption, and the box size is represented by ε . The analysis interval of liquid nitrogen
 20 adsorption data ranges from 0 to 0.99 MPa, while the analysis interval of carbon dioxide
 21 adsorption data is 0.01–0.03 MPa. Therefore, the corresponding value of ε differs [26].

22 In the case of scale variation, the water distribution probability in the interval [A,
 23 B] satisfies

$$24 \quad p_i(\varepsilon) \sim \varepsilon^{-a_i} \quad (3)$$

25 where $P_i(\varepsilon)$ is the mass probability function of the i^{th} box, which is used to quantitatively
 26 analyze the distribution of gas adsorption capacity in each box; a_i reflects the local
 27 singular intensity, and a high value represents the smoothness or regularity of the data.
 28 Conversely, the smaller the value, the greater degree of data variation or the stronger

1 the irregularity will be. a_i is related to the area and reflects the probability of the area.

2 After the singularity index is obtained, the object under investigation is divided
3 into a series of subsets, so that the small units in each subset have the same singularity
4 index. Then, the number of units in this subset is calculated and the relationship between
5 the number of units and the scale is defined as

6

$$N_a(\varepsilon) \sim \varepsilon^{-f(a)}, \varepsilon \rightarrow 0 \quad (4)$$

8 where $f(a)$ is the multifractal spectrum, which is the fractal dimension of the subset with
9 the same singularity index. The curve formed by a and $f(a)$ is called the multifractal
10 singular spectrum, which is used to investigate the uneven distribution of the gas
11 adsorption amount, thereby giving more structural information than the single fractal.
12 If the investigated object is multifractal, $f(a)$ generally represents a unimodal image.

13 The expressions of singularity index a and $f(a)$ are respectively [23]

$$a(q) \propto \frac{\sum_{i=1}^{N(\varepsilon)} [u_i(q, \varepsilon) \lg \varepsilon]}{\lg \varepsilon} \quad (5)$$

$$f(a) \propto \frac{\sum_{i=1}^{N(\varepsilon)} [u_i(q, \varepsilon) \lg u_i(q, \varepsilon)]}{\lg \varepsilon} \quad (6)$$

$$u_i(q, \varepsilon) = \frac{p_i^q(\varepsilon)}{\sum_{i=1}^{N(\varepsilon)} p_i^q(\varepsilon)} \quad (7)$$

17 where q is the order of the statistical matrix. When $q \gg 1$, large concentration
18 information or high degree of aggregation is amplified; when $q \ll 1$, small concentration
19 information or low degree of aggregation is amplified. In this study, q is an integer
20 between -10 and 10, and the step size is 1. a and $f(a)$ can be obtained by linear regression
21 of the above two formulas.

22 The $f(a)$ parameters include a_{min} , a_{max} , a_0 , $a_0 - a_{max}$, $a_{min} - a_0$ and A . The symmetry
23 of the singularity spectrum can be expressed as $A = (a_{min} - a_0) / (a_0 - a_{max})$. The left oblique
24 shape indicates that the measured values are affected by large fluctuations, while the

1 right oblique shape indicates that the measured values are affected by small fluctuations
2 [24, 27, 28].

3 Here $a \sim f(a)$ is a set of basic language describing local features of multifractals,
4 called the multifractal spectrum. The other set is $q \sim D(q)$, which is introduced from
5 the perspective of information theory and is termed the generalized fractal dimension.
6 The parameter D_q includes D_{-10} , D_{10} , D_0 , D_1 , D_2 , $D_{-10}-D_{10}$, D_0-D_{10} , and $D_{-10}-D_0$. D_{-10} is
7 influenced by the lowest probability measure areas, whereas D_{10} is influenced by the
8 highest probability measure areas. D_1 is the information dimension and characterizes
9 the degree of disorder in the PSD. A value of D_1 of 1 represents a uniform pore size
10 distribution. D_2 is the correlation dimension and characterizes the association between
11 the measures contained in the multifractal set. D_0-D_{10} and $D_{-10}-D_0$ are the amplitudes of
12 the right and left branches of D_q , which represent the heterogeneity of the high and low
13 probability measure areas, respectively. For the detailed derivation process, please refer
14 to Zhang et al. [26].

15 **2 Results and discussion**

16 **2.1 Multifractal parameter variation of meso-macropore using LPN₂ GA**

17 2.1.1 Multifractal parameter variation

18 According to the LPN₂ GA data for the representative sample W1, the double
19 logarithm diagram of the partition function $x(q, \varepsilon)$ and the size length ε is calculated
20 using Eqs. 2–7 (Fig. 1a). The results show that $\lg[u_i(q, \varepsilon)]$ value has an obvious linear
21 relationship with $\lg(\varepsilon)$, and Fig. 1b shows that $i(q)$ increases strictly monotonically with
22 the increase of the value of q , which shows that the pore size distribution obtained from
23 the LPN₂ GA data has multifractal characteristics. When the q value is less than 0, there
24 is a negative correlation between $\lg[u_i(q, \varepsilon)]$ and $\lg(\varepsilon)$. When the q value is larger than
25 0, there is a positive correlation between $\lg[u_i(q, \varepsilon)]$ and $\lg(\varepsilon)$. This shows that the
26 nano-pore size distribution in those shale samples is concentrated and the pore
27 distribution interval is smaller [26].

28 By combining with Eqs. 5–7, generalized and singular fractal dimension spectra of
29 meso-macropores of all the samples are obtained (Fig. 1c and d). The results show that
30 the $q \sim D(q)$ spectra of all the shale samples are “anti-s-type”, which is another typical

1 feature of shale pore size distribution in line with multifractals. The spectral lines can
2 effectively characterize the pore size distribution complexity at different pore size,
3 which can then reveal the local differences in the whole pore diameter.

4 To investigate the multifractal variation of all samples, generalized fractal
5 parameters of all the samples were calculated (Table 1). As outlined in Section 1.3, the
6 spectrum width $D_{-10}-D_{10}$ represents the variation degree of overall SNDH. $D_{-10}-D_{10}$ of
7 all the samples is 0.69–1.26, which shows that the pore size distribution difference
8 among those samples is relatively large. The $D_{-10}-D_0$ value is greater than that of D_0-
9 D_{10} , indicating that SNDH in the LAPV was more complex than that in the HAPV. In
10 addition, it should be noted that the D_0-D_{10} values of some samples are 0.57, which
11 indicates that SNDH in the HAPV is consistent, and the overall SNDH is controlled by
12 the pore distribution in the LAPV.

