Journal of Mechanical Engineering Research and Developments
ISSN: 1024-1752
Website: http://www.jmerd.org
Vol. 39, No. 2, 2016, pp. 394-402

Understanding of Gas Storage and Migration Pathways in Shales: Example from Pore Characteristics of Gufeng Formation

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ABSTRACT: To evaluate the hydrocarbon generation capacity and gas storage capacity of shale series，it is important to study the pore structural features of reservoirs. The pore in shales is varied, different pore types are not the same meaning with gas storage and flow capacity. In this paper, we use field emission scanning electron microscopy to take the secondary electron mode to scan shales from the Gufeng Formation in Jianshi, six categories and ten types of pores have been detected. This shows: the main pore types in argillaceous limestone are intercrystal pores within mineral grains, microfracture and brittle mineral are well developed; In siliceous mudstone, we find many pyrite microspherulites and floccules, organopores size from the nanoscale to several microns; A large number of honeycomb holes and microfracture are widely distributed in carbonaceous shale, the main pore types are organic matter nanopores. The nanoscale pore characteristics are quite different with traditional reservoir. In these pore types, the highly possible types to form gas migration pathways are: (1) porous floccules; (2) pores of organic matter; (3) microchannel and microfracture; (4) pores between organic matter and minerals.

KEYWORDS: Shale gas; Pore types; FESEM; Western Hubei; Gufeng Formation.

INTRODUCTION

Shale is generally defined as "fine grains of clastic sedimentary rocks", according to the definition, it refers not only to the simple shale, such as: calcareous shale, iron shale, siliceous shale, carbonaceous shale, black shale and oil shale, but also fine-grained siltstone and fine sandstone, siliceous mudstone, chert, siltmudstone, limestone and dolomite. Shales have a wide variety of mineral compositions, structures and constructions. Carbonaceous shales are made up by thin alternating layers of dark mudstone and light-colored siltstones; rocks are generally composed of clay minerals, quartz particles and organic silty. Recently, the scholars in petroleum geology have made a lot of progress on shale sedimentary processes and diagenesis, shale of micro- sedimentary structure, different lithofacies of coarse grained and fine grain of accumulation model, as well as sequence and parasequence level of the strata [1-6], an important progress in understanding the shale gas storage and flow capacity is embodied in the investigation of the micro and macro pores in the Barnett Shale [7-8]. Along with this study, it is recognized that the shale gas reservoir space is mainly pores and micro fracture [9-11]. Jarvie believes that the pores in the shale are mainly form of organic hydrocarbon generation, if the shale organic matter content is 7%, the volume fraction will be 14%; If 35% of these organic matters are transformed, the rock will increase 4.9% of the pore space [12-13]. Loucks think that this kind of "pores in organic matter" is a dominant pore type in shale, other types of pores are relatively scarce or lack [7]. Roger and Neal think that the pores in organic matter are not the only type found in Barnett shale or other shale. The micro-crack in shale is another major reservoir space, its length is in micron grade to the nanometer level [8]. In the process of hydrocarbon generation, with the increase of hydrocarbon, the internal pressure increases, when the breakthrough pressure is reached, a large number of micro-cracks will be formed, new storage space will be formed also. During diagenesis, mineral phase changes cause micro-cracks formation, many microcracks are formed in the process of tectonic activity as well. Chinese scholars also found that different types of cracks, crack sizes, pore types and pore sizes make different contributions to shale storage and capacity contribution, the role is different too. Pores are the storage space in shale gas reservoirs, that largely determines its storage, while the crack is the main channel of gas migration in shale gas reservoirs, which determines its capacity [11].

