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HYDROCARBON PROCESSING®

Prevent system hydrate formation during sudden depressurization

Using this technique can help size gas plant inhibitor packages

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A hydrate is a physical combination of water (H_2O) and other small molecules producing a solid, which has an ice-like appearance but possesses a different molecular structure. Formation in gas and/or natural gas liquid (NGL) systems can plug pipelines, equipment and instruments, and restrict or interrupt flows. There are three crystalline structures for hydrates. In each, H_2O molecules—host molecules—build the lattice, and hydrocarbons, nitrogen (N_2), xenon (Xe), argon (Ar), carbon dioxide (CO_2) and hydrogen sulfide (H_2S)—guest molecules—occupy the cavities. Smaller molecules such as methane (CH_4), ethane (C_2H_6), CO_2 and H_2S stabilize the body-centered cubic called Structure I. Larger molecules propane (C_3H_8), isobutane (iso-C_4H_{10}) and n-butane (n-C_4H_{10}) form a diamond-lattice called Structure II. Molecules larger than n-C_4H_{10} do not form hydrates as they are too large to stabilize the lattice.[1]

Gas composition determines structure type and mixed gases will typically form Structure II lattices. Limiting hydrate numbers (the ratio of H_2O molecules to molecules of included gaseous component) are calculated using gas molecule sizes and cavity sizes in a H_2O lattice. The structure type does not affect appearance, properties or problems caused by the hydrate; it

