

A Tutorial on:

Generic Fire-Limit Analysis

of

Binary, Ternary, and Quaternary

Gas Systems

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Table of Contents

Introduction	p. 5
Binary System Analysis	p. 7
Ternary System Analysis	p. 13
Practicality Considerations in Fire and Explosion Prevention	p. 31
Quaternary System Analysis	p. 39
Closure	p. 49
References	p. 51

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Introduction

REFERENCE: Werley, B. L., “A Tutorial on Fire-Limit Analysis of Binary, Ternary and Quaternary Gas Systems”, ©2000, ©2017, public domain 15 April 2017, 50 pages.

ABSTRACT: Gas phase fire limit analysis is important to the design of safe systems and the investigation of incidents in such systems. Fires may result from fuel/oxidant combustion or other physical mechanisms such as explosive polymerization, or decomposition all of which have generic similarities. Such analysis is often in large part empirical in basis and can be onerous to accomplish. PC algorithms that help to read ternary diagrams can facilitate such analysis and improve their resulting quality.

KEY WORDS: binary ternary quaternary fire limits, flammability, combustion, hazards, risks

In 2000 and soon after, the writer suggested American Society for Testing and Materials (ASTM) Committees: G4 on *Compatibility and Sensitivity of Materials in Oxygen-Enriched Atmospheres* and E27 on *Hazard Potential of Chemicals* develop standards on fire limit nomenclature, empirical generic fire limit analysis and the validation of simplified PC utility support. Later, the idea was also broached with the leadership of the National Fire Protection Association *Committee 53M*. There was no interest.

Although there is much theory on combustion published, including efforts to theoretically predict fire limits, today it may well still be more common to experimentally measure and empirically estimate fire limits. Indeed, even in cases where theoretic-

cal fire-limits can be predicted there can still be merit to empirical evaluation of them. Furthermore, such interpretation is often not done well, or is avoided and much disdained. In large part this turns out to be a result of the common, and perhaps necessary, use of ternary depictions, which the writer admits has been an unpleasant source of much personal consternation over the years. The writer has seen little tutorial available about the graphical methodology for empirical fire limit estimation and seeks to remedy that.

In 2000 the writer also made a prototype, a demonstration, a proof of concept, PC facility available that could greatly simplify and improve many of these analyses and facilitate some understanding of the vagaries in these data *if validated*.

Typically, fuels are sequestered, and so it has been common to think of fire risk as the addition of fuel to an ambient oxidant. However, there can be cases in which the fuel is the ambient and one need to fear the addition of an oxidant. And there can even be cases in which one need fear the addition of an inert gas to a system. This effort will treat fire limits as generic mathematical parameters and seek to once again interest one of these relevant Committees, or another alternative, into formalizing the practices.

To do this, first simpler binary fire limits will be examined in a more cumbersome method than they require which will then be evolved into the ternary tool that is so widely disdained. Once traditional perspectives on fuels, oxidants and diluents has been introduced (and to some extent shed in favor of a generic treatment) the approach will shift to merely consider how much of a given generic gas (or liquid or solid in special cases) can be added to a system before it enables or prevents a sustained fire or explosion.

Binary System Analysis

The most easily understood fire hazard is when one has a fuel (a flammable gas) and an oxidant (like oxygen), and one is concerned with how much of the fuel can be mixed with the oxidant before it just becomes a sustained-fire risk. Graphically this can be exhibited as in Figure 1, in which two gases (G1 and G2) are shown, and in which “pure” (100%) oxidant gas is shown as gas one (G1) at the origin and the “pure” (100%) fuel is shown as gas two (G2) at the 100% point. The line between them is the linear scale for representing all possible homogeneous mixtures of the two with the origin representing 100% G1, 0% G2 and the right limit representing 100% G2, 0% G1. The scale shown (0-100% G2) is for G2 added to G1, but a second scale (100% to 0%) is also shown to indicate G1 added to G2.

Actual testing might well involve a much different sequence of testing and far fewer tests. However, in this case each series of tests spans the full range of mixtures and some tests produces positive (+) results (meaning above the sustained-fire limit) and some produce negative (-) results (meaning below the fire limit). Note there tends to be a clustering of positive result which may also include negative results, but outside this cluster only negative results are seen.

In traditional fire-limit testing [*1,2,3*], the fire-limit is taken as the point at which self-sustaining (equilibrium) propagation just occurs and therefore a fire might propagate indefinitely. Typically a glass tube 36 inches long is filled with a homogenous mix-

¹ Italic numbers in brackets refer to the reference list at the end of the paper.

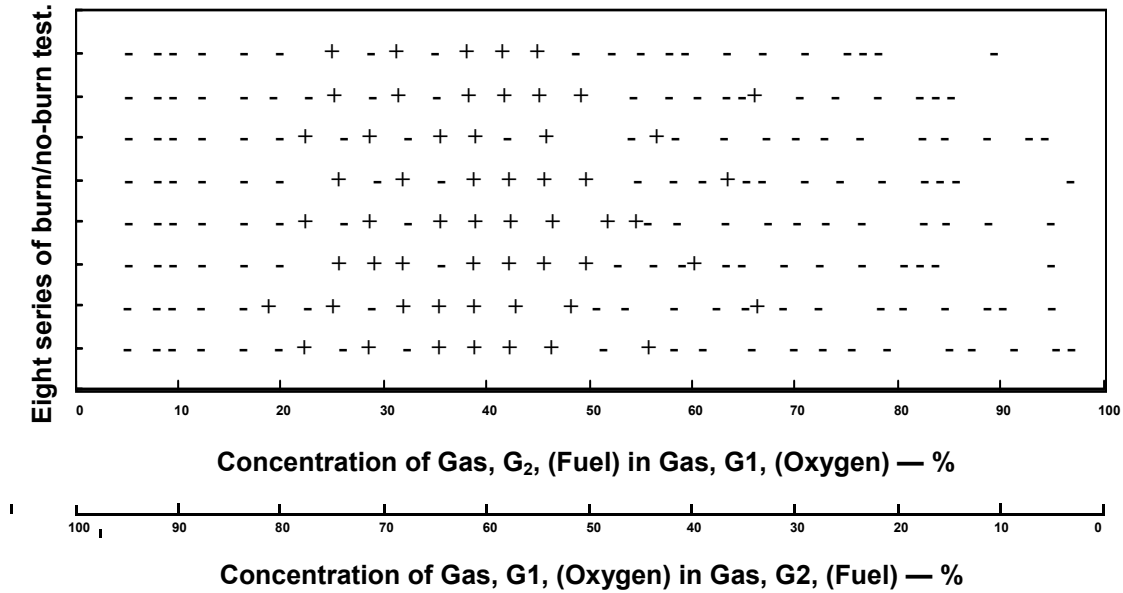


Figure 1—Generic Tests of Two Gases.

ture and exposed to a strong ignition source (often an open flame). However many other vessel geometries are also reported (and can affect the results). If a fire propagates the full length of the tube it is taken as being above the limit. If it does not, even if it propagates a large distance, it is taken as being below the threshold.

However, other data can also affect the interpretation. Self-sustaining combustion requires an equilibrium between the production and dissipation of combustion heat. If a test result produced a fire that progressively decayed for the full 36-inch test length, it *might* be scored as a negative even though it propagated the full length. Similarly, if it burned an appreciable distance with neither acceleration nor deceleration, or if some mechanism produces premature extinguishment, then it *might* be scored as a positive, or at the very least repeated.

To determine the fire limits, one uses a metric such as is shown in Figure 2. In Fig. 2, the data from Fig 1 are replicated and to the left of the figure is the metric: the vertical line with arrows showing its motion to the right. This metric is treated as a filter or screen to separate the positive and negative results. Assume the metric is moved to the right with negative results passing through its pores until it gets to position “A”, about ~18%, where it encounters its first positive result. This concentration is taken as the minimum amount of Gas 2 that can produce a sustained fire. When G₁ is oxidant and G₂ is a fuel, this is called the lower fire limit (LFL) for the binary system of Gas 2 (fuel) in Gas 1 (oxygen). And this is what these data are referred to in the literature for

