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## Gas Condensation and Phase Diagram in Multiphase Flow Systems

Ullas Pathak<sup>1</sup>, Aaron West<sup>1</sup>

<sup>1</sup> Statistics & Control, Inc.

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### ABSTRACT

In the design and operation stages of multiphase pipelines, prediction of flow temperatures and pressure variations is important. Dynamic evaluation of process changes in real-time and calculation of thermodynamic properties of multi-component mixtures is challenging. In this paper a mechanism to counter phase change losses due incorporating real-time dynamics is presented. A case-study for a major oil and gas distribution company with real-world deployment and observation has been included. The model provides notification of changes and displays a PT-phase diagram using different gas compositions and oil assay data to compare the phase changes in model and commercial pipelines in real-time. With density and viscosity considerations in the approach, gas condensation alarms are raised dynamically depending on the total vapor fraction in the mixture.

### INTRODUCTION AND BACKGROUND

Pipelines are used to transport volatile and flammable liquids for widespread applications in chemical and petroleum industries. The design, construction and optimization of pipelines is conducted based on normal operating conditions. However, gas flowing through a pipeline or entering a facility may contain solids and liquids as particulates or in a different phase. In a condensate well, production is usually a high-pressure process, thus the heavy ends are in the liquid state, as the wellhead pressure decreases, more heavy components vaporize into the gas stream increasing the potential amount of

pipeline liquids. Liquid may form during transportation of natural gas in pipelines due to pressure loss, temperature change, or retrograde condensation of heavier hydrocarbons in the gas phase. One of the many challenges faced today with pipeline flow assurance is maintaining lowest pressure losses. Existence of two-phase flow in pipelines can cause different pressure-drop than the intended pressure-drop based on single phase calculations. Separation of solubles and impurities is a critical field process operation. With advent of standards in gas transmission lines, separation becomes necessary to condition the gas. Selecting separation technologies not only requires the knowledge of the process conditions, but also knowledge of the characteristics of the liquid contaminants. Knowledge of phase changes in pipelines, facilitates operations and provides an insight about the process changes with respect to a phase diagram. Oil and gas pipelines encounter multiple “phase diagram disasters” which incur losses and add downtime into pipeline production systems.

The study presented here provides an example of real-time deployment of phase-separation and gas condensation algorithm. The modeling and algorithm have been implemented at a major oil and gas company’s site on a 1700 km (~1056 mi) commercial pipeline network. The network is responsible for transporting ~2700 MMSCF (million standard cubic feet) of gas from 17 different reservoir units. The algorithm delivers a reliable and accurate model to obtain phase changes in oil and gas pipelines using a real-time monitoring system. The approach utilizes a multi-parameter equation-of-state and solves the Rachford-Rice equation to calculate phase compositions. Different gas compositions and oil assay data are used to compare the phase changes in model and commercial pipelines. The study presents an analytical approach for easy prediction of these parameters at any point along the two-phase - gas/gas-condensate transmission lines by applying laws of conservation. Results for the multi-component systems are shown by the design of a gas transmission line with varying compositions and thermodynamic properties presented for an industrial scenario.

## SCIENTIFIC FOUNDATION

Phase Behavior is crucial for accurate prediction of the P-V-T properties of natural gases, especially when dealing with pipeline design, gas storage, measurement and transport. A consideration in gas pipeline design is the differentiation between dry gas and wet gas flow - where multiphase conditions due to condensate dropout are possible.

A phase diagram (Figure 1) describes regions of pure component behavior transitioning between phases. Figure (1) shows that in pure materials, decreasing the pressure at a fixed temperature, results in phase change but just at a point (vapor pressure curve is a line). At extremely high temperatures and pressures, the liquid and gaseous phases become indistinguishable. The phase boundary for liquid and gas does not continue indefinitely, it terminates at a point on the phase diagram called the critical point. For multi-component systems phase diagram is more complex and elaborate than that of pure compounds. Generally, components with widely different structure and molecular sizes comprise the system. P-T diagram is a graphical representation of phase changes of compounds that describes the set of pressure-temperature combinations for the transition zone between the complete liquid and complete vapor phase. Figure (2) shows an idealized P-T diagram for a multi component with a fixed overall composition. There is a region where the two phases are at equilibrium. The two-phase region that is bounded by the bubble point and dew point curves is called "phase envelope".

A two-phase mixture has critical temperature which is between the critical temperatures of the pure components, presented by the locus of all critical temperature points for pure components as shown by the dashed line (Figure 4). The two-phases unlike a pure component can co-exist at a pressure greater than critical pressure and at a temperature greater than critical temperature. For a multicomponent system the maximum pressure that two phases (vapor-liquid) can co-exist in equilibrium and the maximum temperature that they can exist in equilibrium, are known as cricondenbar and cricondentherm, respectively.

The difference between the critical pressure of two component system and each pure component critical pressure increases by increasing the difference between the critical points of the two pure components (Figure 3). A binary mixture cannot exist as a two-phase system outside the region bounded. To have a miscibility of two compounds with any composition, at each temperature the pressure should be higher than the pressure indicated by the locus of critical pressure line for that specific temperature. A minimum miscible pressure for each temperature could be determined according to graph's data (Figure 4). Although a lower pressure might be possible for concentrations when the system will separate into two phases.

