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Title: Insights into fractures and minerals in subbituminous and bituminous coals by X-ray μ -CT and FESEM-EDS

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Abstract: Fractures are the main pathways for fluid transport, which constrain the coalbed methane (CBM) reservoir permeability. Permeability is one of the key petrophysical parameters for CBM production. The fractures in the low and middle rank coals usually filled with minerals, which can significantly reduce the reservoir permeability. In this study, X-ray μ -CT combined with field emission scanning electron microscopy (FESEM) together with energy dispersive spectrometry (EDS) were used to quantitatively evaluate the features of fractures and minerals of FK sample (R_o, \max 0.65%) and ML sample (R_o, \max 1.49%) including morphology, complexity, volume and 3D structure, and its impacts on permeability. The results showed that fractures are well developed as observed in the samples FK and ML by the 2D cross section of X-ray μ -CT. Dolomite and kaolinite are common in the samples FK and ML as confirmed by FESEM images with EDS. Moreover, the 3D fracture structure including the open fracture and mineral filled fracture were reconstructed. The quantitative structural parameters (e.g. length, width, area and volume) of the fractures for these two samples were acquired by the commercial software Avizo 9.0.1. The middle rank coal of sample ML has larger fracture density (181 fractures per mm^3) than that of the low rank coal of sample FK (104 fractures per mm^3). Based on the reconstructed 2D and 3D fracture structure, the fracture complexity was evaluated through fractal dimension. And the impacts of mineralization in coals on petrophysical properties were evaluated, which show that the connectivity of fractures, porosity and permeability were greatly improved after demineralization. Therefore, this study may have implications for enhancing CBM recovery through demineralization.

Highlights

- Fractures and minerals of two Chinese coals were reconstructed by X-ray μ -CT.
- The open and mineralized fractures were quantitatively analyzed.
- Discrepancy of fractures and minerals in medium and low rank coals was evaluated.
- The importance of demineralization to enhance CBM production was illustrated.

1 **Insights into fractures and minerals in subbituminous and** 2 **bituminous coals by FESEM-EDS and X-ray μ -CT**

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12 parameters for CBM production. The fractures in the low and middle rank coals usually filled
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14 μ -CT combined with field emission scanning electron microscopy (FESEM) together with
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16 fractures and minerals of FK sample ($R_{o,max}$ 0.65%) and ML sample ($R_{o,max}$ 1.49%) including
17 morphology, complexity, volume and 3D structure, and its impacts on permeability. The
18 results showed that fractures are well developed as observed in the samples FK and ML by
19 the 2D cross section of X-ray μ -CT. Dolomite and kaolinite are common in the samples FK
20 and ML as confirmed by FESEM images with EDS. Moreover, the 3D fracture structure
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22 structural parameters (e.g. length, width, area and volume) of the fractures for these two

23 samples were acquired by the commercial software Avizo 9.0.1. The middle rank coal of
24 sample ML has larger fracture density (181 fractures per mm³) than that of the low rank coal
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26 structure, the fracture complexity was evaluated through fractal dimension. And the impacts
27 of mineralization in coals on petrophysical properties were evaluated, which show that the
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32

33 **1. Introduction**

34 Coalbed methane (CBM) is a form of unconventional natural gas extracted from coal
35 reservoirs. In the past few decades, CBM has become a substantial source of clean energy, of
36 which the exploration and development have been successfully carried out in many countries
37 such as US, Australia, and Canada [1]. China has rich coal resources and abundant CBM
38 resources. The CBM exploration and development in China has been developing rapidly in
39 recent years. Statistically, the CBM resources below the depth of 2000 meters reach
40 $36.81 \times 10^{12} \text{ m}^3$ and over 16000 CBM wells including vertical and horizontal wells were drilled
41 in China with an annual CBM output of $70.2 \times 10^8 \text{ m}^3$ [2].

42 During the process of CBM production, gas flows through the fractures to the wellbore in
43 response to the pressure gradient [3]. When the reservoir pressure drops below the critical
44 CH₄ desorption pressure, the adsorbed CH₄ gradually diffuses from the micropores in coal

45 matrix to the macropores or fractures [4, 5] Therefore, the fractures are the main pathways for
46 gas and water flow. Normally the in-situ fractures were filling with minerals, which have
47 significant impacts on the fracture properties, and thus on fracture permeability. For instances,
48 minerals that include inherent or extraneous minerals (e.g. quartz, clay minerals, feldspars,
49 carbonates, and sulphide minerals) are often developed in the coal fractures [2]. The inherent
50 or extraneous minerals present differently in coals. Inherent minerals are normally syngenetic,
51 which are generally enclosed by or embraced in coal matrix, while extraneous mineral are
52 filling in fractures [3]. Therefore, the extraneous minerals can substantially reduce the
53 reservoir permeability and affects the process of gas production [4, 5]. The fracture
54 morphology, connectivity and density can be system acquired with multiple techniques (e.g.
55 optical microscope, AFM, FESEM, CT and NMRI) [5-7]. CT as a non-destructive technique
56 has been widely used to investigate the 3D structure of coals, which can obtain the specific
57 fracture parameters including fracture length and surface area [8]. Although the X-ray μ -CT
58 can distinguish the fractures, minerals and coal matrix, the specific minerals (e.g. quartz and
59 feldspars,) cannot be detected [9, 10]. In this study, an X-ray μ -CT combined with field
60 emission scanning electron microscopy and energy dispersive spectrometry will be adopted to
61 acquire the 3D structure information of fractures and minerals in coals.

62 Fracture complexity and irregularity can be described with the fractal geometry [11-14].
63 Fractal dimension (D) is an important characteristic parameter of fractal geometry, which can
64 quantitatively reflect the fracture complex in coals. Therefore, the fractal dimension may
65 indicate the CBM adsorption and flow capabilities [15]. The fractal dimension of fractures in
66 coals can be acquired by scanning electron microscopy (SEM), nuclear magnetic resonance

67 (NMR), mercury intrusion porosimetry and low-temperature N₂ adsorption/desorption
68 isotherm [16-18]. The X-ray μ-CT scanning can capture the detailed fracture structure in both
69 2D slices and 3D reconstruction patterns.

70 In this study, X-ray μ-CT together with FESEM-EDS scanning will be used to accurately
71 characterize the multiscale characteristics of 2D and 3D fracture structure and minerals
72 performance in the low rank coal sample FK and medium rank coal sample ML. Herein, the
73 fractal dimension of fractures in 2D and 3D view will be calculated, and the original and
74 de-mineralized coals will be compared to evaluate the effects of mineralization on fracture
75 properties. Firstly, the morphology of fractures and minerals were identified in 2D
76 FESEM–EDS. Secondly, X-ray μ-CT scanning was combined to accurately characterize the
77 multiscale characteristics of 3D fractures and minerals. And the fracture complexity and
78 irregularity were described with the fractal geometry. Finally, the impacts of fractures and
79 minerals performance on petrophysical properties including porosity and permeability were
80 detail discussed. This study provided a possible way to enhance CBM recovery through
81 demineralization.

82 **2. Experiments and methods**

83 2.1. Coal basic information

84 One subbituminous coal from Fukang (FK) colliery in the southern Junngar Basin and another
85 bituminous coal from Malan (ML) colliery in the northwest Qinshui Basin were collected,
86 which were drilled for cylindrical cores with 25mm in diameter and 50mm in length
87 approximately. With Leitz MPV-3 photometer microscope, maceral composition analysis and
88 the maximum vitrinite reflectance ($R_{o, \max}$) were performed on polished slabs as previous

89 research [7]. The proximate analysis was conducted on the Automatic Proximate Analyzer
90 5E-6600. The results as listed in Table 1 indicated that FK sample was a subbituminous coal
91 with $R_{o, \max}$ of 0.65% that belong to low rank coal and the ML sample was a bituminous coal
92 with $R_{o, \max}$ of 1.49% that belong to medium rank coal.

