THE PFGC EQUATION OF STATE AT THE AGE OF FOURTEEN

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Abstract The purpose of this paper was to review and report the recent advances and progress that have been achieved on the fourteen year old PFGC equation of state. This paper will also discuss the good (pros) and the bad (cons) of this equation of state. Emphasis will be made on the following categories:

A. Prediction of the pure component PVT properties
  1. Hydrocarbons in particular n—alkanes
  2. Selected non—hydrocarbons such as N_2, H_2S, CO_2, H_2O
  3. 1—Alkanols
  4. Refrigerants
  5. Coal derived constituents
B. Vapor—Liquid—Equilibria
  1. Hydrocarbon systems
  2. Hydrocarbon + water systems
  3. 1—Alkanol + water systems
  4. Water + acid gases (e.g. H_2S & CO_2) systems
  5. 1—alkanol + n—alkane systems
C. Vapor—Liquid Hydrocarbon—Liquid water Equilibria
D. Hydrate formation and inhibition by methanol based on the basic model of Präusnitz and Parrish.

In all of the above categories comparison between the experimental data and those predicted by the PFGC equation of state will be presented.

چکیده هدف این مقاله مروری بر تحقیقات انجام گرفته روی معادله حالت (PFGC) است که چهارده سال پیش توسط پروفسورویلسون ارائه شد . در این مقاله نکات ضعف و قوت معادله حالت مذکور و کاربردهای آن مورد بحث قرار گرفته است . تاکید روی موارد ذیل بوده است :

الف) پیش‌بینی خواص ترمودینامیکی اجسام خالص شامل :

۱) هیدروکربورهای اشباع

۲) اجسام غیر هیدروکربوری از قبیل N_2, CO_2, H_2S, H_2O

۳) الکل‌ها

۴) برودت‌زاها

۵) اجسام مستخرج از ذغال سنگ

ب) تعادل بخار و مایع از قبیل :

۱) سیستم های هیدروکربوری

۲) سیستم های آب و هیدروکربور

۳) سیستم های آب و الکل

۴) سیستم های گازهای اسیدی و آب

۵) سیستم های هیدروکربور و الکل

ج) تعادل بخار ــ هیدروکربن مایع ــ آب

د) شرایط تشکیل هیدرات و پیش‌گیری از آن بوسیله متانول بر اساس مدل پریش و پرازنیتز .

در کلیه سیستم های فوق‌الذکر مقایسه‌ای بین عملکرد این معادله حالت و اطلاعات تجربی انجام گرفته است .

INTRODUCTION

Fourteen years ago, at the fifty third Annual Convention of the Gas processors Association in 1974, John Cunningham and Grant Wilson introduced the Parameters From Group Contribution (PFGC) equation of state [1]. The PFGC equation represents the most novel and fascinating recent development within equations of states. The starting point is a description of the Helmholtz energy of a liquid lattice structure with holes:

$$A^{PFGC}/RT = A^{FH}/RT + A^w/RT \quad (A1)$$

A^{FH} is a Flory-Huggins contribution (entropy effect), and A^w is an intermolecular interaction term (enthalpy effect). It is assumed

that all the expressions for the Helmholtz energy are analogous to expressions for the Gibbs energy. Hence any activity coefficient expression may in principle be used for A^w, and they used a modified Wilson equation to describe the interactions between functional groups constituting the molecules rather than between molecules themselves. Based on the appropriate mathematical manipulations and using modified hole theory, Wilson and Cunningham derived the set of equations shown in Table I. These equations from the basic framework of the PFGC equation of state.

Several terms in each equation need further description. The first three terms in equation 1 represent the Flory-Huggins contribution to the compressibility factor; while the last two terms represent the modified Wilson equaiton contributions to the compressibility factor. Similar attributes for each of the other equations (for chemical potential and enthalpy departures) can be deduced from these equations. The mixing rules are described by equations 4 through 10. The term c/b_H appearing in equations 1 through 3 has been labeled as a universal constant by Wilson and Cunningham. They selected a value of 12 for this particular constant.

