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Predicting the compressibility factor of natural gases containing various amounts of CO_2 at high temperatures and pressures

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In recent years, many natural gas reservoirs have been discovered with varying CO_2 contents, many of which are at supercritical conditions. Calculation of compressibility factors for such reservoirs is important. Therefore, this research presents an extensive review of the various methods to calculate the compressibility factor for different natural gases containing CO_2 at various temperatures and pressures. It also provides a comprehensive evaluation of the accuracy of well-known and recently published mixing rules, as well as various Z-factor correlations. Finally, a set of new correlations is presented to calculate the gas compressibility factor with reasonable accuracy. The Z-factor from the proposed correlations, as well as the PR and SRK equations of state are examined against several measured Z-factors for natural gases at supercritical conditions. The proposed correlations have a correlation coefficient of 96% and can be used to calculate compressibility at high pressures and temperatures.

Key words: CO_2, natural gas mixtures, Z-factor, compressibility factor, correlations.

INTRODUCTION

Recently, several natural gas reservoirs containing varying amounts of CO_2 have been discovered in different parts of the world at supercritical conditions. Calculations of Z-factor, density and thermal conductivity of these gases are challenging. These properties are required for the evaluation and planning of CO_2 injections, as well as the design of surface facilities and pipelines (Elsharkawy et al., 2015; Khosravi et al., 2018; Liu et al., 2019). Natural gases with a high CO_2 concentration are highly utilized in gas injection processes to improve oil recovery performance. The gas compressibility factor (often called the Z-factor) is a thermodynamic property that is usually measured as an integral part of any PVT study using reservoir gas samples. Occasionally, samples become difficult and/or experimental data is unreliable, expensive, and/or time consuming. Hence, mathematical tools such as equations of state, corresponding state methods, or empirical correlations are used instead. In many cases, the estimation of the Z-factor of natural gases containing CO2 at supercritical conditions by empirical correlations is subject to significant error.

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After nearly 70 years, the Standing-Katz (1942) chart (SK) is still the core source of Z-factor calculation for natural gases (Standing and Katz, 1942). Based on the theory of corresponding states, the Z-factor in the SK chart is related to the reduced pressure (P_r) and temperature (T_r). This theory simply states that all gases have the same Z-factor at a given T_r and P_r. The reduced pressure and reduced temperature are defined as the pressure and temperature divided by their critical values. The critical pressure and critical temperature (P_c & T_c) of pure components are well known and well documented (Elsharkawy et al., 2001; Elsharkawy, 2004). However, natural gases are multi-component systems containing hydrocarbon and non-hydrocarbon components. Hence, pseudo-critical pressure (P_{pc}) and pseudo-critical temperature (T_{pc}) of natural gas mixtures are needed for the calculation of the reduced pressure and reduced temperature. Nonetheless, there is no agreement on the method to calculate the P_{pc} and T_{pc}. Several mixing rules have been proposed to calculate the pseudo-critical pressure and pseudo-critical temperature of the natural gas mixtures. Thus, in the coming sections, a review of various mixing rules, Z-factor correlations, and equations of state are conducted. This is done to provide an analysis on the accuracy of said correlations on the calculation of Z-factor for natural gas mixtures containing various CO_2 contents at supercritical conditions. The Z-factor correlations are examined using various mixing rules to provide 60 different approaches for Z-factor calculation. Furthermore, a new set of correlations is proposed and examined versus the PR and SRK equations of state for natural gas-CO_2 mixtures at supercritical conditions.

THEORY (LITERATURE REVIEW)

Mixing rules

Kay Mixing Rule (Kay, 1936)

In 1936, Kay introduced the concept of P_{pc} and T_{pc} which can be used in place of the true critical values for hydrocarbon mixtures (Kay, 1936). The Kay mixing rule is expressed by:

$$P_{pc} = \sum y_i \; P_{ci} \qquad (1)$$

$$T_{pc} = \sum y_i T_{ci} \qquad (2)$$

Where, y_i is the mole fraction of the component i and Pc_i and T_{ci} are the critical pressure and critical temperature, respectively. Then, the pseudo-reduced properties (P_r, T_r) are expressed by:

$$P_r = P/P_{pc} \qquad (3)$$

$$T_r = T/T_{pc} \qquad (4)$$

Where, P and T are the system pressure and temperature, respectively.

Stewart-Burkhardt-VOO (SBV) mixing rule

Since the SK compressibility chart was prepared from mixtures of methane with propane, ethane, butane, and natural gases, with a molecular weight below 40, Stewart-Burkhardt-VOO (Stewart et al., 1959) proposed the following mixing rule:

$$J = \left(\tfrac{1}{3}\right)\left[\sum y_i \left(\tfrac{T_c}{P_c}\right)_i\right] + (2/3)\left[\sum y_i (T_c/P_c)_i^{0.5}\right]^2 \qquad (5)$$

$$K = \sum\left[y_i (T_c/P_c^{0.5})_i\right] \qquad (6)$$

$$T_{pc} = K^2/J \qquad (7)$$

$$P_{pc} = T_{pc}/J \qquad (8)$$

Sutton modification of SBV (SSBV)

Sutton (1985) observed that a large deviation in Z-factor occurs in gases with high contents of C_{7+}, therefore he proposed modifying the SBV mixing rule to minimize this deviation as follows (Sutton, 1985):

$$F_j = (1/3)\left[y\left(\tfrac{T_c}{P_c}\right)\right]_{c7+} + (2/3)\left[y(T_c/P_c)^{0.5}\right]_{c7+}^{2} \qquad (9)$$

$$E_j = 0.6081F_j + 1.1325F_j^2 - 14.004F_{jyc7+} + 64.434F_{jyc7+}{}^2 \qquad (10)$$

$$E_k = (T_c/P_c^{0.5})_{c7+}\left(0.3129_{yc7+} - 4.8156y_{c7+}^2 + 27.3751y_{c7+}^3\right) \qquad (11)$$

$$J' = J - Ej \qquad (12)$$

$$.K' = K - E_k \qquad (13)$$

$$.T_{pc} = K'^2/J' \qquad (14)$$

$$.P_{pc} = T_{pc}/J' \qquad (15)$$

Corredor et al. mixing rule

Corredor et al. treated the non-hydrocarbon components and the C_{7+} fractions differently than Sutton (1985), (Corredor et al., 1992). Their mixing rule has the following form:

$$J = \alpha_0 + \sum \alpha_i y_i \left(\tfrac{T_c}{P_c}\right)_i + \alpha_4 \sum y_i \left(\tfrac{T_c}{P_c}\right)_j + \alpha_5\left[\sum y_i (T_c/P_c)_i\right]^2 + \alpha_6(y_{c7+}M_{c7+}) + \alpha_7(y_{c7+}M_{c7+})^2 \qquad (16)$$

$$K = \beta_0 + \sum \beta_i y_i \left(\tfrac{T_c}{P_c^{0.5}}\right)_i + \beta_4 \sum y_j \left(\tfrac{T_c}{P_c^{0.5}}\right)_j + \beta_5\left[\sum y_j (T_c/P_c^{0.5})_j\right]^2 + \beta_6(y_{c7+}M_{c7+})\beta_7(y_{c7+}M_{c7+})^2 \qquad (17)$$

Where, $y_i \in \{yH_2S, y_{CO_2}, y_{N_2}\}$ and $y_i \in \{y_{C_1}, y_{C_2}, \dots\dots, y_{C_6}\}$, α and β are constants.

Piper et al. mixing rule

Piper et al. proposed a modified version of Corredor et al. mixing rule. The difference between the Corredor et al. mixing rule and Piper et al. mixing rule is that each method has different values for the coefficients α and β (Piper et al., 1993).

Elsharkawy's mixing rule

Due to a large deviation between the Z-factor calculation from the SK chart in the presence of non-hydrocarbon components and heptane plus fractions in natural gas and measured Z-factor values, Elsharkawy proposed a simple mixing rule (Elsharkawy, 2004). This mixing rule divided the gas into three parts: a non-hydrocarbon part (such as N_2, CO_2, and H_2S), a hydrocarbon part (such as C_1 to C_6), and a heptane plus part. Knowing the properties of C_{7+}, P_c, T_c, and Mw, the parameters J_{inf} and K_{inf} are calculated as follows:

$$J_{inf} = \alpha_0 + \left[\alpha_1\left(\frac{y_iT_c}{P_c}\right)\right]H_2S + \left[\alpha_2\left(\frac{y_iT_c}{P_c}\right)\right]CO_2 + \left[\alpha_3\left(\frac{y_iT_c}{P_c}\right)\right]_{N_2} + \left[\alpha_4\sum y_i\left(\frac{T_c}{P_c}\right)\right]_{C_1-C_{6+}} + [\alpha_5(y_iMw)]_{C7+} \quad (18)$$

Where, $\alpha_0 = 0.036983$, $\alpha_1 = 1.043902$, $\alpha_2 = 0.894942$, $\alpha_3 = 0.792231$, $\alpha_4 = 0.882295$,
$\alpha_5 = 0.018637$.

$$K_{inf} = \beta_0 + \left[\beta_1\left(\frac{y_iT_c}{P_c^{0.5}}\right)\right]_{H_2S} + \left[\beta_2\left(\frac{y_iT_c}{P_c^{0.5}}\right)\right]_{CO_2} + \left[\beta_3\left(\frac{y_iT_c}{P_c^{0.5}}\right)\right]_{N_2} + \left[\beta_4\Sigma y_i\left(\frac{T_c}{P_c^{0.5}}\right)\right]_{C_1-C_{6+}} + [\beta_5(y_iMw)]_{C7+} \quad (19)$$

Where, $\beta_o = -0.7765003$, $\beta_1 = 1.0695317$, $\beta_2 = 0.9850308$, $\beta_3 = 0.8617653$, $\beta_4 = 1.0127054$, $\beta_5 = 0.4014645$.

