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Micro-nano-scale Stimulation of Coalbed Methane Reservoir: Experimental Hydraulic Fracturing and Pore Connectivity

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Key Points:

- This study provided novel and detailed insights into the microscopic mechanisms of coal hydraulic fracturing
- The changes of pore number resulted in the changes of pore volume and specific surface area after hydraulic fracturing
- The change characteristics of coal pore hydraulic fracturing were affected by elastic modulus and in-situ stress

24 **Abstract**

25 The role of micro-nano-scale stimulation of coalbed methane (CBM) reservoir is to open the
26 fracture and pore channels, improve gas desorption and migration capabilities, and thereby
27 increase CBM production capacity. Based on hydraulic fracturing simulation experiments,
28 combined with liquid nitrogen absorption (LNA) and mercury intrusion porosity (MIP)
29 measurements, the micro-nano-scale pore structure of coal before and after hydraulic fracturing
30 stimulation in coal reservoirs was studied. It is found that significant changes in the number of
31 pores in different pore sizes are the main reason for the changes in coal pore volume (PV) and
32 pore specific surface area (PSSA) after hydraulic fracturing. the micro-nano-scale pore
33 stimulation of hydraulic fracturing includes two forms of pore fragmentation and pore
34 deformation, the coal with larger elastic modulus is mainly manifested as brittle crush.
35 Moreover, larger in-situ stress will inhibit pore fracture and expansion, and even cause further
36 compression of pores.

37 **Plain Language Summary**

38 The application of hydraulic fracturing technology provides more possibilities for the
39 development of coalbed methane (CBM). Hydraulic fracturing mainly acts on the coal matrix to
40 connect the originally isolated pores and fractures, increasing the development potential of CBM.
41 However, previous studies could not explain the effect of hydraulic fracturing on pores
42 accurately. We have studied the pores of coal after hydraulic fracturing, the research results
43 prove that the number of pores in coal changes significantly after hydraulic fracturing, which
44 causes the redistribution of pores. The change forms of pores after hydraulic fracturing are
45 determined by the elastic modulus of coal, the larger the elastic modulus of coal means that the
46 pores are more easily broken to form smaller pores. However, the phenomenon of pore
47 fragmentation or expansion will be limited as the in-situ stress increases.

48 **1 Introduction**

49 The stimulation of micro-nano-scale pore fissures in low-porosity and low-permeability coalbed
50 methane (CBM) reservoirs can increase the connectivity of micro-nanopore fissure structures in
51 CBM reservoirs, improve the permeability of coal reservoirs, increase the recovery rate of CBM
52 and the production of CBM, and promote the effective development of CBM (Ju et al., 2018).
53 The effective development of CBM is of great significance to alleviate the world energy crisis,
54 reduce the emission of methane from coal mine shafts during coal mining and prevent and
55 control coal mine gas disasters.

56 China has abundant CBM resources. As a reservoir for the production and occurrence of CBM,
57 the characteristics of the coal seam are important for CBM production. Considering that coal
58 seams usually have a double pore structure (Liu et al., 2011; Liu et al., 2015), the pore structure
59 of coal directly determines the characteristics of gas adsorption, desorption, and diffusion (Pan et
60 al., 2015), which in turn affects the development of CBM. Therefore, the pore development
61 characteristics of coal seams have been a research focus (Clarkson & Bustin, 1999; Zhang et al.,
62 2012). The development of the original pore structure of coal in China is not ideal, the coal seam
63 pore structure is affected by coal pore pressure, effective stress, and matrix deformation (Liu &
64 Harpalani, 2014; Zhou et al., 2016; Liang et al., 2017; Hou et al., 2017; Li et al., 2018), and coal
65 seams in China have strong heterogeneity and low permeability, which is not conducive to CBM
66 production or to the prevention of mine gas disasters. For the efficient development of CBM and
67 for the prevention and control of coal mine gas outbursts, it is imperative to stimulate CBM
68 reservoirs at the micro-nano-scale (Ju et al., 2017), especially for high rank coal reservoirs with
69 more micropores and less fractures.

70 To improve the efficiency of CBM reservoir stimulation, researchers have proposed various
71 technical approaches, including high-pressure pulsed water jets (Liu et al., 2011), water jet

72 slotting (Shen et al., 2015), enhancement with carbon dioxide injection (CO₂-ECBM) (Busch &
73 Gensterblum, 2011; Gensterblum et al., 2014; Fan et al., 2019; Wang et al., 2020), acoustic
74 shocks (Zhang & Li, 2017), microwave vibration (Kumar et al., 2011), and hydraulic fracturing
75 technology (Lu et al., 2020; Tan et al., 2017). Xie et al. (2020) studied the pore structure of coal
76 after pulsating hydraulic fracturing and found that under the action of pulsating hydraulic
77 fracturing, there are two main changes in pores: expansion of original pores and formation of
78 new pores. Cai et al. (2013) conducted heat treatment on coals of different ranks and found that
79 heat treatment could significantly increase the porosity of coal samples. Shi et al. (2016) found
80 that repeated strong pulse waves had a greater impact on coal pore structure, and that smaller
81 pores were more sensitive than larger pores. For the more commonly used hydraulic fracturing
82 technology in the development of CBM, research has mainly focused on macrofractures caused
83 by hydraulic fracturing: The initiation and expansion of hydraulic fractures is mainly affected by
84 reservoir physical properties, horizontal in-situ stress difference, and construction conditions.
85 Hydraulic fracture initiation is mainly controlled by the physical properties of coal reservoirs.
86 Under a constant water pressure, the greater the original permeability, porosity, elastic modulus,
87 and Poisson's ratio of coal, the easier it was for coal to produce hydraulic fractures (Li & Xing,
88 2015). The extensive development of primitive fractures near the well also reduced the pressure
89 of hydraulic fracture initiation and expansion (Dehghan et al., 2016). The propagation of
90 hydraulic fractures was mainly affected by in-situ stress (Ma et al., 2016; Zhang et al., 2018).
91 Construction conditions (injection pressure, injection rate, fracturing fluid viscosity) have a
92 significant influence on the initiation and expansion of hydraulic fractures (Li et al., 2014). For
93 micro-nano-scale pores, Lu et al. (2020) studied the dynamic evolution of coal pores based on
94 online hydraulic fracturing technology, and found that within 10–120 min of fracturing fluid

95 injection, the total porosity of the coal increased dramatically and pores were formed. As the
96 injection pressure decreases, the number of micropores (< 10 nm) and transition pores (10–100
97 nm) continued to increase. However, the above-mentioned studies mainly focused on the
98 stimulation of CBM reservoirs at the micro-nano-scale by other stimulation methods. For more
99 commonly used hydraulic fracturing technology, studies have focused on fractures caused by
100 hydraulic fracturing, and there is little research on micro-nano-scale pore hydraulic fracturing
101 stimulation of large-scale coal samples, especially the influence of hydraulic fracturing on nano
102 pores.