13 It can be seen from Fig. 2a that there is no obvious correlation between D_{-10} and
14 D_{10} , which is related to the consistent values of the two parameters amongst the samples.
15 In contrast to the results shown in Fig. 2a, $D_{-10}-D_0$ and D_0-D_{10} are positively correlated
16 (Fig. 2b), although there is no clear linear relationship between them. Fig. 2c shows that
17 $D_{-10}-D_{10}$ increases linearly with the increase of $D_{-10}-D_0$. Compared with $D_{-10}-D_0$, the
18 linear relationship between D_0-D_{10} and $D_{-10}-D_{10}$ is not remarkable. Considering the two
19 factors, the overall SNDH is affected by the LAPV.

20 2.1.2 Influencing factors of multifractal parameters

21 Based on the data in Table 1, the influencing factors of nanopore multifractal
22 parameters are examined by integrating the shale maturity, mineral composition,
23 organic matter content and pore structure parameters (Fig. 3). Figs.3a–c show that two
24 fractal parameters $D_{-10}-D_0$ and D_0-D_{10} are weakly correlated with variation in $R_{o,max}$
25 and brittle mineral content. Compared with Fig. 3d and 3e, it can be concluded that the
26 fractal parameter has a strong correlation with the pore structure, which is manifested
27 through the $D_{-10}-D_0$ value decreasing as the pore volume increases, implying that the
28 SNDH of LAPV is weakened. The reason for this is that the increase of total pore
29 volume, accompanied by the decrease of mesopore volume and increase of micro-pore
30 volume, which causes the pore interval in this range change from HAPV to LAPV,

1 results in the distribution of low-value area of pore volume tending to be uniform,
2 subsequently leading to the decrease of $D_{-10}-D_0$ value [26].

3 2.1.3 Pore size distribution heterogeneity obtained from different fractal models

4 By using Table 4 in Yan et al. [16], the correlation of fractal dimension calculated
5 by different fractal models is compared. The above study shows that D_1 calculated using
6 the Frenkel-Halsey-Hill (FHH) model reflects the SNDH of pores >10 nm, and D_2
7 reflects the SNDH of pores between 2 and 10 nm [26]. Fig. 4 shows that there is no clear
8 correlation between the single fractal parameter D_1 and the two multifractal parameters,
9 while the single fractal parameter D_2 and the multifractal parameters show a clear
10 negative correlation that implies that the SNDH of LAPV is weakened. Zhu et al.
11 showed that D_2 (relative pressure corresponding to 0–0.5) characterize adsorption pore
12 surface heterogeneity, and D_1 (relative pressure corresponding to 0.5–1) can reveal the
13 adsorption pore volume heterogeneity [27]. The increase of the D_2 value shows that pore
14 surface heterogeneity increases, and the $D_{-10}-D_0$ value decreases shows that the SNDH
15 of LAPV is weakened. In conclusion, the fractal dimension values calculated using the
16 two fractal models are different. The result shows that the multifractal model represents
17 SNDH in different pore size intervals, and the calculated values are relative values. The
18 single fractal model represents the overall heterogeneity of the pore distribution, which
19 its absolute value is calculated.

20 **2.2 Multifractal parameter variation of micro-pore by using LPCO₂ GA**

21 2.2.1 Multifractal parameter variation

22 According to the LPCO₂ GA data of the representative sample W1, the double
23 logarithm diagram of the partition function $x(q,\varepsilon)$ and the size length ε is calculated (Fig.
24 5a). The results show that $\lg[u_i(q, \varepsilon)]$ value has an obvious linear relationship between
25 $\lg(\varepsilon)$, and Fig. 5b shows that $i(q)$ increases strictly monotonically with the increase of
26 q value, which indicates that the pore size distribution obtained from LPCO₂ GA data
27 has multifractal characteristics. When the q value is less than 0, there is a negative
28 correlation between $\lg[u_i(q, \varepsilon)]$ and $\lg(\varepsilon)$. When q value is greater than 0, a positive
29 correlation is observed between $\lg[u_i(q, \varepsilon)]$ and $\lg(\varepsilon)$. This shows that the micro-pore
30 size distribution in those shale samples is concentrated and the pore distribution interval

1 is smaller.

2 Fig. 5c shows that the $q\sim D(q)$ spectrum of all shale samples is “Anti-S” type, which
3 is also typical feature of multifractal distribution of shale pore-fracture system. The
4 spectrum can effectively represent the complexity of PSDH in different pore stage.
5 Compared with Fig. 1c it can be seen that the singular spectral width of highly mature
6 shale samples is different, and the width of the left branch is larger, which indicates that
7 PSDH is stronger and is related to the HAPV.

8 Table 3 shows that $D_{-10}-D_0$ value is less than D_0-D_{10} , meaning that the SNDH of
9 HAPV is stronger than that of LAPV in micro-pores, which implies that the HAPV
10 controls the micro-pore size distribution heterogeneity. Compared with Fig. 3, the
11 LAPV in the range of 2–100nm controls SNDH within that pore range, indicating that
12 there are obvious differences in the overall distribution of pores <2 nm and 2–100 nm.
13 The $D_{-0}-D_1$ value of all the samples is 0.50–0.72, which is less than that of the value of
14 2–1200 nm pores, indicating that the overall distribution of pores of 2–100 nm is more
15 complex.

16 An analysis of micro-pore multifractal parameters shows that D_{-10} and D_{10} have an
17 obvious linear positive correlation, showing that the minimum and maximum pore
18 volume distribution characteristics are synchronized (Fig. 6a). As shown in Fig. 6b, $D_{-10}-D_0$
19 decrease linearly with the increase of D_0-D_{10} , indicating the distribution
20 heterogeneity of LAPV decreases with the increased HAPV heterogeneity. Fig. 6c
21 shows that $D_{-10}-D_{10}$ and $D_{-10}-D_0$ as well as D_0-D_{10} are linearly negatively and positively
22 correlated, respectively, and the linear fit of the latter is significantly higher than that of
23 the former. In summary, the heterogeneity of full-scale pore distribution is controlled
24 by HAPV.

