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Quantify Non-Equilibrium Flow and Rapid Phase Transitions (RPT)

An ioMosaic Corporation White Paper

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Quantify Non-Equilibrium Flow and Rapid Phase Transitions

Process Safety and Risk Management Practices

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1 Summary

Although non-equilibrium flow and rapid phase transitions (RPT) are well researched, the literature published so far does not explicitly quantify the RPT phenomenon or provide reliable methods for the calculation of non-equilibrium flow for mixtures. The objective of this paper is to provide a clear understanding of how non-equilibrium flow and rapid phase transitions develop and how they should be quantified for pure components and mixtures alike.

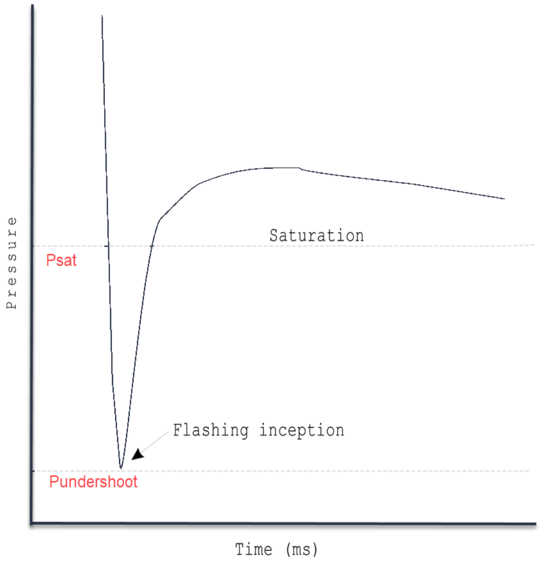
We present a thermodynamic treatment of non-equilibrium flow and rapid phase transitions for pure components and mixtures. We discuss the estimation of hazard potential based on the superheat limit. ioMosaic's SuperChems Expert$^{\text{TM}}$ software is used to model multi-component non-equilibrium flow and LNG spills and to illustrate how mixtures composition influences the development of rapid phase transitions, overpressure generation, and non-equilibrium flow rates.

2 Homogeneous and Heterogeneous Nucleation

Rapid depressuring of a vessel containing saturated liquid can lead to non-equilibrium flow followed by explosive boiling of the liquid contents. Depressuring can be attributed to flow and/or expansion. The same phenomenon, i.e. explosive boiling of liquids, can be induced by rapid heating of the liquid and is sometimes referred to as a rapid phase transition.

As shown to the right for vessel depressuring, the pressure can drop below the saturation point following rapid depressuring. The rate of pressure drop, Σ, influences this pressure undershoot which in turn influences the superheat available for bubble nucleation and growth. A large depressuring rate can lead to a large undershoot and thus a large bubble nucleation and growth superheat.

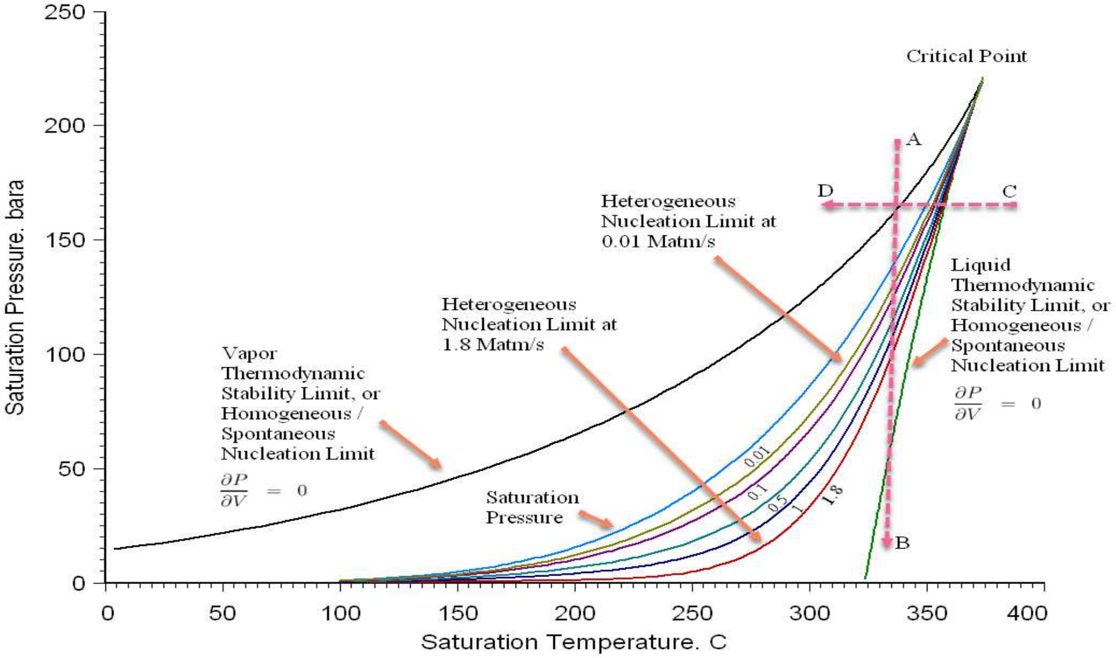
The pressure will recover when the pressure rise caused by bubble generation is equal to the rate of imposed pressure drop at flashing inception. If the rate of pressure drop is large enough, a metastable liquid can form upon depressuring.