In view of the pore types of shale having important effects on reservoir type, gas generation and the characteristics of the gas gathering, domestic and foreign scholars have done some research on the pore structure. Loucks work on siliceous mudstones from the Mississippian Barnett Shale of the Fort Worth Basin, they used Ar-ion-beam milling and scanning electron microscopy to characterize Barnett pores from a number of cores and grouped them into two general categories on the basis of sizes: micropores (pores having diameters ≥0.75um) and nanopores (pores having diameters ＜0.75um). Nanopores are observed in three main modes of occurrence: pores in grains of organic matter as intraparticle pores, interparticle pores between organic matter and nano to micro intercrystalline pores in pyrite framboids [7]. Nie divided the fissure in shale samples of Lower Paleozoic in Sichuan Basin into five classes according to the developing scale, which is giant, large, medium, small and micro; the pore types are divided into organic matter pores, the kerogen network pores, mineral pores and pores between organic matter and various minerals [11]. Roger made a detailed description and classification of pores found in Barnett and Woodford shale, and divided them into the following pore types by their origins: (1) interparticle pores produced by flocculation, (2) organoporosity produced during burial and maturation, (3) fecal pellets, (4) intrapaticle grains from fossil fragments, (5) intrapaticle pores within mineral grains, (6) microchannels and microfractures [8].

This paper works on argillaceous limestone, siliceous mudstone, carbonaceous shale and so on from the Middle Permian Gufeng Formation in Jianshi, Western Hubei. We use high-resolution field emission scanning electron microscopy to study the pore types and characteristics in variety of black shale series, and achieve a certain understanding to be reported for criticism. It is noted that the pores including closed pores and open pores in this paper, namely the pores and the fissures.

REGIONAL GEOLOGY

The study area Luojiaba section belongs to Jianshi County, Hubei Province. It is located in the north of the mountainous southwestern Hubei, the environment refers to steep terrains, with an average 1,800-2000m high altitude. There is a large area of carbonate distribution, Karst landform develop widespread. The highway of Yichang to Enshi is passing through this region (Figure 1), the traffic is very convenient.

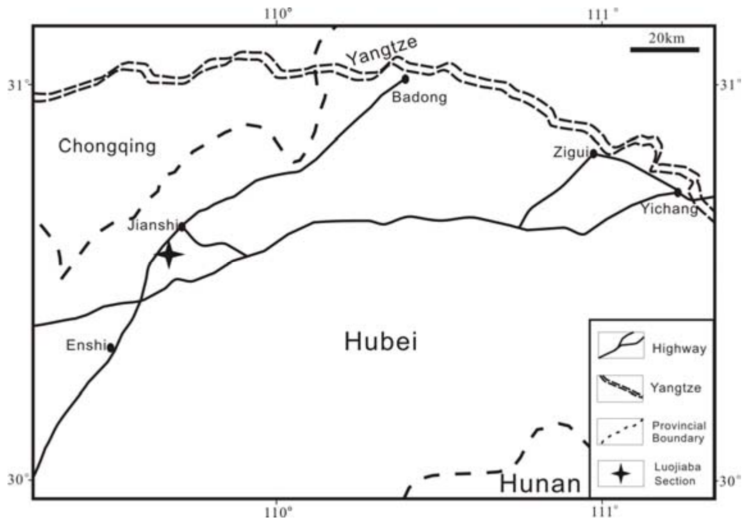

Figure (1). Map showing the Luojiaba section location.

The Luojiaba section is located in the hill on the left side of the road about 10km along the National Road 209 from Jianshi to Enshi, 300m away from the road. The section lies on a large artificial quarry, the outcrop is good and fresh, belonging to the Middle Permian Gufeng Formation. It is 16.35m thick from the bottom up, which can be divided into four lithologic series according to lithological characteristics: siliceous mudstone, argillaceous siliceous rock, argillaceous limestone and carbonaceous shale (Figure 2).

