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Heterogeneous distribution and potential significance of solid bitumen in paleo-oil reservoirs: Evidence from oil cracking experiments and geological observations

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ABSTRACT

The solid bitumen (SB) residing in reservoirs after oil cracking can reduce reservoir quality. To build a quantitative standard for the SB content for paleo-oil reservoir identification and to evaluate the heterogeneous distribution of SB and its effect on reservoir quality, we conducted a cracking experiment on eight dolostone samples injected with oil and analyzed the SB content (vol%) and porosity of 706 reservoir samples from the Sichuan Basin that had undergone oil cracking. The SB contents of the experimental samples (0.95–1.48 %) injected with normal oil were higher than those of the experimental samples (0.22–0.49 %) injected with light oil. Furthermore, for the experimental samples injected with the same type of oil, the SB contents of the samples with higher paleoporosities (PPs) were also higher than those of the samples with lower PPs. For the reservoir samples from the Sichuan Basin, the SB content exhibits a generally positive correlation with PP. When the PP was <2.5 %, the SB content was relatively low (<1.0 %); and when the PP was >2.5 %, the SB content increased significantly and was generally > 1.0 %. The data obtained from the oil cracking experiments and reservoir samples from the Sichuan Basin suggests that SB contents of 1.0 % and 0.4 % can be used as the lower limits for normal and light oil reservoir identification, respectively. The oil cracking experiments verified that SB preferentially resided in the smaller pores and throats, and the SB saturations of the samples with lower PPs were larger than those of the samples with higher PPs. For the reservoir samples from the Sichuan Basin, a paleo-oil reservoir with a PP > 8.0 % can be an effective gas reservoir, but some of the paleo-oil reservoirs with PPs of 2.5 %–8.0 % are ineffective gas reservoirs due to SB filling. As a result, the heterogeneous distribution of SB should be carefully considered in gas reservoir evaluation.

1. Introduction

Many medium to large gas fields, such as the Puguang, Yuanba, Longgang, Dukouhe, and Luojiazhai gas fields, have been identified in the Upper Permian to Lower Triassic marine carbonate strata in the Sichuan Basin, southwest China (Ma et al., 2007, 2010; Guo, 2011; Guo et al., 2018). The hydrocarbon gas in these gas fields was mainly derived from in-situ oil cracking (Li et al., 2005; Hao et al., 2008; Li et al., 2015a, 2015b). Solid bitumen (SB) is commonly found in these gas reservoirs, which provides direct evidence of oil cracking (Li et al., 2005; Hao et al., 2008; Li et al., 2015a, 2015b). The quantitative identification of paleo-oil reservoirs can be used to evaluate the natural gas potential based on the gas oil cracking production model (Barker, 1990; Zhang et al., 2008; Li et al., 2016), if the gas is considered to be mainly derived from in-situ oil cracking. In addition, the correlation between the present gas-water contact and the paleo-oil-water contact based on paleo-oil reservoir identification can be used to analyze natural gas remigration after oil cracking and to predict the position of the gas-water contact (Li et al., 2008, 2015a).

Conventional methods used to identify paleo-oil layers include grains containing oil inclusions (GOI; Eadington et al., 1996; Lisk et al., 2002), quantitative grain fluorescence (QGF; Liu et al., 2007), the amount of soluble organic matter or residual oils (Wang et al., 2006), and SB content (Wang et al., 2006; Li et al., 2015b). The paleo-oil in the

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Permian-Triassic and older deep marine carbonate reservoirs in the Sichuan Basin has experienced complete oil cracking. Oil inclusions are rarely found in these gas reservoirs (Wang, 2008) and the SB displays a high reflectance (pyrobitumen; Sassen, 1988) in current gas reservoirs (Hao et al., 2008; Li et al., 2015a, 2015b). As a result, the conventional GOI and QGF methods, as well as the amount of soluble organic matter or residual oil, cannot be used to identify paleo-oil reservoirs, however, the SB residing in gas reservoirs after oil cracking becomes a direct clue for identifying paleo-oil reservoirs. However, no quantitative lower limit of the SB content has been proposed for the identification of paleo-oil reservoirs. Wang et al. (2006) suggested that a SB content of greater than 2.0 % can be used to identify paleo-oil reservoirs, but they did not present systematic experimental data. Li et al. (2015b) proposed that a SB content of greater than 1.0 % can be used to identify paleo-oil reservoirs based on the SB content vs. PP of samples from the Lower Triassic Feixianguan Formation (T_1f) in the Jiannan gas field. However, this lower limit (SB of 1.0 %) needs to be verified further using data from other gas fields in the Sichuan Basin that have experienced in-situ oil cracking as well as through oil cracking experiments.

As a direct product of oil cracking, SB inevitably resides in the pores of a reservoir, reducing the porosity and permeability of the reservoir and increasing the reservoir's heterogeneity (Jacob, 1989; Lomando, 1992; Li et al., 2015b; Wang et al., 2015; Wood et al., 2015, 2020). Previous studies have primarily focused on the origin and occurrence of SB (Jacob, 1989; Hwang et al., 1998; Walters et al., 2006; Kelemen et al., 2008, 2010; Xiong et al., 2016; Gao et al., 2018; Mastalerz et al., 2018), but little research has been conducted on the distribution of SB and its effect on reservoir quality.

Huc et al. (2000) found that small pores were preferentially invaded by SB compared with larger ones after oil cracking. Li et al. (2015b) also found that the distribution of SB in a T_1f reservoir in the Jiannan gas field in the Sichuan Basin was heterogeneous after oil cracking. The SB saturation (i.e., the proportion of SB filling the paleopores; Wood et al., 2015, 2020) was larger in lower porosity reservoirs than in higher porosity reservoirs, which resulted in some prior reservoirs with lower porosities becoming non-reservoirs after oil cracking (Li et al., 2015b; Wang et al., 2015). As a result, the heterogeneous distribution of SB plays a key role in the quality evaluation of deep reservoirs that have experienced oil cracking in the Sichuan Basin and in other basins worldwide (Li et al., 2015b; Wood et al., 2015, 2020). However, the heterogeneous distribution of SB needs to be further investigated.

To establish a quantitative standard of SB content for paleo-oil reservoir identification, and to confirm the heterogeneous distribution of SB after oil cracking, we injected dolostone samples with different porosities from the Sichuan Basin with two different oils (normal oil and light oil). We then conducted high-temperature oil cracking experiments and analyzed the residual SB of the oil cracking samples and those of actual reservoir samples from typical gas fields in the Sichuan Basin to confirm the lower limit of the SB content for paleo-oil reservoir identification and the heterogeneous distribution of SB.

