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# Authigenic minerals related to wettability and their impacts on oil accumulation in tight sandstone reservoirs: An example from the Lower Cretaceous Quantou Formation in the southern Songliao Basin, China



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#### ABSTRACT

Oil accumulation, being difficult and complicated, is an important issue in petroleum exploration researches. Authigenic minerals, such as carbonate cements and chlorite, can in certain reservoirs alter the wettability of some pore spaces from water-wet to oil-wet. Generally, these oil-wet pore spaces are favorable for oil accumulation. The alteration of reservoir wettability induced by authigenic minerals and the impacts on oil accumulation in tight sandstone reservoirs were investigated using a suite of mineralogical and geochemical characterization techniques, including thin section observation, SEM-EDS, XRD, QEMSEM, CL, quantitative grain fluorescence (QGF), fluorescence spectral analysis, contact angle measurement and sealed coring oil saturation testing on the fourth member of the Lower Cretaceous Quantou Formation  $(K_1q_4)$  in the southern Songliao Basin, China. The study shows that the tight sandstone reservoirs are compositionally immature with detrital grains distributed homogeneously. Quartz, carbonates and clay minerals that show heterogeneous distribution characteristics are the major authigenic minerals in some parts of the intergranular pores. The detrital mineral assemblage suggests that the reservoir rocks at deposition and before diagenesis had characterized by strong water-wet properties. With the development of authigenic minerals, carbonate cements and authigenic chlorite tend to alter the wettability of some parts of the existing pore spaces from water-wet to oil-wet. In the  $K_1q_4$ sandstone reservoirs, oil prefers to accumulate in the cemented residual pore spaces around carbonate cements and chlorite. Reservoirs containing about 4-5% carbonate cements are suggested to be more preferable to oil accumulation. These reservoirs are mainly located between sandstone-mudstone interfaces and central parts of the sand bodies. Chlorites have mainly two effects: on one hand, chlorite alters the wettability of existing pore spaces and provides preferential accumulation sites for oil in the tight sandstone reservoirs, on the other hand it can reduce the adhesion of oil through forming "clay-oil flocs" and promote further migration. Consequently, the reservoirs with moderate amount of carbonate cements or chlorite always show relative high oil saturation in the K<sub>1</sub>q<sub>4</sub> tight sandstones.

#### 1. Introduction

Authigenic minerals, such as quartz, carbonates, albite and clay minerals, are common in clastic reservoirs undergo diagenesis (Bjørlykke, 1998; Hillier, 2003; Omer and Friis, 2014; Yuan et al., 2015a; Mu et al., 2016). Authigenic minerals may influence on reservoir physical and chemical properties (Bjørlykke, 1998; Lai et al., 2016; Kassab et al., 2017). They can also provide valuable records of water-rock interaction during fluid movements in clastic reservoirs (Uysal et al., 2000; Hyodo et al., 2014; Wang et al., 2015; Ma et al., 2015). Authigenic minerals have been studied extensively, and their origins, formation mechanisms, distributions, controls on porosity and permeability have been well documented (Molenaar et al., 2007; Dutton, 2008; Morad et al., 2010, 2012; Lehmann et al., 2011; Yuan et al., 2015a,b; Haile et al., 2015). For example, most of the researches regarded silica cement as an internal authigenic mineral, mainly occurring as quartz overgrowth (Bjørlykke and Egeberg, 1993; Kim and Lee, 2004; Peltonen et al., 2009; Marcussen et al., 2010; Metwally and

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**Fig. 1.** (A) Location map of the study area and sub-tectonic units of the Songliao Basin (From Xi et al., 2015a). (I) Western Slope Zone, (II) Northern Pitching Zone, (III) Central Depression Zone, (IV) Northeastern Uplift Zone, (V) Southeastern Uplift Zone, (VI) Southwestern Uplift Zone, (VI) Kailu Depression Zone; (B) The sub-tectonic units of the study area and well locations.

Chesnokov, 2012). On the contrary, carbonate cements are commonly treated as external authigenic minerals that fill the intergranular pores (Gier et al., 2008; Chen et al., 2009; Yuan et al., 2015b; Wang et al., 2015). Both the silica and carbonate cements could reduce the porosity and permeability (Williams et al., 1997; Dutton and Loucks, 2010; Zhang et al., 2015), but increase rock strength and brittleness at the same time (Zhang et al., 2012; Storvoll et al., 2005; Xi et al, 2015a). Kaolinite as clay minerals can generate from feldspar dissolution and precipitate in the primary pores (Giles and De Boer, 1990; Chuhan et al., 2001; Taylor et al., 2010; Bjørlykke and Jahren, 2012). Smectite that is not stable in sandstone reservoirs can transform into illite as well as chlorite with increasing of burial and thermal evolution (Hower et al., 1976; Lynch et al., 1997; Thyberg et al., 2010; Bjørlykke, 2011; Stroker et al., 2013). Among them, kaolinite and illite mainly occur as pore-filling and pore-lining minerals, strongly reducing reservoir quality (Lander and Bonnell, 2010; Yuan et al., 2015b; Kassab et al., 2017). However, chlorite commonly occurs as grain coating, reducing porosity losses through inhibiting quartz overgrowth (Ehrenberg, 1993; Bloch et al., 2002; Ajdukiewicz and Larese, 2012).

As one of the most important unconventional hydrocarbon resources, tight sandstones encounter complicated and strong diagenetic alterations, forming a variety of authigenic minerals (Karim et al., 2010; Yang et al., 2012; Zhou et al., 2016). Firstly, pore-filling authigenic minerals together with mechanical compaction are the main reason for the formation of the tight reservoirs (Zhang et al., 2009a,b; Fic and Pedersen, 2013). Compared to conventional oil or gas reservoirs, tight sandstone reservoirs are characterized by extremely low porosity and permeability, small pore-throat size and a non-buoyancy dominated charge and accumulation process (Zou et al., 2012, 2013; Jia et al., 2012; Pang et al., 2014). They are always characterized by strong heterogeneous oil even for reservoirs with similar grain fraction. porosity and permeability (Li et al., 2013; Xi et al, 2015b). Hence, except for porosity and permeability, some other controlling factors, such as reservoir wettability, probably play significant roles on oil accumulation in tight sandstones as well (Qi et al., 2015). It is difficult for oil charge into water-wet tight sandstone reservoirs due to small pore spaces and strong capillary resistance, whereas it will become much easier if some pore spaces become oil-wet (Buckley, 2001; Oi et al., 2015). This is because the capillary resistance can turn into the "driving force" through spontaneous imbibition of oil under oil-wet conditions (Qi et al., 2015). Some oil-wet authigenic minerals can facilitate transforming from water-wet to oil-wet in certain reservoirs (Barclay and Worden, 2000; Kumar et al., 2005; Morad et al., 2010; Karimi et al., 2015). Once, moderate oil-wet authigenic minerals precipitate in the pore spaces, the oil will be prone to migrate towards these directions and forming potential oil accumulations (Buckley, 2001; Qi et al., 2015). Although there are many researches focusing on wettability alterations induced by authigenic minerals during the petroleum development phase for enhanced oil recovery (Meybodi et al., 2011; Hadia et al., 2013; Khajepour et al., 2014; Liu et al., 2015), little attention has been paid to wettability alteration by authigenic minerals and their impacts on oil migration and accumulation during the petroleum exploration stage. The tight sandstone reservoir of the Lower Cretaceous Quantou Formation is a prolific oil-producing unit in the southern Songliao Basin. However, oil distribution is very heterogeneous even at small scales (Li et al., 2013; Xi et al., 2015b). In order to predict the "sweet spots" of these tight sandstones accurately and efficiently, more attention should be given to oil accumulation influenced by diagenetic

