

Characterization of Emissions from Diffusion Flare Systems

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ABSTRACT

Emissions from flares typical of those found at oil-field battery sites in Alberta, Canada, were investigated to determine the degree to which the flared gases were burned and to characterize the products of combustion in the emissions. The study consisted of laboratory, pilot-scale, and field-scale investigations. Combustion of all hydrocarbon fuels in both laboratory and pilot-scale tests produced a complex variety of hydrocarbon products within the flame, primarily by pyrolytic reactions. Acetylene, ethylene, benzene, styrene, ethynyl benzene, and naphthalene were some of the major constituents produced by conversion of more than 10% of the methane within the flames. The majority of the hydrocarbons produced within the flames of pure gas fuels were effectively destroyed in the outer combustion zone, resulting in combustion efficiencies greater than 98% as measured in the emissions.

The addition of liquid hydrocarbon fuels or condensates to pure gas streams had the largest effect on impairing the ability of the resulting flame to destroy the pyrolytically produced hydrocarbons, as well as the original hydrocarbon fuels directed to the flare. Crosswinds

were also found to reduce the combustion efficiency (CE) of the co-flowing gas/condensate flames by causing more unburned fuel and the pyrolytically produced hydrocarbons to escape into the emissions.

Flaring of solution gas at oil-field battery sites was found to burn with an efficiency of 62–82%, depending on either how much fuel was directed to flare or how much liquid hydrocarbon was in the knockout drum. Benzene, styrene, ethynyl benzene, ethynyl-methyl benzenes, toluene, xylenes, acenaphthalene, biphenyl, and fluorene were, in most cases, the most abundant compounds found in any of the emissions examined in the field flare testing. The emissions from sour solution gas flaring also contained reduced sulfur compounds and thiophenes.

INTRODUCTION

Flaring has long been used by industry as a means of disposing of waste hydrocarbon products from normal process operations and for venting of process gases during emergency or upset conditions. The combustion processes that occur within these flames are usually complex, and methods for more detailed examination of emissions from them have been limited, resulting in relatively little information about what is contained in the emissions.

Emissions from flaring are influenced by a variety of factors, including flare design, operating conditions, and composition of waste gases. Gas streams with low heating values cannot maintain a stable flame, thereby reducing overall efficiencies of combustion.¹ Gases with varying amounts of liquid hydrocarbons, carbon dioxide, nitrogen, and/or sulfur gases may not only burn with reduced combustion efficiencies, but also produce undesirable components in the emissions.²⁻⁴ Strong crosswinds can produce a significant reduction in the combustion efficiency (CE) of a flare by shedding and/or tearing some of the eddies from the flame that contain incomplete or partially combusted gases.^{5,6}

A literature review on flaring included the topics of detection, control, and analytical techniques of flare gases.⁷ Some preliminary work was also conducted on optimizing sampling systems^{8,9} and on possible effects caused

IMPLICATIONS

This study dealing with emissions from flare stacks used in the upstream oil and gas industry in Alberta concluded that the efficiency of this practice is not as high as originally thought when used to dispose of solution gas. A variety of compounds of concern were being emitted as a result of incomplete combustion. The study addressed the unburned source gases and products of the combustion process that could be attributable to public complaints of odors and health concerns. Subsequently, the Clean Air Alliance in Alberta has made recommendations to reduce the 1.7 billion m³/yr of flared solution gas produced in Alberta. The regulatory Energy and Utilities Board of Alberta responded by incorporating these recommendations into an industry guide that addresses the target levels for flare reduction, starting with a 15% reduction by the end of the year 2000 and a 25% reduction by the end of 2001. Further reductions of 40–50% are required by the end of 2003 with a final reduction of 70% by the end of 2007.

by crosswinds.^{5,6} Little or no research has been carried out on the effects of flaring caused by co-flowing liquid fuels.

There are currently more than 5300 active flares in Alberta, Canada, that burn an estimated $2340 \times 10^6 \text{ m}^3/\text{yr}$ by the upstream oil and gas industry annually.¹⁰ The estimates have some uncertainty because the flow of some waste gases is measured on an infrequent basis. The main contributors to the total flared gases include sweet and sour oil production batteries (with or without hydrogen sulfide) ($1700 \times 10^6 \text{ m}^3/\text{yr}$), followed by acid and sulfur recovery gas plants and gas gathering systems ($700 \times 10^6 \text{ m}^3/\text{yr}$). Most of the gases from sulfur recovery gas plants, however, are incinerated. There are a similar number of emergency or intermittent flares operational in Alberta.

Typical flares used at oil-field sites in Alberta comprise a simple pipe system. The top 2–3 m are usually constructed from stainless steel. The size of the stack and opening is primarily dictated by the necessity to accommodate maximum anticipated gas release caused by a process upset or emergency shutdown at each particular location. These flares are usually equipped with a shroud at the top to reduce blowout from high crosswinds, have an automated ignitor in case of flame blowout, and are rarely equipped with a nozzle because of the low and fluctuating flow that is directed to them. They are designed to function as a diffusion flame—a combustion process in which the fuel and the air are not premixed. These flames are difficult to operate with the necessary proportions of fuel and oxygen for complete combustion.

There are a number of processes that occur in a diffusion flame that can chemically affect the nature of the emissions. These include precombustion, combustion, and post-flame processes. In the precombustion zone, reactions occur before the mixing of fuel with the air and are reducing in character, so products are expected to be pyrolytic in nature.¹¹ The reaction products of these pyrolytic processes are predicted to include unsaturated species such as olefins and acetylenes, and particulate nuclei resulting from polymerization or addition reactions between the unsaturated species. It has been demonstrated that a sequence of pyrolytic reactions can lead to the synthesis of relatively large and complicated polynuclear aromatic hydrocarbons.¹¹

The complexity of the combustion process and the hydrocarbon species that can be produced by these flames, together with the findings of previous studies, indicated that certain aspects of flaring in Alberta needed to be examined in more detail. The research program developed for this study that provided an in-depth characterization of the products of combustion from flaring included (1) the effects of liquid condensates and other liquid droplets during the flaring of gaseous fuels, (2) the effects of crosswinds on flaring, and (3) a reliable sampling and

analytical system to accommodate the measurements of these effects.