2. Geologic setting

The Sichuan Basin is a rhombic basin located in southwestern China with an area of 180,000 km^2 (Fig. 1); the basin has experienced six tectonic cycles: the Yangtze (before 630 Ma), Caledonian (630–320 Ma), Hercynian (320–252 Ma), Indosinian (252–195 Ma), Yanshanian (195–65 Ma), and Himalayan (65–0 Ma) movements (Zhai, 1989). The basin was dominated by subsidence and uplift before the Early Indosinian and by folding and uplift since the Late Indosinian (Zhai, 1989; Ma et al., 2007). The basin can be divided into six secondary units (Zhai, 1989): the eastern complete fault-fold zone, the southern gentle fault-fold zone, the southwestern gentle fault-fold zone, the central flat zone, the northern gentle fault-fold zone, and the western gentle fault-fold zone (Fig. 1). The sedimentary rock cover of the Sichuan Basin is Precambrian to Quaternary in age. The marine carbonate and shale deposits were primarily developed from the Precambrian to the Middle Triassic, and the terrigenous sandstone and mudstone deposits were primarily developed since the Late Triassic. From the bottom to the top, the Permian can be divided into the Liangshan, Qixia, Maokou, Longtan/Wujiaping (P_2w), and Changxing (P_2c)/Dalong formations; and the Triassic can be divided into the Feixianguan (T_1f), Jialingjiang (T_1j), Leikoupo (T_2l), and Xujiahe formations (Zhai, 1989). The gas dryness of the P_2c and T_1f natural gas in these gas fields (e.g., the Puguang, Yuanba,

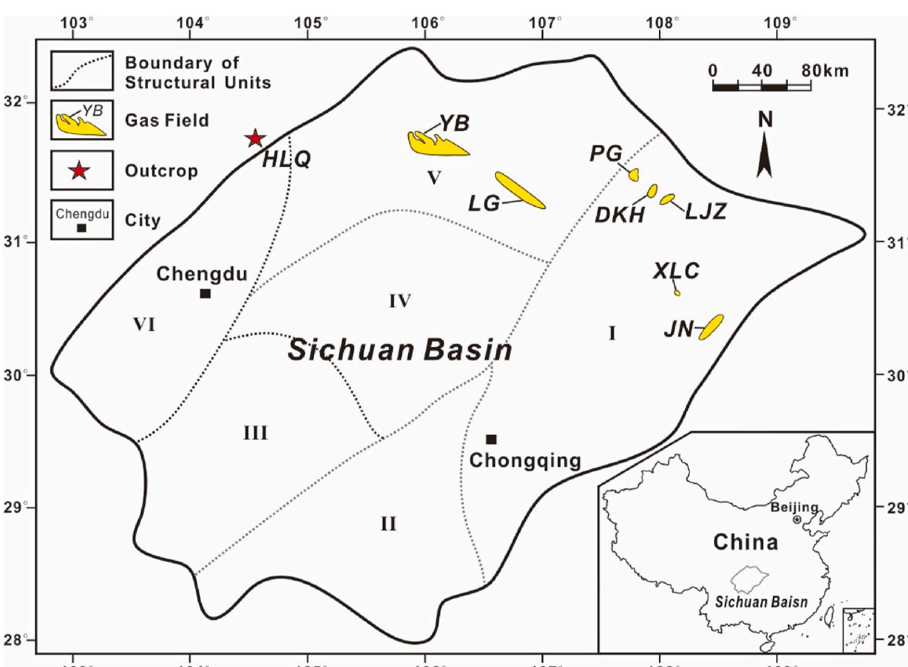

Fig. 1. Structural units and locations of the Puguang (PG), Yuanba (YB), Xinglongchang (XLC), Jiannan (JN), Longgang (LG), Dukouhe (DKH), and Luojiazhai (LJZ) gas fields and the Huanglianqiao (HLQ) outcrop in the Sichuan Basin. I = eastern complete fault-fold zone, II = southern gentle fault-fold zone, III = southwestern gentle fault-fold zone, IV = central flat zone, V = northern gentle fault-fold zone, VI = western gentle fault-fold zone.

Longgang, Dukouhe, and Luojiazhai gas fields) is very high (>0.99), and the maximum burial temperature of these gas reservoirs exceeds 200 °C. SB with a high maturity (reflectance of SB >2.50 %) has been found in the present gas reservoirs (Li et al., 2005, 2015a, 2015b, 2016; Hao et al., 2008), which provides evidence for paleo-oil cracking. For these discovered gas fields, the P_2w shale has been identified as the main source rock generating the oil, the P_2c and T_1f dolostone are the main reservoir rocks, and the late T_1f and T_1j gypsum are the regional caprocks (Li et al., 2005; Ma et al., 2007; Zou et al., 2008; Guo, 2011).

3. Sampling and analytical methods

In this study, we completed oil injection, oil cracking, and residual SB observations following the general procedure presented in Fig. 2. First, we collected eight dolostone samples from the Sichuan Basin, including two P_2c samples (No. 1 and No. 2) from the Yuanba (YB) gas field, two T_1f samples (No. 3 and No. 4) from the Puguang (PG) gas field, and four T_2l samples (No. 5, No. 6, No. 7, and No. 8) from the Huangliangqiao (HLQ) outcrop (104.5857° E, 31.7817° N). Each of these samples was cut into two cylinders (25 mm in diameter, 25–30 mm in length). Second, all the cylinders were extracted using a mixture of benzene and alcohol (2:1) and a Soxhlet extractor to remove soluble organic matter, and then they were dried at 120 °C for 4 h. We created thin sections (Fig. 3) impregnated with blue epoxy using one cylinder from each sample; weighed the other cylinder of each sample (m_0); and measured the pore volume (PV), porosity (ϕ_1), and permeability (K_1) using a Core Lab CMS-300 instrument. The analytical precisions of porosity and permeability are 0.01 % and 0.00005 md, respectively. Third, we injected normal oil (density of 0.814 g/cm^3) into three samples (No. 1, No. 3, and No. 7) under a pressure of 2.5 MPa and injected light oil (density of 0.742 g/cm^3) into five samples (No. 2, No. 4, No. 5, No. 6, and No. 8) under a pressure of 1.5 MPa using a core-holder system. We recorded the oil volume (OV) injected into each cylinder using a flow sensor and calculated the oil saturation (OS) as OV/PV. We weighed the oil-saturated cylinders (m_1) and stored them at 4 °C. Finally, we placed each oil-saturated cylinder into a quartz tube, which we placed in a high-pressure autoclave filled with nitrogen to maintain a confining pressure of 5 MPa. Then, we heated the samples from an initial temperature of 30 °C at a heating rate of 20 °C/h. We held the cylinder at 600 °C for 4 h to ensure that the oil in each cylinder was fully cracked (Tian et al., 2008). Each cylinder was weighed (m_2), and the porosity (ϕ_2) and permeability (K_2) were measured using the same CMS-300 instrument. We then created four thin sections from each cylinder and quantified the SB content (vol. %) using the Adobe Photoshop quantification (PSQ) method as described by Zhang et al. (2014). The analytical error of the SB content is about 0.02 %. To compare the