This mixing rule has the advantage of eliminating the need for estimating the critical properties of heptane plus fractions. The T_{pc} and P_{pc} are calculated as follows:

$$T_{pc} = \frac{K_{inf}^2}{J} \quad (20)$$

$$P_{pc} = \frac{T_{pc}}{J_{inf}} \quad (21)$$

The T_{pr} and P_{pr} are calculated following the standard method.

$$P_{pr} = \frac{P}{P_{pc}} \quad (22)$$

$$T_{pr} = \frac{T}{T_{pc}} \quad (23)$$

In this research all previously mentioned mixing rules are examined. The pseudo-critical properties of various natural gases containing different amounts of carbon dioxide are calculated using the previously mentioned methods.

Effect of non-hydrocarbon components

Natural gases frequently contain CO_2 and H_2S that deteriorate the accuracy of the calculated Z-factor. Wichert and Aziz presented a method to correct the T_{pc} and P_{pc} for natural gases in the presence of H_2S and CO_2 (Wichert and Aziz, 1972). The correction factor is:

$$\epsilon = 120(A^{0.9} - A^{1.6}) + 1.5(B^{0.5} - B^4) \quad (24)$$

Where A = yH_2S + yCO_2 and B =y H_2S in the gas mixture. The corrected P'_{pc} and T'_{pc} are:

$$T'_{pc} = T_{pc} - \epsilon \quad 25)$$

$$P'_{pc} = P_{pc}T'_{pc}/[T_{pc} + B(1-B)\,\epsilon] \quad (26)$$

Z-factor correlations

Many attempts have been made to convert the SK chart into a simplified mathematical form. In this section, all published methods will be studied to assess their accuracy and application for natural gas-CO_2 mixtures, these methods are:

(1) Papay (1968),
(2) Hall-Yarborough (1973 and 1974),
(3) Dranchuk-Abu-Kassem (1975),
(4) Dranchuk-Purvis-Robinson (1974),
(5) Hankinson -Thomas-Phillips (1969)
(6) Londono et al. (2005),
(7) Al-Anazi et al (2010),
(8) Bahadori et al. (2010)
(9) Kamyab et al. (2014),
(10) Aziz et al (2010), 11) Heideryan et al (2010)
(11) Shokir et al. (2012),
(12) Kamari et al. (2013)
(13) Fatoorehchi et al. (2014)
(14) Fayazi et al. (2014),
(15) Ehsan and Nemati (2015),
(16) Mohagheghian and Bahadori (2015)
(17) Khosravi et al. (2018)

Papay method

Papay proposed a simplified equation for calculating the compressibility factor (Papay, 1968):

$$Z = 1 - \frac{p_{pr}}{T_{pr}}\left[0.36748758 - 0.04188423\left(\frac{p_{pr}}{T_{pr}}\right)\right] \qquad (27)$$

The above equation is very simple and does not need iterations; however, it is not accurate (Elsharkawy et al., 2001). For this reason, this method will be excluded from our Z-factor calculations for natural gases containing significant concentrations of CO_2.

Hankinson-Thomas-Philips Method (HTP)

Hankinson, Thomas, and Philips correlated the compressibility factors for natural gas as a function of the T_{pr} and P_{pr} by using the Benedict-Webb-Rubin EOS ((Hankinson et al., 1969). The proposed equation is expressed in terms of the compressibility factor as follows:

$$\frac{1}{z} - 1 + \left[A_4\,Tpr - A_2 \;- \frac{A_6}{Tpr^2}\right]\left(\frac{Ppr}{Z^2Tpr^2}\right) + \;(A_3Tpr - A_1)\left(\frac{Ppr^2}{Z^3Tpr^3}\right) +$$

$$\frac{A_1A_5A_7Ppr^5}{Z^6Tpr^6}\;\left[1 + \frac{A_8Ppr^2}{Z^2Tpr^2}\right]\exp[\frac{-A_8Ppr^2}{Z^2Tpr^2}] = 0$$

It is suggested that the proposed correlation is used only at reduced temperatures (T_{pr}) values greater than 1.1. The Hankinson et al. method proposed a set of coefficients for reduced pressures (P_{pr}) below 5.0 and another set for reduced pressures in the range of 5 to 15. Elsharkawy et al. (2001) studied the accuracy of Hankinson, Thomas, and Philips method in calculating the compressibility factors for gas condensates systems. They found that the equation has reasonable accuracy at reduced pressures below 5 and the second set of constants between 5 and 15 produced unrealistic compressibility factors. They also recommended avoiding the use this method for Z-factor calculation for reduced pressures above 5.0.

Hall-Yarborough method

Hall and Yarborough presented an EOS that accurately represents the Standing and Katz Z-factor chart. The proposed expression is based on the Starling-Carnahan EOS. They proposed the following equation (Hall and Yarborough, 1973; 1974) to calculate the Z-factor:

$$Z = \left[\frac{0.06125\;p_{pr}t}{Y}\right]EXP\left[-1.2\;(1-t)^2\right] \qquad (29)$$

Where, t = T_{pc}/T and Y is the reduced density calculated from the following equation:

$$F(Y) = 0.06125\;\;p_{pr}\,t\,EXP\left[-1.2(1-t)^2\right] + \frac{Y+Y^2+Y^3-Y^4}{(1-Y)^3} -$$

$$(14.76t - 9.76t^2 + 4.58t^3)\;Y^2 + (90.7t - 242.2t^2 + 42.4t^3)\;Y^{(2.18 +2.82t)} = 0 \qquad (30)$$

This method has received great application in the natural gas industry (Elsharkawy et al., 2001; Elsharkawy, 2001). Therefore, it will be used in the next part of this research to assess its accuracy in estimating the Z-factor for mixtures of natural gas with CO_2 at high pressures and high temperatures (HPHT), that is supercritical conditions.

Dranchuk-Purvis-Robinson method

Dranchuk, Purvis, and Robinson developed a correlation based on the Benedict-Webb-Rubin equation-of-state (Dranchuk et al., 1974). The equation has the following form:

$$Z = 1\left[A_1 + \frac{A_2}{T_{pr}} + \frac{A_3}{T_{pr}^3}\right]\rho_r + \left[A_4 + \frac{A_5}{T_{pr}}\right]\rho_r^2 + \left(\frac{A_5A_6}{T_{pr}}\right)\rho_r^5$$

$$\left[\frac{A_7}{T_{pr}^3}\rho_r^2\;(1 + A_8\,\rho_r^2)\;EXP\;(-A_8\;\;\rho_r^2\right] \qquad (31)$$

This method will also be considered in the Z-factor calculations at HPHT for natural gas-CO_2 mixtures.

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DAK method

Dranchuk and Abu-Kassem proposed an eleven-constant EOS for calculating the Z-factor. They proposed the following equation (DAK) (Dranchuk and Abu-Kassem, 1975):

$$Z = \left[A_1 + \frac{A_2}{T_{pr}} + \frac{A_3}{T_{pr}} + \frac{A_4}{T_{pr}^4} + \frac{A_5}{T_{pr}^5}\right]\rho_r \qquad +$$

$$\left[A_6 + \frac{A_7}{T_{pr}} + \frac{A_8}{T_{pr}^2}\right]\rho_r^2 - A_9\left[\frac{A_7}{T_{pr}} + \frac{A_8}{T_{pr}^2}\right]\rho_r^5$$

$$+\;A_{10}(1 - A_{11}\rho_r^2)\frac{\rho_r^2}{T_{pr}^3}EXP[-A_{11}\rho_r^2] + 1 \qquad (32)$$

Where,

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

The above method is also widely used in the petroleum industry to calculate the gas compressibility factor for many gases (Elsharkawy et al., 2015). This method will be considered in the second part of this research.

Londono et al. method

Londono et al. fitted the DAK EOS to a research database. Their modification resulted in an average absolute error (AAE) of 0.412% using that database (Londono et al., 2005). This method was evaluated using a large data bank of natural gas-CO_2 mixtures at supercritical conditions.