93 2.2. X-ray μ -CT scanning and FESEM-EDS

94 The CT measurements of the FK and ML samples were performed on the nanoVoxel-3502E
95 microfocus X-ray CT scan equipment. The cylindrical core samples (9.77mm in diameter and
96 15mm in length for sample FK, and 5.64mm in diameter and 10mm in length for sample ML)
97 was placed perpendicular to the sample couch, and was aligned in the center of the scanner
98 field of view. The X-ray source voltage and current were set to be 120KV, 36.67 μ A and
99 70KV, 40 μ A for the FK and ML samples, respectively. The CT equipment is able to produce
100 a voxel dataset containing a series of 2D slices with the resolution of 500 nm. The detector
101 has a resolution of 2048 \times 2048 pixels, in which each pixel represents an area of
102 $\sim 3.98 \times 3.98 \mu\text{m}^2$ for sample FK and $2 \times 2 \mu\text{m}^2$ for sample ML respectively. The spatial
103 resolution of sample FK and ML are approximately 3.98 μm and 2 μm , with which most
104 fractures can be distinguished.

105 Field emission scanning electron microscope (FESEM) with an energy dispersive
106 spectrometer and X-ray diffractometer were used to aid microfocus X-ray CT method and
107 recognize the mineral matter [19]. Coal samples were polished into lumps approximately 10
108 mm (length) \times 10 mm (width) \times 7 mm (height) with spluttered thin gold coating. The fractures
109 and mineral band in the range of 3nm-50 μm can be measured using the FEI Quanta FEG 450
110 (FEI, America) and EDAX TEAM EDS.

111 2.3. 3D morphology reconstruction

112 For a typical CT grayscale image, each pixel corresponds to a grayscale that equals to specific
113 CT number. Normally, minerals with high CT number (approximately 3000 HU) present
114 white, whereas the fractures with low CT number (<600HU) show black, and coal matrix
115 with medium CT number (100-1600HU) correspond to a certain range of gray [8]. Then 3D
116 threshold segmentation (watershed segmentation), the process of partitioning of the grayscale
117 image into unique phases, was applied according to CT number. After that the partitions were
118 colored and the 3D model of each partition was established. The image data from the X-ray
119 μ -CT was imported into Avizo 9.0.1, the commercial software for 3D visualization,
120 reconstruction and analysis. The representative elemental volume (REV) under the subvolume
121 extraction is important due to the computational capability of the computers and the coal
122 strong heterogeneity [20]. The size of digital core REVs for sample FK and ML was
123 determined to be $2000 \mu\text{m} \times 2000 \mu\text{m} \times 2000 \mu\text{m}$ to assess the properties of organics,
124 fractures and minerals with thousand 2D slices stacked together.

125 2.4. Fractal dimension calculation of fractures

126 The fractals of fractures can be evaluated by box-counting dimension method, fractal Brown
127 motion method and area measurement method [21]. In this study, 2D and 3D box-counting
128 methods were employed to calculate the fractal dimension of 2D slices and reconstructed
129 REV. The box-counting method divides the image space or volume into square boxes of each
130 size δ . By decreasing the size of the square boxes by half, the number of square boxes
131 $N(\delta)$ contained fractures can be counted, which depends on the size of the square box [22]:

$$132 N(\delta) \sim \delta^{-D} \quad (1)$$

133 Then, a log-log plot composed of the number of the boxes $N(\delta)$ versus the different size of
 134 boxes δ can be plotted. Finally, the linear fitting equation can be established below:

$$135 \log(N(\delta)) = -D\log(\delta) + \log(\alpha) \quad (2)$$

136 Where, $N(\delta)$ is the number of square boxes contained fractures, δ is the size of square
 137 boxes, α is constant, and D is the fractal dimension of fractures.

138 2.5. Permeability evaluation

139 The absolute permeability simulation can be conducted through Avizo XLab Extension with a
 140 finite volume method. The equations used in the absolute permeability simulation are
 141 discretized on a staggered grid to allow a better estimation of the no-slip boundary condition.
 142 Before absolute permeability simulation, the isolated fracture space should be removed and
 143 extract the connected fracture space through Avizo software. The aim is to assure that the
 144 fracture space existing in the samples allows fluid pass through. The definition of absolute
 145 permeability is based on Darcy's law:

$$146 \frac{Q}{S} = -\frac{k \Delta P}{\mu L} \quad (3)$$

147 Where Q is the flow rate (m^3 per second); S is the cross section area (m^2); k is the absolute
 148 permeability (m^2); μ is the dynamic viscosity of the fluid ($\text{Pa}\cdot\text{s}$); ΔP is the pressure difference
 149 (Pa); L is the sample length in the flow direction (m).

150 To numerically estimate absolute permeability, the Stokes equations are solved:

$$151 \begin{cases} \vec{\nabla} \cdot \vec{v} = 0 \\ \mu \nabla^2 \vec{v} - \vec{\nabla} P = \vec{0} \end{cases} \quad (4)$$

152 Where $\vec{\nabla}$ is the divergence operator; $\vec{\nabla}$ is the gradient operator; \vec{v} is the velocity of the
 153 fluid in the fluid phase of the material; μ is the dynamic viscosity of the flowing fluid; ∇^2 is
 154 the Laplacian operator; P is the pressure of the fluid in the fluid phase of the material. This

155 simplified Navier-Stokes equations considering an incompressible fluid means the density is a
156 constant; A Newtonian fluid means the dynamic viscosity is a constant; A steady-state flow
157 means the velocity does not vary over time; A laminar flow means the concerned velocities
158 are small enough not to produce turbulence. The simulated flow direction can be adjusted to
159 X, Y or Z direction respectively. The inflow pressure was set to be 1.3×10^5 Pa, the outflow
160 pressure was set to be the atmospheric pressure, and the water viscosity is 1.005×10^{-3} Pa·s.

161 **3. Results and discussion**

162 3.1. 2D morphology of fractures

163 [Fig. 1](#) shows the typical 2D morphology of fractures obtained by X-ray μ -CT, which can be
164 divided into two categories: syngenetic fractures (also named cleats) that formed during the
165 process of coalification, and exogenous fractures that formed during deformation and faulting
166 by tectonic movements [23]. The cleats commonly occur in orthogonal fractures, face cleats
167 and butt cleats (a1-2, b1-2 in [Fig. 1](#)) [24]. Face cleats formed first during coalification, and
168 butt cleats formed later and terminated by face cleats [25]. Face cleats were dominant and
169 extensive, which were wider than butt cleats [26]. The face cleats were basically filled with
170 minerals, whereas no mineralization was observed in butt cleat (a1-2, b1-2 in [Fig. 1](#)).
171 Cleat-filling minerals were epigenetic, which mainly precipitate along the face cleats from the
172 solutions with various compositions and temperatures [27].

173 With the aid of energy dispersive spectrometer, the main fracture filling minerals of sample
174 FK and ML are identified as dolomite (a3 and a4 in [Fig. 1](#)) and kaolinite (b3 and b4 in [Fig. 1](#)),
175 which indicates that the minerals were deposited by the movement of mineralized fluid
176 through the coal seam after the development of major fractures. Gypsum are widely spread in

177 the coal matrix as inherent minerals in the sample FK, and calcite and pyrite occurs as
178 individual grains in coal matrix of the sample ML. Mineral in threshold segmentation can be
179 divided into two categories: fracture-filling minerals and scattered minerals in both sample
180 FK and ML. FESEM imaging (a3 and b3 in Fig. 1) shows the width of open fractures in
181 sample FK and ML are in the range of 0.59-1.83 μm and 1.335-12.6 μm , while mineralized
182 fractures are in the range of 6.07-40.56 μm and 0.13-66.85 μm respectively. The width of
183 mineral-filling fracture varies greatly compared with the open fractures and mineralized fluids
184 mainly flow in large fractures with the width greater than 10 μm .