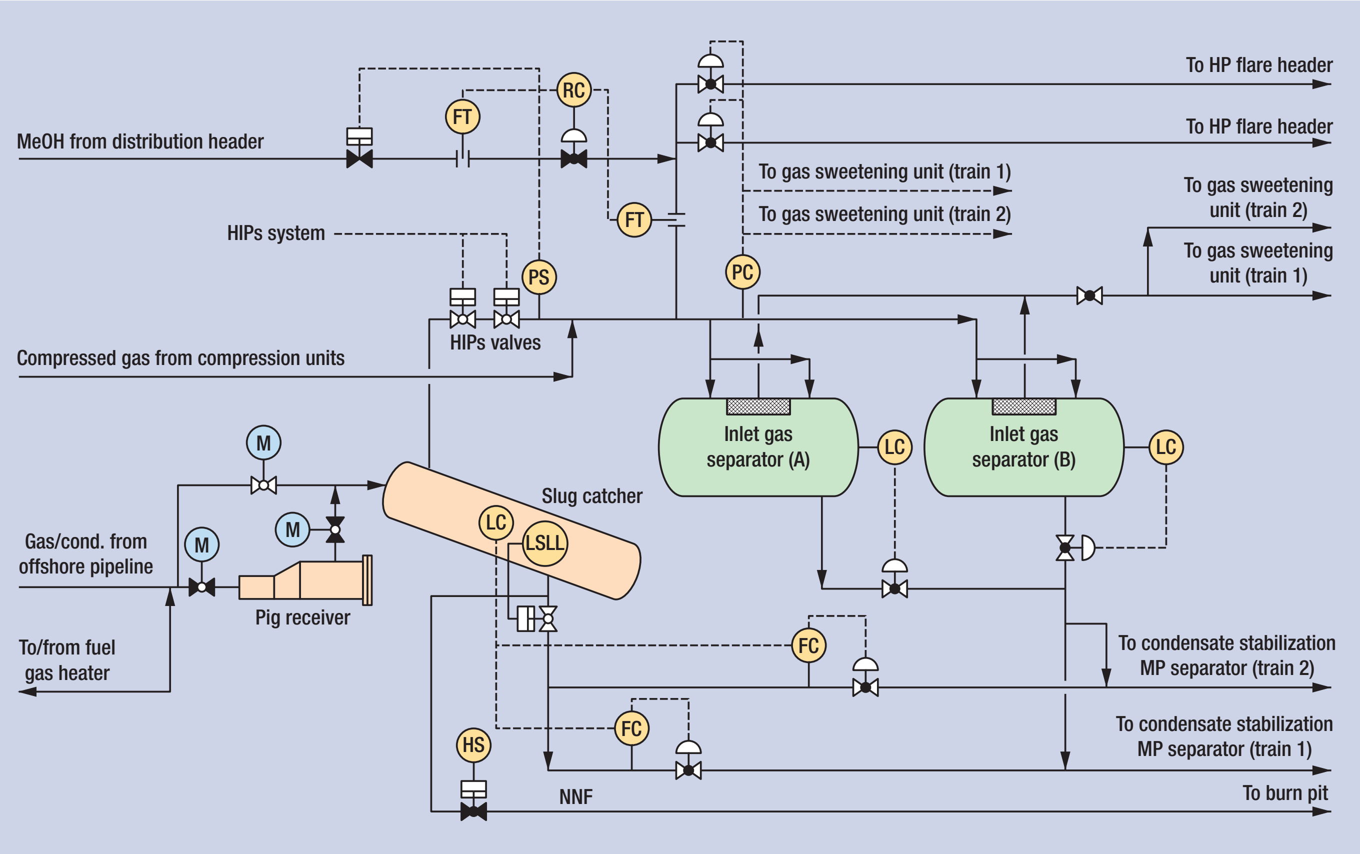

FIG. 1 A typical gas and condensate reception/separation unit PFD.

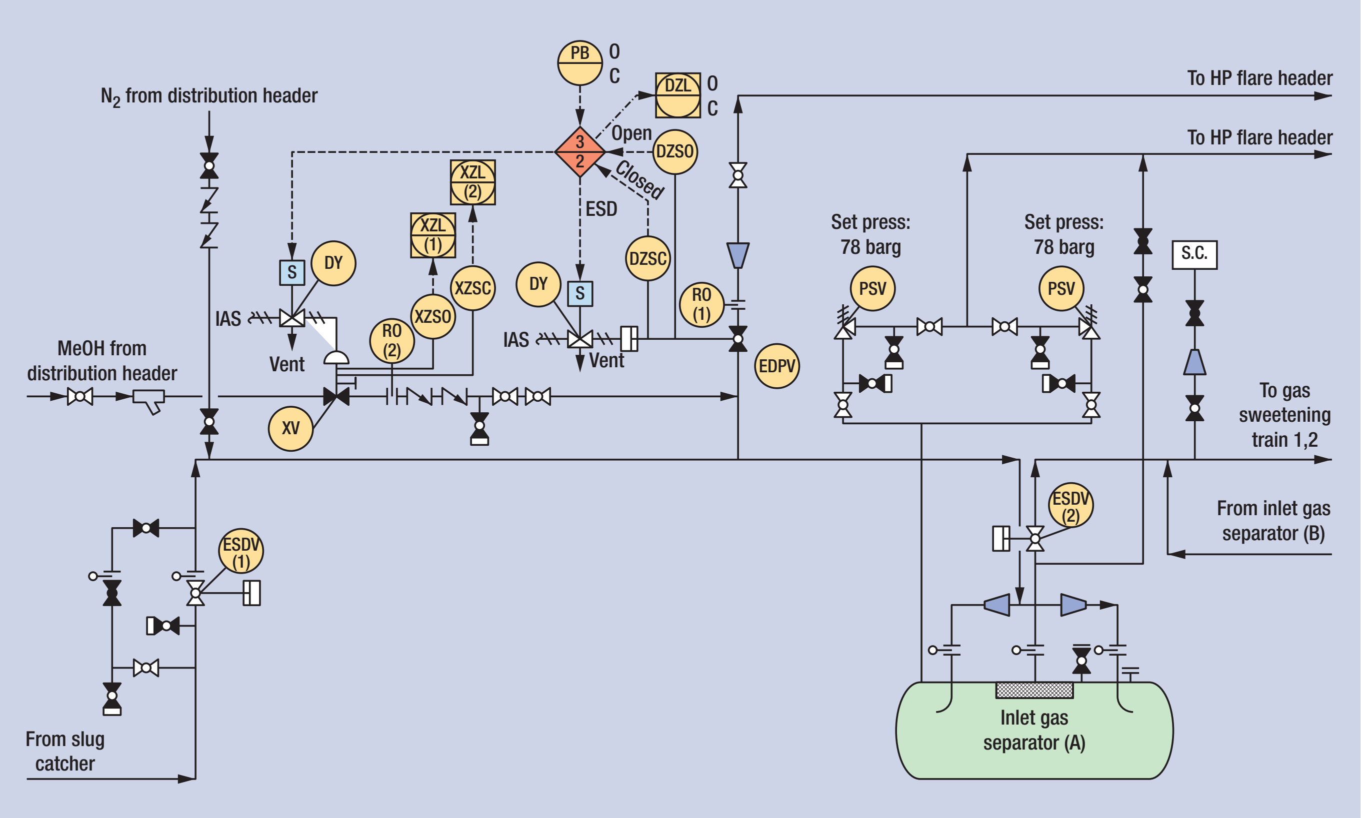

FIG. 2 A typical pipeline between slug catcher and inlet gas separator P&ID.