Al-Anazi and Al-Quraishi method

Al-Anazi and Al-Quraishi proposed another Z-factor correlation based on genetic programming techniques. They stated that their new model allows for accurate determination of Z-factor values both for pure components and gas mixtures. In their method, the factor is calculated in seven-steps as follows (Al-Anazi and Al-Quraishi, 2010):

$$Z = \frac{2E}{1.0482} + F \tag{33}$$

$$F = \frac{D}{E^2} - E \tag{34}$$

$$E = \left[\left(\frac{C + D}{T_{pr}}\right)/1.0474 \right] + 0.9178 \tag{35}$$

$$D = (-2A + B) - C \tag{36}$$

$$C = B - (-2A + B)$$

$$B = \frac{(3A)^2 - 1.427}{T_{pr}} + 0.9178 \tag{37}$$

$$A = \left(2 - \left(\frac{0.05275}{-0.5765}\right) P_{pr}\right) + 0.2360 \tag{38}$$

It is important to note that this method has not been evaluated by any researcher other than the authors, hence this research presents the first assessment of this correlation for calculating the Z-factor for natural gas and CO_2 mixtures.

Bahadori and Vuthaluru method

Bahadori and Vuthaluru proposed another five-step method with sixteen constants to calculate the compressibility factor. In their method, the Z-factor is correlated to the reduced pressure (P_r) and temperature (T_r) as follows (Bahadori and Vuthaluru, 2010):

$$\mathrm{Ln}\,(Z) = \alpha + \frac{\beta}{T_r} + \frac{\gamma}{T_r^2} + \frac{\theta}{T_r^3} \tag{39}$$

Where,

$$\alpha = A_1 + \frac{B_1}{P_r} + \frac{C_1}{P_r^2} + \frac{D_1}{P_r^3} \tag{40}$$

$$\beta = A_2 + \frac{B_2}{P_r} + \frac{C_2}{P_r^2} + \frac{D_2}{P_r^3} \tag{41}$$

$$\gamma = A_3 + \frac{B_3}{P_r} + \frac{C_3}{P_r^2} + \frac{D_3}{P_r^3} \tag{42}$$

$$\theta = A_4 + \frac{B_4}{P_r} + \frac{C_4}{P_r^2} + \frac{D_4}{P_r^3} \tag{43}$$

It is important to note that this method has also not been evaluated by any researcher other than the authors, and this research presents the first assessment of their correlation for calculating the Z-factor for natural gas and CO_2 mixtures at supercritical conditions.

Aziz et al. method

Aziz et al. (2010) proposed a six-step method using twenty five constants to calculate the Z-factor based on the famous Standing-Katz (SK) chart. Their correlation does not require iteration, and is as follows (Aziz et al., 2010):

$$z = A + \frac{B+C}{D+E} \tag{44}$$

Where,

$$A = aT_r^{2.16} + bP_r^{1.028} + cP_r^{1.58}T_r^{-2.1} + dln(T_r)^{-0.5} \tag{45}$$

$$C = iln(T_r)^{-1.28} + jln(T_r)^{1.37} + kln(P_r) + lln\,(P_r)^2 + mln(P_r)\ln(T_r) \tag{46}$$

$$D = 1 + nT_r^{5.55} + oP_r^{0.68}T_r^{0.33} \tag{47}$$

$$E = pln(T_r)^{1.18} + qln(T_r)^{2.1} + rln(P_r) + sln(P_r)^2 + tln(P_r)\ln(T_r) \tag{48}$$

This method has not been evaluated by any researcher other than the authors and this study is the first attempt to study its accuracy and its range application at supercritical conditions.

Heidaryan et al. method

Heidaryan et al. proposed another one-step explicit numerical method for calculating the Z-factor using eleven constants. The Z-factor is calculated from the following (Heidaryan et al., 2010):

$$z = f\left(P_{pr}\,\frac{1}{T_{pr}}\right) \tag{59}$$

$$z = ln\left(\frac{A_1 + A_3 ln(p_{pr}) + \frac{A_5}{T_{pr}} + A_7\left(\ln(p_{pr})^2\right) + \frac{A_9}{T_{pr}^2} + \frac{A_{11}}{T_{pr}}\ln(P_{pr})}{1 + A_2\ln(P_{pr}) + \frac{A_4}{T_{pr}} + A_6\left(\ln(p_{pr})\right)^2 + \frac{A_8}{T_{pr}^2} + \frac{A_{10}}{T_{pr}}\ln(p_{pr})}\right) \tag{50}$$

This method has not been evaluated by any researcher other than the authors, and this research is the first attempt to study its accuracy and range of application at HPHT.

Ehsan-Ebrahim method

Ehsan and Nemati presented two Z-factor correlations based on 5,844 experimentally published data for natural gas mixtures. The gases used to develop their correlations were mostly composed of methane with CO2contents not exceeding 50%, and a maximum pressure of 100.66 Mpa (14,600 psia) and maximum temperature of 598 K. One of the correlations is for low pressures, Ppr <3.0, while the other correlation is used for the reduced pressure range of 3.0 to 15. This empirical correlation was developed using multiple regression analysis based on viral equation of state (Ehsan and Nemati, 2012). It is important to note that this recently published correlation is developed using limited data, low levels of CO_2, and low pressures and temperatures compared to the data bank considered in this study, and therefore, the method is not considered in this research.

Shokir et al. method

Shokir et al. proposed the calculation of gas compressibility factor for various gases using genetic programing. The gas composition is used to calculate the pseudo critical pressures and temperatures via six-steps. The Z-factor is calculated from Ppc, Tpc, pressure and temperature via another six equations (Shokir et al., 2012). The method is quite long, therefore, it not considered in this research.

Kamari et al. Method

Kamari et al. proposed a calculation of the compressibility factor of sour gases using an intelligent approach. The method is based on Least Square Support Vector Machine (LSSVM) (Kamari et al., 2013). They did not present an algorithm nor equations for calculation of the Z-factor. Therefore, the method is not considered in this study.

Fatoorehchi et al. method

Fatoorehchi et al. presented a modification to the Hankinson-Thomas-Phillisps (HTP) correlation for calculation of the Z-factor. Elsharkawy et al. 2001 and Fatoorehchi et al. 2013 reported that the HTP method is not valid for high pressures. This method is also not considered in the evaluation for the previously mentioned reasons.

Kamyab et al. method

Kamyab et al. proposed a method to obtain Z-factors for natural hydrocarbon gases using Artificial Neural Networks (ANN). The input parameters in the ANN are the P_{pr} and T_{pr}. The method needs an engineer who knows how to code the ANN to be able to estimate the Z-factor (Kamyab et al., 2014). This method has also not been evaluated by any researcher other than the authors and this research is the first attempt to study its accuracy and range of applicability at HPHT conditions.

Fayazi et al. method

Similar to the method proposed by Kamari et al., Fayazi et al. also proposed the calculation of the compressibility factor of sour gases using LSSVM. They did not present an algorithm nor an equation for calculation of the Z-factor. This method is not considered in this study.

Mohagheghian and Bahadori (2015)

Mohagheghian and Bahadori calculated the CO2 compressibility factor using an intelligent approach. They did not present any equation or algorithm for the compressibility calculations. Therefore, this method is not considered in this study.

Thus, the previously mentioned methods that are previously mentioned can be classified into three groups:

(1) Iterative methods: (Hall-Yarborough 1973; 1974; Dranchuk-Abu-Kassem 1975, Dranchuk-Purvis-Robinson 1974; Hankinson –Thomas-Phillips, 1969; Londono et al., 2005).
(2) Direct solution methods: Al-Anazi et al. (2010), Bahadori and Vuthaluru (2010), Aziz et al. (2010), and Heideryan et al. (2010).
(3) Intelligent approach methods that need programing or coding techniques: Kamyab et al. (2014), Shokir et al. (2012), Kamari et al. (2013), Fatoorehchi et al. (2013), Fayazi et al. (2014), Ehsan and Nemati (2012), Mohagheghian and Bahadori (2015), and Khosravi et al. (2018).

Description of data used in this study (method)

There are at least 6 mixing rules to calculate the P_{pc} and T_{pc} of natural gas-CO_2 mixtures and 12 methods to calculate the Z-factor. Thus, there are at least 72 possible ways to calculate the Z-factor for natural gases with significant CO_2 content. In this work, various mixing rules and industry standards for Z-factor calculation are evaluated. The accuracy of all the mentioned mixing rules, as well as the Z-factor correlations are studied using a large data bank of 2,200 Z-factor measurements of NG-CO_2 mixtures at supercritical conditions.

Data bank

A total of 2,200 Z-factor measurements representing

Table 1. Ranges of property of the gas mixtures in the data bank used in this study.

