Flammability of Mixtures of CO_2 and Hydrocarbons

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ABSTRACT

Determining if a gas stream is considered flammable is a common problem when performing hazards analyses for facilities associated with CO_2 floods. Guidelines that are based on the number of BTUs per standard cubic foot are sometimes used to determine if a stream is considered flammable. While these guidelines may work well for some streams, they may be either too conservative or not conservative enough in many instances. For example, some mixtures containing CO_2 (a good fire extinguishing agent) and methane are not flammable even if above the Btu/scf limit listed in the guidelines while other mixtures that contain H_2S or heavier hydrocarbons may be flammable well below the guidelines. Knowing better how to predict flammability has several benefits for CO_2 flood operators. Better knowledge of flammability prediction methods can reduce time and costs associated with safety reviews for surface facilities. But most importantly, more accurate prediction of flammability improves safety.

This paper reviews the data available on the flammability of mixtures of CO_2 and N_2 with hydrocarbons and the existing methods for predicting flammability of mixtures. The goal of the work is to help producers and operators appropriately predict the flammability of streams in order to maintain high safety standards.

INTRODUCTION

For design of surface facilities and for conducting Process Hazards Analyses (PHA) and other safety reviews, it is generally important to understand whether or not the material contained in the pipes and vessels is flammable, and, if so, under what concentrations. A typical response to a PHA question of safety about a potential leak might be “could result in accumulation of gases with a potential for explosion or fire, resulting in injury or death to personnel.” This is a serious statement that must be addressed in a PHA. When confronted with a flammable mixture, the PHA team considers potential safeguards that mitigate or prevent such dangers. These safeguards might be LEL (lower explosive limit) monitors, fire eyes, remote location, amount of time personnel are in the area, etc. Then the hazard is risk ranked and recommendations are made to minimize risk. These remedies are very important, but they can add appreciable cost to operations, not only in the purchase of such equipment, but also in the calibration and maintenance of it.

If, on the other hand, the gas mixture is not flammable, then no fire or explosion hazard is found and the team can move on to the next issue. Thus the question of flammability of gas mixtures containing large concentrations of inert gas, including CO_2 or N_2 becomes important, particularly in the Permian Basin where many CO_2 EOR floods are underway or planned and where naturally occuring gas sources contain high nitrogen and/or carbon dioxide levels.

(Note: This paper addresses flammability predictions only. It should be noted that H_2S, CO_2, N_2 and other compounds may have other hazards [such as danger of asphyxiation, toxicity, etc.] that are not covered in this paper.)

The flammability question is important for both the processing plant and for the field operation of injection and producing wells. How does one determine whether a gas mixture is flammable by looking at the gas analysis? Does a heating value greater than 200 BTU/scf really determine if a gas is flammable? And what are the bounds of flammability? The answers may not be obvious when the gas contains appreciable concentrations of carbon dioxide or nitrogen - or - significant amounts of heavier hydrocarbons. Further, in a plant setting, the correct answer to the flammability question may change as the gas passes through the various processes. For example, the gas entering a liquids recovery plant that contains 78+% CO_2 may be deemed flammable because of the heavy hydrocarbons it contains. After NGL recovery, this gas may not be flammable, even if it still contains significant concentrations of methane.

There are numerous resources containing large amounts of gas flammability data (e.g., Bureau of Mines [BOM] bulletins 450, 503, and 627). However, using the published data to predict whether or not a specific gas stream is flammable can often be a confusing and frustrating task for anyone who doesn't use this material routinely. As a result, there is increased chance for errors to be made, or for reliance on subjective methods such as personal opinion or gut feeling.

Even if one uses the documented methods, there is the possibility of some amount of error, especially as one approaches the limits of flammability for complex mixtures. The various methods of estimating flammability discussed here each result in the calculation of a "flammability quotient." When the flammability quotient calculated by a particular method is equal to one or greater, the gas is considered flammable by that method. But, is it actually flammable? Is there any real difference between a flammability quotient of 0.9 vs. one of 1.1? Theoretically, yes, but the accuracy of the determination depends on the quality of the data used in the method. If one were looking at a simple mixture that contained only CO_2 and methane the data appear to be very good. However, for complex mixtures close to a flammability quotient of 1.0, only laboratory flammability studies can definitively answer questions of flammability. Even then, the flammability is determined under ideal conditions, which may not perfectly simulate gas jetting from a flange leak or from a hole in a vessel or pipe.

