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Study on Phase Transition and Gas Deviation Coefficient of Natural Gas with High Carbon Dioxide Content

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Abstract

It is the basic research subject that analyzes and calculate the law and numerical value of phase change and gas deviation coefficient of natural gas with high-CO_2 content in the process of safe and effective development of gas reservoirs, which is obtained by high-pressure physical properties PVT (Pressure-Volume-Temperature) experiments and different calculation methods calculations. Aiming at natural gas with high-CO_2 content in the Dongfang gas field, the phase change characteristics and physical parameters of different PVT samples are simulated and tested by Chandler 3000-GL analyzer and PVT SIM software. The experimental data shows that the phase state of natural gas with different content of CO_2 has not changed in the study range. At the same time, the deviation coefficient calculated by different calculation methods (DPR, DAK, BB, HY, Gopal) are compared with the experimental data, and the applicable scope of different calculation methods are obtained. The results show that the improved Gopal has high accuracy and is suitable for the calculation of the deviation coefficient of natural gas with high-CO_2 content under high temperature and high pressure in the Dongfang gas field.

Keywords

Natural Gas, Phase Transformation, Deviation Coefficient

1. Introduction

The CO_2 content of the gas reservoirs in the Dongfang area is as high as 22.15% - 73.9%, the pressure range is 10 MPa - 100 MPa, and the temperature range is 20˚C - 200˚C. It is the important basic data for natural gas reserves calculation, numerical simulation, dynamic analysis, and formulation of reasonable development plans, which studies the influence of temperature and pressure conditions of natural gas with high-CO_2 content on the phase transformation of natural gas and PVT high-pressure physical property parameters. In this paper, the PVT experimental measurement and software are used to simulate the changes of physical property parameters of natural gas with high CO_2 content during the production process of the Dongfang gas field. At the same time, PVT experimental measurement and different calculation model methods are used to calculate the deviation coefficient of natural gas with high CO_2 content. Through the comparison of calculation results, it is concluded that the calculation method is more suitable for this area.

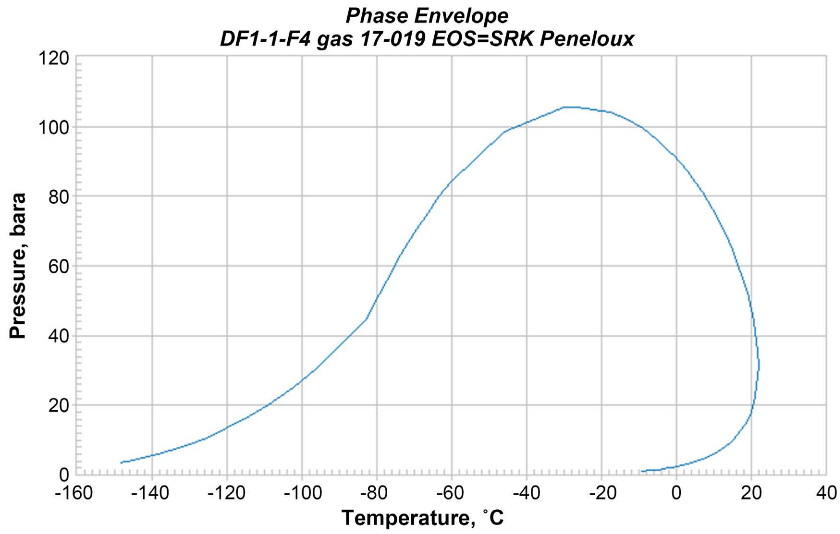
2. Physical Property Parameter Test of Natural Gas with High CO_2 Content