A sharp pressure rise caused by homogeneous (spontaneous) and/or heterogeneous bubble generation follows [1, 2, 3, 4]. Heterogeneous nucleation occurs more frequently in flashing flow than homogeneous nucleation. Heterogeneous nucleation is more likely for dirty fluids with suspended impurities, fluids with dissolved gases, and where vessel and piping walls have rough flow surfaces leading to imperfect wetting. Virtually all liquids contain some dissolved gases. The presence of dissolved gas cause the pressure in the bubble to increase totaling the partial pressure of the gas and saturation pressure of the vapor. As a result, the bubble can grow at liquid pressures greater than the vapor pressure.

One should note that rapid depressuring or rapid heating/cooling can render heterogeneous bubble nucleation sites inactive. As the initial temperature/pressure approach critical conditions, depressuring rates required to cause a metastable liquid to form become smaller. Figure 1 illustrates how the flashing process for water can be influenced by rapid pressure drop or rapid heating leading to heterogeneous and/or homogeneous bubble nucleation and growth.

Figure 1: Flashing inception following a rapid pressure drop for water



Under a near isothermal pressure drop such as is experienced typically during an isentropic expansion of a subcooled liquid (see path A to B in Figure 1), the pressure drops below the saturation pressure, and at $\Sigma = 0.01 \text{ Matm/s}$, flashing will occur at a pressure below but near the saturation pressure. At $\Sigma = 1.8 \text{ Matm/s}$, flashing will occur at a pressure below saturation but near the thermodynamic stability limit. At extremely large values of Σ, flashing will have to occur at the thermodynamic stability limit. Lower values of the undershoot pressure drive more flow for subcooled liquids which is why non-equilibrium liquid flow occurs and is important for pressure relief and vent containment design and evaluation (also see [5]).

The pressure undershoot for water can be estimated from a correlation developed by Lienhard [6, 7]:

$$\psi = 0.1058\left[1+14\Sigma^{0.8}\right]\left(\frac{T}{T_c}\right)^{28.46} \tag{1}$$

$$G_b = \frac{W_{cr}}{k_B T} = \frac{16\pi\sigma^3\psi}{3k_B T\left[1-\frac{\rho_{g,sat}}{\rho_{l,sat}}\right]^2\left[P_{\text{sat}}-P_{\text{undershoot}}\right]^2} = 28.2 \tag{2}$$

$$\Delta P = P_{\text{sat}}-P_{\text{undershoot}}$$

$$= \underbrace{\left(\sqrt{\frac{0.1058\times 16\pi}{3\times 28.2}}\right)}_{0.250}\left(\frac{\sigma^{3/2}}{\sqrt{k_B T_c}}\right)\left(\frac{\left(\frac{T}{T_c}\right)^{13.73}\left[1+14\Sigma^{0.8}\right]^{1/2}}{\left[1-\frac{\rho_{g,sat}}{\rho_{l,sat}}\right]}\right) \tag{3}$$

where ψ is Lienhard's heterogeneity factor regressed from measured data, W_{cr} is the net work required to form a bubble having a critical size from classic homogeneous bubble nucleation theory, $k_B T$ is the kinetic energy of the molecules, G_b is the Gibbs number, ΔP is in Pa, T is the initial temperature in Kelvin, $k_B = 1.380649\times 10^{-23}$ J/K is Boltzmann's constant, σ is the liquid surface tension at T in N/m, Σ is the rate of pressure drop in Matm/s (Mega atmosphere/second), $\rho_{g,sat}$ and $\rho_{l,sat}$ are the mass density of vapor and liquid at equilibrium in kg/m^3, and T_c is the critical temperature in Kelvin. The correlation was developed for:

$$0.62 \leq \frac{T}{T_c} \leq 0.935 \tag{4}$$

$$0.005 \leq \Sigma \leq 1.8 \tag{5}$$

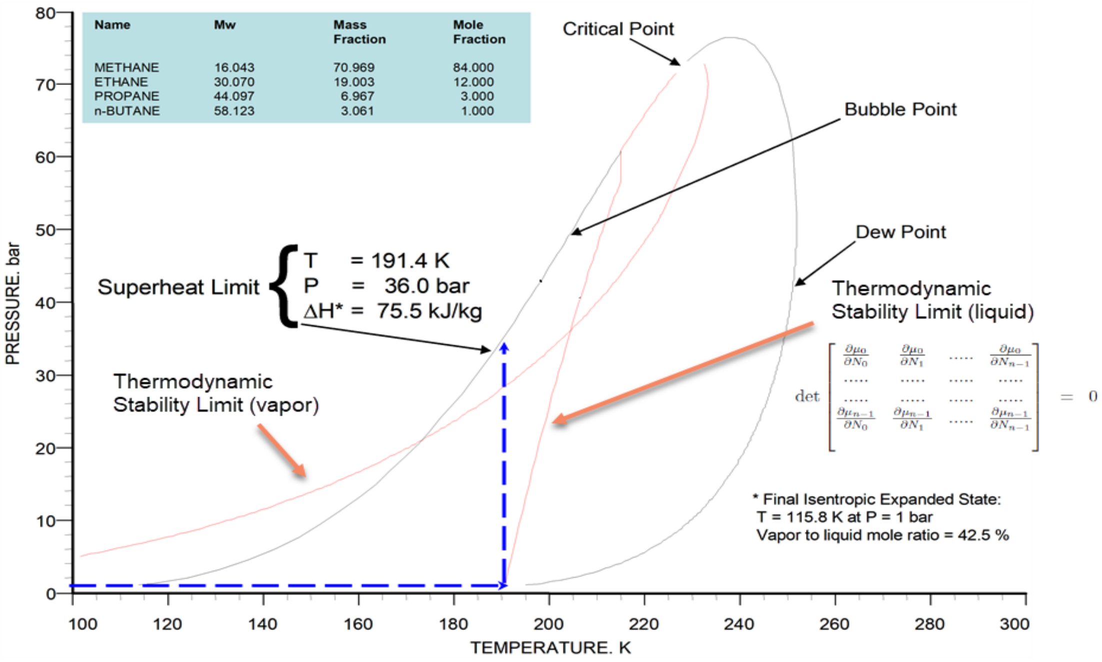
Equation 3 should not be used for lower depressuring rates without changing the equation constants. The rate of pressure drop can include both the contributions of transient vessel blowdown and flow acceleration leading to additional pressure drop in nozzles and/or piping [9]:

$$\Sigma = \underbrace{\left[\frac{\partial P}{\partial t}\right]_z}_{\text{Vessel}} + \underbrace{u\left[\frac{\partial P}{\partial z}\right]_t}_{\text{Nozzle and/or Piping}} \tag{6}$$

$$\underbrace{u\left[\frac{\partial P}{\partial z}\right]_t}_{\text{Nozzle}} \simeq \frac{\dot{m}^3}{\rho^2 A_{\text{noz}}^4}\frac{dA_{\text{noz}}}{dz} = \frac{G^3}{\rho^2}\frac{d\left[\ln A_{\text{noz}}\right]}{dz} \tag{7}$$

where P is the static pressure, z is the flow path length or axial distance, t is time, u is the flow velocity, A_{noz} is the nozzle minimum flow area, G is the mass flux, $\dot{m}$ is the mass flow rate, and ρ is the mass density. Note that Σ is in Matm/s. Flashing inception in nozzles or other flow geometries with restrictions causing flow acceleration will always occur at the throat or plane of minimum flow area. Turbulence may also be generated where flashing occurs. Equation 7 can be used based on the maximum value of dA_{noz}/dz for converging nozzles.

Figure 2: Rapid heating leading to a superheat limit rapid phase transition for an LNG mixture [8]



Name	Mw	Mass Fraction	Mole Fraction
METHANE	16.043	70.969	84.000
ETHANE	30.070	19.003	12.000
PROPANE	44.097	6.967	3.000
n-BUTANE	58.123	3.061	1.000

For flow through nozzles and/or piping, the undershoot pressure, $P_{\text{undershoot}}$, can be used to approximate Burnell's [10] constant C:

$$C \quad = \quad 1 - \frac{P_{\text{undershoot}}}{P_{\text{sat}}} \tag{8}$$

$P_{\text{undershoot}}$ cannot be less than the limit of mechanical/thermodynamic stability often referred to as the homogeneous nucleation limit or superheat limit. For a pure component that limit is established at:

$$\frac{\partial P}{\partial V}\,|_{T,N} \quad = \quad 0 \tag{9}$$

where V is the fluid volume. For a multicomponent mixture, the limit can still be calculated from an equation of state [11] by setting the determinant of the mixture partial molar chemical potential μ to zero:

$$\det \begin{bmatrix} \frac{\partial \mu_0}{\partial N_0} & \frac{\partial \mu_0}{\partial N_1} & \ldots\ldots & \frac{\partial \mu_0}{\partial N_{n-1}} \\ \ldots\ldots & \ldots\ldots & \ldots\ldots & \ldots\ldots \\ \ldots\ldots & \ldots\ldots & \ldots\ldots & \ldots\ldots \\ \frac{\partial \mu_{n-1}}{\partial N_0} & \frac{\partial \mu_{n-1}}{\partial N_1} & \ldots\ldots & \frac{\partial \mu_{n-1}}{\partial N_{n-1}} \end{bmatrix} \quad = \quad 0$$

where N_i is the number of moles of chemical i and n is the total number of chemicals in the mixture.

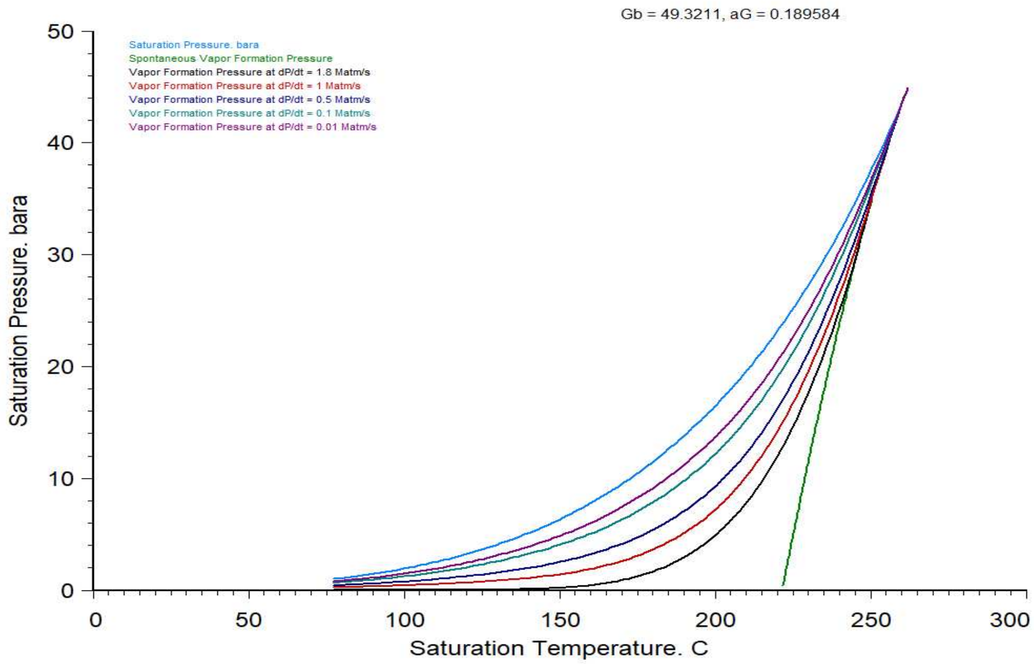
3 Extending Lienhard's Correlation to Other Chemicals and Mixtures