PROJECT DESIGN

The measure of a flare's effectiveness has traditionally been evaluated on the ability of the flare to destroy only the fuels directed to it, resulting in a measure of its destruction efficiency. The combustion products measured in these determinations include the burned species, carbon dioxide, and the unburned species, including carbon monoxide, carbon, and the hydrocarbons that were directed to the flare. The limitations of these measurements do not allow for any reactions that might take place in the burning process other than some oxidative ones. CE measurements, on the other hand, provide a more comprehensive evaluation of the overall effectiveness of a flare, depending on the range of hydrocarbons (HCs) and sulfur compounds (Scmpds) that are measured in the resulting emissions.

The CE of a flare can be measured on the basis of either global or local combustion. Global efficiency measurements require not only the composition and mass flow of the inlet fuel, but also the composition and mass flow of the products of combustion for the entire emissions. The use of this method to evaluate the efficiency of oil-field flares is prohibitive in most field operations, considering that most facilities are not instrumented to measure either flow or fuel composition and that it would be difficult to capture the entire emissions from a flare plume.

Local CE measurements require the determination of relative amounts of products of combustion at a single point that is representative of the average flare emissions. It requires complex analyses of the entire range of combustion products. In the present case, the local combustion approach was used and the resultant efficiency measurements were calculated using either a carbon or sulfur mass balance of all combustion products identified in the emissions. Carbon or sulfur equivalents were calculated from each compound detected and the CE determined as follows:

$$CE\% = \frac{[C]/[CO_2]}{[C]/[CO_2] + [C]/[CO] + [C]/[HCs] + [C]/[soot]} \quad (1)$$

$$CE\% = \frac{[S]/[SO_2]}{[S]/[SO_2] + [S]/[H_2S] + [S]/[COS] + [S]/[CS_2] + [S]/[Scmpds]} \quad (2)$$

METHODOLOGY

The overall flaring study was carried out over 5 years and included three stages of investigations. Each stage required development, construction, and testing of various types of equipment to examine either simulated flaring operations or reliable sampling of emissions from actual flaring. In the first phase, laboratory tests included flaring of various hydrocarbon-based gases, liquids, and combinations of

liquids entrained in gases. Accordingly, the laboratory flaring apparatus was constructed to perform the necessary functions required to simulate industrial flaring conditions.

The laboratory flare test chamber consisted of a vertical steel cylinder, 150 mm in diameter and 600-mm high with a flat quartz window for viewing the flame and an open top. It was enclosed in a fume hood with a water-cooled baffle at the top. The combustion airstream introduced through the bottom of the chamber passed through a flame trap diffuser, a stainless steel wire mesh, a perforated cone, and finally through a honeycomb flow straightener to minimize any turbulence caused by the diffuser. Fuel and fuel mixtures to the flare tip were supplied through a 6.4-mm stainless steel tube that protruded through the center at the bottom of the flare test chamber to accommodate the insertion of various flare tips.

The flare test chamber contained a number of sample probe ports along the height of the cylinder. It allowed for sample acquisition at approximately nine different vertical distances from the top of the flare tip and an infinite number of horizontal distances from the center line of the flare. Integrated samples were collected from the open top of the combustion chamber.

Gaseous fuels were supplied to the flare tip from high-pressure cylinders equipped with a series of control and safety devices. Metering nozzles were used to provide a wide range of flow rates. Liquid fuels were supplied to the flare as fine droplets by two different methods. The first method pressurized the fuel and injected it through a fine diameter (~0.008 mm) nozzle. The second method introduced liquid fuel to the flare system by the use of a laboratory fuel evaporator system. Entrainment of the liquids was achieved by bubbling gaseous fuel through a cylindrical vessel containing the liquid fuel. The amount of liquid fuel entrapped in the gas was varied by changing the liquid fuel temperature.

Flame shape, volume, and length are important characteristics of the flare that are dictated by flare tip design and are directly related to the rate of combustion. Flame length in particular is important because it is an indicator of the reaction time, the extent of exposure of the surroundings to heat transfer from the flame, the size of the combustion zone, and the extent of contact of the combustion gas discharge with the environment. To accommodate flaring of both gaseous fuel and liquid/gaseous fuel mixtures, several different sharp-edged stainless steel flare tips were used. Orifice sizes of the flare tips ranged from 0.1 to 3.0 mm, with appropriate sized nozzles used in conjunction with the type and amount of fuel flared. Nozzle tips were chosen such that maximum velocity of the gas did not exceed a Mach number of 0.2 for continuous flaring.

Sampling of the emissions was carried out in a manner to ensure unaltered samples of gaseous components at the point of sampling. To minimize the disturbance of the flare flow field by either the bulk of the sampler or the withdrawal rate of the emission sample, the probe size was kept to a minimum and the sampling flow rate was kept equal to or less than the flow rate of the main gas stream. The ideal sampling rate is one that is equal to the isokinetic sampling rate, which is accomplished by adjusting the sampling rate so that mass flux through the sampling probe is equal to that of the flaring stream.

The sampling system developed for use in these studies employed both heated and nonheated sampling lines. The continuous sample collected by the sampling probe was split into two streams. One stream was connected to a triple-stage Teflon filter pack, then to a heated Teflon sample line maintained at 125 °C through which the sample was drawn to the on-site analyzers. The second stream was used for obtaining integrated samples collected in Teflon bags and through glass sample traps containing Tenax or carbon adsorbents for further characterization of the emissions by gas chromatography/mass spectrometry (GC/MS). These samples were acquired from the sample stream immediately adjacent to the end of the sampling probe and were, therefore, neither filtered nor heated.

The laboratory flare-testing program consisted of an evaluation of pure gaseous, liquid, and co-flowing gas/liquid flames. A quality assurance/quality control (QA/QC) program was also carried out as an integral part of the laboratory studies on flare emissions. One of the QA/QC aspects of the flaring investigations was to verify that fuels being used, particularly in the laboratory flaring operations, were free from any other hydrocarbon compounds that might be measured as products of combustion. The analyses of both ultrahigh purity methane and propane revealed small quantities of benzene, toluene, and other hydrocarbons. A scrubber containing activated charcoal was used to remove these hydrocarbons from the fuels.