2.1. Phase Transition of Natural Gas with High CO_2 Content

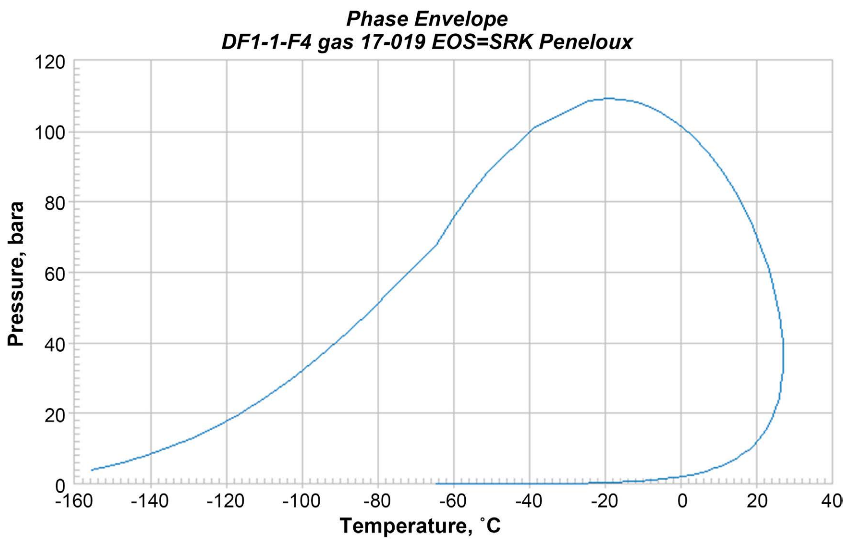
The sample was selected as the 17-019 gas sample from Well DF1-1-F4 in the Dongfang gas field, and different experimental samples were prepared with industrial pure CH_4 and CO_2 in accordance with the set ratio (**Table 1**). The physical property experiment of high temperature and high-pressure natural gas was carried out in the Chandler 3000-GL PVT analyzer [1], and the phase state analysis of different experimental samples was carried out using PVT sim software, and the corresponding phase diagram of the samples was obtained (**Figure 1**). The formation pressure of Well F is 52.880 MPa, the formation temperature is 142.0˚C, the sampling pressure of the separator is 7.316 MPa, and the sampling temperature is 38.5˚C. The gas components of the experimental samples are mainly composed of CO_2, CH_4 and N_2. Through PVT experiments, phase diagrams and vapor pressures of pure substances calculation and analysis, there is no gas-liquid phase change in the studied gas samples [2] [3].

Table 1. Component/% of different natural gas samples.

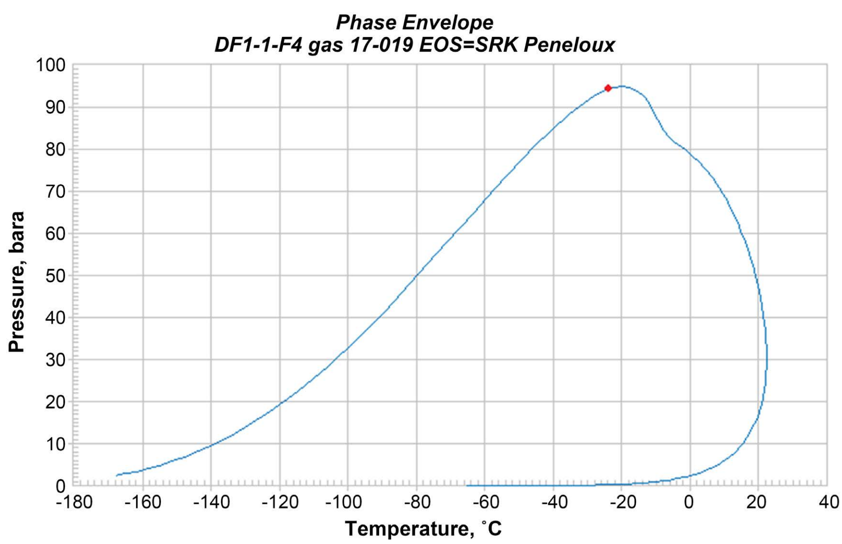
Component	I	II	III	IV
CO_2	5.52	18.02	46.99	73.58
N_2	2.66	9.01	5.93	2.8
C_1	91.22	71.21	45.99	21.84
C_2	0.37	0.94	0.64	0.41
C_3	0.09	0.33	0.2	0.26
iC_4	0.02	0.1	0.06	0.1
nC_4	0.02	0.1	0.05	0.16
iC_5	0.02	0.08	0.04	0.15
nC_5	0.01	0.05	0.02	0.12
C_6+	0.07	0.16	0.08	0.58


Phase Envelope
DF1-1-F4 gas 17-019 EOS=SRK Peneloux
Pressure, bara
Temperature, ˚C
120
100
80
60
40
20
0
-160
-140
-120
-100
-80
-60
-40
-20
0
20
40