When the two-phase mixture enters the phase envelope and tends to move towards the liquid phase a timely notification of these events will help pipeline operators to make effective decisions. If the different pipeline components in the mixture start condensing and form liquids, it causes hinderance to flow assurance. The liquid hold-up not only leads to downtime but also clogging, pressure losses and hence loss in revenue. These losses can range from 10-15% for gas pipelines.

## THERMODYNAMIC MODELING

The thermodynamic behavior of nonideal gas mixtures where liquid and gas are in equilibrium can be used for the real-time detection of gas condensation in pipeline monitoring system. When the gas phase and the liquid phase exist in equilibrium together, the gas phase fugacity and liquid phase fugacity of each component is equal.

i.e.

$$f_i^{gas} = f_i^{liquid} \quad (1)$$

In this work, a gas condensation algorithm (GCA) and dynamically updated pressure-temperature phase diagram (P-T diagram) are developed using this principle along with several other well-known approximations. The Peng-Robinson Equation of State (PR-EOS), Rachford-Rice Equation, and the known experimental properties of pure components in the mixture are all used to model condensation in the pipeline monitoring system.

The following assumptions have been made in order to describe the gas-liquid equilibrium:

- The feed compositions are known.
- Each component that condenses is assumed to form a pure component.
- Quadratic mixing rule for mixtures is used for non-ideal gas mixtures.

### INITIAL PURE COMPONENT PROPERTIES

The acentric factor  $\omega$  is a measurement of molecular centricty of pure components and is used to explain higher boiling points. Acentric factor ( $\omega$ ) can be estimated in various ways. For reduced boiling temperatures  $T_{br} = \frac{T_b}{T_c} \leq 0.8$  where  $T_b$  denotes the normal boiling temperature and  $T_c$  denotes the critical temperature, the Lee-Kesler method<sup>i</sup> was used in this study.

$$\omega = \frac{-\ln \frac{P_c}{1.01325} - 5.92714 + \frac{6.09648}{T_{br}} + 1.28862 \ln T_{br} - 0.169347 T_{br}}{15.2518 - \frac{15.6875}{T_{br}} - 13.4721 \ln T_{br} + 0.43577 T_{br}} \quad (2)$$

where  $P_c$  is in bar.

Otherwise, the Korsten method<sup>ii</sup> was used in this study.

$$\omega = \left[ 0.5899 \frac{T_{br}^{1/3}}{1 - T_{br}^{1/3}} \log \frac{P_c}{1.01325} \right] - 1 \quad (3)$$

where  $P_c$  is in bar.

### PENG ROBINSON EQUATION OF STATE (PR-EOS)

For a pure gas, the PR-EOS<sup>iii</sup> relates pressure  $P$ , molar volume  $V$ , and temperature  $T$  as,

$$P = \frac{RT}{V-b} - \frac{a}{V^2 + 2Vb - b^2} \quad (4)$$

where  $a$  is given by,

$$a = [0.45724R^2T_c^2/P_c] \left[ 1 + \kappa \left( 1 - \left( \frac{T}{T_c} \right)^{1/2} \right) \right]^2 \quad (5)$$

where  $\kappa$  is given by,

$$\kappa = 0.37464 + 1.54226\omega - 0.2699\omega^2 \quad (6)$$

where,  $\omega$  is the acentric factor and  $b$  is given by,

$$b = \frac{0.07780RT_c}{P_c} \quad (7)$$

For a mixture with  $N$  pure components, the PR-EOS can also be expressed in cubic form as,

$$Z_{mix}^3 - (1 - B_{mix})Z_{mix}^2 + (A_{mix} - 2B_{mix} - 3B_{mix}^2)Z_{mix} - AB_{mix} + B_{mix}^2 + B_{mix}^3 = 0 \quad (8)$$

where  $Z_{mix}$  denotes the compressibility of the mixture,

$$A_{mix} = \frac{a_{mix} P}{R^2 T^2} \quad (9)$$

$$B_{mix} = \frac{b_{mix} P}{RT} \quad (10)$$

$$a_{mix} = \sum_{i=1}^N \sum_{j=1}^N a_{ij} n_i n_j \quad (11)$$

$$a_{ij} = \sqrt{a_i a_j} (1 - k_{ij}) \quad (12)$$

$$b_{mix} = \sum_{i=1}^N b_i n_i \quad (13)$$

the  $k_{ij}$  denotes the binary interaction parameter between components  $i$  and  $j$ , the  $n_i$  denotes the mole fraction of the  $i$ th pure component,  $a_i$  and  $b_i$  are given for each component  $i$  by equations (12) and (13), respectively.

In this study, the analytical solution (see reference 4 and reference 5) was used to solve for the compressibility  $Z_{mix}$  of the PR-EOS.

Using the PR-EOS from equation (8), the fugacity coefficient

$\phi_i$  for the  $i$ th component is calculated as,

$$\ln \phi_i = (Z_{mix} - 1) \frac{b_i}{b_{mix}} - \ln(Z_{mix} - B_{mix}) - \frac{A_{mix}}{\sqrt{2B_{mix}}} \left( \frac{1}{a_{mix}} \sum_{i=1}^N x_i a_{ij} - \frac{b_i}{2b_{mix}} \right) \ln \left( \frac{Z_{mix} + (1 + \sqrt{2})B_{mix}}{Z_{mix} + (1 - \sqrt{2})B_{mix}} \right),$$

where  $Z_{mix}$  is the fluid mixture compressibility,  $A_{mix} = \frac{a_{mix} P}{R^2 T^2}$ , and  $B_{mix} = \frac{b_{mix} P}{RT}$ .

