1	1	Source of quartz cement in tight gas sandstone: Evidence from the Upper
2 3	2	Triassic Xujiahe Formation in the western Sichuan Basin, SW China
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Abstract: Quartz cement is a significant authigenic mineral in the tight gas sandstones of the Upper Triassic Xujiahe Formation, fourth member (Xu4 sandstones) in the western Sichuan Basin. Here we report mineralogical, petrological and geochemical data from the Xu4 sandstones to constrain the silica sources of their quartz cements. The quartz cements mainly occur as quartz grain overgrowths and pore-filling quartz cement, with major associated authigenic minerals including calcite, dolomite, illite, kaolinite, and chlorite. The homogenization temperatures of quartz cement fluid inclusions range from 49.2°C to 143.4°C and show a multistage continuous precipitation process. Raman spectrometry results show the presence of methane (CH_4) and CO_2 in quartz cement fluid inclusions. The dissolution of feldspar, the illitization of kaolinite and smectite, as well as the pressure dissolution of quartz grains provided silica sources for quartz cement. The dissolution of feldspar by organic acid (carboxylic acid) is the main silica source for quartz cement in the 80~120°C interval; the illitization of smectite and kaolinite have different initial temperatures, and both consume potassium. The pressure dissolution of quartz grains results in stylolite formation. The low-temperature quartz cement ($<70^{\circ}$ C) is mainly derived from the dissolution of feldspar by meteoric water. At the Late Triassic/Jurassic transition, the uplift of the western basin led to leaching by meteoric water in the upper part of the Xu4 sandstone, forming a weakly acidic fluid with a low K⁺/H⁺ ratio and giving this type of sandstone low potassium and sodium concentrations. Keywords: Quartz cement, silica source, meteoric water, tight gas sandstone, Xujiahe Formation, Sichuan Basin.

32 1. Introduction

Quartz is a major component of authigenic cement in deeply buried (>3.0 km) siliciclastic hydrocarbon reservoirs (Worden and Morad, 2000; French and Worden, 2013). The formation and growth of quartz cement during diagenesis are capable of affecting the reservoir quality either positively or negatively (Giles and de Boer, 1990; Worden and Morad, 2000; Zhang et al., 2012; Götte et al., 2013; Xi et al., 2015). Authigenic quartz overgrowth, pore-filling and fracture-filling quartz cements generally reduce reservoir porosity because they occupy pore spaces and narrow pore throats (Zhang et al., 2012; Bjørlykke, 2014; Xi et al., 2015). In contrast, some scholars claim that authigenic quartz in the form of grain-coating microcrystalline quartz can assist in the preservation of primary pores by inhibiting the growth of quartz overgrowth (Vagle et al., 1994; Ramm et al., 1997; French et al., 2012; French and Worden, 2013). Therefore, accurately understanding the generation and evolution of quartz cement is vital for reservoir characterization and evaluation and for de-risking hydrocarbon resource exploration.

A full understanding of quartz cementation processes in siliciclastic rocks requires identifying the potential silica sources, such as internal sources within sand bodies, or external sources in adjacent shale and mudstone units (Bjørlykke and Egeberg, 1993; Vagle et al., 1994; Walderhaug, 1994; Rezaee and Tingate, 1997; Worden and Morad, 2000; Kim and Lee, 2004; Xi et al., 2015; Lai et al., 2018b), and determining the factors which control quartz cementation during diagenetic processes such as temperature, pressure and paleo-fluids flow properties (Walderhaug, 1994; Worden and Morad, 2000; Pollington et al., 2011; Bjørlykke., 2014; Hyodo et al., 2014). From a broader perspective, understanding the silica source, formation of quartz cement and evolution of paleo-fluids flow can provide important clues for understanding the diagenesis and reservoir quality of hydrocarbon reservoirs (Kim and Lee, 2004; Hyodo et al., 2014; Xi et al., 2015).

This study investigates cementation in a tight sandstone reservoir in the western Sichuan Basin within the fourth member of the Upper Triassic Xujiahe Formation (T_3x^4 ; Xu4 sandstone) at a depth interval of 3000-4500 m, which is one of the main reservoir intervals in the western Sichuan Basin characterized by low ultralow porosity and permeability. Previous studies on the depositional environment and diagenesis of the Xu4 sandstone emphasized the role of carbonate cementation in controlling reservoir quality (Liu et al., 2014; Li et al., 2019; Yu et al., 2019). However, the quartz cement, another significant authigenic mineral in the Xu4 sandstone reservoir, has been less well constrained and few studies ever discussed its source or formation mechanism. Here we report detailed mineralogical, petrological and geochemical characteristics of quartz cement in the Xu4 tight gas sandstone by integrating quantitative mineralogy, fluid inclusions, and in situ geochemistry and isotope techniques, to constrain the possible silica source of the quartz cement.

2. Geological setting

The Sichuan Basin is one of the most important hydrocarbon producing basins in China (Fig. 1; Hao et al., 2008; Lai et al., 2018a; Yu et al., 2019). The metamorphic and granitic crystalline basement of the Sichuan Basin formed during the Proterozoic, and several tectonic movements then occurred in the basin (Ma et al., 2007; Hao et al., 2008). Since the Late Triassic, the tectonic evolution of the Sichuan Basin has been influenced successively by the Indosinian, Yanshanian and Himalayan orogenies (Fig. 2A; Fig. 3). The Indosinian orogeny resulted in a brief uplift of the Sichuan Basin and eroded the strata on the top of the Xujiahe Formation in the western part of the basin, forming a disconformity between the Triassic and Jurassic formations (Fig. 2B; Fig. 3; Ma et al., 2007). The Yanshanian orogeny resulted in folding of the basin border areas during the Jurassic; the Himalayan orogeny has been uplifting the entire basin since the Cretaceous (Fig. 2B; Fig. 3; Ma et al., 2007).