25 2.2.2 Influencing factors of multifractal parameters

26 The influencing factors of the micro-pore multifractal parameters are examined by
27 integrating the shale maturity, mineral composition, organic matter content and pore
28 structure parameters (Fig. 7). The results show that the micro-pore single fractal
29 parameters $D_{-10}-D_0$ and D_0-D_{10} have weak correlations with various influencing factors.
30 Compared to Fig. 3b, Fig. 7b shows that there is no obvious relationship between the

1 two fractal parameters and the micro-pore volume. The reason being that the pore size
2 range corresponding to the low/high pore volume area is not constant. Yan et al. [16]
3 found that there are three peak intervals in the micro-pore distribution of all of the
4 analysed shale samples, which are 0.38 nm, 0.5 nm and 0.85 nm. With the variation in
5 micro-pore volume, the three-peaks of pores corresponding to the high pore volume
6 area are relatively complicated, so the correlation among the two multifractal
7 parameters and pore volume is weak. Zhang et al. [26] analyzed the micro-pore
8 multifractal characteristics of middle and high rank coal samples from western Guizhou
9 and eastern Yunnan, which showed that the micropore [0.72, 0.94] distribution variation
10 was an important interval leading to variation in the micro-pore multifractal. Therefore,
11 the focus of further research should be on exploring the micro-pore interval that affects
12 the distribution of high value area of micropores.

13 Fig.8 shows that there is no correlation between the multifractal parameters of
14 micro-pores and meso-pores, which also explains the strong heterogeneity of shale pore
15 size distribution.

16 **2.3 Dynamic variation of porosity and permeability under the influence of pore** 17 **size distribution heterogeneity**

18 Four representative samples, capturing the variation in the physical characteristics
19 within the reservoir, were selected for further analysis based on their initial porosity,
20 permeability and pore distribution. Fig. 9 shows that the initial permeability of all the
21 samples is different, with the initial permeability of samples W7 and W9 both exceeding
22 0.1 mD while sample W1 reaches 0.29 mD. The overburden porosity results are also
23 shown in Fig. 9. The results shows that the permeability of all samples decreased
24 exponentially when the effective stress increased from 0 to 25 MPa, and the maximum
25 R^2 reached 0.99.

26 In addition, permeability varies in stages with the increase in effective stress.
27 When the effective stress is less than 15 MPa, permeability (including pore volume)
28 decreases rapidly with the increase in stress, and the permeability is in the stage of rapid
29 decline with the average decline of all the samples in the region of 86%. However,
30 when the effective stress is greater than 15 MPa, the permeability is in a slow decline

1 stage and is fairly stable. Initially, the pore volume has not been compressed and so the
2 compressible space is large. The pore space therefore has high compressibility, which
3 leads to the rapid decline of coal permeability during this early phase of applying stress.
4 With the continuous increase in stress, the compressible space of meso-macro-pores is
5 reduced the compressibility coefficient decreases, which results in the coal permeability
6 being stable during this stage.

7 Fig. 9 shows that the permeability loss rate of all samples is between 0.76 and 0.98.
8 Among them, the permeability of sample W1 is the most sensitive to pressure, and the
9 permeability loss rate can reach 98%. In the other three samples, the macro-pores are
10 not developed. As a result, the compressible space in the high stress stage is provided
11 by micro-pores. However, the meso-pores influence the permeability variation, so the
12 permeability loss rate is relatively higher.

13 According to the multifractal parameters and permeability damage rate of samples,
14 $D_{-10}-D_0$ and $D_{-10}-D_{10}$ has obvious linear positive correlation with permeability damage
15 rate (Fig. 10). According to above two parameters, the relationship between D_0-D_{10} and
16 permeability damage rate is not obvious, which implies that the stronger pore
17 heterogeneity, the more obvious the damage effect of stress on permeability is. This
18 conclusion is also consistent with the previous results.

19 Table 4 shows that with the exception of sample W10, the initial diffusion
20 coefficients of samples are $2.14-2.41 \times 10^{-6} \text{ cm}^2/\text{s}$ and are fairly uniform. The diffusion
21 coefficient increases with the increase of confining stress, which indicates that the
22 pressure has a positive effect on the diffusion coefficient. In order to systematically
23 explain the influence of pressure on the diffusion coefficient, the diffusion coefficient
24 variation coefficient D_{20}/D_{10} is introduced to quantitatively characterize the diffusion
25 coefficient variation under pressure. The calculation results indicates that the value is
26 between 2.34 and 8.21. Relevant literature shows that micro-pores play an important
27 role in controlling methane diffusion coefficient. In Section 2.2, the relationship
28 between the multifractal parameters and diffusion coefficient variation is discussed (Fig.
29 11). It indicates that the variation rate of diffusion coefficient decreases with the
30 increase in the multifractal parameters, indicating that the stronger the heterogeneity of

1 pore distribution, the weaker the "transformation effect" of stress on the diffusion
2 coefficient (Table 4) [28-30].

3 **Conclusions**

4 According to low temperature liquid nitrogen and carbon dioxide tests (LPN₂/CO₂
5 GA) of organic rich shale in Qinshui Basin, the multifractal dimension variation of
6 micro-pores (<2 nm) and mesopores (2–100 nm) was studied, and the multifractal
7 factors that affect the distribution of nanopores determined. In addition, the differences
8 between single fractal and multifractal results were compared. Based on this, the
9 dynamic variation of porosity and permeability under the constraints of nanopore
10 structure were discussed from the perspective of multifractal variation. The conclusions
11 are as follows:

12 1) The distribution of nanopores in organic rich shale is a typical multifractal
13 feature. However, there are obvious differences in the multifractal parameters and
14 influencing factors between micropores and mesopores.

15 2) The SNDH in the HAPV of 2–100nm in shale samples tends to be consistent,
16 and the overall SNDH is controlled by the SNDH in the LAPV. The SNDH in the LAPV
17 is affected by the distribution of pores with diameter within 2 and 10 nm.

18 3) The single fractal dimension D_2 calculated using the FHH model has a negative
19 correlation with the multifractal parameters, which implies that the distribution
20 heterogeneity of the LAPV gradually decreases with the increase of D_2 , indicating that
21 the physical meaning of the two models is clearly different.

22 4) In contrast to meso-macropores, the heterogeneity of the micro-pore size
23 distribution is controlled by HAPV, and the correlation between the multifractal
24 variation and pore volume is weak. Moreover, there is no correlation between micro-
25 pore and mesoporous multifractal parameters, which shows the strong heterogeneity of
26 shale pore distribution.