In the same manner, combustion air purchased in cylinders and of "ultra zero" grade quality was also passed through activated charcoal scrubbers and tested prior to usage. Liquid fuels did not contain any detectable levels of hydrocarbon components larger in molecular size than toluene. Additionally, hydrocarbons produced by these flares were found to escape from the flame in sufficient quantities to cause deposits within the combustion chamber that required complete removal and cleaning between runs to eliminate contamination. System blanks consisting of flowing the pre-purified combustion air through the flaring chamber and analyzing it prior to a flare test run were then used to ensure no cross-contamination.

The second stage of the flare-testing program was designed on a slightly larger scale and in an open

atmosphere. The flare system designed and constructed for these studies comprised the same fuel flow and delivery system used in the laboratory tests, but with slightly larger fuel transfer line and flare tip nozzles. Orifice sizes of the pilot-scale flare tips ranged from 2 to 6 mm in diameter, with the size corresponding to the type and amount of fuel flared.

Two different types of sampling systems were used in the pilot-scale tests. The first was simultaneous sampling through a series of probes set out in a horizontal plane perpendicular to the flame. The second was through hood samplers of various diameters that would obtain an integrated sample of the entire emissions from the flame. Once again, samples were either drawn for analyses by the on-site analytical equipment, or were collected on adsorbents or in suitable containers for the confirmatory analyses by combined GC/MS.

The third stage of the program focused on field flaring and included testing at oil-field battery sites that contained both sweet and sour gases. The oil-field battery site containing sweet gas was a central gathering and waterflood facility, fed by 24 wellsites producing both crude oil and solution gas (nonrefined) as well as co-produced water from one of the major oil pools in Alberta. The flare system at this site consisted of a liquid knockout drum (a horizontal drum with interior baffles situated immediately upstream of the flare to remove excess liquid hydrocarbons from the solution gas) and a freestanding flare stack, 12 m in height and 20 cm in diameter at the tip. It was equipped with an auto-ignitor and wind deflector to prevent flame blowout. The top 1.8 m of the stack was fabricated from stainless steel. It also contained an inlet baffle to reduce liquid accumulation. The majority of feed to this flare was solution gas from the treater at $\sim 8000 \text{ m}^3/\text{day}$.

The oil-field battery site containing sour gas was much smaller by comparison. It was fed by two wellsites producing both crude oil and the sour solution gas. The flare system consisted of a liquid knockout drum and a flare stack $\sim 15 \text{ m}$ high and 7.6 cm in diameter at the tip. It was also equipped with an auto-ignitor and a wind deflector to prevent flame blowout. The stack was fabricated from stainless steel. Solution gas from the two wellsites at this battery was directed to the flare, at $\sim 650 \text{ m}^3/\text{day}$.

The sampling system used for the field studies was based on the results obtained during the pilot-scale tests. A single probe sampling system was developed and tested in the laboratory prior to the field sampling. Standard hydrocarbon mixtures were directed through the heated probe and heat traced lines at varying temperatures to determine the most suitable temperature for hydrocarbon recovery.

The sampling system was fitted to a hydraulic basket lift with a 20-m reach. The probe was attached to a boom

that was in turn fastened to the basket. Samples were collected at several locations along the length of the sampling system, which included the sampling probe, a short length of heated sampling line that reached to the basket level, and a longer length of heated sampling line that extended to the ground. Samples were collected immediately after the probe, at the basket level, and at ground level. As in the previous phases of the study, samples were either drawn from this sampling system for the analyses by on-site analytical equipment or through adsorbent samplers for confirmatory analyses by combined GC/MS.

ANALYTICAL EQUIPMENT

There is no single piece of analytical equipment capable of detecting the wide range of potential emission products from flares. A combination of gas chromatographs and combined GC/MS were, therefore, used in these investigations.

A Varian model 3400 gas chromatograph with a flame-ionization detector and automated sample valve, coupled to a high resolution GS-Q megabore fused silica column (J&W Scientific, from Chromatographic Specialties), was used to detect the hydrocarbons ranging from methane to naphthalene. Data collection was carried out using a Hewlett Packard model 3393 computing integrator. This chromatographic system was calibrated using standard hydrocarbon mixtures covering the entire range of hydrocarbons analyzed by the system. The lower limits of detection averaged $\sim 25 \mu\text{g}/\text{m}^3$ and reproducibility of the system was $\pm 10\%$.

A Hewlett Packard model 5890A gas chromatograph with a flame photometric detector and automated sample valve coupled to the same type of fused silica column was used to detect the sulfur-containing compounds ranging from hydrogen sulfide to the light thiophenes. Data collection was carried out using a Hewlett Packard model 3393 computing integrator. This system was calibrated using standard sulfur compound mixtures covering the entire range of compounds detected by the system. The lower limits of detection ranged from 1 to $25 \mu\text{g}/\text{m}^3$, with a reproducibility also of $\pm 10\%$.

Gaseous components, including carbon dioxide, carbon monoxide, hydrogen, oxygen, nitrogen, and the light hydrocarbons were detected on a Microsensor Technologies Inc. model P200 high-speed gas chromatograph. The chromatograph contained two micro-gas chromatographs each with a micro-packed fused silica capillary column that provided high-speed gas separations, with detection by miniaturized thermal conductivity detectors. Complete analyses of these gases were carried out in $\sim 90 \text{ sec}$. Data handling and instrument control were performed with an M200 data system software package on a personal computer.

A Finnigan model 4025 gas chromatograph/mass spectrometer coupled to a dedicated Superincos data system was used in the analyses, data acquisition, and data handling of integrated samples collected on adsorbents. The SPB-5 fused silica gas chromatography column (Supelco Canada Inc.) was selected to provide the necessary separation of individual compounds for identification by the mass spectrometer.¹² Compound identification was accomplished initially by computer matching of the unknown spectrum to the U.S. National Bureau of Standards library of spectral responses to produce the five best "FITS" with confidence limits greater than 80%. Individual sample spectra were manually examined for molecular and fragment ions to ensure identification. Further enhancement of identification for compounds similar in mass spectral response was achieved by comparing gas chromatographic elution times with those of known standards.