The study area is located in the middle of the western Sichuan Basin and is bordered by the Longmenshan thrust belt to the west (Fig. 1B; Fig. 1C). The western Sichuan Basin can be divided into five secondary structure zones (Fig. 1B), and several medium to large gas fields have been discovered in the area (Fig. 1B). The Triassic Xujiahe Formation is siliciclastic rocks dominated by sediment deposited in braided river, deltaic and shallow lacustrine environments. It comprises five members: from bottom to top the 1st to 5th members (Fig. 1C; Fig. 2B), with the main hydrocarbon reservoir intervals being the Xu2 and Xu4 members, whereas the lithology of Xu1 member, Xu3 member, and Xu5 member dominated by fine-grained sediments, including shale, mudstone, coal, and siltstone (Fig. 1C; Fig. 2B), constituted the main hydrocarbon source rocks (Zhang et al., 2016; Yu et al., 2019). The Xu4 member consists of gray sandstone interbedded with

thinly-bedded dark siltstone and mudstone. The occurrence of conglomeratic sediments at the base of the Xu4 member indicate high energy, alluvial fan environment (Luo and Tong, 1989; Lin et al., 2007; Xu et al., 2015). The burial depth of the Xu4 member is approximately 3.0 km to 4.0 km (Fig. 1C; Fig. 3).

3. Methods and database

3.1 Petrology and petrophysics

The Xu4 sandstones analyzed here were sampled from 17 wells (Fig. 1B). Thin sections of the Xu4 sandstone samples, as well as relevant porosity, permeability and burial history data (Fig. 3) were collected from the Sinopec Southwest Oil & Gas Company. Fresh core plugs (2.5 cm diameter) were analyzed using 3020-62 helium porosity analyzer and GDS-9F gas permeability analyzer.

A total of 191 thin sections were selected for mineralogical modal analysis using point-counting techniques under polarizing microscope. At least 300 grains were counted on each thin section. The thin sections were also impregnated with blue epoxy to examine the pore system.

Sixty-one samples and 39 samples were analyzed for whole-rock and clay fraction mineralogy, respectively, by X-ray diffraction (XRD) using a PANalyticl X'pert pro at the Keyuan Engineering Testing Center (Sichuan), to determine the type and content of minerals. For the XRD analysis of whole-rock, 10 g of samples were crushed to 200 mesh, then put the sample powder into the sample mold of the instrument for analyzed. To separate the clay fraction, 2% HCl and dilute H₂O₂ were used to remove carbonate minerals and organic matter, respectively. The clay fraction with particle size less than 2 µm were extracted by centrifugation. After air-dried, the sample mounts were treated with ethylene glycol steam at a constant temperature of 60° C for no less than 8 hours of saturation treatment, and then heated at 550°C for 2 hours. The XRD analysis was performed with CuK α radiation at 40 kV and 40 mA, and a step size of 0.02° 20/s, ranging from 5 to 55°. Fifteen samples were analyzed by a Quanta FEG 250 field emission scanning electron microscope (FE-SEM) with an energy dispersive spectrometer (EDS) to obtain the element compositions of minerals.

3.2 Fluid inclusion and laser Raman analyses

Fluid inclusion microthermometry was performed on quartz cements to determine the temperature at which the quartz cements formed. Fluid inclusion analysis was conducted on 14 doubly polished sections (0.1 mm thick), and the homogenization temperature ($T_h/^{\circ}C$) and final ice-melting temperatures (Tm_{ice}) of the fluid inclusions of quartz cements were determined with a Linkam THMSG-600 heating and freezing stage, the T_h accuracy is $\pm 1^{\circ}$ C. The heating rate was 1°C~5°C/min, and decreased to 0.5°C~1°C/min when it was close to the phase transition point for the initial stages of each heating operation. The fluid inclusions were cooled to the -100°C and then the temperature was increased at a heating rate no more than 0.5°C/min. The salinity of the fluid inclusions was calculated from the measured Tm_{ice} value following ice points of H₂O-NaCl (Bodnar, 1993). Raman spectroscopy analyses of 25 fluid inclusions from 14 sections were performed with a Renishaw inVia Reflex at a laser wavelength of 514 nm and a maximum power of 20 mW. The exposure time was superimposed once every 20 seconds to obtain Raman peaks and identify the composition of the phases. The relative molar fractions (mol%) of gaseous components can be calculated using the following equation (Dubessy et al., 1989; Burke, 2001; Frezzotti et al., 2012):

$$X_a = \frac{A_a/(\sigma_a\xi_a)}{\sum A_i/(\sigma_i\xi_i)}$$

136 where X_a is the molar fraction, A_a is the peak area, σ_a is the Raman cross-section (514 nm), and ξ_a 137 is the instrumental efficiency. And A_i , σ_i , ξ_i represents the sum values of all detected gaseous 138 components in the inclusion. Peak position and peak area were obtained by Gaussian fitting of the 139 Raman spectrum. Note that the Raman spectrum of CO₂ has two peaks and the sum of the two 140 peaks should be taken (Dubessy et al., 1989). The fluid inclusion and Raman spectroscopy 141 analyses were performed at the State Key Laboratory of Oil and Gas Reservoir Geology and 142 Exploitation, Chengdu University of Technology.

3.3 Quantitative mineralogical analysis

Eleven samples were selected for automated quantitative mineralogy studies utilizing the Tescan integrated mineral analyser (TIMA) platform at the John de Laeter Centre (JdLC) at Curtin University, Western Australia. The TIMA integrates a high-resolution field emission scanning electron microscope (SEM) with four silicon drift energy dispersive spectroscopy (EDS) detectors, allowing ultrafast measurements of mineral associations, concentrations and grain sizes, as well as element distributions on multiple samples via SEM-EDS analysis. The Xu4 sandstone samples were cut into mounts of 25 mm in diameter and 10~12 mm in height, well-polished, and conductively coated with a 5~10 nm-thick carbon coat. Samples were analyzed using the acquisition mode of Dot Mapping with a 3.0 µm pixel size. The raw data were processed using the TIMA Software v1.5.47.