27 5) Multifractal variation of pores controls the porosity and permeability variation
28 and diffusion process of shale reservoirs. $D_{-10}-D_0$ and $D_{-10}-D_{10}$ has an obvious linear
29 positive correlation with permeability variation rate, indicating that the stronger the
30 pore heterogeneity, the greater the damage effect of confining stress on permeability.

1 Also, the stronger the heterogeneity of the micro-pore size distribution, the weaker the
2 "reconstruction effect" of stress on the diffusion coefficient.

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Table 1 Sample tests and information.

Sample No.	DP-P tests		DP-F tests	
	Information (cm)	Confine pressure (MPa)	Information (cm)	Confine pressure (MPa)
W1	2.62*2.54	5/10/15/20	1.52*2.54	10/15/20
W7	2.58*2.54	5/10/15/20	1.52*2.54	10/15/20
W9	2.60*2.54	5/10/15/20	1.52*2.54	10/15/20
W10	2.58*2.54	5/10/15/20	1.52*2.54	10/15/20

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Table 2 Generalized multifractal parameters from LPN₂ GA tests.

Sample	D_{-10}	D_{10}	$D_0 - D_2$	$D_{-10} - D_0$	$D_0 - D_{10}$	$D_{-10} - D_{10}$
W1	0.43	1.58	0.36	0.58	0.57	1.15
W2	0.68	1.47	0.17	0.47	0.32	0.79
W3	0.68	1.51	0.17	0.51	0.32	0.83
W4	0.67	1.44	0.18	0.44	0.33	0.77
W5	0.43	1.65	0.36	0.65	0.57	1.22
W6	0.43	1.69	0.36	0.69	0.57	1.26
W7	0.43	1.63	0.36	0.63	0.57	1.20
W8	0.43	1.67	0.36	0.67	0.57	1.24
W9	0.43	1.63	0.36	0.63	0.57	1.20
W10	0.67	1.37	0.18	0.37	0.33	0.69
W11	0.67	1.46	0.18	0.46	0.33	0.78
W12	0.68	1.47	0.18	0.47	0.32	0.80

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Table 3 Generalized multifractal parameters from LPCO₂ GA tests.

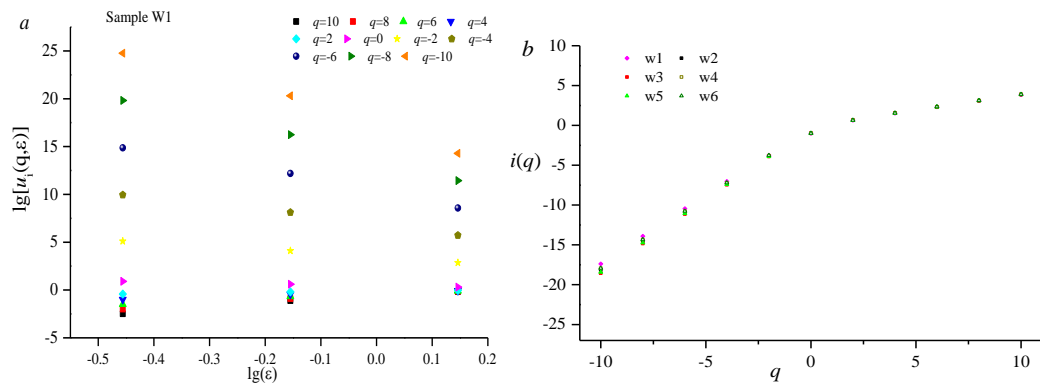
Sample No.	D_{-10}	D_{10}	$D_0 - D_2$	$D_{-10} - D_0$	$D_0 - D_{10}$	$D_{-10} - D_{10}$
W1	0.48	1.18	0.24	0.18	0.52	0.71
W2	0.59	1.23	0.16	0.23	0.41	0.64
W3	0.68	1.23	0.11	0.23	0.32	0.55
W4	0.67	1.21	0.13	0.21	0.33	0.54
W5	0.51	1.18	0.23	0.18	0.49	0.67

W6	0.61	1.21	0.16	0.21	0.39	0.60
W7	0.74	1.26	0.09	0.26	0.26	0.53
W8	0.48	1.20	0.25	0.20	0.52	0.72
W9	0.67	1.22	0.12	0.22	0.33	0.55
W10	0.75	1.25	0.09	0.25	0.25	0.50
W11	0.56	1.19	0.19	0.19	0.44	0.63
W12	0.61	1.21	0.16	0.21	0.39	0.61

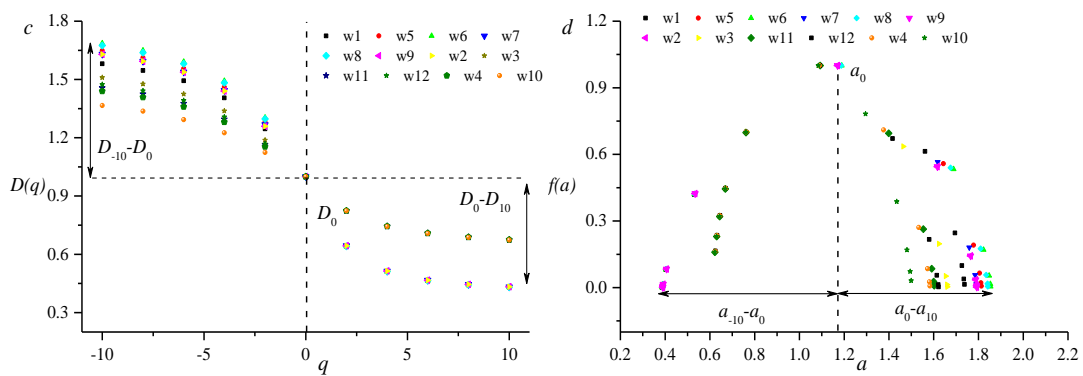
1 Table 4 Diffusion coefficient variation of typical samples under different confining pressures.