Samples obtained for these investigations were collected either by drawing the flare emission sample stream through specific adsorbents, such as Tenax or Carbotrap contained in glass sampling tubes, or by directing the emission sample stream into either Tedlar sample bags or glass sampling bombs. A cryogenic preconcentrator was used to introduce hydrocarbon samples onto the gas chromatograph column. This was done both to allow the preconcentration of sufficient sample by passing specific volumes through the tube, while it was immersed in liquid nitrogen, and to concentrate the sample into the very small volume necessary for injection onto the gas chromatograph. The light hydrocarbons collected on the adsorbents were thermally desorbed from these traps with a helium purge into the cryogenic trap prior to injection into the gas chromatograph/mass spectrometer system.

A suitable range of standard hydrocarbons was tested in the system. Lower detectable limits for these compounds were governed only by the amount of sample collected in the preconcentrator. Additional tests were carried out on available standards to examine for losses or deterioration in sample containers or on adsorbents for up to 2 days. No appreciable losses occurred for standards examined, and repeatability of results fell within a range of $\pm 15\%$.

After thermal desorption, each trap was extracted with a volume of nanograde *n*-hexane. The extracts were reduced in volume, first under vacuum and then under ultrahigh purity nitrogen prior to injection into the GC/MS system. Standard polycyclic aromatic hydrocarbons (PAHs) were introduced and then extracted from blank adsorbent tubes to test for efficiency of recovery and reproducibility. All standards showed no appreciable losses, and recovery and repeatability were within $\pm 15\%$.

PAHs were also sampled using a modified PAH sampler.¹² This was employed as a means of providing higher

volume sampling capabilities. The PAHs identified in these investigations were found to vary widely in molecular weights, along with corresponding physical properties. Vapor pressure was found to be particularly important relative to the sampling techniques. Low vapor pressure compounds are usually associated with particulate matter, while higher vapor pressure compounds are usually found predominantly in the vapor state.^{13,14} The PAH sampling system used in this study to trap both phases of these compounds consisted of a filtration system using glass fiber filters to collect the particulate matter and associated hydrocarbons, followed by a polyurethane foam (PUF)-adsorbent cartridge to collect vapor-phase hydrocarbons that passed through the filter. The entire sampling system was made of Teflon and was used in all field investigations.

Sampling media were extracted with toluene prior to sample collection. The PAH-free filters and PUF-adsorbent cartridges were placed in their respective containers and connected to the sampling line immediately after the sampling probe. Samples were collected at flow rates ranging from ~ 10 to 20 L/min. Samples collected on the PUF cartridges were extracted in a soxhlet extractor using toluene. Extracts were reduced in volume prior to injection into the gas chromatograph/mass spectrometer system. Filter samples were collected both for determining carbon content and for PAH analyses. After weighing the filter to obtain carbon content, a portion of each filter was deposited into a pyrolysis sample inlet system attached to the combined gas chromatograph/mass spectrometer system, heated to ~ 300 °C, and analyzed as previously mentioned.

RESULTS AND DISCUSSION

Flaring of pure methane in controlled laboratory conditions was examined under laminar, transitional, and turbulent flow conditions to evaluate the effects of combustion, including the reactions that occur, and the products that are produced. The most efficient of these flames was under laminar flow (99.98%), followed by the transitional flow (99.91%) and turbulent flow (99.14%). The greatest difference in the combustion of these flames was primarily in the carbon monoxide that they release. Hydrocarbon concentrations in the emissions above the turbulent flame (16 mg/m^3) were double the values detected above the transitional flame (8 mg/m^3) and 4 times the amounts above the laminar flame (4 mg/m^3).

Samples taken from within these flames revealed significant quantities of hydrocarbons that are produced primarily by pyrolytic reactions. Acetylene, ethylene, and benzene were each found in concentrations exceeding 2 g/m^3 that represent over 10% of the conversion from the methane fuel within the flame. Ethenyl benzene, ethynyl

benzene, and naphthalene were also major components produced by the pyrolytic reactions. PAHs were produced in lesser concentrations.

Hydrocarbon concentrations above these flames were, in most cases, below the detection limits of the on-site gas chromatographs. Integrated larger volume samples collected on adsorbents determined that low levels of some of the hydrocarbons were indeed capable of surviving these flames and could be found in the emissions above them. Toluene and naphthalene were found in the highest concentrations (~30 mg/m³). Lesser amounts of benzene and many substituted benzene and naphthalene compounds were also identified.

Propane produced a sooty flame compared with similar methane flame experiments. The carbon particles, along with higher concentrations of hydrocarbons in the emissions, reduced efficiency levels of this flame to 98%. Considerably more hydrocarbons were found within the propane flame, including a number of PAHs.

Flaring of natural or industrial gas streams appeared to have little effect on the overall CE of the flame when compared with pure methane flames. There were, however, nearly twice as many hydrocarbons identified in samples obtained both from within and above the natural gas flame compared with the methane flame. Flaring of natural gas in the open atmosphere under turbulent conditions reduced the efficiency of the flame to 96%. The reduction was caused primarily by the presence of unburned methane and carbon monoxide in the emissions. Generally, hydrocarbon production by pyrolytic reactions within this flame was similar to methane flames. However, the ability of this flame to burn the majority of them was somewhat reduced. Crosswind effects on this natural gas flame produced a positive effect by reducing carbon monoxide levels and increasing the CE to over 99%.

Pure liquid fuels were flared to examine the types of compounds produced and the extent to which these hydrocarbons might be expected in the emissions from subsequent testing of co-flowing gas/liquid flares. Heptane was the cleanest of these flames, followed by cyclohexane, pentane, and toluene. Unburned hydrocarbons, along with the pyrolytically produced hydrocarbons, were the major components. Carbon particles were the next largest component, followed by carbon monoxide, which was particularly high in the emissions from burning toluene.

The hydrocarbons within all of the flames showed a high degree of unsaturation. Ethylene and acetylene were produced in large quantities, each accounting for 3–5% of the conversion products of the original fuel. Further unsaturation was found in aliphatic hydrocarbon chains connected to benzene and naphthalene rings, including large quantities of ethynyl and ethenyl benzene. Benzene

and naphthalene were also found in high concentrations within these flames.

