

GAS FLOW MEASUREMENT & THE IMPACT OF CONTAMINANTS IN THE GAS STREAM

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Abstract.

Significant advances have been made over the past couple of decades with respect to flow metering technology applicable to natural gas transmission measurement. Current flow meter technology is characterised by high reliability, excellent turndown and very low measurement uncertainty. However, the environment in which these flow meters are being placed is becoming more demanding on the meter's operation and performance. This is because of the trend to operate pipelines at higher pressures and transport varying gas composition from multiple pipeline system inlet points. These conditions are conducive to the formation of unwanted contaminants within the gas stream.

Introduction

Flow measurement technology applicable to the measurement of high-pressure natural gas has seen very significant advances over the past few decades providing considerable benefits to the flow measurement process. This is particularly exemplified with the rapid development of both the ultrasonic and Coriolis meters. However, the introduction of new technology and operating criteria can come at a cost – being the presence of unexpected contaminants in the gas stream.



Figure 1. Elemental Sulphur Deposits Downstream of a Turbine Meter.

In natural gas pipeline network operations the most commonly reported contamination related problem is that of the so-called black powder. This black powder is made up of various forms of iron sulphide (FeS), iron oxide (FeO), hydrocarbons and asphalt components. More recently, a new form of contamination has been catching operators' attention

that is mostly sulphur related; it is labelled as the elemental sulphur formation and deposition process.

The elemental sulphur (S₈) deposition mechanism has similarity with the black powder occurrence; however it is difficult to predict due to the lack of uniformity of distribution within a pipeline system and seems to arise even though elemental sulphur as such is not identified in the initial composition of the gas to be transported.

The elemental sulphur formation / deposition process within high-pressure natural gas systems was reported on during 1990 [1] and 1993 [2]. This observation was directly attributed to the decision by TOTAL and STATOIL to collectively develop the Kårstø Metering and Technology Laboratory (K-Lab) in response to concerns as to the accuracy of the custody transfer metering system for the gas coming from the North Sea FRIGG field, and the requirement to independently assess other flow measurement technologies. This FRIGG gas field metering system, measuring one million standard cubic metres of gas per day, was based on orifice plates and located at the Saint Fergus Terminal in Scotland.

The orifice plate meter was not regarded as the most favourable to gas sellers since its accuracy is impacted negatively by a large number of factors, such as:

- Calibration,
- Plate geometry,
- Obstruction by dirt and deposit,
- Presence of liquid,
- Rangeability, and
- Repeatability

However, the presence of elemental sulphur in well bores and production facilities is not new, especially for sour gas wells. One of the imperative problems in production of sour natural gases is precipitation of the sulphur in the formation, well bores, and production facilities especially at high temperatures, pressures, and at high hydrogen sulphide concentrations [3].

The presence of elemental sulphur deposits, together with hydrocarbon liquids is now being more commonly reported by coal-seam gas methane pipeline operators and facilities and gas turbine plant operators having fuel gas sourced directly from high-pressure natural gas transmission pipeline systems. Prior research [4] found that formation of elemental sulphur within transmission pipeline systems can occur through five main processes taking place in order, that of:

- Formation of H₂S from sulphur bearing components,

- Chemical reactions leading to elemental sulphur in the gas phase,
- Desublimation of the sulphur to the solid phase, or
- Condensation formation, and
- Throughout the facilities in the course of the transportation processes, compression, metering and regulation stations, etc.

Desublimation is the direct transformation of vapour to solid phase. As the majority of deposits are found at and immediately downstream of pressure reduction stages, obviously the main elemental sulphur formation process has to be that of desublimation. The potential pipeline chemical reactions can have a variety of contributing sources; however, the majority of these reactions will evolve over time and proceed anywhere in the pipeline system where favourable conditions exist. The condensate mechanism would most likely only occur due to abnormal circumstances such as the accidental injection of liquid hydrocarbons and / or dehydration fluids into the gas stream or favourable retrograde condensation conditions existing.

Operators of transmission pipeline systems become perplexed, and often in conflict with gas producers, over the fact that although the gas at the transfer point is seemingly within specification as regards to sulphur, elemental sulphur is largely encountered several hundred kilometres further downstream. It is precisely because some of the sulphur bearing components not identified / eliminated at the transfer point of the gas producer's plant, have then appeared as elemental sulphur deposits downstream within the pipeline transportation system. To add to the complexity of this problem, it has been found through this research work [5] that the elemental sulphur deposits can be preferentially site selective.

