Origin and controlling factors of chlorite coatings—an example from the reservoir of T₃x Group of the Baojie area, Sichuan Basin, China

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Abstract: The study of the chlorite coatings always attracts scholars in China and other countries because the chlorite coatings play an important role in the preservation of residual primary pores in sandstone reservoirs. At present, the study of the origin and the controlling factors is relatively few. The occurrence, time of formation, genesis, controlling factors, and the mechanism of chlorite coatings inhibiting quartz overgrowths were studied in detail with thin section and SEM analysis. Samples were from the sandstone reservoirs of the T₃x Group in the Baojie area, the transitional zone from the middle to the south of Sichuan Basin. The results indicate that the chlorite coatings on the walls of the pore spaces are oriented perpendicular to grain surfaces in the form of isopachous (even-thickness) grain-coating, while the chlorite coatings at the contacts between adjacent detrital grains are arranged with a preferred orientation tangential to the surface of detrital grains. The chlorite coatings were formed in the eogenetic stage. They were formed by recrystallization of Fe-rich clay films during the syndepositional period, and chlorite cements would be recrystallized after the coatings' formation. The formation of chlorite coatings was mainly controlled by the depositional environment, provenance conditions, and diagenetic environment. The presence of chlorite coatings could result in the preservation of primary pores in deeply buried sandstone reservoirs by effectively inhibiting quartz overgrowths and the development of compaction and pressure solution.

Key words: Chlorite coatings, occurrence, time of formation, origin, controlling factor, quartz overgrowths, T₃x Group

1 Introduction

Chlorite cements are the most observed clay cements in sandstone deposits and have multiple types of occurrence. Grain-coating chlorite cement is defined as chlorite coatings. The study of the origin of chlorite coatings and the mechanism of pore preservation attracts scholars in China and other countries because of the close relation between chlorite coatings and preservation of the primary pores in sandstone reservoirs. Ehrenberg's (1993) studies of the sedimentology and lithology characteristics of Jurassic continental shelf sandstones in Norway indicated that the formation of chlorite coatings was controlled by the depositional environment. The chlorite coatings were mainly formed in an energetic offshore deltaic environment. Besides, several other conditions, such as adequate sources of ferric ions, high content of rigid grains, and the absence of calcite cement, had a great influence on the formation of chlorite coatings. Baker et al (2000) suggested that chlorite coatings, formed in the early diagenetic stage, were a good sign of an onshore fresh water environment and

a sea water deltaic environment. Pittman et al (1992) studied the upper Cretaceous sandstone reservoirs in Louisiana and showed that the formation of Fe-rich chlorite coatings was controlled by the provenance conditions. Lynch's (1996) study of the Fresnian sandstones indicated that chlorite coatings were formed at the eogenetic stage, and their growth continued through the whole sandstone burial history. Zeng's (1996) study suggested that the origin of chlorite coatings was closely related to alteration of sediments from volcanic terrains. Huang et al (2004) indicated that the saline lake delta environment assisted the formation of chlorite coatings. Through thin section analysis and SEM (scanning electron microscope) analysis, the origin mechanism and the controlling factors of chlorite coatings in the sandstone reservoir of T₃x Group in the Baojie area, the transitional zone from the middle to the south of Sichuan Basin were studied in detail in this paper.

2 Regional geological background

The Baojie region is located in three counties, Longchang, Rongchang, and Dazu of Sichuan province, and its area is 1,600 km². The region belongs to the Weiyan-Longnüsi structural group of a gentle palaeohigh in middle Sichuan.

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The Weiyuan-Longnusi structural group is a giant nose structure consisting of many small nose structures. The major gas-producing formation is in the upper Triassic T₃x Group which is a combination of sandstone and mudstone with obvious lithological cyclicity in the vertical direction. The formation can be divided into six sections from bottom to top. The second section, the forth section, and the sixth section are the main reservoir intervals, and the lithology is massive sandstones interbedded with lamellar mudstones, which are delta-front subfacies. The first section, the third section, and the fifth section are secondary reservoir intervals, and the lithology is mainly mudstones and silty mudstones imbedded with thin-medium bedded sandstones, which are prodelta-shallow lake subfacies and delta-front subfacies. The reservoirs consist of mainly feldspathic litharenite, lithic arkose, and partially lithic sandstone. The compositional maturity and textural maturity of rocks are moderate to low. According to grain grade classification, the rock type of reservoirs is mainly medium sandstone, secondly fine sandstone, grit stone, and siltstone. Both the sorting and the roundness are moderate. The shape of the sand grains is primarily subrounded-subangular. Point contact and linear contact are the major contact types between grains. The cements in the rocks are mainly carbonates, authigenic clay minerals, and siliceous minerals, and the average content of cements is approximately 7.1%, which is moderate. The physical properties of reservoirs are generally poor, with extremely low porosity and extremely low permeability. All samples used in the study are from drill cores in the T₃x Formation. Table 1 shows the sample details.

