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RESEARCH ARTICLE

Evaluation of the in-place adsorbed gas content of organic-rich shales using wireline logging data: a new method and its application

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Abstract Adsorbed gas content is an important parameter in shale gas reservoir evaluations, and its common calculation method is based on core experiments. However, in different areas, the correlations between the adsorbed gas content and well logging data might differ. Therefore, a model developed for one specific area cannot be considered universal. Based on previous studies, we studied the relationships between temperature, TOC, organic matter maturity and adsorbed gas content and revealed qualitative equations between these parameters. Then, the equations were combined to establish a new adsorbed gas content calculation model based on depth and total organic carbon (TOC). This model can be used to estimate the adsorbed gas content using only conventional well logging data when core experimental data are rare or even unavailable. The method was applied in the southern Sichuan Basin, and the adsorbed gas content results agree well with those calculated using the Langmuir isothermal model and core experimental data. The actual data processing results show that the adsorbed gas content model is reliable.

Keywords shale reservoir, adsorbed gas, well logging, temperature, pressure, TOC, depth

1 Introduction

Shale gas has attracted researchers' attention in recent decades (Zhang, 2015). Different geophysical methods can be utilized to describe the development of shale gas (Wang et al., 2016a, 2016b; Wang and Fehler, 2018a, 2018b). Advanced methods, specific to borehole geophysics, can be used to acquire formation properties during drilling (Li and Wang, 2016; Wang et al., 2016c, 2017). However, one of the most pressing geophysical exploration tasks associated with shale gas plays is to estimate the components of the gas in place. The total gas in place (GIP) in organic shales includes both adsorbed gas and free gas (Ji et al., 2017). The vast majority of natural gas occurs in micro grain surfaces in an adsorption state (20%–85%) or occurs in pores and fractures as a free gas (Zou et al., 2010). Therefore, adsorbed gas content is a key parameter in calculating shale gas reserves. The adsorption characteristics of shale gas have been studied over the last decade, mainly by experiments and molecular simulations (Clarkson et al., 2013; Mosher et al., 2013; Rexer et al., 2013; Tang et al., 2016; Tian et al., 2016; Xiong et al., 2017). Many studies on methane adsorption capacity and its controlling factors have shown that total organic carbon (TOC) is a main contributor to adsorbed gas capacity (Ross and Marc Bustin, 2009; Weniger et al., 2010; Gasparik et al., 2012; Zhang et al., 2012; Ji et al., 2015; Chen et al., 2017; Zhao et al., 2017) and that the majority of adsorbed gas fills micropores (Li et al., 2017; Zhou et al., 2018).

The adsorbed gas content can be determined from core experimental data or calculated using well logs and other data. Core experimental methods for determining adsorbed gas contents have been developed in recent years. Commonly, adsorbed gas contents can be estimated based on the Langmuir isothermal adsorption equation (Jaroniec et al., 1989); however, this indirect measuring method is affected by many factors and lacks accuracy (Li et al., 2020). There are also direct measurement methods based on on-site desorption data (Bertard et al., 1970). Based on previous studies, Li et al. (2020) proposed a new estimation method for gas-in-place content considering the release behaviors of only free gas and both adsorbed and free gasses during different stages; this method can provide accurate gas content information for cores. However, it is

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still necessary to predict the continuous gas content along a well. Log interpretation models of shale reservoir adsorbed gas content can be built by combining logging data with core experiments to predict the effective adsorption of shale gas and reduce experimental costs. In this way, calculation models of total organic carbon (TOC) and gas content have been established to predict adsorbed shale gas contents based on an isothermal adsorption and volume model. Lewis et al. (2004) presented the method of using Langmuir isotherms to calculate the adsorbed gas content, in which core analysis is required to generate a Langmuir isotherm in each field or subbasin. Cluff (2006) calculated the *in situ* resources of the Delaware Basin Barnett shale and Woodford shale using log interpretation parameters based on isothermal adsorption and volume models, but the parameters in the model were not easy to determine. Li et al. (2012) acquired Langmuir isotherm curves by performing adsorptive experiments at different temperatures and presented a new model for adsorbed gas content calculation by considering the effects of temperature, pressure, and TOC and maturity of organic matter. The correlation coefficient between the calculation results and experiments is greater than 0.9, and this research lays the foundation of continuous adsorbed gas content calculations with logging data. Ji et al. (2017) provided a method of calculating adsorbed gas contents using logging data considering TOC and shale contents. However, detailed isothermal core experiments are needed when using these methods. Based on methane isothermal adsorption experiments and gray correlation theory, Gou and Xu (2019) presented a model for calculating adsorbed gas and free gas contents. This model takes into account the differences in formation temperature, pressure, TOC, mineral composition and their effects on the shale adsorption capacity. However, because of its deep dependence on core data, only a few gas content results at several depth points were calculated. The present methods all deeply rely on local core experimental data. A global usable general model for calculating the adsorbed gas content is still needed.