wettability alterations, especially in reservoirs with similar source rocks, structure locations and migration systems.

The objectives of this paper are primarily focusing on the different aspects of authigenic minerals compared to the previous studies, working only on their genetic mechanisms and controls on reservoir properties: (1) investigate the main types of authigenic minerals and their characteristics; (2) analyze wettability alteration induced by the main types of authigenic minerals; and (3) evaluate the impacts of oilwet authigenic minerals on the oil accumulation process.

## 2. Geological background

The Songliao Basin is a Jurassic - Neogene lacustrine basin in the north-eastern China (Fig. 1), which is located between  $42^{\circ}25'$  to  $49^{\circ}23'$  N and  $119^{\circ}40'$  to  $128^{\circ}24'$  E with an area of about  $26 \times 10^4$  km<sup>2</sup> (Zhang and Zhang, 2013). It can be subdivided into seven first class tectonic zones (Zhou et al., 2012), namely the Western Slope Zone, Northern Pitching Zone, Central Depression Zone, Northeastern Uplift Zone, Southeastern Uplift Zone, Southeastern Uplift Zone, Southeastern Uplift Zone and the Kailu Depression Zone (Fig. 1). The study area, as one of the most oil rich areas, belongs to the Central Depression Zone and consists of three secondary class tectonic units (Li et al., 2013), namely the Changling Sag, Huazijing Terrace and Fuxin Uplift (Fig. 1).

Based on the sediment infilling sequences and structures, the basin evolution can be divided into four phases: (1) a pre-rift phase; (2) a synrift phase; (3) a post-rift phase; and finally (4) a compression phase (Zhang et al., 2009a,b). Sediments that has filled the basin comprise the Lower Cretaceous Huoshiling (K1h), Shahezi (K1sh), Yingcheng (K1yc), Doulouku (K1d) and Quantou (K1q) formations, the Upper Cretaceous Qingshankou (K2qn), Yaojia (K2y), Nenjiang (K2n), Sifangtai (K2s) and Mingshui (K<sub>2</sub>m) formations, the Palaeogene Yian (Ny) formation, the Neogene Daan (Nd) and Kangtai (Nt) formations, and the Quaternary Pingvuan (O) formation. Each formation can be further subdivided into different members (Fig. 2). Previous studies indicated that the Songliao Basin has had a high geothermal gradient throughout most of its history, which makes the sandstone diagenesis more complicated. From about 90.7 Ma to 65 Ma, the geothermal gradient has been estimated at 4.5-5.5 °C/100 m, decreasing to about 4.0 °C/100 m after 65 Ma (Liu, 2004). Presently, the sedimentary sequence is not at its maximum depth and temperature, and the temperature of Quantou Formation has never been more than about 130 °C (Xi et al., 2015a).

The studied section (Fig. 2), the fourth member of the Quantou Formation ( $K_1q_4$ ), was deposited during the tectonic depression period, and consists mainly of deltaic sandstones and some interbedded mudstones (Xi et al., 2015b). According to oil analysis and source rock correlation, the first member of the Qingshankou Formation ( $K_2qn_1$ ), which just overlies  $K_1q_4$ , is the main source rock for the  $K_1q_4$  reservoirs (Zou et al., 2005; Li et al., 2013). Oil generation (Ro = 0.5%) in the K<sub>2</sub>qn<sub>1</sub> began at a depth of about 1350–1450 m and at a temperature of 80–85 °C (Dong et al., 2014). The K<sub>2</sub>qn<sub>1</sub> interval commonly developed over pressures, whereas the K<sub>1</sub>q<sub>4</sub> sandstone reservoir showed normal pressure over most of the studied intervals. This differential pressure may have been a driving force for the oil migration between these formations (Xi et al., 2015b). The K<sub>1</sub>q<sub>4</sub> sandstone reservoir is quite tight and strongly heterogeneous. All wells yield oil, but with low production rates (Li et al., 2013).

## 3. Databases and methods

The data used in this paper was derived from more than 30 wells located in the southern Songliao Basin where most of the producing oil fields are located (Fig. 1). Rock composition data of 743 thin section samples, reservoir porosity and permeability data of 8529 core plug samples, physical properties of 31 crude oil samples were obtained from the Research Institute of Petroleum Exploration & Development of the Jilin Oilfield Company, PetroChina.

Over 300 polished thin sections and about 210 blue or red epoxy resin-impregnated thin sections were prepared for the analysis of rock mineralogy, diagenesis, visual pore and oil micro-occurrence characteristics. Thin sections were partly stained with Alizarin Red S and Kferricyanide for carbonate mineral identification. A total of 37 core samples from 12 wells were prepared as polished thin sections without cover sheet for fluorescent color observation. For the content of authigenic minerals (quartz and carbonate cements), 20 photomicrographs each of 76 thin sections were taken using a Zeiss Axioscope A1 APOL digital transmission microscope. Authigenic minerals in each photomicrograph were then identified under the microscope, and the total area of authigenic minerals in every photomicrograph was obtained using the Image-Pro Plus software. Finally, the percentages of the authigenic minerals were calculated by taking the average of all values in the 20 photomicrographs from each thin section. Quantitative evaluations of minerals by scanning electron microscope (QEMSEM) were done for two representative samples using FEI Quanta 450 combined with the QEMSEM software. Each sample scanned for 36 field of view (FOV) in about six hours, and then all FOVs were stitched together to provide a mineralogical composition map of the scanned area. A total of 105 reservoir sandstone samples were analyzed for whole-rock (bulk) and clay fraction ( $< 2\mu m$ ) mineralogy using XRD. Preparation, analysis and interpretation procedures are modified from Moore and Reynolds (1997) and Hillier (2003). A total of 31 representative samples were viewed under a Quanta FEG 450 and SU 5000 scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDS). Porosity, permeability and oil saturation for 108 sealed coring samples from Well Rang 59 were analyzed. A sub-set of 29 samples were analyzed using the quantitative grain fluorescence (QGF; Liu and Eadington, 2005) method by Petro-China Research Institute of Petroleum Exploration & Development to evaluate the relative oil saturation of the of reservoirs. In addition, contact angles between the oil and rock surface were measured for 36 oil cleaned samples selected from XRD analysis using the pendant drop method (Teklu et al., 2015).