103 In this study, based on mercury intrusion porosity (MIP) and liquid nitrogen absorption (LNA),
104 the changes of coal pore development and pore connectivity before and after hydraulic fracturing
105 were compared, and the mechanism of pore hydraulic fracturing stimulation was analyzed. The
106 conclusions of this study provide guidance for optimizing hydraulic fracturing technology,
107 improving coal seam permeability, increasing CBM production, and for effectively solving mine
108 gas outburst problems.

109 **2 Sample collection and experimental methods**

110 2.1 Characteristics of high-rank coal samples in southern Qinshui Basin

111 The study area is located in the southern Qinshui Basin of Shanxi Province, China, which is one
112 of the most important CBM industrial development base in China. In order to better study the
113 effect of hydraulic fracturing of high-rank coal pores in Qinshui Basin, anthracite samples from
114 Chengzhuang Mine and Sihe Mine were selected for hydraulic fracture simulation experiments.
115 The mean random vitrinite reflectance of the CZ coal sample was 2.405 and that of the SH coal
116 sample was 2.385.

117 The mechanical properties of coal is an important factor affecting the crush characteristics of
118 coal hydraulic fracturing. Mechanical properties of coal were measured by uniaxial compression
119 using a KMT-150 rock mechanics experimental system and performed in reference to national
120 standard GB-T23561.1-16-2009 of the People's Republic of China. The elastic modulus of CZ
121 coal samples was 4.206 GPa and Poisson's ratio was 0.223, and these of SH coal samples were
122 3.432 GPa and 0.297, respectively.

123 2.2 Hydraulic fracturing simulation experiment

124 After coal samples were processed, samples SHF-1, SHF-2, and CZF-1 were wrapped with
125 concrete into $300 \times 300 \times 300$ mm sample blocks, owing to its larger volume, sample CZF-2 was
126 wrapped into a block of $400 \times 400 \times 400$ mm. After the concrete had cured for 24 h, each test
127 block was drilled with a rock drill, with vertically layered drilling directions, the diameter of the
128 borehole was ~ 1 cm to simulate a typical drilling process. The wellbore was put into the hole and
129 then tightly combined with the well wall using epoxy to simulate the cementing process, a space
130 of 3–5 cm was reserved below the wellbore to simulate an open hole section. In this experiment,
131 slickwater (containing 0.5% KCl) with a viscosity of 3 MPa•s was used as the fracturing fluid.
132 Considering the influence of in-situ stress on the experimental results, the in-situ stress of SHF-1
133 and CZF-1 samples was σ_H : 15 MPa, σ_h : 11 MPa, σ_V : 20 MPa, and that of SHF-2 and CZF-2
134 samples was set to σ_H : 19 MPa, σ_h : 11 MPa, σ_V : 20 MPa to simulate the stress conditions of the
135 deep coal seam (about 600 m), where σ_H and σ_h were horizontal in-situ stress, and σ_V was
136 vertical stress. The specific experimental device and procedure Zhou et al. (2008) have described
137 in detail.

138 2.3 Measurement and experimental methods of micro-nano pore structure

139 After completion of hydraulic fracturing experiments, four coal samples were selected in the
140 vicinity of the fracturing wellbore, at the same time, original coal samples of Chengzhuang Mine
141 and Sihe Mine were prepared as the standard. It should be noted that all coal samples were
142 evenly sampled and fully mixed to reduce the influence of coal heterogeneity on the
143 experimental results. Samples were crushed to 3–6 mm and 0.18–0.25 mm, and 10 g from each
144 particle size range was selected and dried to a constant weight at 105°C. MIP experiments were
145 conducted on the coal samples with a diameter range of 3–6 mm using an AutoPore IV 9500
146 mercury injection instrument with a pressure range of 0.00–228.00 MPa and a test aperture range
147 of 5.5 nm–360 μm. The experimental process was conducted according to standard
148 GB/T21650.1-2008. In addition, LNA experiments were carried out on the coal samples with
149 particle sizes of 0.18–0.25 mm using a TriStar II 3flex automatic PSSA and physical adsorption
150 analyzer, the measured pore size distribution range was 0.35–500.00 nm. The experimental
151 process was conducted with reference to standards GB/T 21650.2-2008 and ISO 15901-2:2006.

