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Correlations for Joule-Thomson Coefficients of Geothermal Waters Containing CO_2

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ABSTRACT

Reliable Joule-Thomson (J-T) coefficients of geothermal fluids containing CO_2 are important for non-isothermal reservoir flow simulation, well stream simulation, and process simulation. The J-T coefficients of fluids can be experimentally determined. However, it is difficult to conduct laboratory measurements because precise PVT data for cross interpolation is required for accurate estimation of the coefficients. Thus, experimentally measured J-T coefficients of water-CO_2 mixtures are almost non-existent. Consequently, it is necessary to estimate J-T coefficients by using an accurate equation of state (EoS); however, programming and running a complicated EoS is not an easy task. Accurate EoS modeling requires many parameters, and finding the right volumetric root could be problematic and is not trivial. This paper presents useful correlations for the estimation of J-T coefficients of water-CO_2 mixtures. Isenthalpic expansions are modelled for binary mixtures of water and CO_2 at different concentrations by using Gallagher, Crovetto, and Sengers EoS (1993). The EoS implemented in this study predicts both phase equilibrium and volumetric properties accurately for a temperature range of 400-1000 K and a pressure range of 0-100 MPa and 30% mole fraction of CO_2. Pure component properties are estimated for water and compared with the ones in the literature. In addition, the effects of temperature and pressure on the J-T coefficients are investigated. The EoS given in this study applies for the temperature and pressure conditions which are limited to the ones encountered in typical geothermal reservoirs, and the correlations are derived based on the results of the EoS. The effect of CO_2 content on both the J-T coefficients and the inversion curves is also studied. The proposed correlations are derived in the temperature range of 400-550 K and the pressure range of 8-30 MPa.

1. INTRODUCTION

Interest in temperature transient analysis in recent years has grown rapidly due to the recent advances and improvements in resolution of downhole temperature gauges. Interpretation and analysis of transient sandface temperatures alone or jointly with transient pressure data could yield important information regarding near-wellbore and far-field reservoir characteristics (Onur and Cinar, 2016; Palabiyik et al., 2016). For the majority of well testing applications temperature is measured along with pressure, but analyzing temperature data is not a common practice in well test analysis. Developing techniques for temperature transient analysis could help to acquire more information regarding the well/reservoir system without any additional costs.

There are a number of studies in petroleum and geothermal engineering literature emphasizing certain trends in the temperature changes, when the effects of Joule-Thomson (J-T) heating/cooling, transient adiabatic fluid expansion/compression, conduction, and convection are included in modeling of sandface temperatures during constant-rate drawdown and buildup tests; for instance see, Garg and Pritchett (1977,1984); Sui et al. (2008a, 2008b); App (2009; 2010); Duru and Horne (2010a, 2010b, 2011a, 2011b); Ramazanov et al. (2010); Palabiyik et al. (2013; 2015); App and Yoshioka (2013); Sidorova et al. (2015); and Onur and Palabiyik (2015).

Modeling temperature behavior within the reservoir and in the well requires thermal parameters regarding the fluid/rock system such as thermal conductivity, heat capacity, enthalpy etc. There are two approaches in modelling Joule-Thompson (J-T) effects while modelling temperature behavior: 1) J-T effects are included implicitly through enthalpy variation with respect to temperature and pressure. Eq. 1 shows the energy balance including enthalpy (Palabiyik et al., 2016). 2) Energy balance is derived with respect to temperature and the J-T effects are accounted for explicitly with an additional term in the expression. Eq. 2 shows the energy balance with respect to temperature for a 1D radial heat flow (Palabiyik et al., 2016). The second approach is temperature explicit and needed for analytical solutions of temperature for well test analysis applications. In such a case J-T coefficient is needed for the solution of temperatures (Palabiyik et al., 2016):

$$\frac{\partial}{\partial t}\left[\phi\rho_w H_w + (1-\phi)\rho_s H_s\right] - \frac{\partial(\phi p)}{\partial t} + \nabla\bullet\left(\rho_w H_w \mathbf{v}_w\right) - \nabla\bullet\left(\lambda\nabla T\right) = 0 \tag{1}$$

$$\left[\phi\rho_w c_{pw} + (1-\phi)\rho_s c_{ps}\right]\frac{\partial T}{\partial t} - \phi\left(\rho_w c_{pw}\mu_w + 1\right)\frac{\partial p}{\partial t} - \rho_w c_{pw}\frac{k}{\mu_{vis,w}}\frac{\partial p}{\partial r}\frac{\partial T}{\partial r} + \mu_w\rho_w c_{pw}\frac{k}{\mu_{vis,w}}\left(\frac{\partial p}{\partial r}\right)^2 - \frac{\lambda}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) = 0 \tag{2}$$

The J-T coefficients of fluids can be experimentally determined. However, it is difficult to conduct laboratory measurements because precise PVT data for cross interpolation is required for accurate estimation of the coefficients. Thus, experimentally measured J-T coefficients of water-CO_2 mixtures are almost non-existent. Consequently, it is necessary to estimate J-T coefficients by using an

accurate equation of state (EoS); however, programming and running a complicated EoS is not an easy task. An accurate EoS modeling requires many parameters, and finding the right volumetric root could be problematic and is not trivial. Consequently, quick estimation of J-T coefficients is in need for well testing applications and modelling efforts for geothermal systems containing CO_2. As is well known, the geothermal systems in Turkey, contain considerable amount of CO_2 dissolved in geothermal water.