3.4 SIMS analyses

157 Detailed delineation of thin sections under polarizing microscope ensured the accuracy of 158 targeting. Two of the samples for fluid inclusion studies were selected for secondary ion mass 159 spectrometry (SIMS) analysis with a CAMECAIMS-1280 ion microprobe at the Institute of 160 Geology and Geophysics, Chinese Academy of Sciences, to obtain the *in situ* oxygen isotopes of 161 quartz cements. The reflected and BSE images were used to select appropriate spots for analysis. 162 The beam spot diameter was approximately 20 μ m and the precision of ¹⁸O/¹⁶O ratios is ca 0.2 ‰ 163 (2SE; Li et al., 2012), and other specific analytical details were reported in Li et al. (2013).

165 4. Results

4.1 Sandstone composition

167 The point counting of 191 sandstone samples reveals that the framework grains of the Xu4 168 sandstones are mainly lithic fragments, quartz, feldspar, and other minor minerals. On the 169 sandstone classification diagram (Folk et al., 1968; Fig. 4A), the samples plot in the fields of 170 sublitharenite, litharenite and feldspathic litharenite, with an average framework composition of 171 $Q_{68}F_{5}L_{27}$. The quartz, feldspar and lithic fragments contents ranging from 24 to 89%, 0 to 20%, 172 and 8 to 74%, respectively. The main components of the lithic fragments are sedimentary and 173 metamorphic rock fragments. The automated SEM-EDS data from TIMA indicate that quartz is the most abundant mineral in most sandstones (Fig. 4B, C and Appendix 1), with contents ranging from 29 to 86% (average of 69.1%). Other major minerals reported include calcite (9.7%) and muscovite (5.7%). Minor albite, dolomite, illite, orthoclase and ankerite are also observed (Fig. 4D). Such results are close to those from standard light microscopy petrographic methods. In some litharenites with high rock fragment contents (Fig. 4D), calcite and dolomite lithic grains can account for a higher proportion than quartz. The XRD data of whole-rock (Appendix 2) show that the quartz content ranges from 9.1% to 87.5%, with an average of 63.3%, followed by clay minerals, calcite, dolomite, and plagioclase, with content ranging from 2.1% to 37.1% (average 15.3%), 0 to 56.7% (average 9.6%), 0 to 49.5% (average 7.8%), and 0 to 13% (average 3.1%), respectively. Other minerals, including K-feldspar, siderite, and pyrite, with an average content of less than 1%. The XRD results show minor differences form the mineral contents obtained by SEM-EDS of TIMA.

4.2 Diagenetic mineral analysis

4.2.1 Quartz cement

The microtexureal characteristics of quartz cements in the Xu4 sandstone were investigated by both standard light microscopy petrographic methods and SEM analyses (Fig. 5). Authigenic quartz occurs as overgrowths on detrital grain and as pore-filling cement. In general, quartz overgrowths occur where quartz grains are relatively abundant, and making quartz grains are in concave-convex contact even in stylolite formation (Fig. 5A, B, C, D). The quartz overgrowths exhibiting the same optical orientation as the attached quartz grain (Fig. 5B, D). Most quartz overgrowths can be distinguished by dust rims between detrital quartz grains (Fig. 5A, B, C, D), which are $20 - 50 \mu m$ in thickness and occur along one side or both sides of quartz grains. The morphology of quartz overgrowths varies with the effective space in their growth direction, and partially fills the intergranular pores. Vertical broken sections of quartz overgrowths display clear crystal surfaces (Fig. 5E, F). Pore-filling quartz generally occurs as isolated particles with diameters of 10~100 µm and fills in the primary or secondary pore space of the sandstone (Fig. 5G, H, I). Quartz cement fills in the intragranular pores and can coexist with clay minerals, such as kaolinite (Fig.5H) and illite (Fig.5I).

4.2.2 Carbonate cement

Carbonate minerals also occur in the Xu4 sandstone in frequent association with quartz cement, including calcite (Fig. 5C, D; Fig. 6A, B, C, D) and dolomite (Fig. 6B). In some sandstones, extensive early calcite cement can occupy primary pores and occupy more than 20% of the whole rock volume (Fig. 6A; Fig. 7A), and late calcite and dolomite cements fill primary and secondary pores. The carbonate cement content shows a negative correlation with quartz cement (Fig. 7B), especially in samples with carbonate cement exceeding 10% while quartz cement rare, indicating that the occurrence of carbonate cement can inhibit the formation of quartz cement.

213 4.2.3 Clay mineral cement

Kaolinite (Fig. 5H, Fig. 6B, E), illite (Fig. 5I; Fig. 6E, F, G, H) and chlorite (Fig. 6I) can be observed in the Xu4 sandstone associated with quartz cement. Kaolinite occurs in books or vermicular stacks and is rich in intercrystalline pores (Fig. 6B). Illite occurs as fibers (Fig. 6E, G, H) and filiform variants (Fig. 6F), and is mainly attached to the surface of quartz cement, indicating that illite formed later than authigenic quartz. Chlorite occurs as scaly and vertical growths on the grains (Fig. 6I). The XRD results (Fig. 8) show that the illite component of the clay mineral fraction ranges from 12% to 78%, with an average content of 51.3%. Other clay minerals occurring in lesser abundance include chlorite (0-55%; average 20.95%), kaolinite (0-88%; average 15.47%) and illite/smectite mixed-layer clays (I/S; 0-35%; average 12.44%).