Component	Minimum	Maximum	Average
Temperature (K)	286	478	352
Pressure (Mpa)	0.11	144.43	19.82
Methane	0	0.9222	0.4624
Ethane	0	0.2867	0.0256
Propane	0	0.1316	0.0088
Butane	0	0.0380	0.0036
Pentane	0	0.0285	0.0018
Hexane	0	0.0268	0.0010
Heptane plus	0	0.0817	0.0003
MW_{C7+}	118.0	127.0	122.4
SG_{C7+}	0.7500	0.8050	0.7666
Z-factor	0.0605	2.8743	0.8727
Hydrogen Sulfide	0	0.8104	0.08261
Carbon dioxide	0	0.9393	0.40501
Nitrogen	0	0.1558	0.00342
T_{pr}	0.7527	2.5100	1.4683
P_{pr}	0.0141	31.0187	3.8341

various natural gases with CO_2 content ranging from 0% to as high as 94% have been studied in this work. This data bank was collected from various sources: Robinson et al. (1960), Robinson and Jacopy (1965), DeWitt and Thodos (1966), Buxton and Campbell (1967), Wichert (1970), Simon et al. (1977), Li and Guo, (1991), Assael et al. (2001), Adisoemarta et al. (2004), Bennion et al. (2004), Elsharkawy (2001, 2004), Elsharkawy et al. (2001, 2001, 2015), Rushing et al. (2008), Tabasinejad et al. (2010), Bian et al. (2012), and Li et al. (2016). Various combinations of mixing rules and Z-factor correlations are examined to determine the accuracy of each method. Subsequently, a new method to calculate the Z-factor for natural gas-CO_2 mixtures is presented using the data bank for extremely high pressure and temperature systems. The properties of these mixtures were studied at pressures ranging from 0.11 to 144.43 Mpa (16 to 20,948 psia) and temperatures ranging from 286 to 478 K (55 to 402°F). CO_2 is known to have a critical pressure of 7.4 Mpa and a critical temperature of 304 K (1070 psia and 88°F, respectively). Thus, most of the gases in the data bank exist at supercritical conditions. A detailed description of the various gases used in the data bank is shown in Table 1. The compressibility and density of some of these gases are reported at Pr as high as 31, and Tr as low as 0.7. Thus, deeming most Z-factor correlations and the Standing-Katz chart unsuitable, due to the scope of Pr and Tr covered by these methods (Standing and Katz, 1942).

Limitations of exiting methods

According to the data provided in Table 1, most of the gases in the data bank exist at pressure level of 20 Mpa and temperature of 352K, which exceeds the critical point of CO_2-NG mixtures. There are many published correlations available for the calculation of Z-factor for natural gases. The working condition and limitations of all the Z-factor calculation methods used in this research are briefly summarized in Table 2. This table shows that most of the published methods have temperature and pressure limitations, thus, they cannot be used to calculate the compressibility factor, and hence the density for T_r below 1.0 and reduced P_r above 30.

RESULTS AND DISCUSSION

Evaluation of previously published methods

One objective of this study is to evaluate the validity and accuracy of all well-known and recently published mixing rules, as well as the Z-factor calculation methods. This is done by examining the Z-factor calculations obtained through various combinations of mixing rules and Z-factor correlations against already measured Z-factors for the natural gas-CO_2 mixtures in the data bank. Statistical analysis is conducted to evaluate the performance and the working limits of the calculation methods. The analysis is comprised of average percent relative error (Eave), average absolute error percent (Eabs), root mean square error (Ems), and correlation coefficient. Appendix A provides details of these statistical analyses.

The results of the error analysis are reported in Table 3. It is clear from Table 3 that the Kay mixing rule together with the Wichert-Aziz correction for the presence of non-hydrocarbon components and the Hall-

Table 2. Working ranges of T_r and P_r for various z calculation methods.

Z Calculation method	Range of Tr	Range of Pr
Hall-Yarborough, 1973;1974	no limits mentioned	no limits mentioned
Dranchuk-Purvis-Robinson (2015)	1.05 to 3	0.2 to 30
Dranchuk-Abou Kassem (1975)	1 to 3	0.2 to 30
Londono et al. (2005)	no limits mentioned	no limits mentioned
Bahadori et al. (2010)	1.05 to 2.4	0.2 to 16
Azizi et al. (2010)	1.1 to 2	0.2 to 11
Heidaryan et al. (2010)	1.2 to 3	0.2 to 15
Al-Anazi et al. (2010)	0.974 to1.966	0.174 to 10.195
Kamyab et al. (2014)	1 to 3	0.2 to 30
Ehsan and Nemati (2012)	0.753 to 2.51	0.14 to 31

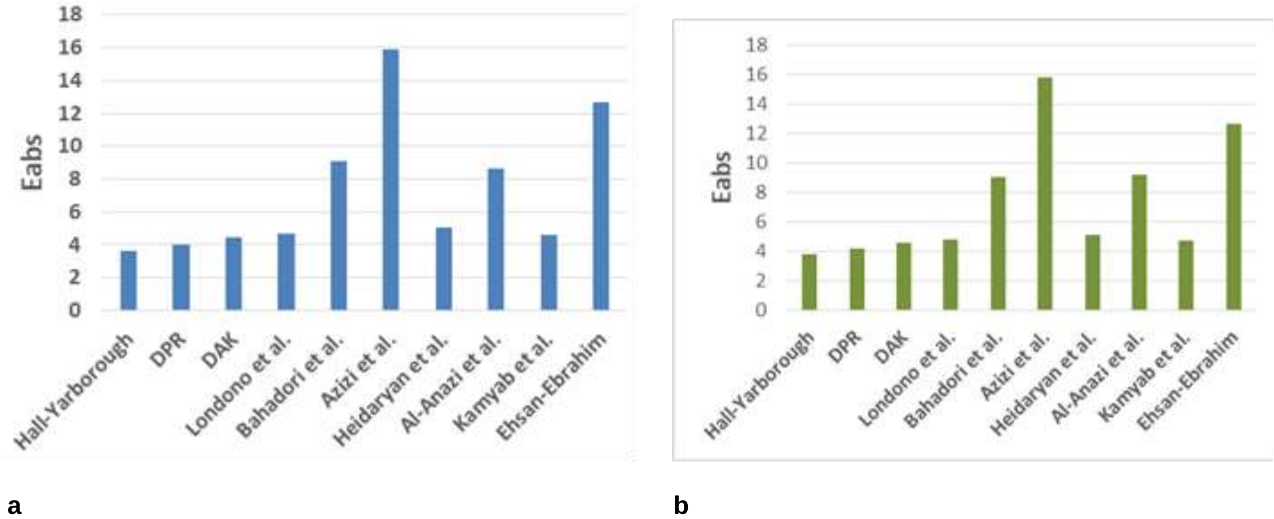

Figure 1(a). E_{abs} Z-Factor Methods (Kay's Mixing Rule) (b). E_{abs} Z-Factor Methods (SBV Mixing Rule).

Yarborough Z-factor correlation resulted in the highest level of accuracy for all gas mixtures considered in this study. The previously mentioned mixing rule, non-hydrocarbon correction method, and Z-factor correlation showed the smallest errors (Eabs= 3%) and highest correlation coefficient (96%). Evidently, recently developed and published methods: Al-Anzi and Alqauraishi, (2010), Bahadori et al. (2010), Aziz et al. (2010), and Ehsan and Nemati (2012) have exceptionally high Eabs of 8.6, 9, 13, and 16%, respectively. It is also observed that the recently published mixing rules presented by Piper et al. (1993), Corredors et al. (1992), and Elsharkawy (2004), which account for the presence of non-hydrocarbon components in natural gases, were not able to reasonably estimate pseudo-reduced properties of the natural gas-CO_2 mixtures in the data bank. It is important to note that some of the gases considered in this study contain as high as 94% CO_2. Therefore, these mixing rules show Eabs in the order of 8 to 20% depending on the selected Z-factor correlation used. Figure 1 (A through F) indicates that among all the mixing rules considered in this study, Kay's mixing rule showed the smallest error level with all the various Z-factor correlations. Furthermore, these figures as well as Table 3, show that Hall-Yarborough 1973;1974 correlations for Z-factor has the smallest average absolute error (Eabs) of less than 4% in comparison to the other methods discussed in this paper. Additionally, Hall-Yarborough correlation has a wider range of application; nearly 1,849 data points (Nd) were predicted out of the total measurements of 2,200.