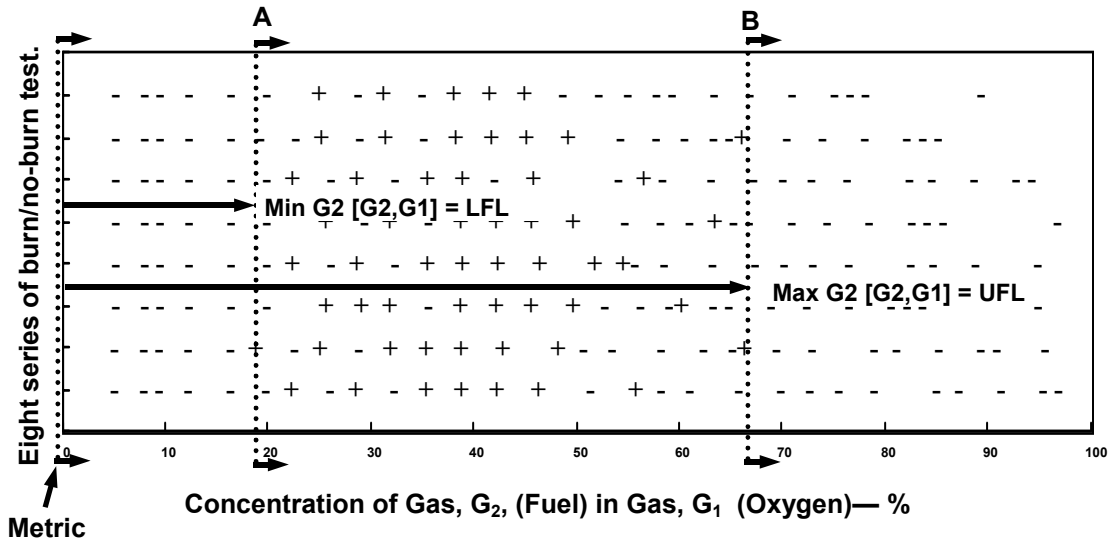


Figure 2—Generic Limit Inference for Two Gases.

a two component system (although there is not a large amount of data for testing in pure oxygen). There does not appear to be a standardized system for designating generic limits, so in this text the LFL will be referred to as the Min G2 (G1,G2), the minimum amount of G2 in the (G1,G2) system. This notation may not be adequate or even desirable for more complex systems, and this is a need worthy of addressing.

If the filter (metric) is allowed to continue moving to the right until it is at the last positive result at point “B” (at ~66%) then this is the greatest concentration of G2 in G1 and is called the Upper Fire Limit (UFL) in the literature, and is the maximum amount of fuel that can yield a sustained fire, the Max G2 (G1,G2). Unfortunately in the literature LFL and UFL data are not always carefully designated as being for a two-component system and data for a three or even four or more component systems may be different.

While, it is common to show the data as fuel being added to oxygen, this is only convention. Figure 1 could have been plotted just as easily with Gas 2, the fuel on the left and Gas 1, the oxygen, on the right. Then the metric would have been determining two different (but functionally related) fire limits. This would have been equivalent to placing the metric to the right of the Figure 2 and moving it to the left to determine the minimum and maximum amount of oxygen in the fuel that just allow sustained combustion, as shown in Figure 3. Namely, the Min G1 [G1,G2] (e.g. Min O₂ [O₂,Fuel]) and Max G1 [G1,G2] (e.g. Max O₂ [O₂, Fuel]).

These latter two fire limits are not routinely published in the literature, perhaps because they can be easily calculated from the prior two limits. To wit:

$$\text{Min G1 [G1,G2]} = \text{Min O}_2 [\text{O}_2,\text{Fuel}] = 100 - \text{Max G2 [G2,G1]} = 100 - \text{UFL}$$

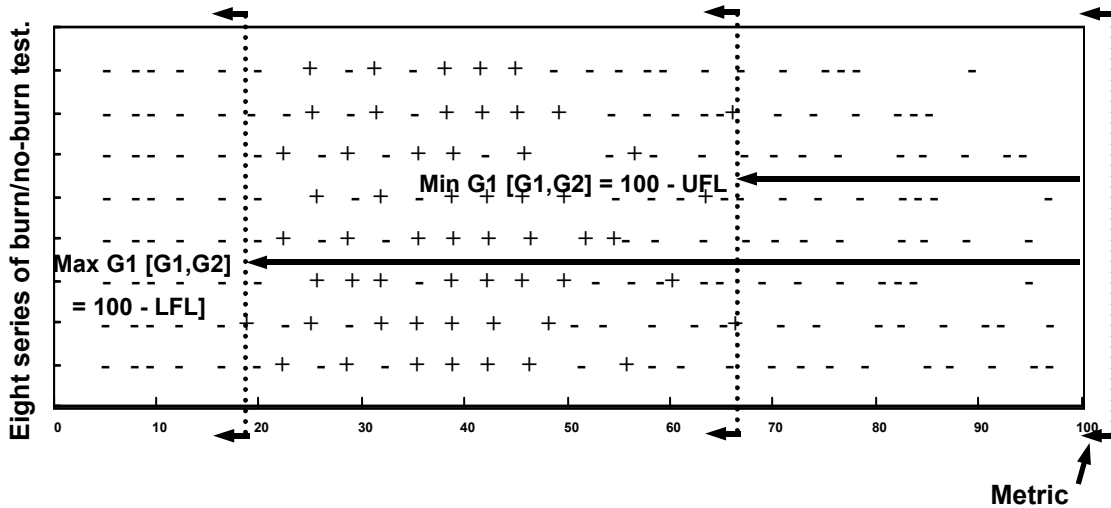


Figure 3—Generic Tests of Two Gases.

$$\text{Max } G_1 [G_1, G_2] = \text{Max } O_2 [O_2, \text{Fuel}] = 100 - \text{Min } G_2 [G_2, G_1] = 100 - \text{LFL}$$

The literature do indeed commonly publish a fire limit: “Minimum oxygen for combustion” but it usually applies to three component systems (that is:

$$\text{Min } O_2 [O_2, G_2, G_3].$$

However, as will be seen later, its value in some (but not all) cases is the same as for:

$$\text{Min } O_2 [O_2, G_2]$$

Things now start to take on complexity. Suppose for instance, that the generic fire limits:

$$\text{Min } G_i [G_i, G_k]$$

and

$$\text{Max } G_i [G_i, G_k]$$

involve mixtures for G_i and/or G_k . This is allowed, as it must be, because in the real world one may encounter mixtures of any gases and one needs to be able to cope with their mutual reactions. Neither mixture is required to be a pure oxidant or fuel or other. Nonetheless, one could simply perform testing as for Figure 1 to determine the relative fire limits for them. In fact, in principle, either might contain two *or more* pure gases yet because of the constancy of the composition would still qualify as a binary system. Furthermore, once you know the percentage of the mixture that yields a fire limit in the greater mixture, you may need to know the limit concentration of a pure constituent of

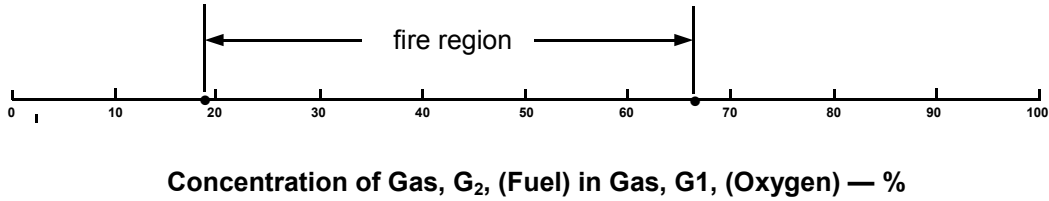


Figure 4—Compact Fire Limit Display.

one of the mixtures rather than the amount of the mixture itself. This is possible to do but not necessarily pleasant.

Today, the preponderance of published fire limits relate to fuel in mixture with air (~21% oxygen, ~79% nitrogen) because so many fire risks are environmental. Although testing may discover the fraction of air in mixture with a fuel that produces a fire limit, if one is operating a system and analyzing its contents, one might need to know the amount of oxygen in the fire-limit mixture rather than the amount of air.

Also, one might be interested in the fire limits of any two gases each of which are mixtures of fuel, oxygen and a non-reactive gas (inert or diluent). However, in a generic sense, the data for any binary system can be shown simply as a scale with the two threshold conditions indicated as in Figure 4 for the example in Figures 1-3..

Figure 4 is a powerful tool. Assume two fuel/oxygen mixtures are of interest: M1 as 10% fuel in oxygen and M2 as 90% fuel in oxygen. One does not need to test these two mixtures to infer their relative fire limits. These two mixtures are a sub-binary

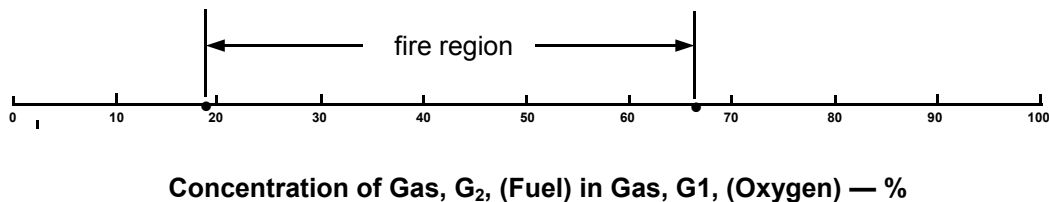
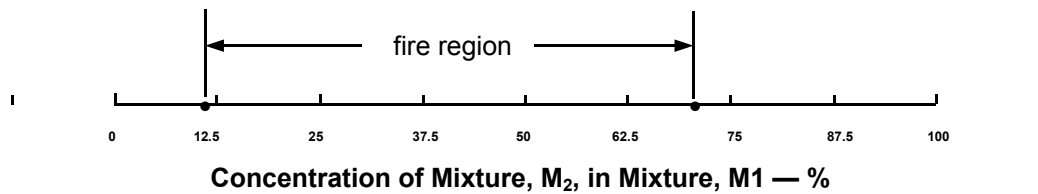


Figure 5—Compact Sub-Binary System.