This paper explains how to use some of the available data and established methods to predict flammability of gas mixtures. It also describes a spreadsheet tool that was developed that takes gas composition as an input and provides flammability predictions by each of three prediction methods.

OVERVIEW OF FLAMMABILITY ANALYSIS TERMINOLOGY AND METHODS

The flammability of gas mixtures in air is frequently characterized by several parameters that are defined below:

- Lower Flammable Limit (LFL) in air – The lowest concentration of a gas or gas mixture in air at which the gas or gas mixture will ignite at atmospheric pressure and 20 ºC. Below this concentration the mixture is too lean to burn (from ASTM E-681-01).
- Upper Flammable Limit (UFL) in air – The highest concentration of a gas or gas mixture in air at which the gas or gas mixture will ignite at atmospheric pressure and 20 ºC. Above this concentration the mixture is too rich to burn, but may become flammable upon mixing with air (from ASTM E-681-01).
- Maximum concentration of a flammable component in a nonflammable binary mixture – For binary mixtures, e.g., methane and nitrogen, this is the maximum concentration of the flammable component (methane) such that the mixture is still not ignitable after mixing with any proportion of air at atmospheric pressure and temperature.

The first two quantities are familiar to many that work in the process safety field. The maximum concentration of a flammable component in a nonflammable binary mixture is used primarily in the determination of flammability of gas mixtures. As an example, a mixture of nitrogen and methane is considered flammable according to standard methods when it contains more the 14.3 % methane (ISO 10156, ASTM E-681-01). Mixtures containing less than 14.3 % are incapable of forming flammable mixtures with air upon mixing in any proportion.

For the purpose of this paper, we are concerned only with mixtures that

- Do not contain oxygen
- Are mixed with air at ambient pressure and near ambient temperatures.

To demonstrate some of the concepts used in flammability determinations, a triangle diagram (shown in Figure 1) is useful. Figure 1 shows the flammability diagram for methane with mixtures of nitrogen and oxygen at atmospheric pressure and 25 ºC. (Note: Although this diagram is for nitrogen mixtures instead of CO_2 mixtures, the concepts illustrated are the same.) Each point within or on the triangle represents a particular composition of the gas mixture; the corners of the triangle correspond to pure methane, pure nitrogen, or pure oxygen.

The “pure methane mixing in air” line shown on the diagram represents the set of all compositions that result when pure methane is mixed with air. This line begins at the corner where the composition is 100 % methane, and then extends to the point at the bottom of the triangle corresponding to the composition of normal atmospheric air (79% N_2 and 21% O2). When the line reaches the bottom of the triangle, it is as if the methane mixed with an infinite amount of air. Notice that the line passes through the flammability envelope. Thus, pure methane can be considered flammable because it forms ignitable mixtures when it mixes with air in certain proportions.

In comparison, a mixture of 10 % methane and 90 % nitrogen cannot form flammable mixtures. The “10% methane, 90% nitrogen mixing with air” line is also shown on Figure 1. It starts at the point that corresponds to the initial mixture of 10% methane in nitrogen and extends to the composition of normal atmospheric air Notice that no matter what proportion it is mixed with air, the resulting composition never enters the flammability envelope. Therefore, the 10% mixture would not be considered to be an ignitable mixture upon release into the air. This is the basis for the methods of determining flammability discussed in the next section.