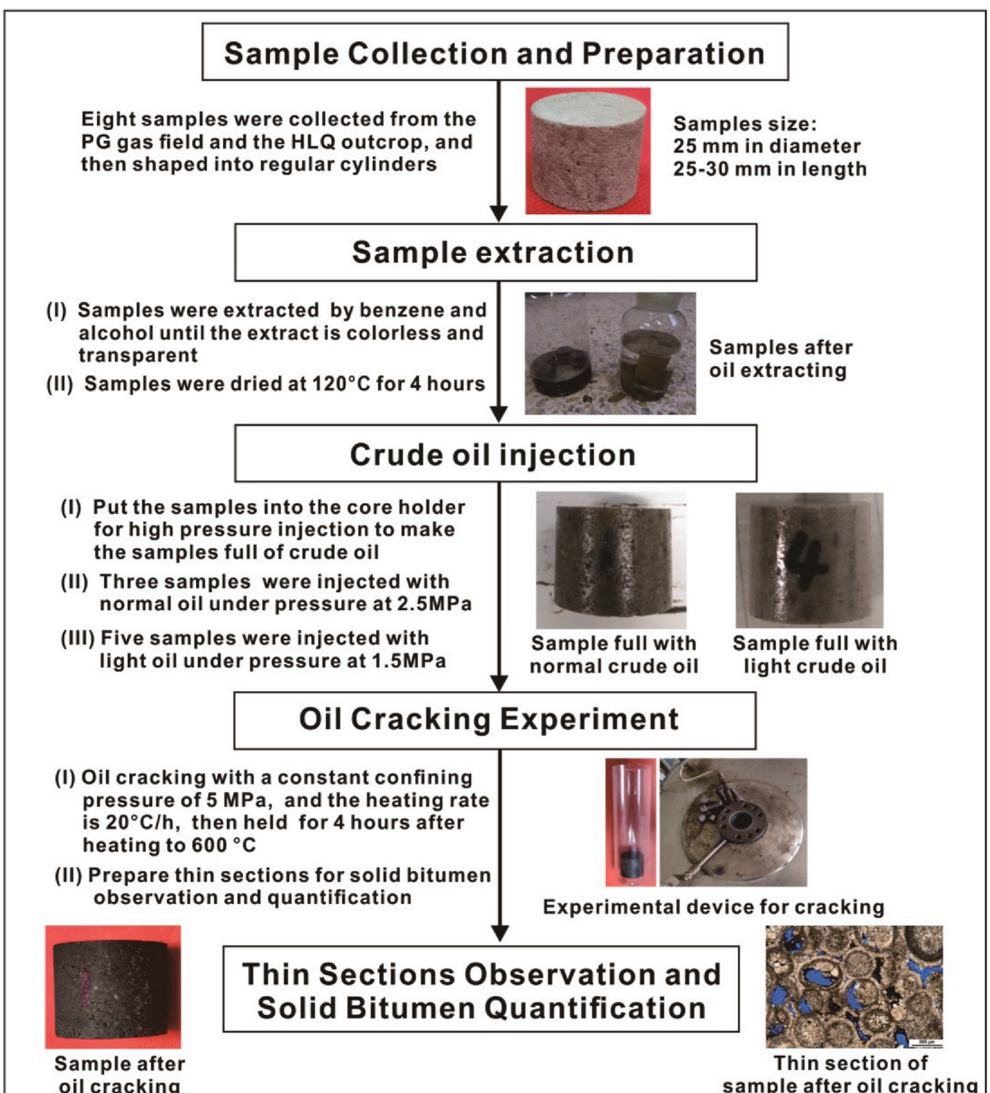

Fig. 2. General flow of the sample preparation, oil injection, oil cracking, and thin section observations.