TABLE 1—*Generic Binary Fire Limits*

Min G2 (G1,G2)	Max G2 (G1,G2)
Min G1(G1,G2)	Max G1 (G1,G2)
Min M2 (M1,M2)	Max M2 (M1,M2)
Min M1 (M1,M2)	Max M1 (M1,M2)

system within the original system.

Figure 5 exhibits the compact display of Figure 4 and adds a second interpretation of the sub-binary-gas system above it. In this case, the mixtures between 0-10% G2 in G1 and 90-100% G2 in G1 are no longer accessible, but everything between 10% and 90% are accessible. So the scale for the upper example is recalibrated from 0-100% Mixture 2, M2, in Mixture 1, M1. This recalibrates the fire limits of 18% and 66% G2 in G1, as 10% and 70% M2 in M1, respectively. No additional testing is required to draw this powerful inference.

As a result, a number of generic fire limits have been defined as listed in Table 1. Among these, those shown in bold-face font are typically tabulated in the literature and the others must be obtained by mathematical or graphical manipulations. Furthermore, the mathematical manipulations required are fairly straight-forward. That is not nearly the case for the next category of ternary systems.

Ternary System Analysis

Whereas common binary systems involve two gases (any two gases) of pure and/or fixed-mixture composition wherein only the relative amounts of each may vary, ternary systems can involve three components and/or mixtures of fixed composition and the relative amounts among them can vary in several ways. Typically, published ternary data are for an oxidant (O), a fuel (F), and a non-reactive (inert or diluent: D) gas, but they can also be for three many-component mixtures of fixed composition.

These many-component mixtures may not be easily categorized as oxidant, fuel or diluent. Nontrivial ternary systems can also be formed of two oxidants and one fuel, two fuels and one oxidant, two diluents and one oxidant/fuel mixture. Some gases may be oxidants in one system and fuels in another. And there are other variations. Some may be null and incapable of producing a fire in any mixture. For example three inert gases would exhibit no fire regions at all.

We shall see that, all complete ternary systems are based upon three binary subsystems. A simple and common published kind can be constructed as shown in Figure 6. In this case, the binary system exhibited in Figures 1-4 has been reduced to a single line at the left between pure G1 (e.g. oxygen), and a pure G2 (e.g. fuel), and with the boundaries for the two fire limits (LFL and UFL) shown as points on the line. Next to it is shown an additional binary test series that involves variables G1 and G2 and a constant fraction of the third (e.g. diluent) gas, G3, equal to ten percent of the original series volume, and the hypothetical fire limits for the series are also indicated with points.

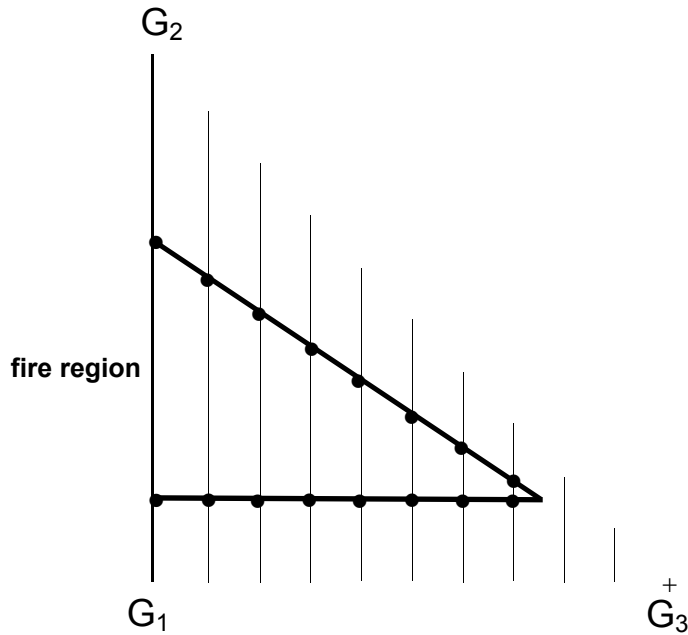


Figure 6—Ten G_1 , G_2 Test Series With Constant Third G_3 Components That Differs in Each Series.

Eight more test series appear to their right each with the constant fraction of G_3 increasing in steps with each series increase equal to ten percent of the original series. Each series is, therefore, shown as being shorter by an amount equal to ten percent of the original test series because each series represents ten percent less of the combined G_1 and G_2 gases. In the tenth series all of the original gases have been replaced with pure G_3 .

Depending upon how well-behaved the gases are, one might now be more than willing to connect the fire-limit dots to interpolate the entire fire region as shown in Figure 6, and thereby to be able to predict behavior between the ten percent increments. Figure 6 is a valid ternary diagram.

Since any two gases can form a binary system, therefore, the pure gases G_1 and G_3 also form another binary system. A line and scale could be placed between them and as one moves from G_1 to G_3 , it would indicate the amount of G_3 in the binary pair. Nine more scales could be formed parallel to the (G_1, G_3) series to represent a constant fraction of G_2 present that increases by ten percent until it achieves 100% G_2 . Similarly G_2 and G_3 form a binary pair. And scales could be placed between them similarly.

Figure 7 removes the lines from Figure 6 related to G_1 and G_3 , leaving the experimental fire-limits in place and creates horizontal lines for the G_1 and G_3 system

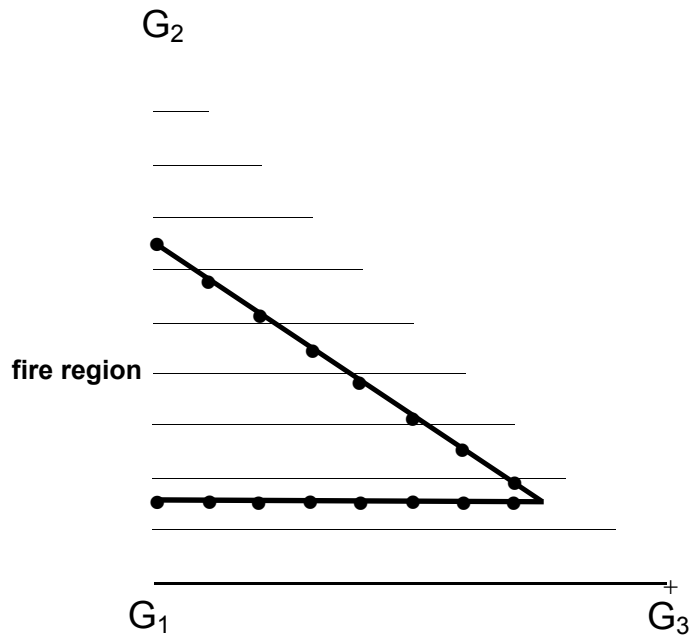


Figure 7—Ten G_1, G_3 Test Series with Constant Third G_2 Component That is Different in Each Series.

with each succeeding line representing the addition of a constant ten percent more of G_2 in its total composition. This is again quite powerful, because experimental tests do not have to be repeated to now know where the fire limits would be found for this series of system variations. Figure 7 is a valid ternary diagram.

Lastly, Figure 8 recognizes that the gases G_2 and G_3 also form a binary system. A new scale could be placed between them and as one moves from G_3 to G_2 , the amount of G_1 in the binary pair in each series of tests increments by ten percent until it achieves 100%. This is a third valid ternary depiction..

Figure 9 combines all three sets of the scales to form a complete valid ternary diagram similar to some that can be occasionally be found in the literature. Figure 9 is just one of many right-triangle formats that are valid. However, right triangles are not the only, nor the most common format that is used. Indeed these data can be morphed (mapped, transformed) into many formats.

Most often one will encounter an equilateral triangle format. Assume the test series of Figure 6 are slanted at 60° as is shown in Figure 10. One could then produce scales like that shown in Figures 7 and 8 at new related angles and when combined with Figure 10, the common complete equilateral ternary diagram of Figure 11 would result.