Table 1 Details of chlorite coatings sampling

Well name	Horizon	Depth, m	Number	Lithology
Baoqian001-1	T ₃ x Group of upper Triassic	1424-1970	42	sandstone
Baoqian001-6	T ₃ x Group of upper Triassic	1299-1842	28	sandstone
Baoqian001-11	T ₃ x Group of upper Triassic	1337-1902	15	sandstone
Baoqian001-16	T ₃ x Group of upper Triassic	1384-1937	20	sandstone
Bao36	T ₃ x Group of upper Triassic	1762-2321	8	sandstone
Bao46	T ₃ x Group of upper Triassic	1318-1862	6	sandstone

3 Crystal structure of chlorite

Chlorite is a layered hydrous aluminosilicate with 2:1 type structure. It has a three-layer tetrahedral-octahedral-tetrahedral structure. The 2:1 type structure of chlorite mineral is made up of a 14Å basic structural unit which is stable at high temperature because the interlayer is filled by positively charged octahedra, which is different from other 2:1 type minerals (Fig. 1). The chemical composition of chlorite is very complex because of extensive isomorphous substitution. The general formula of chlorite is as follows (Liu et al, 1998):

According to the different superposition modes of octahedron and layered hydrous aluminosilicate, chlorite is divided into four types Ia, Ib, II a, and II b. The Ia and Ib types are products of low temperature crystallization. The Ia type is mainly formed during retrograde metamorphism; Ib type is mainly formed in diagenetic environment; IIb type is formed near metamorphic zone (150°C-200°C) (Odin, 1990). Chlorite coatings are usually formed in the eogenetic (early diagenesis) stage. Their structure is type Ib. With the increase of temperature and pressure, Ib type would possibly transform into IIb type through recrystallization (Liu et al, 1998).

Fig. 1 Crystal structure of chlorite (Grim et al, 1960)

X-ray diffraction is a very important means of identifying clay minerals. The X-ray diffraction pattern of well-crystallized chlorite usually demonstrates the basal plane reflection and higher order reflections at 14Å (001), 7Å (002), 4.7Å (003), 3.5Å (004), and 2.8Å (005), which is a main identification criterion of chlorite. Besides, the actual positions and intensity of these reflection peaks are also influenced by the cations in the crystal, so that the diagenetic stage of chlorite can be determined by X-ray diffraction.

4 Occurrence of chlorite cement

The occurrence of chlorite cement in the sandstones of the T₃x Group has two different petrographic types: grain coatings and pore filling. The former is the most important occurrence type. The characteristics are as follows:

1) Chlorite grain coatings are very common in the sandstone reservoirs of the Baojie area. As shown in Fig. 2, the morphology of chlorite coatings formed in different locations in sandstone is quite different. Thin section observation indicated that the chlorite coatings on the wall of the pore spaces were oriented perpendicular to grain surfaces with a fibrous pattern (Fig. 3(a)). SEM observation indicated that the chlorite coatings were oriented perpendicular to grain surfaces with a face-to-edge or face-to-face foliaceous pattern (Fig. 3(b)). The coatings are continuous even-thickness layers. The average thickness of coatings is approximately 5 μm , and the content of coatings in sandstones ranges from 1% to 4%. Compared with the chlorite coatings on the wall of the pore spaces, the chlorite coatings at the contact between adjacent detrital grains are arranged with a preferred orientation tangential to the surface of detrital grains. The coatings are thin and irregular because of compaction (Figs. 3(a) and (b)).

2) Chlorite pore filling is not common in the sandstones of the T₃x Group. The chlorite cement fills the pores of sandstones, showing foliaceous or rosette patterns. The chlorite cement of foliaceous pore filling pattern was itself full of pores, and the crystals became coarser and more euhedral from the grain edge to the centre of the pore (Fig. 3(d)). Chlorite of rosette pore filling pattern is separately distributed in the pores (Fig. 3(e)).