Contrary to a number of successful equations of state such as Soave Redlich Kwong (SRK) [2], Peng Robinson (PR) [3], and Starling Benedict Webb Rubin (BWRS) [4], in PFGC equations, there are no defining parameters in terms of the critical properties, boiling point or any other physical property except type of groups making up the particular molecule. This is a great advantage specially when dealing with heavy fractions such as the C_{6+} fractions commonly encountered in natural gas, crude oil or coal tar liquids systems where the critical temperature and pressure of these compounds must be estimated using empirical correlations. Usually, the estimations are based on knowledge of the "average" molecular weight, "average" boiling point and specific gravity of the fraction. Wilson et al. [5] and later Brule et al. [6] have shown that the existing correlations have extreme sensitivity to the values used for critical properties of the C_{6+} fractions.

The PFGC Parameters

In this equation of state, the parameters are functions only of the groups making up the individual molecules present in the system. For a defined compound such as ethane or propane, the groups present in each molecule are known. For example, there are two CH_3- and one $-CH_2-$ groups in propane. The groups present in the C_{6+} fraction must be determined by some test or correlation.

There are five parameters for each group which must be known before any thermodynamic property calculations can be made. These five parameters are:

b_k = volume of group k

s_k = parameter proportional to external degree of freedom of group k

$E_k^{(0)}$ = first interaction energy coefficient of group k

$E_k^{(1)}$ = second interaction energy coefficient of group k

$E_k^{(2)}$ = third interaction energy coefficient of group k

Wilson and Cunningham presented only four parameters, namely, b_k, s_k, $E_k^{(0)}$, $E_k^{(1)}$ in the original version of the PFGC equation of state, but Moshfeghian et al. [7] introduced the fifth parameter, $E_k^{(2)}$, for better accuracy and extension of capabilities of this equation of state. To distinguish between the Wilson and Cunningham version of the PFGC equation of state, Moshfeghian-Erbar-Shariat have dubbed their version as PFGC-MES.

$$\frac{Pv}{RT} = Z = 1 - \frac{sv}{b} \ln\left(1 - \frac{b}{v}\right) - s + b\left(\frac{c}{b_H}\right) \sum_k \psi_k \left(\frac{b - b\sum_n \psi_n \tau_{nk}}{v - b + b\sum_n \psi_n \tau_{nk}}\right) \tag{1}$$

$$\frac{\mu_i}{RT} = s_i\left(\frac{v}{b} - 1\right)\ln\left(1 - \frac{b}{v}\right) + 1 - \frac{sb_i}{b}\left[\frac{v}{b}\ln\left(1 - \frac{b}{v}\right) + 1\right]$$

$$+ \ln\left(\frac{RT}{v}\right) - \left(\frac{c}{b_H}\right)\left\{\sum_k^g \left[m_{ik} b_k \ln\left(\frac{v - b + b\sum_n^g \psi_n \tau_{kn}}{v\tau_{kk}}\right)\right]\right.$$

$$\left. + b\sum_k^g\left[\psi_k \frac{-b_i + \sum_n^g m_{in} b_n \tau_{kn}}{v - b + b\sum_n^g \psi_n \tau_{kn}}\right)\right]\right\} \tag{2}$$

$$\left(\frac{\Delta H}{RT}\right) = (Z - 1) + \left(\frac{c}{b_H}\right)\frac{b^2}{T}\left\{\frac{\sum_n^g \psi_k\left(\frac{\sum_n^g \psi_n\left(\frac{d\tau_{kn}}{dT}\right)}{v - b + b\sum_n^g \psi_n \tau_{kn}}\right)}{}\right\} \tag{3}$$

$$b = \sum_i^c x_i b_i \tag{4}$$

$$b_i = \sum_k^g m_{ik} b_k \tag{5}$$

$$s = \sum_i^c x_i s_i \tag{6}$$

$$s_i = \sum_k^g m_{ik} s_k \tag{7}$$

$$\tau_{kn} = e^{-E_{kn}/T} \tag{8}$$

$$E_{kn} = k_{kn}[E_k + E_n]/2.0 \tag{9}$$

$$E_k = E_k^{(0)} - E_k^{(1)}\left(\frac{509.7}{T} - 1\right) + E_k^{(2)}\left[\left(\frac{509.7}{T}\right)^2 - 1\right] \tag{10}$$

$$\psi_n = \sum_i^g x_i m_{in} b_n/b \tag{11}$$

Unfortunately, Wilson and Cunningham did not give a complete set of parameters for the various groups in their equation of state. However, Moshfeghian et al. [7-11] developed a set of parameters for a wide variety of compounds/groups and thoroughly evaluated their approach which was different than that of Wilson and Cunningham.