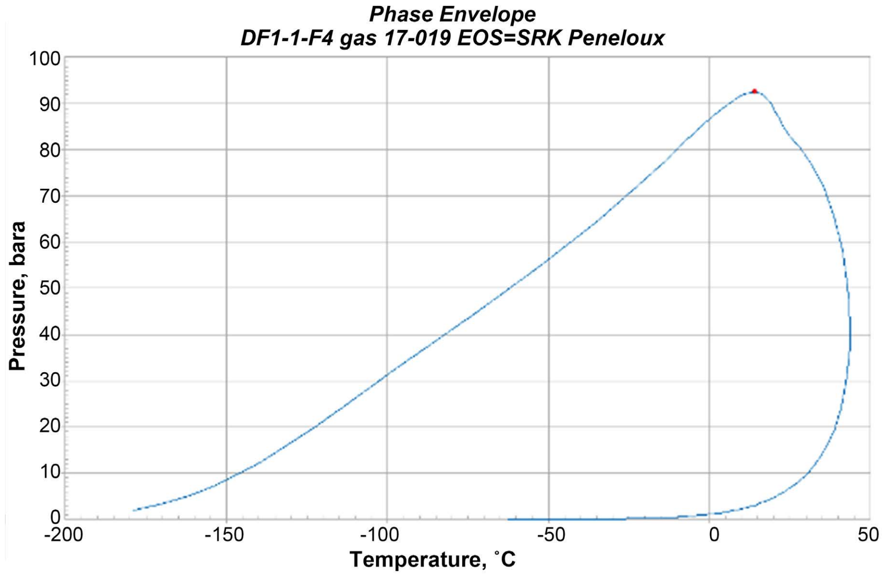
(a)


Phase Envelope
DF1-1-F4 gas 17-019 EOS=SRK Peneloux
Pressure, bara
Temperature, ˚C
120
100
80
60
40
20
0
-160
-140
-120
-100
-80
-60
-40
-20
0
20
40

(b)


Phase Envelope
DF1-1-F4 gas 17-019 EOS=SRK Peneloux
Pressure, bara
Temperature, ˚C
100
90
80
70
60
50
40
30
20
10
0
-180
-160
-140
-120
-100
-80
-60
-40
-20
0
20
40

(c)



(d)

Figure 1. Phase diagram of samples with different CO_2 contents in Well DF1-1-F4. (a) $\chi CO_2 = 5.52\%$; (b) $\chi CO_2 = 18.02\%$; (c) $\chi CO_2 = 46.99\%$; (d) $\chi CO_2 = 73.58\%$.

2.2. The Variation Law of Deviation Coefficient

The PVT experimental test of the coefficient of deviation was carried out with different preparation samples of 17-019 gas samples from well DF1-1-F4. Figure 2 and Figure 3 show the variation of the deviation coefficient with pressure at different CO_2 contents. The deviation coefficient of natural gas decreases first and then increases with pressure. Affected by the content of different gas components, the deviation coefficient of the mole fraction 10% CO_2 gas sample is the smallest when the pressure is about 16 MPa, and the deviation coefficient of the mole fraction 35% CO_2 gas sample is the smallest when the pressure is about 20 MPa. In different pressure ranges, there are differences in the influence of temperature on the deviation coefficient, which changes at about 60 MPa. When the pressure is lower than 60 MPa, the deviation coefficient increases with the rise of temperature. When the pressure is higher than 60 MPa, the deviation coefficient decreases with the rise of temperature.

According to the different sample preparation experiments of 17-019 gas samples from well DF1-1-F4, as shown in Figure 4, the gas deviation coefficient gradually decreases with the increase of CO_2 content, and the difference increases with the increase of pressure. At the same time, the higher the CO_2 content is, the greater the reduction of the gas deviation coefficient is.

3. Calculation Method of Deviation Coefficient of Natural Gas

3.1. Calibration for Non-Hydrocarbon Gases

The content of non-hydrocarbon gases in natural gas in the Dongfang gas field is relatively high, and the average content of CO_2 in PVT samples is ≥35%. The

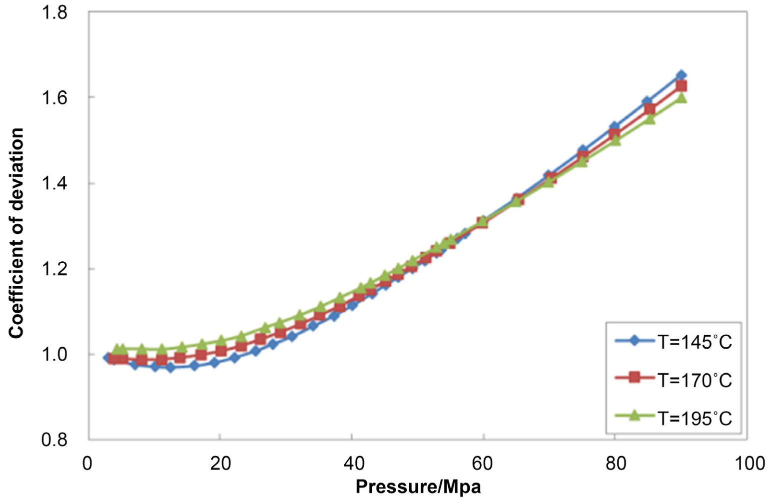


Figure 2. Variation curve of temperature, pressure and deviation coefficient with 10% CO_2 content gas sample.