Fig. 2 Sketch of chlorite coating morphology (Billault et al, 2003)
Q = detrital quartz, QII = quartz overgrowth

5 Time of formation of the chlorite coatings

There is still a variety of opinions on the time of formation of chlorite coatings. Most scholars believe that chlorite coatings were formed in the eogenetic stage (Ehrenberg, 1993; Baker et al, 2000; Zhu et al, 2004; Huang et al, 2004;

Sun et al, 2008; Wang et al, 2007). However, some scholars believe that they are a product of the late diagenetic stage (Liu et al, 1998). In this paper, the study of chlorite coatings in the sandstone reservoirs of the T₃x Group suggested that chlorite coatings were formed in the eogenetic stage. Chlorite recrystallized after chlorite coatings were first formed and the formation and recrystallization continued throughout the whole mechanical compaction stage. Several petrographic characteristics can be used to determine the time of formation of chlorite coatings:

1) Scanning electronic microscopy (SEM) and thin section analysis revealed the presence of chlorite coatings at the contact point between adjacent grains with the chlorite coatings oriented parallel to the grain surface, which were considerably different from the radial orientation of crystals in chlorite coatings on the walls of the pore spaces (Figs. 3(a) and (b)). This suggested that grain-coating chloritization began before the complete compaction of sediment.

2) Illite cement and authigenic quartz overlapped chlorite coatings, which indicated that the chlorite coatings were formed earlier than the illite cement and authigenic quartz (Fig. 3(c)).

3) Moldic pores enclosed with chlorite coatings were commonly seen in SEM observation. This showed that the chlorite formed before the feldspar grains dissolved (Fig. 3(g)).

4) There was a significant difference in arrangement, morphology, and size of the chlorite coatings of foliaceous pore filling pattern from grain edges to the centre of the pores. The chlorite crystals became coarser and more euhedral from grain edges to the centre of the pores, which indicated that they were formed during different periods (Fig. 3(d)). The chlorite coatings on the grain edges were a product of the eogenetic stage, while the second and third generation chlorite cements from grain edges to the centre of the pores resulted from precipitation from percolating formation water during later stages of diagenesis.

6 Origin of chlorite coatings

Most foreign scholars consider that chlorite coatings were formed by recrystallization of Fe-rich clay during the syndepositional period. Ehrenberg (1993) suggested that Fe ions in solution in the delta front zone could result in a gel-like Fe-rich clay layer. Odin's (1990) study indicated that metastable Fe-rich clays (a 7Å mineral) were formed through a syndimentary interaction between seawater and terrigenous sediment. Clays rich in ferrous ions could transform into chlorite coatings during early diagenesis. Grigsby (2001) considered that the radially oriented clay films around detrital grain surfaces could dissolve and recrystallize with increasing formation pressure and temperature. The 7Å layer ferric clays were transformed into 7Å-14Å mixed-layer ferrous clays through the process of dissolution and reprecipitation, and then the clays rich in ferrous ions were transformed into chlorite coatings. Billault et al (2003) suggested that the formation and growth of chlorite coatings occurred in three stages (Fig. 4): 1) deposition of clay films

Fig. 3 Thin section optical micrographs and SEM images of chlorite coatings

(a) Fibrous chlorite coatings on the wall of the pore spaces (CP) oriented perpendicular to the grain surfaces. Chlorite coatings at the contact between adjacent detrital grains (CC) are arranged disorderedly with a preferred orientation tangential to the surfaces of the detrital grains. The grains (G) contact with each other with the convex-concave or sutured shape at the grain contact point where no chlorite coatings (GC) exist. From well Baoqian 001-6, burial depth of 1,781.22-1,781.25 m, crossed polarizer photograph. (b) Foliated chlorite coatings on the wall of the pore spaces (CP) oriented perpendicular to the grain surfaces. Chlorite coatings at the contact between adjacent detrital grains (CC) are arranged disorderedly with a preferred orientation tangential to the surface of detrital grains. From well Baoqian 001-1, burial depth of 1,795.52-1,795.73 m, SEM. (c) Illite cement (I) and authigenic quartz (AQ) overlapped earlier chlorite grain coatings (C). From well Baoqian 001-16, burial depth of 1,777.75-1,777.78 m, SEM. (d) Chlorite pore fillings (CF) become coarser and more euhedral from grain edges to the centre of the pores, which indicated that they were formed during different periods. From well Baoqian 001-11, burial depth of 1,718.70-1,718.93 m, SEM. (e) Foliated chlorite coatings on the wall of the pore spaces (CP) oriented perpendicular to the grain surfaces. Irregular clay minerals (AC) at the contact between adjacent detrital grains were observed. Chlorite rosettes (CR) are separately distributed in the pores. Quartz overgrowths (QII) developed. From well Baoqian 001-6, burial depth of 1,740.3 m, SEM. (f) Biotite alteration (BF) was observed. The pleochroism and twinkling vanished and the interference color faded into first order gray or yellowish. From well Baoqian 36, burial depth of 2,073.31-2,075.12 m, crossed polarizer photograph. (g) Moldic pores (MP) were entirely surrounded with chlorite coatings. From well Baoqian 001-11, burial depth of 1,707.54-1,707.67 m, SEM. (h) Calcite cement (CA) formed before chlorite coatings, which filled the pore spaces and were partly dissolved later. From well Baoqian 46, burial depth of 1,849.27-1,849.29 m, plain light photograph. (i) Quartz overgrowths (QII) occurred at the point (PP) where chlorite coatings were absent. From well Baoqian 001-6, burial depth of 1,784.35-1,784.38 m, plain light photograph.