### GAS CONDENSATION ALGORITHM (GCA)

Using the total mole fractions  $n_i$ , or compositions, of the  $N$  pure components, the operating temperature  $T$ , and the operating pressure  $P$ , GCA ultimately determines the gas mole fractions  $y_i$  and liquid mole fractions  $x_i$  when two phases are present. The mole fractions are all normalized.

$$\sum_{i=1}^N n_i = 1, \quad \sum_{i=1}^N y_i = 1, \quad \sum_{i=1}^N x_i = 1 \quad (15)$$

In turn, the equilibrium constants  $K_i$  are defined through the mixture compressibility  $Z_{mix}$  using the operating pressure  $P$ , operating temperature  $T$ , gas mole fraction  $y_i$ , and liquid mole fraction  $x_i$ .

$$\ln K_i = \ln \frac{y_i}{x_i} = \ln \frac{\phi_i^{gas}(Z_{mix}, y_i)}{\phi_i^{liquid}(Z_{mix}, x_i)}, \quad i = 1, \dots, N \quad (16)$$

Using the equilibrium constants  $K_i$  and the total mole fractions  $n_i$  of the pure components, the total vapor fraction  $\beta$  can be obtained through numerical solution of the Rachford-Rice equation described in reference 6, and can be expressed as,

$$g(\beta) = \sum_{i=1}^N \frac{n_i(K_i - 1)}{1 + \beta(K_i - 1)} \quad (17)$$

The numerical solution is implemented through a combination of the bisection method and Newton-Raphson method, reference 7.

In order to express the Rachford-Rice equation in its form in equation (17), the gas phase composition  $y_i$  and liquid phase composition  $x_i$  must be related to the equilibrium constants  $K_i$  and total mole fractions  $n_i$  through the total vapor fraction  $\beta$  by equations (18) – (20).

$$y_i = \frac{K_i n_i}{1 + \beta(K_i - 1)}, \quad i = 1, \dots, N \quad (18)$$

$$x_i = \frac{n_i}{1 + \beta(K_i - 1)}, \quad i = 1, \dots, N \quad (19)$$

$$n_i = \beta y_i + (1 - \beta)x_i, \quad i = 1, \dots, N \quad (20)$$

If two phases are present, the total vapor fraction  $\beta$  and gas and liquid mole fractions are solved by an iterative procedure. In order to initialize the procedure, the approximate Wilson equilibrium constants  $K_i$  are used and are estimated using generalized correlations, reference 8.

$$\ln K_i = \ln \frac{P_{c,i}}{P} + 5.373(1 + \omega_i) \left(1 - \frac{T_{c,i}}{T}\right) \quad (21)$$

Given the operating pressure  $P$  and temperature  $T$ , critical pressure  $P_{c,i}$ , critical temperature  $T_{c,i}$ , and acentric factor  $\omega_i$  of the  $i$ th component as defined in equation (3), the iterative procedure, reference 9, is performed as follows:

- The total vapor fraction  $\beta$  is determined by solving the Rachford-Rice equation (17).
- The gas phase mole fractions  $y_i$  and liquid phase mole fractions  $x_i$  are calculated with equations (18) and (19).
- The fugacity coefficients  $\phi_i^{gas}$  and  $\phi_i^{liquid}$  are calculated using equations (14) and (16).
- The equilibrium constants  $K_i$  are determined from the fugacity coefficients using equation (21).

The iterations are performed until one of the following conditions is satisfied.

- The total vapor fraction converges between successive iterations:  $|\beta_n - \beta_{n-1}| < 10^{-7}$ .
- Equilibrium is reached:  $\sum_{i=1}^N \left(1 - \frac{\phi_i^{gas}}{\phi_i^{liquid}}\right)^2 < 10^{-14}$
- $\beta = 1$  or  $\beta = 0$

Now that the total vapor fraction is known, pipeline operators can set threshold limits of detection for warnings that too much condensation has occurred in a given pipe.

#### DYNAMIC PRESSURE-TEMPERATURE PHASE DIAGRAM (PT-DIAGRAM)

Using the properties of the pure components in a mixture, a dynamic PT-diagram for the mixture is constructed on-the-fly. First, in order to construct the PT-diagram for the mixture, vapor-liquid equilibrium (VLE) curves must be constructed for the pure components. The critical pressures  $P_c$  and critical temperatures  $T_c$  are known and stored in advance for each pure component. Starting at  $(P_c, T_c)$ , incrementally decreasing the temperature, and using the last determined saturation pressure as an initial guess to the current saturation pressure  $P_{sat}$ , the pure component VLE curves are generated.