In this paper, based on former studies on the relationship between *in situ* temperature and pressure, TOC and adsorbed gas content, we present a new model that quickly predicts the content of adsorbed gas in place using well logging data, which can be used with or without core experimental data.

2 Methods

2.1 Influence of the key factors and the depth-TOC model

2.1.1 Formation temperature and pore pressure model

Formation pore pressure and temperature are related to formation depth. Experience-derived equations for the relationships between pore pressure, temperature and depth are as follows:

$$P = P_0 + d\cdot \mathrm{GP}, \tag{1}$$

$$T = T_0 + d\cdot \mathrm{GT}, \tag{2}$$

where d is the true depth of a formation, m; P_0 is the pore pressure exerted by the ground fluids, MPa; GP is the pressure gradient, MPa/m; T_0 is the ground surface temperature, °C; and GT is the temperature gradient, °C/m.

In practice, the ground pressure can be neglected, i.e., $P_0 = 0$. At a depth of 30 m, temperature is not affected by the ground surface temperature. Therefore, the ground surface temperature can be selected using prior knowledge.

2.1.2 Influence of temperature

When pressure is constant (i.e., isobaric process), with increasing temperatures, the adsorption gas quantities in shale and coal seams show a similar trend (Zhao et al., 2013). According to Chen and Li (2011), the isobaric coalbed methane adsorption experimental results are shown in Fig. 1.

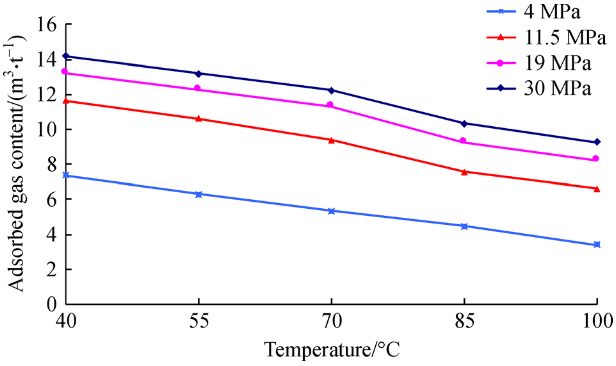

Fig. 1 Isobaric adsorption experiments in coalbeds in the eastern Ordos Basin (Chen and Li, 2011).

According to the experimental data shown in Fig. 1, when the formation pore pressure is 4.0 MPa, 11.5 MPa, 19.0 MPa, and 11.5 MPa, the relationship between temperature and adsorbed gas content can be written as the following equations:

$$V_\mathrm{a} = 9.67 - 0.065\cdot T = 9.67\cdot(1-0.006722\cdot T), \tag{3}$$

$$V_\mathrm{a} = 15.2 - 0.09\cdot T = 15.2\cdot(1-0.005921\cdot T), \tag{4}$$

$$V_\mathrm{a} = 16.78 - 0.088\cdot T = 16.78\cdot(1-0.005244\cdot T), \tag{5}$$

$$V_\mathrm{a} = 17.53 - 0.083\cdot T = 17.53\cdot(1-0.004735\cdot T), \tag{6}$$

where V_a is the adsorbed gas content, m^3/t, and T is the formation temperature, °C. The equations above can be simplified as follows:

$$V_a = V_{mp} \cdot (1 - K \cdot T), \tag{7}$$

where V_a is the adsorbed gas content, m^3/t; V_{mp} is the adsorbed gas content at 0°C under a specific pressure, m^3/t; T is the formation temperature, °C; and K is the temperature coefficient.

From Eq. (7), we can see that under a constant pressure, there is a negative linear relationship between the coalbed adsorbed gas content and temperature. With increasing temperatures, the adsorbed gas content decreases.

According to Li et al. (2012), under a constant pressure, the shale adsorbed gas content also has a negative linear correlation with temperature. The equations of the results from Li et al. (2012) can also be simplified as follows:

$$V_a = 3.441 - 0.0128 \cdot T = 3.441 \cdot (1 - 0.00372 \cdot T), \tag{8}$$

$$V_a = 2.839 - 0.0052 \cdot T = 2.839 \cdot (1 - 0.00183 \cdot T), \tag{9}$$

$$V_a = 2.754 - 0.075 \cdot T = 2.754 \cdot (1 - 0.00272 \cdot T), \tag{10}$$

$$V_a = 2.049 - 0.0057 \cdot T = 2.049 \cdot (1 - 0.00278 \cdot T). \tag{11}$$

From the analysis above, we can deduce that Eq. (7) is suitable for estimating the adsorbed content in shales.