does, however, have a significant effect on the pressure and temperature at which hydrates form. Also Structure II hydrates are more stable than Structure I. Structure H hydrates are a double hydrate that requires two kinds of molecules to be stabilized: a small molecule like Xe, Ar or H_2S and a larger molecule like methylcyclohexane.[2,3]

Natural gas mixtures that have H_2S present result in a substantially warmer hydrate formation temperature at a given pressure. CO_2, in general, has a much smaller impact and often reduces the hydrate formation temperature at fixed pressure for a hydrocarbon gas mixture.[4,5]

Hydrate and hydrate formation processes are well known, and this phenomenon is used as a means to store natural gas and desalinize seawater. However, in the gas industry, hydrate formation in pipelines causes operational problems which ought to be considered during basic design. Consequently, additives like methanol (MeOH), ethylene glycol (EG) and triethylene glycol (TEG)—or their mixtures in hydrate inhibitor packages—are injected into pipelines preventing gas hydrate formation. Due to depressurization in a natural gas plant, produced H_2O naturally contains salts and electrolyte solutions such as sodium chloride (NaCl), potassium chloride (KCl) and calcium chloride ($CaCl_2$). These act as inhibitors but are not used individually as hydrate depressive media.

Being able to calculate adequate amounts of inhibitors to be injected into pipelines for flare networks is important from an economical viewpoint. Determining the type and optimum amount of inhibitor is the first step in designing such packages. Thus, there is an incentive to demonstrate a simple methodology to select inhibitor type, calculate its effectiveness and find an economic flowrate.

Fundamental equations. For a gas system, hydrate and pure H_2O coexist with each other at equilibrium. These equations determine the equilibrium temperature when H_2O is present:[6,7,18]

$$\Delta\mu_w^o/RT_R - \int_{T_R}^{T_o}\left(\Delta h_w/RT^2\right)dT + \left(\Delta\upsilon_w/RT_o\right)P$$
$$-\sum_i \upsilon_i \, ln\left(1+\sum_j C_{ij}f_j\right) - ln\, x_w = 0 \qquad (1)$$

$$\Delta h_w = \Delta h_w^o + \int_{T_R}^{T_o} \Delta C_{p,w}\, dT \qquad (2)$$

$$\Delta C_{p,w} = a + b\left(T_o - T_R\right) \qquad (3)$$

The Langmuir constants in Eq. 1 can be calculated:[8]

$$C_{ij}(T) = 4\pi/kT \int_0^\infty exp\left[-\omega(r)/kT\right] r^2\, dr \qquad (4)$$

In Eq. 4, $\omega(r)$ is the spherically symmetric cell potential which can be expressed as a function of the radius, coordination number and Kihara potential parameters.[8] The gas phase fugacity, f_i, can be calculated using a reliable PVT equation of state.[9]

To suppress hydrate temperature at a given pressure, alcohol and glycols are usually used in inhibition packages. The calculation to suppress hydrate temperature for a gas hydrocarbon plus H_2O is:[1,10,11,14]

$$ln\, a_w = -\frac{\Delta H}{nR}\left(\frac{1}{T} - \frac{1}{T_o}\right) \qquad (5)$$

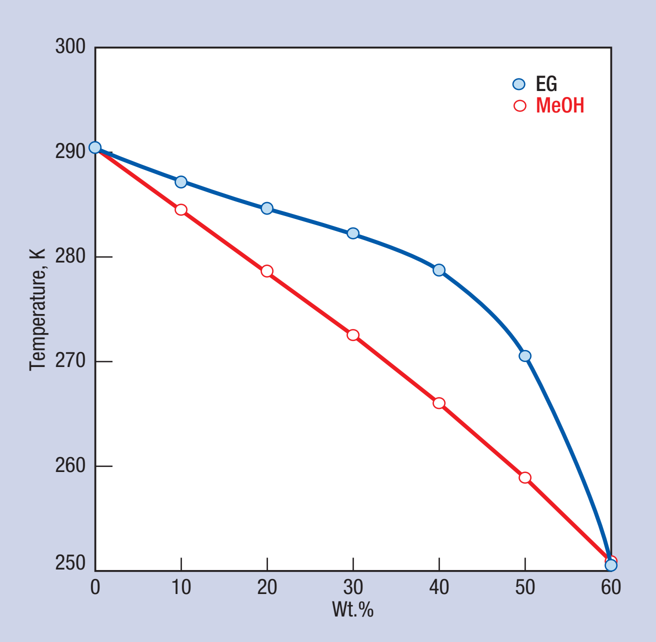

FIG. 3 Gas mixture hydrate-formation temperature comparison in the presence of MeOH and EG at 79 bara (upstream of EDPV).