Equation 3 can be extended to other chemicals and mixtures [12, 13] if we assume that Lienhard's heterogeneity correction ψ which depends on reduced temperature applies equally to other chemicals and mixtures. However, the Gibbs number [14], G_b, has to be scaled relative to water:

$$G_b \simeq \underbrace{G_{b,w}}_{28.2} \left[\frac{\sigma}{\sigma_w}\right]^3 \left[\frac{T_{c,w}}{T_c}\right] \left[\frac{P_{\text{sat,w}}-1}{P_{\text{sat}}-1}\right]^2 \left[\frac{1-\frac{\rho_{v,w}}{\rho_{l,w}}}{1-\frac{\rho_v}{\rho_l}}\right]^2 \tag{10}$$

Where P_{sat} is the saturation or bubble point pressure in bara and ρ is the mass density in $\mathrm{kg/m^3}$ evaluated at a saturation temperature equal to 0.9 times the critical temperature. The surface tension ratio is not very sensitive to temperature and is evaluated at 298.15 K or the normal boiling point. Figure 3 shows a nucleation diagram for Acrylonitrile using Equation 10 yielding $G_b = 49.32$. The scaling proposed in this paper is consistent with the compilation of measured homogeneous nucleation limits for 90 pure substances and 28 mixtures [15].

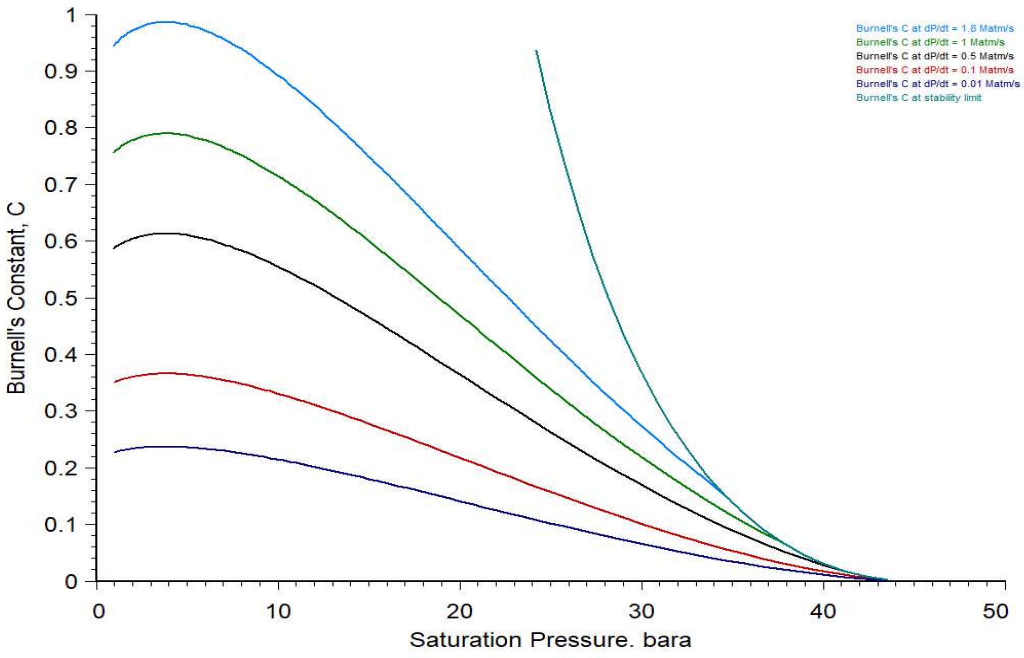
Figure 3: The superheat and nucleation limits for pure acrylonitrile