Addition of liquid fuels and condensates to pure gas streams was found to have the most profound effect on impairing the ability of the resulting flame to efficiently combust all of the fuel and hydrocarbons that are produced within these flames. Methane and natural gas flame emissions contained low levels of benzene, toluene, naphthalene, and the saturated aliphatic substituted benzene compounds. This contrasts the high level of unsaturation found on the alkyl side chains of both benzene and naphthalene compounds emitted from either liquid or co-flowing gas/liquid flames. Hydrocarbons in these emissions were detected at 100 times the amounts found in emissions above methane and natural gas flames.

The degree to which the efficiency of combustion is affected by the addition of liquid fuels to a gaseous stream is primarily dependent on the amount of liquid fuel in the co-flowing stream and by the type of liquid fuel being added. The addition of 15% heptane to a pure methane flame reduced the efficiency to 97%. At 23% heptane, the efficiency was reduced to 93%, and when a 32% mixture with heptane was flared, the efficiency was further reduced to 81%.

The second stage of the study included open atmosphere experiments using a pilot-scale flare to examine the effects of slightly larger scale flaring. The pilot flare was a 10-fold scale above the flaring capability of the laboratory flare. Sampling systems developed and constructed included a multiprobe system and various size hood samplers. Tests were then carried out to determine the area above the flame where emissions were minimally diluted beyond the combustion zone.

Natural gas flaring was carried out under turbulent flow and very calm atmospheric conditions. Compared with laboratory tests, the major difference noted in these emissions was the increased levels of heavier hydrocarbons including benzene, toluene, and the xylenes. Their concentrations averaged 100 times more than amounts found in emissions from laboratory flaring of natural gas. A total of 65 compounds were identified by GC/MS in the more volatile fraction collected from these emissions, and an additional 70 were identified in the less volatile fraction. These compounds ranged from benzene to five-membered PAHs, many of which were previously identified in the emissions above the laboratory flare. The major difference in the character of these emissions was that carbon monoxide and unburned methane primarily accounted for the lowering of the CE in laboratory tests, while the unburned hydrocarbons caused the lower efficiency measurements in the pilot-scale tests. Comparison of emission samples collected above this flame using both the multiprobe and hood samplers revealed that the

results obtained from the hood sampler were very similar to those obtained from the center probe.

The majority of pilot-scale testing was carried out using a combined natural gas/condensate fuel stream. Liquid fuel composition in the gas stream was held constant at ~23% during these tests. Sampling was carried out at two different locations above the flame using both the multiprobe system and various sizes of hood samplers. Sampling was also carried out during calm atmospheric conditions and during crosswinds.

Comparison of the results obtained at 250 and 500 mm above the flame revealed no major differences. Variability in samples collected horizontally across the emissions zone was much lower at 500 mm from the flame tip, indicating a broadening of the emissions. Compared with results obtained from samples collected above the natural gas flame, the emissions from this flame appeared to diffuse much wider and more rapidly. Combustion efficiencies measured throughout this zone were consistently between 88 and 90%, primarily due to the presence of unburned hydrocarbons from the condensate, along with the high levels of hydrocarbons that escaped into the emissions.

The largest effect of the crosswinds on this flame was in the overall reduction of combustion efficiencies, ranging from 82 to 86%, or ~4–6% lower than what was measured under very low wind conditions. These lower efficiencies were a result of greater amounts of unburned condensate components and produced hydrocarbons in these emissions. Benzene, toluene, and xylene levels were ~50% higher than what was found when flaring during low wind conditions.

A total of 119 volatile and semi-volatile hydrocarbons were identified in the emissions above this flame. These compounds ranged from one- to five-ringed PAHs, along with many straight and branched chain saturated and unsaturated aliphatic compounds (see Tables 1 and 2). Some of these compounds were detected in concentrations more than 100 times the amounts measured in comparable laboratory flaring experiments. Results from hood and probe samplers also showed similarities between the hood sampler and what was collected from the center probe.

The field flare-testing program carried out in these investigations was directed to industrial flaring operations at oil-field battery sites in Alberta. Two different sites were examined: an oil-field battery with production from 24 wellsites that contained virtually no hydrogen sulfide, and a battery site that contained ~24% hydrogen sulfide in the fuel stream that was entirely directed to flare. The majority of the field testing was carried out at the oil-field battery site containing no hydrogen sulfide. Flare testing at this site was carried out with two different gas flows directed to the flare, with two different liquid fuel

flows, and with three different liquid fuel levels in the knockout drum.

A single probe sampling system was used for field testing, based on the pilot-scale tests that found comparative results in emission samples collected through center probe and hood samplers. The system was developed and tested in the laboratory prior to the field testing to determine the conditions necessary to recover the majority of hydrocarbons that are generated in the flame. It consisted of a heated probe and heated sampling lines controlled at different temperatures to allow for sampling the different types of hydrocarbons at varying locations along the sampling system.

Emission samples collected while flaring between 1 and 2 m³ fuel/min at the larger oil-field battery site contained ~20 times the levels of hydrocarbon gases than amounts previously detected above natural gas flames in pilot-scale studies. Benzene and other low-molecular-weight aromatic compounds averaged 10 times higher in their concentrations. There was also a much higher degree of unsaturation in alkyl side chains attached to the aromatic rings. The greatest difference in hydrocarbon concentrations between this field test and comparable pilot-scale testing was in the less volatile, higher molecular weight hydrocarbons (concentrations were up to 1000 times higher in the field testing). The higher levels of hydrocarbons in the emissions were the primary reason for the lower combustion efficiencies measured at the field flare (71% compared with 97% efficiency measured on the natural gas pilot flare test). This difference is attributed to the unprocessed solution gas at the battery site that constantly flowed through a knockout drum, re-entraining the liquid hydrocarbons.

Increasing the gas flow to produce a flame 3–4 times the length increased the particulate carbon content in the emissions by ~5 times, the volatile hydrocarbons by ~33%, and the PAH compounds by up to 3 times the levels found in emissions above the smaller flame. Some of the PAHs detected were attached to the particulate carbon collected on the filter, as sampled by the modified PAH sampler. These hydrocarbons, together with those collected on the PUF cartridge, were very similar in both types and quantities, varying by ~10–20% from adsorbent cartridge results. This higher volume flame also produced a decrease in the flaring efficiency to 67%, mainly due to the increased hydrocarbon and particulate carbon levels.