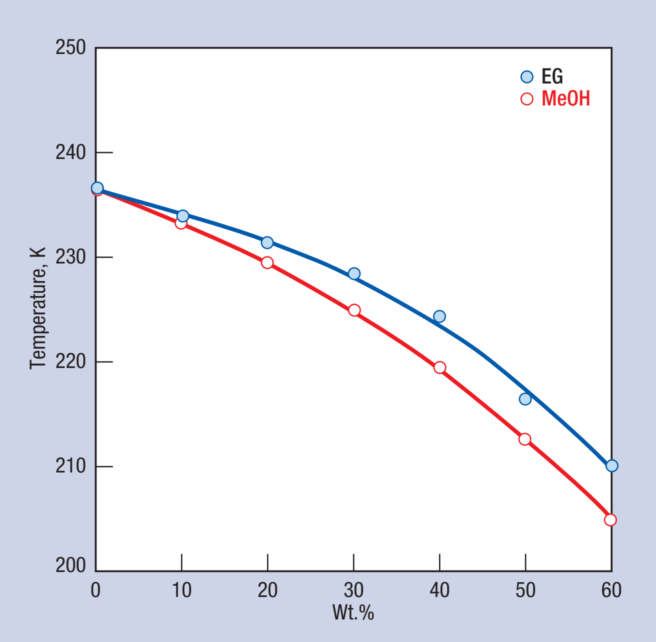

FIG. 4 Gas mixture hydrate-formation temperature comparison in the presence of MeOH and EG at 1.4 bara (downstream of EDPV).

where:

a_w is H_2O activity

ΔH is hydrate formation enthalpy

n is the hydrate number

R is the universal gas constant

T_o is the hydrate temperature in the absence of inhibitors (pure H_2O)

T is the suppressed temperature.

Predicting suppressed hydrate temperature in the presence of alcohol, glycols, electrolytes and mixtures must be outlined.

Alcohol. To solve Eq. 5, when hydrates coexist with an alcohol solution, H_2O activity may be found by the Margules equation:

$$\ln a_w / x_w = (1-x_w)^2 \left[A + 2(A-B)x_w\right] \qquad (6)$$

The expression of $\Delta H/nR$ for alcohol is:[1]

$$\frac{\Delta H}{nR} = \frac{2063}{\gamma_1 + \gamma_2 P + \gamma_3 \ln P} \qquad (7)$$

The parameters in the above equation are also valid for MeOH, EG and glycerol.[6]

Electrolytes. H_2O activity in a hydrate coexisting with an aqueous electrolyte mixtures is expressed:[12]

$$\ln a_w = \sum_k \left(m_k / m_k^{\circ}\right) \ln a_{w,k}^{\circ} \qquad (8)$$

where $\ln a_{w,k}^{\circ}$ is calculated using a developed software model.[13] $\Delta H/nR$ is calculated:[11,14]

$$\frac{\Delta H}{nR} = \frac{\alpha_0 I^{\alpha_1}}{1 + \beta_0 P + \beta_1 \ln P} \qquad (9)$$

Electrolytes and alcohol mixtures. H_2O activity in the case of hydrates coexisting with a electrolytes and alcohol mixture is:[6,18]

$$\ln a_{w,mix.} = \ln a_{w,al} + \ln a_{w,el} \qquad (10)$$

where $\ln a_{w,al}$ is obtained by Eq. 6 and $\ln a_{w,el}$ by Eq. 8.

To find hydrate formation enthalpy, a method that does not consider mixture composition by averaging formation enthalpy in separate mixtures of electrolytes plus H_2O and alcohol plus H_2O should be considered.[6] The harmonic average technique can be used for this purpose:

$$\frac{\Delta H}{nR} = \frac{2\left(\frac{\Delta H}{nR}\right)_{al}\left(\frac{\Delta H}{nR}\right)_{el}}{\left(\frac{\Delta H}{nR}\right)_{al} + \left(\frac{\Delta H}{nR}\right)_{el}} \qquad (11)$$

where $\left(\frac{\Delta H}{nR}\right)_{al}$ and $\left(\frac{\Delta H}{nR}\right)_{el}$ must be calculated using Eqs. 7 and 9, respectively.