Thin section identification and fluorescence observation were done in the Basin Analyses and Reservoir Geology Key Laboratory of China University of Petroleum. XRD, SEM, QGF and QEMSEM analyses were performed in the Key Laboratory of Oil and Gas Reservoirs of PetroChina and University of Oslo. Contact angle were measured in the Huabei Oil field Company of PetroChina.

#### 4. Results

## 4.1. Reservoir lithologies and its initial wettability

#### 4.1.1. Reservoir lithologies

Most of the  $K_1q_4$  sandstones are fine to medium-grained, moderately well to well sorted with sub-angular to sub-rounded detrital grains, which mainly consists of quartz, feldspar and rock fragments. Point counting data show that the relative content of the quartz varies between 32.1% and 62.4% with an average of 42.86%, feldspar ranges from 10.3% to 42.8% with an average of 26%, and rock fragment ranges from 12.4% to 47.6% with an average of 31.14%. The dominated rock fragments are volcanic rock fragments varying from 10% to 47% with an average of 28.32%. The tight sandstone reservoirs have an average framework composition of  $Q_{43}F_{26}L_{31}$  and are compositionally immature (Fig. 3).

QEMSEM photomicrographs can provide mineralogical compositions of the sandstone and their distribution features visually. The results of two typical QEMSEM analyses show that the detrital grains, i.e. quartz, albite and K-feldspar, are nearly homogeneously distributed in the tight sandstone reservoirs (Fig. 4). Authigenic minerals, i.e. calcite, illite and chlorite, however, display heterogeneous distribution characteristics. Generally, authigenic minerals only occur in some parts of the intergranular pores (Fig. 4).



Fig. 2. Generalized Mesozoic-Quaternary stratigraphy of the Songliao Basin, showing major petroleum system elements (from Xi et al., 2015a).

## 4.1.2. Reservoir initial wettability

The contact angle between oil drops and rock surfaces is the major parameter to evaluate the wetting behavior (Barclay and Worden, 2000). Among the detrital grains, quartz and feldspars are commonly water-wet minerals in the studied reservoirs with pH of formation water ranging from about 6–8 (Barclay and Worden, 2000). In the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs, contact angles between oil drop and rock surface ranges from 22.83° to 38.32° with an average of 27.97° (Fig. 5). The relatively low contact angle values suggest that the reservoirs are characterized by strong water-wet properties (Suicmez et al., 2008). Obviously, the distribution of the contact angle can be divided into two subgroups (Fig. 5), even though they have similar detrital grain composition. The first one ranges from about  $22^{\circ}$  to  $28^{\circ}$ , and the second one varies between  $30^{\circ}$  and  $40^{\circ}$  (Fig. 5). This probably shows that although the sandstone reservoirs are still water-wet as a whole, parts of the pore spaces have already became oil-wet, which led the contact angles to increase in these reservoirs.



Fig. 3. Rock composition classification of sandstone in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs.



Well Rang 24, 1846. 09m

Well Rang 24, 1874.02m

Fig. 4. QEMSEM photomicrographs of the typical tight sandstone samples.











Fig. 7. Sealed coring oil saturation distribution of tight sandstone reservoirs in Well Rang59.

## 4.2. Reservoir properties and oil saturation

#### 4.2.1. Reservoir properties

Under laboratory pressure conditions, porosity of core samples ranges from 1.7% to 20% (mainly 2% to 14%) with an average of 8.54% (Fig. 6A). Permeability ranges from 0.01 mD to 44.5 mD (mainly less than 1 mD) with an average of 0.493 mD (Fig. 6B). According to the pressure-controlled mercury injection, the core samples were characterized by high displacement pressure and small pore-throat size. Specifically, most of the displacement pressures were larger than 1.0 MPa (Fig. 6C), and the average pore-throat radii were mainly less than 1.0  $\mu$ m with most frequent ones are populated between 0.1  $\mu$ m and 0.35  $\mu$ m (Fig. 6D). As a whole, the reservoir properties are quite poor, especially the permeability was extremely low, showing the typical features of tight sandstones.



Fig. 8. Correlational relationships between porosity, permeability and oil saturation in the  $K_1q_4$  tight sandstones.



Fig. 9. Photomicrographs of thin section showing characteristics of quartz overgrowth and carbonate cements: (A) Quartz overgrowth; (B) Quartz overgrowth and calcite; (C and D) Ferrocalcite; (E) Ankerite; (F) Calcite zoned and engulfed by ankerite. Qa-quartz overgrowth; Ca-calcite; Fer-ferrocalcite; Ank-ankerire.

## 4.2.2. Oil saturation characteristics

Oil saturation under sealed coring conditions can more accurately reveal the reservoir oiliness. Take the Well Rang 59 as an example, the oil saturation of sealed coring ranges from 5.8% to 48.2% with an average of 29.05% (Fig. 7). Compared to the conventional sandstone reservoirs, these oil saturations were relatively low. Even in small scales with similar sandstones, however, the oil saturations were quite heterogeneous. All these probably imply that oil accumulation in this type

of reservoirs is rather complicated and quite difficult to predict.

From correlational relationships, it is evident that oil saturation in the  $K_1q_4$  tight sandstone reservoirs was not mainly controlled by porosity and permeability (Fig. 8). For example, between the depth of 2112 m and 2115 m, 2117 m and 2120 m, the oil saturation showed relatively high values, whereas the porosity and permeability were relative low (Fig. 8). On the contrary, between the depth of 2105 m and 2107 m, the relative high porosity and permeability was corresponding



Fig. 10. Relationships between the distance to the interface of sandstone-mudstone and the content of quartz overgrowth (A) as well as the content of carbonate cements (B).



**Fig. 11.** Photomicrographs of SEM showing characteristics of clay minerals in the  $K_1q_4$  tight sandstone reservoirs: (A) Kaolinite and mixed-layer illite/smectite; (B) Mixed-layer illite/smectite; (C) Illite; (D and E) Plate-like chlorite; (F) EDS of plate-like chlorite. K-kaolinite; I/S-mixed-layer illite/smectite; I-illite; Ch-chlorite.



Fig. 12. Relationships between the content of chlorite and the content of volcanic rock fragments (A) as well as the content of carbonate cements (B).