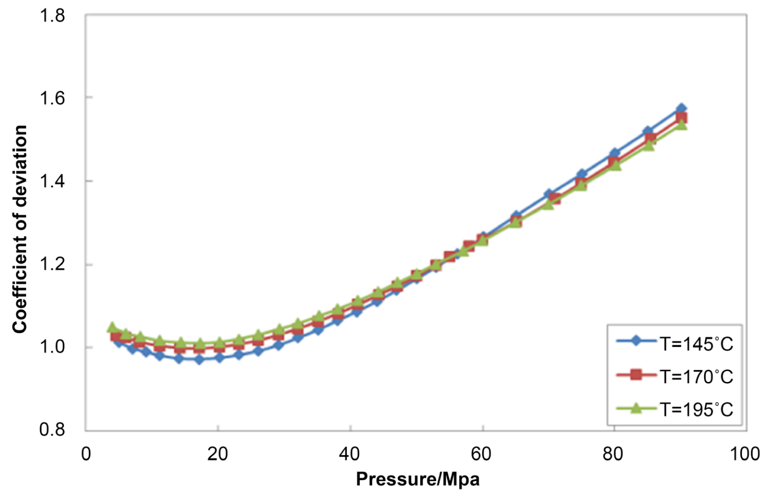


Figure 3. Variation curve of temperature, pressure and deviation coefficient with 35% CO_2 content gas sample.

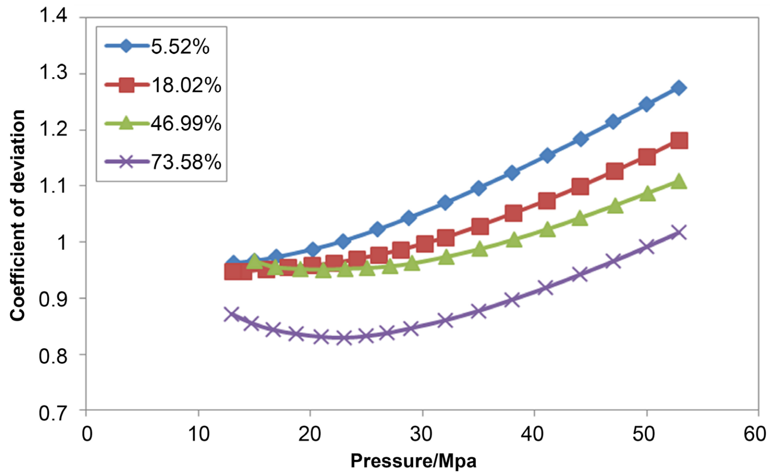


Figure 4. Variation curve of CO_2 content, pressure and deviation coefficient at 170˚C.

presence of sour natural gas in natural gas affects its critical temperature and critical pressure and changes the gas deviation coefficient of natural gas, which leads to deviations in other calculations. Therefore, it is necessary to correct the critical parameter properties of sour natural gas [4] [5]. At present, the methods for correcting the composition of critical parameters of sour gas mainly include the following methods:

3.1.1. Wichert-Aziz Correction Method [6]

$$T'_{pc} = T_{pc} - \varepsilon$$

$$p'_{pc} = \frac{p_{pc} T'_{pc}}{T_{pc} + y_{H_2S}\left(1 - y_{H_2S}\right)\varepsilon}$$

$$\varepsilon = \left[120\left(A^{0.9} - A^{1.6}\right) + 15\left(y_{H_2S}^{0.5} - A^{4.0}\right)\right] / 1.8$$

$$A = y_{H_2S} + y_{CO_2}$$

where T'_{pc} is the corrected pseudo-critical temperature, K; p'_{pc} is the corrected pseudo-critical pressure, MPa; y_{H_2S}, y_{CO_2} are the mole fractions of H_2S and CO_2 in natural gas, respectively; ε is the pseudo-critical temperature correction coefficient, dimensionless .

3.1.2. Car-Kobayshi-Burrows Correction Method [7]

The Car-Kobayshi-Burrows method takes into account the correction for N_2 content.

$$T'_{pc} = T_{pc} - 44.4 y_{CO_2} + 72.2 y_{H_2S} - 138.9 y_{N_2}$$

$$p'_{pc} = p_{pc} + 3.043 y_{CO_2} + 4.137 y_{H_2S} - 1.172 y_{N_2}$$

where $y_{CO_2}, y_{H_2S}, y_{N_2}$ are the mole fractions of CO_2, H_2S and N_2 in natural gas, respectively.

3.2. Calculation Method of Deviation Coefficient of Natural Gas

There are many ways to determine the deviation coefficient of natural gas, including the more reliable experimental measurement method, the plate method [8] and the analytical model solution method.