Starting at the initial guess  $P_{sat}$ , the pressure is lowered until solving the PR-EOS for the compressibility yields two real roots: a gas root and a liquid root. Then, by setting  $N=1$ ,  $n_1=1$ , and  $k_{11}=0$ , equation (22) reduces to the pure component

version. Then, using the gas and liquid compressibility roots, the pure component fugacity coefficients  $\phi_i(Z_{pure}^{gas}, 1)$  and  $\phi_i(Z_{pure}^{liquid}, 1)$  are determined. At this point, the old saturation pressure  $P_{sat}^{old}$  is updated to a new saturation pressure  $P_{sat}^{new}$  as follows.

$$P_{sat}^{new} = (P_{sat}^{old}) (f_{ratio}) \quad (22)$$

where  $f_{ratio}$  is defined as,

$$\text{if } \left|1 - \frac{\phi_i^{liquid}}{\phi_i^{gas}}\right| < \left|1 - \frac{\phi_i^{gas}}{\phi_i^{liquid}}\right|, f_{ratio} = \frac{\phi_i^{liquid}}{\phi_i^{gas}} \quad (23)$$

otherwise,

$$f_{ratio} = \frac{\phi_i^{gas}}{\phi_i^{liquid}} \quad (24)$$

When equilibrium is reached and  $f_{ratio}=1$ , the new saturation pressure  $P_{sat}^{new}$  converges to the saturation pressure  $P_{sat}$  for the pure component.

It is important to note that as gas flows through the pipeline, new components might be added into the mixture. When a new component is added into the mixture, a new pure component VLE is generated and stored in a permanent location.

Once all the pure component VLE's are available for the current mixture, the mixture PT diagram is constructed and consists of the connected bubble and dew curves. The following is performed for each separate pure component. Starting at the largest critical temperature  $T_c$  of all the pure components, if the temperature  $T > T_c$ , a weighted contribution of the critical pressure  $P_c$  based on the mole fractions is made only to the bubble curve. On the other hand, if the temperature  $T \leq T_c$ , then weighted contributions of  $P_{sat}$  are made to both the bubble curve and the dew curve.

Given the PT diagram for the mixture and the operating pressure and temperature, pipeline operators can check dynamically for condensation.

## CASE STUDY: PHASE SEPARATION AND GAS CONDENSATION IN A PIPELINE

The modeling and simulation have been implemented and validated at a major oil and gas company's site on a 1700 km (~1056 mi) commercial pipeline network being used to transport ~2700 MMSCFD (million standard cubic foot) of gas from ~17 reservoir units to different distribution units. The

model helps calculate the total production from suppliers and the amount that reaches the distribution unit of the processing plants. The amount that reached before the deployment of the model was with about 10-11% of loss. Goal of this study was to confirm phase changes and enable gas condensation alarms where occurring in the pipeline in real-time for appropriate actions.

The study has been presented here for simplicity using smaller simulation models with gas samples containing different compositions of hydrocarbons. The study displays three composition categories for gas components in the samples. The first sample displays a gas composition of pure gaseous phase with less than 5% liquid components co-existing in gaseous phase. Second gas composition category contains ~50% of either liquid and gaseous phases of hydrocarbons during transport through the pipeline. The third and final composition is a special case, presented for very high-pressure pipelines where due to high pressure of transportation, C4 and higher, hydrocarbons have the highest probability of liquifying.

The simulation model utilizes multiphase calculations on the process in each segment of the pipeline to provide an integrated approach for phase separation. This results in mass balance, energy conservation and various thermodynamic models which produce compressibility factor, computed density variations, binary interactions, activity and fugacity coefficients, and hence produce the required parameters to output the phase diagram in real-time. Shown in Figure 6, composition with majority of gas components in the current operating condition presents a pure gaseous phase in the phase diagram. For the cases 2 and 3, gas compositions containing higher percentages of liquid components, the shift in the *black dot* on their respective phase diagrams presents phase change in real time with changing phase envelopes due to differing gas components.

For higher compositions of butane, as compared to lower compositions, in the previous two cases, Figure 8 displays a higher inclination towards liquid phase on the PT-diagram. The pipeline transport was conducted at a higher pressure for this case, and increasing the pressure resulted in elevation in boiling point hence the alkane underwent phase change to liquid phase. Figure 12 presents boiling point curve for iso-butane. Phase diagram for second case presents the gas condensation alarm when the current conditions resulted in displacing the *black dot* nearer to the phase envelope.

## CONCLUSIONS

The timely detection and real-time maintenance of gas pipelines is required. Even high-pressure operations do not ensure complete possibility of no liquid hold-up. Gas condensation detection enables operations to have timely notification of liquid hold up and hence slugging of liquids in

gas pipelines. Dynamic behavior of phase diagram for varying operating conditions enabled effective decisions to reduce losses. Based on the results of the case study, the described modeling and simulation techniques effectively detect gas condensation in real-time in multiphase pipelines. This paper discussed the physical, chemical and time variation considerations contributing to precise detection of gas condensation, liquid hold up and shifting of phase diagram. The visual representation of this information enabled operators and operation personnel, on the fly from remote locations, to view critical events.

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## AUTHOR BIOGRAPHY

**Ullas Pathak** is a Technology Manager working at Statistics & Control, Inc. He currently works on developing automation architecture, quantitative modeling, optimization strategies for unit control systems, and process dynamics. Ullas has a Bachelor of Technology in Chemical Engineering from the National Institute of Technology Tiruchchirappali (NIT

Trichy), a Master of Science in Chemical Analytics from Arizona State University, and a Master of Engineering in Biorenewable Technologies from Iowa State University. Ullas has led many global-energy projects, focusing on optimizing operations, process enhancement and product engineering.