Newly proposed method

It is has been proven in Table 3 that Kay's (Kay, 1936) mixing rule combined with the Standing-Katz (Standing and Katz, 1942) chart are able to reasonably estimate the

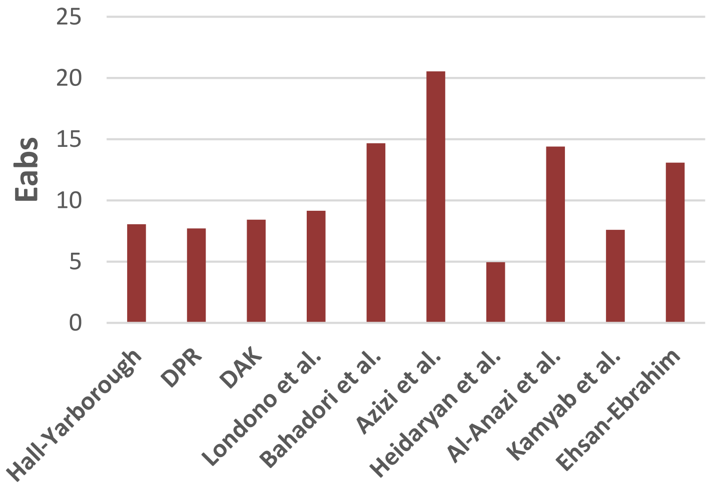

Figure 1c. E_{abs} Z-factor methods (Piper Mixing Rule).

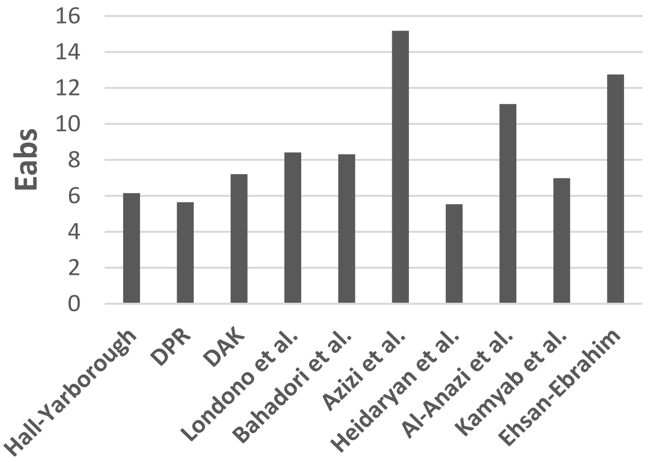

Figure 1d. E_{abs} Z-factor methods (Corredore Mixing rule).

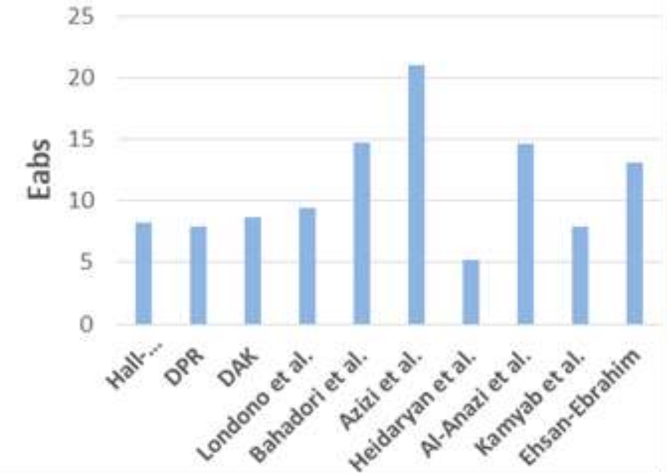

Figure 1e. E_{abs} Z-factor methods (Elsharkawy's).

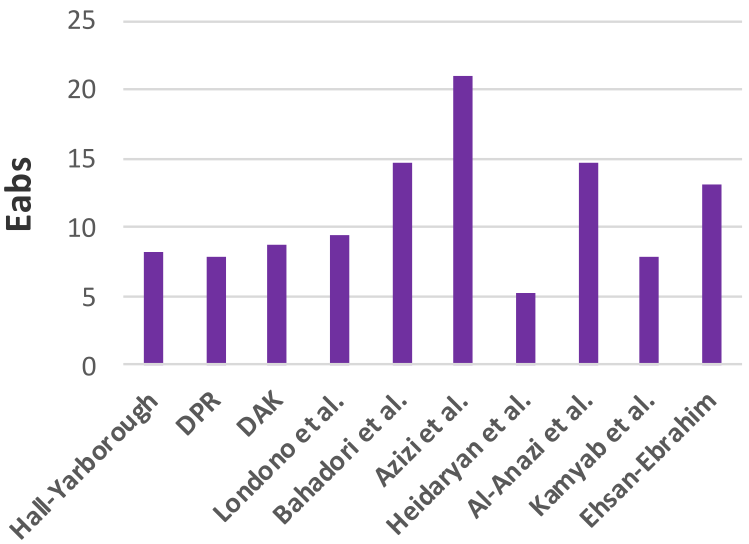

Figure 1f. E_{abs} Z-Factor methods (Bahadori rule).

Z-factor under most conditions. Various mixing rules have been developed for different types of natural gas mixtures. Sutton's (Sutton, 1985) modification of SBV mixing rule was introduced to account for the presence of the heptane plus fraction in gas condensates. Piper et al. (1993), Corredor et al. (1992), and Elsharkawy (2004) mixing rules were recommended for various gases containing non-hydrocarbon components and heptane plus fractions. However, it is clear from Table 1 that the gases considered in the data bank have a high percentage of CO_2 that is beyond the gases used to develop the previously mentioned mixing rules. In this study, three newly proposed correlations have been presented to estimate the Z-factor for natural gases containing significant portion of CO_2. The first correlation proposed for low pressure ranges has the following form:

$$Z = 1 + A_1 P_{pr} + A_2 P_{pr}^2 + \frac{A_3 P_{pr}^{A_4}}{T_{pr}^{A_5}} + \frac{A_6 P_{pr}^{(A_4+1)}}{T_{pr}^{A_7}} + \frac{A_8 P_{pr}^{(A_4+2)}}{T_{pr}^{(A_7+1)}} \qquad (51)$$

Where, A1= -670.272773, A2= -48.271233, A3= 669.298742, A4=0 .999561, A5= -0.002100, A6=48.795104, A7= 0.010294, A8= -0.038756.

Figure 2 shows a cross-plot of calculated versus measured Z-factors for the a-pressure range of $0.01 < P_{pr} < 3.0$. This correlation has a correlation coefficient of 0.93.

The second correlation covers the high-pressure range P_{pr} of 3.0 to 15, where the constants have the following values: A1= 0.686740, A2= 0.000743, A3= 0.751088, A4=0.166734 , A5= 0.802425, A6=0.480604, A7= 0.044629, A8=0.011819.

Figure 3 shows a cross-plot of calculated versus measured Z-factor for the pressure range of $3.0 < P_{pr} < 15$. This correlation has a coefficient of 0.926.

The third correlation covers the entire pressure range (P_{pr}) from 0.01 to as high as 30. The Constants A1 through A8 has the following values: A1= 0.04591366, A2= -0.000673898, A3= -0.597635121, A4=0.811288572, A5= 2.898688651, A6=0.105471654, A7= 4.261541546, A8= -0.002537591 Figure 4 shows a cross-plot of calculated versus measured Z-factor for the entire pressure range. This correlation has a coefficient of 0.96.

Figure 5 shows the error distribution for the three proposed correlations. This figure indicates that at the 2% absolute error level, the three correlations have almost the same cumulative error frequency.

Evaluation of the validity of the newly proposed method

To evaluate the validity of the proposed set of correlations for natural gases with various CO_2 contents at super critical conditions, a few of the gases provided in the data bank were chosen. The Z-factor for the chosen gases was then calculated using the new correlations as well as Soav-Redich-Kowng equation of state, (SRK) and Peng-Robinson equation of state, (PR) for comparison of Z-factor calculations. Both SRK and PR are given in Appendix B. Table 4 shows the selected gases from the data bank that is available in this study. These gases were selected due to large compositional differences.

Gas A is a dry gas containing 20% CO_2. The P-T diagram for this gas is shown in Figure 6A. This figure indicates that this gas has a critical point at 221K and 6.5 Mpa and is at initial reservoir conditions of 477.6 K and

Table 3. Error analysis of mixing rules and Z-factor correlations.