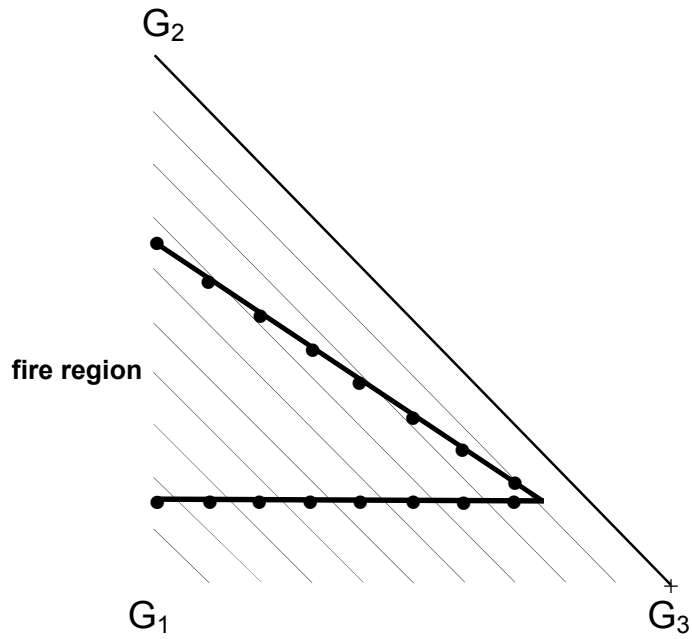


Figure 8—Ten G_2 , G_3 Test Series with Constant Third G_1 Component That is Different in Each Series.

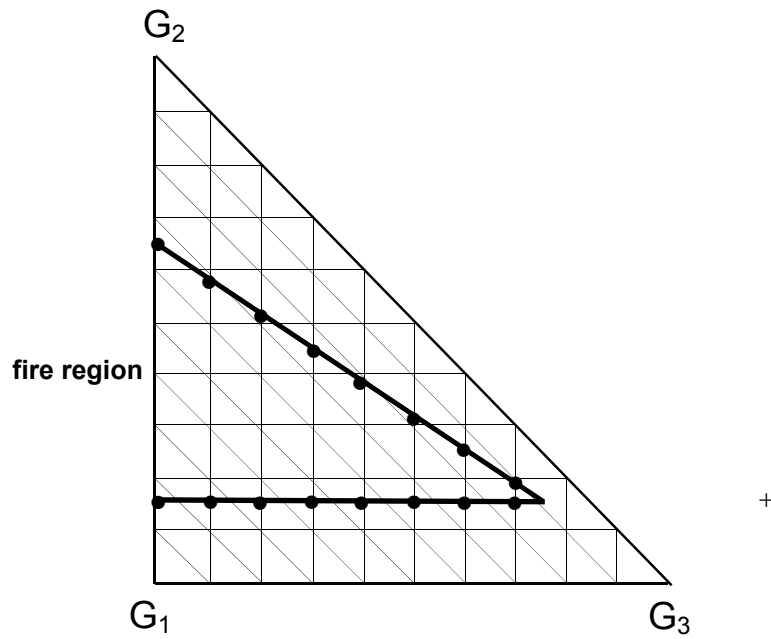


Figure 9—Ten Test Series with Constant Third Component That Increases in Each Series.

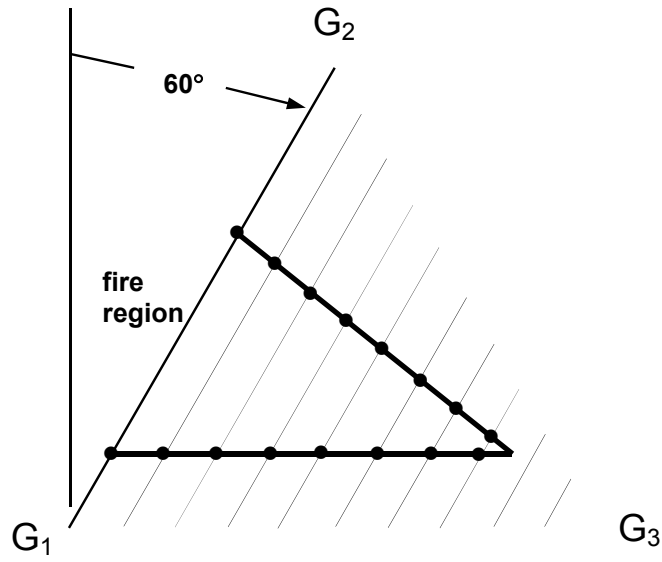


Figure 10—Ten Test Series with Constant Third Component That is Different in Each Series.

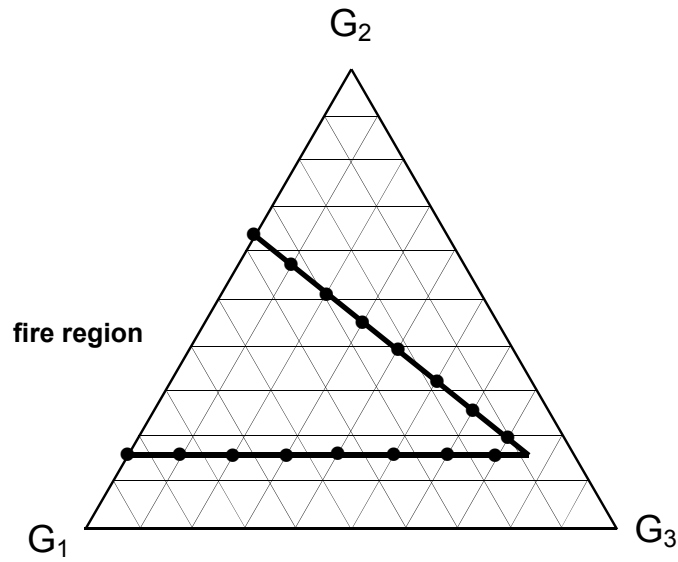


Figure 11—Complete traditional ternary diagram..

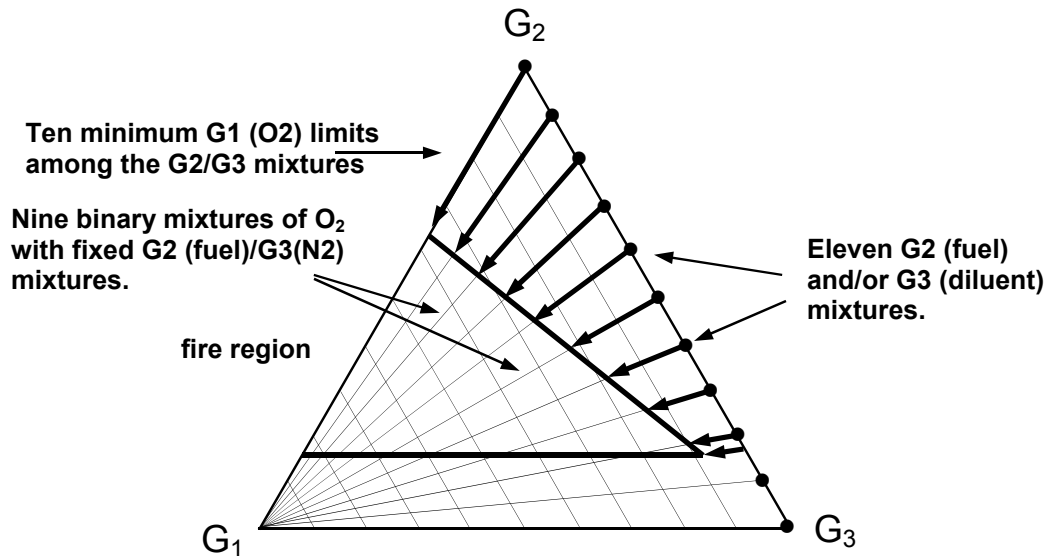


Figure 12—Ten binary-test series to measure *MinG* (e.g. *MinO₂*).

Every possible mixture of the three gases, G1, G2, and G3, is represented by a point on each of these ternary (triangular) plots. Every straight line on a ternary diagram represents a binary system, and all possible mixtures of the two end-point compositions are exhibited, with those possible mixtures that lie in the fire region indicating the location of the fire hazard. Every binary system of interest thus chosen can be analyzed just as for the binary mixtures in the first section of this book.

However, ternary diagrams also allow for the analysis of arbitrary mixtures with up to three variable gases. The most common example of this is when one is going to add a gas to a binary mixture of two other gases that is not fixed. For example: What is the minimum amount of oxygen one can risk adding to *any* mixture of a fuel and diluent before a fire risk presents?

One can address this risk by reducing it to a series of binary cases. In Figure 12, if G1 is taken as oxygen, G2 as a fuel and G3 as a diluent, then one can treat the binary G2, G3 system as a series of mixtures shown as dots on the right leg of the ternary. Then one can examine all of the binary G1 (oxygen) mixtures possible with the series of G2, and G3 gases shown as lines between the binary G2/G3 mixture dots and the G1 apex. Figure 13 plots all of these binary *MinG1* [*MinO₂*] limits. And one can then decide using interpolation what the minimum ternary oxygen limit is and at what binary fuel/diluent composition it occurs. Figures 12 and 13 would indicate that oxygen above 9% could be a risk.