Additional condensate directed to this flare reduced its efficiency by 5 to ~62% in comparison with flames tested with only a gas stream directed to the flare. These combustion efficiencies were, however, ~20% lower than in pilot studies of flaring natural gas with co-flowing industrial condensate. Concentrations of the hydrocarbons identified in the emissions were between 10 and 100 times higher than in the comparative pilot-scale tests.

Table 1. Volatile hydrocarbons identified in emissions from a natural gas flame containing 23% condensate vapor, in crosswinds using the multiprobe and hood samplers (mg/m³).

Compounds	Probes (distance from vertical center in mm)				Hood (dia. mm)	
	150	75	0	75	150	300
PENTANE	1.45	2.18	3.69	2.22	1.15	4.11
3-PENTEN-1-YNE	2.14	2.98	2.97	2.33	1.71	3.16
HEXANE	2.61	3.11	6.81	2.79	1.36	5.81
CYCLOHEXANE	1.85	2.28	4.69	1.91	1.61	4.60
BENZENE	40.23	47.10	74.35	51.45	34.39	86.04
PENTANE, 3,3-DIMETHYL-	10.52	14.36	25.43	14.49	7.13	24.15
HEXANE, 3-METHYL-	7.60	11.47	17.24	9.26	5.18	17.39
CYCLOPENTANE, 1,2-DIMETHYL-	5.43	6.77	12.11	3.69	3.05	13.48
HEPTANE	57.54	70.49	95.66	65.19	28.06	120.67
CYCLOHEXANE, METHYL-	16.35	21.19	34.23	13.40	8.44	44.41
BENZENE, METHYL-	34.97	47.47	71.83	51.33	39.99	82.36
1-HEXENE, 2,5-DIMETHYL-	19.60	24.45	34.67	20.64	14.01	26.85
CYCLOHEXANE, 1,3-DIMETHYL-, CIS-	1.55	3.22	11.34	5.21	3.80	13.32
OCTANE	70.27	117.30	208.93	115.45	50.61	188.43
HEXANE, 3-ETHYL-	1.50	8.33	11.42	8.23	3.62	12.36
CYCLOHEXANE, ETHYL-	11.32	18.83	25.30	17.85	14.89	24.50
HEPTANE, 3,4-DIMETHYL-	14.50	19.88	20.77	16.67	12.97	20.78
CYCLOHEXANE, 1,2,4-TRIMETHYL-	1.83	3.70	7.09	3.96	3.52	7.55
BENZENE, ETHYL-	13.16	16.72	35.85	19.67	14.11	47.86
CYCLOPENTENE, 1-ETHENYL-3-METHYLENE-	50.30	63.03	85.90	62.60	48.50	71.40
2H-PYRAN-2-ONE, TETRAHYDRO-6,6-DIMETHYL-	25.19	31.10	38.61	36.68	28.96	43.22
CYCLOHEXANE, 1,3,5-TRIMETHYL-	6.13	9.40	16.02	13.32	5.84	15.93
BENZENE, 1,4-DIMETHYL-	25.19	26.95	37.11	27.67	22.21	39.93
NONANE	89.16	117.64	154.84	88.96	70.37	131.96
BENZENE, ETHYNYL-	4.21	6.25	9.46	5.79	3.27	15.74
BENZENE, ETHENYL-	8.00	12.22	18.14	13.56	6.33	22.49
NONANE, 3-METHYL-	15.36	16.40	21.23	17.25	8.24	23.90
BENZENE, 1-ETHENYL-2-METHYL-	7.29	11.16	18.13	9.79	6.71	19.21
BENZENE, PROPYL-	20.78	22.38	34.30	12.63	12.49	29.24
BENZENE, (1-METHYLETHYL)-	17.22	25.07	35.01	19.67	14.30	25.17
BENZENE, 1-ETHYL-2-METHYL-	12.04	18.31	29.13	19.38	13.73	20.44

Analyses were also carried out on the vapors drawn from above the liquid knockout while the gas/liquid fuel stream was directed to flare. A total of 88 hydrocarbons were identified in this fuel stream, many of which were also detected in the emissions after flaring these fuel mixtures. The most dominant of the hydrocarbons was *n*-octane with more than five carbon atoms. Other major components included C₆ to C₁₀ aliphatic hydrocarbons along with benzene, toluene, and the xylenes. By contrast, emission samples after combustion of this fuel had much lower quantities of the aliphatic hydrocarbons relative to aromatic compounds. The predominance of unsaturated alkyl side chains attached to aromatic rings in emission samples was in contrast to the saturated alkyl side chains attached to the aromatics found in the fuel mixture prior to flaring. This indicates that a large amount

of the hydrocarbons found in the emissions are not just unburned hydrocarbons, but hydrocarbons produced within the flame by the pyrolytic reactions.

Benzene, styrene, ethynyl benzene, naphthalene, ethynyl-methyl benzenes, toluene, xylenes, acenaphthylene, biphenyl, and fluorene were, in most cases, the most abundant compounds found in any of the emissions examined in the field flare testing. They were found in concentrations exceeding 300 mg/m³. These emissions usually contained between 100 and 150 identified hydrocarbons.

Field testing at the oil-field battery site that contained no hydrogen sulfide also focused on flaring of solution gas through the knockout drum containing three different levels of liquid hydrocarbons. The high and low levels were predetermined by the automated level control

Table 1 (continued).