In this study based on the EoS by Gallagher, Crovetto, and Sengers (1993) J-T coefficients are estimated at different pressures and temperatures as well as CO_2 mole fractions. The EoS used can model thermodynamic behavior of water CO_2 systems up to 30 mole percent CO_2 in a large range of temperature, 400 -1000 K, and pressures, 0-100 Mpa. All thermodynamic properties of the binary mixture with x mole percent of CO_2 are derived from Helmholtz free energy followed by the following equation. The model for Helmholtz free energy of the mixture is based on the principle of generalized corresponding of states and the single phase water thermodynamic properties are based on Kestin et al. (1984).

$$\frac{A(V,T,N,x)}{N\kappa T}=(1-x_{CO2})\frac{A_w^{mol}(N,T)}{N\kappa T}+x_{CO2}\frac{A_{CO2}^{mol}(N,T)}{N\kappa T}-1+\ln\left(\frac{N}{V}\right)+x_{CO2}\ln x_{CO2}+(1-x_{CO2})\ln(1-x_{CO2})+\frac{A^{res}(N,V,T,x)}{N\kappa T} \quad (3)$$

The first two terms are the contributions of each component and the last term is the residual Helmholtz free energy and the other terms account for the ideal gas part. The details of the model are given in the original paper by the authors. The thermodynamic properties of the mixture are estimated using the following relations.

$$p=-\left(\frac{\partial A}{\partial V}\right)_{T,x}=-\left(\frac{\partial A^{res}}{\partial V}\right)_{T,x}+\frac{N\kappa T}{V} \quad (4)$$

$$S=-\left(\frac{\partial A}{\partial T}\right)_{V,x} \quad (5)$$

$$U=A-T\left(\frac{\partial A}{\partial T}\right)_{V,x} \quad (6)$$

$$H=A-T\left(\frac{\partial A}{\partial T}\right)_{V,x}-V\left(\frac{\partial A}{\partial V}\right)_{T,x} \quad (7)$$

$$c_v=c_v^{id}-T\left(\frac{\partial^2 A}{\partial T^2}\right)_{V,x} \quad (8)$$

$$c_p=c_v-\frac{T\left(\frac{\partial p}{\partial T}\right)_{V,x}^2}{\left(\frac{\partial p}{\partial V}\right)_{T,x}} \quad (9)$$

All the required derivatives are estimated using numerical differentiation. Constant volume ideal heat capacities for CO_2 is estimated using the correlations provided by Ely et al. (1989).

2. ESTIMATION OF JOULE-THOMSON COEFFICIENT

For several fluids, J-T coefficients are experimentally determined (Ertle, 1979; Nabil and Wormald, 1977), however, not only it is difficult to conduct laboratory measurements but also precise PVT data for cross interpolation is required for accurate estimation of the coefficients (Hendricks et al., 1972). Similarly, Gunn et al. (1966) concluded that estimating J-T coefficients with an accurate EoS should be more accurate than experimentally measuring J-T coefficients. Nevertheless, to the knowledge of this paper's authors there is no available experimentally measured J-T coefficients for water and CO_2 mixtures in the literature.

In this work, J-T coefficients for water CO_2 mixtures at liquid phase are estimated based on Gallagher, Crovetto, and Sengers EoS (1993). The study is limited with single phase liquid systems, thus, the maximum mole fraction of CO_2 in the mixture at a given temperature and pressure is controlled by the solubility of CO_2. The lower limit for temperature is chosen as 400 K as it is the lower limit of the EoS. The upper limit is determined as 550 K to account for temperatures encountered in geothermal systems. For pressure the lower limit is 8 Mpa; chosen based on the solubility value of CO_2 in water. At lower pressures, the solubility of CO_2 in water is so low that presence of CO_2 in solution do not have a significant effect on the J-T coefficient. The upper limit for pressure is 30 MPa; high enough to cover geothermal applications and provide a decent range for the correlation to account for the mole fraction of CO_2 because at higher pressures the solubility is higher. In summary, the J-T coefficients generated from the EoS are in the range of 400-550 K and 8-30 MPa.

The J-T coefficient is defined by Eq. 10. By definition it is the slope of isenthalpic curve in the pressure temperature plane. Eq. 10 is further manipulated using thermodynamic relations to obtain Eq. 11.

$$\mu = \left(\frac{\partial T}{\partial p} \right)_H \tag{10}$$

$$\mu = -\frac{1}{c_p} \left[T \frac{\left(\frac{\partial p}{\partial T} \right)_V}{\left(\frac{\partial p}{\partial V} \right)_T} + V \right] \tag{11}$$

Initially J-T coefficients are estimated for pure water and compared with IAPWS formulation (Wagner and Pruß, 2002). The thermophysical data generated using IAPWS formulation (Wagner and Pruß, 2002) are available through NIST website (NIST, 2016). This step was necessary since in their original work Gallagher et al. (1993) did not provide any relations for the estimation of J-T coefficients. By comparing the results with the IAPWS formulation (Wagner and Pruß, 2002) the approach is validated as well as the trends with respect to pressure and temperature are determined. The results are compared at the four different pressures; 8, 16, 22, 30 MPa with respect to temperature. Figure 1 shows the match between J-T coefficients by Gallagher et al. (1993) and IAPWS (Wagner and Pruß, 2002). Both formulations yield almost the same results for pure water within the pressure and temperature limits considered in this study.

