Diagenesis and Diagenetic Evolution of Deltaic and Neritic Gas-Bearing Sandstones in the Lower Mingyuefeng Formation of Paleogene, Lishui Sag, East China Sea Shelf Basin: Implications for Depositional Environments and Sequence Stratigraphy Controls

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Abstract: Gas-bearing deposits in the Lower Mingyuefeng Formation of Paleogene, Lishui Sag, East China Sea Shelf Basin consist of shoreface sandstones of the highstand systems tract (HST) and transgressive systems tract (TST), and deltaic sandstones of the lowstand systems tract (LST) and falling stage systems tract (FSST). Detailed petrographic observations suggest that the diagenetic features and related evolution of these deposits cannot be simply characterized and demonstrated in the depth domain. However, the occurrence of diagenetic minerals systematically depends on the studied interval within the HST, TST, LST, and FSST; therefore, diagenesis in this region can be better constrained when studied in the context of the depositional environments and sequence stratigraphic framework. The eogenetic processes in such settings include: (1) microcrystalline sidereite precipitated as concretions in almost all environments and systems tracts, which inhibited further mechanical compaction; (2) grain dissolution and kaolinitization occurred in shoreface HST sandstones and deltaic LST and FSST sandstones; (3) glaucony was locally observed, which did not clearly reflect the controls of facies or sequence stratigraphy; and (4) cementation by pyrite aggregates occurred in the shoreface HST sandstones and deltaic LST sandstones. The mesogenetic diagenesis includes: (1) partial conversion of kaolinite into dickite in deltaic LST sandstones, and minor chlorite cementation in deltaic FSST sandstones; (2) transformation of kaolinite into illite and quartz cementation in deltaic LST and FSST sandstones; (3) frequent precipitation of ankerite and ferroan calcite in shoreface TST sandstones and early HST sandstones, forming baffles and barriers for fluid flow, with common calcite in shoreface HST sandstones as a late diagenetic cement; and (4) formation of dawsonite in the deltaic LST and FSST sandstones, which is interpreted to be a product of the invasion of a CO₂-rich fluid, and acts as a good indicator of CO₂-bearing reservoirs. This study has thus constructed a reliable conceptual model to describe the spatial and temporal distribution of diagenetic alterations. The results may provide an entirely new conceptual framework and methodology for successful gas exploration in the continental margins of offshore China, thus allowing us to predict and unravel the distribution and quality evolution of clastic reservoirs at a more detailed and reliable scale.

Key words: diagenetic alteration, depositional environment, sequence stratigraphy, reservoir quality, Paleogene, East China Sea Shelf Basin

1 Introduction

Depositional and sequence-stratigraphic settings

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1995; Dutton and Willis, 1998; Morad et al., 2010; Ketzer et al., 2002; Lima and De Ros, 2002; Al-Ramadan et al., 2005; El-ghali et al., 2006, 2009; Karim et al., 2010; Kordi et al., 2011; Zhang et al., 2015a, 2015b), providing a feasible way of controlling reservoir quality in sandstones. This integrated concept has been demonstrated to be powerful and successful in identifying the links between reservoir quality and mappable geological features, rather than solely focusing on diagenetic modifications. Various parameters are controlled in the pathways of diagenetic evolution, including pore-water chemistry, rate of deposition, detrital composition and texture, depositional environments or facies, extent of bioturbation, and burial thermal history (Morad et al., 2000, 2010; Ajudwikiewicz et al., 2010; Taylor et al., 2010; Kordi et al., 2011; Zhu et al., 2009; Wang et al., 2014; Yang et al., 2014; Zhang et al., 2007, 2013a, 2014a; Zhang et al., 2014b).

Although hydrocarbon exploration activity has been ongoing in the East China Sea Shelf Basin (ECSB) since the 1970s, few studies have focused on diagenetic and the related evolution pathways in the Lower Paleogene Mingyueng Formation of the Lishui Sag in the ECSB are considered to be important hydrocarbon (particularly gas) reservoir targets. A detailed insight into the reservoir quality evolution of such reservoirs is of great interest owing to their economic importance. In this paper, we report a study based on the connections among diagenesis, sequence-stratigraphic intervals, and depositional environments for the first time, to generate a model for predicting the distribution of diagenetic processes and the related reservoir quality in the ECSB. The results will provide a useful conceptual framework in terms of prospects and play risk assessments.

2 Geological Setting, Depositional Environment and Sequence Stratigraphy

The East China Sea (ECS), a semi-closed sea along the continental margin of China, is located within the convergence area between the Eurasian, Pacific, and Philippine Sea Plates (Yang et al., 2004; Xu et al., 2012; Guo et al., 2013). The ECSB is the largest Cenozoic sedimentary basin within the ECS, covering an area of about 460,000 km² (Zhou et al., 2001; Ye et al., 2007; Xu et al., 2013). The ECSB is composed of a series of depressions (Taipei, Changjiang, Diaobei, Xihu, and Fujiang) separated by the Diaoysuda, Yushan, and Hupijiao uplifts (Fig. 1). The Taipei Depression can be divided into four structural belts: the Lishui Sag, Jiaojiang Sag, Fuzhou Sag, and Yandang lower-uplift.