185 3.2. 3D reconstructed fractures

186 3D fracture reconstructed structures were established by 2D X-ray μ -CT slices to determine
187 the fracture networks. A1 and b1 in Fig. 2 shows that the 3D reconstructed volumes of
188 original FK and ML coals were divided into coal matrix in black, fractures in blue and
189 minerals in yellow. A2 and b2 Fig. 2 shows the 3D structure of FK and ML coals after
190 grayscale threshold segmentation. Fractures are composed of unmineralized fractures in blue,
191 mineral filled fractures in pink. And scattered minerals within coal matrix are presented in
192 yellow. Herein, the mineral filled fractures are the same as fracture filled minerals due to the
193 same threshold segmentation. The volume fraction of each part was calculated through the
194 number of pixels (volume) by CT scanning. The volume fractions of scattered minerals for
195 the samples FK and ML are 0.55% and 13.98% respectively. The mineral-filled fractures or
196 fracture-filling minerals accounts for 10.77% and 7.46% for the samples FK and ML
197 respectively, which occupy 96.42% and 90.31% of the total fractures. Therefore, most
198 fractures in both sample FK and ML were filled with minerals and these fractures that filling

199 minerals may inhibit the fluid flow process due to the decrease in fracture width of the
200 seepage path, which is unfavorable for CBM production, especially when the mineral filled
201 fractures have wonderful connectivity (a2 and b2 in Fig. 2). Therefore, demineralization could
202 be an effective way to increase the fluid flow path and the fractures connectivity, and thus
203 possibly increase the permeability.

204 3.3. The evaluation of fractures complexity

205 3.3.1. Structure evaluation of fractures

206 The fracture density for sample FK was 839 fractures within the volume of 8 mm^3 (149 open
207 fractures and 690 mineral-filling fractures per mm^3), whereas 1451 fractures per 8 mm^3 (1436
208 open fractures and 15 mineral-filling fractures per mm^3) for sample ML, which indicates that
209 more fractures developed in sample ML than that of sample FK. During coalification ($R_{o, \max}$
210 $< 0.65\%$), the dehydration and volatiles loss will result in the formation of microfractures
211 [28]. When $R_{o, \max} > 1.30\%$, the fracture density will decrease slowly and then remain constant
212 with increasing coal rank [29]. The structural parameters (average length, average width,
213 average area and average volume) for open fractures, mineral-filling fractures and total
214 fractures of the two samples were presented in Table 2 and Fig. 3. 3D visualization of open
215 fracture and total fracture in different length scale were presented in Fig. 4. The average width
216 and average volume in sample ML are reduced by 45.94% and 56.78% compared to sample
217 FK and the other average structural parameters including fracture length, area decrease with
218 increasing coal rank (low-rank to medium-rank). At the early coalification of $R_{o, \max}$
219 $= 0.6\% - 1.2\%$, overburden pressure results in structural parameters decreased with the
220 increase of coal rank [30-32]. When $R_{o, \max}$ is in $1.25\% - 2.25\%$, the small-molecule compounds

221 escape from the coal as gases (H_2O , CO_2 , and CH_4) and the average structural parameters
222 continue to decrease because of the overburden pressure and matrix shrinkage, but the rate of
223 decline slows [33]. Mineral-filling fractures occupy 96.42% and 90.31% volume in total
224 fractures for samples FK and ML, which means most fractures are filled with minerals carried
225 by underground fluid. The average width of total fractures increased by 4.46% and 9.87%
226 compared to open fractures and the maximum fracture width changes from $35.95\mu m$ to
227 $55.81\mu m$ and $19.11\mu m$ to $98.96\mu m$ for samples FK and ML respectively, which indicates that
228 the fluid flow space will be greatly improved if mineral-filling fractures are de-mineralized.

229 3.3.2. Fractures complexity by fractal dimension

230 Fractal method was used to evaluate the complexity of fractures [25]. Generally, the
231 microfractures become much more complex as coal rank increase, which leads to the high
232 fractal dimension [31]. The 2D and 3D fractal dimension of open and total fracture of FK
233 sample were calculated by box dimension method as shown in Fig. 5. The following size of
234 square or cube is used (in pixels): 4, 8, 16, 32 and 64. The calculation for obtaining 2D and
235 3D fractal dimension can be seen in Fig. 6. For 2D fractal dimension of fractures, 16 slices
236 were selected evenly to calculate the 2D fractal dimension for sample FK and ML
237 respectively. 2D fractal dimension of fractures varies from slice to slice due to the strong
238 heterogeneity. However, the 3D fractal dimension can represent the fracture complexity of the
239 whole sample. The average 2D and 3D fractal dimensions of open fractures in sample ML are
240 1.16 and 2.172 respectively, which are lower than those of sample FK (1.25, 2.242). Whereas
241 the average value of total fractures for sample ML are 1.50 and 2.519, which are higher than
242 those of sample FK (1.44, 2.403) after demineralization. Therefore, the fractal dimension of

243 fractures in de-mineralized samples, whether 2D or 3D fractal dimensions, increased greatly
244 compared with original open fractures, which means that the value of fractal dimension
245 increases with the degree of fracture development. The 2D and 3D fractal dimension of
246 original open fractures in sample ML are lower than those of sample FK. However, both 2D
247 and 3D fractal dimension of total fractures in sample ML exceed those in sample FK after
248 demineralization for mineral-filling fractures. Therefore, the mineralization of sample ML is
249 more serious.

250 3.4. Insights into petrophysical properties of coals

251 3.4.1 Fracture porosity

252 The fracture porosity of the original open and de-mineralized fractures was numerically
253 calculated to evaluate the effect of minerals on permeability. The fracture porosity of samples
254 FK and ML were obtained from micro-CT data as shown in [Fig. 7](#). For original open fractures,
255 the open fracture porosity in different directions has an average difference of 2.1%, while the
256 total fracture porosity included de-mineralized fractures shows variable porosity with an
257 average difference of 6.3% in different directions, which indicate that the fracture has strong
258 heterogeneity in different directions. The fracture porosity varies from slice to slice. For
259 sample FK and ML, the average total fracture porosity of the de-mineralized samples are
260 6.85% and 11.47%, which increased by 6.39% and 10.37% compared with original open
261 fractures porosity. If the minerals-filling fractures are de-mineralized or acidified, these
262 fractures will be favorable for enhancing CBM recovery. [Fig. 8](#) shows the correlation between
263 the fractal dimensions of fractures and CT fracture porosity. The CT porosity shows an
264 approximately U-shaped change with 2D fractal dimensions or 3D fractal dimensions, which

265 presents similar tendency: porosity firstly decreases from 1.1% to 0.46%, then increases with
266 fractal dimension increasing.

267 3.4.2. Fracture permeability

268 The fracture connectivity is another important factor influencing permeability in coals. Fig. 9
269 show the 3D spatial distribution of connected/unconnected fractures of original samples (a1
270 and b1 in Fig. 9) and de-mineralized samples (a2 and b2 in Fig. 9), respectively. The volume
271 fractions of connected fractures in open and total fractures for sample FK are 33% and 89%,
272 respectively. For sample ML, the volume fractions of the connected fracture are 54.1% and
273 98.4% in open and total fractures, respectively, which indicated that more than half of the
274 open fractures are isolated. And most mineral-filling fractures are well connected with other,
275 which will result in the favorable fluid flow path in fractures after mineral removed. The
276 connective fractures, as effective flow paths, can cause permeability increase for water/gases.
277 In this work, the absolute permeability simulation was conducted based on visualized
278 geometrical properties by micro-CT images [30]. The absolute permeability sharply increase
279 from 400 mD to 450 mD for sample FK and 0 mD to 740 mD for sample ML in the Y axis
280 after demineralization, and increase by 92% and 25% in Z axis after mineral removed, which
281 indicates that more connected fractures were provided for fluid flow. Therefore,
282 de-mineralization or acidification could be very important way for enhancing CBM
283 production. One thing that needs to clear is the result from numerical simulation varied
284 widely from laboratory result, which attribute to the significant interactions of gas, water and
285 fracture-wall that can significantly influence the permeability [31]. Pores that have been
286 neglected in the simulation also occur in coal, which also can make contribution to coal

287 permeability [34]. The difference of permeability indicates the importance of
288 de-mineralization of mineral filling fractures. Therefore, this fracture permeability simulation
289 with and without mineral filling can be conducive to enhancing CBM production.