Group Parameters Determination Procedure

To determine the best values of group parameters, the following procedure has been used by the researchers [11, 12]:

As an example, consider the CH_3- group of ethane.

1. Values for b_k, s_k, $E_k^{(0)}$, $E_k^{(1)}$ and $E_k^{(2)}$ for CH_3- were assumed.

2. For a selected temperature below the critical, the vapor pressure, saturated vapor volume, saturated liquid volume, and heat of vaporization of the component were calculated.

3. The values calculated in step 2 were compared with the corresponding experimental values [13] and the square of the relative error for each calculated value was summed up:

$$SOE = C_1 (CVP/EVP - 1)^2$$
$$+ C_2 (CSVV/ESVV - 1)^2$$
$$+ C_3 (CSLV/ESLV - 1)^2$$
$$+ C_4 (CHV/EHV - 1)^2 \qquad (A2)$$

C_1 through C_4 are the weighting factor for each calculated error.

A value of 2 was used for C_1 and 1 was used for C_2 through C_4.

4. Step 3 repeated for other temperatures and an objective function was defined:

$$OF = \Sigma (SOE)_i \qquad (A3)$$

5. The nonlinear regression computer program developed by Chandler [14] was used to find the values of b_k, s_k, $E_k^{(0)}$, $E_k^{(1)}$ and $E_k^{(2)}$ which minimized the objective function.

This procedure for determination of group parameters was used for a series of components typical of those found in n-alkanes, selected non hydrocarbons such as N_2, H_2S, CO_2, and H_2O, 1-alkanols [7-11], refrigerants [10], coal derived compounds [11]. Table II (a) presents the previously unpublished parameters for several groups. The interaction group parameters are also presented in Tables II (b) and II (c).

Prediction of Pure Component PVT Properties [7, 10, 11, 15]

References [7, 15] summarize the results of the fitting process and some predicted deviations for the PFGC-MES equation of state. The results shown in these references are for selected non-hydrocarbon compounds. Parameters for most polar and non-hydrocarbon components were derived by considering the components as a single functional group. The remaining compounds were fitted using the group contribution approach defined earlier and a single interaction parameter. For the most part, these are predicted properties rather than fitted properties.

With the exception of the data of the vapor pressure predictions for carbon tetrachloride, the predicted properties are in reasonable agreement with the experimentally determined values. The data for monochloromethane are particularly outstanding in that these data were predicted completely using only the group contribution. No fitting was done for this particular compound.

Reference [7] also reports the absolute average deviation between predicted and experimental values for hydrocarbon components fitted in that study. With the exception of trans-2-butene, the predicted values appear