4.3 Fluid inclusions in quartz cement

Fluid inclusions (2-8 μ m diameter) scattered throughout the quartz cements are two-phase (v-l), liquid-rich (90-95%) and irregular to elliptical in shape (Fig. 9A, B). Homogenisation temperatures (T_h) range from 49.2°C to 143.4°C (Table 1) with a homogenisation peak in the 80-100°C interval (Fig. 9C). The wide range in T_h may indicate that the authigenic quartz formed during a thermally dynamic process. The salinities range from 0.18 to 14.15 wt.% NaCl eq. and shows no correlation with T_h (Fig. 9D). The Raman spectroscopy results show that the vapour is dominated by methane (CH₄) with minor CO₂, ranging from 0 to 77.4% (Table 2; Fig. 10). The liquid composition is dominated by H₂O. The statistical results show that the T_h distribution range of inclusions containing CH₄ is 65.8~130.8°C (Table 2; Fig. 10A); the T_h distribution interval of inclusions containing CH₄ and CO₂ is 80~100°C (Fig. 10A).

236 4.4 In situ oxygen isotopes of quartz cement

237 In situ oxygen isotopes of the quartz cement samples from the fluid inclusions were determined by 238 SIMS analysis (Fig. 11 and Appendix 2). The $\delta^{18}O_{SMOW}$ (SMOW: standard mean ocean water) 239 values of detrital quartz grains range from 8.44~10.12‰ with an average of 9.42‰. Compared 240 with detrital quartz, quartz cement has higher $\delta^{18}O$ values ranging from 12.08 to 18.21‰ (Fig. 241 11G).

243 5. Discussion

Internal silica sources of sandstone include dissolution of detrital feldspar, biogenic and volcanic
grains; transformation of smectite to illite and chlorite; and pressure dissolution of quartz grains
(Bjørlykke and Egeberg, 1993; Vagle et al., 1994; Walderhaug, 1994; Rezaee and Tingate, 1997;
Worden and Morad, 2000; Kim and Lee, 2004; Xi et al, 2015). In contrast, the silica-rich flow
from adjacent mudstones or several deeply buried rocks is regarded as the external source for
sandstones (Worden and Morad, 2000).

250 5.1 Organic acid dissolution of feldspar

Based on the thin section and SEM observations, secondary intragranular dissolution pores are
primarily the dissolution products of feldspar minerals (Fig. 5A, G, H), and silica is one of the
dissolution products of feldspar (Fig. 5H, I).

254 Mineralogical observations show that quartz and illite precipitated next to the residual albite in
255 the original grain (Fig. 12). This process can be well interpreted by the reaction of albite or
256 K-feldspar with H⁺ from the fluid (Giles and de Boer, 1990; Bjørlykke and Jahren, 2012):

257
$$2NaAlSi_{3}O_{8} + 2H^{+} + H_{2}O \rightarrow Al_{2}Si_{2}O_{5}(OH)_{4} + 4SiO_{2} + 2Na^{+}$$
Albite Kaolinite (R1)

258
$$\frac{2KAlSi_{3}O_{8} + 2H^{+} + H_{2}O \rightarrow Al_{2}Si_{2}O_{5}(OH)_{4} + 4SiO_{2} + 2K^{+}}{\text{K-feldspar}}$$
(R2)
K-feldspar Kaolinite

Previous studies have shown that H⁺ in fluid has inorganic and organic sources; the inorganic source is meteoric water, and the organic source is organic acid, which is related to the maturity of the organic matter during the late stage of eodiagenesis or mesodiagenesis (Surdam and Crossey, 1987; Bevan et al., 1989; Bjørlykke et al., 1992; Ehrenberg et al., 2001; Seewald, 2003; Yuan et al., 2019). When temperature reaches 70~80°C, the process of organic matter maturation leads to the formation of organic acid (carboxylic acid) and CO₂ (dissolves in the water to produce carbonic acid); whereas when temperature reaches over 120°C, the concentration of organic acid in the fluid decreases due to thermal decarboxylation, which generates CO₂ and CH₄ (Surdam and Crossey, 1987; Ehrenberg et al., 2001; Ong et al., 2013; Yuan et al., 2019). In the temperature range of 80°C to 120°C, the concentration of organic acids in the fluid is the highest, which is consistent with the peak temperature distribution (80~100°C, Fig. 9C) of the quartz cement in the Xu4 sandstones. Moreover, the temperature peak of the fluid inclusions containing CO₂ also lies within this interval (80~120°C Fig. 10A). Therefore, combined with the mineralogical observations (Fig. 5H, I, Fig. 12), it is shown that feldspar dissolution could account for the source of silica in quartz cement in the 80~120°C range.

However, it should be noted that according to formula R1 and R2, the direct product of feldspar
dissolution should be kaolinite rather than the illite as observed here. This result is not unusual
because when the temperature rises to a certain range (generally 110~130°C; Berger et al., 1997;
Bjørlykke, 2014) and the K⁺/H⁺ ratio in the fluid increases, kaolinite is unstable and forms illite in
a close diagenetic system or reacts with K⁺ in the diagenetic fluid in an open system (Berger et al., 1997), as R3.

280
$$Al_{2}Si_{2}O_{5}(OH)_{4} + KAlSi_{3}O_{8} \rightarrow KAl_{3}Si_{3}O_{10}(OH)_{2} + 2SiO_{2} + H_{2}O$$
kaolinite K-feldspar illite
(R3)

282 5.2 Illitization of kaolinite and smectite

Illitization of Kaolinite as formula R3 is a potassium-consuming reaction that accelerates the
dissolution rate of K-feldspar in sandstones. The XRD results show that the content of kaolinite
decreases with the increase of burial depth (Fig. 8C), especially when the depth exceeds 3800 m,
the content of kaolinite decreases sharply (Fig. 8C), making 3800 m the maximum depth at which
kaolinite can exist in large quantities in the study area. The temperature reached approximately

110°C~130°C when kaolinite transformed into illite (Bjørlykke, 2014), which was during the middle stage of mesodiagenesis, and the porosity of sandstone decreased, which resulted in a relatively closed diagenetic system; thus, quartz cement and illite precipitated near the original feldspar grains (Fig. 12). The relative contents of illite show a positive correlation with depth (Fig. 8A); moreover, the SEM image (Fig. 6E) shows that the conversion of kaolinite to illite is further evidence to support this reaction. Because of the relatively high reaction temperature, the illitization of kaolinite is one of the silica sources of quartz cement with precipitation temperature above 110°C.