Siliceous mudstone series is mainly made up by black siliceous mudstone, carbonaceous shale and mudstone, which the thickness is 3.63m; Argillaceous siliceous rock series is made up by the black muddy cherts, carbonaceous shale and mudstone, which the thickness is 5.82m; Argillaceous limestone series is mainly composed of black argillaceous limestone, lenticular limestone, carbonaceous shale and thin-bedded limestone, which the thickness is 5.3m; Carbonaceous shale series is mainly composed of black carbonaceous shale and clay shale, which the thickness is 1.6m. We collect a total of 34 samples, after processing, select 10 representative lithology to do research on pore characteristics.

TYPES AND CHARACTERISTICS OF PORES

In this study, we use field emission scanning electron microscope taking the secondary electron imaging mode to observe and scan shales in the Key Laboratory of Geological Processes and Mineral Resources, discover that a variety of pore types are existed in the black shales of Luojiaba section. On the basis of pore types division made by former scholars, we divide pores in this section into six categories and ten types according to shale components(clay, organic matter, minerals, fossils, detritus).

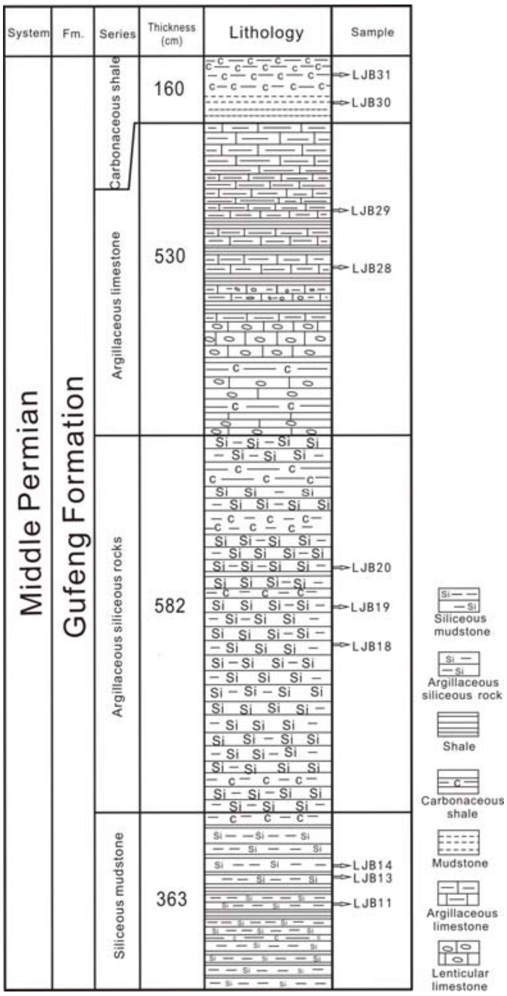

Figure (2). Stratigraphic sequence, rock characteristics and sampling location of the Gufeng Formation in Luojiaba section, Jianshi, Western Hubei.

Intergranular Pores Produced by Flocculation

The floc is massive electrostatic charge clay fragments of ion-rich sinking into the sea, it is a typical representative of the pores of clay. O'Brien and Slatt shows an example of flocculent micro-bedding in several ancient shale, but cannot explain how this fabric openwork was preserved after burial and diagenesis for billions of years[14]. Although this situation is relatively difficult to understand, but the fact that flocs can be preserved is common in the internal structure of many buried ancient shale, this pores were found in Barnett and Woodford shale also.

The results of the observation have its significance, this open network or paper-room-like structure may provide pores larger than the methane molecular diameter (0.38nm) between floc. We find that the floc are most irregular flakes distribution under the microscope (Figure 3a), parallel to the bedding plane, pore size change from the nanoscale to several micrometers. These pores can connect together to form a porous channel (Figure 3b). Therefore, the open or partly collapsed floc structure preserved in the shale may be the location of intergranular pores between the clay fragments.