Today natural gas transmission pipelines are operating at higher pressures with ANSI Class 900 systems now becoming more common. Market forces pushing for the increased deregulation of pipeline networks is resulting in multiple gas supply entry points with greater variability in the gas composition transported. Both of these operational features have the potential to incur retrograde condensation and hence liquid drop-out. For example smaller gas producers may now have access to the market; however by necessity the smaller producer may use glycol as the gas dehydration process, with the subsequent potential carry-over in the gas stream to the transmission network.

Elemental Sulphur Formation & Deposition Processes.

The following points [6] provide a simplified overview of the 'elemental sulphur' formation and deposition process for a process plant or pipeline pressure reduction facility that would have the necessary gas composition and operating conditions.

1. Sulphur vapour already in gas stream at sub parts per million (sub-ppm) levels. [Typical concentration around just a few to low tens of parts x 10⁻⁹]
2. The sulphur vapour becomes supersaturated due to the rapid cooling of the gas mixture rapidly flowing through the pressure control valve cage (mechanism), nozzle or like pressure restriction/control device.

3. The supersaturated sulphur vapour molecules form nuclei – this is the commencement of the very rapid nucleation process. The formed nuclei being new, minute particles.
4. Concurrent possibility of retrograde condensation occurring for some of the heavier hydrocarbon components in the gas stream. This is also due to the rapid cooling of the gas stream.
5. Other molecules (retrograde condensation components) are attracted to the sulphur particle surface through mechanism of condensation. An analogy to this condensation phase is the process of water in the atmosphere condensing around suitable air-borne nuclei. These nuclei could be a dust, combustion produce or salt particle, generally of a size of less than 1.0 μ
6. The resulting larger particles, which will have a very high velocity, will collide with other particles in the gas stream forming larger particles. This is the coagulation process.
7. There may be other deposits on the internal pipe-walls or fittings, or travelling within the gas stream. Again due to the high gas velocities and turbulence, there will be a high probability of collision with these other particles – this resulting in the agglomeration phase.

Figure 2 demonstrates the elemental sulphur deposition at a point of high-pressure reduction on a transmission pipeline. Note that the deposition material texture is relatively fine with a high concentration of elemental sulphur.

The conversion of the sulphur vapour to the particle phase during the homogeneous nucleation phase is influenced by pressure, temperature, flow and the supersaturation state of the sulphur vapour in suspension in the gas stream. The nucleation rate is the number of nuclei formed per unit volume and time.



Figure 2. Elemental Sulphur deposition.
(Composition predominantly S₈ and hydrocarbons)

A natural gas stream exiting a large gas process plant into a transmission pipeline system will most likely have passed through a coalescing filter and therefore be free of any liquid. However, as this gas stream is most likely exiting the plant at a temperature well above the pipeline ground temperature, cooling of the gas stream will occur in the pipeline until the gas becomes at, or near equilibrium with the ground temperature. There will also be a pressure drop due to this cooling together with the pipeline wall frictional effect on the gas stream. This means that there is the potential for some

retrograde condensation to occur, especially if the gas stream exiting the gas plant has small quantities of heavier hydrocarbons in the gaseous state. There is the potential for desublimation of any sulphur vapour to elemental sulphur. However, the potential formation of these liquid hydrocarbon droplets and minute elemental sulphur particles will not necessarily suddenly occur when the respective vapours are brought to their saturation pressure and temperature.

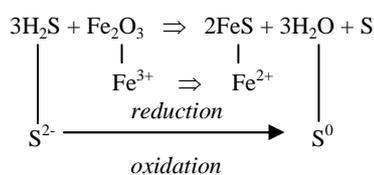
Research has shown [7] that a vapour will condense at its saturation point only in the presence of sufficient amounts of "foreign" condensation nuclei, such as particles, ions, and extended surfaces (heterogeneous nucleation). In the case of homogeneous nucleation, that is when the vapour is free of all impurities, condensation may not occur at the saturation point and the vapour can exist in a state of supersaturation. This means that the presence of existing particles in the gas stream may assist in the condensation process of the referenced vapours.

As heterogeneous nucleation requires less energy than homogeneous nucleation, it would be expected that heterogeneous nucleation will be more dominant within the pipeline system. This is borne out by the majority of elemental sulphur deposits being coagulated with other solid and liquid particle matter. Elemental sulphur deposits observed at affected pipeline sites tend to demonstrate finer particles are formed where there is a substantial pressure reduction. This can be explained by the fact that the greater the supercooling, the smaller will be the formed nuclei critical radius and thus the less energy required to form it.