The illitization of smectite is common in siliciclastic rocks (Worden and Morad, 2000), and research based on Gibbs free energy (ΔG) calculations has confirmed that this is a low-energy reaction (Berger et al., 1997). Unlike the illitization of kaolinite, which can directly precipitate pure illite, the conversion of smectite to illite forms an intermediate product: I/S; the I/S mixed layers are usually disordered during eodiagenesis, while the degree of I/S disorder decreases and the content of illite increases during burial; these changes are often used as diagenetic stage indicators in clastic rocks (Worden and Morad, 2000; Worde and Morad, 2009). The composition of smectite is complex, and the reaction for one type of smectite transformation into illite can be simplified as:

 $8Al^{3+} + 4.5K^{+} + smectite \rightarrow illite + Na^{+} + 2Ca^{2+} + 2Mg^{2+} + 2.5Fe^{3+} + 3Si^{4+}$ (R3)

The initial temperature of this reaction is approximately 70°C (Peltonen et al., 2009; Thyberg et al., 2010; Xi et al., 2015). Similar to the illitization of kaolinite, this is also a potassium-consuming reaction; Ca, Mg and Fe are discharged while silica is generated. The relative content of I/S shows a negative correlation with the depth (Fig. 8D); the higher the temperature is, the lower the content of the I/S mixed layer and the higher the illite content in the Xu4 sandstone, implying that the illitization of smectite may be another silica source for the sandstones. The conversion of smectite to illite stops when the I/S mixed layer is completely converted into illite or there is no K^+ or Al^{3+} in the diagenetic system. The illitization of smectite could cause dehydration and the release water may result in the decrease of fluid salinity (Colten-Bradley, 1987; Hüpers and Kopf, 2012; Tremosa et al., 2020), however, no correlation is observed in Fig. 9D suggesting that no clearly evidence of the smectite dehydration.

The illitization of smectite in the adjacent mudstone layers is the external silica source for the Xu4 sandstones. Previous studies have documented that when limited by fluid rates and by the solubility of silica and Al³⁺, silica cannot be transported massive quantities into the sandstone; thus, the silica source must be derived from the sandstone reservoir itself or from adjacent mudstones (Giles et al., 2000; Bjørlykke and Jahren, 2012).

The illitization of kaolinite or smectite is a reaction of potassium consumption, which can lead to the much lower content of K-feldspar than the plagioclase subgroup (i.e., albite-anorthite series) in the present Xu4 sandstones. The XRD results and mineral phase images obtained by TIMA show that the content of K-feldspar is generally less than that of plagioclase (Fig. 13), with K-feldspar absent in more than half of the samples (Fig. 13A). Although the absent of K-feldspar may be related to the provenance of the Xu4 sandstone, considering the widespread existence of illite, it is proved that a large number of K⁺ exist in the diagenetic system and the source can only be provided by the dissolution of K-feldspar.

331 5.3 Pressure dissolution of quartz grains

Under certain conditions of pressure and temperature, pressure dissolution occurs at the contact points of quartz grains with dissolved silica forming quartz cement elsewhere in the sample (Worden and Morad, 2000). Silica solubility, dissolution, transport and precipitation rates increase with temperature (Bjørkum, 1996; Renard et al., 2000). In addition, pressure influences the relative silica solubility between the grain contacts and the kinetics of the contact grain surfaces (Renard et al., 2000). The pressure dissolution of detrital quartz grains can provide a silica source for a portion of the high-temperature quartz cement in the Xu4 sandstones. Evidence of this process can be detected from the optical observations, quartz cements, especially quartz overgrowths, tend to occur in areas where quartz grains are in stylolite formation or concave-convex contact with other quartz grains (Fig. 5A, B, C, D). In contrast, the occurrence of quartz cement is rarely where the quartz grains in contact with ductile rock fragments (including mudstone, mica, mud intraclast) because of their weak compaction resistance. In some samples with relatively numerous quartz grains, pressure dissolution may be the significant silica source.

5.4 Meteoric water dissolution of feldspar

The acid dissolution of feldspar, the illitization of smectite and kaolinite, and the pressure dissolution of quartz grains can all provide silica sources for quartz cement in the Xu4 sandstones. However, these reactions require relatively high temperatures; even the illitization of smectite which requires the lowest temperature, cannot begin until the temperature reaches 70°C. Therefore, other mechanisms may be required to explain the precipitation of early quartz cement between 49.2°C and 70°C (Fig. 9C). Previous study has shown that there are quartz cement inclusions with a minimum T_h between 50°C and 60°C in the Xu2 and Xu4 sandstones (Luo et al., 2019). One possible interpretation is that the early quartz cement with low precipitation temperature could be a byproduct of the dissolution of feldspar by meteoric water. Because CO₂ dissolves in the water, forming carbonic acid and releasing H⁺, meteoric water is weakly acidic and can lead to the dissolution of feldspar (França et al., 2003; Bjørlykke and Jahren, 2012; Yuan et al., 2019), according to reaction R1. In an open diagenetic system, products such as silica, sodium or potassium (depending on the type of feldspar minerals dissolved), can be removed with the diagenetic fluid and make this reaction proceed further (Bjørlykke and Jahren, 2012).