In addition, flocculation may contribute to long-range carrying a lot of mud in the marine environment. Barnett Shale, including the ancient mudstone and shale, numerous micro-pore structure is clearly support the mechanism that the coastal sediments are carried by ocean currents. Schieber compared modern mud ripple marks and the scour surface with the analogues in stratum, he conclude that: flocculation may make the sediment particles thicker in a hydraulic manner, as high-density flow or turbidite component to move coastally [3]. This effect may be linked to the trapping capacity by internal silt particles in floc [15]. A modern high-density flow has been demonstrated to travel along the Sea of Japan 700 km from the river source [16]. Soyinka and Slatt demonstrated the muddy high-density flow in the Cretaceous continental slope and bottom stratum [17].

Pores of Organic Matter

Pores of organic matter including pores of organic fossils and pores of asphalt/kerogen network.

Pores of organic fossils

The pores of organic fossils are organic fossil material (such as algae floc) preserved in the rock. Due to the influence of underground temperature and the increased pressure by burial diagenesis, organic matter gradually become porous during the cracking hydrocarbon conversion process, these pores become the preservative place where can generate gas [7,18,19]. It is common to see siliceous sponge spicules in shales, they have central cavity initially, which may contain decomposed organic matter during burial, or the central cavity may be partially or completely filled by secondary quartz, clay or organic particles. There are similar silicified spicules, sometimes with open chamber in Cowley Formation, Kansas, Mississippi [20]. In the Barnett Shale, the spicules are usually concentrated in a thin strip the same way with feces. The generation of such pores may due to bioturbation of the sediments, generation of fecal pellets and porosity of organic skeleton or shell [21].

There are rich fossils in shales from the Middle Permian Gufeng Formation in Jianshi, Western Hubei. Some of these biological fossils have porosity, microns and sub-micron pores have also found in compressed algae residues, methane bacteria and sporangium. Pores of organic fossils seen in this paper are mainly fossil spores and the sponge-like organic matter floc (Figure 3c, d), their shape are bubble-like irregular oval and round, pore size are between 1-6μm. It is common in the siliceous mudstone.

Pores of asphalt/kerogen

Such pores of organic matter are formed during burial and mature period, including asphalt pores and kerogen pores.The aperture is mainly nanoscale, which can be reservoir space for adsorbed natural gas. Asphalt residue in black shales belonging to this kind of pores, adsorbed or even dissolved gas occurs in the asphalt. Organic matter in the source rock is not scattered, which mostly distribute along the micro-bedding plane. Micro-bedding plane have relatively good permeability , the relative enrichment of the organic matter can make it good lipophilicity, if kerogen was connected in gas generation stage, it is easy to connect lipophilic network with each other that free of capillary resistance [22]. According to pore size, pores can be divided into micropores (diameter less than 2nm), mesopores (diameter 2-50nm) and macropores (diameter typically more than 50nm); with the increasing of porosity, pore structure will change (micropore turn into mesopore, even macropore), surface area in pores also increased [23].

The asphalt pores are more flocculent round shape and massive texture(Figure 3 e, f), the kerogen pores appear network structure (Figure 3 g), different size and shape, dispersed distribution in the matrix, in which the number of nanopores is rich, it may contain hundreds to thousands of nanopores within a piece of organic matter. In addition, due to the strong activity of the adsorbent on the surface of dispersed organic matter, it can greatly improve the adsorption capacity of shale, along with the increase of the maturity, the thermal evolution of hydrocarbon generation will produce some micropores.

Pores between the Mineral

It is mainly including intergranular (interparticle) pores, transgranular (intragranular) pores, pores of dissolution and miscellaneous pores. In shales from Gufeng Formation in Jianshi, we found a great quantity of intragranular pores in pyrite and intergranular pores in mineral grains.