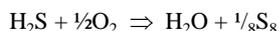
Other Potential Pipeline Sources of Elemental Sulphur

(i). Mill Scale (Rust).

Pipeline internal wall rust is a potential oxidant, per the following equation:



(ii). Introduction of Extraneous Air



Extraneous air may enter the pipeline system through a variety of means, such as:

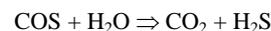
- Pipeline pigging operations,
- Maintenance activities,
- Recovery of compressor casing vent gas, and
- Compressor dry gas seals.

(iii) Other Reactions within the Pipeline System

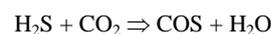
Conversion of carbonyl sulphide (COS) to hydrogen sulphide (H₂S), and hence an additional source for the formation of elemental sulphur.

Field studies conducted [4] have found that very low levels of H₂S can exist within a pipeline system immediately

downstream of a gas treatment facility, however at a point many kilometres downstream of the gas entry point a noticeable increase in the H₂S level can be observed. This increase is due to the conversion of carbonyl sulphide COS to H₂S in the presence of water vapour according to the following reaction:



Of particular interest is the potential for the reversal of this reaction. In the operation of particular molecular sieves used at large gas plants for gas dehydration, there is the ability for COS to form according to the reaction:



It has been stated [8] that the phenomenon has been identified as the simultaneous H₂S adsorption and rate-limited catalytic reaction of H₂S and CO₂ to form COS and water.

(iv). Microbial Reduction of Sulphate



Other Pipeline Contaminants and their Potential Source.

Within a natural gas stream there is the potential for a variety of vapour, liquid and solid contaminants to be present. The liquids can be represented by hydrocarbons, dehydration fluids such as glycols, inhibitors, and free water. Extensive inductively coupled plasma – mass spectrometer (ICP-MS) analysis [6] has demonstrated a vast number of elements and other matter such as grease, anti-seize compounds, asphaltenes, waxes and corrosion products. Table 1 provides a summary of elements found within elemental sulphur deposits from differing transmission pipeline systems.

Element	Sample A	Sample B	Sample C	Sample D	Sample E
Fe	7235	10471	26377	117	509
Li	0.97	2.2	10	0.1	0.41
Mg	24	47	7896	16.7	13
Al	68	465	11997	37.9	97
Si	33	128	1133	173	33
Ca	14	73	7681	10.4	12
Ti	12.8	8.6	3700	7.2	8.6
Cr	499	21.4	379	7.1	34.1
Mn	78	66	1700	12.3	11.8
Ni	120	14.6	200	17.8	32.6
Cu	92	25.5	1100	4.6	47.8
Zn	25.9	100	8099	26.9	57
Cd	0.04	0.11	1.7	0.1	1.2
Ba	1.6	5.2	170	1.3	9.9
Pb	1.71	3	1600	4.6	11.9
S	10267	5798	61091	6239	4484
Totals	18473	17229	133135	6676	5363

Table 1. ICP-MS Detected Elements (ppm) in Elemental Sulphur Deposits.

The majority of the contamination samples have been taken from pressure reduction and metering facilities. As such there is expected to be a variety of differing metals, greases, oils and derivative products present together with varying gas velocities. It will be noted that iron (Fe) is by far the second most common element in the analysed elemental sulphur samples. For the sample B case, Fe was the dominant element and gave the sample a dark olive-black colour.

Pipeline ‘Black Powder’.

From the results of an initial industry survey, it was determined that Black Powder is the least understood and most prominent contamination problem in pipelines and their compression equipment [9]. It appears that this statement still holds true today. This author also states that once sulphur enters the pipeline at any point, conversion to iron sulphide is prompt and that the material may be wet and have a tar-like appearance, or dry and be a very fine powder, sometimes like smoke.

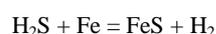
Black powder describes a material that collects in natural gas pipelines and creates wear and reduced efficiency in compressors, clogs instrumentation and valves and leads to flow losses in long pipelines. Chemical analysis of the material has revealed that it is any of several forms of iron sulphide or iron oxide. Further, it may be mechanically mixed or chemically combined with any number of contaminants such as water, liquid hydrocarbons, salts, chlorides, sand, or dirt. Iron sulphide has both a chemical and a microbial source in pipelines. Microbiologically influenced corrosion (MIC) is a form of corrosion that is initiated by microbes that find a habitable environment in natural gas piping. MIC is pitting of the pipe wall as a result of the activities of microbial communities in areas that provide their required habitat.