3.2.1. Experimental Method

The samples with 10% and 35% ratio were analyzed and tested by high temperature and high pressure PVT instrument, the relationship between the relative temperature and the relative pressure and the variation coefficient is relatively good and corresponds to the characteristics of the relationship curve of the Standing-Katz deviation coefficient (**Figure 5**, **Figure 6**).

3.2.2. Analytical Model Methods

1) Dranchuk-Purvis-Robinsion method [9]

According to the Benedict-Webb-Rubin equation of state, Dranchuk, Purvis and Robinson converted the deviation coefficient into a function of reduced

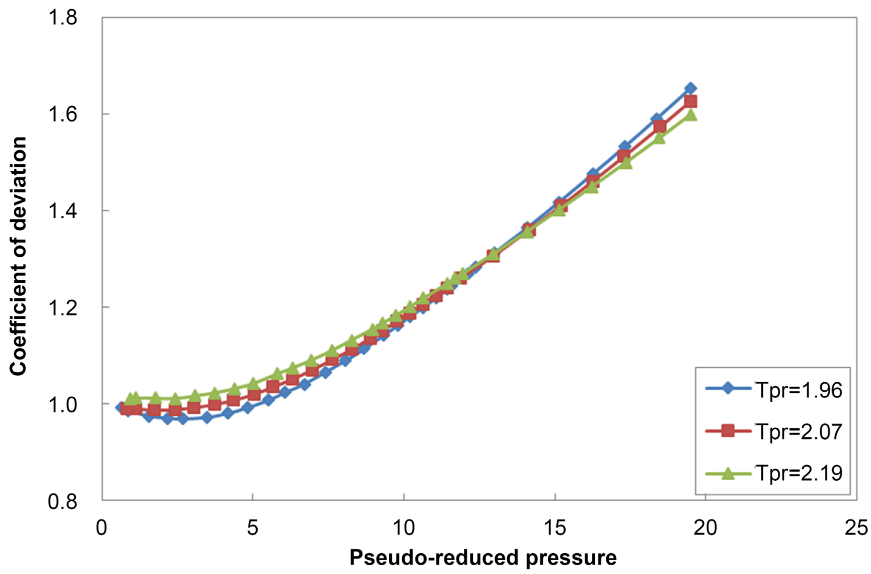


Figure 5. Variation curve of 10% pseudo-reduced temperature, pseudo-reduced pressure and deviation coefficient.

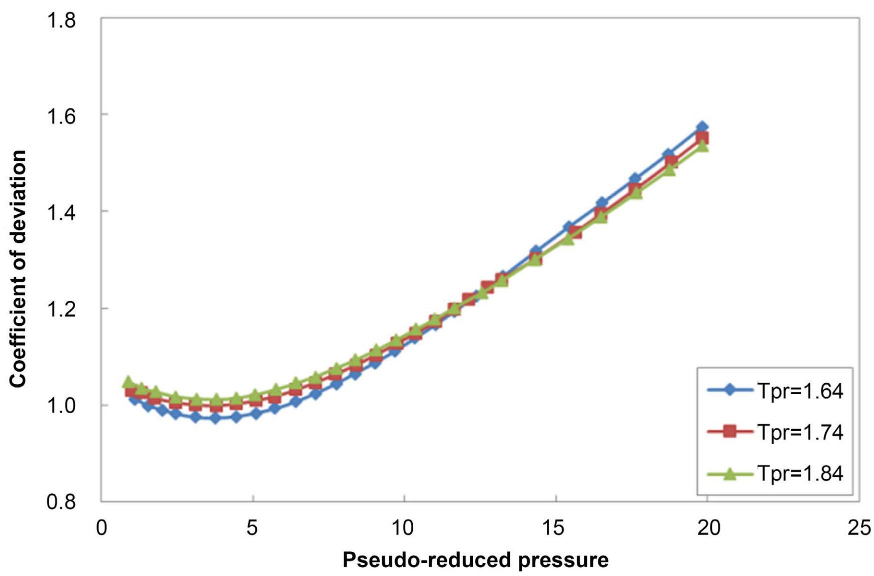


Figure 6. Variation curve of 35% pseudo-reduced temperature, pseudo-reduced pressure and deviation coefficient.

pressure and reduced temperature, and in 1974 derived an empirical formula containing 8 constants, which is in the form:

$$Z = 1 + \left| A_1 + \frac{A_2}{T_{pr}} + \frac{A_3}{T_{pr}^3} \right| \rho_r + \left| A_4 + \frac{A_5}{T_{pr}} \right| \rho_{pr}^2 + \left| \frac{A_5 A_6}{T_{pr}} \right| \rho_r^5 - \frac{A_7}{T_{pr}^3} \rho_r^2 \left(1 + A_8 \rho_r^2\right) \exp\left(-A_8 \rho_r^2\right)$$

$$\rho_r = \frac{0.27 p_{pr}}{Z T_{pr}}$$

whrere A_i is a given coefficient, A_1 = 0.31506237, A_2 = −1.04670990, A_3 = −0.57832729, A_4 = 0.53530771, A_5 = −0.61232032, A_6 = −0.10488813, A_7 = 0.68157001, A_8 = 0.68446549;

p_{prv} is the pseudo-reduced pressure, dimensionless;

T_{pr} is the pseudo-reduced temperature, dimensionless.