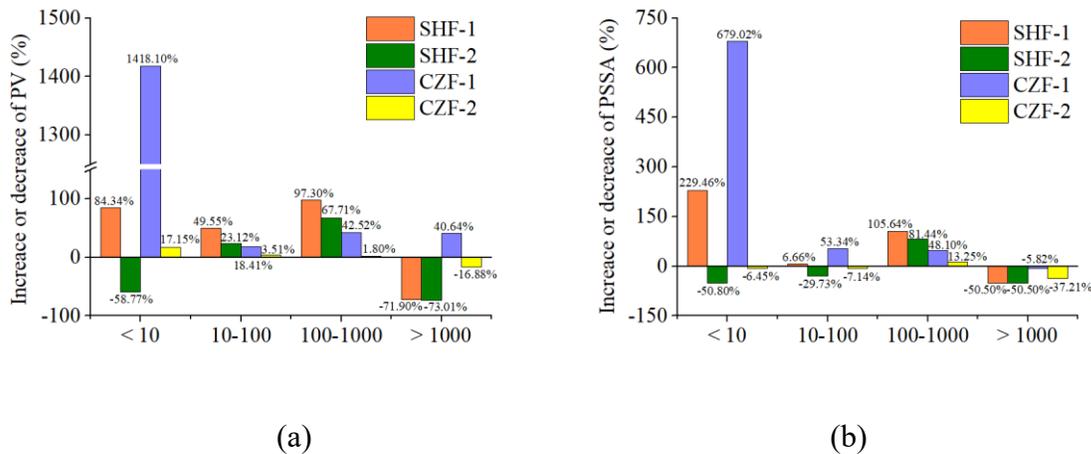
152 **3 Results and analysis**

153 3.1. Changes of coal pore before and after hydraulic fracturing

154 The characteristics and distribution of pores in coal vary widely and it is difficult to determine
155 the multi-scale pore distributions of coal samples using a single experimental method. LNA and
156 MIP are commonly used methods in the study of coal pore structural characteristics (Mastalerz et
157 al., 2012; Li et al., 2012). The combination of LNA and MIP is an effective method for
158 comprehensive and quantitative characterization of coal pore structure. LNA experiments
159 following the BJH method are relatively accurate for pore diameters of < 50 nm, while the MIP
160 method has a large error (coal matrix compression effect) in the range of < 30 nm (Li et al.,

161 1999). For the greatest accuracy, we took 50 nm as the threshold (i.e., for pores of > 50 nm
 162 diameter the MIP approach was used, for pores of < 50 nm the LNA approach was used) (Barrett
 163 et al., 1951).

164 The pore size division is very important for the study of pore distribution characteristics, and the
 165 targeted pore size division method can clearly reflect the distribution characteristics of pores
 166 with different pore sizes. In this study, the Hodot classification scheme was used (Hodot, 1966),
 167 with pores divided into micropores (< 10 nm), transition pores (10–100 nm), mesopores (100–
 168 1000 nm) and macropores (> 1000 nm).



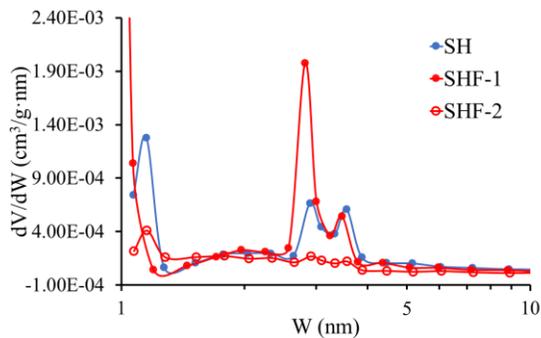
169
 170 (a) (b)
 171 **Figure 1.** Variations in the pore volume (PV) and pore specific surface area (PSSA) of coal for different pore
 172 sizes ranges before and after hydraulic fracturing experiments. (a): Increase or decrease of PV, (b): Increase or
 173 decrease of PSSA.

174 After hydraulic fracturing, the total PV of coal pores showed a downward trend. Under different
 175 in-situ stress conditions, the SHF coal samples of showed the same trend for transition pores,
 176 mesopores, and macropores, that is, the volume of transition pores and mesopores increased by
 177 varying degrees after hydraulic fracturing (Figure 1a). For micropores, the PV of SHF-1
 178 increased by 84.338% while that of SHF-2 fell by 58.772%. The PV of CZF-1 in the micropores,
 179 transition pores and mesopores was the same as that of SHF-1 while that of macropores was the

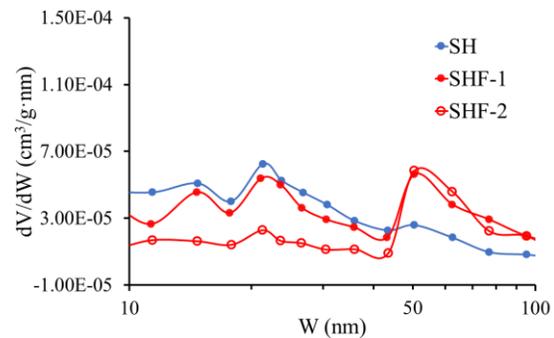
180 opposite. However, the change trends of CZF-2 were consistent with that of CZF-1 in PV of
 181 micropores, transition pores and mesopores, while the change trend of macropores was opposite,
 182 the PV of the macropores was reduced by 16.883%. The change amplitude of PV of CZF-2
 183 sample in each aperture section was smaller than that of CZF-1 sample obviously.

184 The gas adsorption capacity of a coal seam is closely related to the development characteristics
 185 of coal PSSA (Clarkson & Bustin, 1999; Ju et al., 2009). After hydraulic fracturing, the total
 186 PSSA of coal showed different trends. Compared with the original coal samples (SH: 2.2047
 187 m^2/g , CZ: 0.7356 m^2/g), the total PSSA of SHF-1 (6.5679 m^2/g) and CZF-1 (4.4415 m^2/g)
 188 increased significantly, both coal samples showed that PSSA of micropores, transition pores and
 189 mesopores increased, while that of macropores decreased (Figure 1b). For the SHF-2 (1.1694
 190 m^2/g) and CZF-2 (0.6904 m^2/g) samples, the total PSSA was reduced 46.960% and 6.141%,
 191 respectively, the main manifestations were that the PSSA of micropores, transition pores and
 192 macropores were reduced to varying degrees, and the PSSA of mesopores increased
 193 significantly.

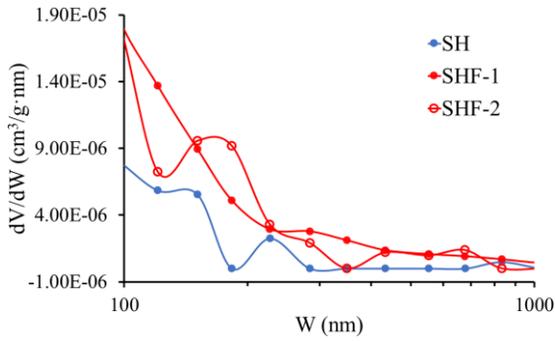
194 In general, the change characteristics of PV and PSSA are directly related to the change of pore
 195 number. In this study, differential PV versus pore size was used to construct pore size
 196 distribution diagrams of coal samples (Figure 2).