The microbes that cause pipe corrosion are basically two families:

- Sulphate reducing bacteria (SRB), and
- Acid producing bacteria (APB).

Although the SRB directly produces H₂S instead of FeS, the conversion of the first to the second is direct and prompt at the site of the microbial activity where iron is present. Iron sulphide is not easily filtered out of the flow stream, and is pyrophoric under some conditions. Some pipelines have black powder problems and others do not – it can be a major source of unwanted pipeline particles.

Black powder, at least the iron sulphide and oxides, are known to be created inside natural gas and other similar wells and pipelines. The components and conditions necessary to create the materials can be found at some point in many pipelines. Iron sulphide, and apparently many of its variations, can be quickly and efficiently created in a gas pipeline from the chemicals naturally available in the system. Hydrogen sulphide (H₂S) easily reacts with the iron in piping to form iron sulphide as in the formula:



In addition to hydrogen sulphide, sulphur can also react to form iron sulphides. Sulphur compounds are present in relative abundance in gas and oil wells. Older fields might have sulphate reducing bacteria that have grown in the formation.

Iron sulphide, either attached to the wall of a pipe, or collected in the bottom of a pipe, has the negative effects of

increasing roughness, decreasing flow area, and increasing pressure drop. Furthermore, over time its formation thins the pipe wall and reduces the margin of safety against compromise of the pressure boundary.

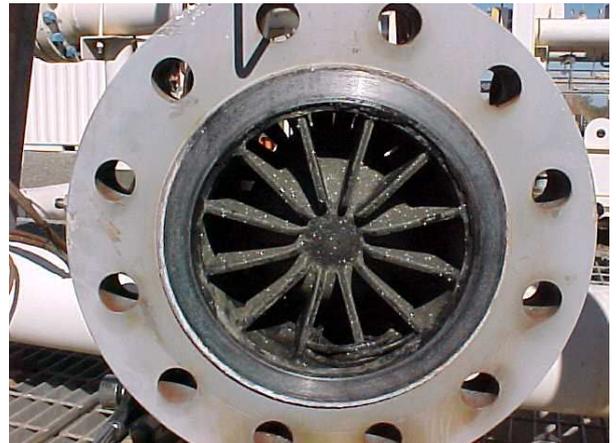


Figure 3. Black Powder at Inlet to a Turbine Meter.

The black powder deposits, which can contain particles of less than one micron, can be readily transported through a transmission pipeline system.

Field observations associated with this research work have shown that fine black powder can be found immediately downstream of in-line correctly functioning filters designed to remove particles of just a few microns in size.

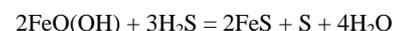
The ‘Black- Powder’, Pipeline Rust Contribution.

The corrosion of steel usually produces goethite, FeO(OH), which is more commonly referred to as rust. Common pipeline rust is formed according to the following equation:



Therefore, even before first gas travels down a pipeline system there is a potential source of oxygen bonded to the pipe walls. As a source of oxygen, any chemical reactions with the goethite and sulphur compounds within the pipeline are of interest.

It has been demonstrated [10] that goethite will react with H₂S, per the following equation:



The reaction of FeO(OH) and H₂S has to be considered as yet another potential source of sulphur within the transmission pipeline system.

Steps to Minimize Threat of Particle Formation / Deposition.

In pipelines where the operator suspect, or have identified sulphur and / or black powder as an operational problem, the following recommendations could minimise the contamination formation / deposition threat for both elemental sulphur and black powder.

- Reduce the potential for retrograde condensation occurring at a pressure reduction facility.

- Minimize the entry and transmission of liquids and solid particles in the gas stream.
- Minimize the sources of water (moisture) and oxygen that can enter the gas stream.
- Ensure carry-over from glycol processing plants is kept to a minimum.
- At commissioning of new pipelines ensure dewatering processes are complete and thorough. Do not permit 'puddling' of odorants or other like additives. Make sure pipe work is free of particle matter.
- Review type of material used in molecular sieve beds does not favour the conversion of H₂S to COS.
- Minimize H₂S levels. 4.0 ppm is seen as a realistic target taking into account the economic / technical requirements for gas producers / processors and the transmission pipeline operator. However, a lower level is more preferable.
- Minimize site conditions suitable for the colonization and maintenance of SRB.
- Care needs to be exercised in the interpretation of on-line and wellhead analysis results. This may be due to sampling techniques, absorption of components by the sampling apparatus (tubing, valving and container), or due to the variations between sampling and analysis pressures (and temperatures).