The study area, the Lishui Sag, occurs in the southwestern portion of the Taipei Depression and consists of the Western Subsag and Eastern Subsag, trending generally northeast–southwest (Fig. 1). It has been recognized as a ridged sag, covering an area of approximately 14,600 km² (Ge et al., 2003; Jia et al., 2006). Tectonic events in the Lishui Sag can be subdivided into an Upper Cretaceous rift basin, Paleogene rifting, an Eocene depression, and Neogene subsidence (Jia et al., 2006). The maximum thickness of sediments in the Lishui Sag can reach up to 13,000 m (Ge et al., 2003). The studied stratigraphic interval is the Lower Mingyueng Formation of Paleogene age, which overlies the Lingfeng Formation and is overlain by the Oujiang Formation (Fig. 2). Organic-rich mudstones of the Yueguifeng Formation were believed to be the main gas source rocks for the Mingyueng deposits, and elemental analysis has indicated a predominant type-II kerogen within the Yueguifeng Formation (Ge et al., 2007).

The deposition of Lower Paleogene Mingyueng sandstones has been interpreted to have occurred in shoalface and deltaic environments (Fig. 2; Zhang et al., 2014c). The associated systems tracts are outlined in Fig. 2. The sequence-stratigraphic model of sequence IV has been used to fit the field observations, in combination with a tectono-stratigraphic analysis, sequence stratigraphy, and seismic geomorphology (Fig. 2; cf., Hunt and Tucker, 1992; Helland-Hansen and Gjelberg, 1994; Catuneanu, 2002; Catuneanu et al., 2009; Catuneanu et al., 2011; Zhang et al., 2014c).

3 Samples and Methods

Sandstone samples were collected from cored intervals of the wells drilled within the study area, which cover variations in the depositional environments and systems tracts. Thin sections were prepared for selected samples and impregnated with blue epoxy under vacuum; the mineralogy, diagenetic relationships, and porosity were determined after the samples had been cleaned to remove oil. Modal analyses were obtained for the representative sandstones, and 300 points were recorded for each thin section. A Quanta 200 scanning electron microscope (SEM) was used to visualize rock morphology and investigate the habits and textural relationships of diagenetic minerals, with an accelerating voltage of 20 kV and beam current of 10 nA. X-ray diffraction analyses were performed on each collected sample by using a D/max-1200 X-ray diffractometer, particularly intended to determine the presence and composition of clay minerals.

4 Results

4.1 Detrital mineralogy
Reservoir rocks are typically characterized by fine- to medium-grained sandstones. The modal detrital composition of the Lower Paleogene Mingyufeng sandstones indicates that the sandstones are predominately feldspathic litharenite and less commonly lithic arkose (sensu Forl, 1980) with an average composition of Q_{23}F_{10}R_{57} indicated by the modal analysis (Fig. 3). Component grains were found to be mostly subangular to subrounded, with moderate sorting in general. The quartz content is relatively low in the Lower Mingyufeng Formation, and monocrystalline quartz grains are more abundant. Feldspars are relatively less abundant in the rock by volume, although K-feldspar (average: 7.2 vol%) is more common than plagioclase (average: 3.4 vol%). Modal analyses revealed that rock fragments (igneous, metamorphic, and sedimentary) are the most abundant detrital minerals in all sandstone samples, with the igneous component predominant (average: 47.2 vol%). Mica is rare, with only trace amounts found (average: <0.1 vol%).

4.2 Diagenetic alterations
Carbonates observed in the studied sandstones include ankerite, ferroan calcite, siderite, and calcite. Ankerite and ferroan calcite are common in the shoreface TST and early HST, both as pore-filling cement and as a replacive mineral, reaching up to 28% of the rock volume, and typically exhibit poikilotopic or blocky texture (Figs. 4a, b). Ankerite engulfs, and thus postdates, ferroan calcite (Fig. 4b). Ankerite is less abundant in the deltaic LST and FSST sandstones. Siderite lines pore spaces, forming as concretions in all depositional facies and systems tracts (Fig. 4c). Calcite is most abundant in shoreface HST
sandstones, and post-dates quartz overgrowths and the dissolution of feldspars. Moreover, trace amounts of calcite cement are present, filling intergranular pores in deltaic LST sandstones.