For the temperature coefficient K, from the experimental results shown above, in coalbeds, the value of K is 0.005–0.007, while in shales, it is 0.002–0.004. The difference in K between coalbeds and shales is likely driven by TOC. There is a higher TOC in coalbeds than in shales. This means that temperature has a more substantial impact in coalbeds. By analyzing the value range of K and experimental data from Li et al. (2012), the relationship between K and TOC is shown in Eq. (12).

$$K = 0.003 \cdot \lg(100 \cdot \mathrm{TOC}). \tag{12}$$

To verify the relationship shown in Eq. (12), we set TOC = 80% under coal seam conditions and TOC = 5% under shale conditions. The temperature influence coefficients of K are 0.00571 and 0.0021, respectively, and both are within the range of the experiments. The verification results show that Eq. (12) is reliable. When the TOC content is less than 1%, we assume that temperature does not have a strong effect, and the value of K is set to a very low value rather than 0 or a negative value to ensure that it has physical meaning. The following equations containing K are reasonable.

According to Eqs. (7) and (12), in the isobaric process, the adsorbed gas content V_{ap} can be written as shown in Eq. (13).

$$V_{ap} = V_{mp} \cdot \left(1 - \lg(100 \cdot \mathrm{TOC}) \cdot k \cdot T\right), \tag{13}$$

where k is an experience constant with a value of 0.003.

2.1.3 Influence of pressure

When temperature is constant (i.e., isothermal process), changes in the adsorbed gas content with pressure follows the Langmuir isothermal model, as shown in Eq. (14) (Langmuir, 1918; Do, 1998).

$$V_a = V_{mt} \cdot \frac{p}{p + 1/b}, \tag{14}$$

where V_{mt} is the highest adsorbed gas content at a certain temperature, m^3/t; p is the pressure, MPa; and b is the affinity constant, which is a measure of the attachment of an adsorbate molecule attracted onto a surface, MPa^{-1}.

According to Eq. (14), in the isothermal process, the adsorbed gas content V_{at} can be written as Eq. (15):

$$V_{at} = V_{mt} \cdot \frac{p}{p + 1/b}. \tag{15}$$

2.1.4 Joint influence of temperature and pressure

Under isothermal and isobaric conditions, the adsorbed gas content has different laws of change. In an actual well, temperature and pressure are variable. Ideally, changes in temperature and pressure are both related to depth. Therefore, according to the analysis above, considering the influence of temperature and pressure, the adsorbed gas content calculation equation can be written as Eq. (16).

$$V_a = V_m \cdot (1 - \lg(100 \cdot \mathrm{TOC}) \cdot k \cdot T) \cdot \frac{p}{p + 1/b}, \tag{16}$$

where V_m is the adsorbed gas content at 0°C under infinite pressure. By substituting Eq. (1) and Eq. (2) into Eq. (16), the equation can be written as a function of depth as follows:

$$V_a = V_m \cdot (1 - \lg(100 \cdot \mathrm{TOC}) \cdot k \cdot d \cdot GT) \cdot \frac{d \cdot \mathrm{GP}}{d \cdot \mathrm{GP} + 1/b}. \tag{17}$$

The influences of temperature and pressure on the adsorbed gas content are opposite. There must be a certain depth at which the influences of pressure and temperature are in equilibrium. This depth is referred to as the critical depth (noted as CD). At the critical depth, the adsorbed gas content can be referred to as the balance gas content (noted as V_{bag}). For different shale formations or coal seams, CD can change. At the CD, V_{bag} can be written as follows:

$$V_{bag} = V_m \cdot (1 - \lg(100 \cdot \mathrm{TOC}) \cdot k \cdot \mathrm{CD} \cdot \mathrm{GT}) \cdot \frac{\mathrm{CD} \cdot \mathrm{GP}}{\mathrm{CD} \cdot \mathrm{GP} + 1/b}, \tag{18}$$

where CD is the depth of the maximum point of the adsorbed gas content curve, at which point the first derivative of the adsorbed gas content versus d of Eq. (17)

is 0. Then, CD can be calculated as Eq. (19).