Additional recommendations to minimise elemental sulphur formation / deposition are:

- When a large pressure reduction is required use a dual stage pressure cut. Due to the potential for temperature recovery between stages, a much lower temperature gradient at each stage, and lower retrograde condensation rate (if applicable) will decrease the overall nucleation, condensation and coagulation rates.
- Where possible, avoid the application of labyrinth type pressure control / regulation valves.
- Try to minimize large temperature excursions along pipeline route.
- Maintain flowing gas temperature as high as practically possible. This will not only help maintain the sulphur gaseous form and hence in solution, but will also assist in the minimization of retrograde condensation.
- Take care when applying commercial models to gas processing, transportation conditions. Ensure all gas component parameters are within the specified design criteria of the model.
- Minimize pipeline pigging operations and maintain pig speeds at realistic controlled levels. Pigging operations are a cause of oxygen ingress. Such operations also have the potential to transport a slug of liquid down the pipeline to pressure reduction facilities. High speed pigging operations, due to the result of the Joule-Thomson effect, can actually result in heavier hydrocarbons condensing.

Impact on Flow Meter Performance.

Unwanted contaminants, both liquids and solids, can come at a cost to the pipeline operator – varying from nuisance value to causing complete failure of equipment and stoppage of the gas flow. The impact of the presence of elemental sulphur not

only translates to the potential for gas supply interruption, damage to equipment and general reliability issues, but also very significant and costly demands on system maintenance.

Some of the more common locations for elemental sulphur deposits in pipelines and associated systems are:

- Deposition on internals of flow meters:
Impact; Loss of gas measurement accuracy. Erratic flow readings due to flaking and shedding of deposits.
- Deposits around pressure control valves:
Impact; Adverse impact on stem movement. Potential for plugging of valve orifice.
- Coating on thermowells, pipe walls and flow conditioning elements:
Impact; General degradation of performance. Potential to stop gas flow for case of flow conditioner element.
- Deposition in the throat of critical flow nozzles:
Impact; Nozzle can no longer be used for intended purpose.
- Coating on in-line filters and on filter housing internals:
Impact; Increase in differential pressure across filter elements with potential for complete plugging, and/or filter collapse.
- Coating of sour gas exchangers at natural gas treatment plants:
Impact; Due to plugging, plant shut down required.
- Coating on in-line filters and on filter housing internals:
Impact; Increase in differential pressure across filter elements with potential for complete plugging, and/or filter collapse.
- Coating of sour gas exchangers at natural gas treatment plants:
Impact; Due to plugging, plant shut down required.
- Downstream of gas turbine control valves:
Impact; Valves starting to plug with output reduced. Periodic shedding of the uncontrolled sulphur deposits into the gas fuel nozzles. This has potential to cause flashback and flame holding of the secondary and tertiary pre-mixing system resulting in physical damage to equipment.

Flow Meters Observations.

Investigations into the contamination build-up on flow meters has been limited to turbine, ultrasonic and Coriolis type meters, although contamination has also been observed on a small number of orifice plate and vortex meters. Turbine, ultrasonic and Coriolis meters are now generally the meters of choice for high-pressure natural gas and coal-seam methane gas transmission pipelines. These meters may have to respond to the resulting gas flow distortions due to particle deposition processes depicted in Figures 1 to 5 inclusive. The following general discussion is limited to these three meter types.

Turbine Meters

As the discovery of elemental sulphur formation and deposition in natural gas transmission pipelines was first identified in the late 1980's, turbine meters were replacing orifice plate meters for custody transfer applications. This has resulted in reasonable in-depth analysis of the impact of such contamination on the turbine meter. In the majority of case

studies, turbine meters with reasonably uniform and extensive deposition have been able to be cleaned and returned to service without any repair or recalibrations performed.



Figure 4. Elemental Sulphur Deposits on Flow Straightening Tubes

The deposits on the turbine meter internals with reasonably uniform, non-conglomerate particle deposits, flow deviations of up to +2 % have been observed by placing the effected meters in series prove with a field based ‘master’ meter. For such cases the particles are generally less than 20 μm . For larger particles, and in particular for non-uniform deposits on the meter internals, the flow deviations can be erratic with under-registration of flow being possible. The modern turbine meter has demonstrated that it is robust to small particle contaminants in the gas stream and can be very repeatable over a long period of operation.