The nucleation limits shown in Figure 3 can be used to calculate Burnell's C constant from Equation 8 at different pressures as shown in Figure 4. Burnell [10] used C to approximate the impact of non-equilibrium on mass flux by modifying the standard Bernoulli flow equation:

$$G = \sqrt{2\rho_o \left[P_o - (1-C)\, P_{\text{sat}}\right]} \tag{11}$$

Figure 4: Burnell's C constant for pure acrylonitrile



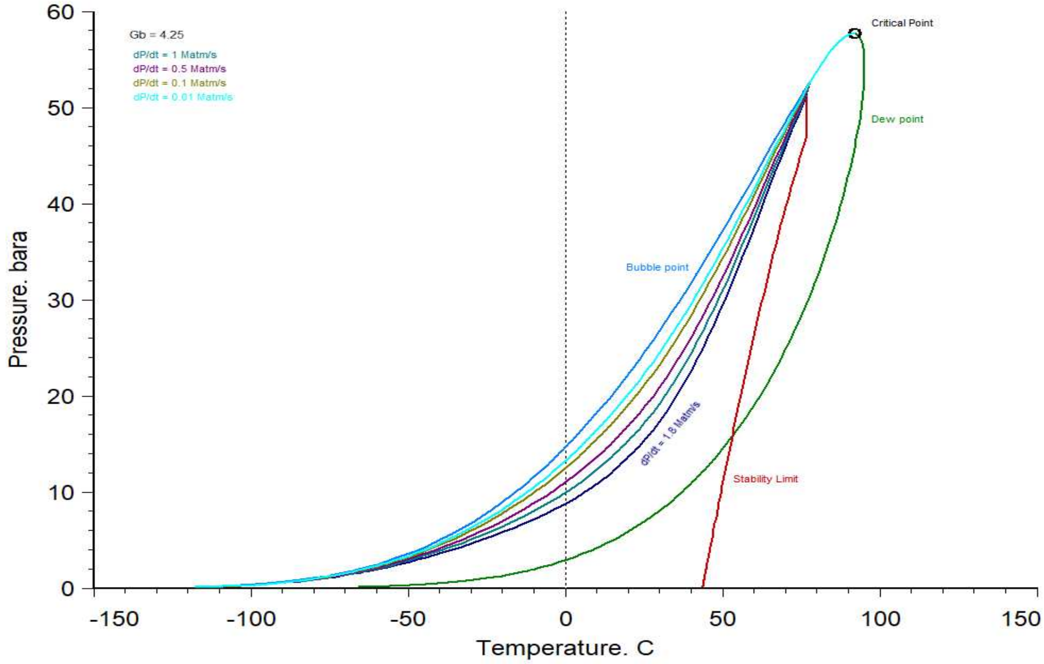
where G is the mass flux, ρ is the mass density, P_o is the system pressure, and P_{sat} is the saturation pressure. As discussed earlier, C is directly related to the bubble nucleation and typically ranges from 0.2 to 0.3 for low depressuring rates. The magnitude of C determines the pressure undershoot at the choke point/exit due to the superheating of the liquid. C tends to 0 as the starting initial temperature approaches the critical temperature. Depending on the flow conditions, C can also depend on the length of pipe and initial vapor quality and should be established for the correct rate of pressure drop using Equation 7. For pipe flow, it has long been recognized that a pipe flow length of approximately 4 inches [16] is required for equilibrium two-phase flow to develop, $C = 0$. Non-equilibrium is most important for nozzle flow (PRV flow) and for short piping. The above equation can be corrected to reflect the important of friction on nozzle flow:

$$G = \sqrt{\frac{2\rho_o\left[P_o - (1-C)\,P_{\text{sat}}\right]}{1+4f\frac{l}{d}}} \tag{12}$$

where f is the Fanning friction factor and l/d is the length to diameter ratio of the nozzle and/or piping. More information about non-equilibrium flow and retrograde and phase change flow can be found in references [17, 18].

Nucleation limits are shown in Figure 5 for a 50/50 by weight mixture of ethane-butane as calculated by SuperChems Expert. In order for the scaling to work properly for mixtures, the scaling vapor properties must be obtained at the vapor mole fractions in equilibrium with the liquid mole fractions at the bubble point.

Figure 5: The superheat and nucleation limits for a 50/50 by weight mixture of ethane-butane



4 Rapid Phase Transitions (RPT)

A fluid can also be made metastable by rapid heating or rapid cooling at constant pressure as shown in Figure 1 by going from point C to D (rapid cooling) or by going from point D to C (rapid heating). In the case of extremely rapid cooling, a vapor will have to condense and partially or fully convert to liquid when it crosses the vapor thermodynamic stability limit. Under rapid heating, a liquid will have to generate vapor when it crosses the thermodynamic stability limit for liquid as is usually experienced in LNG rapid phase transitions [8, 19] or steam explosions. Figure 2 from reference [8] illustrates how an LNG mixture can be rapidly heated to the superheat limit at ambient pressure causing a rapid phase transition.

A rapid phase transition is the very rapid (near spontaneous) generation of vapor as the cold LNG is vaporized from heat gained from the underlying spill surface or from large volumes of water contacting LNG in a storage tank. Because the vapor is evolved very rapidly, localized overpressure is created. This is also sometimes described as a physical explosion.

Following a release of liquefied natural gas (LNG) from a ship or storage tank a liquid pool forms and spreads on the surrounding spill surface. Rapid phase transitions have been shown to occur during or following an LNG spill. The hazard potential of rapid phase transitions can be severe, but is highly localized within or in the immediate vicinity of the spill area.