Intragranular pores in pyrite

Pyrites in shales from the Middle Permian Gufeng Formation in Jianshi are well developed. The pyrite framboids are found commonly, which are composed by many small strawberry-shaped pyrite crystals (Figure 3 h), during which there are micropores. The pyrite grains within the pyrite framboids in argillaceous limestone are cube, the edge length is about 100-400nm. There are certain controversies about the significance of shalegas on this particular type of intragranular pores. The majority of pyrite grains in siliceous shale are separate distribution, the intergranular pores produced by shedding of pyrite are common, the remaining organic residues (mucus) usually exist around the pyrite framboids (Figure 3 i). Loucks think that these pores in the shale matrix are usually exist in isolation, the connectivity between the pores is poor, they are not conducive to the shale gas stored [7]. While some scholars have found the

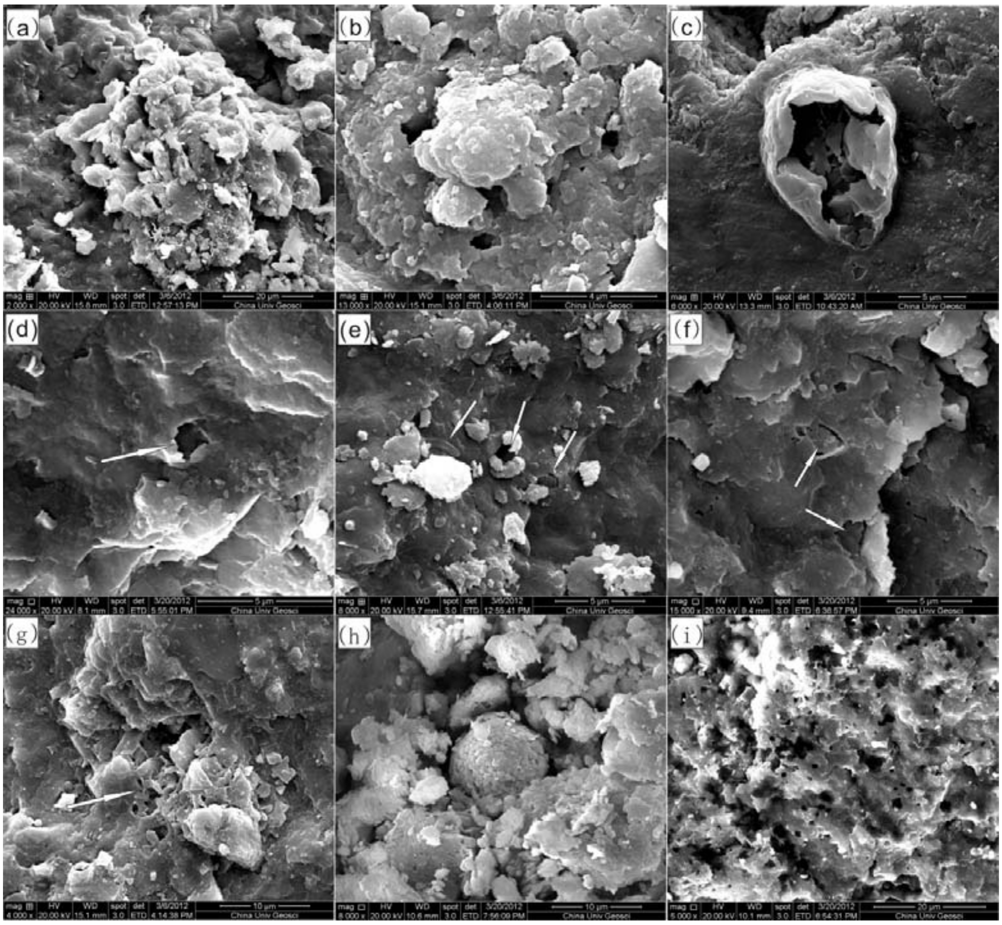

Figure (3). Pore types from the Middle Permian Gufeng Formation in Jianshi (1).

a. sheet structure of the floccule; b. permeability channel by flocculation; c. sporangium in argillaceous limestone; d. pore from broken spores; e, f. pores from residual organic matter debris; g. honeycomb pores from kerogen; h. pyrite framboids crystals; i. residual pores from pyrite framboids

remnants of biological organic matter in the vicinity of the pyrite framboids, with the strong ability of adsorbing natural gas [24]. They proved that these pores have some relationship with source of organic matter, and affirmed the shale gas significance of the intragranular pores in pyrite to some extent.