Compounds	Probes (distance from vertical center in mm)				Hood (dia. mm)	
	150	75	0	75	150	300
OCTANE, 2,6-DIMETHYL-	7.91	13.35	19.44	13.67	6.64	23.70
BENZENE, 1,3,5-TRIMETHYL-	8.98	14.26	15.85	7.88	4.80	21.13
CYCLOPENTANE, 1-METHYL-3-(2-METHYLPROPYL)-	3.15	7.61	8.25	6.74	3.60	10.40
BENZENE, 1,2,4-TRIMETHYL-	10.39	15.24	20.84	17.38	12.46	20.97
DECANE	64.96	89.61	127.93	73.60	45.19	102.11
BENZENE, DIETHYL-	3.75	3.80	6.78	5.56	1.64	7.10
NONANE, 4,5-DIMETHYL-	1.27	2.41	5.62	4.61	1.32	4.68
BENZENE, 1-METHYL-2-PROPYL-	6.60	11.63	15.07	9.08	5.20	19.06
CYCLOHEXANE, (2-METHYLPROPYL)-	9.87	10.37	16.44	11.40	7.60	18.69
DECANE, 3-METHYL-	2.26	2.84	3.98	2.59	1.93	4.06
BENZENE, 1-ETHYL-2,3-DIMETHYL-	16.79	24.43	24.75	18.85	15.09	20.71
BENZENE, METHYL(1-METHYLETHYL)-	3.66	5.51	12.21	9.69	4.05	18.78
BENZALDEHYDE, 4-METHYL-	4.79	5.44	12.64	8.71	4.73	10.37
BENZENE, 1,2,3,4-TETRAMETHYL-	2.40	3.69	9.66	7.57	2.64	8.88
BENZENE, 1,2,3,5-TETRAMETHYL-	4.16	5.49	9.54	6.02	2.69	12.87
BENZENE, 4-ETHYL-1,2-DIMETHYL-	15.29	22.94	32.88	29.63	15.31	31.77
UNDECANE	1.71	2.59	3.67	2.31	1.77	2.36
BENZENE, 1,2,4,5-TETRAMETHYL-	0.66	0.60	1.67	0.78	0.82	1.78
BENZENE, 1-METHYL-4-(1-METHYLETHYL)-	0.91	1.35	1.65	1.47	1.29	1.92
BENZENE, (1,1-DIMETHYLPROPYL)-	0.88	1.01	2.11	1.65	0.71	1.87
BENZENE, (1-ETHYLPROPYL)-	0.60	0.84	2.17	1.39	0.70	1.75
BENZENE, 1,3-DIETHYL-5-METHYL-	0.87	0.86	1.68	1.18	0.39	1.74
NAPHTHALENE	27.21	46.22	61.88	38.05	25.18	79.95
DODECANE	1.94	3.74	7.86	3.99	1.65	7.40
NAPHTHALENE, 2-METHYL-	5.32	8.15	11.84	6.74	3.20	12.90
NAPHTHALENE, 1-METHYL-	1.55	5.83	6.69	3.42	2.61	7.84
1,1'-BIPHENYL	2.36	5.45	6.47	3.27	2.47	6.14
BIPHENYLENE	1.93	4.20	5.00	2.96	2.72	6.67
ACENAPHTHYLENE	1.62	3.13	3.78	2.23	1.73	3.88
9H-FLUORENE, 9-METHYLENE-	0.91	2.08	3.16	2.01	1.22	3.21

system that was a component of the knockout drum. Tests were conducted with the drum as full as it could operate, as low as the pump could remove, and approximately halfway between. Operating conditions of the flare system were very uniform between tests, where ~6 m³ of solution gas/min was directed to flare. This produced a flame of ~4–5 m. Winds were slightly variable throughout the tests with mean speeds ranging from 1.5 to 1.9 m/sec.

The CE of the flame when directing solution gas through the knockout drum while full was 64% (Table 3). Reducing the amount of liquid hydrocarbons in the knockout to approximately one-half of the high level had the effect of slightly reducing the hydrocarbon concentrations in the resulting emissions. Only acetylene and benzene were found in slightly larger amounts in this test. The reduction in liquid level also had the effect of slightly increasing the CE of this flare to 65%. Reduction of liquid hydrocarbons to the lowest possible level in the knockout

drum reduced hydrocarbon emissions from the resulting flame by ~25% below amounts found in tests with higher liquid levels in the knockout drum. Carbon levels were ~40% lower in these emissions and the CE was 5–6% higher than what was measured when flaring with larger amounts of liquid in the knockout.

These tests were conducted over a 2-year period and the hydrocarbons, including both volatile and nonvolatile fractions, identified by GC/MS were similar in types to comparable flare emissions examined in the previous year's study. Reducing the liquid hydrocarbon level in the knockout drum also reduced the pyrolytically produced hydrocarbon concentrations in the emissions in some cases by as much as 25–50%. Individual concentrations of these hydrocarbons varied up to a maximum of ~150 mg/m³. These emissions usually contained between 80 and 100 hydrocarbons identified by the gas chromatograph/mass spectrometer system, as well as the gases detected by on-site

Table 2. Semi-volatile hydrocarbons identified in emissions from a natural gas flame containing 23% condensate vapor, in crosswinds ($\mu\text{g}/\text{m}^3$).