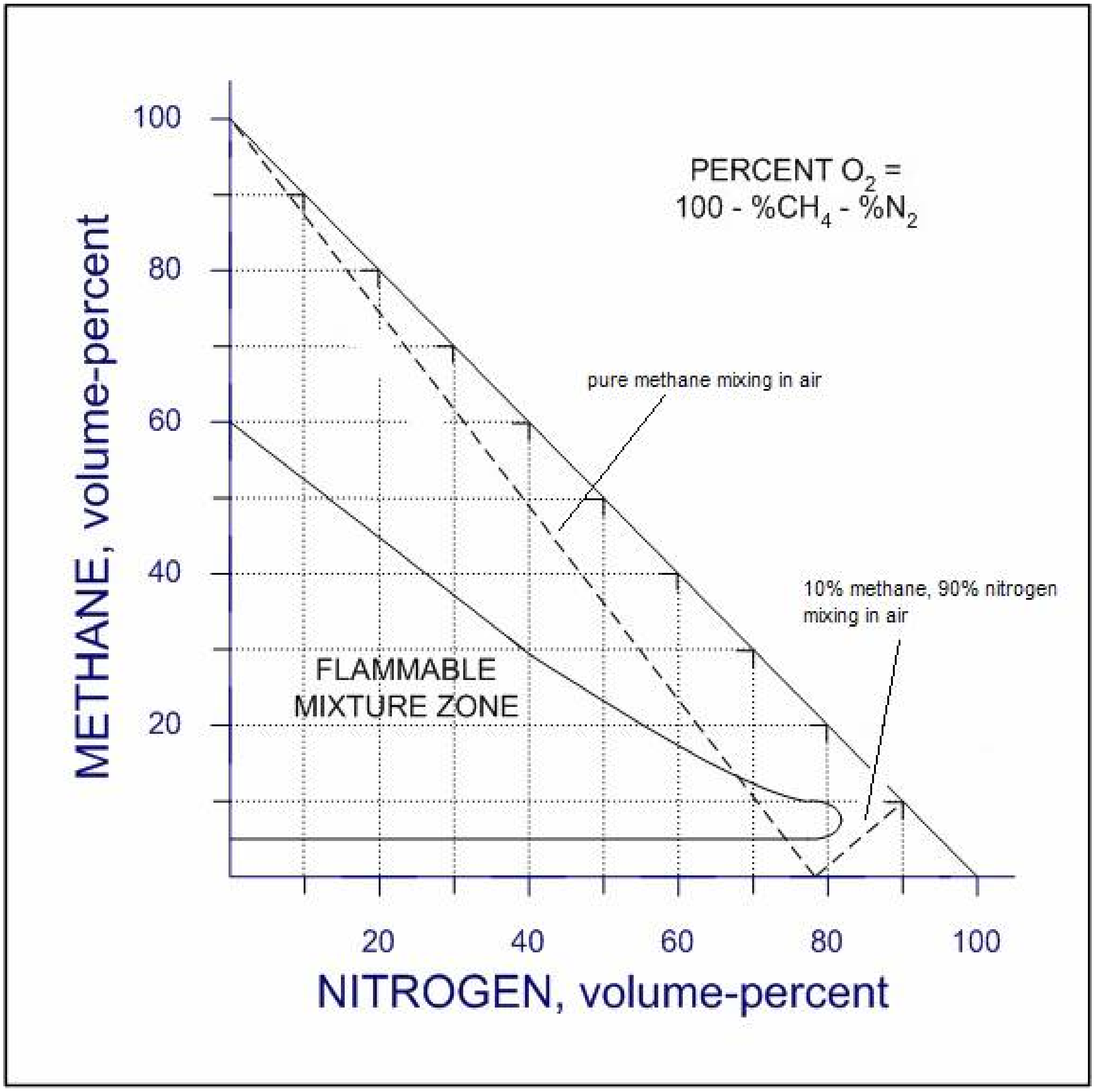

Figure 1. Flammability Diagram for Mixtures of Methane, Nitrogen, and Oxygen. (For use as an example only; for any other purpose, consult original source [Source: Zebetakis, 1965])

METHODS FOR PREDICTING FLAMMABILITY OF GAS MIXTURES

Determining if a particular gas mixture is flammable is an essential step in completing a process hazard analysis or determining the proper electrical classification of a process area. Ideally, one should always use actual experimental data if a particular gas mixture is questionable or close to the limit. There are several firms available to measure flammability limits of gas mixtures given samples of the gas or a gas composition to work with. However, in the absence of specific experimental data, we found that there are methods to predict whether the gas mixture is expected to be flammable or not.