The oxygen isotope of authigenic quartz can provide information about the nature of the fluids precipitating the quartz (Harwood et al., 2013; Hyodo et al., 2014; Yuan et al., 2017). The development of high-precision in situ analyses of isotopes by SIMS has made it possible to obtain the oxygen isotopes of micron-sized quartz cement, and such analysis has been successfully utilized in many works (Rezaee and Tingate, 1997; Chen et al., 2001; Pollington et al., 2011; Hyodo et al., 2014). Clayton et al. (1972) domonstrated that the δ^{18} O composition of quartz $(\delta^{18}O_{\text{quartz}})$ is a function of temperature and the $\delta^{18}O$ composition of the fluid which it precipitated ($\delta^{18}O_{fluid}$). Based on the measured fluid inclusion temperatures and $\delta^{18}O_{quartz}$ of quartz cements from well Y3 (3133.18 m) and XC27 (4023 m), the $\delta^{18}O_{\text{fluid}}$ can be to range from -12.1‰ to -5.4‰ (Fig. 14), showing that $\delta^{18}O_{\text{fluid}}$ value increases with the increase of precipitation temperature. This result supports the interpretation that the Xu4 sandstones were affected by meteoric water during early diagenesis and that the leaching by meteoric water caused $\delta^{18}O_{\text{fluid}}$ to be lower than -10%. Due to the influence of meteoric water leaching, early quartz cement with lower precipitation temperatures has low $\delta^{18}O_{\text{fluid}}$ values.

Previous studies have shown that leaching by meteoric water can affect strata with a burial depth of approximately 2000m (Giles and de Boer, 1990; Yuan et al., 2017). The Sichuan Basin experienced a major movement during transition from the Late Triassicto the Jurassic, resulting in the uplift of the western basin (Fig. 3; Ma et al., 2007). During this process, meteoric water seeped into the surface, and entered and affected the upper part of the Xu4 member at burial depths less than 1500 m at this time. Diagenetic fluid mixed with meteoric water to form a weakly acidic fluid with a low K⁺/H⁺ ratio, which resulted in dissolution of feldspar and precipitation of kaolinite and early quartz cement, as shown in R1. Because the low K^+/H^+ ratio is one of the reaction conditions, the dissolution of K-feldspar would increase the K⁺/H⁺ ratio of the fluid; however, in this case, the dissolution of feldspar by meteoric water tended to dissolve plagioclase. In addition, the sandstone had not experienced strong diagenesis, and there was enough porosity to ensure that the diagenetic system remained relatively open. Hence, if there were a sufficient volume of diagenetic fluid with H⁺, the feldspar in sandstone would have dissolved until it completely disappeared, and kaolinite would have precipitated. Some silica, K⁺, and Na⁺ would have flowed out with the fluid, giving this type of sandstone extremely low potassium and sodium contents. During this process, the content of Al remained relatively stable, so the ratio of Al/(K+Na) in sandstones can be used to indicate the influence of meteoric water on the leaching of sandstone (Fig. 15). However, in most of the sandstones, leaching by meteoric water is not obvious because meteoric water mainly affected the sandstones in the upper part of the Xu4 unit, and the degree of influence decreases with increasing burial depth.

6 5.5 Amorphous silica

397 Amorphous silica mainly consists of biogenic and volcanic origins (Worden and Morad, 2000).
398 Biogenic silica has been suggested to be one of the silica sources of quartz cement (Vagle et al.,
399 1994; Hendry and Trewin, 1995; Weibel et al., 2010). However, there is no evidence for biogenic
400 silica (such as siliceous sponge spicules and diatoms) exist in the Xu4 member which was
401 deposited under the lacustrine delta environment. Another type of amorphous silica, volcanic
402 origin materials, could provide silica sources through dissolution or devitrification (Worden and
403 Morad, 2000). Although volcanic materials are rare in the present Xu4 sandstone, the alteration of

volcanic materials occurred during the eodiagenesis (Worden and Morad, 2000; Huang et al.,
2007), and the related minerals have already disappeared. In addition, there was volcanic activity
in the western Sichuan Basin during the same period (Ran et al., 2016). Therefore, it is possible
that amorphous silica of volcanic origin provided silica source for the Xu4 sandstone, but no direct
evidence to support and further research is needed.

410 5.6 Synthetic model for silica sources

Based on the petrologic characteristics and the textural relationships of minerals and fluid inclusions, the diagenetic sequence of the Xu4 sandstones can be constructed (Fig. 16) and can provide useful information for discussion on the silica sources. The precipitation history of quartz cement in the Xu4 sandstones spanned the eodiagenetic and mesodiagenetic processes (Fig. 16). The dissolution of feldspar by meteoric water occurred during eodiagenesis, forming the earliest quartz cement and kaolinite. The presence of CH₄ in the fluid inclusions indicates that the hydrocarbon migrated into reservoir. At the end of eodiagenesis, when the temperature reached approximately 70°C~80°C, the illitization of smectite began, and illite and quartz cement precipitated. Next, organic acid and CO₂ entered the fluid, dissolved the feldspar, and formed kaolinite and quartz. These two reactions started almost simultaneously; the dual silica sources caused a large amount of quartz cement to precipitate within the 80°C~120°C interval (Fig. 9C). Pressure dissolution proceeded at a sufficiently high temperature, and the silica dissolved at the quartz grain contacts, forming stylolites between quartz grains (Worden and Morad, 2000). When the temperature reached 110°C~130°C, kaolinite could react with K-feldspar and form illite and quartz cement.