$$\mathrm{CD} = \frac{\sqrt{K^2\cdot \mathrm{GT}^2 + b\cdot K\cdot \mathrm{GT}\cdot \mathrm{GP}}}{b\cdot K\cdot \mathrm{GT}\cdot \mathrm{GP}} - \frac{1}{b\cdot \mathrm{GP}}. \tag{19}$$

To verify the reliability of Eq. (17), assuming the conditions of shale gas reservoirs and coal seams, a numerical simulation is performed. In the shale gas reservoir, we set b = 1 MPa^{-1}, V_m = 10 m^3/t, GP = 0.01 MPa/m, GT = 0.03°C/m, and TOC = 10%. In addition, under the coal seam condition, we set b = 0.5 MPa^{-1}, V_m= 10 m^3/t, GP = 0.01 MPa/m, GT = 0.03°C/m, TOC = 80%. Under both coal and shale conditions, the temperature of the isothermal case is 0°C, and the pressure of the isobaric case is infinity. The simulation results are shown in Figs. 2 and 3.

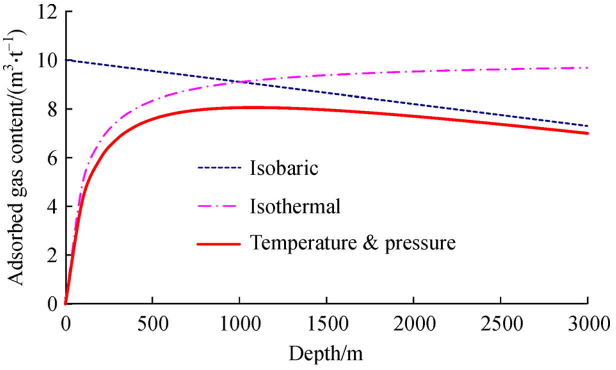

Fig. 2 Simulation results of the relationship between adsorbed gas content and depth in the shale formation.

In Figs. 2 and 3, the red dashed-dotted lines and blue dotted lines represent isothermal and isobaric processes, respectively, which indicate the effect of a single pressure or temperature, and the solid red lines are the results of simulating how adsorbed gas contents vary with depth. The curves are similar to those that describe how adsorbed gas contents change with depth in coal seams in the eastern Ordos Basin (Chen and Li, 2011; Fig. 4). In Fig. 4, the curves in different colors represent different TOC and organic matter contents. In most cases, regardless of how much TOC is present or how mature the organic matter is, the critical depth in this area is approximately 900–1600 m. Above the critical depth, the adsorbed gas content increases with depth; below the critical depth, the adsorbed gas content decreases with depth.

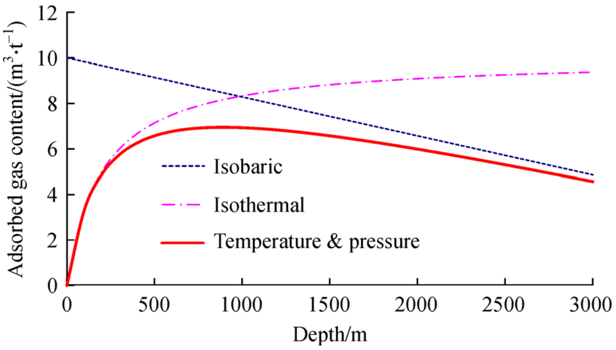

Fig. 3 Simulation results of the relationship between adsorbed gas content and depth in the coal seam.

2.1.5 Influences of TOC and organic matter maturity

In the calculation model, the adsorbed gas content under the condition of 0°C and infinite pressure V_m is needed. Through the above discussion, from Eq. (18), the relationship between V_m and V_bag is as follows:

$$V_\mathrm{m} = \frac{V_\mathrm{bag}}{C}, \tag{20}$$

where $C = (1-\lg(100\cdot \mathrm{TOC})\cdot k\cdot \mathrm{CD}\cdot \mathrm{GT})\cdot\frac{\mathrm{CD}\cdot \mathrm{GP}}{\mathrm{CD}\cdot \mathrm{GP}+1/b}$.