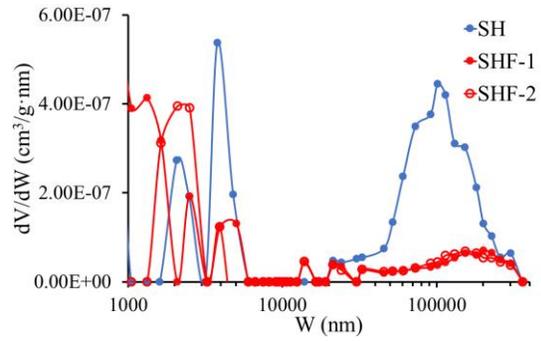
197 (a) 1–10 nm



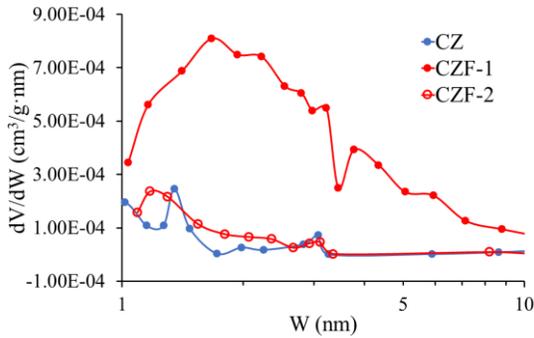
198 (b) 10–100 nm



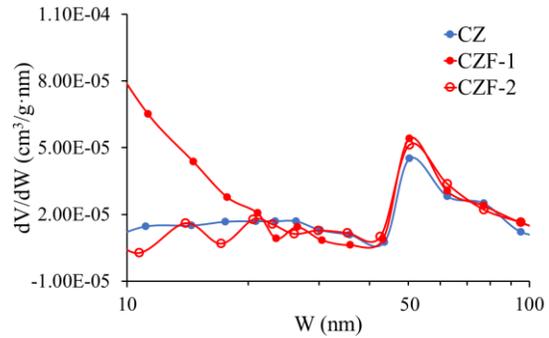
(c) 100–1000 nm



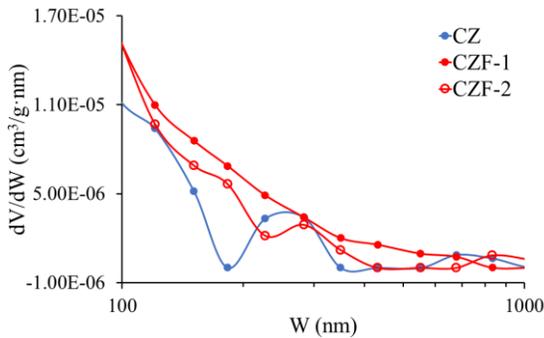
(d) 1000–400000 nm



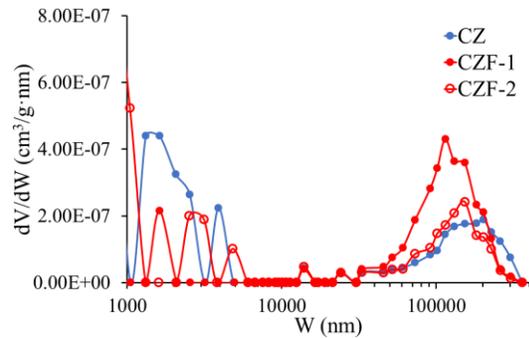
(e) 1–10 nm



(f) 10–100 nm



(g) 100–1000 nm



(h) 1000–400000 nm

Figure 2. Distribution of coal pore size before and after hydraulic fracturing experiments. (a), (b), (c) and (d) are pore diameter distribution characteristics of SH samples before and after hydraulic fracturing, (a) 1–10 nm, (b) 10–100 nm, (c) 100–1000 nm, (d) 1000–400000 nm. (e), (f), (g) and (h) are pore diameter distribution characteristics of CZ samples before and after hydraulic fracturing. (e) 1–10 nm, (f) 10–100 nm, (g) 100–1000 nm, (h) 1000–400000 nm.