**Dr. Aaron C. West** is a Software Engineer working at Statistics & Control, Inc. He has a Bachelor of Arts in Chemistry from Augustana College, Bachelor of Arts in Biology from Augustana College, and Doctor of Philosophy in

Physical Chemistry from the Iowa State University of Technology. Aaron has over 10 years of experience in the development and use of computer software for modeling complex physical processes.

**FIGURES**

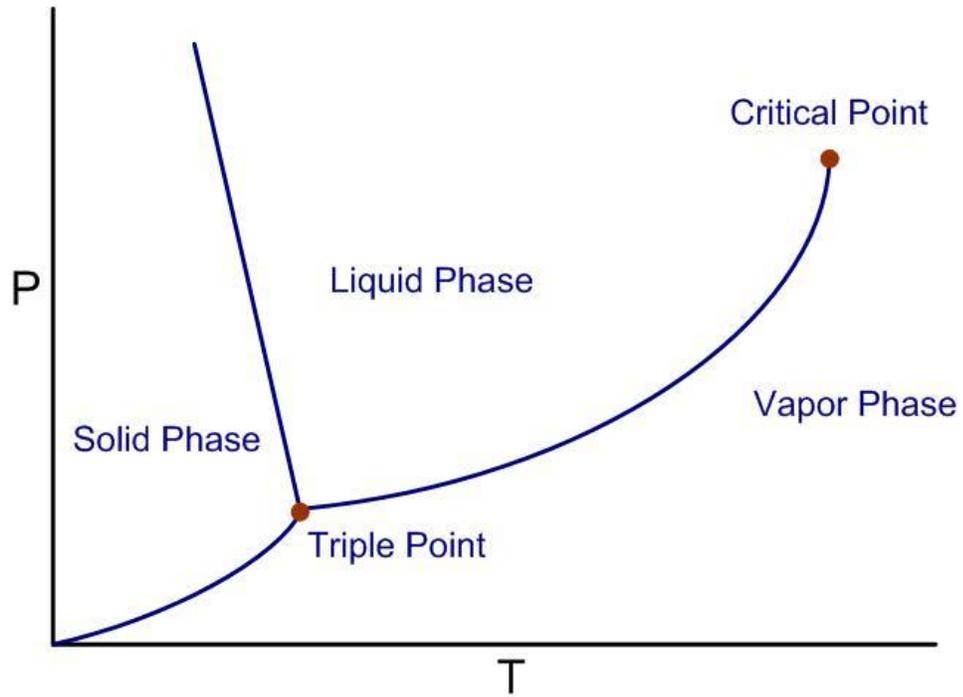


Figure 1 Phase Diagram of any Pure Component

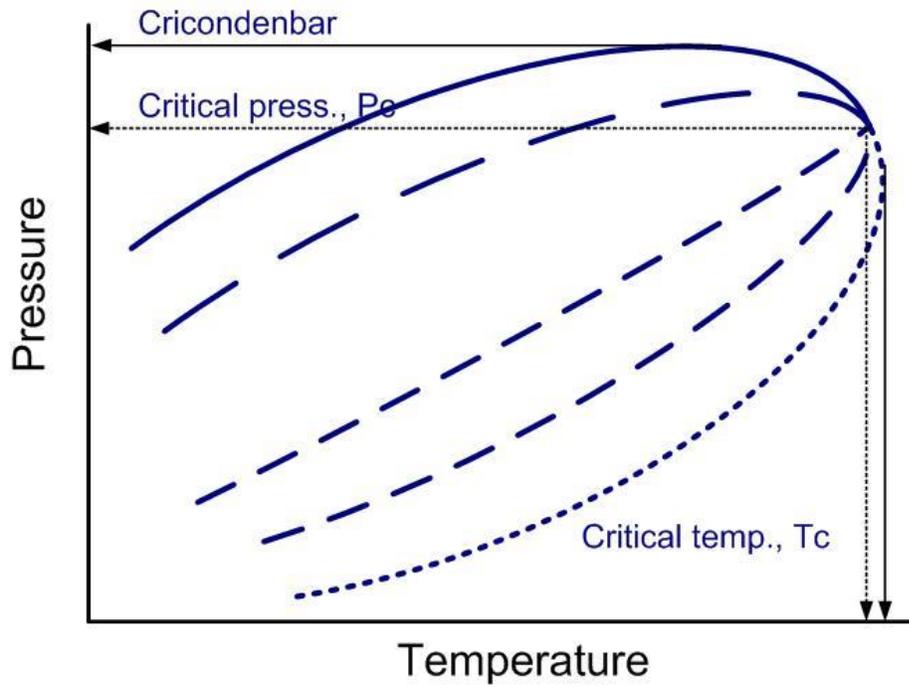


Figure 2 PT-Diagram for a multicomponent system