Organic matter maturity is one of the most significant controlling factors of pore development and structure (Gao et al., 2020). Figure 4 reveals that V_bag is related to organic

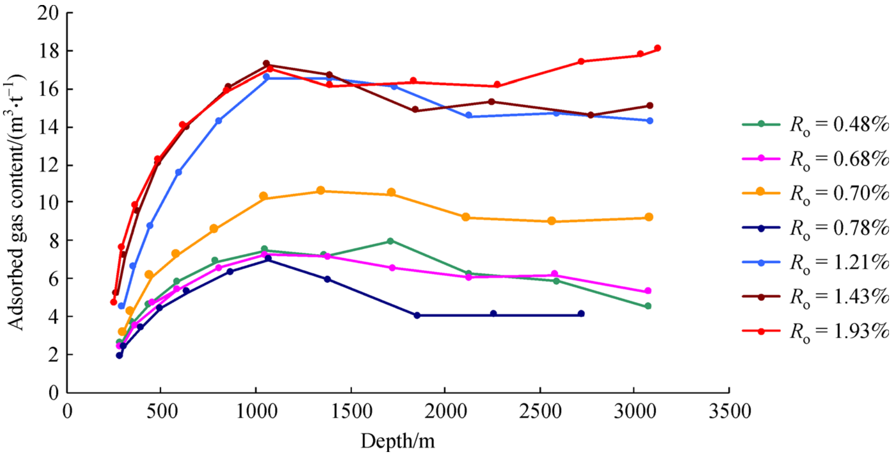

Fig. 4 The relationship between coal seam adsorbed gas contents and depth in the eastern Ordos Basin (modified from Chen and Li (2011)).

matter maturity. From the experimental data from Fig. 4, we can see that the following equation is satisfied:

$$V_{\mathrm{bag}} = (r \cdot 100 \cdot \mathrm{TOC})^n, \tag{21}$$

where $n = 0.5556$ and r is the coefficient of organic matter maturity. In shale formations, the default value of r is 3, and it can be adjusted based on experimental data or prior experience. Substitute Eq. (21) into Eq. (20), we obtain the final equation used to calculate V_{m}:

$$V_{\mathrm{m}} = \frac{(r \cdot 100 \cdot \mathrm{TOC})^n}{C}. \tag{22}$$

2.1.6 TOC-Depth model

Equations (17) and (22) constitute our depth-TOC calculation model of the adsorbed gas content. The model can be written in a final form as shown in Eq. (15).

$$V_{\mathrm{a}} = ((r \cdot 100 \cdot \mathrm{TOC})^n / C) \cdot (1 - \lg(100 \cdot \mathrm{TOC}) \cdot k \cdot d \cdot \mathrm{GT}) \cdot \frac{d \cdot \mathrm{GP}}{d \cdot \mathrm{GP} + 1/b}. \tag{23}$$

The physical meaning of this model suggests that when TOC and organic matter maturity are constant, the adsorbed gas content depends on the joint influence of temperature and pressure. Above the critical depth, the curve can be approximately viewed as a Langmuir isothermal process, in which pressure is a major factor. Below the critical depth, the formation is close to being saturated with adsorbed gas. This increase in pressure barely exerts any influence on the adsorbed gas content, and the temperature becomes the main influencing factor.

2.2 TOC calculation model

The estimation of TOC can be calculated by using well logging data (e.g., Passey et al., 1990; Tan et al., 2015; Nie et al., 2017). Then, the adsorption gas content can be calculated. The dual-V_{sh} model presented by Nie et al. (2017) is as follows.

The formation bulk model was simplified into two parts: brine-bearing shale and organics-bearing shale. Because clay minerals have a stronger ability to absorb radioactive substances than other minerals, mud rock formations always show high values of natural gamma radiation in well logs. Therefore, V_{sh} can be calculated using gamma ray logs. The classic formulas used to calculate the total V_{sh} with gamma ray logs are as follows:

$$SH = \frac{\mathrm{GR} - \mathrm{GR}_{\mathrm{min}}}{\mathrm{GR}_{\mathrm{max}} - \mathrm{GR}_{\mathrm{min}}}, \tag{24}$$

$$V_{\mathrm{sh}} = \frac{2^{gcur \cdot SH} - 1}{2^{gcur} - 1}, \tag{25}$$

where SH is the original shale content; $\mathrm{GR}_{\mathrm{min}}$ and $\mathrm{GR}_{\mathrm{max}}$ are the GR values of sandstone and pure mudstone formations, respectively; and $gcur$ is the correction coefficient, which is 2 in old strata and 3.7 in new strata.

The V_{sh} values calculated from the GR logs actually contain the volume of shales bearing both brine and organic matter. The water bearing shale content can be calculated with electrical conductivity measured through well logging. Under ideal conditions, the content of water bearing shale (V_{shw}) and effective resistivity (R_{t}) can be written as follows:

$$R_{\mathrm{t}} = R_{\mathrm{sh}} V_{\mathrm{shw}}^{-a}, \tag{26}$$

where R_{sh} is the resistivity of pure shale formations, which is always set as the R_{t} value of the formation that has the highest shale content, and a is an index that is related to the conductivity of clay minerals, gas-bearing conditions and porosity values, and its value varies between 1–2.