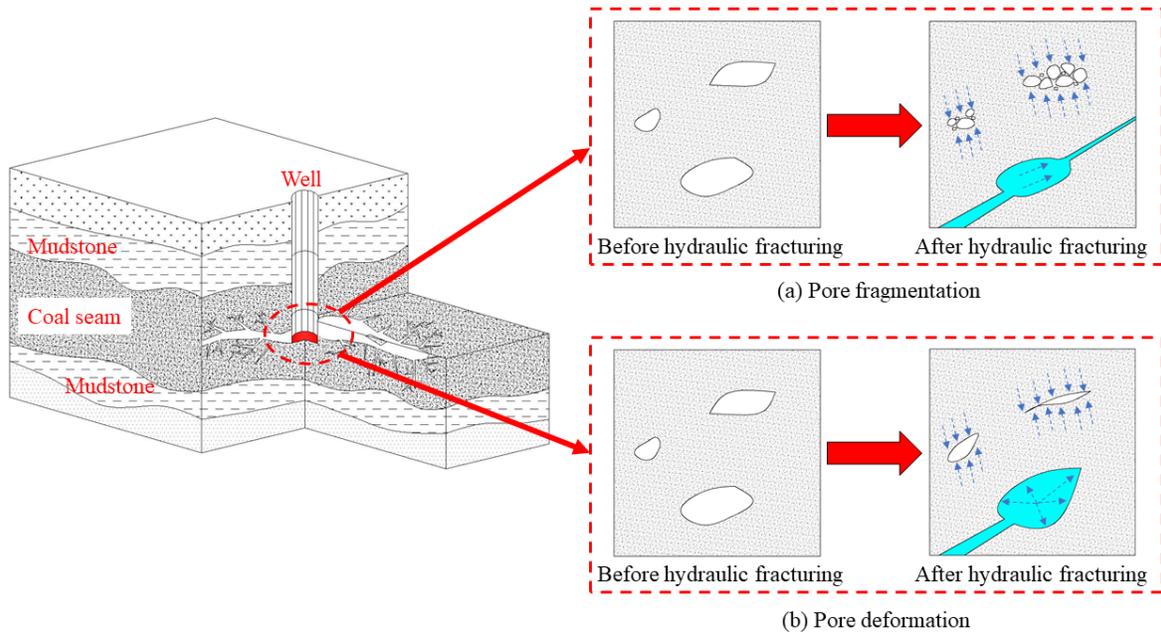
210 After hydraulic fracturing, pores changed significantly in the range of 1–100 nm, other aperture
211 segments also had small changes, which could be seen from the order of magnitude of the
212 ordinate in Figure 2. There were differences in the stimulation of different coal samples, this
213 difference was mainly concentrated in the pore size of 1–50 nm and > 1000 nm. The number of
214 micropores in SHF-1 and SHF-2 samples was slightly different from the original coal (SH), the
215 number of pores in most pore segments decreased slightly after hydraulic fracturing, and there
216 was a relatively obvious inflection point within the aperture range between 3–4 nm. CZ ore
217 samples also had obvious inflection points between 3–4 nm, the number of micropores in CZF-1
218 sample was significantly increased, the CZF-2 sample only began to occupy a smaller advantage
219 in the pore size below 4 nm. After hydraulic fracturing of SH coal, the number of pores in the
220 mesopore section could be divided into two stages: The number of pores in the 50–100 nm
221 aperture section was dominant while the number of pores in the original coal (SH) was more
222 developed in the 10–50 nm aperture section. The CZ samples were more complex, especially the
223 CZF-1 sample, there was a decrease in the number of pores between 20–50 nm, and the number
224 of pores in the pore size range of 10–20 nm and 50–100 nm had an absolute advantage. CZF-2
225 had a dominant pore size in the 50–100 nm pore size range, but the number of pores was
226 significantly reduced in the 10–50 nm pore size range. The number of macropores of SH coal
227 samples and CZ coal samples showed two opposite trends after hydraulic fracturing: taking 3000
228 nm as the boundary, the pore number of SHF-1 and SHF-2 samples was dominant at < 3000 nm,
229 and SH pore number was dominant at > 3000 nm. On the contrary, the pore number of CZF
230 samples dominated at > 3000 nm, and CZ sample dominated at 1000–3000 nm pore diameter
231 section. In the mesopore section, the samples after hydraulic fracturing occupied the dominant
232 position.

233 3.2. Hydraulic fracturing stimulation form of micro-nano-scale pore

234 From the experimental results, under the same hydraulic fracturing conditions, after hydraulic
235 fracturing of coal samples, the change trends of pore number, PV and PSSA of different samples
236 showed great differences. Considering the different mechanical properties of coal samples, there
237 may be great differences in the form of coal pore stimulation.

238 Based on the above-mentioned pore change characteristics and connectivity changes of different
239 samples after hydraulic fracturing, it is believed that there are two main forms of pore
240 stimulation during hydraulic fracturing: pore fragmentation and pore deformation (Figure 3), the
241 former mainly occurs in the macropores and mesopores that may be connected during hydraulic
242 fracturing, the latter mainly occurs in transition pores and micropores that are difficult to be
243 connected. The specific form of pore change is also related to the mechanical properties of coal.
244 Considering that SH coal has a larger elastic modulus (4.206 GPa) and a smaller Poisson's ratio
245 (0.223), SH samples were more easily broken with the increase of stress in coal seam. Therefore,
246 in the process of hydraulic fracturing, the macropores and mesopores that could be connected
247 were crushed as the hydraulic pressure in the coal seam gradually increased (Lu et al., 2020),
248 part of them was transformed into microfractures, others was transformed into macropores and
249 mesopores with smaller pore diameters, resulting in a decrease in the number of macropores with
250 a pore diameter > 3000 nm (Figure 2d), while the number of mesoporous segments increased
251 slightly (Figure 2c). On the one hand, the increase in the number of 50–100 nm was caused by
252 the fragmentation of macropores and mesopores. On the other hand, as the pore size range of
253 fracturing fluid could be affected was limited in the process of hydraulic fracturing, fracturing
254 fluid accumulated pressure within the smallest pore size range, and then destroyed this part of the
255 pores, causing a slight increase in the pore size in this range (Figure 2b). In addition, both the
256 expansion of pores and the formation of microfractures required space, it resulted in the

257 compression of some pores and an increase in the number of micropores (Figure 2a) (Shi et al.,
 258 2020). As for the decreased in the number of micropores in the SHF-2 sample, it might be related
 259 to the in-situ stress conditions.



260
 261 **Figure 3.** Change model of coal pore structure and connectivity before and after hydraulic fracturing. (a) Pores
 262 fragmentation, which mainly includes pores are cut through by fractures and pores are broken into smaller
 263 pores. (b) Pore deformation, which mainly includes pore expansion and pore compression. The blue parts in
 264 the figures are fracturing fluid.

265 The CZ coal samples have a smaller elastic modulus (3.432 GPa) and a larger Poisson's ratio
 266 (0.297), it means that under the same stress conditions, CZ coal samples are more likely to be
 267 squeezed and deformed than SH coal samples (Figure 3b). During the hydraulic fracturing
 268 process, as the hydraulic pressure in the coal seam gradually increased, the pores of the CZ coal
 269 gradually expanded and deformed. Part of the pores increased to 3000–400000 nm, resulting in
 270 an increase in the number of pores in the > 3000 nm (Figure 2h), the other pores continued to
 271 expand to reach the crushing strength of CZ coal, which was transformed into microfractures.