Intergranular pores in mineral grains

The micropores between mineral grains include pores between minerals and pores between the mineral and other particles. Generally, the aperture is a few microns, the individual may be up to a dozen microns, even millimeters, mainly developed in the mineral assemblages which have fine and thick crystals(Figure 4 a). The morphological manifestations are particles surrounding by cyclic annular, clear boundary, pore size ranging from 1-20um, the specific shape and size depending on the surrounded particles. The annular pores around the pyrite framboids mentioned above should also be part of the intergranular pores. Other minerals in which may developed intergranular pores are illite, kaolinite, calcite, quartz, montmorillonite and so on. The size, shape and amount of pores depends on the mineral formation time, also depending on the mineral grains are protogenetic or secondary. For example, illite showed curved lamellar, irregular lath-shaped, honeycomb shape and silk shape in the scanning electron microscope (Figure 4 b), it is a product under the conditions of the high thermal evolution, with relatively high concentration. Therefore, it is available to judge the maturity of marine shales in Early Paleozoic according to illite crystallinity. These pores are mainly contributed to gas storage, since there are many clay minerals on the surface, which have a strong adsorption capacity to natural gas.

Pores between Organic Matter and Minerals

Mainly refers to various pores between organic matter and minerals(Figure 4 c). Such pores have connected organic matter (asphalt) and minerals, join the two types of pores together, making the gas generated by organic matter can migrate into the mineral pores, it is very important for shale gas accumulation and output. This type of pores is only a small part of the pores in shale, but significant in that they can play the role of microfissure to some extent.

Microchannels and Microfractures

Microchannel

Various sizes and shapes of microchannels are existed in the shale matrix (Figure 4 d). They can provide favorable permeability channel and micro-pores, they are usually wavy, discontinuous, and nearly parallel to the bedding plane. Under SEM, they are usually not extends to the outside viewing area of the shale samples. These cracks show that the microchannels are not generated during the sample handling and sample preparing period, but it represents the opening of the original micro-channel stored in the undisturbed shale matrix. Microchannel is usually a width of less than 0.3mm, but enough for gas molecules to penetrate the channel. The causes of these micro-channels are difficult to determine, which may be the bioturbation between texture, residues of micro-erosion surface, or the top surface of micro wavemark.

Microfracture

The fracture size in shales from the Middle Permian Gufeng Formation is different. The microfractures are generally larger than the micropores, and the sinuosity is small, relatively flat. Small fractures in shales may fill with asphalt or partially open (Figure 4 e).

Such microfractures are the main channel in which the adsorbed natural gas in shale gas reservoir resolve to the free state (Figure 4 f), and it's important in the study of the shale fabrics.