From Eq. (26), we can deduce the V_{sh} calculation formula using R_t logs.

$$V_{\mathrm{shw}} = 10^{\left(\frac{\lg R_{\mathrm{sh}} - \lg R_t}{a}\right)}, \tag{27}$$

where V_{shw} is the brine bearing shale content. The organic-bearing shale content (V_{sho}) can be calculated as follows:

$$V_{\mathrm{sho}} = V_{\mathrm{sh}} - V_{\mathrm{shw}}. \tag{28}$$

Then, the relative volume content of TOC (simplified as V_{TOC} in Eq. (29)) in the oil shale formation can be calculated by multiplying V_{sho} by the apparent porosity of the formation (ϕ_{tsh}).

$$V_{\mathrm{TOC}} = V_{\mathrm{sho}} \phi_{\mathrm{tsh}}. \tag{29}$$

The volume fraction can then be transformed into the mass fraction:

$$\mathrm{TOC} = \frac{V_{\mathrm{TOC}} \rho_{\mathrm{TOC}}}{\mathrm{DEN}}, \tag{30}$$

where ρ_{TOC} is the density of the organic matter (g/cm^3) and DEN is the density logging value (g/cm^3). This is the final equation used to calculate the TOC content using the dual-V_{sh} method. The calculation model has no parameters that can only be acquired from core data. Therefore, it can be used under the condition of a lack of core experimental data.

3 Application and discussion

The W well used in this study is a shale gas well in the southern Sichuan Basin. In this research area, gas-bearing organic shales mainly developed in the Qiongzhusi Group of the Lower Cambrian and Longmaxi Group of the Lower Silurian. The shales mainly consist of black-gray to gray shales, sandy shales and carbonaceous shales, with relatively developed pyrite and a large amount of graptolite. The Qiongzhusi and Longmaxi Formations showed average clastic quartz contents of 38.7% and 31.8%, respectively, consisting of pyrite and idiogenous carbonate minerals such as dolomite and calcite. These results suggest that the Qiongzhusi and Longmaxi Formations formed in a strongly reductive marine sedimentary environment, which is favorable for the accumulation and preservation of marine aquatic organic matter and provides favorable conditions for shale gas reservoir formation (Qi et al., 2011). The depth-TOC model and Langmuir isothermal model are used to calculate the adsorbed gas content in this well. The processing results are shown in Fig. 5.

In Fig. 5, the first track is depth, and tracks 2–4 show the original well logging data. Track 5 is the TOC calculated by using well logs. From the results, we can see that the lower part of this depth range has a higher TOC due to its high GR and RD. Track 6 shows comparisons of the adsorbed gas content calculated by using the depth-TOC model (pink) and Langmuir isothermal model with core experimental data (blue). The two logs of adsorbed gas contents show similar trends and agree well. Figure 6 shows the comparison of these results. The horizontal axis represents the adsorbed gas content calculated by using the depth-TOC model, and the vertical axis represents the adsorbed gas content calculated by using the Langmuir isothermal model. Throughout the whole depth interval of 1490–1540 m, the results are all within the error of 0.5 m^3/t, and the standard deviation between the adsorbed gas content calculation results of those two models is 0.158 m^3/t. This result shows that the depth-TOC model is reliable. However, there are several parameters that need to be adjusted according to prior knowledge of the research area, especially the organic matter maturity coefficient r. If core data are available, both the results of TOC and adsorbed gas content analysis can be well constrained and calibrated, thereby producing more confidence in the results.

The X Well is another shale gas well in the southern Sichuan Basin. In this well, the interval of interest is much deeper here than in the W well. At depths of 3615.5–3712.8 m, organic shales developed in the Longmaxi Group of the Lower Silurian and Wufeng Group of the Ordovician (Yan et al., 2019). The depth-TOC model is used to calculate the adsorbed gas content in this well with the same parameters of the W well. The processing results are shown in Fig. 7. In track 6 of Fig. 7, we can see that the calculated results are in good agreement with the core experimental results. This demonstrates that the method is strong and stable.