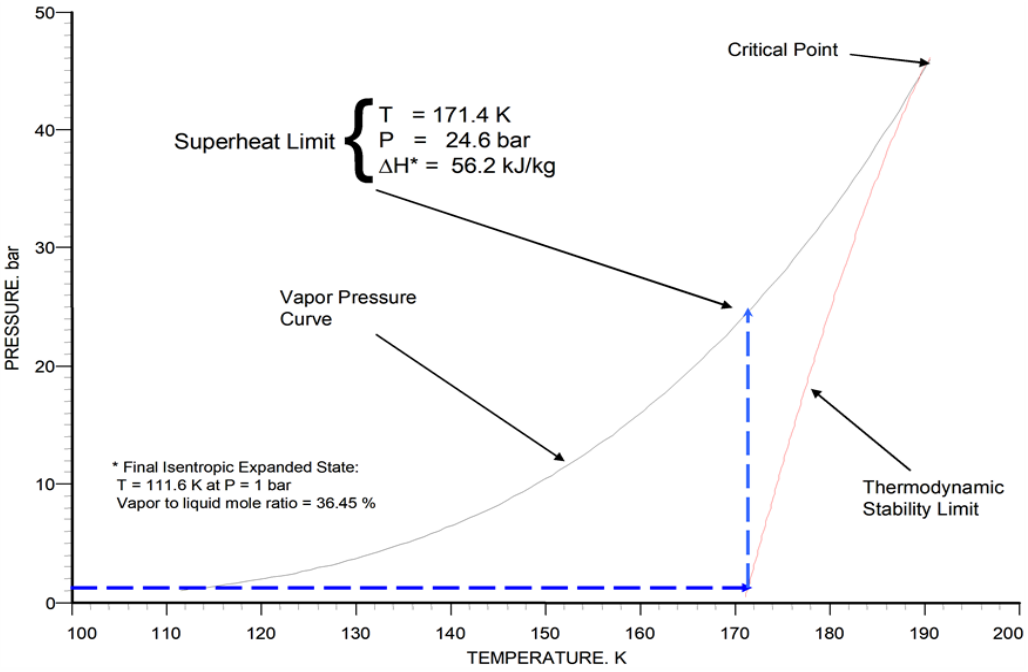
5 Prediction of RPT Hazard Potential

Rapid phase transitions are also referred to as physical explosions. This type of explosion does not involve combustion or a chemical reaction to create mechanical explosion energy. Instead, mechanical or explosion energy is created from the rapid expansion of a high pressure metastable fluid to ambient pressure. As illustrated in Figure 1, a liquid can be made thermodynamically unstable (metastable) by rapidly changing its temperature and pressure such that it cannot exist at those conditions in its initial state (all liquid).

Even during very rapid heating or very rapid depressuring, all fluids must change phase ultimately. These phase change limits (also called the thermodynamic stability limits) can be determined accurately using an equation of state. An LNG rapid phase transition can be explained using the thermodynamic stability limit (also called the superheat limit).

We illustrate the rapid heating process of LNG leading to a rapid phase transition on a phase diagram. LNG consists predominantly of methane. Certain LNG compositions will contain higher fractions of ethane and some propane and as a result their phase diagram is different from that of pure methane.

Figure 6: The superheat limit for pure methane [8]



First lets look at how the superheat limit is reached for pure liquid methane. This is illustrated graphically in Figure 6.

Follow the dashed blue line at the bottom of Figure 6. Pure liquid methane boils at 111.6 K (-258.8 °F) at ambient pressure. Rapid heating at ambient pressure causes methane to reach the thermodynamic stability limit of 171.4 K (-151.15 °F). Once heated to that temperature, methane becomes a superheated liquid, i.e. a saturated liquid with a vapor pressure of 24.6 bars. Methane reaches the superheated state and has to give up its superheat by expanding because the ambient pressure is 1 bar. If we assume that the expansion process is reversible/isentropic (we can bring methane back to its superheated state by adding back the same amount of energy it lost when it expanded) methane will expand to 1 bar and exert 56.2 kJ/kg in mechanical work (physical or pressure-volume) or energy (on the surroundings) that can be used to create overpressure, i.e. explosion energy.

In reality, the expansion process is not reversible and its efficiency at best is around 50 % as established by actual testing. This is because the expansion process loses energy as it creates turbulence and as the liquid flashes to vapor. As a result, the maximum possible rapid phase pressure that methane can reach is 24.6 bars and its mechanical explosion energy is 28.1 kJ/kg. This is equivalent to burning 0.56 grams of methane vapor. In other words, on per unit mass basis, the methane combustion process produces 1,780 times more energy than a rapid phase transition. This is why, historically, rapid phase transition overpressure estimates were excluded from LNG risk assessments and considered to be negligible and localized.

Now lets repeat the same process for an LNG mixture. An LNG mixture containing high fractions of ethane and propane is more likely to undergo a rapid phase transition than pure methane. This is observed in real LNG spills and can also be proven theoretically as illustrated later in this document.

Instead of a vapor pressure curve, an LNG mixture has a phase envelope consisting of a bubble point curve and a dew point curve as illustrated in Figure 2. Follow the dashed blue line at the bottom of Figure 2. This LNG mixture boils at 115.8 K at ambient pressure. Rapid heating at ambient pressure causes the LNG mixture to reach the thermodynamic stability limit of 191.4 K. Once heated to that temperature the LNG mixture becomes a superheated liquid, i.e. a saturated liquid with a bubble point pressure of 36.0 bars. The LNG mixture reaches the superheated state and has to give up its superheat by expanding because the ambient pressure is 1 bar. If we assume that the expansion process is reversible/isentropic, the LNG mixture will expand to 1 bar and exerts 75.5 kJ/kg in mechanical work or energy that can be used to create overpressure, i.e. explosion energy.