## Table 1

Composition,	salinity	and pH	of some	typical	formation	water	samples	in the	$K_1q_4$	tight	sandstone reservoir	s.
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Well	Depth, m	Ion concentrat	Total salinity, mg/l	pH					
		$K^+ + Na^+$	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl-	SO4 <sup>2-</sup>	$HCO_3^-$		
Cha10	2281.1	2724.1	60.1	23.1	3014.3	516.3	1684.8	8022.7	7
Cha19	2368.4	2527.1	149.1	36.2	3813.7	95.1	657.8	7279	7
Cha22	1678.6	2089.6	39.7	18.1	2466.6	619.6	724.9	5958.5	7
Gu101	1588.2	6511.5	57.5	64.2	2310.6	1314.1	12133.8	22391.9	7
Gu24	1627	10780.9	10	30.2	3032.4	2596.5	20278.2	36728.2	6.8
Gu25	1840.3	6250.8	29.1	23.5	2733.5	1203.6	10562.6	20803.1	8
Gu26	1262.1	10743.9	69.5	36.2	5445.5	1798.7	17251.6	35345.4	6.6
Gu27	1188.8	11,695	129.3	30.2	7126.2	1381.8	17,564	37926.5	7
Gu24	1553.7	10780.9	10	30.2	3032.4	2596.5	20278.2	36728.2	6.8
Gu26	1627.4	10743.9	69.5	36.2	5445.5	1798.7	17251.6	35345.4	6.6
Gu27	1602.2	11,695	129.3	30.2	7126.2	1381.8	17,564	37926.5	7
Gu31	1512.6	5881.5	13.4	48.1	2793.5	941.4	9819.9	19497.8	7
Gu32	1392.8	5637.1	63.1	36.5	6125.8	1568.2	2800.8	16231.5	7
Qian102	2324	11788.6	35.1	20.7	4492.9	7583	14132.2	38052.5	7
Qian153	2300.9	12,230	99.8	236.1	3430.5	6589.7	19672.8	42258.9	6.9
Qian163	2381.2	10597.2	41.7	25.3	3109.7	3696.4	18332.2	35802.5	7
Qianshen10	2180.7	17382.7	39.3	3.9	3589.3	6166.1	32264.3	59445.6	7
Qianshen4	2168.3	15822.9	15	9.5	1698.1	8075.3	28908.2	54,529	7
Qianshen214	2350.4	4482.6	21.4	70.7	3649.9	3401	1617	13242.6	6
Qian218	1822.55	2580.4	24.9	46.1	2895.2	86	2021.6	7654.2	7.5
Qian219	1836.4	4324.4	23	162.1	4644	2164.2	1343.7	12661.4	7
Qian220	2091.7	12542.4	13.4	12.8	1744.1	1220.9	28842.9	44376.5	8
Qian221	2183.1	11366.3	12.4	15.4	2205	2459.1	23357.8	39,416	7
Rang11	1840	3918.2	38.2	157.1	5026.8	1280.5	790.2	11,211	7.2
Rang15	1935.4	3292.2	32.8	63.9	3495.4	1765.1	838.4	9487.8	7.4
Rang24	1853.7	4187	36.2	129.3	5095.9	1119.6	1495	12063.2	7
Rang24	1858.9	3910.8	30.2	99.4	4826.2	1000.5	1255.8	11122.9	7
Rang58	2107.15	3460.5	8.8	110.8	4236.3	878	1158.8	9853.2	7
Xin251	1767.6	4579.6	22.1	75.2	5843.2	425.5	1896.5	12842.1	7
Xin254	1685.4	6719.3	18.1	39.7	2580.4	1298.3	11,955	22610.8	6.9
Xin256	1683.8	4532	18.1	39.7	2882.8	631.1	6476.7	14580.4	6.6
Xin256	1660.1	4306.3	18.1	29.9	2747.4	643.1	6065.4	13810.2	6.9
Xin257	1608.5	11,385	30	66.9	5193.1	476.5	21028.7	38180.3	7.2



**Fig. 13.** Characteristics of oil micro-occurrence in the  $K_1q_4$  tight sandstone reservoirs: (A, B and C) Photomicrographs of thin section showing oil accumulated around carbonate cements; (D) Photomicrographs of fluorescence showing oil accumulated around carbonate cements; (E) Photomicrographs of thin section showing oil accumulated around chlorite; (F) Idem with E but SEM Photomicrographs showing plate-like chlorite. Ca-carbonate cements; Ch-chlorite.

to the relative low oil saturation (Fig. 8). This was quite different from conventional sandstone reservoirs.

## 4.3. Main types of authigenic mineral

Several authigenic minerals are present in the sandstones of the  $K_1q_4$  tight sandstone reservoirs, but quartz, carbonates (Fig. 9) and clay minerals are the major cementing agents.



Fig. 14. Illustration of the influence of pH on the surface charge of common minerals (modified from Barclay and Worden, 2000).

## 4.3.1. Quartz cement

Quartz cement was one of the most common authigenic mineral in the studied tight sandstone reservoirs. Authigenic quartz mainly occurred as syntaxial overgrowths with approximately  $30-100 \,\mu\text{m}$  in size (Fig. 9A and B), and less commonly, as macrocrystalline euhedral crystals within the intergranular pores. Abundances of quartz cement range from 1.55% to 8.38% with an average of 5.62% (Fig. 10A). Furthermore, authigenic quartz distribution was scattered showing no obvious variation with samples location in the tight sandstone reservoirs (Fig. 10A).

## 4.3.2. Carbonate cements

Calcite, dolomite, ferrocalcite and ankerite are the main types of authigenic carbonate minerals in the  $K_1q_4$  tight sandstones (Fig. 9B–F). They mainly show as partially or completely pore-filling blocky or mosaic aggregates around the euhedral quartz overgrowth or partly replacing the authigenic quartz (Fig. 9B). Ferrocalcite and ankerite are more abundant than calcite and dolomite in the studied reservoirs (Fig. 9C-F). Additionally, calcite and dolomite are always zoned and engulfed by ankerite (Fig. 9E and F). In total, the content of the carbonate cements ranges from 1.97% to 12.93% with an average of 5.72% (Fig. 10B). However, it shows significant differences in distribution mode compared to quartz cement (Fig. 10). Carbonate cementation is commonly extensive at or within about 1.0 m of sandstone and mudstone contact surface, where carbonate cements are more than 8% (Fig. 10B). Generally, the carbonate cements decrease with increasing distance from sandstone-mudstone interface (Fig. 10B). The carbonate cements was reduced to less than 3%, when the distance from sandstone-mudstone interface was more than 7.0 m (Fig. 10B). Between the distance of about 3.5 m and 7.0 m, the reservoirs contained moderate amount of carbonate cements ranging from about 3.0% to 6.0% (Fig. 10B), where the sandstones still reserved some cemented residual pore spaces around carbonate cements.