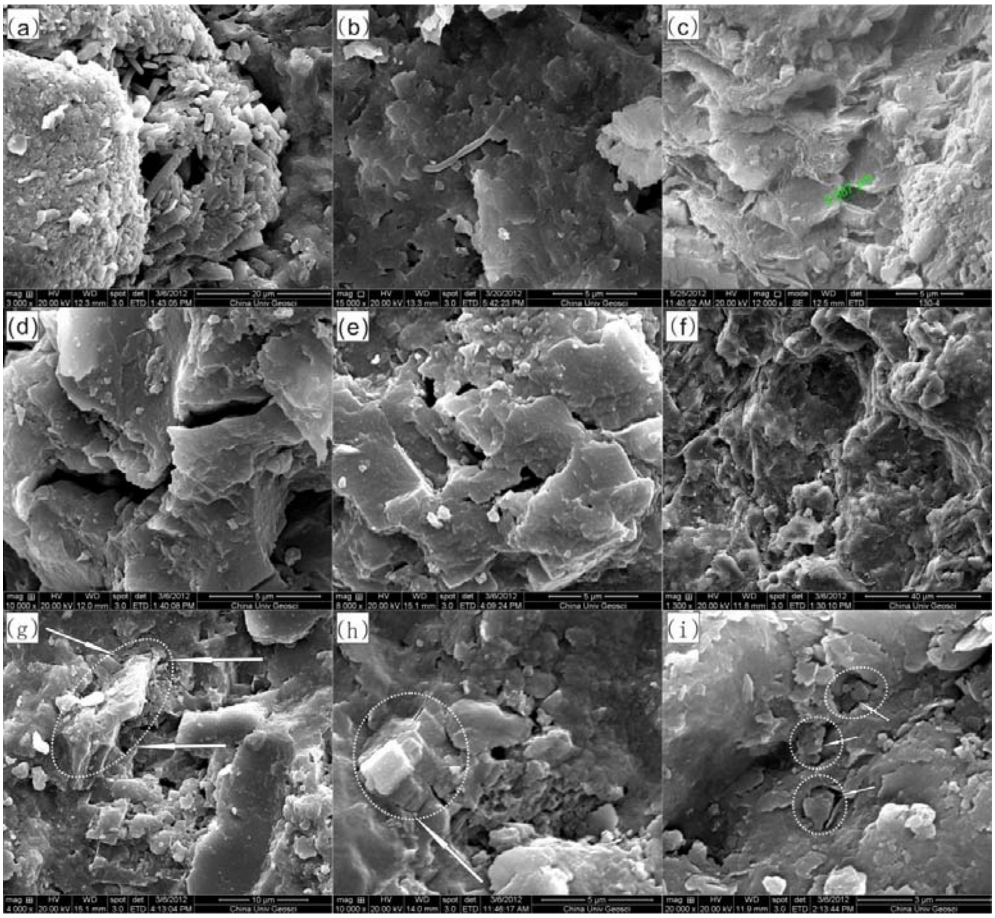

Figure (4). Pore types from the Middle Permian Gufeng Formation in Jianshi (2).

a. pores between organic matter and minerals; b. micropores between the mineral grains; c. lamellar mineral Intercrystal pores; d. microchannel in the shale matrix; e. natural microfissures; f. microfissures in carbonaceous shale; g, h. pores between calcareous fossil fragments and surrounding rock; i. pores between detrital particles and surrounding rock

Pores between the Detrital Particles

Under the field emission scanning electron microscope, we have found some pores formed by the detrital particles in shales from the Middle Permian Gufeng Formation in Jianshi. These pores including pores between calcareous fossil fragments and surrounding rock, pores between detrital particles and surrounding rock.

Pores between calcareous fossil fragments and surrounding rock

The majority of the invertebrate fossils are composed of the calcium carbonate, pores may form between these fossil fragments and surrounding rock. There are many microfossils in shales from Gufeng Formation, and thus this type of pores is numerous (Figure 4 g, h), they can provide better conditions for the accumulation and preservation of the natural gas.

Pores between detrital particles and surrounding rock

In addition to the calcareous fossil fragments, shale also contains a certain amount of detrital material, these detritus have different shapes and sizes, pores may form within the detrital particles or between the surrounding rock(Figure 4 i), such as the tiny pores with different origin in quartz and feldspar. There are pores of irregular polygons, ranging in size (generally less than 1um) in quartz, feldspar and other detrital mineral grains, part of the pores represent the characteristics of mineral particle shedding and dissolution traces, indicating the different origins of pores[5,7]. To a certain extent, the content of quartz, feldspar and other brittle mineral in shales can determine the number of detrital particle pores, and determine the shale gas storage subsequently. This reflects the influence of rock composition on the pore development.