Table II (a) PFGC—MES Group Parameters

NO.	GROUP	b, ft³/lbmole	s	$E^{(0)}/R$, °R	$E^{(1)}/R$, °R	$E^{(2)}/R$, °R
1	H_2	0.3296	1.8729	−53.6787	8.2008	1.3592
2	CH_4	0.590	1.871	−129.0	−57.2	7.0
3	CH_3-	0.332	1.983	−319.0	−80.0	0.0
4	$-CH_2-$	0.268	0.507	−266.0	−63.7	0.0
5	$>CH^-$	0.2178	−0.8975	−102.26	−86.2	0.0
6	$>C<$	0.191	−3.39	482.0	−115.0	0.0
7	$CH_2=$	0.354	1.340	−221.6	−54.0	0.0
8	$-CH_2-$cyc	0.3744	0.6333	−198.645	−7576	0.3
9	$-CH=$ rin	0.258	0.3471	−257.1	−193.8	30.0
10	$>C=$ rin	0.0545	0.0855	−1167.7	−53.8	159.0
11	$-CH=$	0.344	−0.708	−100.6	−119.0	0.0
12	N_2	0.445	2.3695	−118.3	−33.2	3.0
13	CO_2	0.330	3.6985	−614.7	−201.2	20.2
14	CO	0.4053	2.5993	−136.27	−57.1	6.5
15	H_2S	0.405	3.4335	−609.6	−172.9	16.0
16	H_2O	0.200	2.200	−2651.3	−2779.3	858.5
17	$-OH$	0.1011	2.1264	−2593.30	−3053.10	1449.90
18	MeOH	0.3732	5.5992	−1407.77	−789.31	108.03
19	NH_3	0.277	2.7065	−985.6	−838.0	210.0
20	$-NH_2$	0.083	3.000	−2177.1	−626.5	271.5
21	Cl^{-}	0.3032	1.4585	−441.9	−113.0	0.0
22	$-CH=$ cyc	0.268	0.507	−271.0	−115.0	0.0
23	CCl_3F	0.994	6.5677	−453.33	−47.09	2.88
24	CCl_2F_2	0.8530	6.0670	−404.87	−41.44	1.17
25	$CClF_3$	0.8472	4.0990	−253.46	−70.78	9.00
26	CF_4	0.8472	3.5000	−203.10	−69.76	8.44
27	$CHCl_2F$	0.8454	5.2309	−464.19	−110.10	12.09
28	$CHClF_2$	0.6723	4.8861	−446.54	−135.12	22.57
29	CHF_3	0.5764	3.9402	−365.24	−153.16	23.20
30	CH_3Cl	0.6606	3.4159	−445.93	−137.70	13.01
31	$C_2Cl_3F_3$	1.4009	6.3884	−344.77	−64.46	8.03
32	C_2ClF_5	1.2747	6.7136	−32363	−5389	7.39
33	$C_2H_3ClF_2$	0.8834	−0.4543	−440.33	−164.25	60.00
34	$C_2H_4F_2$	0.7705	4.7970	−419.39	−209.43	60.00
35	R500*	0.8200	5.8308	−400.32	−88.60	16.30
36	R502*	0.7264	6.6788	−455.71	−57.14	7.51

Table II (a). continue

No.	Group	b, ft^3/lbmole	s	$E^{(0)}/R$, oR	$E^{(1)}/R$, oR	$E^{(2)}/R$, oR
37	R503*	0.5905	5.2765	−394.93	−57.88	2.25
38	R504*	0.6305	4.8257	−428.95	−172.13	39.47
39	> N−	0.0812	4.6608	−2286.0	1000.0	84.9

rin = ring; cyc = cyclo; * = azeotropic mixgures, treated as pure compounds

Table II (b) PFGC—MES Vapor Phase Binary Group Interaction Coefficient

K_{ij}	VK_{ij}	K_{ij}	VK_{ij}	K_{ij}	VK_{ij}
102	0.691	313	0.850	1318	0.840
103	0.411	315	0.830	1618	0.900
104	0.720	316	0.335	221	1.430
105	0.460	318	0.750	621	5.500
108	0.6951	405	1.395	107	0.550
109	0.950	407	0.940	716	0.380
110	0.050	408	1.050	1116	0.300
112	0.600	410	0.600	916	0.250
203	0.945	412	0.650		
204	0.900	413	0.900		
205	0.500	415	0.750		
207	1.050	416	0.290		
208	0.880	418	0.750		
212	0.945	420	0.677		
213	0.765	510	0.300		
215	0.720	521	0.950		
216	0.2635	713	0.895		
218	0.500	918	0.805		
304	1.032	1113	0.980		
307	0.989	1213	0.450		
308	1.020	1215	0.450		
312	0.945	1218	0.500		

K_{ij} = 100 × (ID No. for group i) + (ID No. for group j)

Table II (c) PFGC—MES Liquid Phase Binary Group Interaction Coefficient

K_{ij}	LK_{ij}	K_{ij}	LK_{ij}	K_{ij}	LK_{ij}
102	0.691	313	0.850	1318	0.840
103	0.411	315	0.830	1618	0.900
104	0.720	316	0.210	221	1.430
105	0.460	318	0.750	621	5.500
108	0.695	405	1.395	107	0.550
109	0.950	407	0.940	716	0.250
110	0.050	408	1.050	1116	0.420
112	0.600	410	0.600	916	0.470
203	0.945	412	0.950		
204	0.900	413	0.900		
205	0.500	415	0.750		
207	1.050	416	0.310		
208	0.880	418	0.750		
212	0.945	420	0.677		
213	0.765	510	0.300		
215	0.720	921	0.950		
216	0.125	713	0.895		
218	0.500	918	0.805		
304	1.032	1113	0.980		
307	0.989	1213	0.450		
308	1.020	1216	0.450		
312	0.945	1218	0.500		

K_{ij} = 100 × (ID No. for group i) + (ID No. for group j)

to be in reasonable agreement with the literature data.