#### 4.3.3. Clay minerals

Kaolinte, mixed-layered illite/smectite, illite and chlorite are the major types of authigenic clay minerals in the  $K_1q_4$  studied tight sandstones. Kaolinite, being a minor phase, occurs as pseudohexagonal crystals (Fig. 11A). Foliated or honeycomb mixed-layer illite/smectite (I/S) with R = 1 or R = 3 acts as the dominating authigenic clay

minerals in primary pores and grain surfaces (Fig. 11A and B). Illite mainly occurs as flaky debris within some primary and secondary pore spaces (Fig. 11C). Occasionally, small amounts of fibrous illite are developed around kaolinite as well in the deeper buried intervals. Authigenic iron rich chlorites appear as scattered flakes in the studied reservoirs (Fig. 11D-F). Generally, chlorite is distributed uneven in the pore spaces, and its content is associated with volcanic rock fragments. The content of chlorite increases with increasing amount of volcanic rock fragments, when its content is lower than about 35-40% (Fig. 12A). Afterwards, the content of chlorite shows no obvious trend with respect to the amount of volcanic fragments (Fig. 12A). This may be caused by extensive compaction with limited pore spaces reserved for pore fluids flow and authigenic minerals alteration when the amount of ductile lithic grains are high. There is additionally a negative correlation between the content of chlorite and carbonate cements in the  $K_1q_4$  sandstone reservoirs, especially when the carbonate content is less than about 5% (Fig. 12B).

## 4.4. Characteristics of formation water

Data of 236 formation water samples from the studied reservoirs reveal that about 98.1% of the samples are characterized by NaHCO<sub>3</sub> water and just a few formation water samples are characterized by MgCl<sub>2</sub> (about 1.52%) and Na<sub>2</sub>SO<sub>4</sub> (about 0.38%). The salinity of formation water is moderate to high, varying from 5958.5 mg/L to 59445.6 mg/L with an average of 24311.61 mg/L (Table 1). The pH of most formation water ranges from 6 to 7 with only a few samples from 7 to 8, showing very weak acidic to circum-neutral pH (Table 1).

## 4.5. Oil micro-occurrence characteristics

Based on the thin section evidence, SEM analyses and fluorescence observations, oil was distributed quite heterogeneously in the studied sandstones. Interestingly, oil preferred to accumulate in the residual pore spaces around carbonate cements (Fig. 13A–D). Furthermore, in the pores with moderate amount of scattered flakiness chlorite, oil also performed as preponderant accumulation compared to other pore spaces (Fig. 13E and F). The fluorescence colors of oil and hydrocarbon inclusions are all primarily blue and white (Fig. 13D), indicating a relatively mature and light oil (Zhang et al., 2009a,b; Dong et al., 2014).



**Fig. 15.** Distribution characteristics of divalent ion in the carbonate cements of the  $K_{1}q_{4}$  tight sandstones reservoirs: (A) Photomicrograph of the studied sample; (B) Photomicrograph of SEM showing Ca<sup>2+</sup> distribution; (C) Photomicrograph showing Mg<sup>2+</sup> distribution; (D) Photomicrograph of SEM showing Fe<sup>2+</sup> distribution; (E and F) Photomicrograph of SEM and EDS of the marked site showing ankerite. An-ankerite.

## 5. Discussions

## 5.1. Formation of the main authigenic minerals

Previous studies of the authors had focused on authigenic minerals formation mechanisms, i.e., quartz, carbonates and some clay minerals, extensively (Xi et al., 2015a, b). Firstly, quartz cementation was a continuous process from about 60 °C to 130 °C, centering on 70 °C to 100 °C (Xi et al., 2015a). One of the most important silica sources was explained to be smectite to illite reaction (Xi et al., 2015a; Lynch et al., 1997):

10.93 I/S (20%I) + 0.91 discrete illite + 2.75 kaolinite + 0.86 K-feldspar + 1.46 plagioclase + 2.11 A1<sub>2</sub>O<sub>3</sub> + 2.66 K<sub>2</sub>O → 12.59 I/S (85% I) + 0.27 chlorite + 3.16 quartz + 1.98 albite + 4.70 SiO<sub>2 (aq)</sub> (1)

During this process, the decrease in smectite-layer and increase in illite-layer make the random mixed-layer I/S transform to ordered

mixed-layer I/S or illite, and precipitated the chlorite and authigenic quartz at the same time. This reaction occurred relatively earlier (mainly from 70 °C to 90 °C) and provided about 50–60% silica source for the quartz cements (Xi et al., 2015a). Therefore, quartz cementation and authigenic clay minerals formation (i.e., flaky illite and chlorite) are the related reactions at a relative earlier stage in the diagenetic sequence of the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs. Pressure solution (chemical compaction) between detrital quartz was another major silica source for the authigenic quartz (Xi et al., 2015a).

Carbonate cements characterized by the relatively low negative  $\delta^{13}$ C values mainly originated from organic matter decarboxylation of adjacent mudstones or source rocks (Xi et al., 2015b). Conversion of volcanic rock fragments and the smectite to illite reaction could release extra Ca<sup>2+</sup>, Mg<sup>2+</sup> and Fe<sup>2+</sup> into pore water (Boles and Franks, 1979; Stroker et al., 2013), which provided cations for carbonate cements, such as ferroclacite and ankerite. The concentrations of Ca<sup>2+</sup>, Mg<sup>2+</sup> and Fe<sup>2+</sup> along the sandstone and mudstone contact surfaces are higher



Fig. 16. Relationship between carbonate content and contact angle in the  $K_1q_4$  tight sandstone reservoirs.

than in the central parts of the sandstones, thus carbonate cements extensively occurred along the sandstone-mudstone interface, and decreased with increasing of the distance from mudstone (Xi et al., 2015b). According to the fluid inclusion thermometry and oxygen isotope analysis, the precipitation temperature of the carbonate cements ranges from 83.78 °C to 130.96 °C with an average of 108.59 °C, showing a relative later stage in the diagenetic sequence (Xi et al., 2015b).

By comparison, oil emplacement in the  $K_1q_4$  tight sandstone reservoirs was mainly posterior to quartz cementation and associated authigenic clay minerals, including ordered mixed-layer illite/smectite, illite and chlorite (Xi et al., 2015b). However, carbonate cementation occurred at late stage of the diagenetic sequence accompanied by oil emplacement, when the reservoir porosity reduced to about 10% (Xi et al., 2015b). Therefore, oil charge and accumulation behavior in such tight reservoirs might be impacted by associated authigenic minerals.