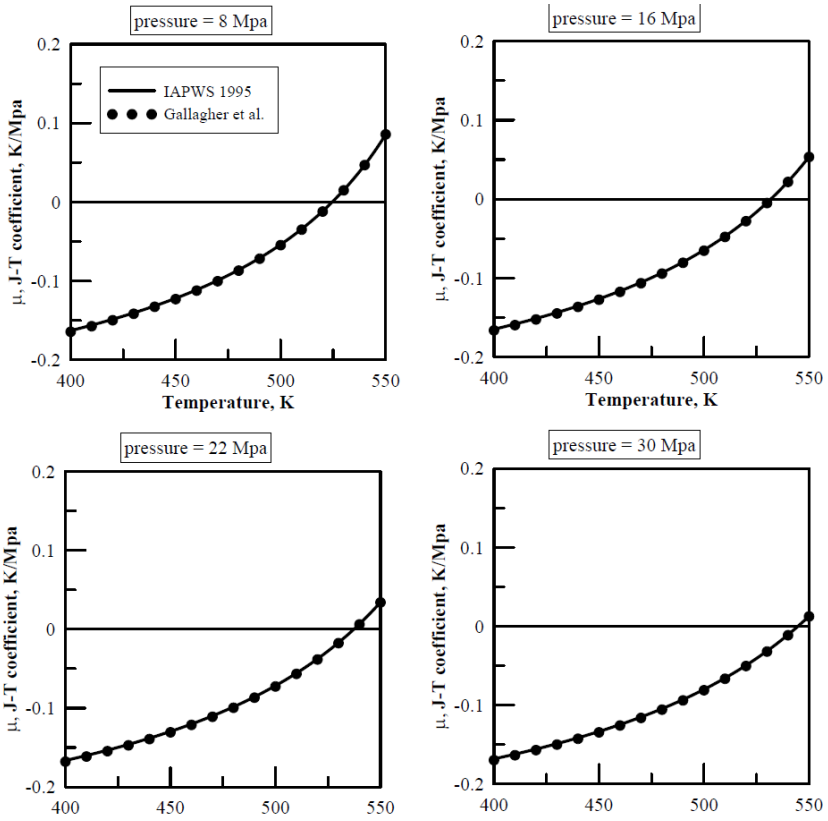

Figure 1: Comparison of J-T coefficients estimated from Gallagher, Crovetto, and Sengers EoS (1993) EoS with IAPWS 1995 formulation (Wagner and Pruß, 2002) at various pressures with respect to temperature.

Next the effect of temperature on J-T coefficients of pure water at constant pressure is investigated. As shown in Fig. 1 as temperature increases at a constant pressure, J-T coefficient also increases. The temperature dependence could be represented with an exponential function of the following form. Note that using 5 or 6 order polynomials provides better fits than Eq. 12 but to reduce the number of parameters Eq. 12 is chosen as it gives the best fit with minimum number of parameters.

$$\mu_w(T) = A_1 T^{B_1} - 0.2 \tag{12}$$

Figure 2 gives the effect of pressure on J-T coefficient of pure water. In general, as pressure increases J-T coefficient of pure water decreases. Besides, as temperature increases pressure dependence becomes more pronounced and J-T coefficient gets greater values such that for the temperatures shown in the figure only at 550 K J-T coefficient is positive. On the other hand, for temperatures 400,

450, and 500 K for the ranges of pressures considered, the J-T coefficient is negative. This is particularly important as the sign of the J-T coefficient dictates weather a cooling or heating occurs. If the J-T coefficient is negative the isenthalpic expansion causes a temperature rise, however, a positive J-T coefficient would result in a cooling. The point where isenthalp is a maximum (that is Eq. 10 is equal to zero) is called the inversion curve.

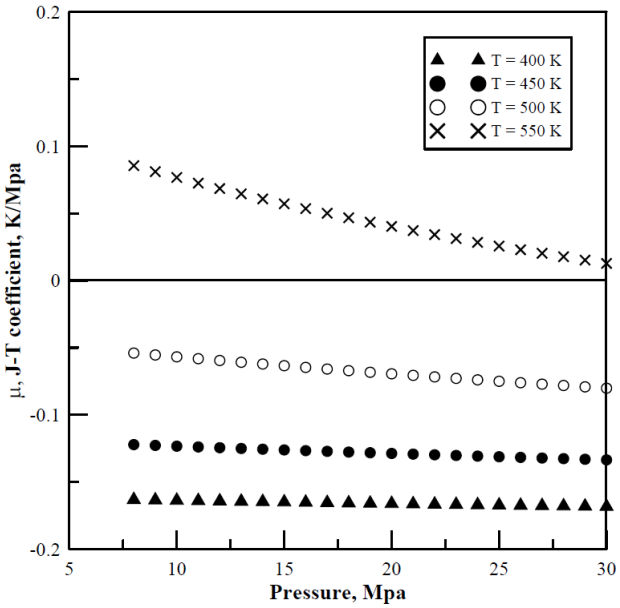

Figure 2: Change of J-T coefficients of water with respect to pressure at various temperatures.