Pressure (MPa)	Sample W1	Sample W7	Sample W9	Sample W10
10	2.15E-06	2.14E-06	2.41E-06	6.80E-08
15	3.45E-06	3.39E-06	2.25E-05	1.35E-07
20	4.85E-06	6.58E-06	8.89E-05	5.62E-07

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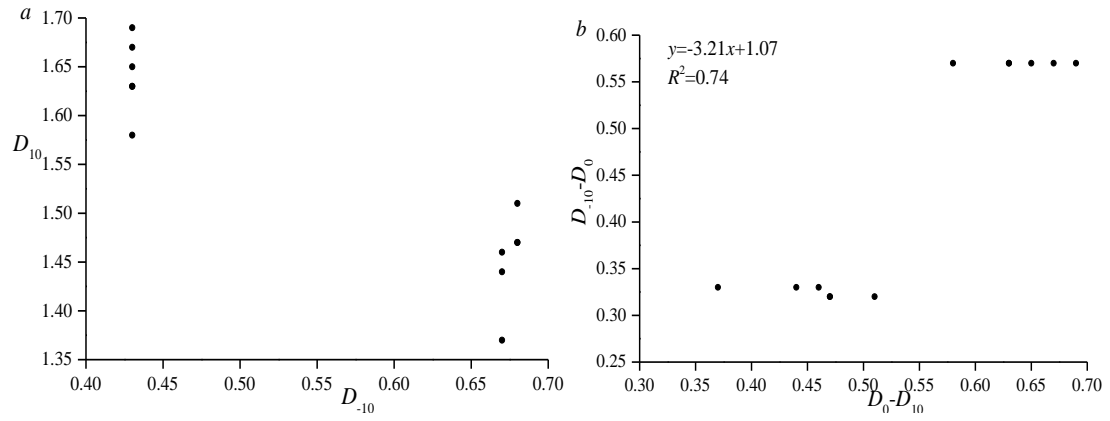


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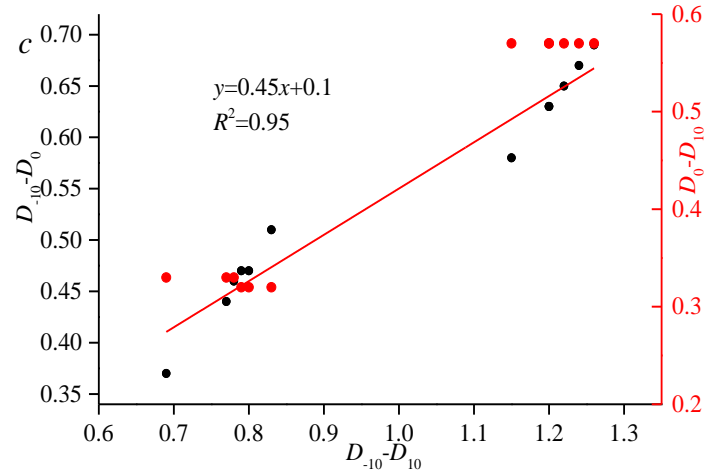


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4 Fig.1 Characteristics of generalized multifractal curves by using LPN₂ GA tests. a, relationship
5 between $\lg(\varepsilon)$ and $\lg[u_i(q, \varepsilon)]$; b, relationship between q and $i(q)$; c, relationship between q and
6 $D(q)$; d, relationship between a and $f(a)$.



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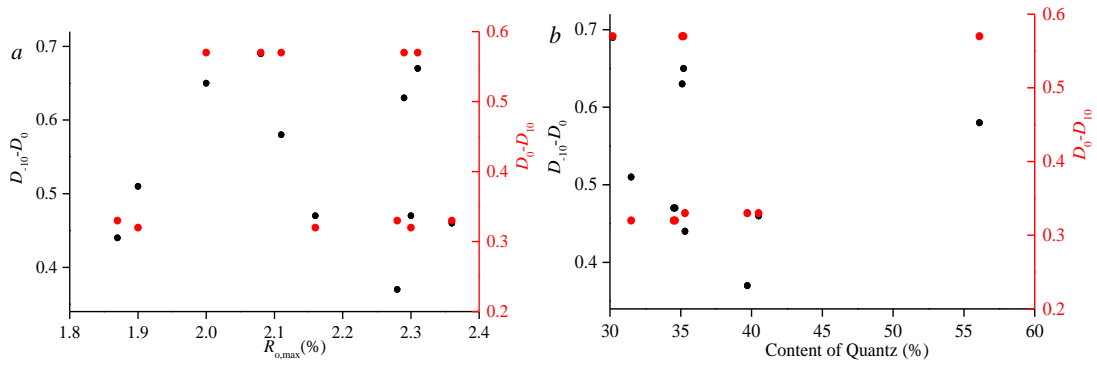
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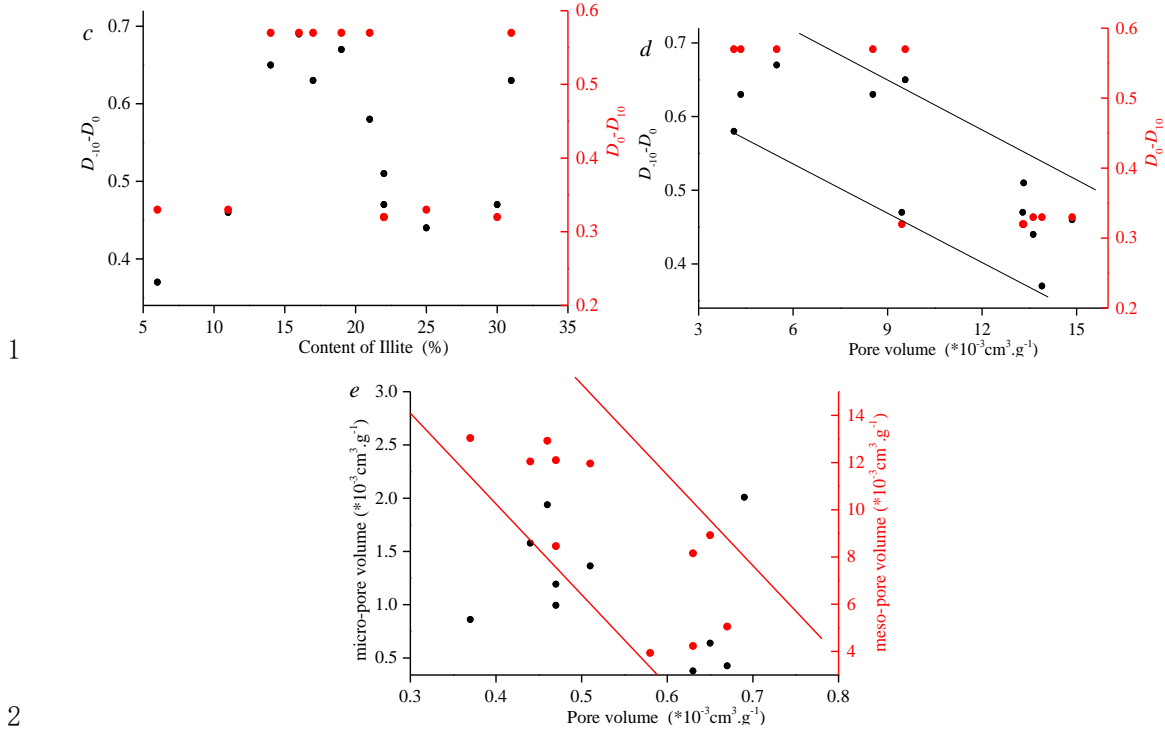
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Fig. 2 Correlation analysis of generalized fractal parameters by using LPN₂ GA tests. a, relationship between D_{10} and D_{-10} ; b, relationship between $D_{-10}-D_0$ and D_0-D_{10} ; c, relationship between $D_{-10}-D_{10}$ and $D_{-10}-D_0$.