Notes: The SEM used is a Philips XL30 and the secondary electron images were taken at 20kV (SY/T 5126-1997 standard)

around detrital grain surfaces; 2) transformation of clay films into chlorite coatings on the surface of detrital grains; 3) the continuous growth of chlorite cement after formation of chlorite coatings. At present, there is not a definitive interpretation of the growth mechanism of chlorite coatings. Jahren's (1991) study showed that the chlorite crystals in sandstone pores were chemically zoned. The shape, size, Si/

Al ratio, and isotopic characteristics of chlorite crystals varied from grain edges to the center of pores. Jahren suggested that the dissolution and recrystallization of chlorite crystals would occur with an increase of burial temperature because of the instability of chlorite crystals formed at low temperature. This could be an important factor in the formation of chemical zonation of chlorite crystals. There are many related reports

about chlorite coatings preserving pores of sandstone reservoirs in Chinese literature (Zhu et al, 2004; Wang et al, 2007; Li et al, 2004; 2007; Zhou et al, 2004; Wang et al, 2005a; 2005b; 2005c; Zhao and Guo, 2006; Guo et al, 2007), but reports about the origin of chlorite coatings are rare.

Fig. 4 Sketch map of the formation process of chlorite coatings (Billault et al, 2003)

The genesis of chlorite coatings in the sandstone reservoirs of the T₃x Group in the Baojie area was studied in detail in this work. We found that irregular clay material existed on the grain surfaces and the bottom of chlorite coatings, which was persuasive evidence of Fe-rich clay films (Fig. 3(e)). Chlorite coatings were not simply from deposition of the minerals from pore water, but were formed by recrystallization of Fe-rich clay films during the syndepositional period. Firstly, large amounts of Fe ions were released by weathering of volcanic detritus brought to the delta area through fluvial transportation. In the last stage of early Permian, large-scale volcanic eruption took place in the southwest China. The study area was located in the transitional zone from the middle to the south of Sichuan Basin, and the sediments mainly came from the south and northeast of the study area where a lot of volcanogenic rocks were distributed (The lake in the Sichuan Basin during late Triassic period was not a freshwater lake but brackish (Huang et al, 2004)). The ferric clay material flocculated and deposited, forming Fe-rich clays in the delta-front area. Detrital grains rolled in the Fe-rich clays under the impact of water flow and the Fe-rich clays became coated on the grain surfaces. The Fe-rich clay films further formed chlorite coatings by recrystallization. The formation process of chlorite coatings is demonstrated in Fig. 4.

7 Factors controlling the formation of chlorite coatings

The formation of chlorite coatings in the study area was controlled by the depositional environment, provenance conditions, and diagenetic environment.

7.1 Depositional environment

The development of chlorite coatings is obviously controlled by the depositional environment. Chlorite coatings mainly developed in sediments in subaqueous distributary channel and river mouth bar delta front facies. Because of the strong hydrodynamic conditions, high content of the coarse rigid detrital grains, and low content of matrix and plastic debris in these regions, a great quantity of primary pores in the sediments were preserved in early diagenesis, which provided a wide space for the growth of chlorite coatings (Huang et al, 2004; Zhou et al, 2004; Wang et al, 2005c). Besides, the ferric ions dissolved in the river could precipitate as clay minerals in delta front environment because of the increase of electrolytes in the lake water and the weak alkaline and weak reducing conditions, which provided abundant ferric ions for the formation of chlorite coatings (Billault et al, 2003). Grigsby (2001) proposed that water level cycles also controlled the vertical distribution of chlorite coatings. During delta progradation, a large amount of detrital materials were discharged and deposited in the delta zone because of strong fluvial action and weak coastal current action. During delta retrogradation, strong coastal currents and tidal action resulted in a wide sediment dispersal, reducing the size of the delta.