When the behavior of J-T coefficient is investigated for pure water with respect to pressure at a constant temperature, it is observed that pressure dependence could be represented with a second order polynomial. This trend is clearly shown in Figure 3. At lower temperatures the dependence shifts towards a linear trend. Besides Figure 4 gives the dependence of J-T coefficients on temperature at pressures of 8, 16, 22, and 30 MPa. Again it is observed that at lower temperature, pressure dependence is weak and as pressure increases pressure dependence becomes stronger. Our further investigation suggests that below 8 MPa, there is almost no pressure dependence. J-T coefficients generated at 6 and 8 MPa with varying temperature are almost identical. The values change sign at a certain temperature for a specific pressure. At a constant pressure, there is a certain temperature beyond which J-T coefficient changes sign. Based on the aforementioned observations, a correlation in the form given by Eq. 13 is proposed. Here the form given by Eq. 12 is assumed for temperature dependence and parameters for A_1 and B_1 are treated as pressure dependent.

$$\mu_w(T,p) = A_1(p)T^{B_1(p)} - 0.2 \tag{13}$$

If the pressure dependence of parameters is represented with a second degree polynomial, the resulting equation would be in the following form. There are six parameters of the correlation to be determined.

$$\mu_w(T,p) = \left(A + Bp + Cp^2\right)T^{\left(D+Ep+Fp^2\right)} - 0.2 \tag{14}$$

While finding the parameters in Eq. 14, the Levenberg-Marquardt (LM) algorithm is used for non-linear regression. For estimation of the initial guesses, the following procedure is considered: 1) The values of J-T coefficients are generated using the EoS with respect to temperature at given values of constant pressures. Pressure increments are 2 MPa, and the increment in temperature is 1 K. Each curve at a given pressure value is fitted by Eq. 12 and parameters A_1 and B_1 is determined. 3) As a result of second step, a list of A_1 and B_1 parameters are obtained as a function of pressure. 4) Second order polynomials are fitted to determine the dependence of A_1 and B_1 on pressure. At the end, a list of parameters is obtained and these parameters are used as initial guesses for the LM algorithm. The parameters estimated with their 95% confidence intervals are given in Table 1. The agreement is quite good and the resulting root mean square error (RMSE) is 0.003072. The match of the correlation with the EoS model is shown in Figure 5. The proposed correlation represents J-T coefficients that are generated with the EoS quite good.

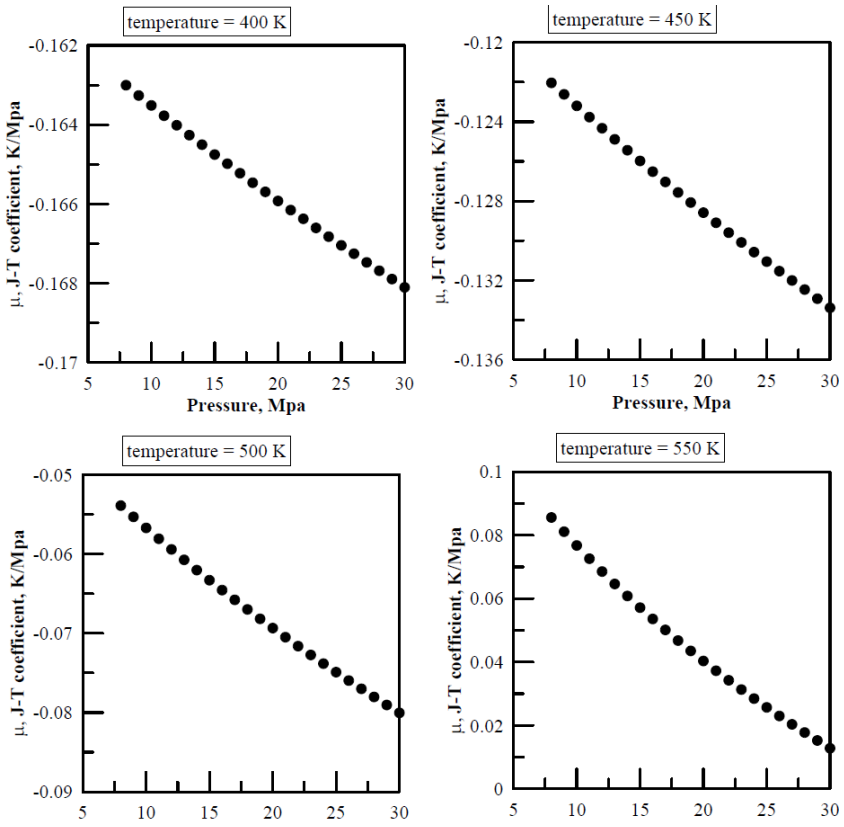

Figure 3: Change of J-T coefficient values of water with respect to pressure at temperatures, 400, 450, 500, 550 K individually.