2) Dranchuk-Abu-Kassem method [10]

This model is calculated in the same way as Dranchuk-Purvis-Robinson, but the relative density is calculated using Newton iteration from:

$$\begin{aligned} Z = & \left| A_1 + \frac{A_2}{T_{pr}} + \frac{A_3}{T_{pr}^3} + \frac{A_4}{T_{pr}^4} + \frac{A_5}{T_{pr}^5} \right| \rho_r + \left| A_6 + \frac{A_7}{T_{pr}} + \frac{A_8}{T_{pr}^2} \right| \rho_r - \left| \frac{A_7}{T_{pr}} + \frac{A_8}{T_{pr}^2} \right| \rho_r^5 \\ & - A_9 \left| \frac{A_7}{T_{pr}} + \frac{A_8}{T_{pr}^2} \right| \rho_r^5 + \frac{A_{10}}{T_{pr}^3} \rho_r^2 \left(1 + A_{11}\rho_r^2\right) \exp\left(-A_{11}\rho_r^2\right) + 1 \end{aligned} \quad (3)$$

where A_1 = 0.32650, A_2 = −1.07000, A_3 = −0.5339, A_4 = 0.01570, A_5 = −0.05165, A_6 = 0.5475, A_7 = −0.7361, A_8 = 0.18440, A_9 = 0.10560, A_{10} = 0.6134, A_{11} = 0.7210.

3) Brill-Beggs method [11]

$$Z = A + \frac{1-A}{e^B} + Cp_r^D$$

where A, B, C and D are functions of the pseudo-reduced pressure and the pseudo-reduced temperatures.

$$A = 1.390\left(T_{pr} - 0.920\right)^{0.5} - 0.360T_{pr} - 0.101$$

$$B = \left(0.62 - 0.23T_{pr}\right)p_{pr} + \left(\frac{0.066}{T_{pr} - 0.86} - 0.037\right)p_{pr}^2 + 0.132 \times 10^{-9\left(T_{pr}-1\right)} p_{pr}^6$$

$$C = 0.132 - 0.32\lg\left(T_{pr}\right)$$

$$D = 10^{\left(0.3106 - 0.49T_{pr} + 0.1824T_{pr}^2\right)}$$

4) Hall-Yarborough method [12]

The method is based on the Starling-Carnahan equation of state, and the following relationship is obtained by fitting the Standing-Katz plate:

$$Z = 0.06125\left(\frac{p_{pr}}{\rho T_{pr}}\right)\exp\left[-1.2\left(1 - \frac{1}{T_{pr}}\right)^2\right]$$

ρ_r is the *pseudo-reduced* density, which can be obtained from the following formula by the Newton iteration method:

$$\begin{aligned} & \frac{\rho_r + \rho_r^2 + \rho_r^3 - \rho_r^4}{\left(1-\rho_r\right)^3} - \left| \frac{14.76}{T_r} + \frac{9.76}{T_{pr}^2} + \frac{4.58}{T_{pr}^3} \right| \rho_r^2 \\ & + \left| \frac{90.7}{T_{pr}} + \frac{2422}{T_{pr}^2} + \frac{42.4}{T_{pr}^3} \right| \rho_r^{2.18 + 2.82/T_{pr}} \\ & - 0.06125\left(\frac{p_{pr}}{T_{pr}}\right)\exp\left[-1.2\left(1 - \frac{1}{T_{pr}}\right)^2\right] = 0 \end{aligned}$$

5) Gopal method

Gopal fits the curve segment of the Standing-Katz gas deviation coefficient chart with a straight line equation:

$$Z = p_{pr}\left(AT_{pr} + B\right) + CT_{pr} + D$$

According to the range of p_{pr} and T_{pr}, different parameters are used to calculate the Z value (**Table 2**).

4. Deviation Coefficient Calculation

4.1. Computational Comparison

By applying different methods, the deviation coefficient of natural gas in the Dongfang Area is obtained, and the error analysis is carried out. It can be seen from **Figure 7** that under different pressure ranges, the calculation results of different methods have large errors, and the average errors are all >10%. Taking the relative error calculated by the DAK method as an example (**Figure 8**), it also shows that the error distribution is extremely uneven, which brings difficulties to the accurate calculation of the deviation coefficient.