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Fig. 3 Correlation analysis of generalized fractal parameters with maturity and pore structure parameters by using LPN₂ GA tests. a, relationship between $R_{0, \max}$ and $D_{-10}-D_0$; b, relationship between quartz content and $D_{-10}-D_0$, D_0-D_{10} ; c, relationship between illite content and $D_{-10}-D_0$, D_0-D_{10} ; d, relationship between pore volume content and $D_{-10}-D_0$, D_0-D_{10} ; e, relationship between pore volume content and micro-pore/meso-pore volume.

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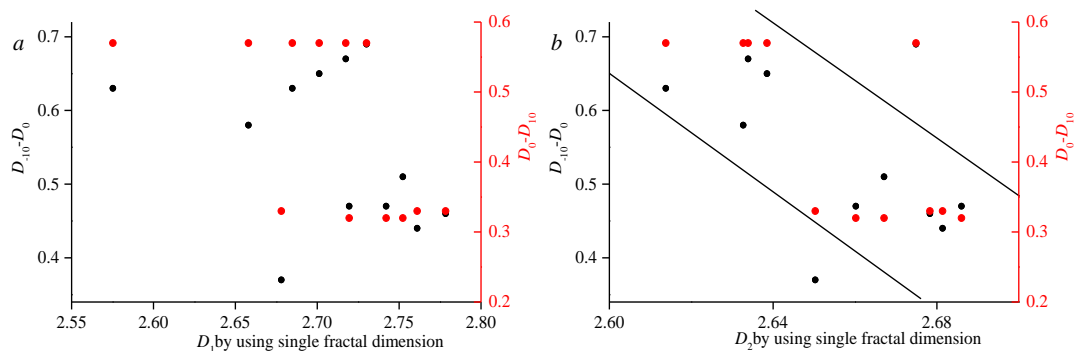
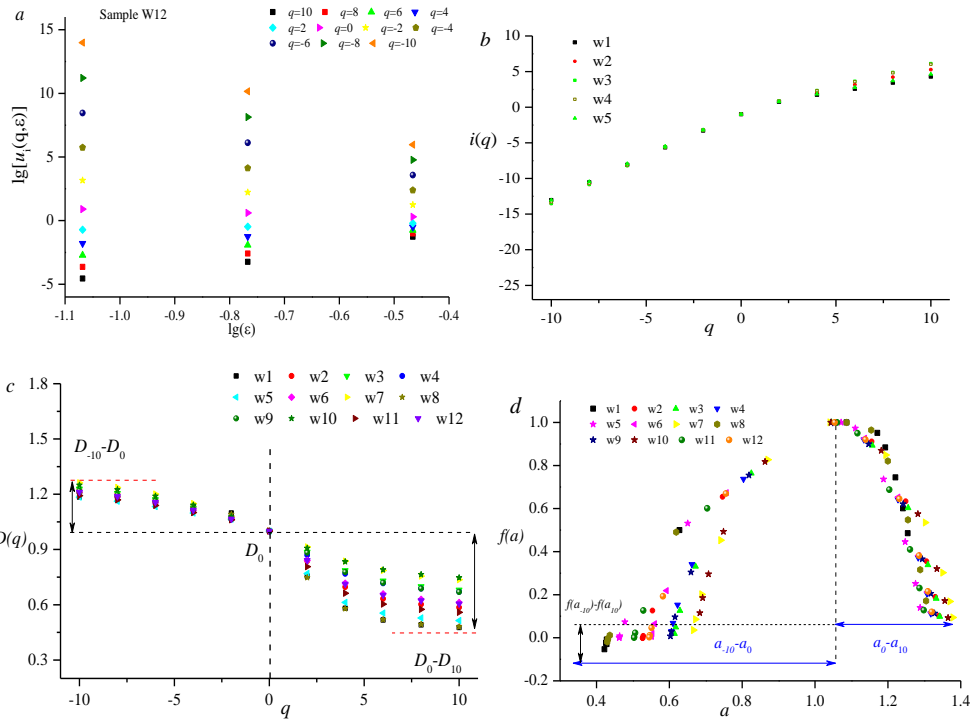


Fig. 4 Correlation analysis of fractal parameters by combining single with multifractal calculations. a, relationship between D_1 by using single fractal dimension and $D_{-10}-D_0$; b, relationship between D_2 by using single fractal dimension and D_0-D_{10} .