As a starting point, the EPA has published guidelines for flares (40 CFR, CHAPTER I – PART 60) that state that flares shall only be used with gases with a net heating value of 200 Btu/SCF for non-assisted flares and 300 Btu/SCF for steam-assisted or air-assisted flares. This guideline seems to suggest that gas mixtures with a net heating value of 200 Btu/SCF or greater are always to be considered flammable, and that gas mixtures with a net heating value of less than 200 are always to be considered nonflammable. This may not always be the case, as we shall see.

To further investigate available flammability prediction methods, we searched the engineering literature, Internet, and interviewed several process safety experts. We found several approaches for making flammability predictions as listed below:

- Application of Bureau of Mines Flammability Data Bulletins
- Application of Industry Standards ISO 10156 and CGA P-23
- Application of Guidelines Based on Calculated Adiabatic Flame Temperature (CAFT)

Each of these approaches is described in the following subsections:

Application of Bureau of Mines Flammability Data Bulletins

The United States Bureau of Mines (BOM) published many reports on fire and explosions resulting from flammable gas mixtures. For our purposes in investigating mixtures of CO_2 and light hydrocarbons, we found that BOM bulletins 503 and 627 were most helpful, along with BOM Technical Publication 450. To summarize, the BOM bulletins present flammability data for mixtures of single hydrocarbons with air in the presence of inert gases nitrogen and CO_2. Flammability limits for gas mixtures that contain multiple hydrocarbons are then calculated based on limits for the individual components using Le Chatelier's rule. (Le Chatelier's rule is a mole fraction weighting method; for further explanation, see Mashuga & Crowl, 2000.) Binary mixture data were taken directly from BOM bulletins and tabulated below:

Table 1. Flammability of Binary Mixtures of Alkanes with Nitrogen and CO_2

Binary Mixture	Maximum Amount of Hydrocarbon in Nonflammable Mixture, (mole percent)	Bureau of Mines Reference (Bulletin No. and Figure)
Methane/CO_2	23	503, Fig. 1
Ethane/CO_2	12	503, Fig. 2
Propane/CO_2	11	627, Fig. 30
n-Butane/CO_2	9.6	627, Fig. 31
n-Pentane/CO_2	7.9	627, Fig. 32
n-Hexane/CO_2	6.8	627, Fig. 33
Methane/N_2	14	503, Fig. 1
Ethane/N_2	7.2	503, Fig. 2
Propane/N_2	6.3	627, Fig. 30
n-Butane/N_2	5.7	627, Fig. 31
n-Pentane/N_2	4.4	627, Fig. 32
n-Hexane/N_2	3.8	627, Fig. 33

Multicomponent mixtures can be evaluated using the approach described on pages 5 – 8 of BOM Bulletin 503. This method requires that the multicomponent mixture be "dissected" into binary mixtures. The binary mixtures are then evaluated to determine their flammability limits. These flammability limits are then averaged according to the proportion of the dissected mixtures. As an example, a ternary mixture of 50% CO_2, 25% methane, and 25% ethane could be dissected into two binary mixtures. Although an infinite combination of mixtures could be constructed, the first binary mixture could possibly be 50% CO_2 and 50% methane, and the second binary mixture 50% CO_2 and 50% ethane. The upper and lower flammability limits of 50:50 CO_2/methane mixture and the 50:50 CO_2/ethane mixture are determined using the data. Then, these two sets of upper and lower flammability limits for the binary mixtures are combined using Le Chatelier's rule to predict the flammability limits of the original mixture. In this case, since both dissected mixtures are flammable, the original mixture is also flammable.

For some mixtures, such as 1 % methane, 1% ethane, and 98% nitrogen, the multicomponent mixture cannot be dissected into a set of flammable binary mixtures. It could be dissected into one flammable mixture, say 50 % methane and 50 % nitrogen, but the other mixture of containing mostly nitrogen (98.98%) and the balance ethane (1.02%) is not flammable. The key is that it cannot be dissected into a group of binary mixtures whereby each mixture is flammable. The B.O.M. Bulletin 503 states on page 8 that “if the amount of inert gas is so great that a complete series of flammable mixtures cannot be dissected, the air-free mixture is not flammable.” This is the basis for applying the BOM Bulletin data to determine whether a gas mixture is flammable or not.

