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Experimental Study of Supercritical CO_2 Injected into Water Saturated Medium Rank Coal by X-ray MicroCT

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Abstract

Carbon dioxide geosequestration into deep unmineable coal seams is a technique which can mitigate anthropogenic greenhouse gas emissions. However, coal composition is always complex, and some minerals such as calcite chemically react when exposed to the acidic environment (which is created by $scCO_2$ mixing with formation water). These reactive transport processes are still poorly understood. We thus imaged a water-bearing heterogeneous coal (calcite rich) core before and after $scCO_2$ injection in-situ at high resolutions (3.43 µm) in 3D via X-ray in-situ microCT flooding system. Indeed, the calcite-coal mixed layer was partially dissolved, and absolute porosity and connectivity significantly increased. We thus suggested that such process could be used as an acidizing method in CO_2 ECBM. However, such dissolved damage also can significantly affect the rock mechanical properties and potentially induce geohazards.

Keywords: geosequestration; coal; ECBM; microCT; core flooding

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1. Introduction

CO_2 injected into deep unmineable coal seams for enhanced coalbed methane recovery and combine with carbon storage have gained substantial interest in recent years [1- 6]. Technically, CO_2 migrates into coal matrix micro/nano pores and displaces the original methane, at the same time, the CO_2 traps inside the matrix by adsorption trapping mechanism and closes the potential leakage route (cleats/factures) by swelling effect. Such coal matrix – CO_2 interaction induced swelling is complex and can significantly change the physical properties of the host rock, e.g. many studies proved that the CO_2 injection reduces the coal seam permeability [7-12]. Zhang et al., 2016 [12] injected $scCO_2$ into coal sample with permeability dramatically reducing recorded and micro fractures closed were observed by X-ray tomography. However, most of the experiments were conducted in the dry conditions and the previous models did not care about the existed formation water influence [13-16].

Note that the injected CO_2 dissolved into formation water called CO_2 – saturated brine is acidic at reservoir conditions where the pH values could be dropped to 3-4 [17, 18]. Moreover, the coal material is always heterogenous and consists of organic carbon and inorganic materials such as carbonate, quartz and clay [19, 20]. Especially for the coal from the low to medium rank group, such inorganic materials are abundant. However, such inorganic materials (mainly carbonates) may sensitive to the acid environment and the effect to the sample and the related petrophysical properties change need to be clarified. In this study, we thus used the newly in-situ microCT flooding system [21-23] has been used to investigate how the coal microstructure change by $scCO_2$ injected into water-bearing coal medium rank coal (calcite rich) sample in reservoir condition.

2. Methodology

2.1. Materials

The special coal sample which contained calcite was selected in this experimental study, obtained from the Pingdingshan coal mine, China. The coal had been identified as sub-bituminous containing 36 % (± 1%) volatile matter and 54 % (± 2%) fixed carbon content by Chinese standard GB/T 212 -2008 and DL/T 1030-2006. Fig. 1 showed the SEM image of the coal microstructure. A cylindrical coal plug (5 mm diameter and 10 mm length) was drilled for the following microCT in-situ flooding test.

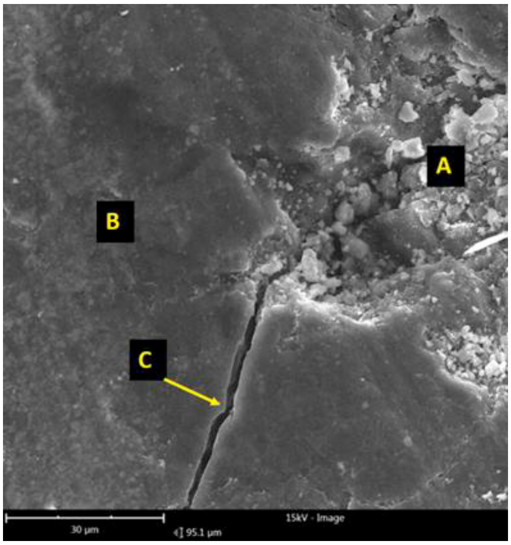

Fig. 1. The SEM image of the coal sample; (A) the calcite particles inside the coal matrix, (B) the coal matrix, (C) the micro fractures.