Method	Kay's mixing rule					SBV modified mixing rule				
	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2
Hall-Yarborough (1974)	1849	-1.744	3.638	1.480	0.957	1850	-1.288	3.796	1.519	0.957
DPR (1974)	1505	-2.179	4.025	1.580	0.939	1506	-1.704	4.231	1.633	0.939
DAK (1975)	1622	-2.673	4.426	1.627	0.961	1624	-2.160	4.603	1.675	0.960
Londono et al. (2005)	2044	-1.709	4.701	1.565	0.974	2044	-1.218	4.838	1.611	0.974
Bahadori et al. (2010)	1410	-1.644	9.052	2.694	0.881	1411	-1.257	9.025	2.678	0.879
Azizi et al. (2010)	1121	-10.337	15.870	4.805	0.734	1112	-10.11	15.822	4.749	0.728
Heidaryan et al. (2010)	1159	-2.419	5.055	1.684	0.811	1163	-2.044	5.137	1.712	0.807
Al-Anazi et al. (2010)	1394	-0.299	8.674	2.134	0.889	1401	0.234	9.166	2.251	0.884
Kamyab et al. (2014)	1848	-2.636	4.583	1.630	0.954	1849	-2.195	4.710	1.669	0.954
Ehsan and Nemati (2012)	2039	3.530	12.671	2.885	0.958	2039	3.502	12.650	2.891	0.958

Method	Piper et al. mixing rule					Corredor mixing rule				
	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2
Hall-Yarborough (1974)	1949	4.538	8.048	2.156	0.963	1785	-2.978	6.155	2.245	0.926
DPR (1974)	1666	3.732	7.716	2.135	0.963	1422	-2.075	5.649	2.074	0.902
DAK (1975)	1722	4.538	8.424	2.187	0.965	1570	-3.591	7.206	2.427	0.928
Londono et al. (2005)	2044	5.936	9.153	2.203	0.965	2044	-3.699	8.415	2.798	0.943
Bahadori et al. (2010)	1557	5.826	14.665	3.358	0.884	1310	-2.330	8.310	2.662	0.836
Azizi et al. (2010)	1300	-2.549	20.548	5.471	0.749	1047	-11.332	15.182	4.594	0.666
Heidaryan et al. (2010)	1235	-1.174	4.954	1.660	0.842	1142	-1.547	5.530	1.830	0.780
Al-Anazi et al. (2010)	1488	9.479	14.406	2.804	0.873	1340	-4.570	11.102	2.863	0.864
Kamyab et al. (2014)	1948	3.637	7.598	2.101	0.961	1784	-4.083	6.985	2.380	0.921
Ehsan and Nemati (2012)	2034	3.581	13.080	2.983	0.955	2040	3.787	12.748	2.936	0.955

Method	Elsharkawy's mixing rule					Bahadori mixing rule				
	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2	N_d	E_{AVE}	E_{ABS}	E_{RMS}	r^2
Hall-Yarborough (1974)	1953	4.744	8.268	2.184	0.963	1296	-1.713	3.842	1.517	0.882
DPR (1974)	1666	3.934	7.945	2.164	0.962	1179	-1.705	4.120	1.588	0.876
DAK (1975)	1726	4.781	8.684	2.220	0.965	1196	-2.006	4.305	1.605	0.885
Londono et al. (2005)	2044	6.162	9.398	2.236	0.965	1336	-1.679	4.179	1.575	0.908
Bahadori et al. (2010)	1556	5.856	14.708	3.357	0.883	1066	-2.358	6.408	1.968	0.821
Azizi et al. (2010)	1307	-1.997	21.048	5.530	0.748	890	-13.028	15.186	4.367	0.721
Heidaryan et al. (2010)	1260	-0.928	5.180	1.704	0.855	1031	-2.085	5.095	1.630	0.775
Al-Anazi et al. (2010)	1488	9.839	14.646	2.831	0.873	924	0.081	6.359	1.659	0.854
Kamyab et al. (2014)	1952	3.784	7.871	2.136	0.961	1294	-1.838	3.982	1.544	0.881
Ehsan and Nemati (2012)	2034	3.584	13.123	2.993	0.955	2039	3.578	12.846	2.934	0.957

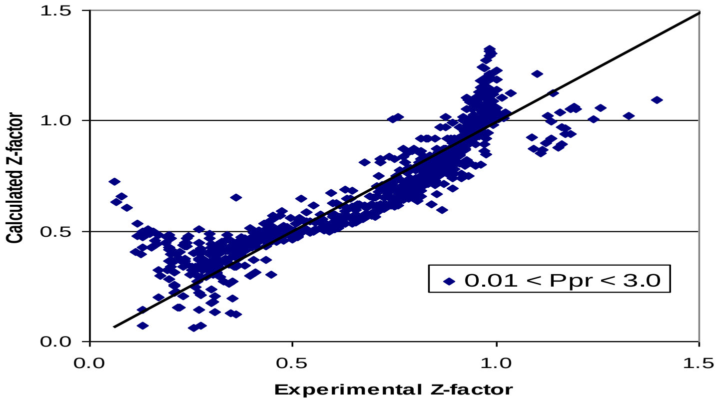

Figure 2. Crossplot of Z-factor for the range of P_{pr} 0.01 to 3.0.

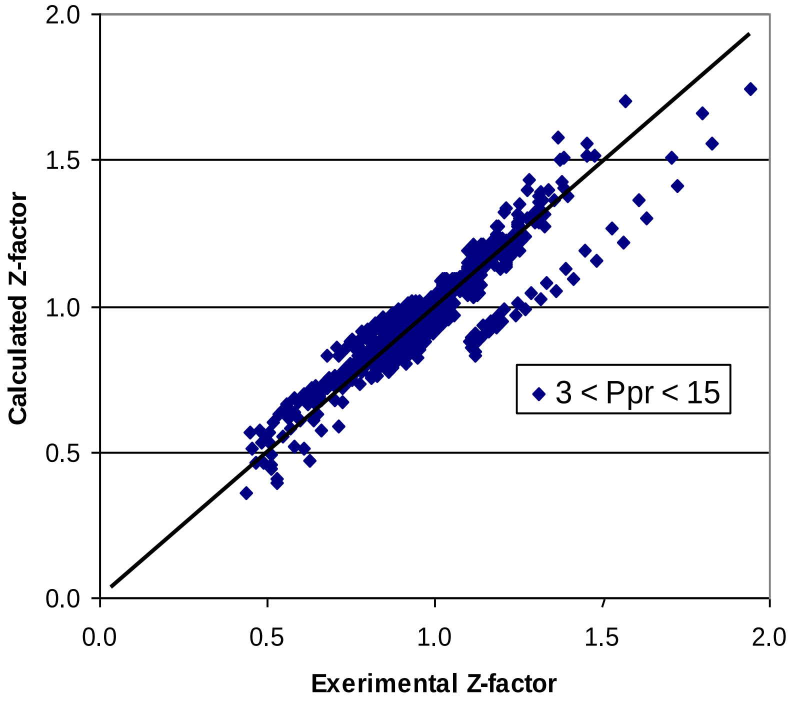

Figure 3. Crossplot of Z-factor for the range of P_{pr} 3 to 15.

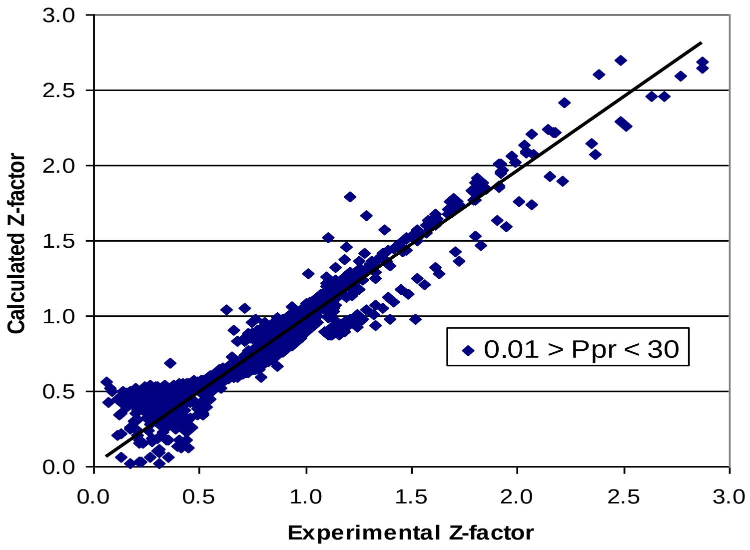

Figure 4. Crossplot of Z-factor for the range of P_{pr} 0.01 to 30.

136.7 Mpa. The measured Z- factor is reported for this gas at supercritical conditions of 477.6 K (400°F) and pressures up to 137.89 Mpa (20,000 psia). Figure 6B shows a comparison between measured and calculated Z-factors for Gas A at pressures up to 41.37 Mpa (6000 psia) using SRK equation of state, PR equation of state, and the correlation presented in this paper. It is clear from this figure that the calculated Z-factor from the newly proposed correlations is much closer to the experimental value than SRK equation of state, i.e. at 51.8 Mpa the experimentally measured Z-factor is found to be 1.152, whereas, the model presented in this study finds it to be 1.178 while SRK obtains 1.203. Figure 6C shows a similar comparison at a high pressure range of 41.37 to

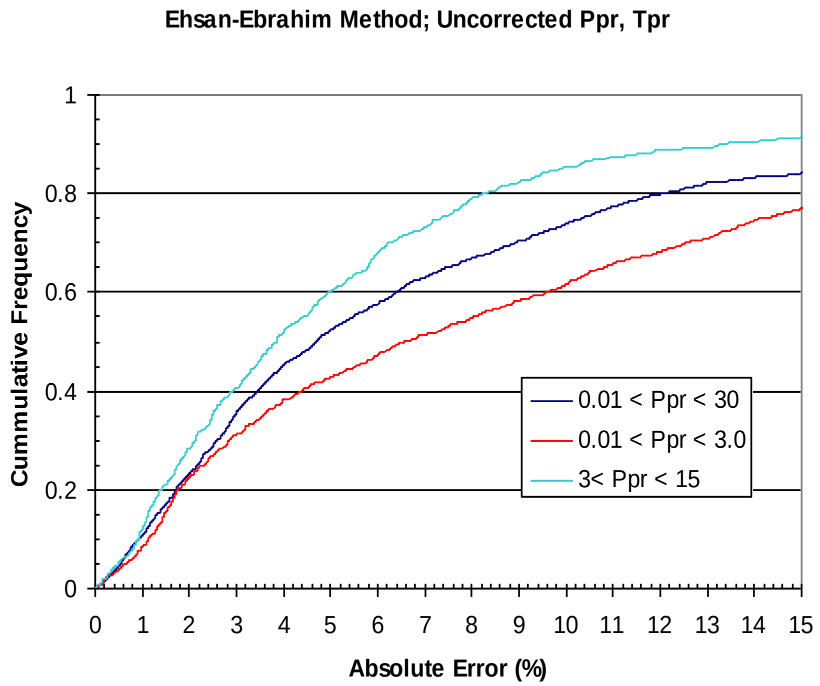

Figure 5. Error Distribution for the Three Proposed Correlations.