The expression coefficient in the Gopal method is obtained from the experimental data of Gopal's own research block, so there is a deviation in the calculation of the deviation coefficient in the Dongfang area. According to the high temperature, high pressure, high CO_2 characteristics of the Dongfang area, using the pseudo-reduced pressure and the pseudo-reduced temperature to perform a piecewise multiple regression, the fitting empirical formula of each piece is obtained (**Table 3**), and the error analysis effect is good (**Figure 9**).

Table 2. Gopal formula parameters.

pseudo-reduced pressure	pseudo-reduced temperature	parameters A, B, C, D			
		A	B	C	D
0.2 - 1.2	1.05 - 1.2	1.6643	−2.2114	−0.3647	1.4385
	1.2 - 1.4	0.5222	−0.8511	−0.0364	1.049
	1.4 - 2.0	0.1391	−0.2988	−0.0007	0.9969
	2.0 - 3.0	0.0295	−0.0825	−0.0009	0.9967
1.2 - 2.8	1.05 - 1.2	−1.357	−1.4942	1.8315	4.7
	1.2 - 1.4	0.1717	−0.3232	−0.5869	0.1229
	1.4 - 2.0	0.0984	−0.2053	−0.0621	0.858
	2.0 - 3.0	0.0211	−0.0527	−0.0127	0.9549
2.8 - 5.4	1.05 - 1.2	−0.3278	−0.4752	1.8223	−1.9036
	1.2 - 1.4	−0.2521	0.3871	1.6027	−1.6635
	1.4 - 2.0	−0.0284	0.0625	0.4714	0.0011
	2.0 - 3.0	0.0041	0.0039	0.0607	0.7927

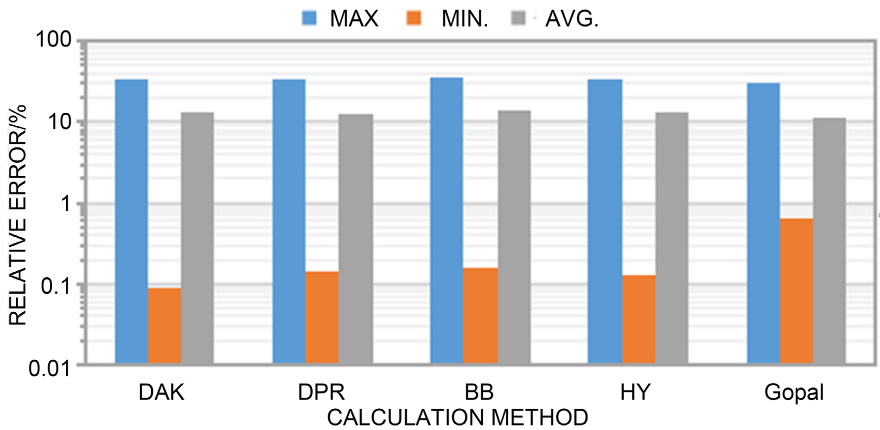


Figure 7. Errors in the results of different deviation coefficient calculation methods.

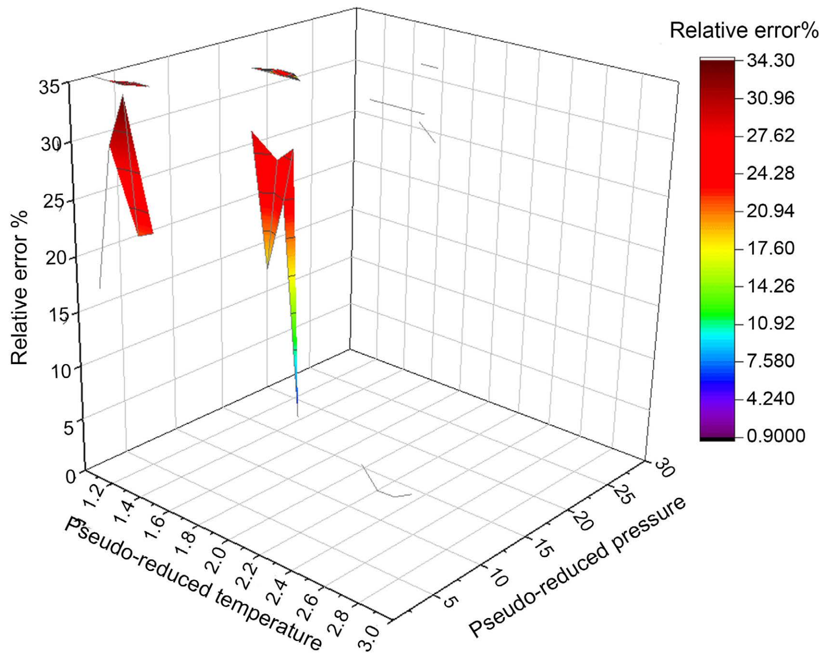


Figure 8. DAK calculation relative error distribution.