According to SEM observations, kaolinite (average: 3.7 vol%) occurs as booklet-like aggregates partially replacing feldspars, particularly in shoreface HST and deltaic LST sandstones (Fig. 4d). In some cases, kaolinite transforms into blocky and thicker crystals of dickite in deltaic LST sandstones (Fig. 4d). Trace amounts of thin pore-lining chlorite cement forming thin fringes have developed in deltaic FSST sandstones. Minor amounts of pore-lining illite, observed in deltaic LST and FSST sandstones, have formed by the replacement of kaolinite according to the morphology of illite (Fig. 4d).

As one of the minor diagenetic minerals, pyrite is identified as frambooidal aggregates (<2 μm) and always fills intergranular pores. Pore-filling pyrite is overall abundant in shoreface HST and deltaic LST sandstones.

Quartz cement (average: <0.5 vol%) occurs as syntaxial overgrowths on detrital quartz grains and less commonly as pore-filling quartz overgrowths. The quartz overgrowths partly fill the adjacent intergranular pores (Fig. 4e), whereas pore-filling quartz overgrowths occur on quartz grains and post-date illite crystals (Fig. 4f). Authigenic quartz cement has developed preferentially where clay rims are discontinuous or nearly absent, covering detrital quartz grains. Quartz cement is slightly more abundant in the deltaic LST and FSST sandstones than in other samples.

Dawsonite (NaAlCO₃(OH)₂) is present as a fibrous and locally pore-filling cement (Fig. 4g) comprising up to 8% of the rock volume, as observed by a point-count analysis, in both deltaic LST and FSST sandstones. In some cases, dawsonite has replaced detrital plagioclase feldspar grains locally (Fig. 4h).

Glaucophy occurs in trace amounts, observed in few samples with a dark green color. However, the limited amounts of glaucony do not display a systematic distribution according to the depositional environments or the systems tracts.

Dissolution of detrital framework grains, mainly feldspar and less commonly rock fragments, is common in shoreface HST and deltaic LST and FSST sandstones, which has resulted in secondary porosity. A considerable volume of
Fig. 4. Photomicrographs of authigenic minerals in Lower Mingyuefeng sandstones of Paleogene.
(a), Ankerite (Ank) fills pore spaces and replaces framework grains; (b), Pore-filling ankerite engulfs ferroan calcite (F-cal); (c), Pore-filling siderite (Si) sits around detrital grains; (d), Partial booklet-like kaolinite (K) forms blocky crystals of dickite (Di) with smooth surfaces, along with illitization (I) of kaolinite; (e), Euhedral quartz overgrowths (Qo) around detrital quartz grains; (f), Isolated quartz overgrowths (Q) occur as pore-filling crystals engulfsing, and thus post-dating, illite; (g), Dawsonite (Daw) exhibits fibrous texture; and (h), Dawsonite has replaced detrital grains.
both secondary intergranular and intragranular porosity is evident, with good connectivity with primary pores.

5 Discussions

5.1 Paragenetic sequence

On the basis of the petrographic examinations, the paragenetic sequence of the Lower Paleogene Mingyuefeng sandstones has been reconstructed to characterize the variable relationships of diagenesis to the depositional environments and systems tracts during eogenetic and mesogenetic regimes (Fig. 5).

Siderite, favored under reducing conditions and at relatively high Fe/Ca ratios (Berner 1981), indicates that pore spaces are available for precipitation owing to its euhedral shape (Fig. 4c). The meteoric waters have resulted in the initial dissolution of feldspars. The absence of feldspar kaolinitization implies that the formation of eogenetic kaolinite is caused by the dissolution of feldspars (Fig. 4d). The sulfide ions, which are supplied by pyrite precipitation with framoidal texture, are almost certainly related to bacterial sulfate reduction.

Partial transformation of kaolinite to dickite indicates a neomorphic transformation to a more well-ordered and stable structure under mesogenetic conditions (Fig. 4d; cf., Ehrenberg et al., 1993; McAulay et al., 1994; Kordi et al., 2011). Extensive mesogenetic cementation, particularly carbonate cementation, inhibits mechanical compaction to some extent and forms baffles and barriers for fluid flow (Figs. 4a, b). The petrographically determined quality of the carbonates has been plotted versus their porosity, illustrating that the carbonate cements exert strong controls on reservoir quality (Fig. 6). The occurrence of mesogenetic carbonates requires mechanisms of Ca and Mg mass transfer, which can be generated by shale diagenesis (cf., Boles, 1978; Land et al., 1987; Moraes and Surdam, 1993). The lack of quartz cement in shoreface HST and TST sandstones is due primarily to pervasive carbonate cementation. In general, the major source of silica for the mesogenetic quartz cement of the studied sandstones is likely to be the reaction of detrital feldspar minerals or illitization (cf., Barclay and Worden, 2000;

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Fig. 5. Paragenetic sequence and relative timing of the main diagenetic features that occurred in all depositional environments and systems tracts of the Lower Mingyuefeng sandstones.
Porosity (%)

Carbonates (%)