Inhibition package sizing procedure. In hydrocarbon processing industries (HPIs), an important aspect in basic engineering is to perform safety and risk analyses.[15] Fire, mal-operation, equipment failure and other such events are summarized in a report after a hazard and operational study (HAZOP), and/or safety meetings handled by experienced engineers, are performed. The primary goal is to pinpoint where and how dangerous situations may arise and, secondly, to propose the best possible preventive actions.

One consideration is to perform a study on gas vapor buildup in process equipment. To design an effective and reliable safety instrumented system (SIS), many standards such as API must be considered.[16] During abrupt pipeline or equipment depressurization, contents are normally released to the atmosphere or through a flare network. Clogging emergency depressurizing valves (EDPVs), pressure safety valves (PSVs) and flare header

TABLE 1. Raw natural gas and its composition used in this study

Component	Mole%
N_2	3.12
CO_2	1.85
H_2S	0.50
CH_4	86.08
C_2H_6	5.45
C_3H_8	1.79
Iso-C_4H_{10}	0.30
N-C_4H_{10}	0.45
Iso-C_5H_{12}	0.13
N-C_5H_{12}	0.12
Light Aromatics	0.01
C_6^+	0.20

TABLE 2. The blowdown rates calculated for EDPV based on API RP 521

Step	Time, h	Vent rate, kg/h	Vent rate, m^3/h
1	0.000	22,209.7	278.9
	0.125	18,719.1	275.2
2	0.375	11,205.1	268.0
	0.500	6,132.5	263.0
3	0.625	4,345.4	261.0
	0.750	3,095.9	258.6
4	0.875	2,218.2	256
	1.125	1,405.5	252.3
5	1.375	731.8	246.3

TABLE 3. P-T history calculated for upstream and downstream of EDPV and also predicted hydrate formation temperature at each step of depressurization process

Step	Time, h	Upstream pres., bara	Upstream temp., K	Upstream[2] hydrate temp., K	Downstream pres., bara	Downstream temp., K	Downstream[2] hydrate temp., K
1	0.000	79.001[1]	287.2	290.4	1.40	236.2	236.5
2	0.375	24.18	228.7	281.2	1.32	206.4	233.3
3	0.500	16.97	216.3	277.6	1.31	200.1	231.4
4	0.750	8.47	196.0	263.9	1.30	187.8	227.6
5	1.125	3.03	173.4	237.7	1.30	171.2	223.1
6	1.375	1.57	162.7	224.3	1.30	162.3	221.2

[1] Design pressure of inlet gas separator. Operating pressure and temperature are 71.5 barg and 287.2 K, respectively.
[2] Hydrate temperatures computed by created software and counter-checked with industry-available software.

TABLE 4. Released gas analysis at each step of P-T history of depressurization process

			Mole fraction			
Component	Step 1	Step 2	Step 3	Step 4	Step 5	Step 6
N_2	0.0312	0.0319	0.0321	0.0324	0.0325	0.0322
CO_2	0.0185	0.0185	0.0185	0.0184	0.0182	0.0183
H_2S	0.0050	0.0048	0.0046	0.0042	0.0036	0.0034
C_1	0.8608	0.8742	0.8801	0.8898	0.8998	0.9030
C_2	0.0545	0.0527	0.0512	0.0476	0.0423	0.0405
C_3	0.0179	0.0141	0.0113	0.0068	0.0034	0.0024
iC_4	0.0030	0.0016	0.0010	0.0004	0.0001	0.0001
n-C_4	0.0045	0.0018	0.0010	0.0003	0.0001	0.0001
iC_5	0.0013	0.0002	0.0001	0.0001	0.0000	0.0000
n-C_5	0.0012	0.0001	0.0001	0.0000	0.0000	0.0000
Lt. Arom.	0.0001	0.0000	0.0000	0.0000	0.0000	0.0000
C_6^+	0.0020	0.0001	0.0000	0.0000	0.0000	0.0000
Total	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**

inlets with hydrates that are formed due to pressure reduction can impose grave and perilous operating problems.

Injecting an inhibitor into the system is an accepted industry practice simplifying the pressure reduction process and preventing hydrate formation. To demonstrate sizing procedures for inhibition packages, a gas and condensate reception/separation unit for an onshore refinery is used as an example.