2.2. MicroCT in-situ flooding test

The microCT in-situ scanning techniques developing in recent years give us the opportunity to observe the microstructures change by fluid-rock interaction in 3D at reservoir conditions, where the resolution could achieve as high as less than 10 µm voxel size (e.g. [24-30]). Such technique successfully overcomes the shortcomings of the traditional imaging tools such as SEM which only can obtain 2D surface images at laboratory conditions. Here the cylindrical coal plug was mounted inside an novel X-ray transparent core holder [12, 22, 23], which as a part of the microCT in-situ imaging core flooding system in Curtin University, Australia, see Fig. 2. This core flooding system included two parts: the microCT instrument itself (Xradia VersaXRM instrument, a 2000 × 2000 pixel detector was used and the X-ray accelerating voltage set as 60 kV in this experiment), and the high pressure – high temperature (HPHT) flooding system. The in-situ microCT core flooding test was then conducted by the below steps (also see [21, 23]):

1. The coal plug was saturated with 5 wt% NaCl brine before the coal sample amounted into the coal holder (coal plug merged into the brine water with vacuumed 1 week).
2. After coal plug mounted into core holder, the tubes and coal plug were injected 5 wt% NaCl brine by backpressure pump (Teledyne ISCO 500D, B in Fig. 2), then the coal plug and tubing system were vacuumed for 24 hours to air removed; all flow tubes and pumps were continuously isothermally heated to 50°C (323 K) with heat jackets by continuously circulating warm water. The core holder was heated by electric tape.
3. Initially the saturated coal plug was imaged (voxel size: 3.43 µm) under a confining pressure of 5 MPa (without fluid injection, thus 5 MPa effective stress). The confining pressure was applied by compressing deionized (DI) water by pump C in Fig. 2.
4. Then the coal plug was flooded by supercritical CO_2 ($scCO_2$) at 10 MPa pore pressure (backpressure pump, B in Fig. 2), and a 15 MPa confining pressure was applied (i.e. the experiment was conducted at a constant effective stress of 5 MPa). The injection flow rate was 0.25 ml/min with approximately 100 pore volumes were flooded.
5. The sample was then microCT imaged again at the same condition with 15 MPa confining pressure, 10 MPa pore pressure, and temperature of 50°C / 323K.

All the obtained grayscale tomograms were then filtered by 3D non-local means filter [31] method to image denoising, and the watershed algorithm [32] was used for phase segmentation according to their (different) relative radio-densities. Finally, the different phase could be extracted in 3D for the further qualitative and quantitative analysis [24]. All the images processing used the Avizo 9.2 software which provided by the Pawsey Supercomputing Centre, Australia.

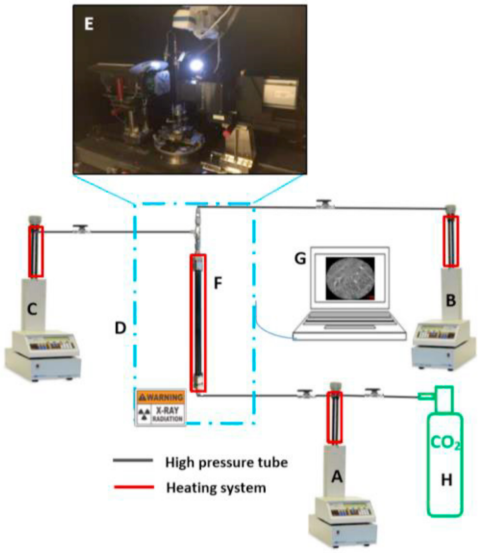

Fig. 2. High pressure-High temperature (HPHT) in-situ microCT coreflooding apparatus: (A) CO_2 injection pump, (B) back pressure pump, (C) confining pressure pump, (D) microCT (Xradia VersaXRM instrument), (E) the photo for inside microCT, (F) the X-ray transparent core holder assembly, (G) microCT image processing, (H) CO_2 cylinder.

3. Results and discussion

The microCT images showed a very highly heterogenous morphology for such coal sample. In the water saturated tomography (see Fig. 3), it can be distinguished as three parts: the coal matrix (dark grey – low CT number), the calcite mineral (white – high CT number), and calcite-coal mixed layer (grey – medium CT number). Not that, this CT image did not show any micro fractures/cleats system which were presented in the former studies for dry coal sample [33, 34] and also the dry SEM image (Fig. 1), this may due to the micro fractures/cleats are closed due to water adsorption by coal matrix swelling effect [24].