290 **4. Conclusions**

291 X-ray μ -CT and field emission scanning electron microscope (FESEM) together with energy
292 spectrum experiment (EDS) can be used as effective means to evaluate fracture structure and
293 mineral performance in different rank coals (samples FK and ML). The structure, fracture
294 porosity, connectivity and permeability of open fracture and total fractures was quantitatively
295 evaluated by 2D and 3D fracture model and calculated to illustrate the effect of minerals on
296 flow capability. The following conclusions are made:

297 1) The well developed microfractures and dolomite and kaolinite filled microfractures present
298 in samples FK and ML, which were confirmed by FESEM images and 2D slices of X-ray
299 μ -CT together with EDS. The 3D reconstructed open fractures and total fractures of samples
300 FK and ML show the significantly changes in fracture structure.

301 2) The variable structure parameters (e.g. length, width) indicate that coal rank has significant
302 influence on fracture structure. The fracture density in medium rank coal (sample ML) is two
303 times than that in low rank coal (sample FK), while the structural parameters (average
304 fracture length, width, and volume) reduce as coal rank increase. The fracture structure
305 becomes more complex with the growth of fractal dimensions due to the matrix shrinkage and
306 overburden pressure during the coalification jump.

307 3) The permeability simulation reveals that the structural parameters (e.g. fracture porosity,
308 connectivity and permeability) of the fractures and the minerals have important impacts on

309 permeability. The volume of connected fractures and the porosity sharply increased after
310 mineral removal, which can significantly improve the permeability. Therefore,
311 demineralization or acidification can be used to increase the void space and connectivity of
312 the mineral-filling fractures, and will be favorable for enhancing CBM production.

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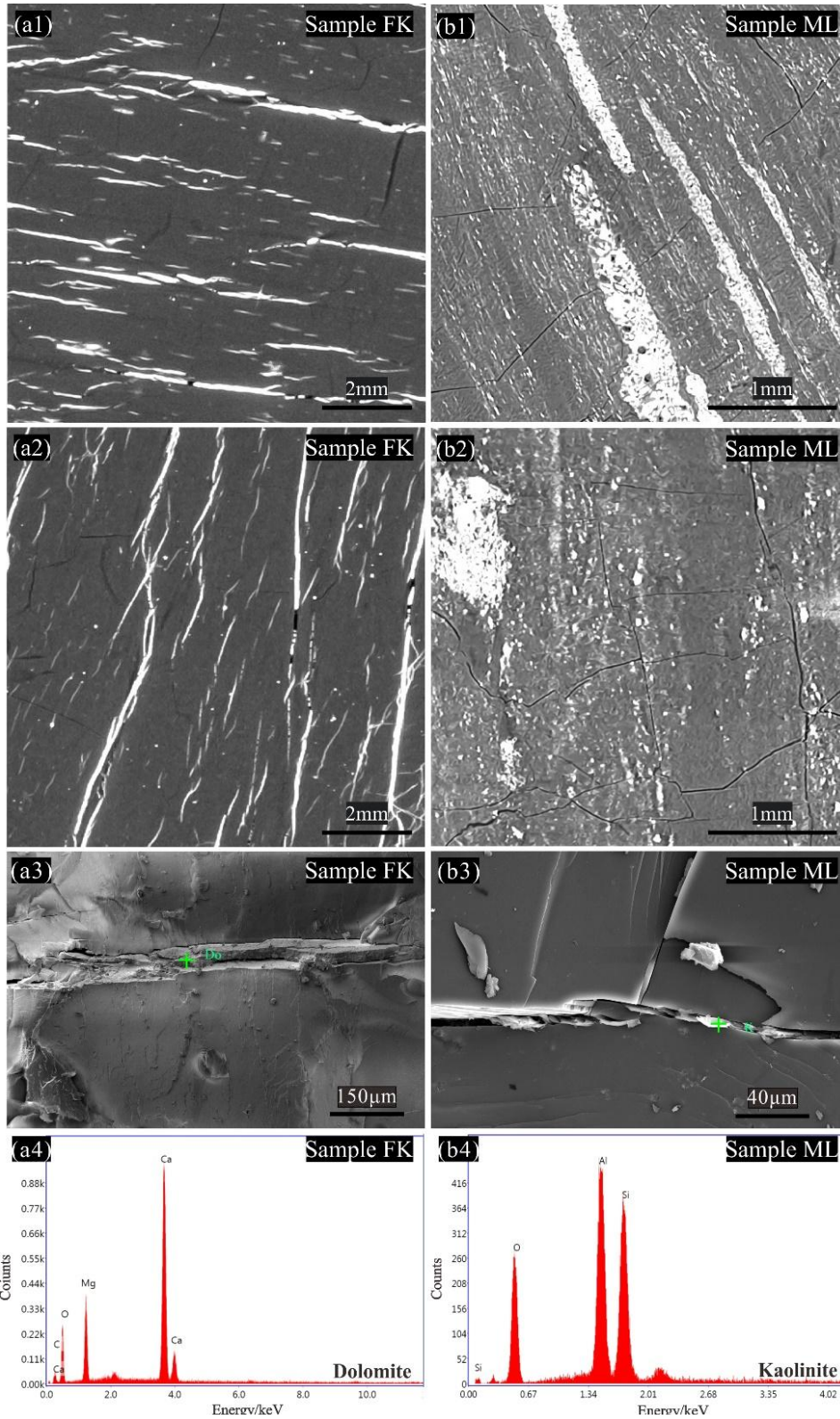
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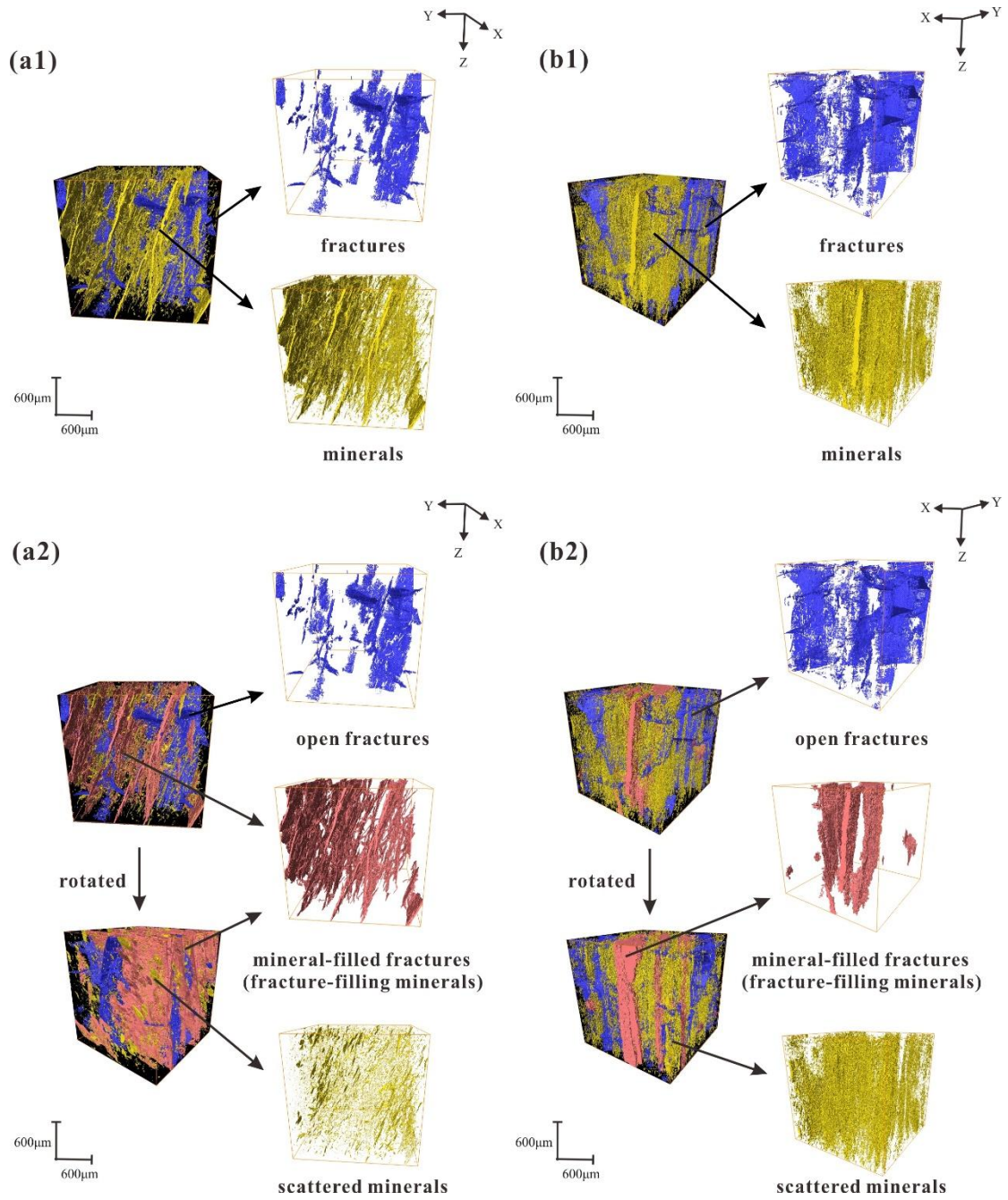
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 402 Fig.1 Typical 2D morphology of fractures obtained by X-ray μ -CT(a1, a2, b1,b2) and FESEM (a3,b3). X-y plane (a1,b1) and y-z
 403 plane (a2,b2) cross-section of sample FK and ML. Fractures in sample FK filled with dolomite (a3) and the spectrum obtained
 404 from a point on dolomite's surface (a4). Fractures in sample ML filled with kaolinite (b3) and the spectrum obtained
 405 from dolomite's surface (b4).