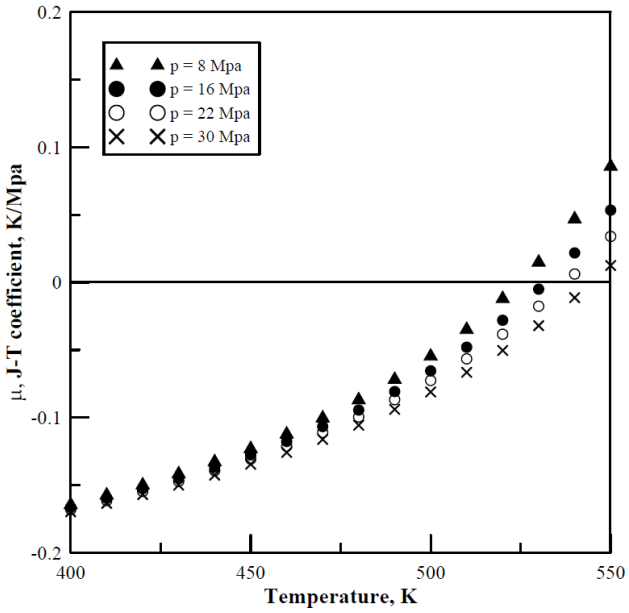

Figure 4: Change of J-T coefficient values of water with respect to temperature at various pressures.

Table 1: Estimated parameters for pure water J-T correlation together with confidence limits.

Parameter	Estimated Value	95% Confidence Limits
A	1.134E-19	9.288e-20, 1.34e-19
B	3.825E-21	-1.828e-22, 7.833e-21
C	9.541E-22	6.535e-22, 1.255e-21
D	6.767	6.716, 6.818
E	-0.02006	-0.02368, -0.01644
F	0.0001641	0.0001153, 0.0002129

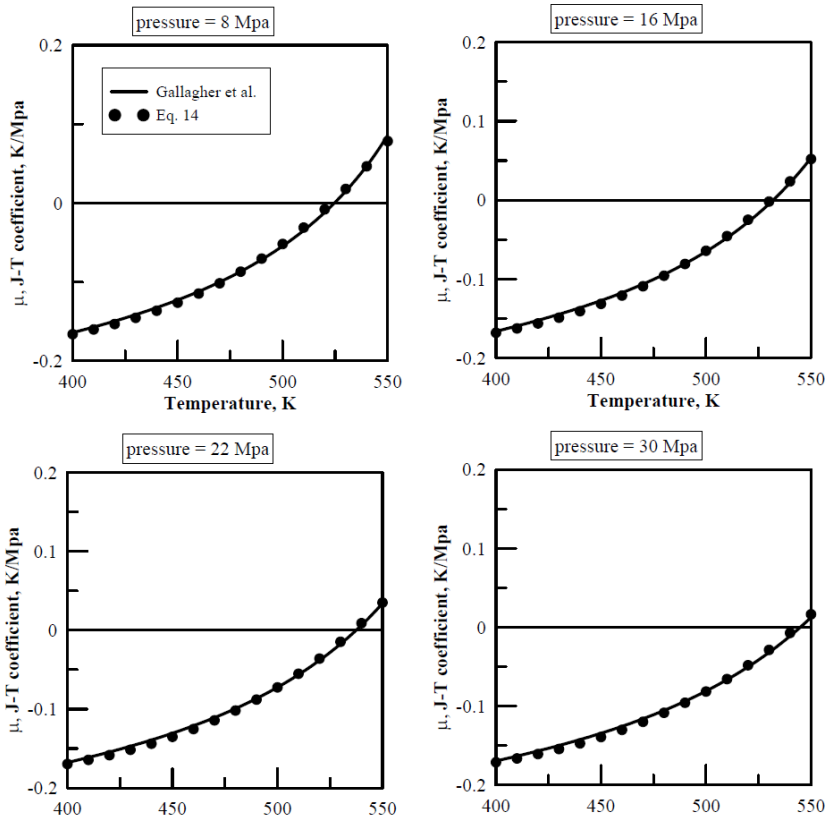

Figure 5: Match of the proposed correlation for pure water (Eq. 14) with the EoS used at pressures 8, 16, 22, and 30 MPa.

2.1 Effect of CO_2 on the inversion curve

The inversion curve is defined as the locus of points for which the Joule-Thomson coefficient is zero. For the temperature and pressure range of interest the effect of CO_2 content on the inversion curves is investigated. Figure 6 compares the inversion curves of water with water – CO_2 mixtures having different CO_2 content. It is clearly that CO_2 added to water as a component in solution shifts the inversion curve to the right. Note that, the solid line represents the inversion curve for water. At higher temperatures the J-T coefficient is positive so the upper part of the inversion curve gives the region where J-T coefficient is positive so that a decrease in pressure would result in a cooling. The lower part of the curve is the region where J-T coefficient is negative that is a decrease in pressure would result in a heating. The presence of CO_2 in the solution widens the region where the J-T coefficient is positive.

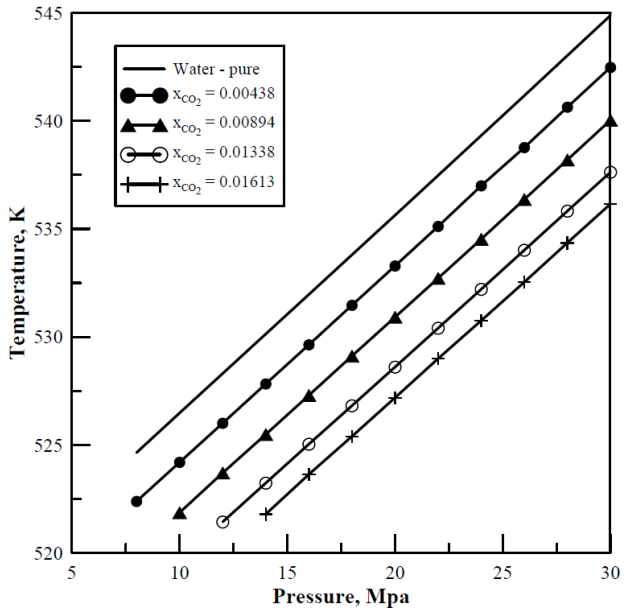

Figure 6: Effect of CO_2 content on the inversion curves of water-CO_2 systems.