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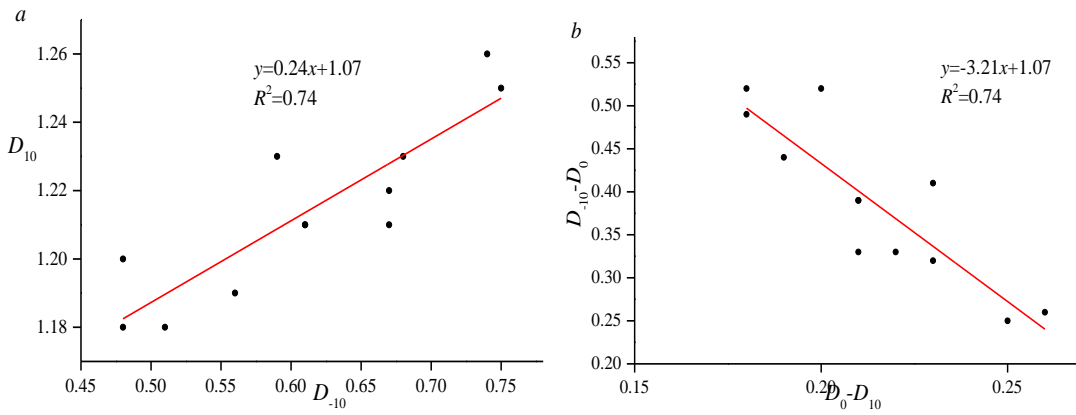
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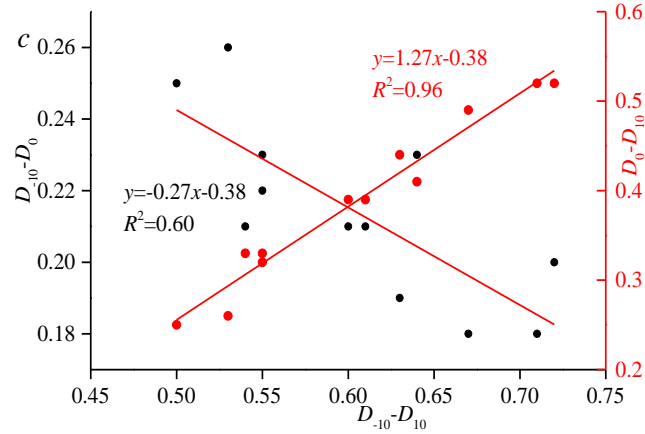
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Fig. 5 Characteristics of generalized multifractal curves by using LPCO₂ GA tests. a, relationship between $\lg(\varepsilon)$ and $\lg[u_i(q, \varepsilon)]$ in sample W12; b, relationship between q and $i(q)$ of sample W12; c, relationship between q and $D(q)$ of all the samples; d, relationship between a and $f(a)$ of all the samples.



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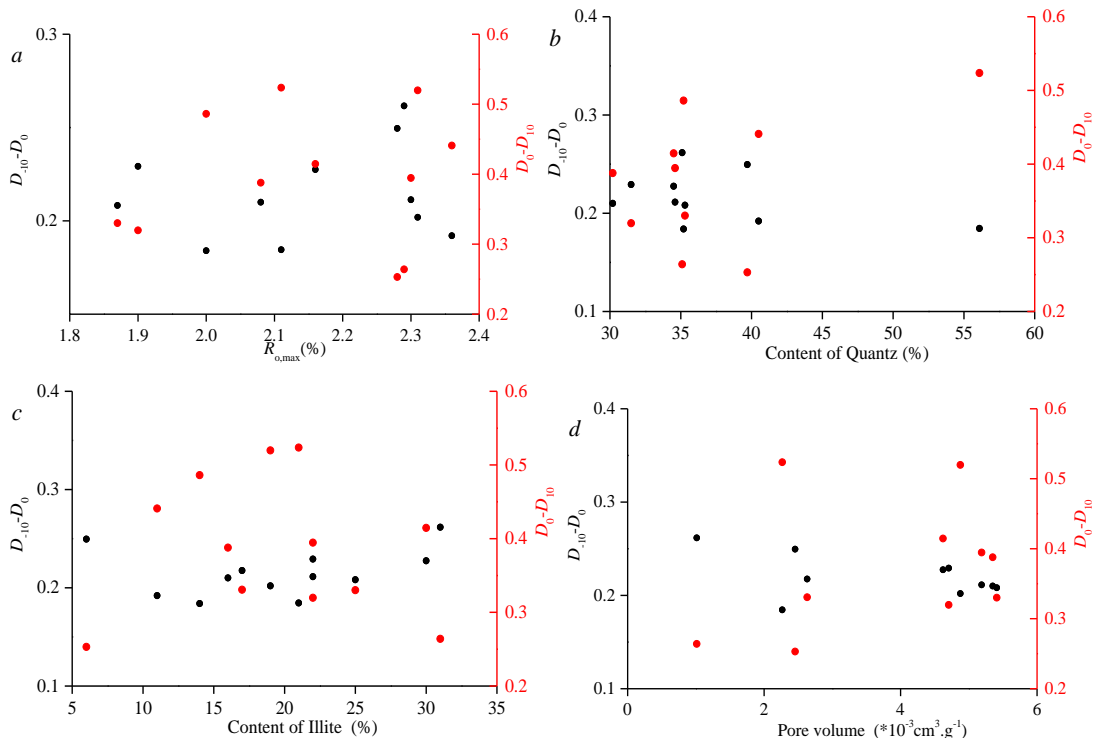
Fig. 6 Correlation analysis of generalized fractal parameters by using LPCO₂ GA. a, relationship

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between D_{10} and D_{-10} ; b, relationship between $D_{-10}-D_0$ and D_0-D_{10} ; c, relationship between $D_{-10}-$

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D_{10} and $D_{-10}-D_0$.



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Fig. 7 Correlation analysis of generalized fractal parameters, maturity and pore structure

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parameters by using LPCO₂ GA tests. a, relationship between $R_{o, \max}$ and $D_{-10}-D_0$; b, relationship

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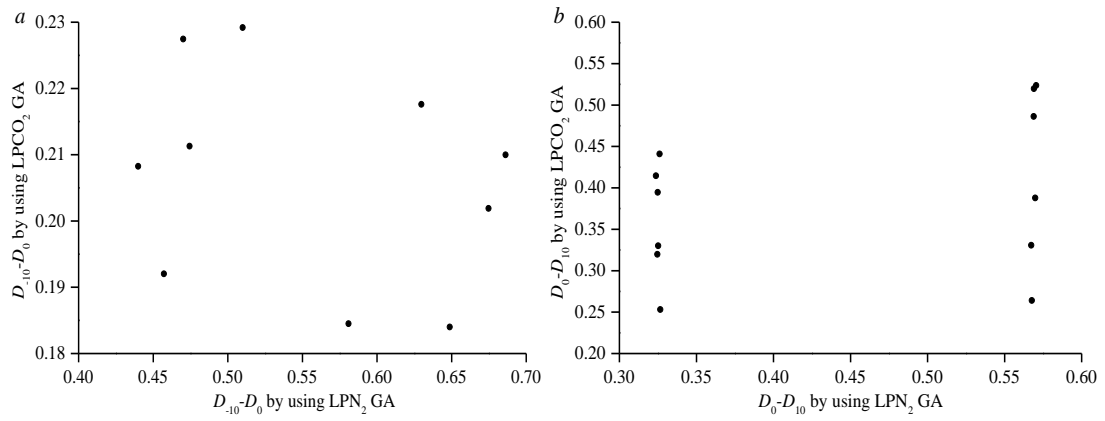
between quartz content and $D_{-10}-D_0$, D_0-D_{10} ; c, relationship between illite content and $D_{-10}-D_0$, D_0-