272 Pore expansion compressed the space of pores that cannot be connected, the pores caused by
273 crushing and pore compression were the reasons for the increase in the number of pores in the
274 50–1000 nm. Similarly, during the hydraulic fracturing process of CZ coal, the pore size range
275 that the fracturing fluid can affect was limited. CZF-1 showed a decrease in the number of pores
276 in 20–50 nm, it might be the minimum pore diameter range that hydraulic fracturing can affect.
277 Under the action of hydraulic pressure, some of the pores in the 20–50 nm expanded, and the
278 pore diameter became larger, at the same time, more pores were compressed, resulting in a sharp
279 increase in the number of pores < 20 nm (Figure 2e and Figure 2f). For CZF-2 sample, the
280 smallest pore size range that can be affected might be smaller (4–50 nm). Part of the 4–50 nm
281 pores expanded, and the pore diameter became larger, resulting in an increase in the number of
282 pores at > 50 nm (Figure 2f), the other pores were compressed, resulting in an increase in the
283 number of pores at < 4 nm (Figure 2e). Obviously, the change in the number of pores in CZF-2
284 was smaller than that in CZF-1, which might be related to the in-situ stress conditions.

285 3.3 Coal elastic modulus and coal pore stimulation after hydraulic fracturing

286 To quantitatively analyze the influence of coal physical properties on hydraulic fracturing,
287 considering that the microscopic material composition of coal and the development of original
288 pore fractures will affect the elastic modulus of coal (Pan et al., 2013), the elastic modulus was
289 chosen as the representative of the physical property of coal reservoir.

290 With reference to Figure 1, the PV of SHF coal changed drastically, while the PSSA of CZF coal
291 changed drastically after hydraulic fracturing. Considering that SH coal has a larger elastic
292 modulus (4.206 GPa), it is mainly because the greater the elastic modulus, the worse the
293 resistance of coal to deformation under a given pressure, the coal is easily broken during
294 hydraulic fracturing (Li et al., 2012). The crushing of pore will mainly affect the mesopore and

295 macropore with larger scale. The fragmentation and connection of mesopores and macropores
296 increased the pore connectivity of SHF samples, which in turn connected more transition pores
297 and micropores. However, the hydraulic pressure was not enough to have a greater impact on the
298 micropores and transition pores due to the larger elastic modulus of coal. CZ coal has a smaller
299 elastic modulus (3.432 GPa), pore deformation was the main form of hydraulic fracturing micro-
300 nano-scale stimulation of CZ coal, which was mainly manifested in the expansion and
301 compression of pores. In the process of hydraulic fracturing, less pore fragmentation determined
302 that the connectivity between pores had little change. The extension and expansion of
303 microfractures squeezed the pore space, resulting in the tendency of pores to become smaller
304 pores after the formation of hydraulic fractures. Therefore, the PV of micropores of CZF-1 and
305 CZF-2 samples increased after hydraulic fracturing.

306 In addition, considering that coal has extremely strong heterogeneity (Gibbins et al., 1999; Day
307 et al., 2008; Chen et al., 2013), it cannot be ruled out that the significant increase in macropores
308 PV of CZF-1 was caused by heterogeneity, although the influence of heterogeneity on the
309 experiment was minimized when sampling.

310 3.4 Influence of in-situ stress on pore stimulation after hydraulic fracturing

311 The effect of high confining pressure on the fracture of low stiffness results in the rapid decrease
312 of hydraulic aperture, permeability and conductivity (Shu et al., 2020). Similarly, the larger in-
313 situ stress will inhibit the pore crushing and expansion, and even cause the pore further
314 compression.

315 Considering that the macropores were directly affected by in-situ stress and hydraulic pressure, it
316 was difficult to define the influence of in-situ stress on macropores, the changes of micropores,
317 transition pores and mesopores were strongly affected by in-situ stress. According to Figure 2, it

318 was obvious that under the condition of larger in-situ stress, the number of pores increased less,
319 or even decreased. According to Figure 1, the change ranges of PV and PSSA of micropores,
320 transition pores and mesopores of SHF-1 and CZF-1 samples (σ_H : 15 MPa, σ_h : 11 MPa, σ_V : 20
321 MPa) were slightly larger than that of SHF-2 and CZF-2 samples (σ_H : 19 MPa, σ_h : 11 MPa, σ_V :
322 20 MPa) after hydraulic fracturing. Considering that the same coal sample initiation pressure was
323 basically at the same level under different in-situ stress conditions, according to the principle of
324 effective stress, the greater the in-situ stress, the greater the effective stress that the coal will
325 withstand under the similar condition of pore pressure, resulting in the suppression of pores
326 expansion and crushing, and even the compression of pores.

327 **4 Conclusions**

328 In order to find out the influence of hydraulic fracturing on the micro-nano-scale pores of
329 coalbed methane (CBM) reservoirs, which is the core issue for the efficient development of
330 CBM and the prevention of coal mine gas outburst, on the basis of hydraulic fracturing
331 experiments on Chengzhuang (CZ) and Sihe (SH) coal samples, liquid nitrogen absorption and
332 mercury intrusion porosity were used to analyze the pores before and after hydraulic fracturing.
333 It was found that the changes in coal pore volume (PV) and pore specific surface area (PSSA)
334 were caused by changes in the number of pores after hydraulic fracturing. In particular, the
335 number of transition pores and mesopores increased significantly. The changes of pore number,
336 PV and PSSA of micropores and macropores were controlled by the elastic modulus,
337 heterogeneity and in-situ stress conditions of coal: The difference of elastic modulus determines
338 the hydraulic fracturing stimulation form of micro-nano-scale pore. The larger elastic modulus
339 means that coal is dominated by crushing, it is conducive to the improvement of coal pore
340 connectivity under the same in-situ stress conditions. Smaller elastic modulus means that pore

341 deformation is the main factor in the process of hydraulic fracturing. In-situ stress will affect
342 pore crushing, expansion and compression, it is mainly shown that the larger in-situ stress will
343 inhibit the pore crushing and expansion, and even cause the pore further compression.

344 **Acknowledgments**

345 This study was sponsored by the National Natural Science Foundation of China (Grant No.
346 41772162), the National Science and Technology Major Project of China (2017ZX05064,
347 2016ZX05066) the Fundamental Research Funds for the Universities of Henan Province
348 (NSFRF180103), the Program for Innovative Research Team of Henan Polytechnic University
349 (T2015-1) and the “Climate Change: Carbon Budget and Related Issues” Strategic Priority
350 Research Program of the Chinese Academy of Sciences (Grant No. XDA05030100).