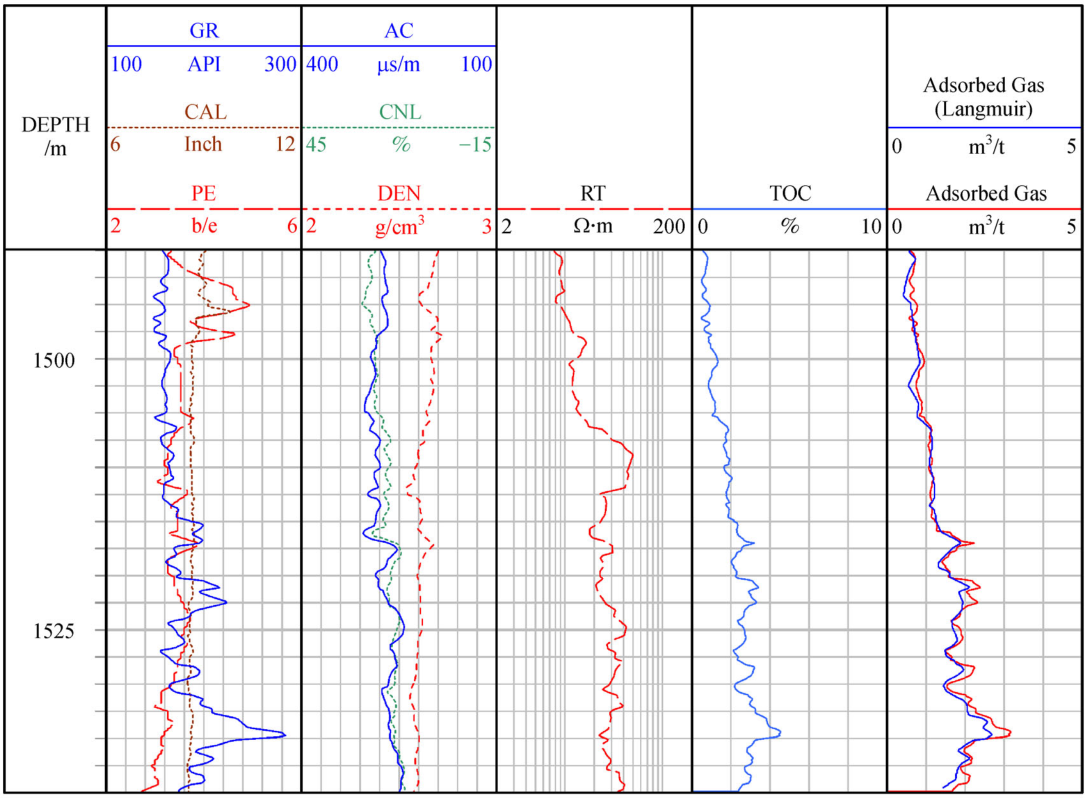

Fig. 5 The adsorbed gas content calculation results obtained using different methods of the W well.

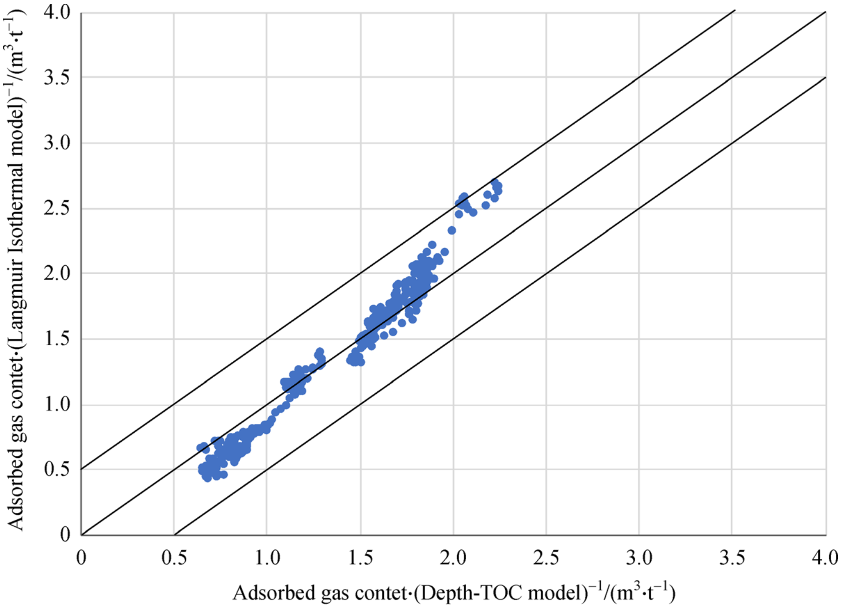

Fig. 6 Comparison of the adsorbed gas content calculation results of the W well using different methods.