Further consideration of this method reveals that a multicomponent mixture can be dissected into binary mixtures whereby each flammable component is allocated an amount of CO_2 corresponding to the leanest mixture of that component in CO_2 that is still flammable. After doing this, if there is still some CO_2 left over, then it is impossible to dissect the multicomponent mixture into flammable binary mixtures and thus the original multicomponent mixture is nonflammable.

As an example, consider 100 moles of the above mixture containing 15 % methane, 2 % ethane, and 83 % CO_2. From Table 1 above, we find that the leanest ethane/CO_2 mixture that would be considered at the border of flammability should be 12 % ethane and 88% CO_2. Thus, the 2 moles of ethane can be combined with a maximum of approximately 14.5 moles CO_2 and still be considered flammable. Similarly, from Table 1 above, we find that the leanest methane/CO_2 mixture that would be considered at the border of flammability should be 23% ethane and 77% CO_2. Thus, the 15 moles methane can be combined with a maximum of about 48.9 moles CO_2 and still be flammable. The total CO_2 required is thus 63.4 moles. This leaves about 29.6 moles of CO_2 left over. Since there is CO_2 left over, the original mixture is therefore nonflammable.

This method is easily extended to more components by using the appropriate maximum ratio of CO_2 to flammable component for each alkane from methane to n-hexane. These ratios are given in the following table (obtained from the data in Table 1).

Table 2. Maximum Ratio of CO_2 to Hydrocarbon for Two-Component Flammable Mixtures Based on Bureau of Mines Data

Compound	**Maximum Ratio of CO_2 to Hydrocarbon**
Methane	3.25
Ethane	7.25
Propane	7.69
n-Butane	9.31
n-Pentane	11.6
n-Hexane	13.5

For mixtures containing both nitrogen and CO_2, an additional complication is introduced in deciding how to allocate the nitrogen. For most of the gases in this study, the nitrogen amount is much less than the CO_2 amount. Therefore, one arbitrary method is to allocate the nitrogen to methane, and remove a methane/nitrogen mixture of 14 % methane in nitrogen and then proceed to dissect the remaining CO_2 and hydrocarbon mixture.

The preceding methodology of calculation has been implemented in a simple spreadsheet model. The user simply inputs the gas composition, and the spreadsheet calculates the maximum CO_2 required. If this maximum is higher than the actual CO_2, then the mixture can be dissected into a series of flammable binary mixtures and thus the mixture is flammable. The ratio of this maximum to the actual CO_2 required is defined as the flammability quotient. Therefore if the quotient is greater than one, the mixture is predicted to be flammable; and if the quotient is less than one, the mixture is predicted to be nonflammable.

One significant drawback to using the BOM method is the lack of data on iso-hydrocarbons, and alkanes greater in molecular weight than hexane. As will be shown later, excluding these compounds or lumping them together may affect whether or not a mixture is determined to be flammable, if it is close to the limit of flammability.

Application of Industry Standards ISO 10156 and CGA P-23

The second approach for determining flammability of gas mixtures is to apply industry standards. The International Organization for Standardization (ISO) and the Compressed Gas Association (CGA) have both published standards for categorizing gas mixtures containing flammable and nonflammable components. These standards both give a formula and detailed instructions for calculating a flammability quotient. Both of these methods are easily implemented with a spreadsheet. The methodology for both standards is similar, but not identical to the method previously described for application of the BOM data and will not be described in detail here.

The main advantage with using the industry standard approaches is that these methods have tables that allow the method to be extended to over 50 compounds, including many chemicals and non-hydrocarbon components, as well as inert diluents other than nitrogen or CO_2 such as argon, helium, or neon. The two methods produce similar results, although there are some important differences for mixtures containing certain components, such as isobutane or isopentane.

The ISO standard provides flammability data for an iso-alkane (e.g., isobutane or i-butane) and the corresponding normal alkane (e.g., normal butane or n-butane) together as one number which is used in the calculations. The CGA standard has data for the iso-alkanes that is separate from that for the normal alkanes. Since the iso-alkanes have different flammability limits than the normal alkanes and since iso-alkanes may be present in different ratios depending on the stream considered, the CGA standard may be the more useful of the two standards for oil and gas producers.