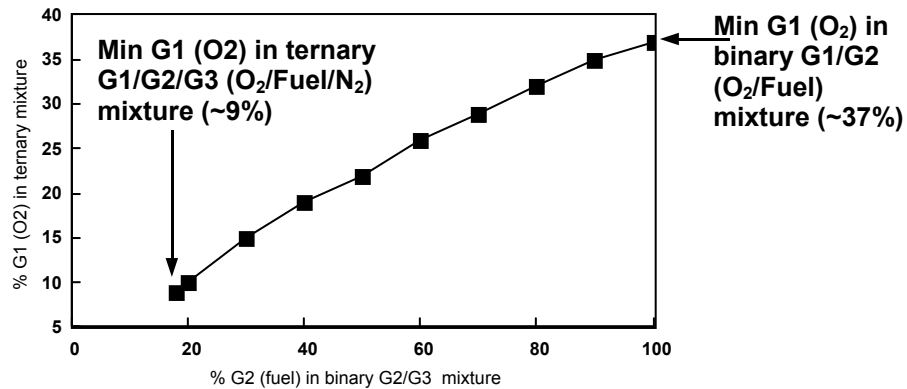


Figure 13—Minimum Oxygen (*G1*) for Combustion in Ten Binary Systems of Oxygen (*G1*) With a Fixed Mixtures of Fuel (*G2*) and Diluent (*G3*).

Unfortunately, it is too easy to misread ternary scales and come up with a wrong answer. Some scales are read left to right others right to left, some top to bottom, other bottoms up. Some on angles.

Notice that constructing the plot of Figure 13 to identify Min *G1* is equivalent to using a movable metric as shown in Figure 14. Being parallel to the right side of the ternary, its *G1* (e.g. oxidant) level is the same everywhere on the metric. If one moves it towards the O₂ apex one can stop it when it first contacts the fire region at any point as shown. This is the Min *G1* or Min O₂ (ternary case). In this hypothetical case, the minimum occurs at the “nose” of the fire region. This is not always the case. Figure 15 exhibits a different fire region shape that shows how a Min *G1* condition can occur when diluent in the mixture is zero (and hence the binary pure-gas Min *G1* and the ternary Min *G1* would be the same value. In comparison, with the gas of Figure 14, the ternary-gas mixture Min *G1* is significantly less than for the Min *G1* of the binary-gas (e.g. O₂/Fuel) mixture. The reason for this is rooted in the thermodynamic and chemical equilibrium properties of the gases and is well suited to a paper on the behavior of diluent gases. Very importantly, this illustrates how *the addition of a diluent, even an inert, gas can increase the fire hazard.*

Similarly, one can return to Figure 14, and observe a Max *G1* metric on the left side, parallel to the right side, that can be moved into the ternary to measure the ternary Max *G1* which in this case is the same value and at the same point as the Max *G1* for the binary pure (*G1,G2*), (e.g. fuel and oxygen) system would be. And again Figure 15, shows how a differently-shaped fire region might identify a Max *G1* for the ternary case that that is located in the interior of the ternary diagram.

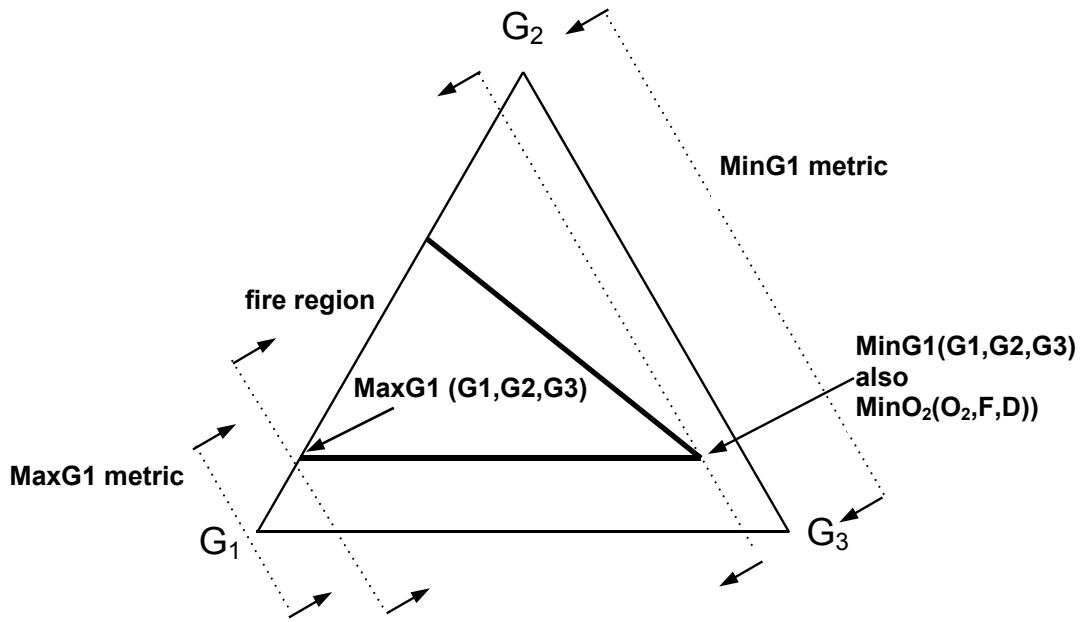


Figure 14—Min and Max G1 metrics.

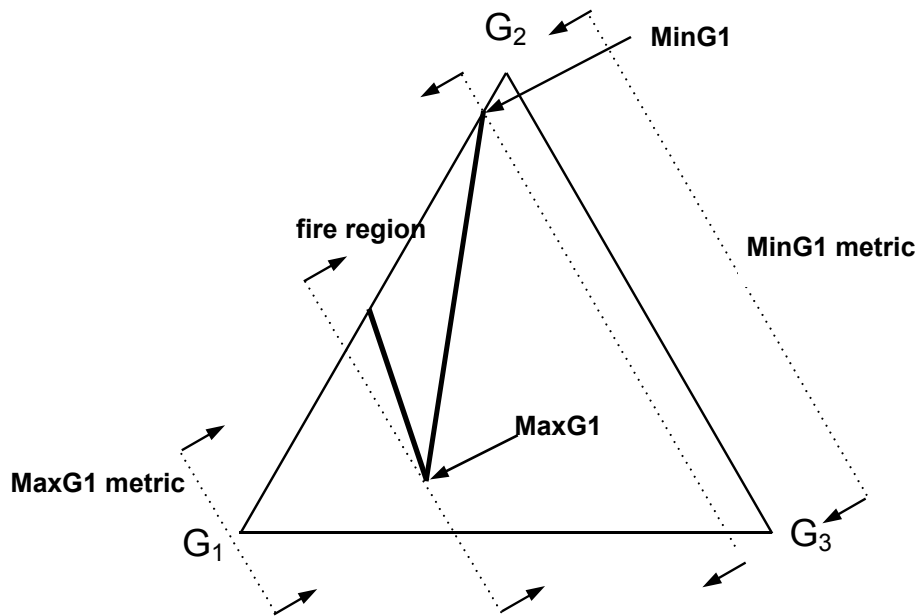


Figure 15—Min and Max G1 metrics with different fire region..

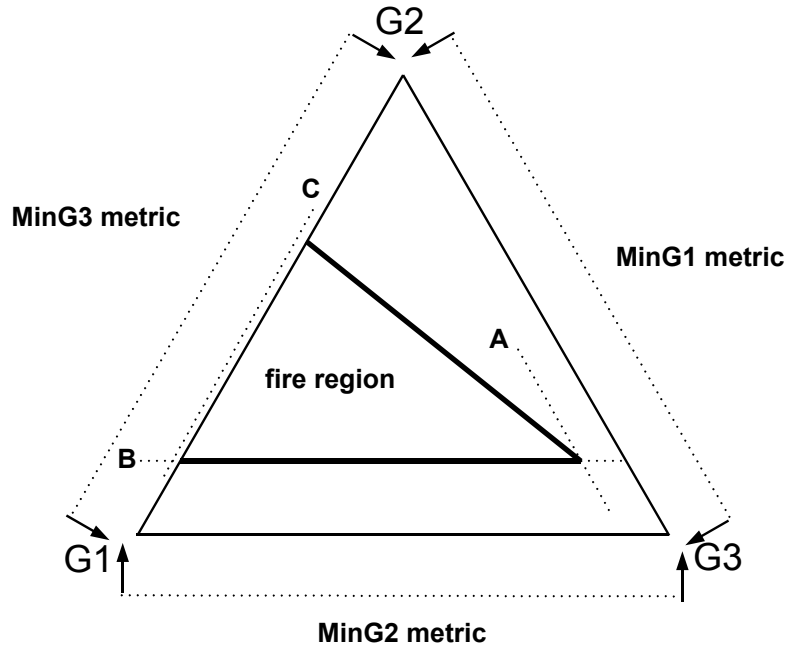


Figure 16—*MinG1, MinG2 and MinG3 measurements at A, B, and C.*

There are three related pairs of ternary metrics of this kind: three Min and three Max. Figure 16 exhibits the three Min metrics. Short dotted lines show where the three metrics would contact the fire region at points A, B, and C. Figure 17 exhibits the three max metrics and short dotted lines show where the three metrics would first contact the fire region at points D, E, and F. These metrics are used this way regardless of the shape of the fire region and whether it is for an oxidant, fuel, diluent or other gases or mixtures.