To further demonstrate the power of the PFGC-MES equation of state, Moshfeghian et al. [7] determined the group parameters for the OH— group in the normal alcohols by fitting ethanol, n-propanol and n-butanol. After they determined the OH— group parameters, the vapor pressure of remaining alcohols up to nC_{20}—OH was predicted. Their results of these predictions are shown in Table III. With the possible exception of the higher molecular weight alcohols, C_{17} and above, the predicted and experimental results were in good agreement. In that study, the upper reduced temperature limit for the pure component fitting process was of approximately 0.95.

Reference [10] reports the average absolute deviation between preicted and the experimental values for pure and azeotropic mixtures of refrigerants by the PFGC-MES equation of state. The predicted values appear to be in excellent agreement with the experimental data. In this reference the capabilities of the PFGC-MES are also compared with that of the SRK [16] equation of state for the same components. Even though PFGC-MES covers a wider range of for reduced temperature, it gives better results.

The comparison of the predicted results and the experimental PVT data for coal derived compounds and those predicted by PFGC-MES are reported in reference [11]. For some of the compounds studied in that reference, there was no experimental data available to make a complete comparison. For example, the only data on quinoline that they could find was its vapor pressure reported by Van De Rostyne and Praushitz [17]. However, the predicted properties such as the saturated liquid volume, saturated vapor volume and heat of vaporiza-

tion for this compound was reported in this reference. The overall comparison results between the experimental PVT data and those predicted by PFGC-MES is very good. The maximum average absolute deviation is 5.71 percent which corresponds to that of saturated vapor volume of benzene.

Vapor-Liquid-Equilibria [7-11, 15, 18]

Data for binary mixtures of carbon dioxide, nitrogen, hydrogen sulfide, methane, ethane, propane, benzene, toluene, methanol, glycols and a variety of cycloparaffins with light hydrocarbons have been used by researchers to derive the vapor and hydrocarbon-liquid phase binary group interaction coefficients, k_{kn}, used in Equation (9). Mixtures of water with light hydrocarbons, carbon dioxide, hydrogen sulfide, nitrogen and carbon monoxide have also been used by researchers to derive the binary group interaction coefficients for the water-rich liquid phase. Results for selected systems reported by Moshfeghian et al. [7, 8], Majeed et al. [15] and later by Wagner et al. [18] are in good agreement with experimental data.

Using the parameters defined in the binary fitting process, researchers [7, 15, 18] predicted the behavior of multicomponent systems. Figure 1 presents one such comparison based on Yarbrough's data [19]. The quality of agreement shown here is typical for petroleum/natural gas systems. These results are comparable with SRK and PR predictions for this system.

The ability of the PFGC-MES equation of state to describe the behavior of more non-ideal systems is illustrated in Figures 2 and 3. These diagrams are typical of the behavior that one can expect for these kind of systems. The prediction of the liquid phase composition

Table III. Summary of the absolute average percentage error for vapor pressures of aliphatic alcohols [7]

Alcohol	Temperature Range, °F	Absolute Average Percent Error
Methanol	273–483	1.16
Ethanol	323–503	1.55
Propan–1–01	343–523	2.13
Butan–1–01	358–550	1.19
Pentan–1–01	410–535	5.22
Hexan–1–01	450–545	5.38
Octan–1–01	485–561	4.87
Nonan–1–01	485–661	6.44
Decan–1–01	500–678	5.74
Undecan–1–01	520–600	2.91
Dodecan–1–01	535–617	2.15
Tridecan–1–01	550–633	0.84
Tetradecan–1–01	567–650	1.39
Pentadecan–1–01	578–667	3.32
Hexadecan–1–01	594–678	5.54
Heptadecan–1–01	611–689	7.76
Octadecan–1–01	628–706	10.38
Nonadecan–1–01	633–717	12.79
Eicosan–1–01	644–728	15.22

for methanol-benzene system [20] in Figure 2 are not particularly good at low concentrations of benzene. However, the higher concentrations of benzene are predicted equite well.