As mentioned earlier, the expansion process is not reversible and its efficiency at best is around 50 %. As a result, the maximum possible rapid phase pressure that the LNG mixture can reach is 36.0 bars and its mechanical explosion energy is 37.75 kJ/kg. An LNG mixture rapid phase transition produces 1,325 times less overpressure energy per unit mass than the combustion process.

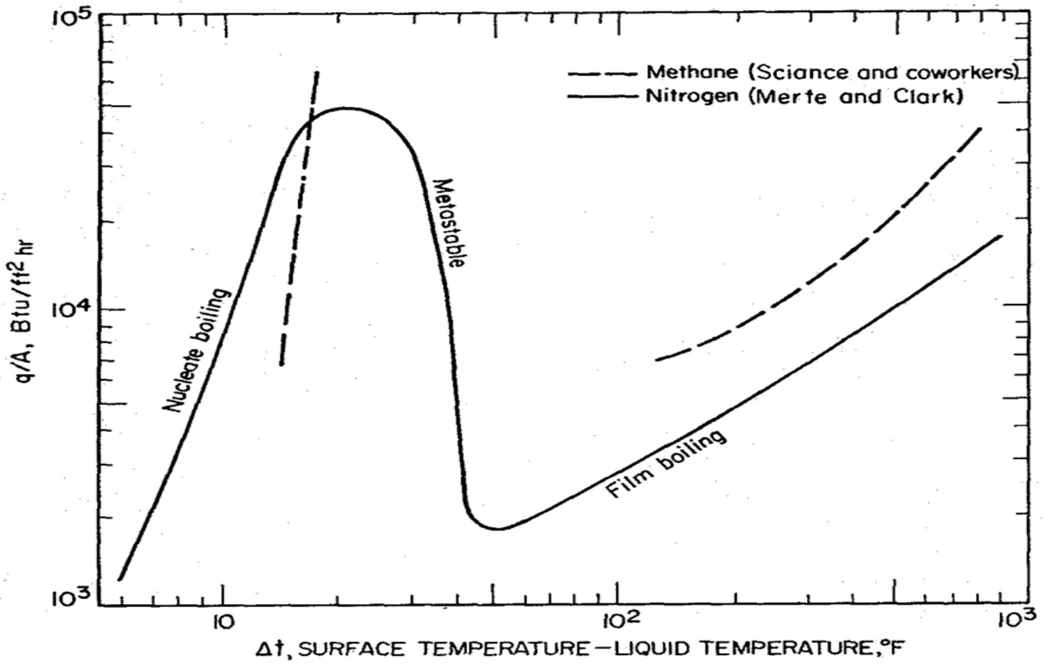
The explosion energy predicted by the superheat limit at 37.75 kJ/kg or (20.7 kJ/liter) is consistent with recent spill data measured by Shell [20] at 5.6 kJ/liter. Until a more detailed model is developed to better represent the rapid phase transition process, we recommend the use of the superheat limit explosion yield of 20.7 kJ/liter. This number can easily be established for other LNG compositions of interest. Although not recommended by the authors, the explosion yield of 20.7 kJ/liter can be used with a simple TNT equivalency method to predict overpressure contours from a rapid phase transition with a specified amount of LNG. Note that TNT equivalence will over predict near field overpressure values and is therefore considered to be a conservative method.

Even if we were to consider the physically impossible, i.e., the entire contents of one LNG storage tank (say 25,000 m3) participated in a single RPT at the same time (only a small portion of the liquid spilled on water that is in intimate contact with the spill surface has been shown to participate in an RPT in large scale field trials), the overpressure hazard radius to 1.0 psi would be estimated at 0.82 miles from the center of the RPT. The RPT hazard radius is well within distances of concern of LNG flammable dispersion to 1/2 LFL for releases from hole sizes ranging from 1 to 5 meters.

6 Predicting RPTs from LNG Spills

Existing modeling methods fall short from being able to identify with accuracy what fraction of an LNG spill will participate in a rapid phase transition. However, there are existing advanced modeling techniques that can tell us if a rapid phase transition will occur and at what approximate time during the spill it will occur.

Figure 7: Boiling regimes for LNG and nitrogen [8]



Before discussing RPT modeling, one needs to understand the different boiling regimes based on the temperature difference between the heating surface and the liquid. Figure 7 illustrates the various boiling regimes for LNG and nitrogen spills.

The process of forming vapor in all liquids (also referred to as flashing) usually involves what is called nucleation sites. For example, in a process vessel, these nucleation sites can be small imperfections on the vessel inner surface or tiny colloidal suspensions of dirt or dissolved gas in the liquid. Nucleation is a process where vapor bubbles start to form in these surface imperfections when a liquid is heated to a boiling state. The nucleation process requires mass and heat transfer in order to produce vapor. If heating occurs at an extremely rapid rate, these nucleation sites are rendered inactive as they do not have enough time to complete the mass and energy transfer/exchange required to generate the vapor bubbles, i.e. nucleate. The same effect can be produced by dropping the pressure of a saturated fluid very fast as shown in Figure 1.

When LNG is spilled on land or water, LNG is initially very cold (110 K or -261.67 °F). The spill surface (land or water) is initially very hot compared to the temperature of LNG. Even cold ocean water is typically around 60 °F or 289 K. The initial difference between the LNG and the water surface is 289-110 or 179 K (322 °F). This high temperature difference causes the LNG to start boiling. Because the difference in temperature is so high initially, a vapor film is formed at the contact point between the LNG and the underlying spill surface (see Figure 7).