Plagioclase (%)

Dawsonite (%)

Worden and Morad, 2000; Zhang et al., 2012). Illite precipitation in the Lower Mingyuefeng Formation appears to be a result of the reaction of kaolinite and K-feldspar, which is considered to be the source of potassium for illitization (cf., Wilson, 1982; Chuhan et al., 2000; Franks and Zwingmann, 2010). The formation of chlorite as rims around framework grains may have occurred as a result of the transformation of detrital or cogenetic clays (cf., Gier et al., 2008; Zhang et al., 2008; Zhang et al., 2013b). Several studies have shown that dawsonite may be a routine consequence of CO₂ injection (e.g., Du, 1982; Xu et al., 2003; Sun and Xi, 2003; Worden, 2006; Bai and Keene, 2007). As a late-stage diagenetic mineral of the subsurface environment in the studied sandstones, it is regarded to reflect the presence of CO₂-rich fluids, thus helping to locate CO₂-bearing reservoirs. The negative correlation between dawsonite and plagioclase feldspar may reflect the dissolution of plagioclase feldspars as a source for dawsonite in the deltaic LST and FSST sandstones (Fig. 7).

5.2 Model for the distribution of the primary diagenetic alterations associated with reservoir quality evolution

A conceptual model for the spatial and temporal distribution of diagenetic alterations in the Lower Paleogene Mingyuefeng sandstones is shown in Fig. 8, which indicates that the diagenetic alterations can be linked to depositional environments and systems tracts. In addition, the characteristics of reservoir generation, destruction, and preservation are intimately linked to diagenetic processes. Thus, the reservoir quality of the studied sandstones is determined by variations in diagenetic processes within depositional environments and the sequence-stratigraphic framework. The main diagenetic alteration generating reservoir quality is the dissolution of detrital framework grains, which results in the formation of secondary pores. Secondary porosity resulting from such dissolution is well documented to be important to the storage and producibility of deeply buried sandstones (e.g., Ehrenberg and Jakobsen, 2001; Taylor et al., 2010). Diagenetic alterations that destroy reservoir quality includes mechanical compaction, carbonate cementation, and quartz cementation. In particular, local but significant carbonate cementation events occurring in shoreface TST sandstones can occlude most of the pore spaces, thus playing the primary role in precluding gas accumulation. Diagenetic alterations that help preserve reservoir quality contribute to the conversion of kaolinite into dickite, illitization, and the formation of chlorite. Moreover, the formation of pyrite, siderite, and dawsonite and grain kaolinitization have a slight impact on reservoir quality.

A plot of the total intergranular volume versus intergranular cement demonstrates that the primary porosity loss owing to compaction was more important than cementation in both LST and FSST sandstones (Fig. 9). This can be attributed to the presence of minor amounts of eogenetic cements capable of preventing mechanical compaction (cf., Salem et al., 2005).

Linking the impact of diagenetic alterations on reservoir heterogeneity to the depositional environments and sequence stratigraphy, it can be predicted that deltaic LST and FSST sandstones will be important targets for gas accumulation, with relatively good reservoir quality. This knowledge and the establishment of an evolution model of reservoir quality within the framework of different depositional environments and systems tracts will provide a valuable means of predicting and ranking prospects in hydrocarbon exploration and production.

6 Conclusions

The results of this study facilitate the prediction of the distribution of diagenetic alterations based on the links between such alteration and the depositional environments and the sequence-stratigraphic framework among the
Fig. 8. Schematic diagenetic model showing evolution and diagenetic alterations of the depositional environments and the sequence-stratigraphic framework of the Lower Mingyuefeng Formation.

Fig. 9. Cross-plot of cement volume versus total intergranular volume (after Houseknecht, 1987) for the deltaic LST and FSST sandstones, indicating that compaction is more important than cementation in porosity reduction.

shoerface HST and TST sandstones and deltaic LST and FSST sandstones of the Lower Paleogene Mingyuefeng Formation in the Lishui Sag. The main diagenetic alteration affecting the studied sandstones include the following: (1) grain dissolution, primarily in the deltaic LST and FSST sandstones; (2) mechanical compaction in all systems tracts; (3) quartz cementation, primarily in shoerface HST and TST sandstones; (4) carbonate cementation, primarily in shoerface HST and TST sandstones; (5) illitization in deltaic LST and FSST sandstones; (6) transformation of kaolinite into dickite in deltaic LST sandstones; and (7) formation of chlorite fringes in deltaic FSST sandstones.

Deltaic LST and FSST sandstones are expected to be the most important targets for gas accumulation in the study area. This case study has demonstrated that synthesis of diagenetic alterations, depositional environments, and sequence stratigraphy can help predict reservoir quality as a cumulative product. Thus, this method will be applicable in
the exploration and production of gas-bearing sandstones and the generation of reservoir quality evolution models.

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