## 5.2. Authigenic minerals and wettability alteration

Wettability alteration is an important issue in oil exploration and exploitation researches, especially for the unconventional reservoirs. During the oil recovery, most studies have focused on changing reservoirs from oil-wet to water-wet (Meybodi et al., 2011; Hadia et al., 2013; Khajepour et al., 2014; Liu et al., 2015; Karimi et al., 2015). On the contrary, wettability changes of some pore spaces from water-wet to oil-wet has been less studied and there is a potential to better understand the oil charge and accumulation processes, especially for tight sandstones (Wardlaw, 1982; Qi et al., 2015). Essentially, wettability alteration is the results of zeta potential changes under a specific

reservoir conditions (Hiorth et al., 2010; Alotaibi et al., 2011), with oil having a negative surface potential (Hiorth et al., 2010). Therefore, if the pore surfaces charge is positive, the water film will collapse immediately and the surface will be oil-wet. In the reservoirs with high salinity formation water, monovalent and divalent cations can interact with reservoir fluids and rock surface, which will disturb the charges' stability (Zhang et al., 2007; Alotaibi et al., 2011). Previous studies showed that extra divalent cations, i.e.,  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Fe^{2+}$ , in the reservoirs would produce more positive charges on the rock surface, which acted as attractive sites for negative ends of polar components in crude oil (Kia et al., 1987; Xu et al., 2016; Zhang et al., 2007; Alotaibi et al., 2011). Furthermore, Mg<sup>2+</sup> are more effective for wettability alteration than Ca<sup>2+</sup> due to the faster increase of zeta potential with increasing of their concentrations in the reservoirs (Zhang et al., 2006; Zhang et al., 2007; Alotaibi et al., 2011). Moreover, samples that contained more Fe<sup>2+</sup> showed stronger lipophilicity than others (Toledo et al., 1996). Except for divalent cations, the surface charge of a mineral is also sensitive to the pH of formation water, which can indirectly control the wettability of a reservoir (Barclay and Worden, 2000). For example, the surface charge of quartz and feldspar are positive at very low pH, and negative at high pH. Carbonate minerals are positively charged until pH reached to about 8 (Barclay and Worden, 2000). In particular, Iron-rich silicates and carbonates, such as chlorite, siderite or ankerite, are always positively charged (Barclay and Worden, 2000). Formation water of the studied sandstone reservoirs are characterized by weak acid to circum-neutral pH and high salinity. So, the surface charge of common minerals is summarized in Fig. 14.

Accordingly, although the studied tight sandstone reservoirs were originally characterized by water-wet and detrital grains distributed relatively uniform, diagenetic reactions may have altered the wettability through mass transformation and ion migration. In the studied reservoirs, carbonate cements and scattered flakiness chlorites are the main types of authigenic minerals with divalent cations rich.

## 5.2.1. Carbonate cement and wettability alteration

According to the thin section observation and XRD analyses, carbonate cements in the  $K_1q_4$  sandstone reservoirs were mainly ferrocalcite and ankerite that only occurred in parts of the pore spaces (Fig. 9C–F). Furthermore, pore-filling carbonate cements were mainly characterized by  $Mg^{2+}$  and  $Fe^{2+}$  rich (Fig. 15). In these reservoirs with weak acid to circum-neutral pH and high salinity formation water,  $Mg^{2+}$  and  $Fe^{2+}$  caused extra divalent cations on carbonate cement surfaces, especially the  $Fe^{2+}$  enrichment, leading these pore spaces positively charged. When negatively charged crude oil injected into the reservoirs, the pore spaces filled with moderate carbonate cements would attract them preferentially. Therefore, carbonate cements in the  $K_1q_4$  sandstone acted as an important medium to alter the wettability of some pore spaces, where the water-wet pore spaces surfaces would turn



Fig. 17. Zeta potential of clay minerals under different salinity (Toledo et al., 1996) and pore water salinity distribution in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs.



Fig. 19. Relationships between content of carbonate cements and oil saturation in the  $K_1q_4$  tight sandstone reservoirs.

into oil-wet.

It was found that contact angles between the oil and reservoir rocks increase with increasing amount of the carbonate content when the carbonate content is lower than about 6–7% (XRD results) (Fig. 16). This was because, with the increasing of carbonate cements, more and more pore spaces occupied by authigenic carbonates. In such content ranges, there are still some residual pore spaces preserved around carbonate cements. Consequently, these pore space surfaces turn into oil-wet, causing the contact angle of the whole reservoir rocks to increase. When the content of carbonate cements is more than about 6–7% (XRD result), however, contact angles decrease with increasing amount of carbonate cements and the plots were more scattered (Fig. 16), indicating complete cementation in this part of the rock and no residual pore space for oil emplacement. The mount of carbonate redistribution and newly growth therefore appears to play a key role in oil charge and accumulation.

## 5.2.2. Authigenic chlorite and wettability alteration

As to the clay minerals, zeta potential has been determined by both mineral types and pore water salinity (Alotaibi et al., 2011). In de-ionized water, all of the clay minerals, i.e., smectite, kaolinite, illite and chlorite, showed negative charges, whereas chlorite became positively charged in seawater conditions(Alotaibi et al., 2011) (Fig. 17A). For the  $K_1q_4$  sandstone reservoirs, formation water performed as high salinity, varying from 5958.5 mg/L to 59445.6 mg/L with an average of 24311.61 mg/L (Table 1; Fig. 17B); and authigenic chlorite appeared as plate-like with  $Mg^{2+}$  and  $Fe^{2+}$  rich (Fig. 11D–F). Hence, chlorite usually had positively charged edges in the reservoir conditions and acted as oil-wet behavior (Toledo et al., 1996; Zhang et al, 2000; Alotaibi et al., 2011). Similar with the carbonate cements, negatively charged crude oil would prefer to adsorb in the pore spaces with authigenic chlorite. Conversely, other clay minerals, such as kaolinite, smectite and illite are negatively charged both in de-ionized water and



Fig. 20. Relationship between oil saturation, QGF index and carbonate cements content in the  $K_1q_4$  tight sandstone reservoirs.



Fig. 21. Relationship between the distance to interface of sandstone-mudstone and oil saturation in the  $K_{1}q_4$  tight sandstone reservoirs.

seawater (Fig. 14), which can't alter the wettability of the studied reservoirs.

Relationship between the content of the clay minerals and contact angle also showed that it is the clay mineral types rather than its total content to influence the reservoirs wettability (Fig. 18). Firstly, there is no obvious correlativity between the total content of clay minerals and contact angle (Fig. 18A). When referred to specific authigenic clay minerals, the relative content of illite and mixed-layer illite/smectite increased with decreasing contact angle (Fig. 18B and C). On the contrary, it is showed a positive correlativity between the relative content of chlorite and contact angle (Fig. 18D). It indicates that authigenic chlorite can alter the wettability of some pore space surfaces from water-wet to oil-wet in certain extent.