351 **Data Availability Statement**

352 Data can be found in the Global Change Research Data Publishing and Repository (GCdataPR)
353 at <http://www.geodoi.ac.cn/WebEn/doi.aspx?Id=1616>

354 **References**

- 355 Barrett, E. P., Joyner, L. G., & Halenda, P. P. (1951). The Determination of PV and Area
356 Distributions in Porous Substances. I. Computations from Nitrogen Isotherms. *Journal of*
357 *the American Chemical Society*, 73(1), 373–380.
- 358 Busch, A., & Gensterblum, Y. (2011). CBM and CO₂-ECBM related sorption processes in coal:
359 A review. *International Journal of Coal Geology*, 87(2), 49–71.
- 360 Cai, Y. D., Liu, D. M., Pan, Z. J., Yao, Y. B., Li, J. Q., & Qiu, Y. K. (2013). Petrophysical
361 characterization of Chinese coal cores with heat treatment by nuclear magnetic resonance.
362 *Fuel*, 108, 292–302.

- 363 Chen, Z. W., Liu, J. S., Elsworth, D., Pan, Z. J., & Wang, S. G. (2013). Roles of coal
364 heterogeneity on evolution of coal permeability under unconstrained boundary conditions.
365 *Journal of Natural Gas Science and Engineering*, 15, 38–52.
- 366 Clarkson, C. R., & Bustin, R. M. (1999). The effect of pore structure and gas pressure upon the
367 transport properties of coal: a laboratory and modeling study. 1. Isotherms and PV
368 distributions. *Fuel*, 78(11), 1333–1344.
- 369 Day, S., Fry, R., & Sakurovs, R. (2008). Swelling of Australian coals in supercritical CO₂.
370 *International Journal of Coal Geology*, 74(1), 41–52.
- 371 Dehghan, A. N., Goshtasbi, K., Ahangari, K., & Jin, Y. (2016). Mechanism of fracture initiation
372 and propagation using a tri-axial hydraulic fracturing test system in naturally fractured
373 reservoirs. *European Journal of Environmental and Civil Engineering*, 20(5), 560–585.
- 374 Fan, C. J., Elsworth, D., Li, S., Zhou, L. J., Yang, Z. H., & Song, Y. (2019). Thermo-hydro-
375 mechanical-chemical couplings controlling CH₄ production and CO₂ sequestration in
376 enhanced coalbed methane recovery. *Energy*, 173, 1054-1077.
- 377 Gensterblum, Y., Merkel, A., Busch, A., Krooss, B. M., & Littke, R. (2014). Gas saturation and
378 CO₂ enhancement potential of coalbed methane reservoirs as a function of depth. *AAPG
379 Bull*, 98(2), 395–420.
- 380 Gibbins, J. R., Beeley, T. J., Crelling, J. C., Scott, A. C., Skorupska, N. M., & Williamson, J.
381 (1999) Observations of Heterogeneity in Large Pulverized Coal Particles. *Energy & Fuels*,
382 13(3), 592–601.
- 383 Hodot, B. B. (1966). *Outburst of Coal and Coalbed Gas (Chinese Translation)*. Beijing: China
384 Industry Press, p. 318.

- 385 Hou, S. H., Wang, X. M., Wang, X. J., Yuan, Y. D., Pan, S. D., & Wang, X. M. (2017). Pore
386 structure characterization of low volatile bituminous coals with different particle size and
387 tectonic deformation using low pressure gas adsorption. *International Journal of Coal
388 Geology*, 183, 1–13.
- 389 Ju, Y. W., Huang, C., Sun, Y., Wan, Q., Lu, X. C., Lu, S. F., et al. (2017). Nanogeosciences:
390 Research history, current status, and development trends. *Journal of Nanoscience and
391 Nanotechnology*, 17(9), 5930–5965.
- 392 Ju, Y. W., Huang, C., Sun, Y., Zou, C. N., He, H. P., Wan, Q., et al. (2018). Nanogeology in
393 China: A review. *China Geology*, 1, 286-303.
- 394 Ju, Y. W., Jiang, B., Hou, Q. L., Tan, Y. J., Wang, G. L., & Xiao, W. J. (2009). Behavior and
395 mechanism of the adsorption/desorption of tectonically deformed coals. *Chinese Science
396 Bulletin*, 54(1), 88-94.
- 397 Kumar, H., Lester, E., Kingman, S., Bourne, R., Avila, C., Jones, A., et al. (2011). Inducing
398 fractures and increasing cleat apertures in a bituminous coal under isotropic stress via
399 application of microwave energy. *International Journal of Coal Geology*, 88(1), 75–82.
- 400 Li, C. W., Xie, B. J., Yang, W., & Wei, S. Y. (2012). Characteristics of Transient Magnetic
401 Nearby Field in Process of Coal Impact Damage. *Chinese Journal of Rock Mechanics and
402 Engineering*, 31(05), 973–981.
- 403 Li, D. Q., Zhang, S. C., & Zhang, S. A. (2014). Experimental and numerical simulation study on
404 fracturing through interlayer to coal seam. *Journal of Natural Gas Science and Engineering*,
405 21, 386–396.

- 406 Li, Q. S., & Xing, H. L. (2015). Numerical analysis of the material parameter effects on the
407 initiation of hydraulic fracture in a near wellbore region. *Journal of Natural Gas Science and*
408 *Engineering*, 27, 1597–1608.
- 409 Li, S., Tang, D. Z., Xu, H., & Yang, Z. (2012). The pore-fracture system properties of coalbed
410 methane reservoirs in the Panguan Syncline, Guizhou, China. *Geoscience Frontiers*, 3(6),
411 853–862.
- 412 Li, X. C., Kang, Y. L., & Haghghi, M. (2018). Investigation of pore size distributions of coals
413 with different structures by nuclear magnetic resonance (NMR) and mercury intrusion
414 porosimetry (MIP). *Measurement*, 116, 122–128.
- 415 Li, Y. H., Lu, G. Q., & Rudolph, V. (1999). Compressibility and Fractal Dimension of Fine Coal
416 Particles in Relation to Pore Structure Characterisation Using Mercury Porosimetry. *Particle*
417 *& Particle Systems Characterization*, 16(1), 25–31.
- 418 Liang, M. L., Wang, Z. X., Gao, L., Li, C. L., & Li, H. J. (2017). Evolution of pore structure in
419 gas shale related to structural deformation. *Fuel*, 197, 310–319.
- 420 Liu, J. C., Wang, H. T., Yuan, Z. G., & Fan, X. G. (2011). Experimental Study of Pre-splitting
421 Blasting Enhancing Pre-drainage Rate of Low Permeability Heading Face. *Procedia*
422 *Engineering*, 26, 818–823.
- 423 Liu, J. S., Chen, Z. W., Elsworth, D., Qu, H. Y., & Chen, D. (2011). Interactions of multiple
424 processes during CBM extraction: A critical review. *International Journal of Coal Geology*,
425 87(3-4), 175–189.
- 426 Liu, Q. Q., Cheng, Y. P., Zhou, H. X., Guo, P. K., An, F. H., & Chen, H. D. (2015). A
427 Mathematical Model of Coupled Gas Flow and Coal Deformation with Gas Diffusion and
428 Klinkenberg Effects. *Rock Mechanics and Rock Engineering*, 48(3), 1163–1180.