Compound	Amount	Compound	Amount
BENZENE, 1,2,3,4-TETRAMETHYL-	4599.0	BENZENE, 1-METHYL-2-[(3-METHYLPHENYL)METHYL]-	617.5
PHENOL, 2-METHYL-	3047.0	BENZENE, 1-METHYL-3-[(4-METHYLPHENYL)METHYL]-	648.1
BENZENE, 4-ETHYL-1,2-DIMETHYL-	504.7	9H-FLUORENE, 9-METHYLENE-	1660.0
PHENOL, 4-METHYL-	9668.0	9H-FLUORENE, 1-METHYL-	398.7
PHENOL, 2-ETHYL-	794.6	9H-FLUORENE, 2-METHYL-	353.4
PHENOL, 2,5-DIMETHYL-	3396.0	ANTHRACENE	957.8
PHENOL, 3,5-DIMETHYL-	6502.0	9H-FLUORENE, 2,3-DIMETHYL-	1070.0
NAPHTHALENE (ACN)(DOT)	5145.0	BENZENE, 1-METHYL-3-(2-PHENYLETHENYL)-, (E)-	94.2
PHENOL, 3,4-DIMETHYL-	2136.0	BENZENE, 1-METHYL-2-(2-PHENYLETHENYL)-	125.3
BENZOIC ACID (ACN)	6749.0	1,1'-BIPHENYL, (1-METHYLETHENYL)-	92.9
BENZENE, (3-METHYL-2-BUTENYL)-	1634.0	PHENANTHRENE, 3-METHYL-	774.2
1H-INDENE, 2,3-DIHYDRO-1,2-DIMETHYL-	2267.0	ANTHRACENE, 2-METHYL-	929.4
PHENOL, 2-ETHYL-4-METHYL-	1130.0	4H-CYCLOPENTA(DEF)PHENANTHRENE	471.9
BENZENE, PENTAMETHYL-	3047.0	PHENANTHRENE, 2-METHYL-	363.7
NAPHTHALENE, 2-METHYL-	842.5	PHENANTHRENE, 2,5-DIMETHYL-	583.9
NAPHTHALENE, 1-METHYL-	457.1	PHENANTHRENE, 3,6-DIMETHYL-	716.8
1,1'-BIPHENYL	4413.0	PHENANTHRENE, 2,3-DIMETHYL-	221.9
NAPHTHALENE, 1,5-DIMETHYL-	895.1	FLUORANTHENE	526.9
NAPHTHALENE, 2-ETHYL-	775.2	NAPHTHALENE, 2-(PHENYLMETHYL)-	94.8
NAPHTHALENE, 1,7-DIMETHYL-	1254.0	BENZENE, 1,1'-(1,3-BUTADIENE-1,4-DIYL)BIS-	236.5
NAPHTHALENE, 2,3-DIMETHYL-	680.2	PYRENE	825.0
BIPHENYLENE	931.9	PHENANTHRENE, 2,3,5-TRIMETHYL-	451.3
NAPHTHALENE, 1,4-DIMETHYL-	785.2	11H-BENZO[A]FLUORENE	96.2
1,1'-BIPHENYL, 3-METHYL-	349.1	11H-BENZO[B]FLUORENE	230.3
NAPHTHO[2,1-B]FURAN	498.4	PYRENE, 2-METHYL-	130.7
NAPHTHALENE, 1,4,5-TRIMETHYL-	558.3	2,5-CYCLOHEXADIENE-1,4-DIONE, 2,5-DIPHENYL-	2480.0
BENZENE, 1-METHYL-2-(PHENYLMETHYL)-	825.5	CHRYSENE	131.3
BENZENE, 1-METHYL-4-(PHENYLMETHYL)-	278.4	TRIPHENYLENE	183.1
NAPHTHALENE, 1,4,6-TRIMETHYL-	380.4	BENZENE, 1,2-DIPHENOXY-	94.7
9H-FLUORENE	541.6	BENZENE, 1,4-DIPHENOXY-	101.4
BENZENE, 1,1'-METHYLENEBIS[4-METHYL-	431.1	BENZO[B]THIOPHENE, 3-(2-NAPHTHALENYL)-	667.8
1,1'-BIPHENYL, 2-ETHYL-	661.3	BENZO[E]PYRENE	219.8
		BENZO[A]PYRENE	346.1

gas chromatographs. The majority of the compounds identified were aromatic hydrocarbons, many of which had unsaturated alkyl side chains. A large number of these were PAHs ranging to coronene, a multiringed compound.

The sour gas flaring operation was considerably different from the operations at the larger battery site. This battery site was fed by only two wells with all gases directed to the flare. After passing through the separator, the solution gas was much lower in liquid hydrocarbon content, particularly from the butanes upward. Flow rates to this flare were about one-tenth of the average volume directed to the flare at the larger battery site. The resulting flame was much lower in luminosity, approximately one-third of the length sampled from most of the larger battery flaring tests, and contained no visible amounts of carbon.

The on-site characterization of the emissions from this plume consisted of both carbon- and sulfur-containing

compounds. Hydrocarbons were ~50% lower in concentrations in these emissions compared with amounts detected in the larger battery flare emissions. The carbon content was between 50 and 70% lower and the CE as calculated by the carbon mass balance was 84%, compared with the range of 64–71% at the larger battery site.

Sulfur compounds in these emissions included sulfur dioxide, hydrogen sulfide, carbonyl sulfide, carbon disulfide, and lower-molecular-weight mercaptans and sulfides. The sulfur dioxide concentration at the point of sampling was 6900 mg/m³. The carbon disulfide concentrations (480 mg/m³) were higher than any single hydrocarbon concentration measured in any of the flaring operations. The CE of this flame as measured by the sulfur mass balance was 82%, compared to 84% in the carbon mass balance.

A total of 31 volatile compounds were identified in the emissions from this flare and approximately one-third were

Table 3. Characterization of hydrocarbon emissions from the sweet oil-field battery flare with different levels of liquid hydrocarbon in the knockout drum (mg/m³ except where otherwise noted).

Liquid Hydrocarbon Level	High	Medium	Low
O ₂ (%)	20.4	20.8	20.8
N ₂ (%)	78.2	77.9	78.4
H ₂	15	20	10
CO	18	16	10
CO ₂	5050	4890	4720
Carbon	58	54	32
Hydrocarbon gases	224	215	168
Volatile hydrocarbons	340	320	215
Semi-volatile hydrocarbons	235	206	170
CE (%)	64	65	71

sulfur compounds. The major components were carbon disulfide, thiophene, benzothiophene, and benzene. An additional 43 less volatile compounds were identified, consisting primarily of polycyclics, thiophenes, and oxygenated compounds. Elemental sulfur was detected at 157 mg/m³.

CONCLUSIONS

Diffusion flares used to burn solution gases at oil-field battery sites in Alberta were investigated under a program that included laboratory, pilot, and field studies. In the laboratory studies, all fuels, including methane, produced large quantities of pyrolytic products within the oxygen void portion of their respective flames. The amounts that escape these flames, however, are directly related to the type of fuel flared. In the case of methane, these compounds are efficiently burned in the outer combustion zone and, hence, their combustion efficiencies are high (>99%). Other fuels do not burn quite so efficiently, and slightly higher amounts of these hydrocarbons as well as some of the unburned fuel can be found in their emissions. The addition of liquid fuel such as condensates to gaseous fuel streams had the most profound effect on impairing the ability of the resulting flame to efficiently combust all of the hydrocarbon fuel as well as the hydrocarbons produced within these flames, lowering combustion efficiencies to 80%.

Field studies conducted at two oil-field battery sites that contained various amounts of liquid fuel directed to flare resulted in reduced combustion efficiencies. Both unburned fuel and other hydrocarbons were found in their emissions. CEs of 64–71% were measured at one of the oil-field battery sites. The CEs were directly related to either how much liquid fuel was directed to flare, or how much liquid fuel was contained in the knockout drum. The emissions usually contained up to 150 identified hydrocarbons at concentrations exceeding 300 mg/m³.

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