## 5.3. Authigenic minerals impact on oil accumulation

Among authigenic minerals in the  $K_1q_4$  sandstone reservoir, carbonate cements and authigenic chlorite can alter the initial wettability from water-wet to oil-wet in parts of the pore spaces. Thus, they might have some significant impacts on tight sandstone oil accumulation processes. In this study, oil saturation and Quantitative grain fluorescence (QGF) were regarded as the main parameter to reflect the oil accumulation processes. QGF can accurately detect both current and palaeo oils in petroleum reservoirs; and QGF index can be regarded as a quantitative indication to determine the reservoirs oiliness like oil saturation (Liu and Eadington, 2005).

## 5.3.1. Carbonate cements impact on oil accumulation

The relationship between oil saturation and content of carbonate cements in the studied sandstone reservoirs is rather complicated. The oil saturation increased with increasing of carbonate cements, when the content is less than about 6–8% (XRD results) (Fig. 19). However, if the content of the carbonate cements is more than this, the reservoirs mainly show lower oil saturation around 10% (Fig. 19). This suggested that carbonate cements could serve a dual function in the tight sandstone reservoirs. On one hand, they could destroy the pore spaces, making the reservoir properties poorer; on the other hand, carbonate cements might alter wettability of some pore spaces in a moderate content ranges, promoting the oil accumulation in tight sandstone reservoirs.

In order to further study the impacts of authigenic carbonates on oil accumulation, relationships between the sealed coring oil saturation, QGF index and the content of carbonate cements (image analysis results of the thin section) were analyzed. It shows that the highest oil saturation and QGF index correspond to the carbonate cements content of 4-5% in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs (Fig. 20). When the content of carbonate cements is less than about 5%, the oil saturation and QGF index both correlates positive with carbonate cement content (Fig. 20). When the content of carbonate cements is higher than about 5% (image analysis results), on the contrary, the oil saturation and QGF index decrease with increasing carbonate cement content (Fig. 20). The relationship is very similar to the one observed between the contact angle and content of carbonate cements (Fig. 16). Accordingly, in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs, if the content of carbonate cements is larger than about 5% (image analysis results), there are probably very limited pore spaces preserved for oil accumulation. When carbonate cements are less than about 5% (image analysis results and petrographic observations), more and more residual pore spaces around the cements can turn into oil-wet with increasing carbonate cements. Thus, more crude oil would be attracted into these related reservoirs by oil-wet carbonate cements, showing higher oil saturation. These lead to a conclusion that the reservoirs containing about 4-5% (image analysis results) carbonate cements are most preferable for oil accumulation in the K<sub>1</sub>q<sub>4</sub> sandstones. Petrological evidences showed that oil preferred to accumulate in the cementation residual pore spaces around carbonate cements (Fig. 13A-D), also support the favorable impacts of oil-wet carbonate cements on oil accumulation in the K1q4 sandstone reservoirs. In addition, the differences in inflection point values of the carbonate cements content in Figs. 16, 19 and 20 were mainly caused by different analysis methods.

The content of carbonate cements was closely related to the distance of the samples from sandstone-mudstone interface (Fig. 10B), which probably could influence the distribution of oil saturation in tight sandstone reservoirs. According to the quantitative statistics, the highest oil saturation appeared in the samples with about 4–6 m far



Fig. 22. Schematic illustration of wettability alteration caused by carbonate cements and the impacts on oil accumulation in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs.

away from the distance to the sandstone-mudstone interface, where the content of carbonate cements ranged from 3 to 6% (Fig. 10B, Fig. 21). When the distance to the sandstone-mudstone interface was less than about 6 m, oil saturation increased with increasing of the distance, whereas oil saturation decreased with the distance further increasing (Fig. 21). The reservoirs closest to the mudstones were extensively cemented by carbonates with limited pore space for oil accumulation. Although, the reservoirs far away from the mudstones had relative high porosity with less carbonate cements, the pore space surfaces were lack of oil-wet behavior with high capillary resistance, performing as low oil saturation. Consequently, in the  $K_1q_4$  tight sandstone reservoirs, oil mainly accumulated in the reservoirs between sandstone-mudstone interface and central parts of the sand body, where the reservoirs contain about 4–5% (image analysis results) carbonate cements.

In conclusion, the impacts of carbonate cements on oil accumulation could be summarized as follows: ① After compaction and quartz cementation, the sandstones became tight with poor porosity, permeability and small pore-throat size. Most of the detrital grains were surrounded by irreducible water films and water-wet (Fig. 22A). Thus, the whole reservoir rocks showed strongly water-wet properties with high capillary resistance for crude oil emplacement initially. Compared to the major occurring time, oil emplacement was later than carbonate cementation (Xi et al., 2015b). At the beginning of the cementation, oil saturation was so low that couldn't prevent the carbonate cements precipitation. Therefore, carbonate cements extensively occurred along the sandstone-mudstone interface and decreased with increasing distance to sandstone-mudstone interface (Fig. 22A). 2 When the oil generated from source rocks and migrated into the sandstone reservoirs through oil source faults, it would choose the pore spaces larger than oil charging threshold to enter into the reservoirs that farther away from sandstone-mudstone interface (Xi et al., 2016). Generally, quite few oil could inject into the sandstones along sandstone-mudstone interface (mainly less than 1-2 m far away from mudstone) due to extensive cementation with limited pore spaces preserved (Fig. 22B). ③ Previous studies showed that the controlling of wettability was stronger than pore-throat size for oil accumulation processes in tight oil reservoirs,



Fig. 23. Relationship between QGF index, oil saturation and clay mineral contents in the  $K_1q_4$  tight reservoirs.



Fig. 24. Resin and asphaltene distribution characteristics of the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs.

moreover, the oil would migrate easily in the pore spaces with oil-wet surfaces, forming preferred migration pathways (Buckley, 2001). Therefore, the oil was firstly prone to migrate and accumulate in the pore spaces around moderate amount of carbonate cements (about 4–5%) after enter into the K<sub>1</sub>q<sub>4</sub> sandstone reservoirs (Fig. 22C). ④ Then, with the increasing of oil emplacement, some oil could also enter into the reservoirs with higher porosity and less carbonate cements in central part of the sand bodies (Fig. 22D). ⑤ Afterwards, oil would further adjust to the adjacent oil-wet pore spaces with moderate amount of carbonate cements from the water-wet pore spaces in central part of the sand bodies through oil spontaneous imbibition induced by carbonate cementation as well as preceding oil accumulation (Fig. 22E). Finally, oil mainly accumulated in the reservoirs with 4–5% carbonate cements (Fig. 22E).