These metrics now allow for the determination of the generic fire limits listed in Table 2. Notice that only the fire limits in bold face type, for typical G1 as oxidant, G2 as fuel, G3 as diluent, are typically published. Yet any of the others may be equally important to any specific system.

These limits all apply to any system in which all three of the constituents are variable. When you add any one constituent to a system, the other two may be in any combination at all. However, there are other ways in which chemicals can be mixed and that prospect will be covered next.

Variable Binary Streams Feeding Ternary Systems

Consider next the risk involved in altering the amount of a gas, G1 (e.g. O₂), in

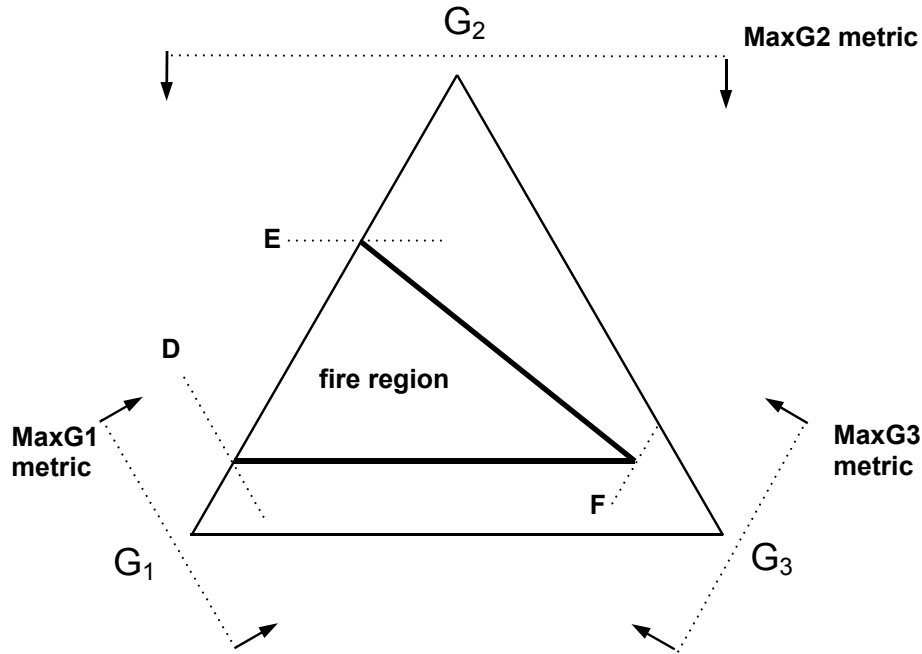


Figure 17—Ten Test Series with Constant Third Component That is Different in Each Series.

TABLE 2—Generic Ternary Fire Limits

MinG1 (G1,G2,G3)	MaxG1 (G1,G2,G3)
MinG2 (G1,G2,G3)	MaxG2 (G1,G2,G3)
MinG3 (G1,G2,G3)	MaxG3 (G1,G2,G3)

Where the boldface limits are most commonly tabulated and:
 MinG1 (G1,G2,G3) is usually tabulated as **MinO₂ (O₂, F, D)**
 MinG2 (G1,G2,G3) is usually tabulated as **MinF (O₂, F, D)**
 MaxG2 (G1,G2,G3) is usually tabulated as **MaxF (O₂, F, D)**

a variable binary (G1,G3) stream, (e.g. O₂ with D) before mixing it into a G₂, (e.g. fuel) stream and asking the question: What is the minimum or maximum amount of G₃, diluent, required in the binary (G1,G3) mixture to avoid a fire risk in any subsequent ternary mixture with the G₂ (fuel gas)? This scenario is schematically shown in Figure 18.

Here again a series of binary analyses similar to those of Figure 12 can be performed. Consider the oxygen/nitrogen binary stream first. All binary mixtures of oxidant and diluent on Figure 12 are represented by the binary line connecting G₁ and G₃.

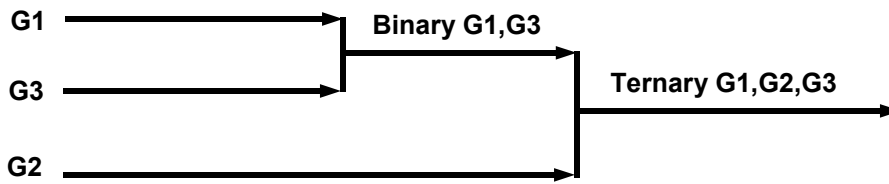


Figure 18—*Variable G1,G3 Stream Forming G1,G2,G3 Ternary Stream*

When a (G1,G3) mixture is chosen, then all binary mixtures of that mixture with the fuel supply are present on a binary line connecting it with the G2 apex. One could choose a series of points between G1 and G3 and connect each point to the G2 apex, then inspect the lines to identify those that contact the fire region and those that don't on either side, if any.

Even better, would be to have a mobile line attached to pivot on the G2 apex at 100% G2 that can be rotated (pivoted) along the G1 to G3 binary range until it just contacts the fire region at any point on each side as shown on Figure 19. Dots (at points A and B) on the G1 (O₂) and G3 (N₂) side indicate the maximum and minimum amounts of oxygen (0% and ~10%) in the binary mixtures that just allow combustion to occur in the ternary mixture.

One could also confront a related system in which a binary mixture of oxidant with fuel is then mixed with diluent, or a binary mixture of diluent and fuel that might then be mixed with oxygen. Hence two more sets of rotary metrics might be used as are illustrated in Figure 20.

As before, here again a mobile pivoting metric can be used. Consider a metric pivoted at the G3 apex. This would rotate from bottom up or top down until it just contacts the fire region. The mixtures indicated on the G1/G2 scale at either point of contact (D or C) with the fire region are the threshold amounts of G1 with G2 before a fire is possible. Similarly metrics pivoted from the G1 apex indicate the threshold amount (at A and B) of G2 with G3 required to present a fire risk. As a result, there are six pairs of rotary metrics as shown generically in Figures 19 and 20.

These twelve metrics analyze three-component gas systems in terms of limits in binary streams among those that feed them. And this illustrates why the three traditional commonly-tabulated fire limits are inadequate. One might need to know any of these twelve assorted generic fire limits. Unfortunately, there does not seem to be a standard notation system, however Table 3 uses the fractional format, $G_X/(G_X + G_Y)$, for the twelve which do not appear to have any significant amounts of tabulated data in the lit-

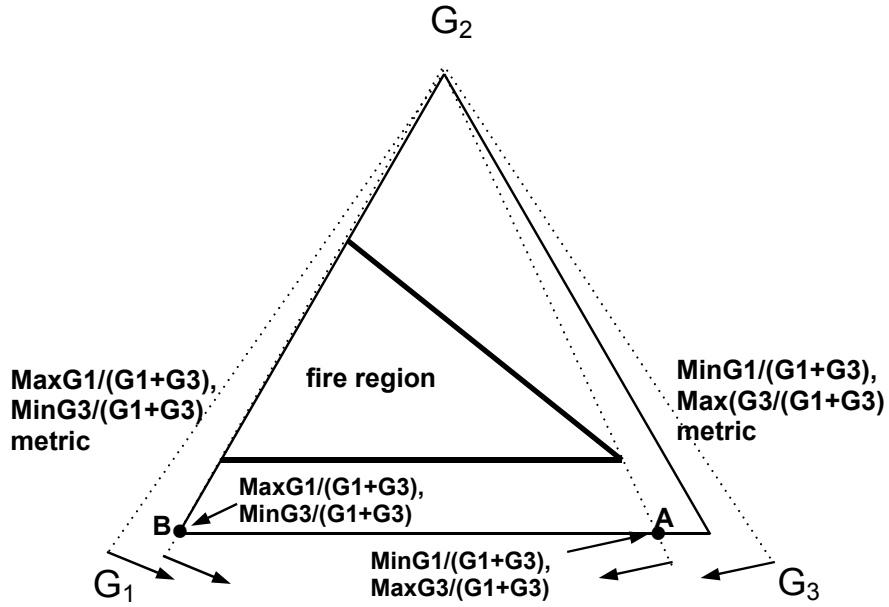


Figure 19—Limits on a Binary Mixture That is Mixed with a Third (Ternary) Component.