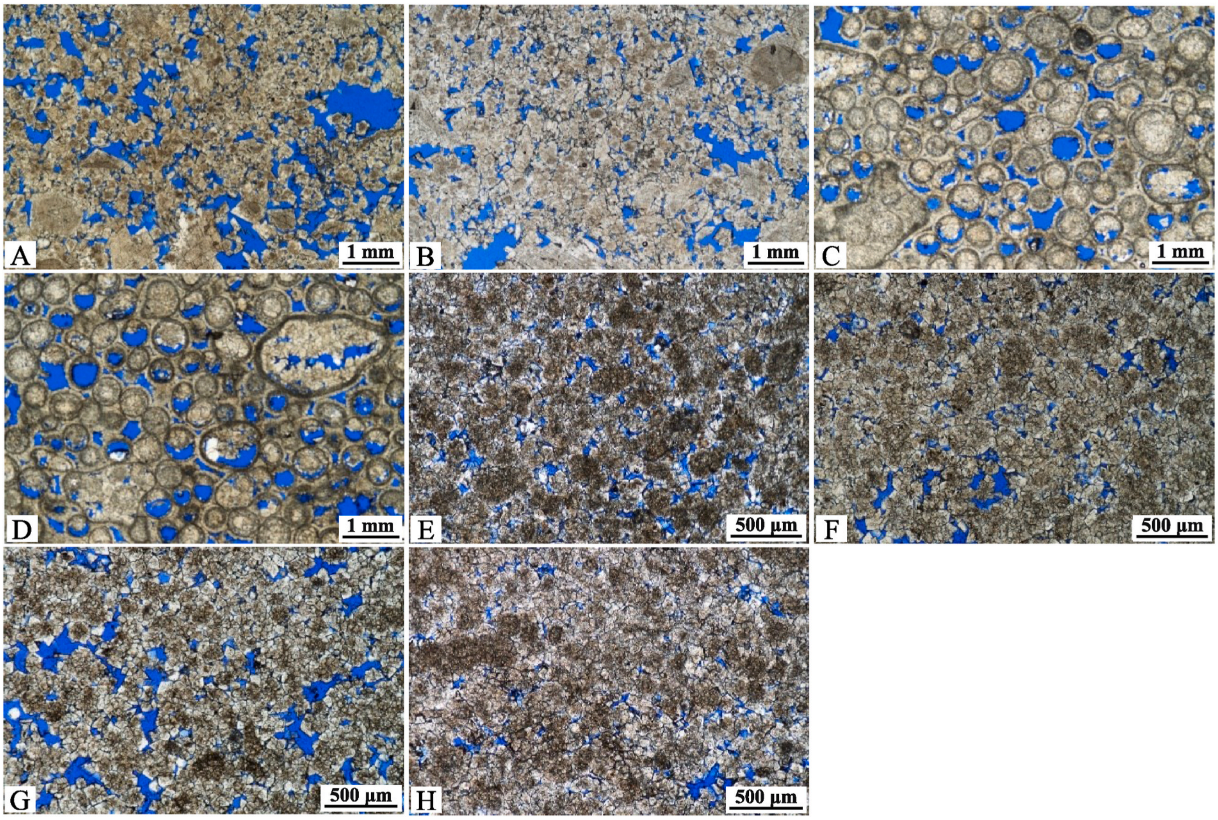

Fig. 3. Thin-section photomicrographs of dolostone samples after extraction using benzene and alcohol. (A, B) Samples No. 1 and No. 2 from the Upper Permian Changxing Formation in the Yuanba (YB) gas field, with porosities of 15.10 % and 14.40 %, respectively. (C, D, E) Samples No. 3 and No. 4 from the Lower Triassic Feixianguan Formation in the Puguang gas field, with porosities of 12.17 % and 13.68, respectively. (F, G, H) Samples No. 5, No. 6, No. 7, and No. 8 from the Middle Triassic Leikoupo Formation from the Huangliangqiao (HLQ) outcrop, with porosities of 6.87 %, 3.27 %, 7.69 %, and 2.56 %, respectively.

samples that experienced oil cracking in the Sichuan Basin, we made 309 thin sections using the samples from the YB, Xinglongchang (XLC), and PG gas fields and quantified their SB contents using the same PSQ method (Zhang et al., 2014). In addition, we also included 397 data points of porosity and SB content from the Jiannan (JN) gas field that were previously published by Li et al. (2015b).

We calculated the paleoporosity (PP, i.e., the porosity available at the time of oil charging) as the sum of present-day porosity and SB content (Li et al., 2015b; Wood et al., 2015, 2020). The SB saturation (i.e., the fraction of PP filled by SB) was calculated as the ratio of SB to PP (Wood et al., 2015, 2020).

Table 1
The weight, porosity, and permeability of the samples before and after oil cracking, and the solid bitumen (SB) content of the samples after oil cracking.

Sample ID	Sample location	Strata	Lithology	Oil Type	m_0 (g)	m_1 (g)	m_2 (g)	PV (ml)	OV (ml)	OS (%)	K_1 (mD)	K_2 (mD)	ϕ_1 (%)	ϕ_2 (%)	SB (%)	SB/ϕ_1 (%)
1	YB gas field	P_2ch	Dolostone	Normal oil	25.821	26.691	25.870	1.072	1.069	99.70	66.76	11.91	15.10	13.54	1.48	9.80
3	PG gas field	T_1f	Dolostone	Normal oil	29.163	30.114	29.217	1.184	1.167	98.54	13.09	2.27	12.07	10.60	1.31	10.85
7	Outcrop HLQ	T_2l	Dolostone	Normal oil	31.562	32.143	31.595	0.723	0.712	98.49	11.45	2.14	7.69	6.75	0.95	12.35
2	YB gas field	P_2ch	Dolostone	Light oil	23.961	24.792	23.964	1.140	1.119	98.15	62.82	46.10	14.40	13.90	0.49	3.40
4	PG gas field	T_1f	Dolostone	Light oil	27.883	28.736	27.886	1.165	1.146	98.36	15.10	6.97	13.68	13.18	0.48	3.51
5	Outcrop HLQ	T_2l	Dolostone	Light oil	31.932	32.467	31.934	0.734	0.714	97.33	9.92	3.22	6.77	6.32	0.42	6.65
6	Outcrop HLQ	T_2l	Dolostone	Light oil	32.784	33.095	32.785	0.429	0.418	97.46	0.09	0.05	3.27	3.03	0.24	7.34
8	Outcrop HLQ	T_2l	Dolostone	Light oil	29.531	29.732	29.532	0.272	0.270	99.24	0.07	0.03	2.56	2.16	0.22	8.59

Note: P_2ch = Upper Permian Changxing Formation, T_1f = Lower Triassic Feixianguan Formation, T_2l = Middle Triassic Leikoupo Formation. m_0 = weight of samples after extraction and drying, m_1 = weight of samples after oil injection, m_2 = weight of samples after oil cracking. PV = pore volume, OV = oil volume, OS = oil saturation. K_1 and ϕ_1 are the permeability and porosity of the samples after extraction and drying, respectively; and K_2 and ϕ_2 are the permeability and porosity of the samples after oil cracking, respectively.

4. Results

4.1. Porosity, permeability, and SB content of experimental samples after oil cracking

Thin section photomicrographs of the dolostone samples after extraction using benzene and alcohol are shown in Fig. 3. The SB cannot be observed in these photomicrographs because all the samples were collected from non-paleo-oil intervals. Intercrystalline pores can be observed in the crystallized dolostone from the YB gas field (Fig. 3A–B); intragranular and intergranular pores can be observed in the oolitic dolostone from the PG gas field (Fig. 3C–D); and intergranular pores can be observed in the intraclastic dolostone from the HLQ outcrop (Fig. 3E–H). The oil saturation (OV/PV) of the samples after oil injection was 97.33–99.7 %. The weights, porosities, permeabilities, and SB contents of experimental samples before and after oil cracking are presented in Table 1.

The residual SB content (volume of SB) after oil cracking was related to the type of oil injected and the sample's porosity before oil injection. The SB contents of the experimental samples (0.95–1.48 %) injected with normal oil were higher than those of the experimental samples (0.22–0.49 %) injected with light oil (Fig. 4). For example, samples No.1 and No. 4, which were injected with normal oil and light oil, respectively, had similar porosities before oil injection (15.10 % and 14.40 %, respectively), but the SB content of No.1 (1.48 %) was remarkably higher than that of sample No.4 (0.49 %) after oil cracking (Table 1). In addition, for the experimental samples injected with the same type of oil, the SB contents of the samples with higher porosities were also higher than those of the samples with lower porosities before oil injection. For example, the porosity of sample No. 1 (15.1 %) was higher than and that of sample No. 7 (7.69 %) before normal oil injection; and the SB content of sample No. 1 (1.48 %) was also higher than that of sample No. 7 (0.95 %) after oil cracking (Table 1). The porosity of sample No. 2 (14.2 %) was higher than that of sample No. 8 (2.56 %) before light oil injection; and the SB content of sample No. 2 (0.49 %) was also higher than that of sample No. 8 (0.22 %) after oil cracking (Table 1).