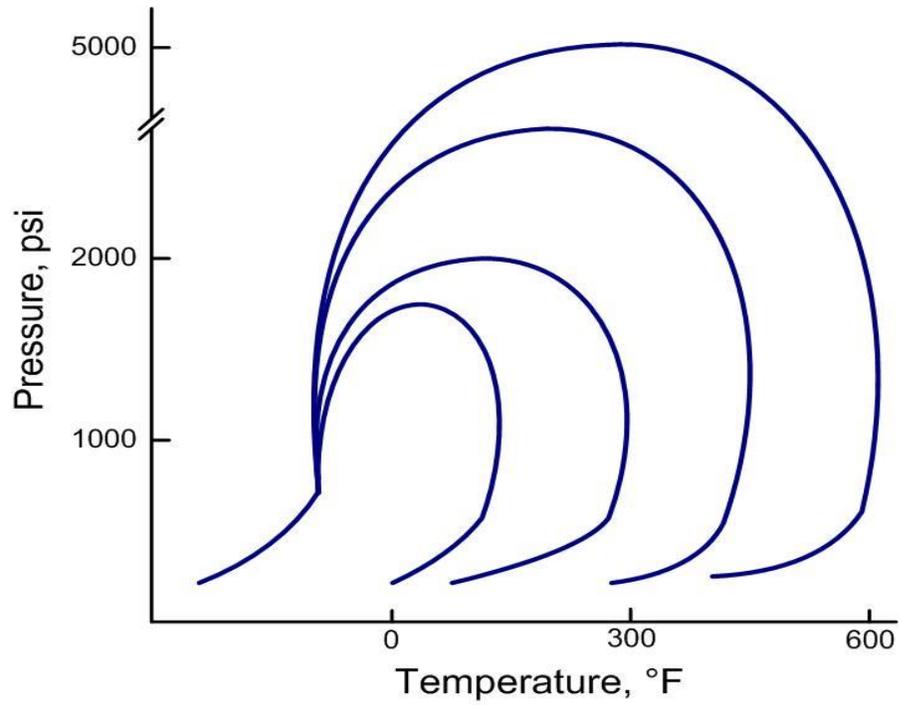


Figure 3 PT-Diagram for different hydrocarbon mixtures

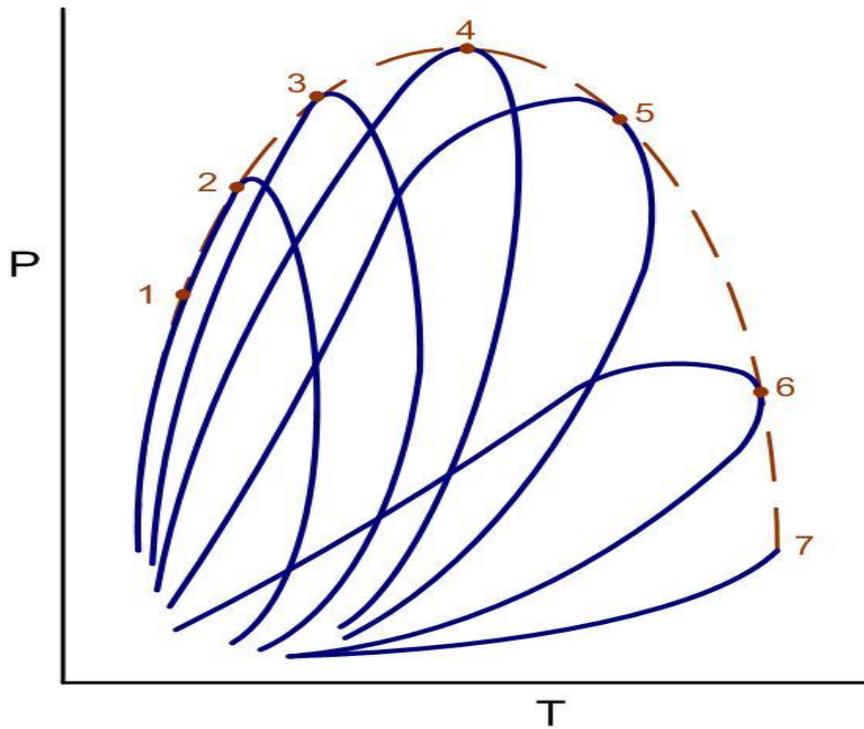


Figure 4 Locus of Temperature based on mixture composition

Methane	0.93
Ethane	0.013
Propane	0.0052
i Butane (Isobutane)	0.0123
n Butane	0.0156
i Pentane	0.01
n Pentane	0.01
N-Hexane	0.0023
N-Heptane	0.0001
Nitrogen	0.0015

Figure 5 Gas Composition for vapor phase

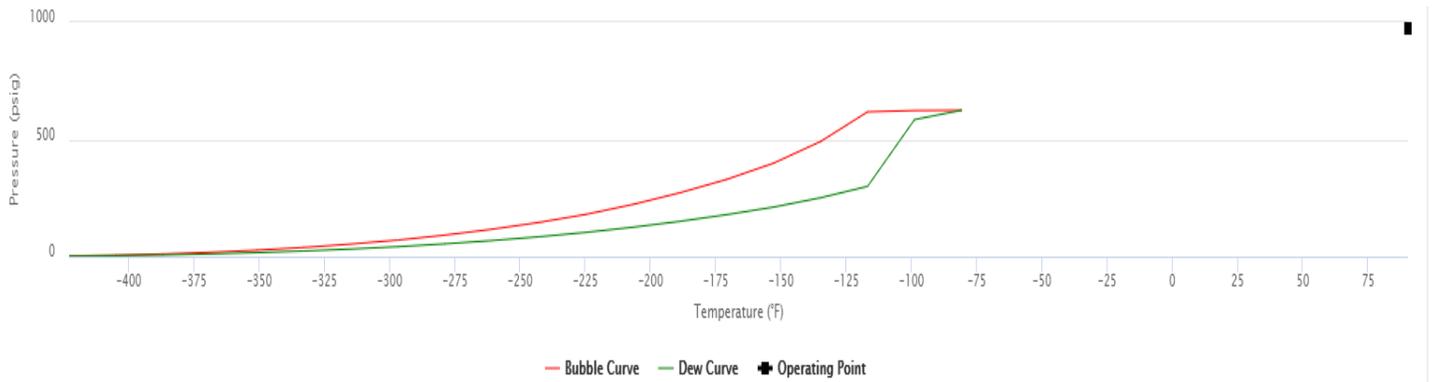


Figure 6 Phase Diagram for vapor phase

Methane	0.1
Ethane	0.2
Propane	0.1
N-Hexane	0.2
N-Heptane	0.2
n Octane	0.2
Water	0