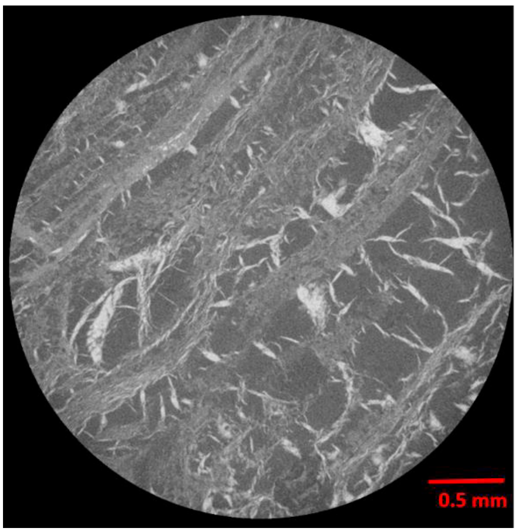

Fig. 3. The microCT image (3.43 μm voxel size) for the coal sample (brine saturated) where coal matrix is dark grey (low CT number), the calcite mineral is white (high CT number), and calcite-coal mixed layer is grey (medium CT number).

After $scCO_2$ injected into the water-bearing coal sample, the chemical– dissolved effect was significant (see Fig. 4). The dissolved wormhole clearly presented in the 3D images (B in Fig. 4). Obviously, the CT scanning results by $scCO_2$ flooded into water-bearing coal were totally different with the previously dry coal – $scCO_2$ injection (see [12] where $scCO_2$ induced swelling effect on same coal sample at dry condition); the significant dissolution happed from our experiments. The calcite minerals chemical reacted during the carbon geosequestration by the following equations [35, 36]:

$$H_2O + scCO_2 \leftrightarrow H_2O + CO_2(aq)$$
$$H_2O + CO_2(aq) \leftrightarrow H_2CO_3$$
$$H_2CO_3 \leftrightarrow HCO_3^- + H^+$$
$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+$$
$$CaCO_3 + H^+ \leftrightarrow Ca^{2+} + HCO_3^-$$
$$CaCO_3 + H_2CO_3 \leftrightarrow Ca^{2+} + 2HCO_3^-$$

The dissolved area showed the heterogenous characteristics where most located in the calcite-coal mixed layer. This could be explained by the less consolidated of this phase in nature. The 3D segmented dissolved hole presented in Fig. 5, which represented 2.78% volume fraction.

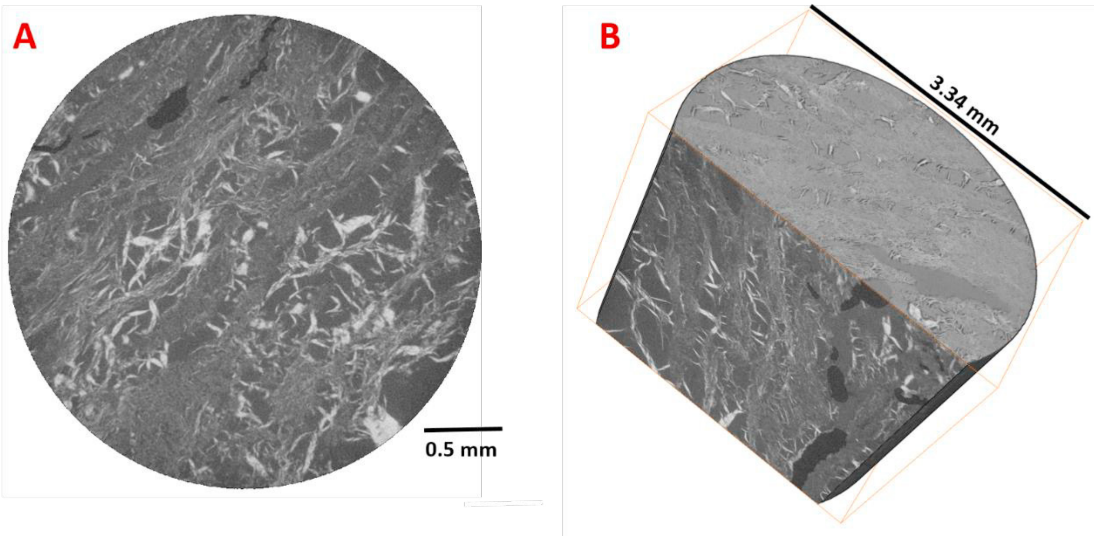

Fig. 4. The microCT image (3.43 μm voxel size) for the coal sample after $scCO_2$ flooding where the dissolved area is black (lowest CT number), coal matrix is dark grey (low CT number), the calcite mineral is white (high CT number), and calcite-coal mixed layer is grey (medium CT number); (A) is the 2D slice, (B) is the 3D cut view.