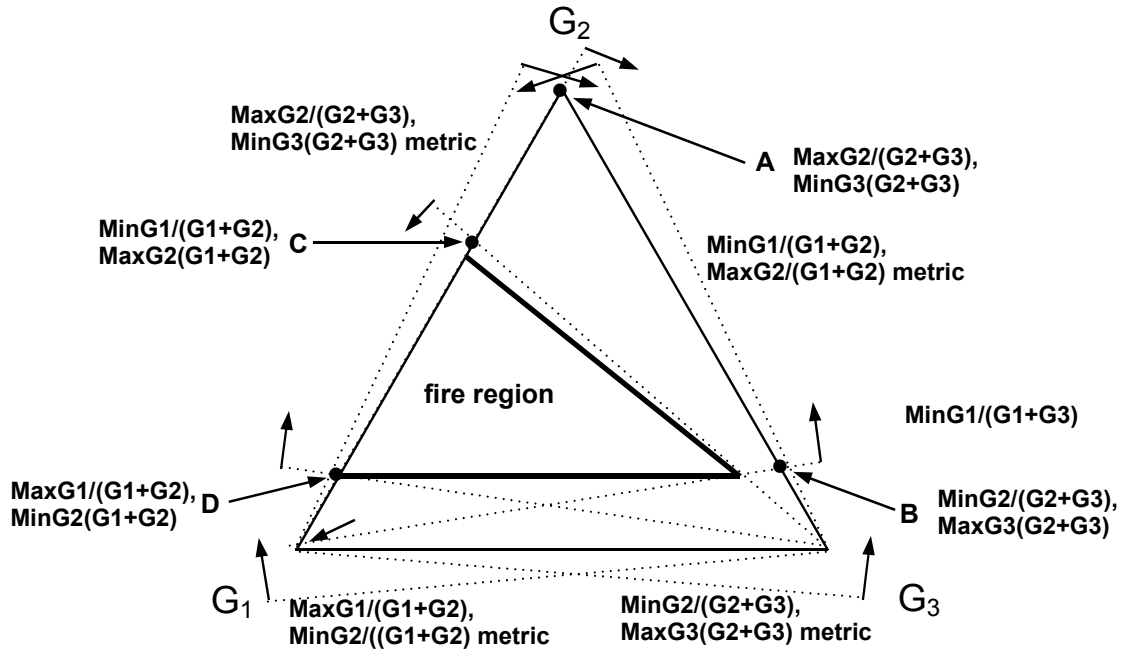


Figure 20—Limits on a Binary Mixture That is Mixed with a Third (Ternary) Component.

TABLE 3—Fire Limits for Binary Streams Feeding Ternary (G1, G2, G3) Systems

<u>G1 + G3 Feedstream</u>	
MinG1/(G1+G3)	MaxG1/(G1+G3)
MinG3/(G1+G3) MaxG3/(G1+G3)	
<u>G1 + G2 Feedstream</u>	
MinG1/(G1+G2)	MaxG1/(G1+G2)
MinG2(G1+G2) MaxG2(G1+G2)	
<u>G2 + G3 Feedstream</u>	
MinG2/(G2+G3)	MaxG2/(G2+G3)
MinG3(G2+G3) MaxG3(G2+G3)	

erature under any notation system.

More Sophisticated Systems: Sub-Ternary Diagrams

The same way that any two gases including gas mixtures can form a binary system, any three gases including gas mixtures can form a ternary system. The same way that picking any two points in a binary system defines a new binary system obeying the same fire properties, when you pick any three points on a ternary system, you define a new sub-ternary system again obeying the same fire properties.

Hence if you take a traditional ternary diagram and designate any three points on it, you have defined a new sub-ternary system. The three points may include apex (pure) concentrations, perimeter (binary) concentrations, or interior (ternary) concentrations. And indeed, if you have a process system that uses these three new apex gas mixtures, then its fire hazards can be analyzed in these exact same ways.

Figure 21 exhibits a parent ternary and five sub-ternary systems. Since there are fire-limit data for the parent ternary, all six can be analyzed with metrics analogous to those examined in Figures 14 thru 20. Indeed each can be transformed into regular normal triangular form. Each of the new apexes are considered to be 100% gas mixture and the scales along the legs are recalibrated to be linear in mixture percentage.

The most common sub-ternaries are those for which pure air (as G1) is juxtaposed with a pure fuel (as G2) and a pure diluent (most often nitrogen as G3). This configuration is shown in Figure 22, page 27. In this case, the mixture point approximating air (~21% oxygen and ~79% nitrogen) is indicated on the bottom side and labeled 100% Air. A binary component line from this “air” point to the G2 (fuel) apex divides the par-

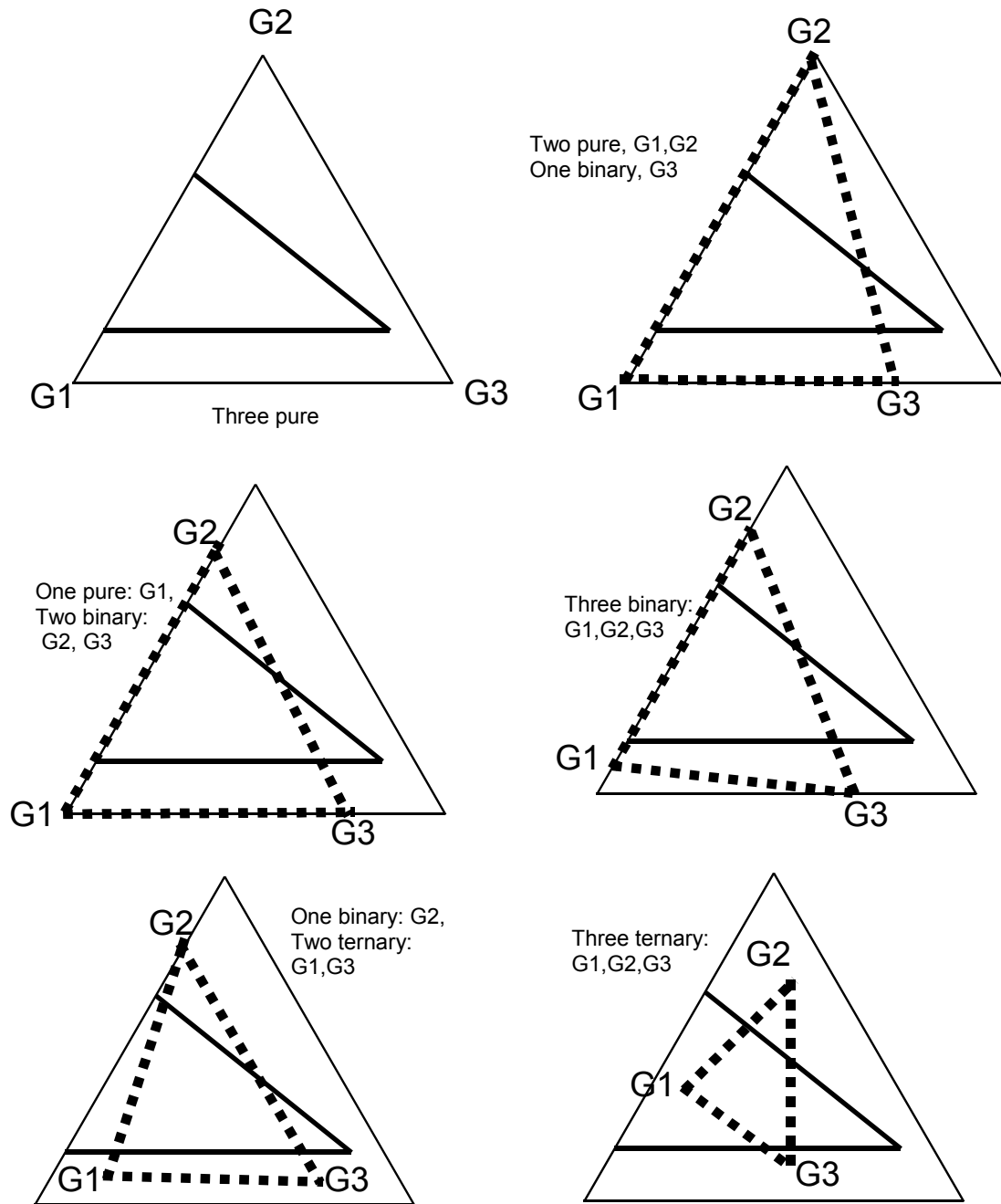


Figure 21—Examples of Sub-Ternary Systems

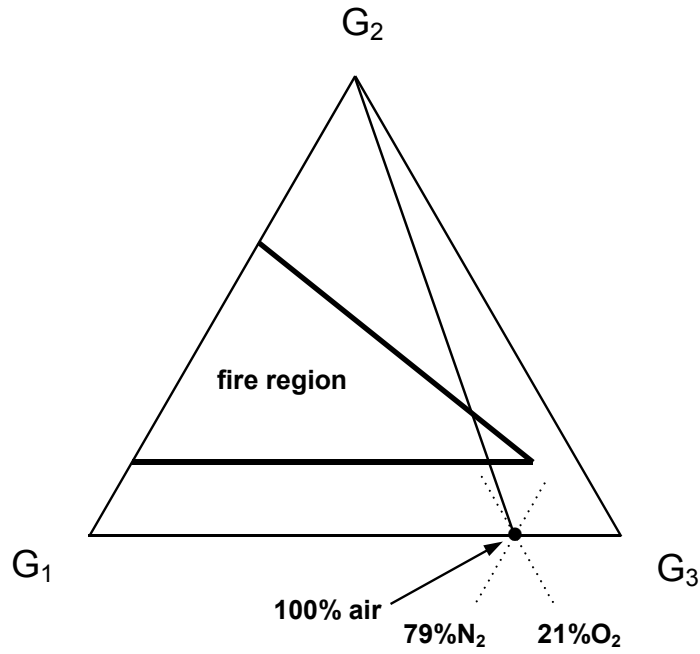


Figure 22—Two Sub-Ternaries (G_1, G_2, Air) and (Air, G_2, G_3), formed by the Binary Mixture: 100% Air = 21 oxygen, 79% nitrogen.

