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 CO$_2$ 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.

**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:

$$P_{pc} = \sum y_i P_{ci}$$  \hspace{1cm} (1)

$$T_{pc} = \sum y_i T_{ci}$$  \hspace{1cm} (2)

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

$$P_r = P/P_{pc}$$  \hspace{1cm} (3)

$$T_r = T/T_{pc}$$  \hspace{1cm} (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:

$$J = \left( \frac{1}{3} \right) \left[ \sum y_i \left( \frac{T_r}{T_{ci}} \right) \right] + \left( \frac{2}{3} \right) \left[ \sum y_i \left( \frac{(T_r/P_{pc})^{0.5}}{T_{ci}} \right) \right]^2$$  \hspace{1cm} (5)

$$K = \sum y_i \left( \frac{T_r}{T_{pc}} \right)^{0.5}$$  \hspace{1cm} (6)

$$T_{pc} = K^2/J$$  \hspace{1cm} (7)

$$P_{pc} = T_{pc}/J$$  \hspace{1cm} (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):

$$F_j = (1/3) \left[ y_i \left( \frac{T_{pc}}{P_{pc}} \right) \right] + (2/3) \left[ y_i (T_r/P_{pc})^{0.5} \right]$$  \hspace{1cm} (9)

$$E_j = 0.6081 F_j^2 + 1.1325 F_j^2 - 14.004 F_j y_{c7}\% + 64.434 F_j y_{c6}\%$$  \hspace{1cm} (10)

$$E_k = (T_{pc}/P_{pc})^{0.5} (0.3129 y_{c7}\% - 4.8156 y_{c6}\% + 27.3751 y_{c5}\%)$$  \hspace{1cm} (11)

$$J^* = J - E_j$$  \hspace{1cm} (12)

$$K^* = K - E_k$$  \hspace{1cm} (13)

$$T_{pc} = K^*/J^*$$  \hspace{1cm} (14)

$$P_{pc} = T_{pc}/J^*$$  \hspace{1cm} (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:

$$J = a_0 + \sum a_3 y_i \left( \frac{T_r}{P_{pc}} \right) + \sum a_4 y_i \left( \frac{T_r}{P_{pc}} \right)^2 + \sum a_5 \left[ \frac{y_i (T_r/P_{pc})^{0.5}}{T_{pc}} \right]^2 + a_6 \left( y_{c7+} M_{c7+} \right)$$  \hspace{1cm} (16)

$$K = b_0 + \sum b_3 y_i \left( \frac{P_{pc}}{P_{pc}} \right) + \sum b_4 y_i \left( \frac{P_{pc}}{P_{pc}} \right)^2 + \sum b_5 \left[ \frac{y_i (T_r/P_{pc})^{0.5}}{T_{pc}} \right]^2 + b_6 \left( y_{c7+} M_{c7+} \right)$$  \hspace{1cm} (17)

Where, $y_i \in \left\{ y_{H2S}, y_{C6+}, y_{Y_2} \right\}$ and $y_i \in \left\{ y_{C2}, y_{C3}, \ldots, y_{C6} \right\}$, $a$ and $b$ 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 $\alpha$ and $\beta$ (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$_2$S), 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:

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

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$_2$S 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$_2$S and CO$_2$ (Wichert and Aziz, 1972). The correction factor is:

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

Where $A = y_{H_2S} + y_{CO_2}$ and $B = y_{H_2S}$ in the gas mixture.

The corrected $P_{pc}'$ and $T_{pc}'$ are:

$$T_{pc}' = T_{pc} - \varepsilon \quad (25)$$

$$P_{pc}' = P_{pc} T_{pc}' / [T_{pc} + B(1 - B)] \quad (26)$$

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):

\[
Z = 1 - \frac{P_{pr}}{T_{pr}} \left( 0.36748758 - 0.04188423 \left( \frac{P_{pr}}{T_{pr}} \right) \right)
\]  

(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 CO₂.