2.1 Effect of CO_2 on J-T coefficients

The solubility of CO_2 in water is a function of pressure, temperature and salinity. In this study we neglect the effect of various salts on the solubility of CO_2 in water. The solubility of CO_2 at a certain pressure and temperature would give the maximum amount of CO_2 that a single phase water could contain. Thus, at a certain temperature and pressure the CO_2 content of single phase water is limited with its solubility. In this study, this limit is considered and the maximum CO_2 mole fraction at each temperature and pressure is chosen accordingly. Figure 7 shows the effect of CO_2 content on J-T coefficients at various pressures with respect to temperature.

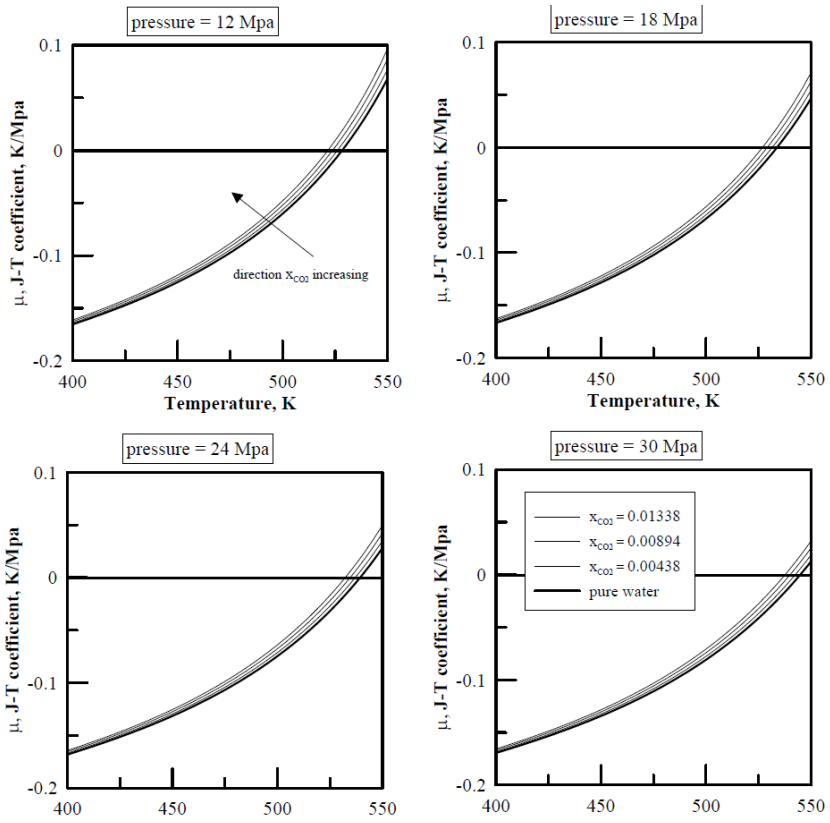

Figure 7: Effect of CO_2 on J-T coefficient at various pressures, 12, 18, 24, 30 MPa.

In general, as the CO_2 content increases J-T coefficient at a certain temperature and pressure increases as well. The dependence is more pronounced at higher temperatures. At lower temperatures effect of CO_2 content on J-T coefficient is not significant. Also at higher pressures, the effect of temperature on J-T coefficients are slightly higher for the pressures and temperatures considered.

It is not possible to consider a wide range for mole fractions of CO_2 as it is limited with the solubility of CO_2 in water. The maximum amount of CO_2 dissolved in water at a certain temperature and pressure depends on its solubility. If CO_2 content is greater than its solubility, a second phase is inevitable. Thus, the J-T coefficients along the saturation line are estimated. The solubility of CO_2 at a given temperature and pressure is estimated using the correlations provided by Duan and Sun (2003). Then giving temperature, pressure, and mole fraction of CO_2 as inputs J-T coefficients are estimated using the EoS. This is repeated for all pressure and temperatures considered in this study. Interestingly, J-T coefficients along the saturation line exhibit a negligible pressure dependence. Figure 8 compares J-T coefficients along the saturation line with respect to temperature at 8 and 30 Mpa. Even between lower and upper bounds of the study pressure dependence is negligible. This is due to two contrary effects; as pressure increases J-T coefficients decrease, however, with increasing pressure solubility also increases resulting in a higher CO_2 dissolved in solution increase of which increases the values of J-T coefficients. The changes with respect to pressure and CO_2 content are similar in magnitudes thus pressure and CO_2 content counterbalances each other resulting in a negligible pressure dependence along the saturation line. Assuming that the J-T coefficients are only dependent on temperature on the saturation line Eq. 15 is obtained. Parameters are estimated by matching all the data to the equation below. Total of 1396 data points are used. The RMSE value is 0.0018 and the 95% confidence intervals for the estimated parameters are: 0.0002297-0.0002346, for the multiplier and 0.01297-0.01301for the exponent. The J-T coefficient on the saturation line is critical as it provides an upper limit for the J-T coefficients at a certain temperature and pressure.