Figure 3 shows a plot of the predicted and experimental K-values for the hydrogen sulfide-water system [21]. Again, agreement between experiment and prediction appears to be satisfactory.

Figure 4 shows the effect of pressure on the solubility of water in the propane vapor phase and the solubility of propane in the liquid water phase at the approximate propane critical isotherm [22]. The agreement between experimental and calculated values

must be considered excellent for this system. Researchers [7, 15, 18] report similar agreement between predicted and experimental hydrocarbon vapor and liquid water phase solubilitues for most of the available data.

Vapor-Liquid-Liquid Equilibria and Hydrate Formation [9,15,18,23,24]

The capability of this equation of state to handle three-phase calculations has been evaluated by several investigators. The PFGC ability to predict hydrate formation condition and hydrate inhibition have been also studied [18, 24]. The Hydrate model described by Parish and Prausnitz [25] is used
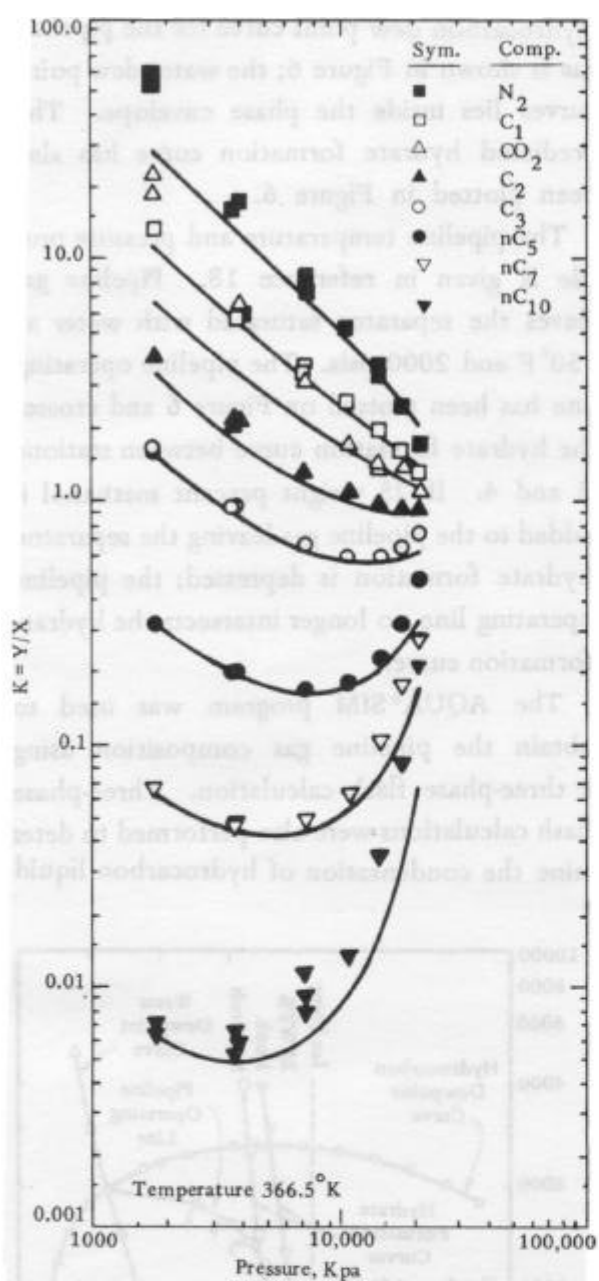
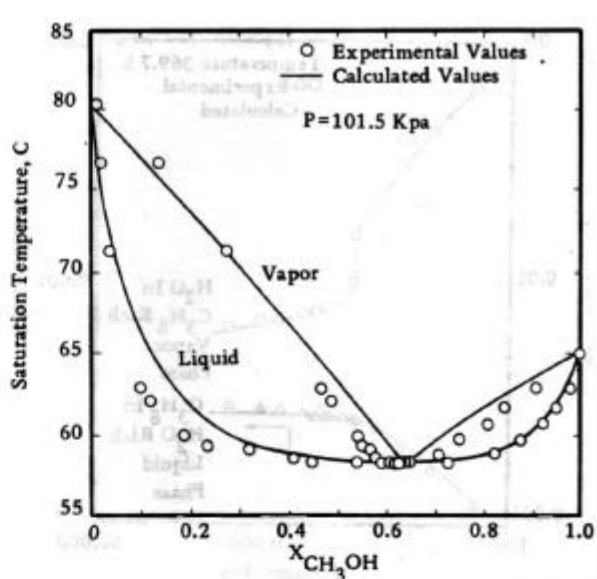Figure 1. Comparison of predicted and experimental K values-yarborough data [7].