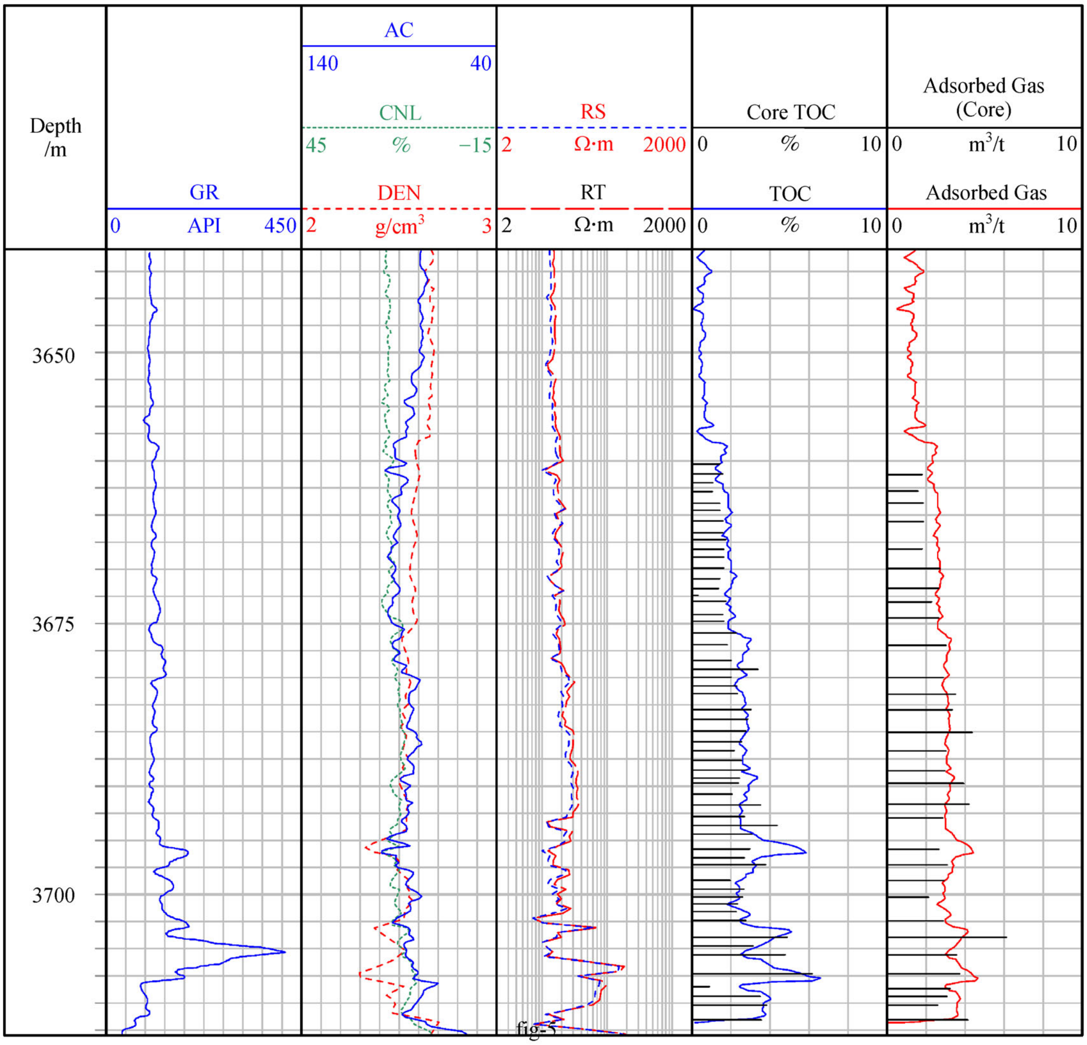

Fig. 7 The adsorbed gas content calculation results of the X well by using the depth-TOC model.

4 Conclusions

The adsorbed gas content is an important parameter for reserve evaluation in shales. Based on previous studies, we present a new method of calculating the adsorbed gas volume of shale reservoirs. The main conclusions of this paper are listed as follows.

1) Based on the Langmuir isothermal model and an isobaric model, the relationships between depth, TOC and adsorbed gas content are revealed. Depth controls pressure and temperature, and adsorbed gas content has a positive correlation with pressure and a negative correlation with temperature. When the effect of temperature on the adsorbed gas volume is considered, as the depth increases, the effect of temperature is greater. TOC plays a very important role in determining adsorbed gas contents. All the factors interactively affect the adsorbed gas content.

2) Based on the revealed relationships, a new depth-TOC model for calculating the adsorbed gas content is presented. This calculation model can be used to estimate the adsorbed gas content only with well logging data when core experimental data are unavailable. According to the analysis of the practical data processing results, the adsorbed gas content calculation model is convenient and reliable.

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