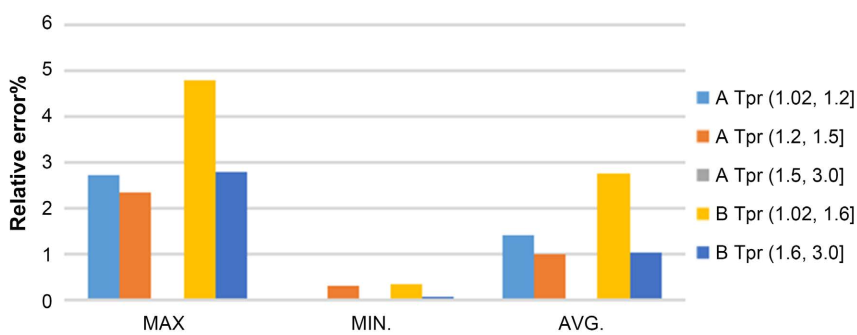


Figure 9. Relative error diagram of the calculation results of the improved Gopal formula.

4.2. Sample Calculation Verification

By selecting 4 samples of natural gas in the research field, different deviation

Table 3. Gopal experience improvement formula.

p_{pr}	T_{pr}	Expression formula
A (2.0, 5.4]	(1.02, 1.2]	$Z = 0.553 p_{pr} T_{pr} - 1.0338 p_{pr} + 2.0943$
	(1.2, 1.5]	$Z = 1.1527 p_{pr} T_{pr} - 1.5183 p_{pr} - 3.3389 T_{pr} + 5.2344$
	(1.5, 3.0]	$Z = 0.0258 p_{pr} T_{pr} - 0.042 p_{pr} + 0.8797 T_{pr} - 0.6195$
B (5.4,30.0)	(1.02, 1.6]	$Z = 0.0202 p_{pr} T_{pr} - 0.0002 p_{pr} + 0.0227 T_{pr} + 0.741$
	(1.6, 3.0]	$Z = 0.046 p_{pr} T_{pr} - 0.0503 p_{pr} - 0.4054 T_{pr} + 1.5685$

Table 4. Calculation comparison of natural gas PVT blind samples.

Blind sample number		Ⅰ	Ⅱ	Ⅲ	Ⅳ
	p_{pr}	2.615	11.725	11.565	19.727
	T_{pr}	1.34	1.85	1.79	1.94
Gas deviation coefficient	Experimental value	0.8242	1.2397	1.2224	1.5564
	DAK	0.6744	1.2278	1.2236	1.6466
	DPR	0.6737	1.2297	1.2249	1.6444
	BB	0.6974	1.2424	1.2328	1.7670
	HY	0.6756	1.2305	1.2270	1.6428
	Improved Gopal	0.8291	1.2259	1.2128	1.5492
Relative error/%	DAK	18.17	0.96	0.09	5.79
	DPR	18.26	0.81	0.20	5.66
	BB	15.39	0.21	0.85	313.53
	HY	18.03	0.74	0.37	5.55
	Improved Gopal	0.60	1.11	0.79	0.46

coefficient methods were used for calculation. The DAK, DPR, BB, HY and LXF methods have great limitations, and the calculated values within the distribution range of $1.7 < T_{pr} < 2.2$ and $9 < p_{pr} < 16$ have good accuracy. However, for the wide distribution of T_{pr} and p_{pr} in the research field, the calculation effect cannot be satisfied. By the Gopal method, the calculation error is smaller and has a good effect (Table 4).

5. Conclusions

1) Within the research range, the gas-liquid phase transition did not occur in the natural gas with high-CO_2 content during the experimental changes of temperature and pressure.

2) As the pressure increases, the deviation coefficient of natural gas first decreases and then increases, and the minimum pressure changes according to the CO_2 content. When the pressure is the same, the deviation coefficient changes with the temperature change law. When the pressure is less than 60 MPa, the deviation coefficient increases with the rise of temperature. When the pressure is

more than 60 MPa, the deviation coefficient decreases with the rise of temperature.

3) Different deviation coefficient calculation methods are used to calculate the deviation coefficient of natural gas with high-CO_2 content in the Dongfang area, and the calculation results have a large deviation. Through the improved Gopal method, the calculation effect is good.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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