Referring to the process flow diagram (PFD) in Fig. 1, the incoming feed facilities are designed to receive hydrocarbon gas and liquid from an offshore pipeline. Feed is then separated into gas and liquid streams so that it can be further processed in the gas sweetening and condensate stabilization units. It also provides the ability to handle random slugs formed during normal operation and those generated by periodic pigging operations.

The hydrocarbon fluid from the offshore pipeline flows into the slug catcher where the hydrocarbon gas and liquid are coarsely separated. The gas flows under control via the inlet gas separator to the gas sweetening units. The inlet gas separator is located downstream of the slug catcher and is designed to remove liquid carry-over from the coarse gas liquid separation in the slug catcher. The liquid flows under level control to the condensate stabilization units.

To prevent a worst-case scenario for a safety integrity level (SIL), the plant will be provided with an emergency shutdown (ESD) system. This ensures safe isolation and equipment shutdown under fault conditions. In gas industries, due to H_2O coexisting with gas and condensate hydrocarbons, the probability of hydrate formation in process pipelines is high during emergency depressurization. Therefore, all pipelines between the pig receiver, slug catcher and inlet gas separators (Fig. 1) must be well appointed with an effective and reliable ESD system.

All pipelines connected to an ESD valve, which is equipped with a hydrate inhibitor injection system, are part of the ESD system. A piping and instrument diagram (P&ID) shows details between the slug catcher and one of the inlet gas separators (Fig. 2). The ESD valve is designed to depressurize and protect the inlet gas separator. Process thermodynamics manifests while a pressurized system is relieving its contents as gas, thus the system's pressure and temperature (P-T) changes with respect to time. Therefore, P-T history must be acknowledged and realized before a hydrate formation calculation at any given pressure can be done. Table 1 shows gas composition used and the C_6^+ fraction is characterized as n-decane and the light aromatics as benzene.

P-T history. When metal is exposed to fire on one side with vapor/liquid on the other, the metal temperature may reach a level at which a stress rupture may occur; even though the pressure within the vessel does not exceed the allowable over-pressure (110% of the design pressure). It is on this basis that an emergency vapor depressurizing (blowdown) system is used.

TABLE 5A. Upstream inhibitor flowrate calculation at step 1 of EDPV depressurization process (*P* = 79 bara)

Inhibitor[1] wt%	Hydrate[2] temp., K	Depression[3] value, K	Flowrate,[4] kg/h		
			100%[5]	96%[5]	90%[5]
0	290.4	–3.2	–	–	–
10	284.5	2.7	0.11	0.12	0.13
20	278.6	8.6	0.25	0.26	0.29
30	272.5	14.7	0.43	0.45	0.50
40	266.0	21.2	0.67	0.71	0.80
50	258.9	28.3	1.00	1.09	1.25
60	250.9	36.3	1.50	1.67	2.00

[1] MeOH is selected as inhibitor in inhibition package. Also, wt% in this column shows inhibitor wt% in the condensed liquid H_2O phase in the pipeline.
[2] Hydrate temperatures are computed by created software and counter-checked by industry-available software.
[3] Depression value is defined as system temperature (i.e., 287.2 K) minus hydrate temperature.
[4] Inhibitor flowrate based on 1 kg/h condensed H_2O phase.
[5] Wt% indicates inhibitor purity used in inhibition package.

TABLE 6A. Upstream inhibitor flowrate calculation at step 6 of EDPV depressurization process (*P* = 1.57 bara)

Inhibitor[1] wt%	Hydrate[2] temp., K	Depression[3] value, K	Flowrate,[4] kg/h		
			100%[5]	96%[5]	90%[5]
30	213.0	–50.3	–	–	–
40	207.4	–44.7	0.11	0.12	0.13
50	200.8	–38.1	0.25	0.26	0.29
60	193.0	–30.3	0.43	0.45	0.50
70	183.8	–21.1	0.67	0.71	0.80
80	172.7	–10.0	1.00	1.09	1.25
90	156.5	6.2	1.50	1.67	2.00

[1] MeOH is selected as inhibitor in inhibition package. Also, wt% in this column shows inhibitor wt% in the condensed liquid H_2O phase in the pipeline.
[2] Hydrate temperatures are computed by created software and counter-checked by industry-available software.
[3] Depression value is defined as system temperature (i.e., 162.7 K) minus hydrate temperature.
[4] Inhibitor flowrate based on 1 kg/h condensed H_2O phase.
[5] Wt% indicates inhibitor purity used in inhibition package.