427 6. Conclusions

428 Authigenic quartz is a significant cement in the Xu4 tight gas sandstone and affects the reservoir 429 quality. In this study, integrating mineralogical, petrological, and geochemistry techniques to 430 constrain the source of quartz cement in the Xu4 sandstone, the principal conclusions are as 431 follows:

432 a) The combination of meteoric water and organic acid dissolution of feldspar, illitization of clay

minerals (kaolinite and smectite), and pressure dissolution of quartz grains provided the silica
source for the Xu4 sandstone. Among them, meteoric water dissolution of feldspar constituted
the main source of low-temperature (<70°C) quartz cement, silica sources for quartz cement
with temperatures greater than 70°C were provided by the other three. Oxygen isotope values
obtained by SIMS confirm that the presence of meteoric water in the diagenetic fluid during
the eodiagenesis in the sandstone, which was associated with the tectonic movement during
the transition from the Late Triassic to the Jurassic.

b) Whether the illitization of clay minerals or feldspar dissolution, both are potassium
consumption reactions, the difference is that illitization of kaolinite and smectite and feldspar
dissolution by organic acid cause a reduction in the K-feldspar content, the K⁺ remains in the
diagenetic system. The meteoric water dissolution of feldspar causes the K⁺ or Na⁺ to be
discharged with the diagenetic fluid, resulting in extremely low K⁺ or Na⁺ contents of the
sandstone.

c) The results of this study demonstrate that quantitative mineralogy analysis technique (not
limited to the TIMA used herein) has great potential for the study of diagenesis, although the
accuracy of the data obtained on mineral content and element concentrations are not
particularly good, it can effectively reflect the trend of variation among samples and visualize
diagenetic appearance.

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658 Figure captions

Fig. 1. (A) The location and geological map of the Sichuan Basin; (B) Location map of gas fields
and wells in the western Sichuan Basin; (C) Northwest southeast cross section along A-A' in (B)
illustrating the Upper Triassic strata and tectonic structure characteristics of the study area
(modified from Yang and Zhu, 2013).

Fig. 2. Generalized stratigraphic columns of the Mesozoic (A) and the Xujiahe Formation (B) inthe Sichuan Basin.

Fig. 3. Typical burial history of the Xu4 member in the Xinchang gas field (orange line) and
Fengguchang gas field (blue line) in the western Sichuan Basin (modified from Leng et al. (2011)
by simulation using vitrinite reflectance data).

Fig. 4. (A) Classification of the Xu4 sandstones according to Folk's scheme (Folk et al., 1968); (B)
Mineral content of sandstones by TIMA SEM-EDS analyses; image of mineral phases by TIMA
analyses; (C) Image of TIMA mineral phases in sample from CM 39 well, 3106.8 m; (D) Image of
TIMA mineral phases in sample from CH 139 well, 3781.79 m.

Fig. 5. Images of petrographic features of quartz cement in the Xu4 sandstones. (A) Plane-polarized light and (B) cross-polarized light photomicrographs of quartz overgrowth and secondary intragranular pore, CF563 well, 3912.71 m; (C) Plane-polarized light and (D) cross-polarized light photomicrographs of quartz overgrowth and carbonate cement, GM3, 3784.2 m; (E) Quartz overgrowth with clear crystal surface, CF563 well, 3913.6 m; (F) Quartz overgrowth with iilite adhere to the surface of quartz, MS1 well, 4584.5 m; (G) Pore-filling quartz cement in primary and secondary pores, HL1 well, 3472.92 m, plane-polarized light; (H) Pore-filling quartz cement and kaolinite in intragranular pores, CX568 well, 3408.59 m, plane-polarized light; and (I) Pore-filling quartz cement and illite in intergranular pores, CF563, 3882.7 m, SEM image. Q: quartz grain; QO: quartz overgrowth; PFQ: pore-filling quartz cement; C: carbonate cement; K: kaolinite; I: illite.

683 Fig. 6. Photomicrographs of associated authigenic cements in the Xu4 sandstone. (A)
684 Plane-polarized light and (B) cross-polarized light images showing calcite and dolomite cements
685 and kaolinite associated with quartz overgrowth, CM39 well, 3108.8 m; (C) Pore-filling quartz
686 cement and calcite cement in dissolutio pores, GM3 4084.1 m; (D) Growth of calcite wrapped

quartz cement, MS1, 4393.1 m; (E) Kaolinite and illite fill in intergranular pores, X11 well, 3446.37 m; (F) Filiform illite growing on quartz cement, CF563 well, 3913.6 m; (G) and (H) Quartz overgrowth and fibrous illite fill in intergranular pores, CF563 well, 3882.7 m, SEM image; and (I) Idiomorphic columnar quartz cement and leaf-like chlorite, CF563 well, 3511.8 m; QO: quartz overgrowth; PFQ: pore-filling quartz cement; C: calcite cement; D: dolomite cement; K: kaolinite; I: illite; C; chlorite.

Fig. 7. (A) Plot of cements versus intergranular volume (modified from Houseknecht, 1987); (B) Plot of quartz cement content versus carbonate cement content in the Xu4 sandstones.

Fig. 8. Clay mineral vertical distribution characteristics in the Xu4 sandstones.

Fig. 9. Photomicrographs, frequency histograms of T_h (°C) and plot of salinity versus T_h (°C) of fluid inclusions in the quartz cements: (A) GM4 well, depth 4088.89 m; (B) XC27 well, depth 4023 m; (C) Frequency histogram of T_h (°C) and (D) plot of salinity versus T_h (°C). QG: quartz grain; OC: quartz cement; FIs: fluid inclusions.

Fig. 10. (A) T_h (°C) versus CH₄ content (mol%) with CO₂ content (mol%); (B) Raman spectra with peaks at $1282/\text{cm}^{-1}$, $1386/\text{cm}^{-1}$ and $2912/\text{cm}^{-1}$ indicating the presence of CO₂ and CH₄ in the quartz fluid inclusions, CG561 well, depth 4009.01 m, where the red line is the spectrum of the gas phase and the black line is the spectrum of the liquid phase; (C) Raman spectra with peaks at 1282/cm⁻¹ and $1386/\text{cm}^{-1}$ and at $2914/\text{cm}^{-1}$ indicating the presence of CO₂ and CH₄ in the quartz fluid inclusions, respectively, GM3 well, depth 3781.5 m, where the red line is the spectrum of the gas phase and the black line is the spectrum of the liquid phase.