Figure 7 Gas Composition for liquid phase

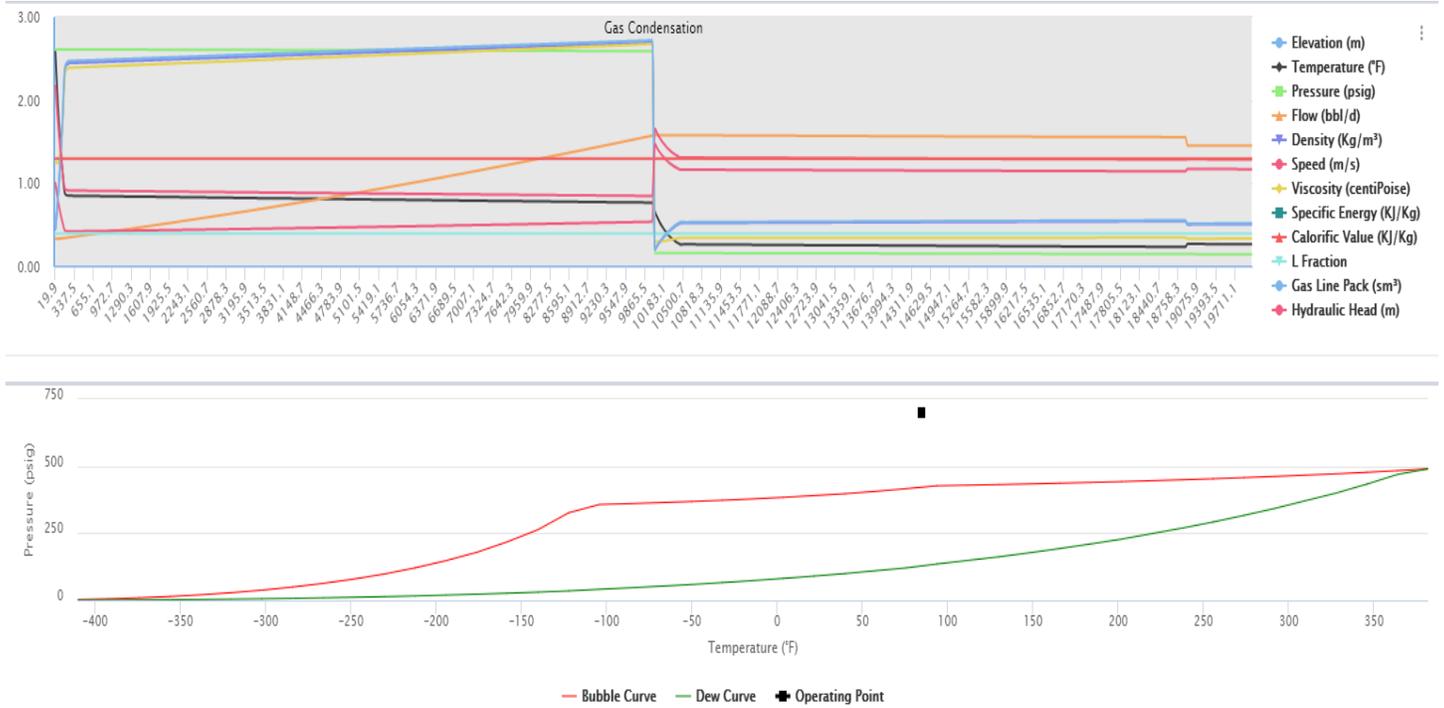


Figure 8 Pipeline Profile with Gas Condensation; Phase Diagram for liquid phase in pipeline

Methane	0.1
Carbon Dioxide	0.063
Ethane	0.1
Propane	423039375203384
i Butane (Isobutane)	0.1400065082980
n Butane	126911812561015
i Pentane	569476082004556
n Pentane	0.2
N-Hexane	0.2
N-Heptane	0.07
Nitrogen	0.05