The porosities and permeabilities of the experimental samples both decreased after oil cracking (Fig. 5), and residual SB was observed in the pores and throats of the samples (Fig. 6). The decreases in the porosities and permeabilities of the experimental samples injected with normal oil were larger than those of the experimental samples injected with light oil after oil cracking (Fig. 5). Specifically, the decreases in the porosities and permeabilities of the experimental samples injected with normal oil were 0.94%–1.56 % and 9.31–54.85 mD, respectively, whereas those of the experimental samples injected with light oil were 0.24%–0.5 % and 0.04–16.72 mD, respectively (Table 1). In addition, for the experimental samples injected with the same type of oil, the decrease in permeability was larger than the decrease in porosity. For example, the porosity of sample No.1 decreased from 15.10 % to 13.54 %, but the permeability of sample No.1 decreased from 66.76 mD to 11.91 mD (Table 1).

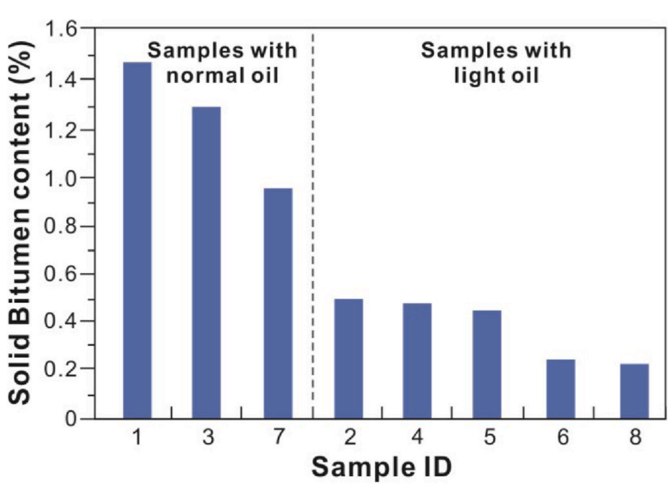

Fig. 4. The residual solid bitumen contents of the samples with normal and light oil after oil cracking.

4.2. SB contents and distributions of the reservoir samples from the Sichuan Basin

The porosities and SB contents of 706 reservoir samples from the JN, PG, YB, and XLC gas fields in the Sichuan Basin are presented in Supplementary Table S1. The porosities of these reservoir samples were 0–12.0 %, and the corresponding SB contents were 0–8.0 %. In general, we did not observe a significant positive correlation between the SB content and the present-day porosity (Fig. 7). In contrast, the SB content and PP exhibited a rough positive correlation (Fig. 8). When the PP was less than 2.5 %, the SB content was relatively low and was generally less than 1.0 %. In contrast, when the PP was greater than 2.5 %, the SB content increased rapidly and was generally greater than 1.0 % (Fig. 8). The SB saturation exhibited strong heterogeneity. When the PP was less than 2.5 %, the SB saturation ranged from 0 to 100 %; and when the PP was greater than 2.5 %, the SB saturation decreased as the PP increased (Fig. 9).

5. Discussion

5.1. Lower limits of the SB content for paleo-oil reservoir identification

In gas reservoirs that have experienced complete oil cracking, oil inclusions are rare; however, SB inclusions are common (Wang, 2008). Thus, the conventional GOI and QGF methods (Eadington et al., 1996; Lisk et al., 2002; Liu et al., 2007) cannot be used to identify paleo-oil reservoirs. In theory, SB inclusions can be used to identify paleo-oil reservoirs, but at present, no SB inclusion index similar to the GOI has been proposed to identify paleo-oil reservoirs, and it is relatively time-consuming to quantify these SB inclusions. In comparison, the residual SB content of the reservoir is easy to quantify based on thin sections of reservoir samples (Zhang et al., 2014; Li et al., 2015b), which can be effective indicators for quantitatively evaluating whether paleo-oil reservoirs have developed (Hao et al., 2008; Zhang et al., 2008; Li et al., 2015b).

Based on the oil cracking experiments conducted in this study, the SB contents quantified using the thin sections were generally slightly lower than the decrease in porosity (Table 1). The difference between the SB content and the decrease in porosity was within 10 % (Table 1), which demonstrates that the SB content data obtained from the cracking experiments are acceptable. In addition, the maximum heating temperature (600 °C) of the oil cracking experiments conducted in this study is much lower than the initial temperature of the thermal decomposition of dolostone (above 800 °C; L'vov, 2002); thus, the changes in the porosity and permeability after oil cracking were most probably not caused by the heating process but by the SB filling produced by the oil cracking. Furthermore, the porosity data of the experimental samples can be compared with the reservoir porosity data at subsurface condition, because dolostone has strong resistance to mechanical compaction in a relatively deeply buried environment (Schmoker and Halley, 1982).

The SB contents of the experimental samples injected with normal oil (0.95–1.48 %) were higher than those of the experimental samples injected with light oil (0.22–0.49 %) (Fig. 4). This is reasonable because the content of the resins and asphaltenes, which are the main components of SB, of the normal or heavy oil is higher than that of the light oil (e.g., Trejo et al., 2004; Cheshkova et al., 2019); thus, the SB produced by normal-oil cracking should be higher than the SB produced by light-oil cracking. The average residual SB content of the experimental samples injected with normal oil was 1.25 %, whereas that of the experimental samples injected with light oil was about 0.46 %

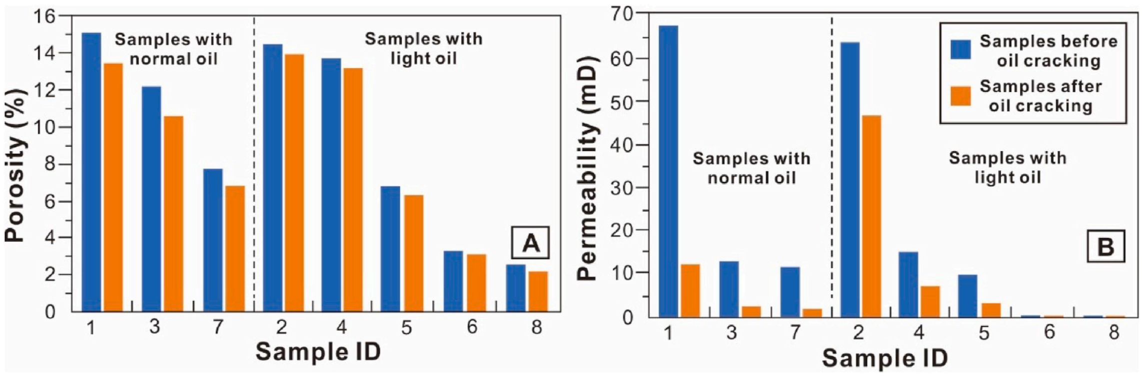

Fig. 5. A comparison of the (A) porosity and (B) permeability of the samples before and after oil cracking.