7.2 Sediment provenance

The formation of chlorite coatings needs a lot of ferrous ions, while alteration of sediments of volcanic provenance can produce a large amount of ferrous ions (Zeng, 1996; Zhou et al, 2004; Tian et al, 2008; Li et al, 2006). Much diagenesis, such as the alteration of volcanoclastics and biotite hydrolysis generally takes place in reservoir rocks (Fig. 3(f)). Biotite hydrolysis can produce a large number of ferrous (Tian et al, 2008; Li et al, 2006). The statistical analysis of thin section data indicated that the content of volcanoclastics was relatively high in sandstone reservoirs where chlorite coatings develop. Therefore, the development of chlorite coatings is closely related to alteration of fragments of volcanic rocks.

7.3 Diagenetic environment

Chlorite coatings are formed in diagenetic environments. Therefore, diagenetic temperature, lithological characteristics, and pore water medium have a great impact on the formation of chlorite coatings.

1) Diagenetic temperature: the formation of chlorite coatings needs suitable temperature condition. The study of Grigsby (2001) showed that the temperature for the beginning of chlorite growth in sandstones was approximately 20-40 °C by X-ray diffraction and stable isotope analysis. The result of our study indicated that chlorite coatings were formed in the early diagenesis stage. The diagenetic temperature in this

stage is between normal temperature and 65 °C according to the diagenetic stage of clastic sediment classified by Ying (2003).

2) Lithological characteristics: the formation of chlorite coatings needs growing space. Plastic fine sands with a large amount of matrix lost pore space because of compaction, so that it is unfavorable for the growth of chlorite coatings. Moreover, the early calcite cement formed before chlorite coatings, and then the presence of large amounts of calcite cement would block up the pores in sandstones and occupy the growth space required for chlorite coatings (Fig. 3(h)). Statistical analysis of thin section data indicated that the sandstones with chlorite coatings are mainly medium-coarse grained rigid feldspathic litharenite or lithic arkose, and the content of calcite cement, matrix, and plastic debris is relatively low.

3) Pore water medium: the formation of chlorite coatings needs weak alkaline and reducing conditions (Tian et al, 2008; Du et al, 2006). The pH and Eh values in the water of delta front environment could satisfy this kind of condition (Tian et al, 2008). Moreover, the pore water reflects the sedimentary environment in the diagenetic process, which was favorable to the formation of chlorite coatings.

8 Mechanism of chlorite coatings inhibiting quartz overgrowths

Three causes were proposed to explain the absence or scarcity of quartz overgrowths in the sandstones with chlorite coatings: 1) quartz overgrowths did not occur because pressure solution was restrained (Pittman et al, 1992); 2) chlorite coatings on the surface of clastic grains significantly inhibited the growth of secondary quartz crystals (Billault et al, 2003); 3) the surfaces of the clastic quartz grains were isolated from pore water rich in SiO₂ and thus the nucleation of quartz overgrowths was not possible. The syntaxial cement would not be formed if the nucleation of quartz grains now surrounded by chlorite coatings was lost (Ehrenberg, 1993; Pittman et al, 1992; Huang et al, 2004; Bloch et al, 2002; Berger et al, 2009; Pe-Piper and Weir-Murphy, 2008). Thin section and SEM analysis showed that quartz overgrowths were rarely found on the quartz grains surrounded by chlorite coatings and the contact way between the detrital grains was mainly dot-line which indicated that pressure solution was weak (Figs. 3(a) and (b)), but if the chlorite coatings around quartz grains were discontinuous, secondary quartz would nucleate and eventually form quartz overgrowths in the place where detrital quartz surfaces were not isolated from the pore fluids by chlorite coatings (Fig. 3(i)). A large number of primary pores could therefore be preserved by chlorite coatings inhibiting quartz overgrowths.

9 Conclusions

1) Chlorite cement is mainly present as grain coatings. The chlorite coatings on the wall of the pore spaces orient perpendicularly to grain surfaces, while the chlorite coatings at the contact between adjacent detrital grains are arranged with a preferred orientation tangential to the surface of

detrital grains.

2) Chlorite coatings were formed in the eogenetic stage. Chlorite crystals would be recrystallized after chlorite coatings were formed and the formation continued through the whole mechanical compaction.

3) Chlorite coatings were formed by recrystallization of Fe-rich clay films during the syndepositional period.

4) The formation of chlorite coatings was controlled by the depositional environment, provenance conditions, and diagenetic environment.

5) Chlorite coatings inhibited quartz overgrowths by restraining pressure solution, the growth of secondary quartz crystals, and the nucleation of quartz on the surface of grains.

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