Figure 9 Gas Composition for both liquid and vapor phase

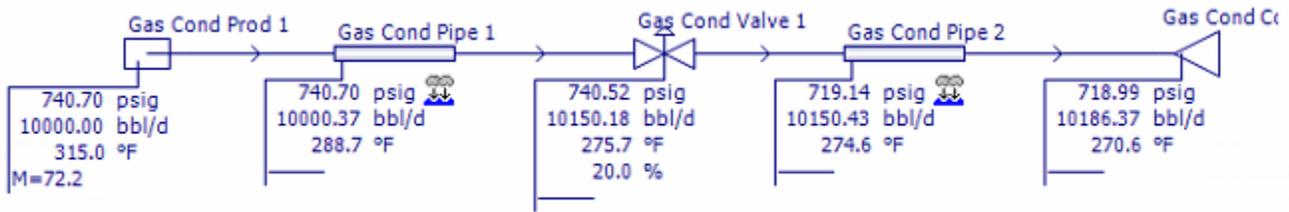


Figure 10 Simulation displaying high-pressure operation

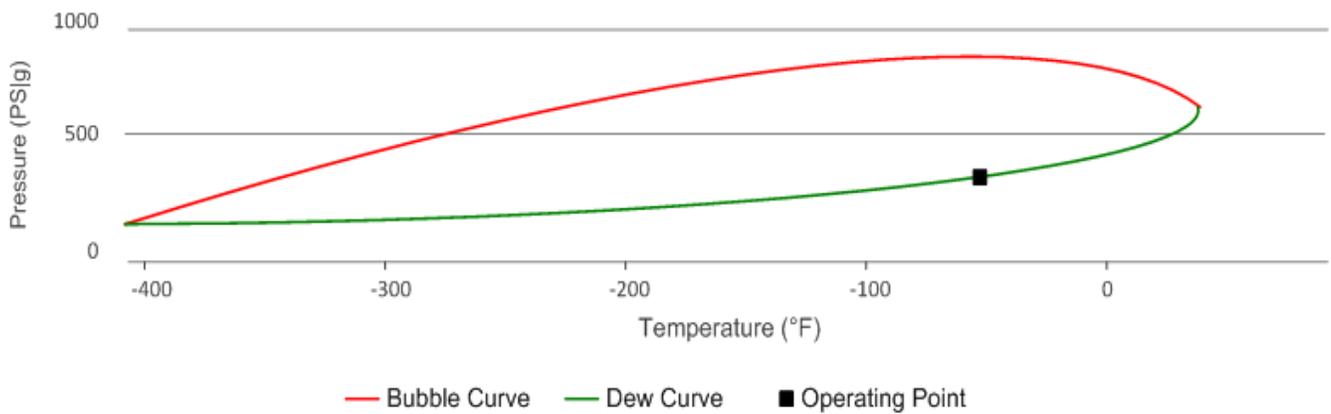
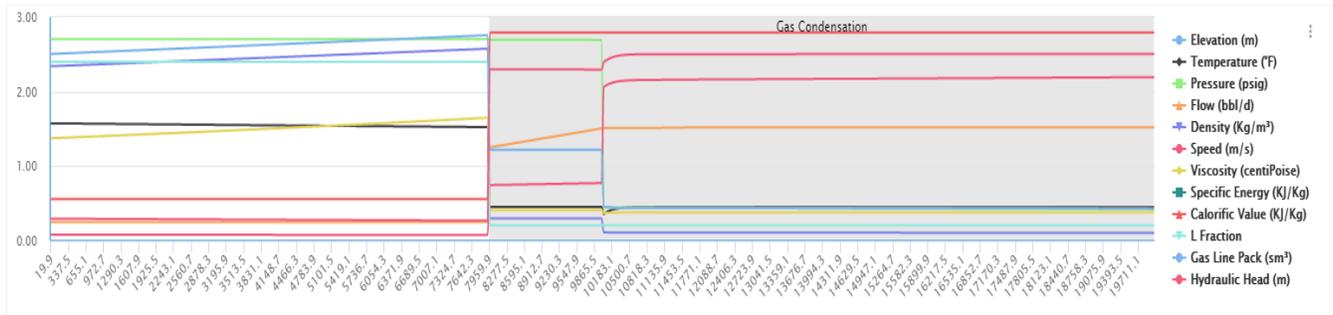


Figure 11 Pipeline Profile with Gas Condensation; Phase Diagram for liquid phase in pipeline

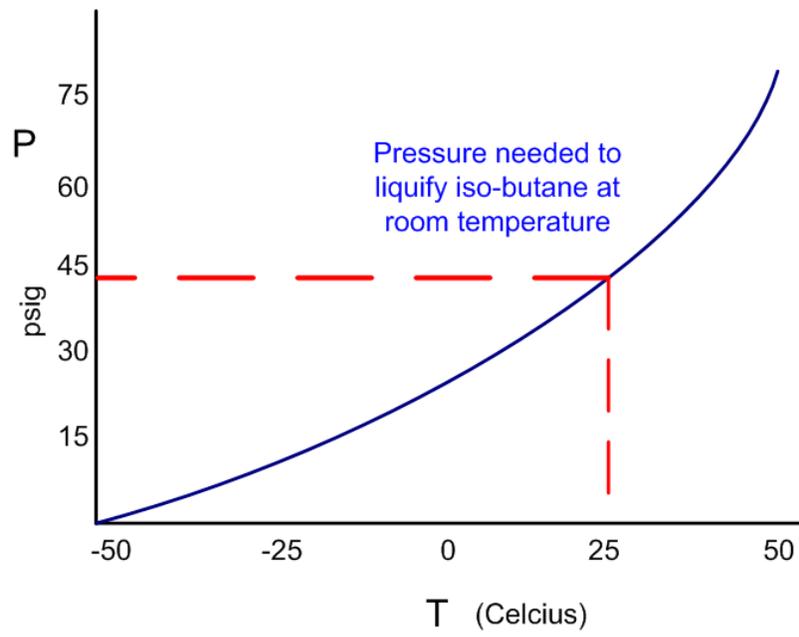


Figure 12 Iso-butane PT diagram