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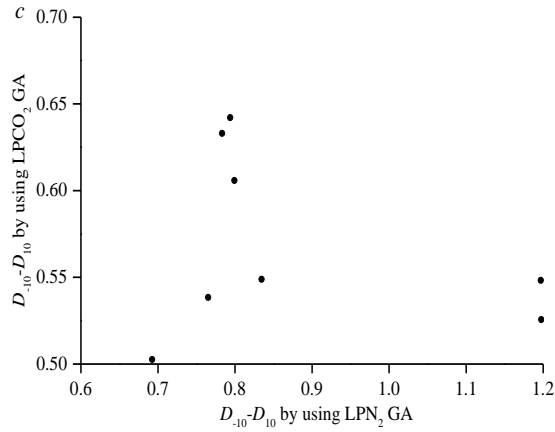
D_{10} ; d, relationship between pore volume content and $D_{-10}-D_0$, D_0-D_{10} ; e, relationship between

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pore volume content and micro-pore/meso-pore volume.

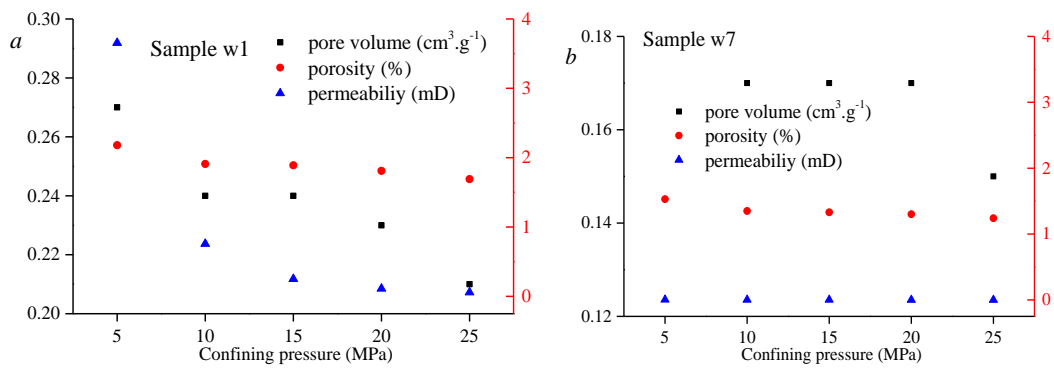


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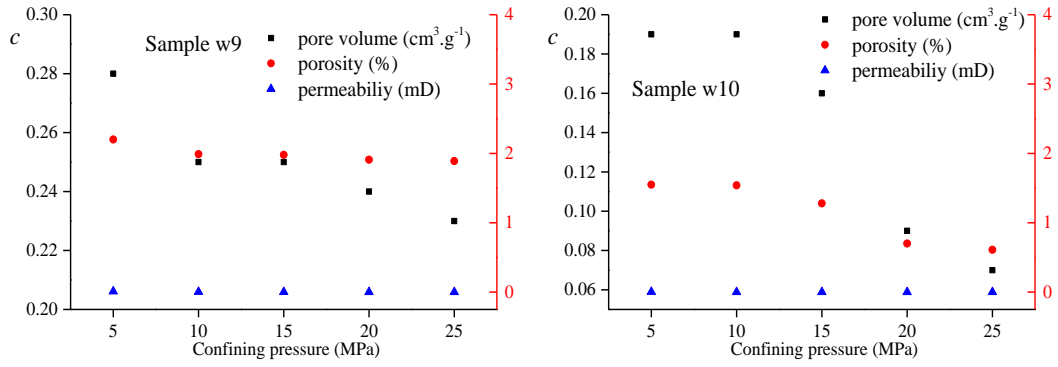


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3 Fig.8 Correlation analysis of generalized fractal parameters by using LPCO₂ and N₂ GA tests. a,
 4 relationship between $D_{-10}-D_0$ by using LPCO₂ GA and $D_{-10}-D_0$ by using LPN₂ GA; b, relationship
 5 between D_0-D_{10} by using LPCO₂ GA and D_0-D_{10} by using LPN₂ GA; c, relationship between $D_{-10}-$
 6 D_{10} by using LPCO₂ GA and $D_{-10}-D_{10}$ by using LPN₂ GA.

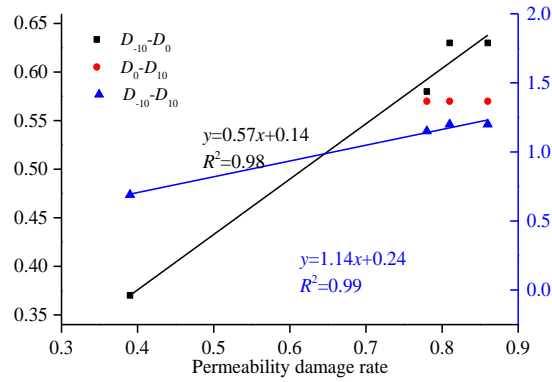


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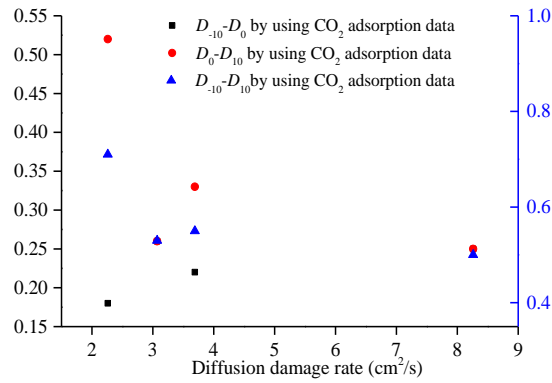
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Fig. 9 Dynamic parameter variation of porosity and permeability under different confining pressures for a, sample W1; b, sample W7; c, sample W9; d, sample W10.



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Fig. 10 The relationship between multifractal parameters and permeability damage rate based on LPN₂ GA tests.



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Figure 11 The relationship between multifractal parameters and damage rate of diffusion coefficient based on LPCO₂ GA tests.