- 429 Liu, S. M., & Harpalani, S. (2014). Determination of the Effective Stress Law for Deformation in
430 Coalbed Methane Reservoirs. *Rock Mechanics and Rock Engineering*, 47(5), 1809–1820.
- 431 Lu, Y. Y., Wang, L., Ge, Z. L., Zhou, Z., Deng, K., & Zuo, S. J. (2020). Fracture and pore
432 structure dynamic evolution of coals during hydraulic fracturing. *Fuel*, 259, 116272.
- 433 Ma, S., Guo, J. C., Li, L. C., Xia, Y. J., & Yang, T. (2016). Experimental and numerical study on
434 fracture propagation near open-hole horizontal well under hydraulic pressure. *European
435 Journal of Environmental and Civil Engineering*, 20(4), 412–430.
- 436 Mastalerz, M., He, L. L., Melnichenko, Y. B., & Rupp, J. A. (2012). Porosity of Coal and Shale:
437 Insights from Gas Adsorption and SANS/USANS Techniques. *Energy & Fuels*, 26(8),
438 5109–5120.
- 439 Pan, J. N., Meng, Z. P., Hou, Q. L., Ju, Y. W., & Cao, Y. X. (2013). Coal strength and Young's
440 modulus related to coal rank, compressional velocity and maceral composition. *Journal of
441 Structural Geology*, 54, 129–135.
- 442 Pan, J. N., Zhu, H. T., Hou, Q. L., Wang, H. C., & Wang, S. (2015). Macromolecular and pore
443 structures of Chinese tectonically deformed coal studied by atomic force microscopy. *Fuel*,
444 139, 94–101.
- 445 Shen, C. M., Lin, B. Q., Sun, C., Zhang, Q. Z., & Li, Q. Z. (2015). Analysis of the stress–
446 permeability coupling property in water jet slotting coal and its impact on methane
447 drainage. *Journal of Petroleum Science and Engineering*, 126, 231–241.
- 448 Shi, Q. M., Qin, Y., Li, H. L., Qiu, A. C., Zhang, Y. M., & Zhou, X. T., et al. (2016). Response
449 of pores in coal to repeated strong impulse waves. *Journal of Natural Gas Science and
450 Engineering*, 34, 298–304.

- 451 Shi, R., Liu, J. S., Wang, X. M., Elsworth, D., Liu, Z. H., Wei, M. Y., et al. (2020). Experimental
452 observations of heterogeneous strains inside a dual porosity sample under the influence of
453 gas-sorption: A case study of fractured coal. *International Journal of Coal Geology*, 223,
454 103450.
- 455 Shu, B., Zhu, R. J., Elsworth, D., Dick, J., Liu, S., Tan, J. Q., et al. (2020). Effect of temperature
456 and confining pressure on the evolution of hydraulic and heat transfer properties of
457 geothermal fracture in granite. *Applied Energy*, 272, 115290.
- 458 Tan, P., Jin, Y., Hou, B., Han, K., Zhou, Y. C., & Meng, S. Z. (2017). Experimental
459 investigation on fracture initiation and non-planar propagation of hydraulic fractures in coal
460 seams. *Petroleum Exploration and Development Online*, 44(3), 470-476.
- 461 Wang, K., Pan, J. N., Wang, E. Y., Hou, Q. L., Yang, Y. H., & Wang, X. L. (2020). Potential
462 impact of CO₂ injection into coal matrix in molecular terms. *Chemical Engineering Journal*,
463 401, 126071.
- 464 Xie, J. N., Xie, J., Ni, G. H., Rahman, S., Sun, Q., & Wang, H. (2020). Effects of pulse wave on
465 the variation of coal pore structure in pulsating hydraulic fracturing process of coal seam.
466 *Fuel*, 264, 116906.
- 467 Zhang, D. F., Gu, L. L., Li, S. G., Lian, P. C., & Tao, J. (2012). Interactions of Supercritical CO₂
468 with Coal. *Energy & Fuels*, 27(1), 387–393.
- 469 Zhang, J. W., & Li, Y. L. (2017). Ultrasonic vibrations and coal permeability: Laboratory
470 experimental investigations and numerical simulations. *International Journal of Mining
471 Science and Technology*, 27(2), 221–228.

472 Zhang, Y. S., Zhang, J. C., Yuan, B., & Yin, S. X. (2018). In-situ stresses controlling hydraulic
473 fracture propagation and fracture breakdown pressure. *Journal of Petroleum Science and*
474 *Engineering*, 164, 164-173.

475 Zhou, J., Chen, M., Jin, Y., & Zhang, G. Q. (2008). Analysis of fracture propagation behavior
476 and fracture geometry using a tri-axial fracturing system in naturally fractured reservoirs.
477 *International Journal of Rock Mechanics and Mining Sciences*, 45(7), 1143–1152.

478 Zhou, S. D., Liu, D. M., Cai, Y. D., & Yao, Y. B. (2016). Fractal characterization of pore–
479 fracture in low-rank coals using a low-field NMR relaxation method. *Fuel*, 181, 218–226.