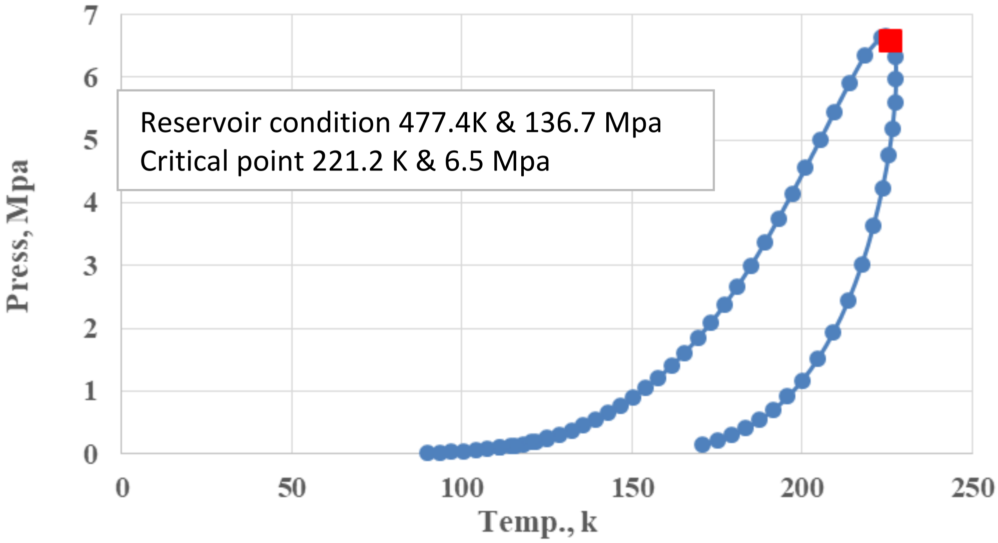

Figure 6A. P-T Diagram of Gas A (20% CO_2).

137.89 Mpa (6000-20,000 psia). This figure also indicates that the presented model is much closer to the experimental data. However, in this case the prediction by PR is much more accurate than SRK equation of state.

Gas B is another dry gas containing 75% CO_2. The P-T diagram for this gas is shown in Figure 7A. This figure shows that this gas has a critical point at 283K and 8.45 Mpa, and initial reservoir conditions of 377 K and 34 Mpa. The measured Z-factor for this gas is shown in Table 4 at 377K and pressures up to 34.77 Mpa, which are supercritical conditions. Figure 7B shows a comparison between the measured and predicted Z-factors via the correlation presented in this study as well as SRK and

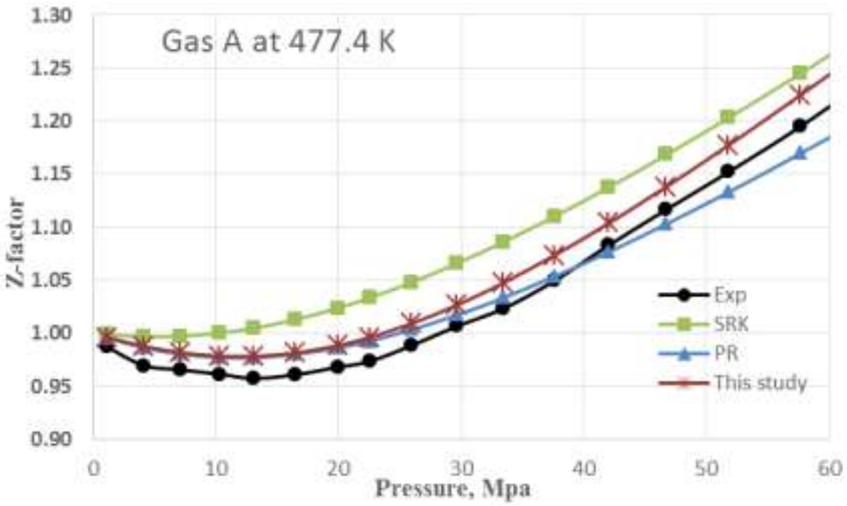

Figure 6B. Measured and Predicted Z-Factor for Gas A at Low P.

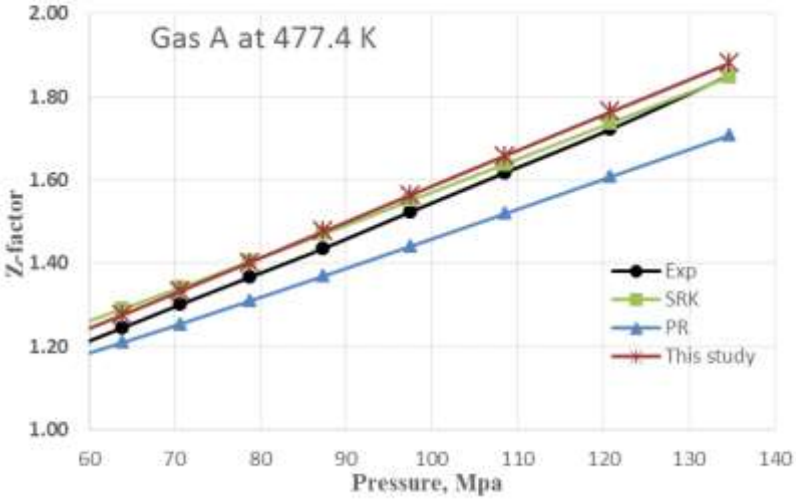

Figure 6C. Measured and Predicted Z-Factor for Gas A at High P.

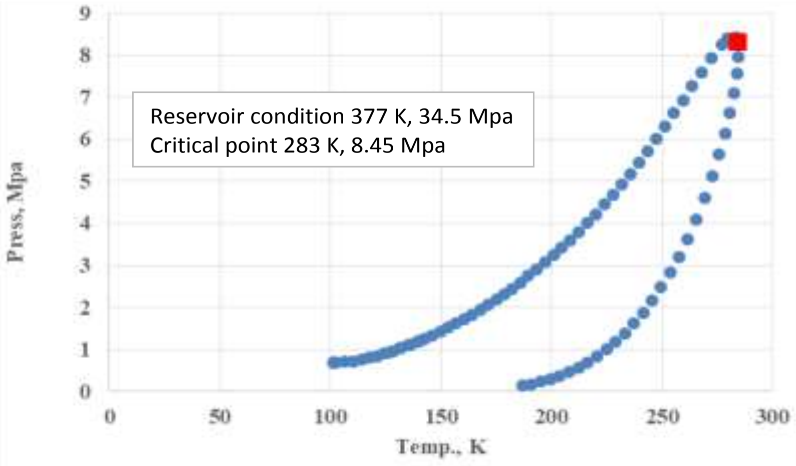

Figure 7A. P-T Diagram of Gas B (75% CO_2).