Ultrasonic Meters.

The diagnostic capability and design of the modern ultrasonic meter means that it can detect swirl, jetting and contamination. These capabilities, coupled with the ability to perform remote diagnostics on the meter, make the multi-path ultrasonic meter an attractive gas flow meter. However, as with all technology, development and knowledge about these meters continues, resulting in changes and revisions regarding the operation of these meters. This can be reflected in the relevant industry technical guide to the application of these meters, namely AGA-9 [11] and [12]. With respect to the meter internal surface there are differences in the recommendations made between the two editions.

The AGA-9 (1998) edition [11] states with respect to the meter internal surface that “the internal surface of the UM should be kept clean of any deposits due to condensates or traces of oil mixed with mill-scale, dirt or sand, which may affect the meter’s cross-sectional area. The UM’s operation depends on a known cross-sectional area to convert mean gas velocity to a flow rate. If a layer of deposits accumulates inside the UM, the cross-sectional area will be reduced, causing a corresponding increase in gas velocity and a positive measurement error”. Examples of the measurement errors that can be introduced are given in Table 2.

It is to be noted that the above information has been deleted from the current edition of AGA-9 [12], with a recommendation now given that the maximum surface roughness of the meters bore be no greater than 250 μinch . However it is still acknowledged that a change in the meter’s cross-sectional area will result in a fluid velocity increase and

a positive measurement error. However for any contamination build-up on the transducer faces, the advice has been modified; acknowledging that a measurement error will not necessarily occur in all contamination deposition situations.

Meter ID (mm)	Deposit layer on meter bore (mm)	Percentage error
152.4 (6 inches)	0.203 (0.008 inches)	+ 0.53 %
508 (20 inches)	0.203 (0.008 inches)	+ 0.16 %

Table 2. Meter Error due to Uniform Deposits on Meter Bore

[Adapted from [11]]

Although there has been extensive research into the design and operation of ultrasonic meters, including the impact of contaminants, there does not appear to be any published data on how these meters react to the total contamination issues presented by the presence of elemental sulphur or other contaminants in the gas stream. As already referenced, the elemental sulphur deposits can coat the pipe bore, meter bore, upstream fixtures such as flow straighteners and flow conditioners and downstream fixtures such as thermowell housings. To add to the complexity, the coatings appear to vary over time with flaking of the deposits regularly occurring. This flaking can result in discrete, prominent deposits along the metering and pressure regulating facility. Figures 1 and 4 demonstrate this elemental sulphur deposit flaking phenomenon.

Coriolis Meter.

Coriolis meters are fast gaining acceptance as a high-pressure gas meter. Although a lesser number of Coriolis meters (than turbine and ultrasonic meters) have been installed at transmission pipeline facilities known to be impacted by elemental sulphur deposition, there is little evidence that they are directly impacted by the presence of contaminants in the gas stream. This situation is probably due to the high gas velocity through these meters, however direct plugging could be a possibility. It is possibly a little early to determine if there could be any long term mechanical impact on these meters by particle matter in the gas stream.



Figure 5. Upstream View to Pressure Regulator
[Note clear indication of liquid (hydrocarbon) flow]

Conclusion.

As the boundaries of flow measurement continued to be extended with regard to the introduction of new technology, increased accuracy and operating parameters, there is a possibility that these benefits may come at a cost – that of the formation and deposition of contaminants due to desublimation and/or chemical reactions. With gas transmission pipeline systems now tending to have multiple gas entry points through increased deregulation, the potential to have greater variations in gas composition transported increases. This could also contribute to an increase in occurrence of retrograde condensation.

The entry of liquids into a gas transmission pipeline system, together with other particles and contributing elements, such as hydrogen sulphide and oxygen to the formation of elemental sulphur and black dust deposits, need to be tightly controlled. This also applies to the control of conditions within the pipeline system that could be conducive to particle and/or liquid formation. Good engineering design and pipeline operational modifications have demonstrated that these contamination issues can be minimised. There are many benefits to be realised by applying the current modern flow measurement technologies – however the pipeline operator must be prepared to make engineering design and operational changes to the pipeline system to minimize the potential contamination threats and to protect the end users gas usage process.

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