Figure 2. Comparison of predicted and experimental T-X diagrams for the methanol-benzene system [7].
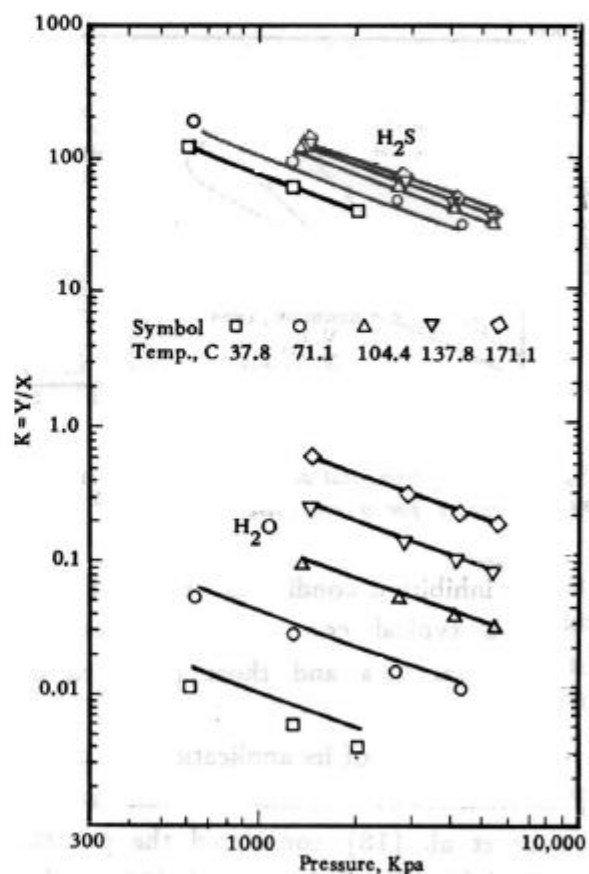
Figure 3. Comparison of experimental and calculated K values for the H_2S-H_2O system [7].

as the basis for hydrate formaiton calculations. The activity coefficient corrections suggested by Menten et al. [26] were included by Wagner and co-workers to account for the effects of inhibitors such as methanol and glycols. Wagner, Erbar and Majeed [23] have developed a computer simulation program, AQUA*SIM, for process design involving three-phase calculations and hydrate formation as

described above, based on PFGC. All of these investigators report good agreement between experimental and predicted hydrate forma-
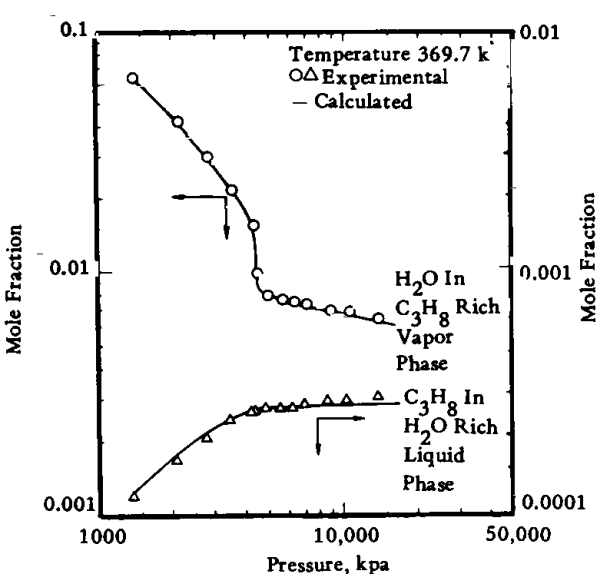
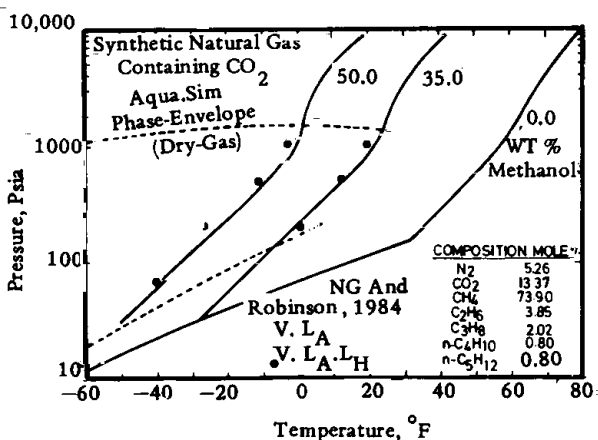Figure 4. *Mutual solubility of C_3H_8 and H_2O [7].*