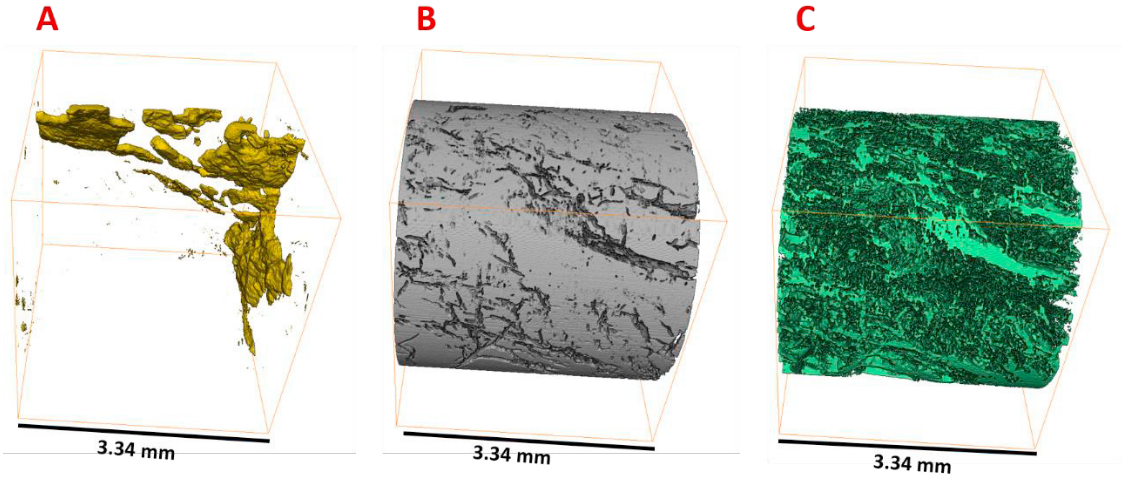

Fig. 5. The 3D segmented microCT image (3.43 µm voxel size) for the coal sample after $scCO_2$ flooding, (A) dissolved area, (B) coal matrix and calcite-coal mixed layer, (C) calcite mineral phase.

In summary, such dissolved area largely increasing the porosity and improved the connectivity of the coal sample. We thus suggested such process could be an environmental friendly acidizing method in the CO_2 ECBM. At the same time, such damaged can significant affect the rock mechanical properties of the coalbed – may cause geohazards, e.g. the layer collapse and fault reaction – which has been seriously considered at the carbon geosequestration in carbonate reservoir.

4. Conclusion

Carbon geosequestration in deep geological formations has been suggested as the most efficient ways to mitigate climate change with reducing the CO_2 concentration from the atmosphere [37-40]. The deep unmineable coal seams are some of the main targets; however, the injected CO_2 with formation water induce an acidic environment – may impact some acidic sensitive minerals (such as calcite) in coal seam, which are still poorly investigated.

We thus investigated such interactions in pore-scale by 3D microCT in-situ core flooding experiments, the $scCO_2$ was injected into a heterogeneous water-bearing bituminous coal at reservoir conditions (15 MPa confining pressure, 10 MPa pore pressure, and 323 K). The coal's microstructure (calcite-coal mixed layer) partially dissolved after the flooding test. We thus concluded that such dissolved area largely increasing the porosity and improved the connectivity of the coal seam. We also suggested such process could be an environmental friendly acidizing method in the CO_2 ECBM in some calcite rich coal seams. Moreover, such dissolved damage also can significant affect the rock mechanical properties of the coalbed which can induce potential geohazards, e.g. the layer collapse and fault reaction

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References

[1] Al-Yaseri AZ, Roshan H, Xu X, Zhang Y, Sarmadivaleh M, Lebedev M, et al. Coal Wettability After CO2 Injection. Energy & Fuels. 2017;31(11):12376-82.

[2] Zhang Y, Zhang Z, Sarmadivaleh M, Lebedev M, Barifcani A, Yu H, et al. Micro-scale fracturing mechanisms in coal induced by adsorption of supercritical CO2. International Journal of Coal Geology. 2017;175:40-50.