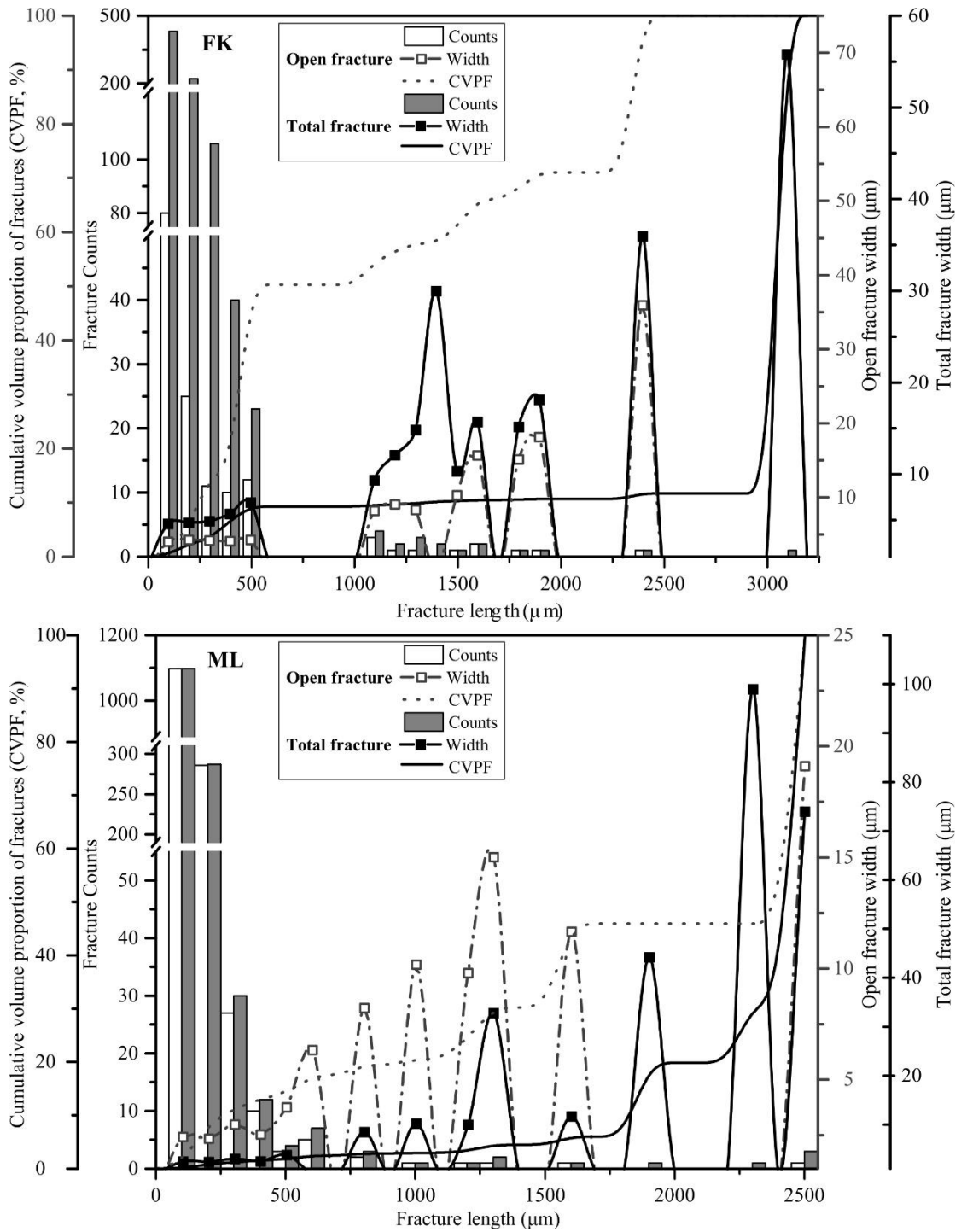


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Fig.2 3D visualization of original sample FK (a1) and ML (b1), re-segmented sample FK (a2) and sample ML (b2). For original samples (a1,b1), coal matrix are shown in black, fractures are shown in blue and minerals components are shown in yellow. For re-segmented samples (a2, b2), open fractures are shown in blue, mineral-filled fractures (fracture-filling minerals) are shown in pink, and scattered minerals are shown in yellow.

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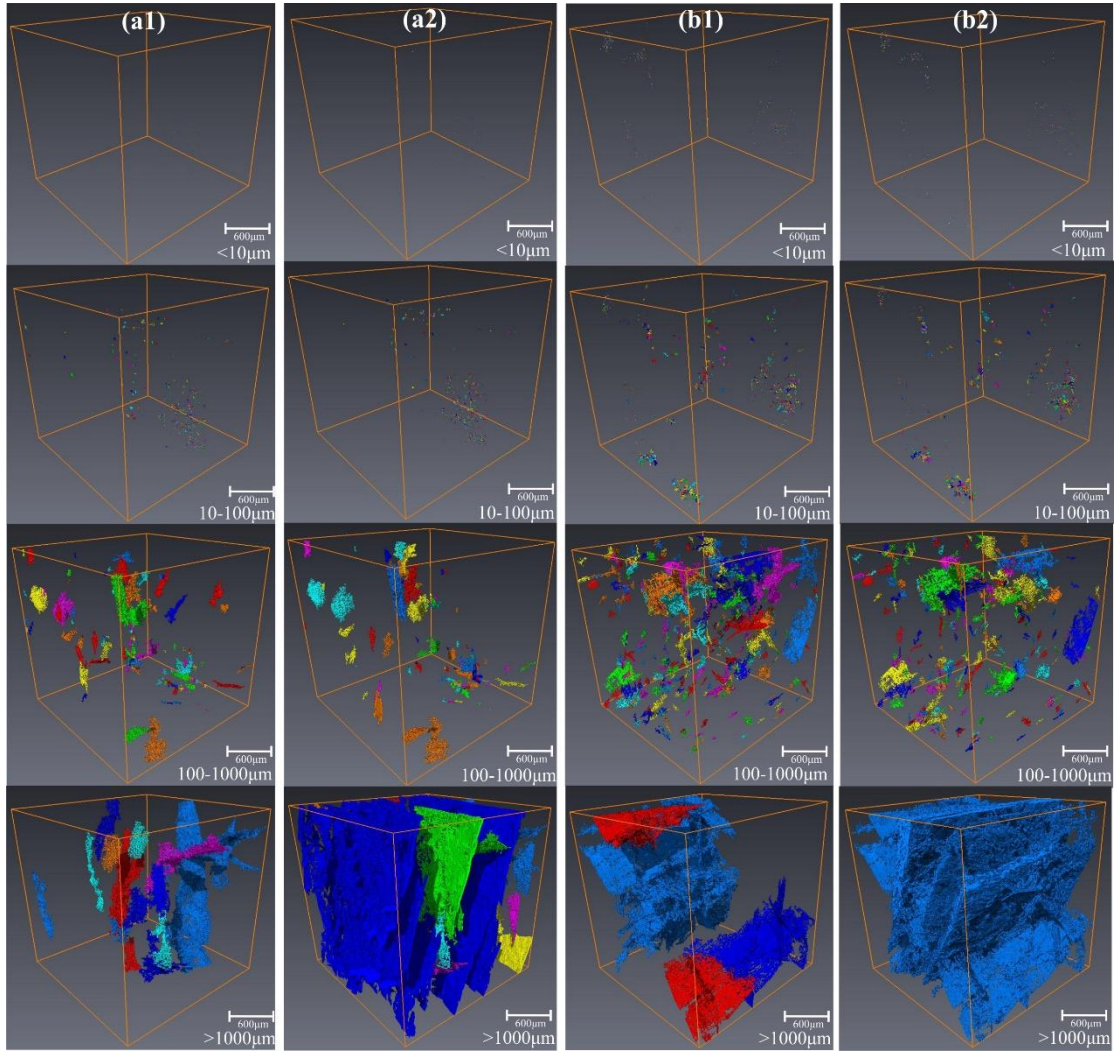
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418 Fig.3 The counts, average width and cumulative volume proportion of open fracture and total fracture in different length scale.