Both the ISO and the CGA methods were also implemented in the same spreadsheet with the BOM method, so that the spreadsheet shows and compares flammability predictions for all three methods when applicable.

Application of Guidelines Based on Calculated Adiabatic Flame Temperature (CAFT)

More recently, flammability research has been directed at developing more rigorous methods of determining flammability limits, especially for mixtures at high pressures and/or high temperatures with oxygen present in the mixture. Crowl (1999) describes a fundamental method for determining whether mixtures are expected to be flammable. The method involves calculating the adiabatic flame temperature of the mixture at equilibrium. If the calculated adiabatic flame temperature (CAFT) is greater than 1200 K higher than the initial temperature, then the mixture is declared flammable.

The CAFT method is more conservative for many mixtures. For example, some CAFT calculations were made for mixtures of methane and CO_2 to determine the maximum amount of methane possible in a nonflammable mixture. Methane/CO_2 mixtures have been thoroughly characterized and it is known that these mixtures are nonflammable up to about 23 % methane. The CAFT method is overly conservative in predicting that these mixtures become flammable starting at about 15 % methane. For some applications, this could result in increased costs if operators had to treat the stream as flammable when in fact it was not a flammable stream.

For mixtures of H_2S and air, the CAFT method appears to underestimate the flammability. For example, a mixture of 6.5% H_2S in air is above the LEL for H_2S, but the calculated adiabatic flame temperature is only about 600 K higher than the starting temperature. Therefore, the CAFT method suggests that this mixture is not ignitable when in fact it is ignitable.

COMPARISON OF PREDICTIONS USING DIFFERENT METHODS FOR PROCESS STREAMS AT OXY FACILITIES

Occidental (Oxy) Permian Ltd operates three plants near Denver City, Texas that process gas streams with high concentrations of CO_2. One goal of this project was to apply flammability models to various process streams at these plants to predict their flammability characteristics based on the known compositions of these streams. Table 3 shows the composition of four process streams, and Table 4 shows the estimated heating value and flammability characteristics.

Table 3. Composition of Streams at Oxy Permian Facilities, mol %

	DUCRP HP Inlet	**DUCRP RA Overhead**	**Willard Mixed Inlet**	**WCRP West Inlet**	**WCRP West Inlet (iso and normal lumped together, hexanes+ lumped together, no H_2S)**
Methane	10.65	13.99	8.50	4.39	4.39
Ethane	3.68	3.14	2.29	2.63	2.63
Propane	1.95	0.00	1.34	1.49	1.49
i-Butane	0.31	0.02	0.21	0.23	
n-Butane	0.80	0.07	0.61	0.64	0.87
i-Pentane	0.23	0.01	0.21	0.25	
n-Pentane	0.24	0.00	0.25	0.29	0.54
n-Hexane	0.23	0.00	0.33	0.43	0.71
n-Heptane	0.20	0.00	0.16	0.21	
n-Octane	0.04	0.00	0.05	0.07	
H_2S	0.22	0.00	0.11	0.22	
N_2	1.30	1.30	1.08	0.42	0.42
CO_2	80.15	81.48	84.87	88.73	88.94

One of the process streams (WCRP West Inlet) is shown with a more complete analysis and then also with a simplified analysis. In the simplified analysis, 1) the concentration of each iso-alkane is added to that of the corresponding normal alkane and treated as if it were a normal alkane, 2) concentrations of compounds heavier than hexane are added in with the hexane concentration, and 3) H_2S concentration is omitted. The two WCRP West Inlet analyses are shown to illustrate how all three calculation methods compare, and also to show how using a simplified analysis may artificially cause a stream to appear not flammable when it would otherwise appear to be flammable.