## 5.3.2. Authigenic chlorite impacts on oil accumulation

In the reservoirs with carbonate cements lacking, oil-wet authigenic chlorite probably has some considerable impacts on oil accumulation. In particular, the content of chlorite increase rapidly with decreasing carbonate content in some reservoirs when the carbonate was less than about 5% (Fig. 12B). Firstly, oil saturation and the QGF index are negatively related to relative content of illite and I/S in the  $K_1q_4$  tight sandstone reservoirs (Fig. 23A–D). Although plate-like chlorite could, as illite and I/S, destroy the porosity and permeability, positive correlation relationships exist between the relative content of chlorite and oil saturation as well as QGF index (Fig. 23 E, F). This correlations match well with the relationships between contact angle and relative content of clay minerals (Fig. 18; Fig. 23), which support that wettability alteration caused by authigenic clay minerals precipitation has played a significant roles on the oil charging and accumulation in tight sandstone reservoirs. Specifically, oil-wet plate-like chlorite was more favorable to oil accumulation than water-wet illite and I/S in the  $K_1q_4$  tight sandstone reservoirs.

In addition, clay minerals act as adsorbents and reactants with crude oil components in sandstone reservoirs (Wu et al., 2012). Wettability alterations of sandstone reservoirs, particularly those containing oil-wet



Fig. 25. Relationship between content of clay minerals and oiliness in the K1q4 tight sandstone reservoirs of Well Rang 59.

clay minerals, have been attributed to the adsorption of the heavy polar ends of the crude oil onto mineral surfaces (Wu et al., 2012; Lebedeva and Fogden, 2011). In the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs, the crude oil was mainly free oil characterized by low content of resins and asphaltenes, ranging from 9.8% to 19.5% with an average of 14.84% (Fig. 24). When this type of crude oil entered into the tight sandstone reservoirs, it would migrate to the pore spaces with oil-wet authgenic minerals firstly. Afterwards, oil could interact with oil-wet authigenic clay minerals, i.e. chlorite, and the heavy polar ends could absorb onto clay mineral surfaces, leading to form "clay-oil flocs" (Wu et al., 2012). This way, the adhesion of oil to other minerals was reduced and allowed it to be easily removed by gentle water motion in following migration process (Owens, 1999; Wu et al., 2012). Hence, not only has chlorite altered the wettability of related pore spaces and provided preferential accumulation spaces for oil in the tight sandstone reservoirs, but also it can reduce the adhesion of oil through forming "clay-oil flocs" and promote further migration. This means that plate-like iron rich chlorite may have acted as a favorable factor for oil enrichment in the K1q4 sandstone reservoirs.

Take the typical tight sandstone reservoirs from 2100 m to 2130 m of Well Rang59 as an example (Fig. 25). In the entire reservoir intervals, the porosity and permeability were extremely low (Fig. 25). It showed insignificant relationships between the relative content of clay minerals and reservoir porosity, permeability. Moreover, porosity and permeability were not the main controlling factors for sealed coring oil saturation and QGF index (Fig. 25). Instead, the distribution of sealed coring oil saturation and QGF index were positively associated with the content of chlorite in the K<sub>1</sub>q<sub>4</sub> tight sandstone reservoirs with similar

amount of carbonates (Fig. 25), indicating that authigenic chlorite actually contributed to oil accumulation.

Similar with carbonate cements, the impacts of authigenic chlorite on oil accumulation could be summarized as follows: ① Most of the detrital grains were surrounded by irreducible water films and waterwet. Alteration of the volcanic rock fragments could generate various clay minerals, such as smectite, illite, I/S and chlorite. The pore spaces with illite and I/S mainly showed strong water-wet surfaces, however, plate-like chlorite have altered some pore-throat surfaces to be oil-wet (Fig. 26A). <sup>(2)</sup> After the oil generated from source rocks and migrated into the sandstone reservoirs through oil source faults, it would choose the pore spaces larger than oil charging threshold to enter into the reservoirs (Fig. 26B). 3 Then, crude oil preferred to migrate into the pore spaces filled with oil-wet chlorite. Contrarily, it is difficult for oil to charge and accumulate in the pore spaces with illite and I/S rich since strong water-wet and high capillary resistance (Fig. 26C). (4) Afterwards, crude oil interacted with oil-wet chlorite and formed "clay-oil flocs". Thus, the adhesion of oil to other minerals reduced (Fig. 26D). S Finally, crude oil with lower adhesion could migrate more easily into other oil-wet pore spaces with plate-like chlorite and formed relative high oil saturation parts in the tight sandstone reservoirs (Fig. 26E).

#### 6. Conclusions

(1) The  $K_1q_4$  tight sandstone reservoirs are compositionally immature with an average framework composition of  $Q_{43}F_{26}L_{31}$ . Framework grains distribute homogeneously in the tight sandstones. Authigenic minerals, however, only occur in some parts of the intergranular



Fig. 26. Schematic illustration of wettability alteration caused by the formation of authigenic chlorite and the impacts on oil accumulation in the  $K_1q_4$  tight sandstone reservoirs.

pores and show heterogeneous distribution characteristics. Initially, the reservoirs rocks are characterized by strong water-wet properties.

- (2) The  $K_1q_4$  tight sandstone reservoirs are characterized by poor reservoir properties and low oil saturations. Porosity and permeability are not the main factors controlling oil saturation any more. Even in small scales with similar sandstones, the oil saturations were quite heterogeneous.
- (3) Several authigenic minerals are present in the sandstones of the  $K_1q_4$  tight sandstone reservoirs, but quartz, carbonates and clay minerals are the major cementing agents. Authigenic quartz distribution is scattered showing no obvious variation with samples location in the tight sandstone reservoirs. The content of carbonate cements are, however, closely related to the distance from sandstone-mudstone interface. Kaolinte, mixed-layered illite/smectite, illite and chlorite are the major types of authigenic clay minerals. Among them, authigenic chlorite appeared as scattered flakiness crystals with iron rich.
- (4) Carbonate cements and authigenic chlorite in the  $K_1q_4$  sandstone may help modifying the wettability of some parts of the pore spaces through divalent cations surplus, where the water-wet pore-throat surfaces will turn into oil-wet surfaces.
- (5) In the  $K_1q_4$  sandstone reservoirs, oil prefers to accumulate in the cemented residual pore spaces around the carbonate cements and

chlorite. The reservoirs locating between sandstone-mudstone interfaces and central parts of the sand body are more preferable for oil accumulation, where the content of carbonate cements is about 4–5% (image analysis result). Not only does chlorite alter the wettability of related pore spaces and provide preferential accumulation spaces for oil in the tight sandstone reservoirs, but it can also reduce the adhesion of oil through forming "clay-oil flocs" and promote further migration.

(6) Oil charge and accumulation in tight sandstone reservoirs is quite difficult and rather complicated due to poor porosity and permeability, small pore space sizes and high capillary resistance. Once, moderate oil-wet authigenic minerals precipitate in the pores, oil will be prone to migrate to these directions and forming prepotent oil accumulation (Buckley, 2001; Qi et al., 2015). Therefore, it is essential to look for reservoirs with moderate amount of oil-wet authigenic minerals during tight oil exploration, especially when the source rocks, structure locations and migration systems are similar.

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