Figure 5. *Experimental and predicted hydrate formation curves for a synthetic natural gas[5] [18].*

tion or inhibition conditions. Figure 5 [18] shows a typical comparison between the experimental data and those predicted by PFGC.

As an example of its application for phase equilibrium and hydrate inhibition calculations Wagner et al. [18] considered the process described by Moshfeghian et al. [23]. The natural gas feed enters a separator operating at 150°F and 2000 psia where liquid water and hydrocarbon are knocked out. The composition of the pipeline gas leaving the separator is also given in reference 18. The

Hydrocarbon dew point curve for the pipeline gas is shown in Figure 6; the water dew point curves lies inside the phase envelope. The predicted hydrate formation curve has also been plotted in Figure 6.

The pipeline temperature and pressure profile is given in reference 18. Pipeline gas leaves the separator saturated with water at 150°F and 2000 psia. The pipeline operating line has been plotted on Figure 6 and crosses the hydrate formation curve between stations 3 and 4. If 25 weight percent methanol is added to the pipeline gas leaving the separator, hydrate formation is depressed; the pipeline operating line no longer intersects the hydrate formation curve.

The AQUA*SIM program was used to obtain the pipeline gas composition using a three-phase flash calculation. Three-phase flash calculations were also performed to deter mine the condensation of hydrocarbon liquid
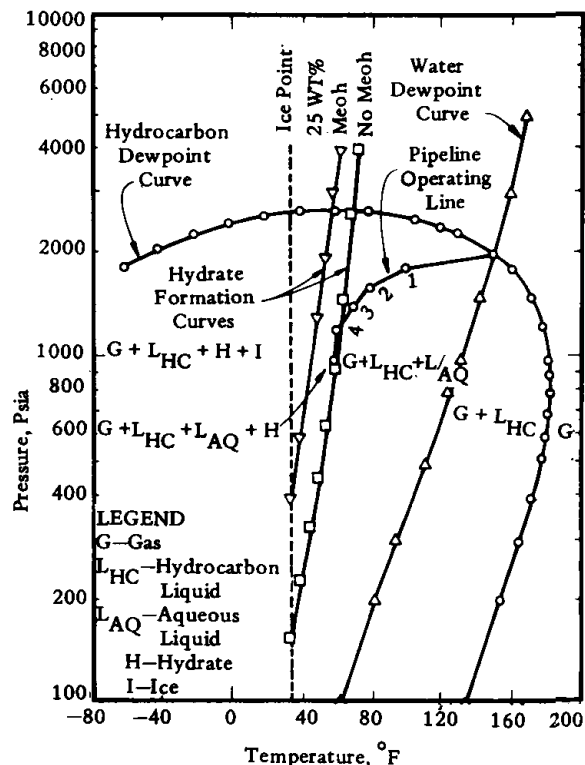

Figure 6. *Example of phase behavior and process/ pipeline conditions for a natural gas system [18].*

and distribution of water and methanol in the aqueous and vapor phases along the pipeline. The results which were obtained by Wagner et al. [18] provide an estimate of

$E_k^{(2)}$ = Energy coefficient for group k; used in Eq; (10)

E_k = Energy term for group k; defined by Eq. (10)

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