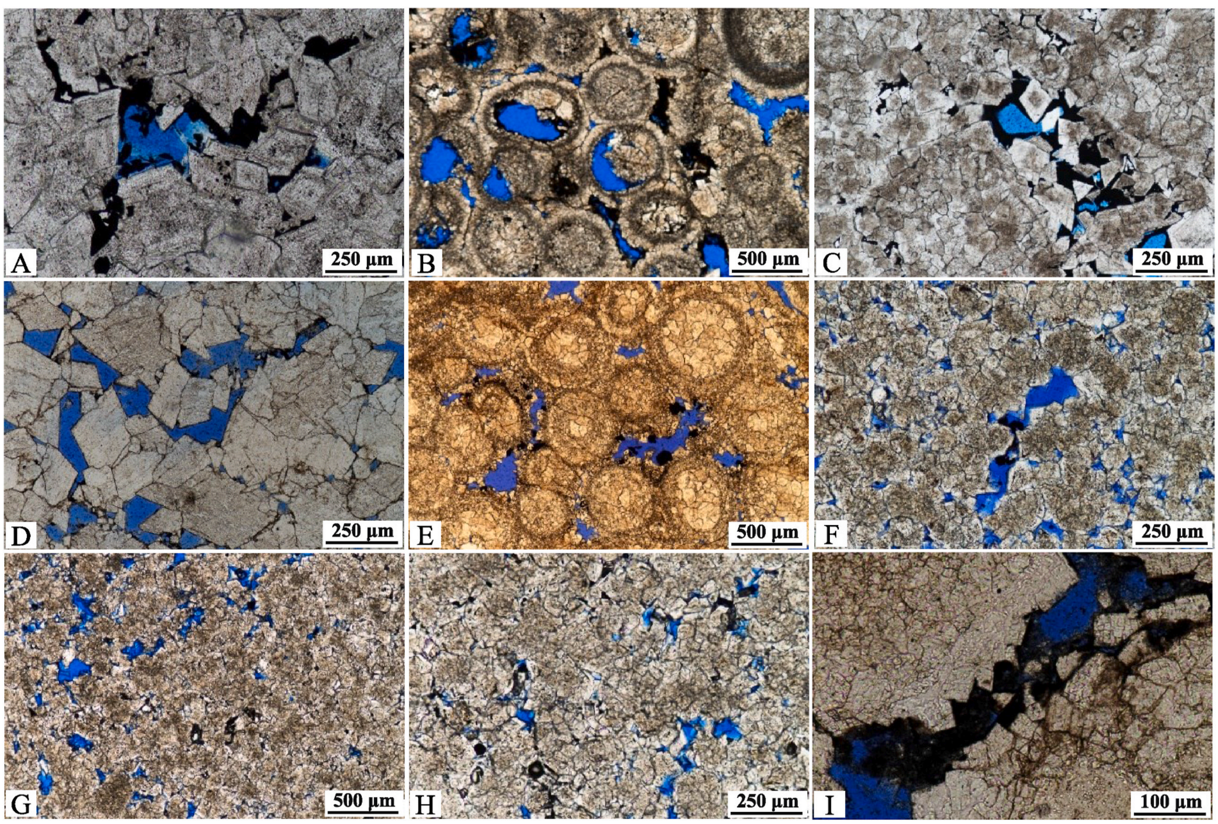

Fig. 6. Thin-section photomicrographs of dolostone samples showing the residual solid bitumen distribution after oil cracking. (A, B, C) Samples No. 1, No. 3, and No. 7 were injected with normal oil, and their solid bitumen contents were relatively high. Some of the smaller pores are filled with solid bitumen, whereas some of the larger pores are partially filled with solid bitumen. (D, E, F, G, H) Samples No. 2, No. 4, No. 5, No. 6, and No. 8 were injected with light oil, and their solid bitumen contents were relatively low. The solid bitumen is mainly developed in the smaller pores and throats (I, sample No. 4).

(excluding the two samples with abnormally low porosities).

According to the data for the reservoir samples from the Sichuan Basin, the SB content is positively correlated with the PP (Fig. 8), which is consistent with the study of the JN gas field conducted by Li et al. (2015b) and the results obtained from the oil cracking experiments conducted in this study. Whether the experimental samples were injected with normal oil or light oil, there was a positive correlation between the SB content after oil cracking and the porosity before oil cracking (PP) (Table 1). Moreover, when the PP was lower than 2.5 %, the SB content of reservoir samples was relatively low (<1.0 %); and when the PP was greater than 2.5 %, the SB content of reservoir samples rapidly increased to greater than 1.0 % (Fig. 8). The obvious difference in SB content of reservoir samples with PP lower and higher than 2.5 %, suggested oil may accumulated in the reservoir with PP greater than 2.5 %. Therefore, a SB content of 1.0 % can be used as a lower limit for identifying paleo-oil reservoirs in the Sichuan Basin.