[3] Yang D, Koukouzas N, Green M, Sheng Y. Recent development on underground coal gasification and subsequent CO2 storage. Journal of the Energy Institute. 2016;89(4):469-84.

[4] Perera M, Ranjith P, Ranathunga A. Challenges and issues for CO2 storage in deep coal seams. Rock Mechanics and Engineering Volume 4: CRC Press; 2017. p. 87-119.

[5] Pan Z, Ye J, Zhou F, Tan Y, Connell LD, Fan J. CO2 storage in coal to enhance coalbed methane recovery: a review of field experiments in China. International Geology Review. 2018;60(5-6):754-76.

[6] Zhang Y. Microstructure and Nanoscale Rock Mechanical Properties of Coal: Applications to CO2 Storage: Curtin University; 2017.

[7] Siriwardane H, Haljasmaa I, McLendon R, Irdi G, Soong Y, Bromhal G. Influence of carbon dioxide on coal permeability determined by pressure transient methods. International Journal of Coal Geology. 2009;77(1-2):109-18.

[8] Perera M, Ranjith P, Choi S, Airey D. The effects of sub-critical and super-critical carbon dioxide adsorption-induced coal matrix swelling on the permeability of naturally fractured black coal. Energy. 2011;36(11):6442-50.

[9] Ranathunga A, Perera M, Ranjith P, Ju Y, Vishal V, De Silva P. A macro-scale experimental study of sub-and super-critical CO2 flow behaviour in Victorian brown coal. Fuel. 2015;158:864-73.

[10] Vishal V. In-situ disposal of CO2: Liquid and supercritical CO2 permeability in coal at multiple down-hole stress conditions. Journal of CO2 Utilization. 2017;17:235-42.

[11] Ranathunga A, Perera M, Ranjith P, Wei C. An experimental investigation of applicability of CO2 enhanced coal bed methane recovery to low rank coal. Fuel. 2017;189:391-9.

[12] Zhang Y, Lebedev M, Sarmadivaleh M, Barifcani A, Iglauer S. Swelling- induced changes in coal microstructure due to supercritical CO2 injection. Geophysical Research Letters. 2016;43(17):9077-83.

[13] An H, Wei X, Wang G, Massarotto P, Wang F, Rudolph V, et al. Modeling anisotropic permeability of coal and its effects on CO2 sequestration and enhanced coalbed methane recovery. International Journal of Coal Geology. 2015;152:15-24.

[14] Liu H-H, Rutqvist J. A new coal-permeability model: internal swelling stress and fracture–matrix interaction. Transport in Porous Media. 2010;82(1):157-71.

[15] Wang G, Massarotto P, Rudolph V. An improved permeability model of coal for coalbed methane recovery and CO2 geosequestration. International Journal of Coal Geology. 2009;77(1-2):127-36.

[16] Liu S, Wang Y, Harpalani S. Anisotropy characteristics of coal shrinkage/swelling and its impact on coal permeability evolution with CO2 injection. Greenhouse Gases: Science and Technology. 2016;6(5):615-32.

[17] Deng H, Fitts JP, Crandall D, McIntyre D, Peters CA. Alterations of fractures in carbonate rocks by CO2-acidified brines. Environmental science & technology. 2015;49(16):10226-34.

[18] Menke H, Bijeljic B, Blunt M. Dynamic reservoir-condition microtomography of reactive transport in complex carbonates: Effect of initial pore structure and initial brine pH. Geochimica et Cosmochimica Acta. 2017;204:267-85.

[19] Zhang Y, Lebedev M, Al-Yaseri A, Yu H, Xu X, Iglauer S. Characterization of nanoscale rockmechanical properties and microstructures of a Chinese sub-bituminous coal. Journal of Natural Gas Science and Engineering. 2018;52:106-16.

[20] Yu H, Zhang Y, Lebedev M, Han T, Verrall M, Wang Z, et al. Nanoscale geomechanical properties of Western Australian coal. Journal of Petroleum Science and Engineering. 2018;162:736-46.

[21] Zhang Y, Lebedev M, Al-Yaseri A, Yu H, Nwidee LN, Sarmadivaleh M, et al. Morphological evaluation of heterogeneous oolitic limestone under pressure and fluid flow using X-ray microtomography. Journal of Applied Geophysics. 2018;150:172-81.