Fig. 11. Photomicrographs and $\delta^{18}O_{SMOW}$ distribution of quartz cements: (A) Sample panoramic image, (B) plane-polarized light and (C) BSE image, Y3 well, 3133.18 m; (D) Sample panoramic image, (E) plane-polarized light and (F) BSE image, XC27 well, 4023 m; and (G) Distribution of $\delta^{18}O_{SMOW}$ value.

Fig. 12. BSE (A), mineral phases (B), Na (C) and K (D) elemental distribution images showing the dissolution of albite to form illite and quartz cements, 4194.72 m, DY1 well.

Fig. 13. (A) Feldspar compositions and vertical distribution characteristics of the Xu4 sandstones; mineral phases from TIMA showing that the content of the plagioclase subgroup is much higher

than that of K-feldspar, (B) HL1, 3477.3 m and (C) GM4, 4088.89 m.

- **Fig. 14.** Diagram of T_h (°C)- $\delta^{18}O_{quartz}$ - $\delta^{18}O_{fluid}$ of quartz cements in the Xu4 sandstones. The $\delta^{18}O_{fluid}$ ratio of quartz cement can be calculated based on equation of Clayton et al. (1972), where $\alpha_{quartz-fluid}$ is the fraction factor and defined as $\alpha_{quartz-fluid} = (1+(\delta^{18}O_{quartz}/1000))/(1+(\delta^{18}O_{fluid}/1000)).$
- 719 Fig. 15. Diagram of the Al/(K+Na) and Ca/Al ratios of the whole rock analysed by TIMA showing
- the intensity of meteoric water leaching in the Xu4 sandstones.
- Fig. 16. Synthetic schematic diagram of the silica sources and diagenetic sequence of the Xu4sandstones.

Table captions

Table 1. Homogenization temperatures (°C) and salinity (wt% NaCl eq.) of fluid inclusions in quartz cements.

Table 2. Concentration (mol%) of the gaseous components of fluid inclusions in quartz cements.

728	Table 1

Well	Depth(m)	Size(µm)	T_h (°C)	Salinity (wt% NaCl eq.)	No.
CG561	4009.01	1×2~ 5×10	60.7~ 143.4	4.18~12.05	11
HL1	3477.65	2×4~ 4×10	71.5~ 98.8	0.18~10.11	9
HL1	3478.65	1×2~ 5×10	62.4~120.5	1.4~10.35	9
XC22	3553.8	3×4~ 3×8	66.8~115.3	10.24~11.57	3
Y3	3133.18	1×2~ 6×8	49.2~130.8	1.91~10.98	9
GH2	4011.66	2×3~ 6×12	83.6~130.2	0.18~11.22	7
GH2	4008.86	1×2~ 4×6	67.6~94.6	4.03~13.62	8
GM3	3781.5	3×5~ 4×5	80.4~82.3	0.53~5.11	2
GM3	3784.2	2×4~ 3×8	81.7~118.1	5.26~14.15	5
GM4	4088.89	1×2~ 5×7	58~99.5	0.35~13.07	13
XC31	3740.38	1×2~ 6×8	63.9~90.7	1.4~9.6	5
CH139	3781.79	3×4~ 2×7	63.2~75.3	8.14~9.08	2
CJ566	3478.61	1×2~ 3×10	52.4~91.3	6.59~9.47	6
XC27	4023	2×3~ 7×12	52.1~102.9	3.39~13.18	10

Well	Depth(m)	$T_h(^{\circ}\mathrm{C})$	Composition	Gaseous components (mol%)	
				CH ₄	CO_2
CG561	4009.01	65.8	$CH_4(g), CO_2(g), H_2O(l)$	61.3	38.7
00501		85.7	CH ₄ (g), H ₂ O(l)	100	0
Ш 1	3477.65	71.5	CH ₄ (g), H ₂ O(l)	100	0
ΠLΙ		98.8	CH ₄ (g), H ₂ O(l)	100	0
HL1	3478.65	115.4	CH ₄ (g), H ₂ O(l)	100	0
VC22	3553.8	66.8	CH ₄ (g), H ₂ O(l)	100	0
AC22		115.3	CH ₄ (g), H ₂ O(l)	100	0
W2	2122 19	130.8	CH ₄ (g), H ₂ O(l)	100	0
15	3133.18	115.9	CH ₄ (g), H ₂ O(l)	100	0
GH2	4011.66	95.9	CH ₄ (g), H ₂ O(l)	100	0
GH2	4008.86	82.1	CH ₄ (g), H ₂ O(l)	100	0
GM3	3781.5	82.3	CH ₄ (g), CO ₂ (g), H ₂ O(l)	53.4	46.6
CM2	3784.2	86.7	CH ₄ (g), H ₂ O(l)	100	0
GM3		118.1	CH ₄ (g), H ₂ O(l)	100	0
CM4	4099 90	99.5	CH ₄ (g), CO ₂ (g), H ₂ O(l)	22.6	77.4
GM4	4088.89	85.8	CH ₄ (g), H ₂ O(l)	100	0
XC31	3740.38	90.7	CH ₄ (g), CO ₂ (g), H ₂ O(l)	42.0	58.0
CH139	3781.79	114.3	CH ₄ (g), H ₂ O(l)	100	0
CJ566	3478.61	83.7	CH ₄ (g), H ₂ O(l)	100	0
VC07	4023	100.8	CH ₄ (g), H ₂ O(l)	100	0
AC27		92.5	CH ₄ (g), H ₂ O(l)	100	0

731 Note: g = gas; l = liquid.

Highlights

- Quantitative mineralogical analysis based on SEM-EDS was used to describe diagenesis appearance and trace silica sources
- Meteoric water dissolution in the sandstone was recognized by oxygen isotope of quartz cement and low temperature fluid inclusions
- Synthetic model for silica sources was established in the tight gas sandstone















Figure8

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Supplementary Interactive Plot Data (CSV)

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Liangbiao: Supervision, Writing - Review & Editing, Resources. Li Zhen: Writing - Review
& Editing, Methodology. Chen Hongde: Project administration, Resources.