The residual SB content obtained from the normal oil cracking experiments was approximately 1.25 %, which is slightly higher than the lower limit (1.0 %) for paleo-oil reservoirs based on actual reservoir samples from the Sichuan Basin. Because the samples used in this experiment were basically dried without water and were injected with oil under high pressure, the oil saturation of these samples exceeded 95 % (Table 1), which is much higher than the oil saturation of actual reservoirs. Therefore, it is reasonable that the SB content obtained in the experiments was slightly higher than the SB content of the actual reservoir samples. Based on data for the Tahe oilfield in the Tarim Basin in northwest China, the oil saturation of carbonate reservoirs is about 80 % (Zhao et al., 2006). Therefore, if the oil saturation of the samples used

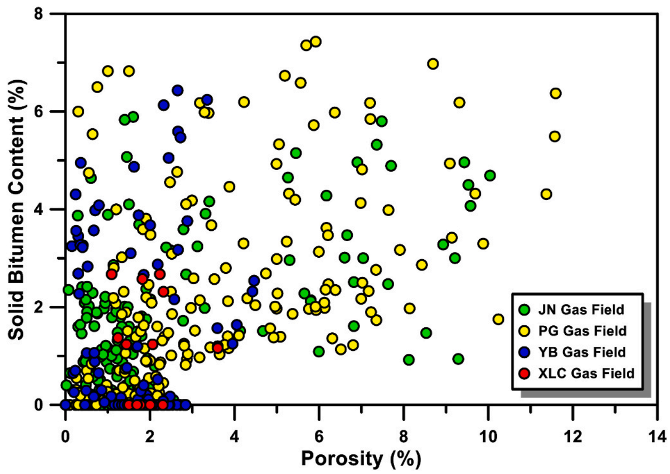

Fig. 7. Solid bitumen content versus porosity of the P_2c and T_1f samples from the Jiannan (JN), Puguang (PG), Yuanba (YB), and Xinglongchang (XLC) gas fields in the Sichuan Basin.

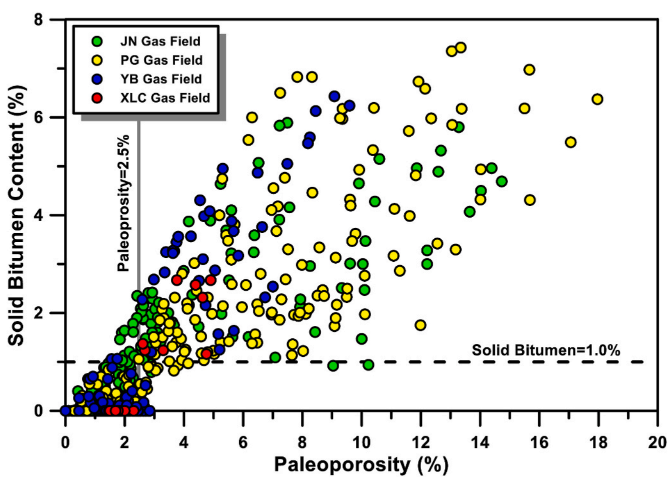

Fig. 8. Solid bitumen content versus paleoporosity of the P_2c and T_1f samples from the Jiannan (JN), Puguang (PG), Yuanba (YB), and Xinglongchang (XLC) gas fields in the Sichuan Basin.

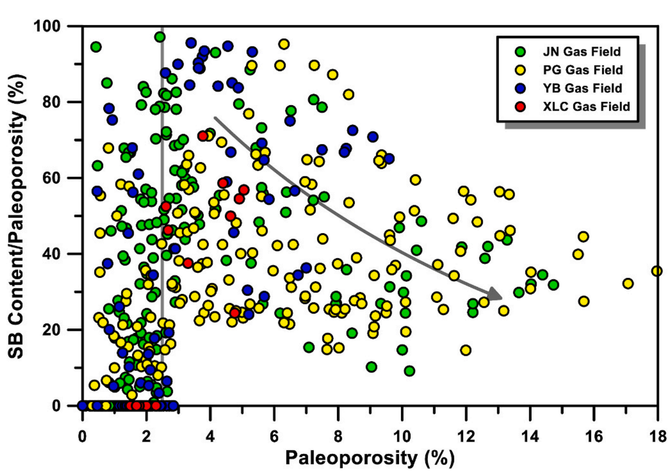

Fig. 9. Solid bitumen (SB) content/paleoporosity versus paleoporosity of the P_2c and T_1f samples from the Jiannan (JN), Puguang (PG), Yuanba (YB), and Xinglongchang (XLC) gas fields in the Sichuan Basin.

in this experiment was approximately 80 %, the residual SB content of the experimental samples injected with normal oil and light oil should have been about 1.0 % (1.25 × 0.8) and 0.4 % (0.46 × 0.8), respectively. No diagram of solid bitumen and PP was established due to limited number of the experimental samples, and the lower limit value of SB content cannot be determined similar to the method discussed above using the reservoir samples from the Sichuan Basin. However, the SB content of the experimental samples (about 1.0 %) injected with normal oil is quite similar to the lower limit value (1.0 %) of paleo-oil reservoir determined by the actual reservoir samples from the Sichuan Basin. So the SB content of the experimental samples can be used as the lower limit values of normal and light oil reservoir.

Based on the oil cracking experiments conducted in this study and the actual reservoir samples from the Sichuan Basin, the lower limits of the SB content for the identification of normal and light oil reservoirs are 1.0 % and 0.4 %, respectively. These values can also serve as reference values for the identification of paleo-oil reservoirs in other basins worldwide. However, it is essential that an evaluation of the probable types of paleo-oil be made if the lower limits of the SB content are to be used to identify paleo-oil reservoirs.

5.2. Heterogeneous distribution of the SB and reservoir effectiveness after oil cracking

As a direct product of oil cracking, the residual SB in the pores of a reservoir reduces the reservoir's porosity and permeability (Lomando, 1992; Li et al., 2015b; Wang et al., 2015; Wood et al., 2015, 2020). Previous studies of the heterogeneous distribution of SB have shown that more small pores are invaded by SB than large pores (Huc et al., 2000; Li et al., 2015b). We identified two types of heterogeneous SB distribution in the oil cracking samples and reservoirs samples from the Sichuan Basin.

Type I was the heterogeneous distribution of SB in the same reservoir interval, in which the SB tended to reside in small pores and throats. Whether the experimental samples were injected with normal or light oil, we observed relatively little SB in the larger pores (Fig. 6). In contrast, we observed more SB in the small pores and throats (Fig. 6), some of which were completely filled with SB (Fig. 6A, B, C, G, I). The decrease in permeability was much larger than the decrease in porosity after oil cracking (Fig. 5, Table 1), which also suggests that the SB was mainly left in the small pores and throats. However, the morphology of the pores cannot be accurately distinguished when using the photo statistics of porosity. As a result, the relationship between the SB content and the morphology of the pores is not discussed further in this paper.