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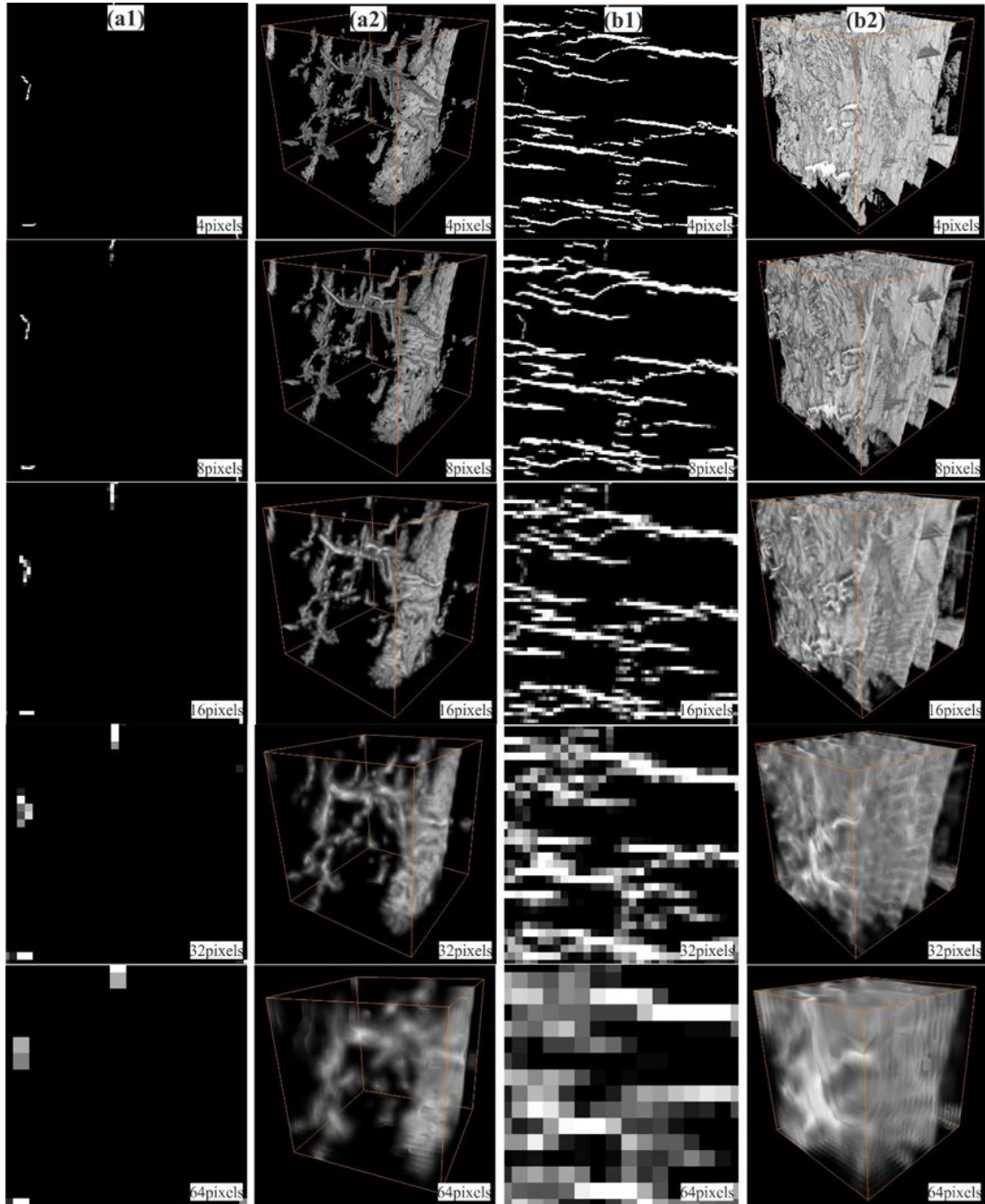


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421 Fig.4 Different length scale of open fractures and total fractures in sample FK and ML: Fractures length $<10\mu\text{m}$, 10-100 μm ,

422 100-1000 μm and $>1000\mu\text{m}$. (a1) Open fractures in sample FK; (a2) Total fractures in sample FK. (b1) Open fractures in sample