Depressurization is required for both operational reasons and emergency situations. During an emergency, it is used to rapidly reduce pressure in the plant significantly below operating pressures. To facilitate a swift and controlled blowdown, the process plant is segregated by ESDVs into discrete sections. Each section has at least one emergency depressurizing point (Figs. 1 and 2).

Each system emergency depressurization is made possible via a dedicated emergency blowdown line connected to the plant flare network. In the blowdown line, a restriction orifice (RO) is installed, across which pressure reduction is carried out. The RO should be sized to achieve the required blowdown rate. The ESDV is upstream of the RO and is equipped with on/off action and an inhibitor injection package.

Blowdown rates. These are calculated according to API RP 521, using a process simulation program depressurization module.[16, 17] This can be used in a fire case mode with a supersonic valve, simulating blowdown under heat input from fire conditions. In this example, the depressurizing stream is modeled from system operating pressure to system design pressure and heat input at a constant system volume (i.e., constant stream density). Table 2 shows these results.

TABLE 5B. Downstream inhibitor flowrate calculation at step 1 of EDPV depressurization process (*P* = 1.4 bara)

Inhibitor[1] wt%	Hydrate[2] temp., K	Depression[3] value, K	Flowrate,[4] kg/h		
			100%[5]	96%[5]	90%[5]
0	236.5	–0.3	–	–	–
10	233.3	2.9	0.11	0.12	0.13
20	229.4	6.8	0.25	0.26	0.29
30	224.8	11.4	0.43	0.45	0.50
40	219.3	16.9	0.67	0.71	0.80
50	212.7	23.5	1.00	1.09	1.25
60	205.0	31.2	1.50	1.67	2.00

[1] MeOH is selected as inhibitor in inhibition package. Also, wt% in this column shows inhibitor wt% in the condensed liquid H_2O phase in the pipeline.
[2] Hydrate temperatures are computed by created software and counter-checked industry-available software.
[3] Depression value is defined as system temperature (i.e., 236.2 K) minus hydrate temperature.
[4] Inhibitor flowrate based on 1 kg/h condensed H_2O phase.
[5] Wt% indicates inhibitor purity used in inhibition package.

TABLE 6B. Downstream inhibitor flowrate calculation at step 6 of EDPV depressurization process (*P* = 1.30 bara)

Inhibitor[1] wt%	Hydrate[2] temp., K	Depression[3] value, K	Flowrate,[4] kg/h		
			100%[5]	96%[5]	90%[5]
30	210.3	–48	–	–	–
40	204.9	–42.6	0.11	0.12	0.13
50	198.4	–36.1	0.25	0.26	0.29
60	190.7	–28.4	0.43	0.45	0.50
70	181.8	–19.5	0.67	0.71	0.80
80	171.0	–8.7	1.00	1.09	1.25
90	155.5	6.8	1.50	1.67	2.00

[1] MeOH is selected as inhibitor in inhibition package. Also, wt% in this column shows inhibitor wt% in the condensed liquid H_2O phase in the pipeline.
[2] Hydrate temperatures are computed by created software and counter-checked by industry-available software.
[3] Depression value is defined as system temperature (i.e., 162.3 K) minus hydrate temperature.
[4] Inhibitor flowrate based on 1 kg/h condensed H_2O phase.
[5] Wt% indicates inhibitor purity used in inhibition package.

Blowdown temperature. For process equipment design, minimum blowdown temperature should be determined. This identifies construction materials and also the required hydrate inhibitor. Similar to determining blowdown rate, blowdown temperatures are computed using a depressurization module in a process simulation program. It is used in the adiabatic mode, which simulates user blowdown, selected isentropic efficiency and heat transfer area. Upstream and downstream temperatures of the relieved gas related to EDPVs and its composition are shown in Tables 3 and 4.

Hydrate formation. By using a calculation software package and process simulation, hydrate formation temperatures at each depressurization process step can be obtained. Hydrate formation conditions were calculated and reported in Table 3. Here saturated hydrocarbon gas mixture with H_2O is defaulted.

Package specification. Two main characteristics for any hydrate inhibition package are:

- Type
- Flowrate.

Determining these two parameters must be done by P-T history prediction and hydrate formation temperature.