**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:

\[
\frac{1}{2} - 1 + \left[ A_4 T_{pr} - A_3 \left( \frac{P_{pr}}{Z^{2 T_{pr}}} \right)^2 + \left( A_2 T_{pr} - A_1 \right) \left( \frac{P_{pr}}{Z^{2 T_{pr}}} \right) \right] \exp \left[ \frac{-A_0 P_{pr}^2}{Z^2 T_{pr}^2} \right] = 0
\]

(28)

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:

\[
Z = \left[ \frac{0.06125 P_{pr}^{4/3}}{Y} \right] \exp \left[ -1.2 \left( 1 - t \right)^2 \right]
\]  

(29)

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

\[
F(Y) = 0.06125 P_{pr} t \exp \left[ -1.2 \left( 1 - t \right)^2 \right] + \frac{Y + 2.97 + 2.97 - Y^2}{\left( 1 - Y \right)^2} - \left( 14.76t - 9.76t^2 + 4.58t^3 \right) Y^2 + \left( 90.7t - 242.2t^2 + 42.4t^3 \right) Y^3
\]

(30)

This method has received great application in the natural gas industry (Elsharkawy et al., 2001; Elsharkawy, 2001). Therefore, it will be used in this part of the research to assess its accuracy in estimating the Z-factor for mixtures of natural gas with CO₂ 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:

\[
Z = 1 + A_1 \frac{T_{pr}}{T} + A_3 \left( \frac{T_{pr}}{T} \right)^2 + \left[ A_2 \frac{T_{pr}}{T} - A_1 \right] \left( \frac{T_{pr}}{T} \right)^2 + \left[ A_6 \frac{T_{pr}}{T} + A_5 \right] \rho_r^2 + \left[ A_8 A_5 \right] \left( \frac{T_{pr}}{T} \right)^2 \rho_r^5 + \left( A_7 \frac{T_{pr}}{T} + A_6 \right) \rho_r^2 \exp \left[ -A_9 \rho_r^2 \right]
\]

(31)

This method will also be considered in the Z-factor calculations at HPHT for natural gas-CO₂ mixtures.

**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):

\[
Z = \left[ A_1 + A_2 \frac{T_{pr}}{T} + A_3 \left( \frac{T_{pr}}{T} \right)^2 + \left[ A_4 \frac{T_{pr}}{T} + A_5 \right] \rho_r + \left[ A_6 \frac{T_{pr}}{T} + A_7 \right] \rho_r^2 + \left[ A_8 \frac{T_{pr}}{T} + A_9 \right] \rho_r^5 + \left[ A_{10} \left( 1 - A_{11} \rho_r^2 \right) \right] \rho_r^2 \exp \left[ -A_{12} \rho_r^2 \right] + 1 \right]^{1/2}
\]

(32)

Where,

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):

\[
Z = \frac{2E}{1.0482} + F
\]

\[
F = \frac{D}{E^2} - E
\]

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

\[
D = (-2A + B) - C
\]

\[
C = B - (-2A + B)
\]

\[
B = \left(\frac{3A}{T_{pr}}\right)^2 - 1.427 + 0.9178
\]

\[
A = \left(2 - \frac{0.05275}{-0.5765}\right)P_{pr} + 0.2360
\]

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):

\[
\ln(Z) = \alpha + \frac{\beta}{T_r} + \frac{\gamma}{P_r} + \frac{\theta}{T_r^3}
\]

Where,

\[
\alpha = A_1 + \frac{B_1}{P_r} + \frac{C_1}{P_r^2} + \frac{D_1}{P_r^3}
\]

\[
\beta = A_2 + \frac{B_2}{P_r} + \frac{C_2}{P_r^2} + \frac{D_2}{P_r^3}
\]

\[
\gamma = A_3 + \frac{B_3}{P_r} + \frac{C_3}{P_r^2} + \frac{D_3}{P_r^3}
\]

\[
\theta = A_4 + \frac{B_4}{P_r} + \frac{C_4}{P_r^2} + \frac{D_4}{P_r^3}
\]

(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):

\[
z = A + \frac{B+C}{D+E}
\]

Where,

\[
A = aT_r^{2.16} + bP_r^{1.028} + cP_r^{1.58}T_r^{-2.1} + d\ln(T_r)^{-0.5}
\]

\[
c = iln(T_r)^{-1.30} + jln(P_r)^{3.37} + kln(P_r) + lln(P_r)^2 + mln(P_r)\ln(T_r)
\]

\[
D = 1 + nT_r^{5.55} + oP_r^{0.69}T_r^{0.33}
\]

\[
E = p\ln(T_r)^{2.09} + q\ln(T_r)^{2.1} + r\ln(P_r) + s\ln(P_r)^2 + t\ln(P_r)\ln(T_r)
\]