$$\mu_{sat}(T) = 0.0002322 \times e^{T \times 0.01299} - 0.2 \tag{15}$$

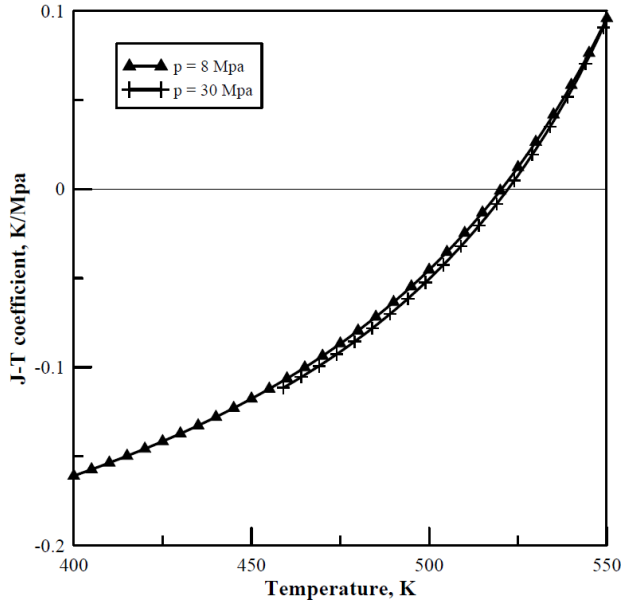

Figure 8: J-T coefficient along the saturation line with respect to temperature at 8 and 30 MPa.

Equation 15 gives the coefficients along the saturation line, however, if the CO_2 content is lower than the maximum solubility at a certain temperature and pressure a new correlation is in need. Thus, a general correlation is proposed that accounts for the variation in CO_2 content based on the pure water correlation. However, it is not trivial as there are three variables, temperature, pressure, and CO_2 mole fraction. As explained above effect of CO_2 content and pressure is similar in magnitude but in opposite directions. The general correlation within the pressure and temperature limits considered in this study is built based on this fact. An equivalent pressure is calculated as a function of CO_2 mole fraction to be used in equation 1, to account for variations in CO_2 content. Through investigation of the behavior of J-T coefficients with respect to pressure and CO_2 content, the following relation (Eq. 16) is found useful for the estimation of such a pseudo or equivalent pressure.

$$p_{pseudo}\left(x_{CO_2}, p\right) = p - p \times \left(V - Y \times p\right) \times {x_{CO_2}}^{Z} \tag{16}$$

Equation 16 used in Eq. 14 (replacing pressure with pseudo pressure) gives the general correlation that would account for the variation in CO_2 content. If the CO_2 content is zero pseudo pressure would be equal to pressure itself so the general correlation accounts for pure water as well. The parameters are estimated by matching J-T data generated by the EoS at different mole fractions of CO_2 (0.01, 0.02, 0.03, 0.04) within the pressure and temperature limits by using LM algorithm. Note that at a specific temperature and pressure the amount of CO_2 dissolved in single phase water is controlled by its solubility. Thus, it is not possible to estimate the values at each temperature, pressure and CO_2 mole fraction given above. Table 2 gives the number of data points at each pressure and CO_2 mole fractions used for matching. Total of 2827 data points are used for the estimations.

Table 2: Number of data points used for the estimation of parameters in Eq.16.

p, MPa	x_{CO2}, mole fraction				Total
	0.01	0.02	0.03	0.04	
8	87				
10	144				
12	151				
14	151				
16	151	74			
18	151	76			
20	151	79	5		
22	151	81	42		
24	151	83	58		
26	151	81	71	21	
28	151	87	82	28	
30	151	89	90	39	
Sum	1741	650	348	88	2827

The match is quite good with an RMSE value of 0.0037. The values of the obtained parameters with the 95% confidence limits is given by Table 3. The match of the proposed equation with the EoS values at different pressures are given in Figure 9. Also Figure 10 gives the match at 30 MPa with respect to temperature at different CO_2 mole fractions. All the matches are good considering the fact that the changes in the J-T values are small in magnitude.

Table 3: Estimated parameters for Eq. 16 together with confidence limits.

Parameter	Estimated Value	95% Confidence Limits
V	19.4370	18.2193, 20.7354
Y	0.3393	0.3112, 0.3694
Z	0.8065	0.7920, 1 0.8212