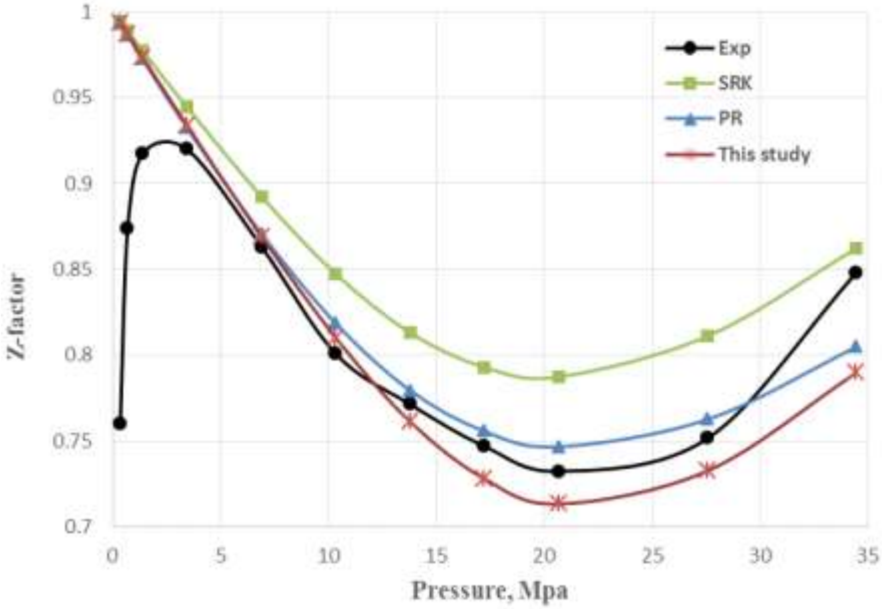

Figure 7B. Measured and Predicted Z-Factor for Gas B at 377 K

Table 4. Gas composition and measured Z-factors for gases containing high amounts of CO_2.

Composition	Gas A	Gas B	Gas C	Gas A at 477.6 K		Gas B at 377.6K		Gas C at 322K	
	MF	MF	MF	P (Mpa)	Z	P (Mpa)	Z	P (Mpa)	Z
H_2S	0	0	0	134.64	1.8511	34.47	0.8481	20.15	0.4725
CO_2	0.2	0.75	0.8994	108.45	1.6173	27.58	0.7517	19.04	0.4571
N_2	0	0	0.0004	87.35	1.436	20.68	0.7325	17.94	0.4441
C1	0.768	0.242	0.0944	70.68	1.3015	17.24	0.747	17.04	0.4351
C2	0.024	0.007	0.0021	57.66	1.1951	13.79	0.7715	15.94	0.4253
C3	0.008	0.001	0.001	51.80	1.1522	10.34	0.8008	14.84	0.4194
iC4	0	0.001	0	42.04	1.0832	6.89	0.8632	13.67	0.4184
nC4	0	0	0.0006	33.45	1.0235	3.45	0.9198	12.96	0.4273
iC5	0	0	0	25.90	0.9884	1.38	0.9173	10.58	0.5098
nC5	0	0	0.0005	19.93	0.9682	0.69	0.8737	9.45	0.5781
C6	0	0	0.0016	13.04	0.9576	0.34	0.7603	8.00	0.6623
C7	0	0	0	7.08	0.9657			6.56	0.7382
C8				4.10	0.9698			5.41	0.789
Total	1	1	1	1.12	0.9872			4.69	0.8191

MF is the mole fraction, Gas A is from Rushing et al. (2008), Gas B is from Adisoemarata et al. (2004), and Gas C is from Simon et al. (1977).

PR equations of state. This figure indicates that the measured Z-factors at pressures below 3.48 Mpa (500 psia) are unreliable; as all predictions via the various methods fall close to each other. However, at pressures greater than 3.48 Mpa, the calculations by the proposed correlations in this study are in agreement with PR equation of state and much closer to the measured values than predicted by SRK.

Gas C is a CO_2 rich gas which contains 90% CO_2. The P-T diagram of this gas is shown in Figure 8A. This figure indicates that this gas has a critical point at 296K and 7.86 Mpa and initial reservoir conditions of 322K and 20.7 Mpa. The compressibility factors of this gas at supercritical conditions of 322 K (120°F) and pressures up to 20.15 Mpa are reported in Table 4. Figure 8B shows a comparison of measured and predicted Z-factors by SRK, PR, and this study's proposed correlations. Again the calculated Z-factors in this study match the

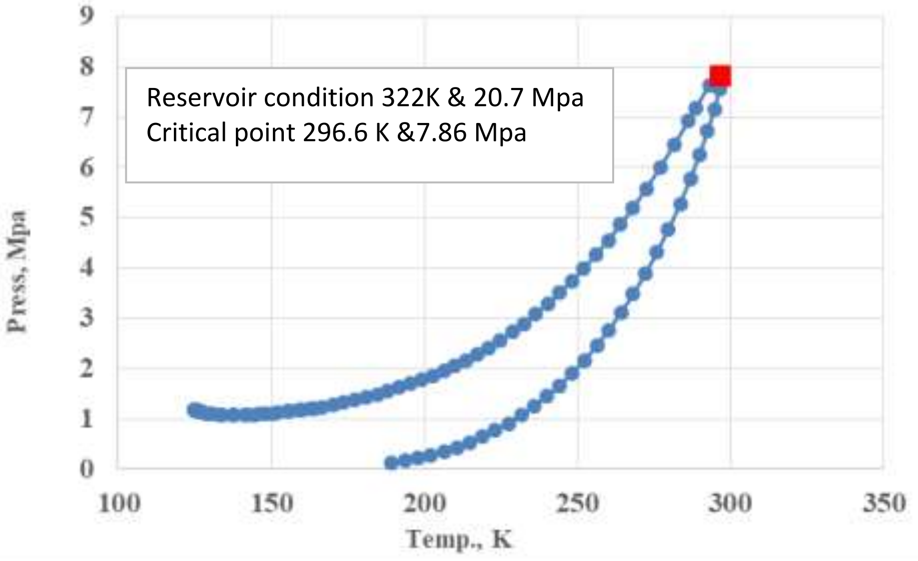

Figure 8A. P-T Diagram of Gas C (90% CO_2).

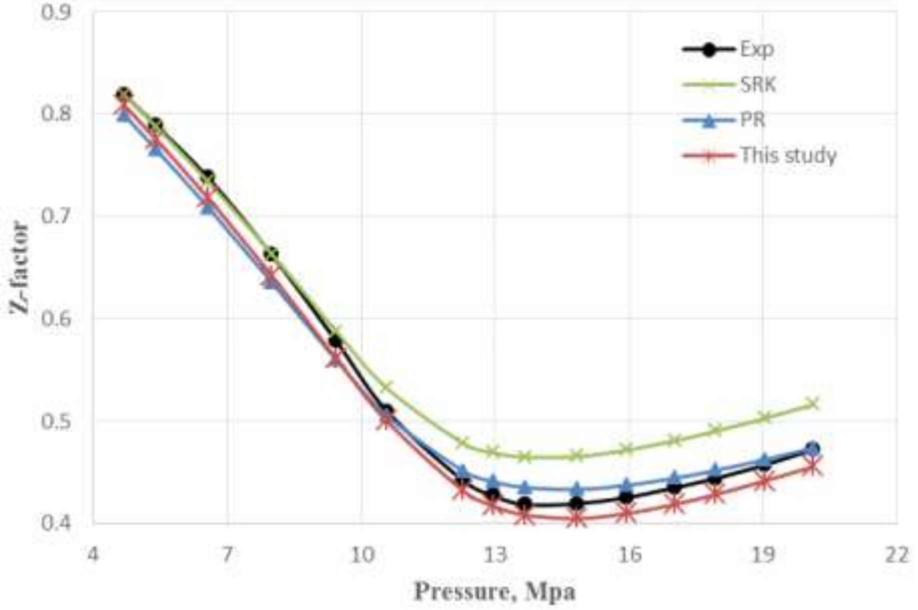

Figure 8B. Measured and Predicted Z-Factor for Gas C at 322 K.

experimental values well and agree with both SRK and PR equations of state. Once again, as the pressure increases, SRK EOS predictions become less reliable.

Conclusion

A large data bank of Z-factor measurements was collected for natural gas-CO_2 mixtures with exceptionally high contents of CO2, ranging from 0 to 94%, at pressures higher than any previously used data. The pressures of the gas mixtures range from 0.11 to 144.43 Mpa (16 to 20,948 psia) and temperatures range from 286 to 478 K (55to 402°F).

The accuracy of previously published mixing rules and Z-factor correlations were examined using a large data bank of natural gas systems with varying temperatures, pressures, and CO_2 content. This study considered 60 possible techniques (through the numerous combinations of mixing rules and Z-factor correlations) to estimate the Z-factor knowing the composition of the natural gas. It was found that Kay's mixing rule and Wichert and Aziz method for correction for non-hydrocarbons combined with Hall-Yarborough correlation produces the highest accuracy in predicting compressibility factor for natural gas-CO_2 mixtures, with a correlation coefficient of 0.96.

New correlations were proposed with the capability to predict the Z-factor for natural gas-CO2 mixtures. The new method is simple, does not require iterations or coding, and can easily be used. The Z-factor predictions at supercritical conditions by the newly proposed correlations were tested against measured experimental data for some selected gases as well as predictions by PR and SRK equations of state. The comparisons indicated that the new proposed correlations closely match the experimentally measured Z-factor at extremely high temperatures and pressures, with correlation coefficients ranging from 0.926-0.96. The data obtained from these correlations will prove helpful for the pipeline design, transport of natural gas, and planning for gas processing facilities. The newly proposed correlations are also useful for the design of carbon capture and storage plants and the determination of carbon storage sites.

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CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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