SUMMARY AND DISCUSSION

Table 1 summarized and compared the types and characteristics of the pores in shales from Gufeng Formation, in these pore types, the types may most contribute to permeability are: (1) porous floccules; (2) pores of organic matter; (3) microchannel and microfracture; (4) pores between organic matter and minerals. If fossil residues, such as sponge spicules and spores, are connected together and arranged in very thick shale formations, they may also provide some permeability. Particles and crystals dispersed in the shale matrix, such as pyrite, of which the intragranular pores may not contribute to the permeability largely. In addition to microfractures and the organic pores, pores produced by flocculation of clays may have the greatest potential to provide storage space and penetration channel for migration of hydrocarbon molecules. Sample observation and description, SEM and FESEM, EDX, back scattering imaging and electronic microprobe spectroscopy provide identification and verification to the existence of these pore types. Although the discussion of this paper is mainly restricted to the shales from the Middle Permian Gufeng Formation in Jianshi, similar pore types also exist in shales of other unconventional oil and gas.

Table 1. Comparison of pore types and characteristics in shales from Gufeng Formation in Jianshi.

Characteristic / Type	Morphology	Pore Size	Connectivity
Porous floccules	Distribution of network or paper-shaped structure	Nanoscale intergranular pores, pore size is about 0.1-5μm, the particles are nearly parallel arrangement	Pores may be connected
Pores of organic fossils	Oval, broken spores particles	Aperture is about 1-6 μm	Pores are usually isolated, the internal chamber is empty, or it may be filled by detritus or authigenic mineral
Pores of asphalt/kerogen	Asphalt pores are honeycomb, floc or massive； kerogen pores are network structure	The pore diameters are from nanoscale to 10μm	The distribution of asphalt pores in the matrix is more dispersed, the connectivity of kerogen pores is better
Intragranular pores in	Framboids aggregate, tightly	Diameters are about 200-400nm, the framboids	Particles usually scattered in

pyrite	packed	are about 8-10μm	the shale matrix
Intergranular pores in mineral grains	Thin sheet or irregular lath structure; aggregates are honeycomb or silk thread shape	Carbonate minerals diameter <10μm, in clay minerals, intergranular pore diameter is generally a few microns.	General connectivity
Pores between organic matter and minerals	Less distribution, irregular polygon	Aperture size is about 1-3 μm	With good connectivity
microchannel	Usually wavy, discontinuous, nearly parallel to the bedding plane	Slit width from nanometer to micron level, generally less than 0.3mm	Distribution is typically concentrated but commonly not connected
microfracture	Mostly flat seam and inclined seam, obvious dissolution and expand effect along the fissures, and then form the dissolved pores	Natural fractures, some may be asphalt filling, the width is 0.5-2μm, the length is less than 2mm	With a high degree of cluster distribution characteristics, and easy to connect
Pores produced by calcareous fossil fragments	Irregular shape, oval, triangle and long strip	Length from 2 μm-5 μm	The connectivity is pretty average
Pores between detrital particles and surrounding rock	Widely distributed, nearly circular micropores	Aperture between 3-6 μm	Medium connectivity

The study of pores is the core of shale reservoir researching. The pore is the main aspect to evaluate the hydrocarbon generation capacity, gas storage capacity and exploitation value of shales. Pores are a crucial influencial factor to shale reservoir types, characteristics of the gas gathering and gas exploitation. Shales and pores in shale are varied. Different rock types have different pore types, size, abundance, morphology and connectivity, thus the effects to natural gas storage and flow capacity of shales are not the same. We discovered many nanopores and micropores under FESEM in black shales from the Middle Permian Gufeng Formation in Jianshi, western Hubei. They are small, widely distributed, high abundance, very different from traditional reservoir pores, and of great significance to shale gas storage.

ACKNOWLEDGEMENTS

This work was financially supported by National Natural Science Foundation of China (40839903), China Geological Survey Bureau (1212011220796), State Key Laboratory of Geological Processes and Mineral Resources project jointly funded.

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