Table 4. Comparison of Flammability Characteristics of Oxy Permian Streams using Different Methods

	DUCRP HP Inlet	**DUCRP RA Overhead**	**Willard Mixed Inlet**	**WCRP West Inlet**	**WCRP West Inlet (iso and normal lumped together, hexanes+ lumped together, no H_2S)**
Net Heating Value, BTU/SCF	276	181	213	198	195
Flammability Quotients, BOM Method	(1.22)*	(0.84)*	(0.88)*	(0.77)*	0.77
Flammability Quotient, ISO 10156 Standard Method	1.50	1.00	1.12	1.04	0.96
Flammability Quotient, CGA P-23 Standard	1.59	0.90	1.22	1.16	0.84
Flammable – BOM Method?	(Yes)*	(No)*	(No)*	(No)*	No
Flammable – ISO 10156 Standard?	Yes	Yes	Yes	Yes	No
Flammable – CGA P-23 Standard?	Yes	No	Yes	Yes	No

* This result comes from using the BOM method with iso-alkanes combined with and treated like normal alkanes, with compounds heavier than hexane combined with and treated like hexane, with H_2S excluded, and with the composition renormalized. It is shown here to illustrate the result one might get if one used only the BOM method.

The results in Table 4 show general agreement between the CGA and ISO methods. However, the methods may not agree exactly, as shown by the DUCRP RA Overhead column, when the stream is borderline flammable as indicated by flammability quotients that are very near 1.0 . For this stream, the ISO 10156 Standard Method predicts that the mixture is flammable, while the CGA method predicts that it is nonflammable.

Results obtained by simplifying the data and then using the BOM method are also shown in Table 4 within parentheses and with an asterisk following the parentheses. Comparing the result so obtained from the BOM method with the results obtained with the CGA and ISO standards shows that if one simplifies the data and uses the BOM method in this fashion, doing so may artificially show some streams as nonflammable when they are in fact flammable. For streams that have a flammability quotient near one, it would be more conservative to use a method (e.g., the CGA or ISO standards) that can make use of all the available data, or better yet, get actual flammability data from a qualified source for the stream in question.

The last two columns show results for all three methods when applied to a simplified analysis. This column is presented in order to show that when the three methods are all used on a data set that is amenable to the BOM method, all three methods show general agreement.

EXPERIMENTAL MEASUREMENT OF FLAMMABILITY FOR OXY'S GAS STREAMS USING ASTM METHODS

Samples of several high-CO_2 streams were also taken at Oxy Permian's DUCRP facility and sent to a qualified laboratory for determination of flammability limits. While the data were not available in time for inclusion in this paper, they may be available in the future.

CONCLUSIONS

- Flammability of gases is best determined by experimental measurements using standard test methods, especially for mixtures that are close to their flammability limits. These tests may be conducted by commercial labs, but less rigorous tests could be performed in the field, for example in a plant lab under a vent hood using Bunsen or Fisher burners, an ignition source, and rotameters to generate gas mixtures, assuming proper safety precautions were made and applicable test procedures followed.

- When measurement data are not available, flammability of gas mixtures can be estimated using one of several methods, including analysis of Bureau of Mines flammability data, ISO and CGA standard methods, and application of calculated adiabatic flame temperature (CAFT) methods. Of these, the CGA standard is the one that breaks out iso-hydrocarbons and normal hydrocarbons separately and thus may be more applicable for oil and gas production situations.

- For streams whose composition allows application of all three methods, results from each of the methods are generally in agreement, with the exception of gas mixtures that are very close to the limit between flammable and nonflammable. When the methods are close to the limit between flammable and nonflammable, one can conservatively assume that it is flammable for safety purposes, or get better data. Whenever in doubt, flammability tests are recommended.

- Mixtures of hydrocarbons with CO_2 are less flammable than the same mixtures with nitrogen, even though the net heating value may be the same for both mixtures. For example, anything greater than 14.3% methane in nitrogen is flammable, but 14.3% methane in CO_2 is clearly nonflammable as shown by the literature data. (Data show methane at concentrations up to ~23% in CO_2 to be nonflammable.) This is because CO_2 has a higher heat capacity, a better ability to absorb heat and quench a flame than nitrogen.

- Certain mixtures, e.g. methane in nitrogen, or hydrogen sulfide in CO_2, may be ignitable even though the mixture's heating value is less than 200 Btu/scf.

Other potential uses of these types of flammability determinations are in determining amounts of assist fuel that must be added to a flare in order to support combustion and in determining the volumes of inert gas needed to safely purge vessels below the flammable limit. With respect to purging, other published resources should also be consulted (e.g., Kinsley, 2002 & Tomkins, 1934).

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