This vapor film will persist until the spill surface cools enough and/or until the LNG bubble point temperature gets high enough as methane is preferentially depleted from the liquid LNG spill. As long as the vapor film exists between the LNG and the spill surface, heat transfer is greatly reduced (vapor layer acts as an insulator also). When the difference in temperature between the LNG and the spill surface gets smaller, the vapor film is destroyed and a different (faster) heat transfer mode begins (see Figure 7). The rate of heat exchange between the cold LNG and the warmer spill surface is now orders of magnitude larger than it was with the vapor film intact. As a result, the LNG is heated very rapidly (almost instantaneously to the superheat limit) and a rapid phase transition occurs.

7 Conclusions

This brief technical note provided an overview of why and how non-equilibrium flow and rapid phase transitions occur. The rate of pressure drop (or the rate of heating and/or cooling) is critically important for the creation of a fluid metastable state leading to non-equilibrium flow and/or a rapid phase transition.

How can we help?

In addition to our deep experience in process safety management (PSM) and the conduct of large-scale site wide relief systems evaluations by both static and dynamic methods, we understand the many non-technical and subtle aspects of regulatory compliance and legal requirements. When you work with ioMosaic you have a trusted ISO certified partner that you can rely on for assistance and support with the lifecycle costs of relief systems to achieve optimal risk reduction and PSM compliance that you can evergreen. We invite you to connect the dots with ioMosaic.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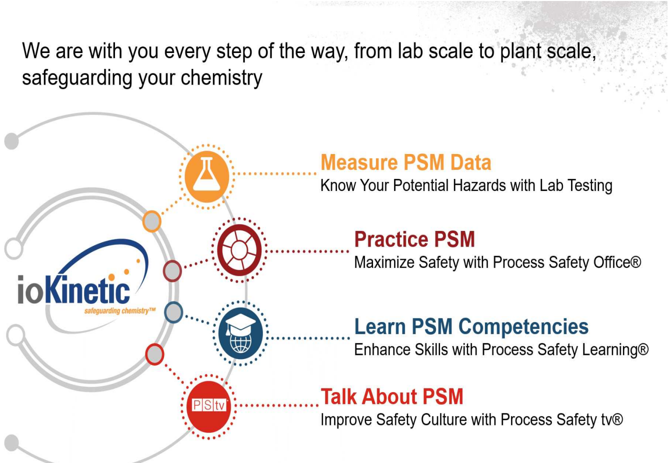
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About the Author

Dr. Melhem is an internationally known pressure relief and flare systems, chemical reaction systems, process safety, and risk analysis expert. In this regard he has provided consulting, design services, expert testimony, incident investigation, and incident reconstruction for a large number of clients. Since 1988, he has conducted and participated in numerous studies focused on the risks associated with process industries fixed facilities, facility siting, business interruption, and transportation.

Prior to founding ioMosaic Corporation, Dr. Melhem was president of Pyxsys Corporation; a technology subsidiary of Arthur D. Little Inc. Prior to Pyxsys and during his twelve years tenure at Arthur D. Little, Dr. Melhem was a vice president of Arthur D. Little and managing director of its Global Safety and Risk Management Practice and Process Safety and Reaction Engineering Laboratories.

Dr. Melhem holds a Ph.D. and an M.S. in Chemical Engineering, as well as a B.S. in Chemical Engineering with a minor in Industrial Engineering, all from Northeastern University. In addition, he has completed executive training in the areas of Finance and Strategic Sales Management at the Harvard Business School. Dr. Melhem is a Fellow of the American Institute of Chemical Engineers (AIChE) and Vice Chair of the AIChE Design Institute for Emergency Relief Systems (DiERS).

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Contact Us

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About ioMosaic Corporation

Through innovation and dedication to continual improvement, ioMosaic has become a leading provider of integrated process safety and risk management solutions. ioMosaic has expertise in a wide variety of areas, including pressure relief systems design, process safety management, expert litigation support, laboratory services, training and software development.

As a certified ISO 9001:2015 Quality Management System (QMS) company, ioMosaic offers integrated process safety and risk management services to help you manage and reduce episodic risk. Because when safety, efficiency, and compliance are improved, you can sleep better at night. Our extensive expertise allows us the flexibility, resources, and capabilities to determine what you need to reduce and manage episodic risk, maintain compliance, and prevent injuries and catastrophic incidents.

Our mission is to help you protect your people, plant, stakeholder value, and our planet.

Consulting Services

- Asset Integrity
- Auditing and Due Diligence
- Combustible Dust Hazard Analysis and Testing
- Facility Siting
- Fault Tree/SIL/SIS Analysis
- Fire and Explosion Dynamics
- Incident Investigation, Litigation Support and Expert Testimony
- Liquefied Natural Gas Safety
- Pipeline Safety
- Process Engineering Design and Support
- Process Hazards Analysis (PHA)
- Process Safety Management (PSM)
- Reactive Chemicals Evaluation and Testing
- Relief and Flare Systems Design and Evaluations
- Risk Management Program Development
- Quantitative Risk Assessments (QRA)
- Software Solutions
- Structural Dynamics
- Training

Laboratory Testing Services (ISO accredited)

- Chemical Reactivity
- Combustible Dust
- Battery Safety
- Specialized Testing