[22] Lebedev M, Zhang Y, Mikhaltsevitch V, Inglauer S, Rahman T. Residual trapping of supercritical CO2: direct pore-scale observation using a low cost pressure cell for micro computer tomography. Energy Procedia. 2017;114:4967-74.

[23] Lebedev M, Zhang Y, Sarmadivaleh M, Barifcani A, Al-Khdheeawi E, Iglauer S. Carbon geosequestration in limestone: Pore-scale dissolution and geomechanical weakening. International Journal of Greenhouse Gas Control. 2017;66:106-19.

[24] Zhang Y, Lebedev M, Sarmadivaleh M, Barifcani A, Rahman T, Iglauer S. Swelling effect on coal micro structure and associated permeability reduction. Fuel. 2016;182:568-76.

[25] Zhang Y, Xu X, Lebedev M, Sarmadivaleh M, Barifcani A, Iglauer S. Multi-scale x-ray computed tomography analysis of coal microstructure and permeability changes as a function of effective stress. International Journal of Coal Geology. 2016;165:149-56.

[26] Roshan H, Masoumi H, Zhang Y, Al-Yaseri AZ, Iglauer S, Lebedev M, et al. Microstructural effects on mechanical properties of shaly sandstone. Journal of Geotechnical and Geoenvironmental Engineering. 2017;144(2):06017019.

[27] Shi X, Pan J, Hou Q, Jin Y, Wang Z, Niu Q, et al. Micrometer-scale fractures in coal related to coal rank based on micro-CT scanning and fractal theory. Fuel. 2018;212:162-72.

[28] Jing Y, Armstrong RT, Ramandi HL, Mostaghimi P. Coal cleat reconstruction using micro-computed tomography imaging. Fuel. 2016;181:286-99.

[29] Iglauer S, Lebedev M. High pressure-elevated temperature x-ray micro-computed tomography for subsurface applications. Advances in Colloid and Interface Science. 2017.

[30] Khanamiri HH, Torsæter O. Fluid Topology in Pore Scale Two- Phase Flow Imaged by Synchrotron X- ray Microtomography. Water Resources Research.
[31] Buades A, Coll B, Morel J-M, editors. A non-local algorithm for image denoising. Computer Vision and Pattern Recognition, 2005 CVPR 2005 IEEE Computer Society Conference on; 2005: IEEE.
[32] Schlüter S, Sheppard A, Brown K, Wildenschild D. Image processing of multiphase images obtained via X- ray microtomography: a review. Water Resources Research. 2014;50(4):3615-39.
[33] Zhang Y, Lebedev M, Al-Yaseri A, Yu H, Xu X, Sarmadivaleh M, et al. Nanoscale rock mechanical property changes in heterogeneous coal after water adsorption. Fuel. 2018;218:23-32.
[34] Ramandi HL, Mostaghimi P, Armstrong RT, Saadatfar M, Pinczewski WV. Porosity and permeability characterization of coal: a micro-computed tomography study. International Journal of Coal Geology. 2016;154:57-68.
[35] Busenberg E, Plummer L. A comparative study of the dissolution and crystal growth kinetics of calcite and aragonite. Studies in diagenesis. 1578: US Geol. Surv. Bull; 1986. p. 139-68.
[36] Iglauer S. Dissolution trapping of carbon dioxide in reservoir formation brine–a carbon storage mechanism. Mass Transfer-Advanced Aspects: InTech; 2011.
[37] Metz B. Carbon dioxide capture and storage: special report of the intergovernmental panel on climate change: Cambridge University Press; 2005.
[38] Al- Khdheeawi EA, Vialle S, Barifcani A, Sarmadivaleh M, Zhang Y, Iglauer S. Impact of salinity on CO2 containment security in highly heterogeneous reservoirs. Greenhouse Gases: Science and Technology. 2018;8(1):93-105.
[39] Al-Khdheeawi EA, Vialle S, Barifcani A, Sarmadivaleh M, Iglauer S. Influence of injection well configuration and rock wettability on CO2 plume behaviour and CO2 trapping capacity in heterogeneous reservoirs. Journal of Natural Gas Science and Engineering. 2017;43:190-206.
[40] Al-Yaseri A, Zhang Y, Ghasemiziarani M, Sarmadivaleh M, Lebedev M, Roshan H, et al. Permeability Evolution in Sandstone Due to CO2 Injection. Energy & Fuels. 2017;31(11):12390-8.