423 ML. (b2) Total fractures in sample ML.

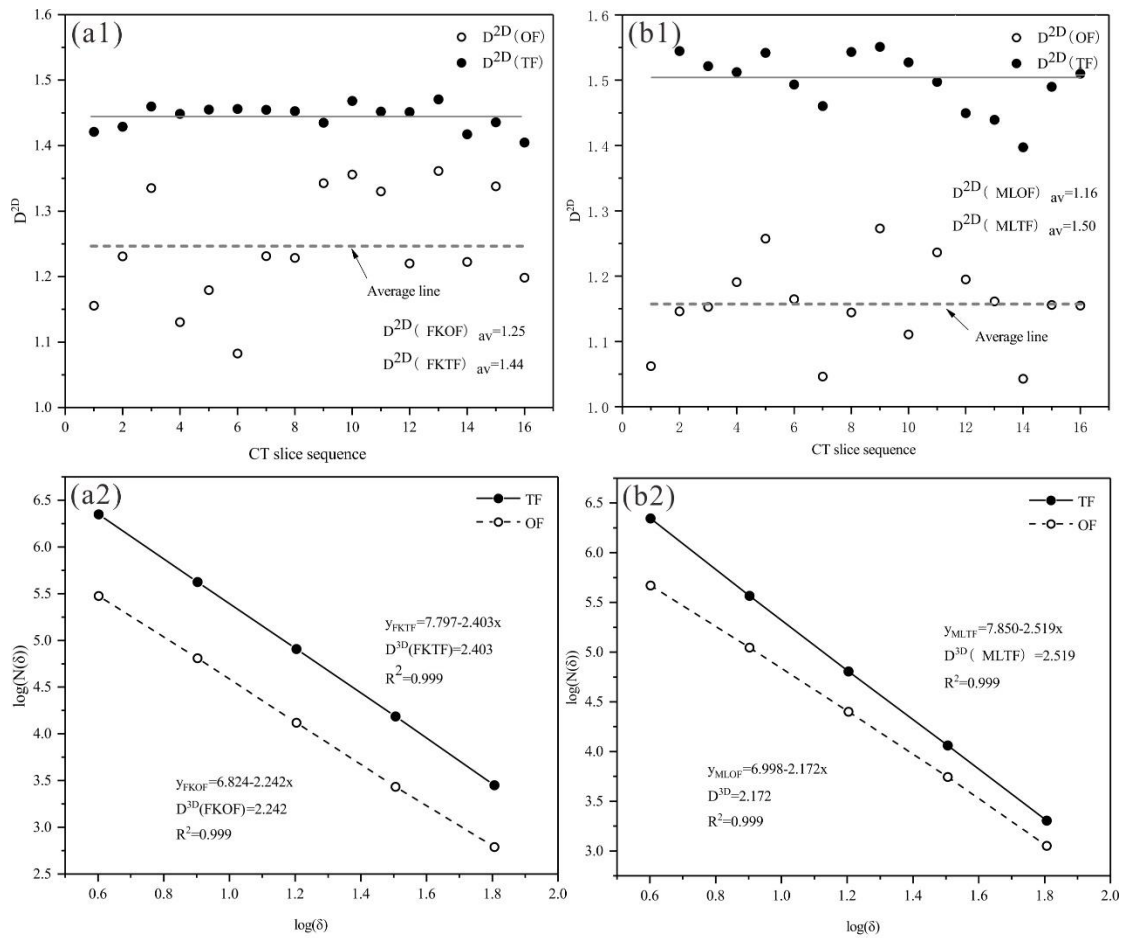


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425 Fig.5 Two dimension (2D) and three dimension (3D) fractal dimension calculation processes of fracture in sample FK. (a1) 2D
 426 fractal dimension calculation processes of open fracture, taking the 1st section along Z axis as example; (a2) 3D fractal dimension
 427 calculation processes of open fracture in sample FK; (b1) 2D fractal dimension calculation processes of total fracture, taking the 1st
 428 section along Z axis as example; (b2) 3D fractal dimension calculation processes of total fracture in sample FK. The size of square
 429 or cube were used (in pixels):4,8,16,32,and 64,espectively.

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Fig.6 The results of two dimension(2D) and three dimension(3D) fractal dimension of samples FK and ML. (a1)The 2D fractal dimension of open and total fractures in sample FK along Z axis direction;

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(a2)The 3D dimension of open and total fractures in sample FK;

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(b1) The 2D fractal dimension of open and total fractures in sample ML along Z axis direction;

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(b2) The 3D dimension of open and total fractures in sample ML. Notes: OF: Open fracture; TF: Total fracture.

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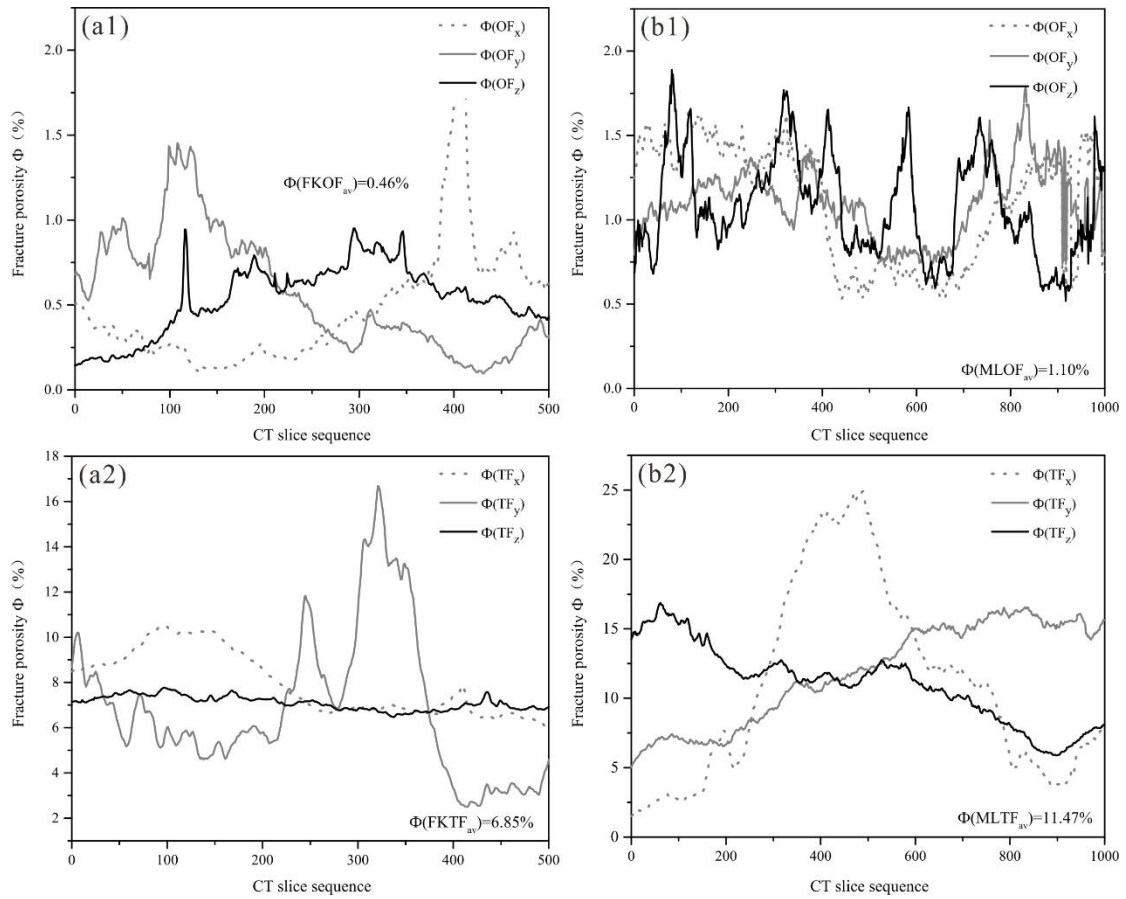
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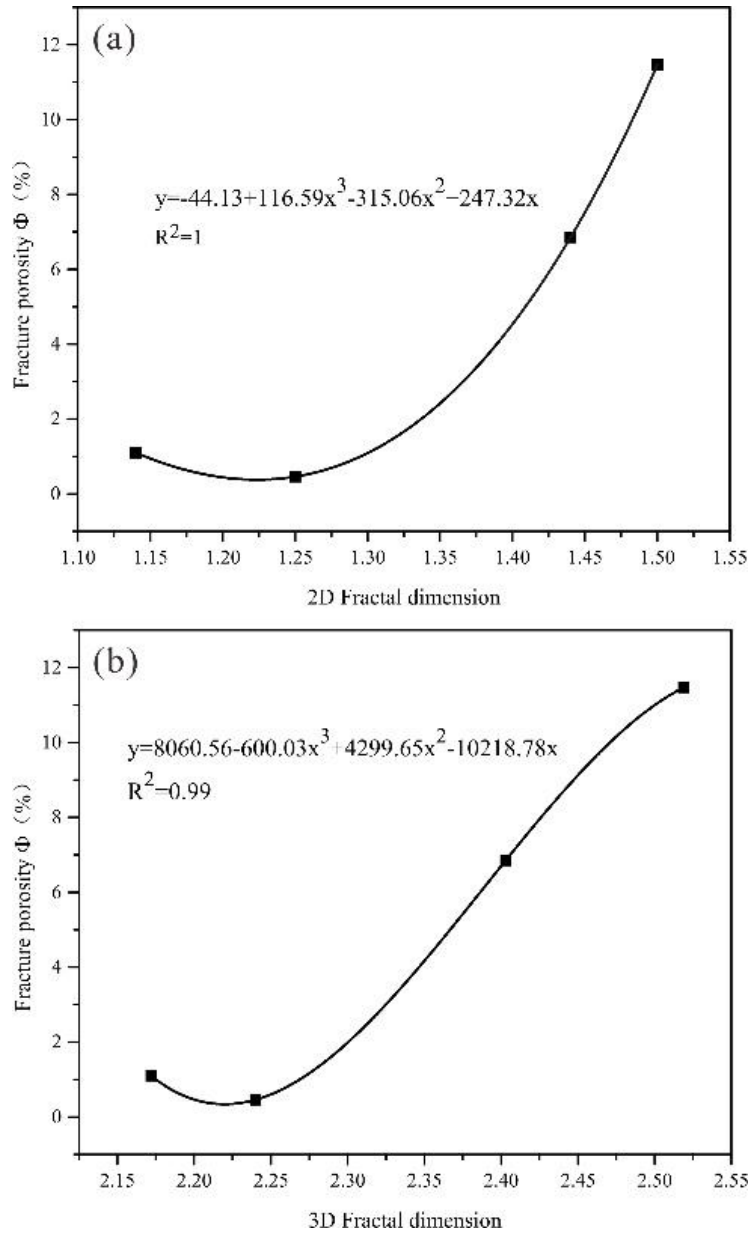
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Fig.7 Fracture porosity of original (Φ (OF)) and de-mineralized (Φ (TF)) of samples FK and ML in different directions. (a1) Fracture porosity of original sample FK (Φ (FKOF)); (a2) Fracture porosity of de-mineralized sample FK (Φ (FKTF)); (b1) Fracture porosity of original sample ML (Φ (MLOF)); (b2) Fracture porosity of de-mineralized sample ML (Φ (MLOF)). Notes: OF: Open fracture; TF: Total fracture.