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 of 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):

\[
z = f\left(\frac{1}{P_{pr}T_{pr}}\right)
\]

\[
z = \ln\left(1 + \frac{A_1}{P_{pr}^{2.92}} + \frac{A_3}{P_{pr}^{2.92}} + \frac{A_5}{P_{pr}^{2.92}}\right)
\]

\[
z = \ln\left(\frac{4A_2 + 4A_4 + 4A_6 + 4A_8 + 4A_10}{4A_2 + 4A_4 + 4A_6 + 4A_8 + 4A_10} + 1\right)
\]

(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 CO2 contents 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 virial 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 CO2, 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 programming. 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 an equation 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-Phillips (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 Ppr and Tpr. 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 Ppc and Tpc of natural gas-CO2 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 CO2 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-CO2 mixtures at supercritical conditions.

Data bank

A total of 2,200 Z-factor measurements representing
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 Tr below 1.0 and reduced Pr 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 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  |
Table 2. Working ranges of $T_r$ and $P_r$ for various $z$ calculation methods.

| Z Calculation method                  | Range of $T_r$ | Range of $P_r$ |
|---------------------------------------|----------------|----------------|
| 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 to 1.966 | 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     |

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 ($E_{abs}$= 3%) and highest correlation coefficient (96%). Evidently, recently developed and published methods: Al-Anzi and Alqauraishi, (2010), Bahadori et al. (2010), Azizi et al. (2010), and Ehsan and Nemati (2012) have exceptionally high $E_{abs}$ 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 $E_{abs}$ 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 ($E_{abs}$) 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).
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 CO₂ 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 CO₂. The first correlation proposed for low pressure ranges has the following form:

\[
Z = 1 + A_1 P_{pr} + A_2 P_{pr}^2 + \frac{A_3 P_{pr}^4}{T_{pr}^4} + \frac{A_4 P_{pr}^{6(A_{1+1})}}{T_{pr}^{6(A_{1+1})}} + \frac{A_5 P_{pr}^{8(A_{1+2})}}{T_{pr}^{8(A_{1+2})}}
\]

(51)

Where, A₁ = -670.27277, A₂ = 48.271233, A₃ = 669.298742, A₄ = 0, A₅ = 0.999561, A₆ = -0.002100, A₇ = 48.795104, A₈ = 0.010294, A₉ = 0.000743, A₁₀ = 0.751088, A₁₁ = 0.811288572, A₁₂ = 2.898688651, A₁₃ = 0.105471654, A₁₄ = 4.261541546, A₁₅ = -0.002537591

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 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 CO₂ 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-Kwong 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% CO₂. 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                          | \( N_d \) | \( \bar{E}_{AVE} \) | \( E_{ABS} \) | \( E_{RMS} \) | \( r^2 \) | \( N_d \) | \( \bar{E}_{AVE} \) | \( E_{ABS} \) | \( E_{RMS} \) | \( r^2 \) |
|--------------------------------|-----------|-------------------|--------------|-------------|------|-----------|-------------------|--------------|-------------|------|
| 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 |

The error analysis was performed using Kay’s mixing rule and SBV modified mixing rule. The following table compares the error metrics for different mixing rules: Hall-Yarborough, DPR, DAK, Londono et al., Bahadori et al., Azizi et al., Heidaryan et al., Hall, Yarborough, Anazi et al., Londono et al., Yarborough, Kamyab et al., and Ehsan and Nemati.

Figure 2. Crossplot of Z-factor for the range of \( P_{pr} < 3.0 \).
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
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% CO₂. 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% CO₂).
Table 4. Gas composition and measured Z-factors for gases containing high amounts of CO₂.

| 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      |
| H₂S         | 0     | 0     | 0     | 134.64  | 1.8511 | 34.47   | 0.8481 | 20.15   | 0.4725 |
| CO₂         | 0.2   | 0.75  | 0.8994 | 108.45  | 1.6173 | 27.58   | 0.7517 | 19.04   | 0.4571 |
| N₂          | 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 CO₂ rich gas which contains 90% CO₂. 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
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 CO$_2$, 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 (55 to 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-CO₂ 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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