Type II was a heterogeneous distribution of SB in different reservoir intervals. The SB saturation of the reservoir intervals with higher porosities was generally lower than that of the reservoir intervals with lower porosities (Fig. 9). We also verified this Type II heterogeneous distribution of SB using the oil cracking samples injected with normal and light oil. The SB saturations of samples No. 1 and No. 7 (porosities of 15.1 % and 7.69 % before oil injection, respectively), which were injected with normal oil, were 9.80 % and 12.35 % after oil cracking, respectively. The SB saturations of samples No. 2, No. 4, No. 5, No. 6, and No. 8 (porosities of 14.4 %, 13.68 %, 6.77 %, 3.27 %, and 2.56 % before oil injection, respectively), which were injected with light oil, were 3.40 %, 3.51 %, 6.20 %, 7.34 %, and 8.59 % after oil cracking, respectively. The SB saturations obtained in our oil cracking experiments (3.51–12.35 %; Table 1) were lower than those of the reservoir samples from the Sichuan Basin likely because a small amount of SB resided on the quartz tube wall after oil cracking, not in the pores of the samples. However, we verified the heterogeneous distribution of SB in the thin sections and the SB saturation of the experimental samples after oil cracking.

The heterogeneous distribution of SB may be related to the pressure increase during oil cracking and the differences in the viscosities of oil and gas. Solid bitumen is predominantly produced in high thermal maturity environments associated with the dry gas stage of oil cracking (Xiong et al., 2016). As the temperature increases and the oil cracks, the reservoir pressure increases (Barker, 1990; Zhang et al., 2008). In addition, according to Darcy's law ($\nu = k/\mu \times dp/dx$, where ν is the flow velocity, k is the permeability constant, μ is the fluid viscosity, and dp/dx is the hydraulic gradient), the flow velocity is inversely proportional to the viscosity of the fluid. Thus, the viscosity of the gas produced by oil cracking is much lower than that of the oil, and the flow velocity of the gas is much greater than that of the oil. Consequently, the gas will preferentially occupy the larger pores with smaller resistance, while the remaining oil will be squeezed into smaller pores and throats by overpressure. The oil will then be completely cracked to gas and the residual SB will mainly be left in the smaller pores and throats that previously contained oil. This results in the Type I heterogeneous distribution of SB. Similarly, oil-cracked gas preferentially occupies high porosity and high permeability intervals, causing the SB to preferentially reside in low porosity and low permeability intervals, leading to SB saturation differences between the different intervals (Type II heterogeneous distribution of SB).

In addition, in this study, the reservoir samples from the Sichuan Basin exhibited similar heterogeneous distributions of SB compared with the oil cracking samples. When the PP was greater than 2.5 %, the SB content of reservoir samples was greater than 1.0 %; therefore, this interval should be considered a paleo-oil reservoir (Fig. 8). Generally, the SB saturation decreased as the PP increased for samples with a PP of greater than 2.5 % (Fig. 9). The SB saturation was less than 50 % when the PP was greater than 8 %, and the SB saturation was greater than 50 % when the PP was 2.5–8.0 %. The SB contents of the reservoir samples from intervals with PPs of less than 2.5 % were generally less than 1.0 % (Fig. 8), and these intervals should be considered non-paleo-oil reservoirs. However, the SB saturations of some of the reservoir samples were 50–100 % due to the heterogeneous distribution of SB.

The heterogeneous distribution of SB resulting from oil cracking causes differential destruction of reservoir quality. Taking the Sichuan Basin as an example, its present porosity of 2.0 % is the lower limit for effective gas reservoirs (Li, 2013). When the PP was greater than 8.0 %, the residual SB saturation was less than 50 %, resulting in a present porosity that remained greater than 5.0 %. As such, it could be an effective gas reservoir after oil cracking (Fig. 10). When the PP was 2.5–8.0 %, some paleo-oil reservoirs became ineffective gas reservoirs because the present porosity would be less than 2.0 % due to the heterogeneous distribution of SB after oil cracking (Fig. 10). According to Li et al. (2015b), in an oil reservoir, the intervals with relatively low porosities and the intervals with relatively low porosities that are adjacent to intervals with relatively high porosities can become bad or ineffective gas reservoirs after oil cracking. As a result, it is necessary to evaluate the heterogeneous distribution of SB and its effect on reservoir quality by using systematic logging data to calculate the SB content (Li et al., 2020).

6. Conclusions

The SB content was found to be related to the type of cracked oil and the PP in oil cracking experiments. The SB contents of the experimental samples injected with normal oil (0.95–1.48 %) were higher than those of the experimental samples injected with light oil (0.22–0.49 %). For the experimental samples injected with the same type of oil, the SB contents of the samples with higher PPs were also higher than those of the samples with lower PPs before oil injection. The SB contents of the reservoir samples from the Sichuan Basin increased with increasing PP. When the PP was <2.5 %, the SB content was relatively low and was generally <1.0 %. When the PP was >2.5 %, the SB content increased significantly and was generally >1.0 %. The data obtained from the oil cracking experiments and for the reservoir samples from the Sichuan Basin revealed that SB contents of about 1.0 % and 0.4 % are the lower limits for normal and light oil reservoir identification, respectively.

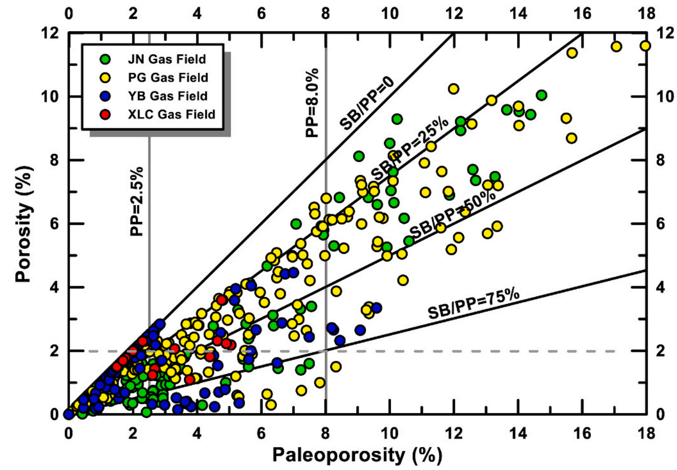

Fig. 10. Porosity versus paleoporosity of the P_2c and T_1f samples from the Jiannan (JN), Puguang (PG), Yuanba (YB), and Xinglongchang (XLC) gas fields in the Sichuan Basin. SB = solid bitumen content, PP = paleoporosity.

During oil cracking, the generated gas preferentially occupied the relatively larger pores, resulting in the residual SB tending to reside in the small pores or throats that previously contained oil, which reduced the reservoir quality and increased the reservoir heterogeneity. For the reservoir samples from the Sichuan Basin, paleo-oil reservoirs with PPs of >8.0 % can be effective gas reservoirs, while some of the paleo-oil reservoirs with PPs <8.0 % may be ineffective gas reservoirs after oil cracking. Thus, it is necessary to evaluate the heterogeneous distribution of SB and its differential destruction in gas reservoir quality after oil cracking.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.petrol.2021.109340.

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