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Fig.8 The correlation between the fractal dimensions ((a):2D fractal dimension; (b)3D fractal dimension). Porosity firstly decreases and then increases with fractal dimension increasing, displaying a 'U-shaped' trend.

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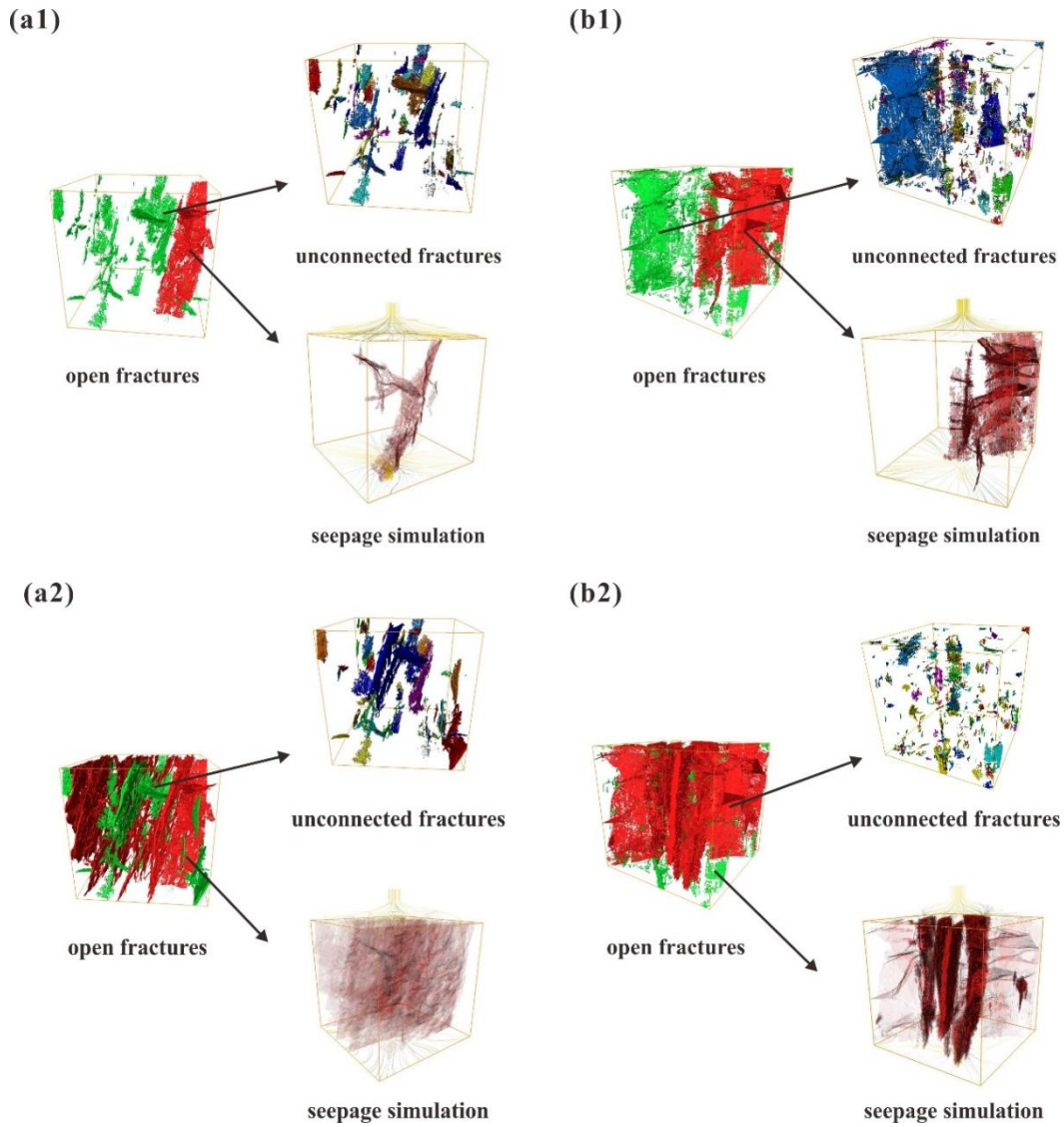
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Fig.9 Results of connectivity analysis showing 3-D distribution of connected-unconnected fractures in original and de-mineralized sample FK (a1 and a2) and sample ML (b1 and b2). Further seepage simulation of connected fractures and 3-D distribution analysis of unconnected fractures were carried out. Connected fractures are shown in red and unconnected fractures are shown in green.

483 Table 1 Petrographic and proximate analysis results for two Chinese coals

Sample No.	R _{o,max} (%)	Coal rank	Coal composition (Vol. %)			Proximate analysis (wt. %)			
			Vitrinite	Inertinite	Exinite	Mad	Aad	Vad	FCad
FK	0.65	subbituminous coal	83.9	1.7	14.5	3.76	8.86	39.21	48.17
ML	1.49	bituminous coal	67.6	32.4	0	0.29	9.97	14.73	75.01

484 R_{o,m} = maximum vitrinite reflectance under oil immersion; Mad = moisture content (wt. %, air dry basis); Aad = ash yield (wt. %, air dry basis); Vad = volatile matter (wt. %, air dry basis); FCad = fixed carbon (wt. %, air dry basis).

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Table 2 The average structural parameters of open fractures and mineral-filled fractures in sample FK and ML.

Sample no.	Structural parameter	Fracture type		
		Open fractures	Mineral-filled fractures	Total fractures
FK	AL (μm)	67.55	81.56	76.80
	AW (μm)	4.83	5.10	5.05
	AA (μm^2)	5.23×10^4	1.00×10^5	8.91×10^4
	AV (μm^3)	2.16×10^5	1.33×10^6	1.07×10^6
ML	AL (μm)	58.82	934.30	67.87
	AW (μm)	2.48	26.21	2.73
	AA (μm^2)	2.20×10^4	5.57×10^6	7.93×10^4
	AV (μm^3)	4.46×10^4	4.03×10^7	4.61×10^5

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Notes: AL=average length; AW=average width; AA=average area; AV=average volume.

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556 Table 3 Comparison of structural parameters between open fractures and total fractures, sample FK and ML.

Relative growth rate between OAT (%)	Sample no.		Relative decline rate between FAM (%)	Fracture type	
	FK	ML		Open fracture	Total fracture
AL	15.84	15.39	AL	12.92	13.27
AW	4.46	9.87	AW	48.74	45.94
AA	67.69	261.39	AA	58.71	11.01
AV	390.07	933.39	AV	79.51	56.78

557 Notes: AL=average length; AW=average width; AA=average area; AV=average volume; OAT=open fractures and total fractures;
 558 FAM=FK and ML sample.