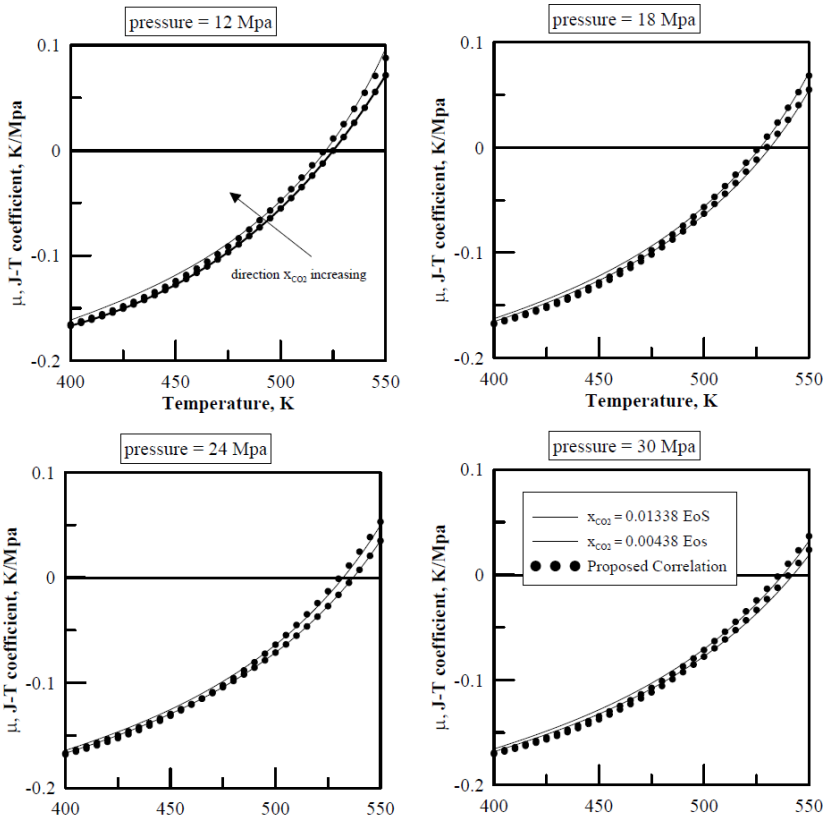

Figure 9: Matches of EoS estimated J-T coefficients with proposed correlation at different pressures and mole fractions of CO_2.

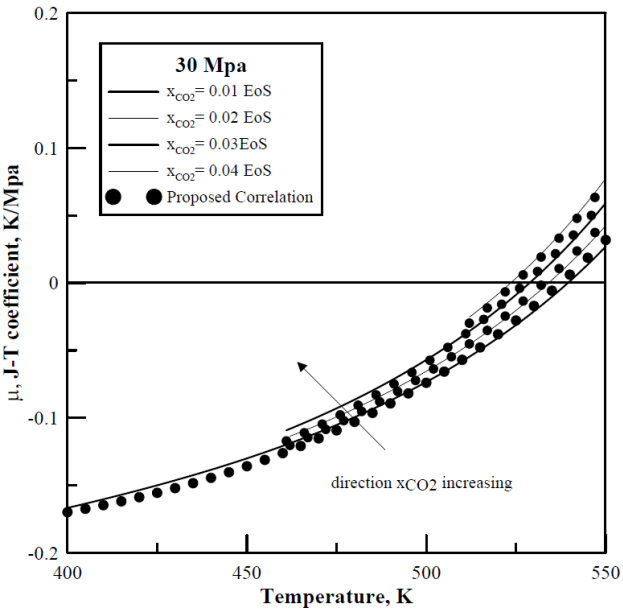

Figure 10: Matches of EoS estimated J-T coefficients with proposed correlation at 30 MPa and at different mole fractions of CO_2.

3. CONCLUSIONS

In this study, the effect of CO_2 content on the J-T coefficients is investigated for water-CO_2 systems as well as the inversion curves by using the EoS proposed by Gallagher et al. (1993). Note that the results are not supported with experiments because such data are not available. All the findings are based on the EoS used in this study. In this study, it is observed that as the amount of CO_2 dissolved in water increases at a certain temperature and pressure, the values of J-T coefficients also increases resulting in a shift in the inversion curve such that the inversion temperature at a certain pressure decreases as the CO_2 content increases. In addition, effects of temperature and pressure on the J-T coefficients of pure water are investigated. A positive correlation with temperature and a negative correlation with pressure are observed.

The J-T correlations for single phase water-CO_2 systems in the temperature range of 400-550 K and the pressure range of 8-30 MPa are proposed. The correlations are also valid for pure water within the same range of temperatures and pressures. In addition, a correlation along the saturation line is proposed. It is observed that the pressure dependence along the saturation line is negligible.

Proposed correlations fit the EoS quite good when considering the confidence intervals of the estimated parameters and RMSE values. However, care is advised when using the correlation as those are valid only for single phase water-CO_2 systems thus the solubility limits the maximum amount of CO_2 to be considered in the correlations. The users are advised to use the correlation for the saturation line as limiting value giving the maximum values of the J-T coefficient at the pressure and temperature in the consideration. This value should provide an upper limit for using the proposed general correlation. Using the correlation without knowing the solubility value could give erroneous results as there is no implicit limit for CO_2 mole fraction.

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NOMENCLATURE

A	Helmholtz free energy, Joule.
c_p	Heat capacity at constant pressure, Joule/ mol K.
c_v	Heat capacity at constant volume, Joule/ mol K.
H	Enthalpy, Joule.
G	Gibbs free energy, Joule.
k	Permeability, m^2
N	Number of molecules
p	Pressure, MPa (In Eqs. 1 and 2 Pal).
r	Radial distance, m
S	Entropy, Joule.
T	Temperature, K.
t	Time, s.
U	Internal energy, Joule.
V	Volume specific, m^3/mol.

v Velocity, m/s
x Mole fraction

Greek symbols

ϕ Porosity
κ Mixing parameter for volume
λ Overall thermal conductivity of rock and fluid, W/m.K.
μ Joule-Thomson coefficient, K/MPa (In Eqs. 1 and 2 K/Pa)
μ_{vis} Viscosity, Pa s.
ρ Density, mol/m^3.

Subscripts

w water
s solid

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