Inhibitor type. Alcohol, glycols and their mixture can be used as inhibitors in inhibition packages. Gas composition and pressure heavily influence effectiveness and cost. Comparing different media on a hydrate formation temperature diagram can easily select the most suitable inhibitor. Hydrate formation temperature is a function of the inhibitor's wt% coexisting in equilibrium with gas and hydrate phases. Figs. 3 and 4 are a comparison between EG and MeOH for hydrate formation in the example synthetic gas mixture. For equal inhibitor weights, MeOH decreases hydrate formation temperature more than EG and, as a result, for this specific example, it appears to be the more suitable inhibitor.

Inhibitor flowrate. The injected inhibitor amount must be sufficient to prevent freezing the inhibitor H_2O phase. There must also be enough solution for the equilibrium vapor phase content of the inhibitor and its solubility in any liquid hydrocarbon. MeOH's vapor pressure is high enough that significant amounts will vaporize. EG, diethylene glycol (DEG) and TEG have been used for hydrate inhibition. The most popular is EG due to its lower cost, lower viscosity and lower solubility in liquid hydrocarbons.

Based on the material balance, inhibitor mass flowrate required in the H_2O phase is:

$$X = \frac{\varphi_2}{\varphi_1 - \varphi_2} \tag{12}$$

where:

X is injected inhibitor mass flowrate based on 1 kg/h condensed H_2O phase

φ_1 is inhibitor purity

φ_2 is inhibitor weight fraction in the condensed H_2O phase.

To select effective and suitable inhibitor flowrates, at each P-T history step, inhibitor quantity can be calculated based on the above outlined procedure. Tables 5 and 6 show typical inhibitor flowrates at steps 1 and 6 of the depressurizing process related to EDPV. The inhibitor flowrate extracted from Tables 5 and 6 should be doubled to compensate for losses to the hydrocarbon liquid phase and vaporization.

Using the method. Hydrate formation must be considered by many HPI facilities as it can impose serious and crucial operating hazards. Using inhibitors such as alcohol and glycols prevent formation effectively and economically.

A methodology for selection and sizing inhibition packages is a useful tool in HPI facilities. The chosen emergency depressurization valve is responsible for rapid and safe system depressurization. System P-T history should therefore be known before and after the pressure reduction valve. Having established this information, hydrate formation temperature at each depressurization step can then be computed using calculations and simulation software packages.

A suitable inhibitor can then be selected by comparing different media on a hydrate formation temperature diagram. Based on the material balance the mass flowrate of the chosen inhibitor is computed by using Eq. 12. Having established the inhibitor type and flowrate, the entire inhibition package can then be accurately and reliably sized, thus improving facility operations and economics. **HP**

NOMENCLATURE

a = Constant in Eq. 3
a_w = Activity of H_2O
$a^{o}_{w,k}$ = Activity of H_2O in a binary solution containing H_2O and electrolyte k with the same ionic strength as the mixture of all electrolytes and H_2O
A = Constant in Eq. 6
b = Constant in Eq. 3
B = Constant in Eq. 6
C = Langmuir constant, MPa
C_p = H_2O heat capacity, J/mol K
f = Fugacity, MPa
h = Enthalpy of pure H_2O, J/mol K
I = Ionic strength
k = Boltzman constant, J/K molecule
m_k = Molality of electrolyte k in the electrolyte solution, mol/1,000 g solvent
m^{o}_k = Molality of electrolyte k in an aqueous mixture containing only electrolyte k with the same ionic strength as the mixture, mol/1,000 g solvent
n = Hydrate number
P = Pressure, MPa
r = Separation distance between molecule cores, Å
R = Gas constant, 8.314×10^{-3}
T = Temperature, K

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x = Mole fraction in liquid H_2O phase
X = Inhibitor mass flowrate provided by inhibition package based on one kg/h condensed H_2O phase, kg/h

GREEK LETTERS

α_0, α_1 = Constants in Eq. 9
β_0, β_1 = Constants in Eq. 9
$\gamma_1, \gamma_2, \gamma_3$ = Constants in Eq. 7
υ = Molar volume, m^3/mol
φ_1 = Inhibitor purity used in inhibition package
φ_2 = Inhibitor weight fraction in the condensed H_2O phase
μ = Chemical potential, J/mol
Δ = Indicates difference in thermodynamic property value of H_2O in two phases
ω = Cell potential, J/molecule

SUPERSCRIPT

° = Reference state

SUBSCRIPTS

al = Alcohol
el = Electrolyte
i, j = Indicate gas species
mix = Mixture
o = Indicates hydrate temperature in the presence of pure H_2O
R = Reference state
w = H_2O phase

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