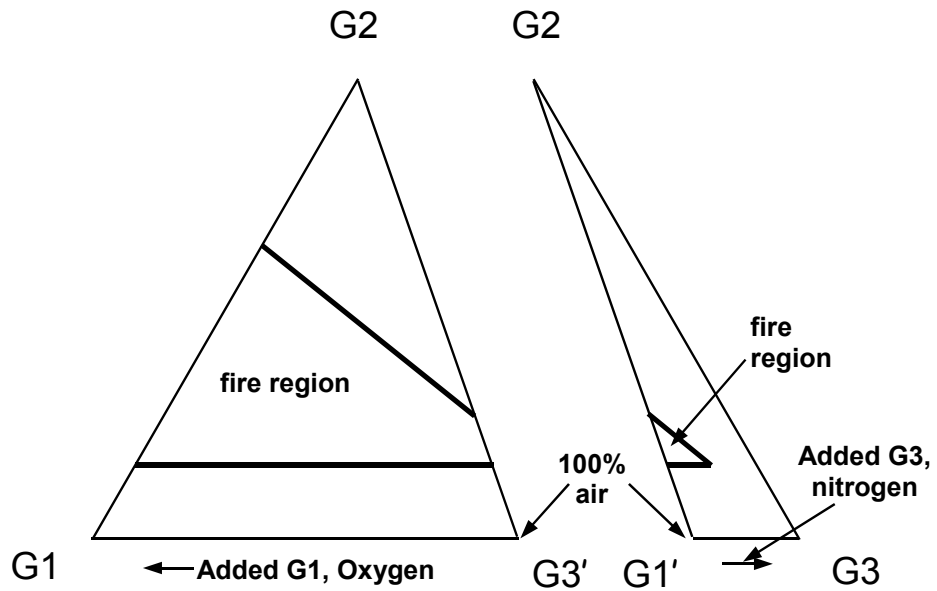


Figure 23—Sub-Ternaries of Figure 20 Separated..

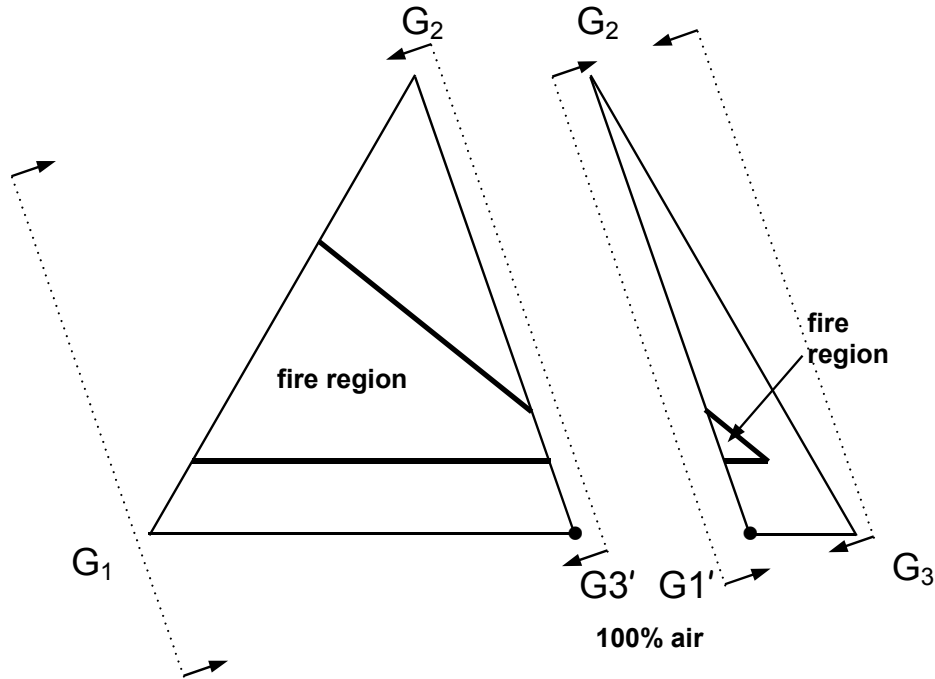


Figure 24—New metrics for sub-ternaries of Figure 23.

ent ternary into two valid sub-ternary systems.

Figure 23, page 27, shows the two sub-ternaries separated. The left sub-ternary has 100% air as its G3' apex. The right sub-ternary has 100% air as its G1' apex. The new scales on the bottom sides would now range (right to left) from 0-100% added G1 (added oxygen which does not include the oxygen in the air) and which relates to 0-100% air (left to right) on the left sub-ternary. The right sub-ternary is scaled from 0-100% added G3 (added nitrogen which does not include the nitrogen in air) corresponding to 0-100% added nitrogen on the right sub-ternary.

Figure 24 exhibits the new metrics that would be used to analyze the portions of the new reference mixture (air in the example). They are parallel to the binary reference-mixture-to-fuel line, which is Air-to-Fuel in the example, or (G3',G2) and (G1',G2).

On the left sub-ternary the new metric measures added oxygen (0-100%, right to left) in the sub-ternary, while to measure total oxygen, one still uses the metric parallel to the parent ternary right side (0-100%, right to left). The metric parallel to the left side measures both air (0-100%) and total nitrogen (0-79%).

On the right sub-ternary, the new metric measures added nitrogen (0-100%, left to right) in the sub-ternary, while to measure total nitrogen, one still uses the metric par-

allel to the parent ternary left side (0-100%, left to right). The metric parallel to the right side measures both air (0-100%) and total oxygen (0-21%).

Similar new metrics form whenever a reference mixture of G1 and G2 or of G2 and G3 are selected to define other sub-ternaries, or whenever interior ternary mixtures are chosen to define alternative sub-ternaries.

The Ternary Nightmare

Keeping track of all these metrics can and does prove confusing, even daunting, for a large fraction of those who need to do these analyses. And there is no small number of people have indicated great aversion to using these diagrams and who avoid it whenever possible. And the writer agrees.

However, once the general and generic tactics of ternary analysis are understood, PC software can provide much simplification by keeping track of which metrics go with each measurement and reporting the assorted concentrations at each point accurately. The next section, examines prototype, proof-of-concept, software that does some of this much-needed simplification for the most common oxidant/fuel/diluent form of ternary analysis and sub-ternaries that may involve user-selectable oxygen/nitrogen mixtures. Hopefully it, or similar software, can someday be expanded and standardized for truly generic use.

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Practicality Considerations in Fire and Explosion Prevention

The writer has background in oxidant fire and explosion prevention and incident investigation. Principal among this experience were the hazards related to oxygen. The quest for mastery of oxygen hazards achieved a watershed moment when ASTM Committee G4 on Compatibility and Sensitivity of Materials in Oxygen-Enriched Atmospheres was formed in 1975.

G4 was born in a climate in which antitrust exemption allowed industrial competitors to cooperate on efforts to address oxygen fire safety. Industry was willing to cooperate because fire related deaths, injuries, equipment losses, interrupted production injured not only its victims but injured the industry as a whole. Everyone and every company was hurt. G4 provided a way to share hard-won lessons in safe use of oxygen, but also to facilitate efficiency and returns to scale that support even better (continuous improvement in) safety practices. The writer has observed inspiring episodes of cooperation through the years.

As a result, G4 defined a target audience as being those “qualified technical personnel” (QTPs), such as engineers and scientists who, by way of education, training or experience know how to apply physical and chemical principles involved in the reactions between oxygen and other materials. G4’s QTPs span a wide range of talents in all these areas..

Fire limit analysis was not the first priority. Selection of materials of construction was most crucial. Then came attention to contamination which is most commonly

oils. However, in the investigation of oxygen hardware fires and explosions, there have been incidents in which gaseous contamination was present, and when that happens, it is important to assess if these gaseous mixtures might have been flammable.

Fire-limit analysis, especially that involving ternary systems, has been onerous even to many QTPs with extreme education. It's been the same for the writer, and many among the G4 QTP community. However, ternary analysis is very powerful when mastered, and so making it practical for use by a greater number of QTPs, even if not every one, is a worthwhile goal. Indeed, much of the onus in ternary fire limit analysis can be abated with PC algorithms that can support analysis (reducing both the onus and error rates) and serve to familiarize and train by taking some of the risk of error out of the process.

In the 1990s the writer began work on an algorithm in Visual Basic language to serve as a proof of concept, as a prototype, and as a starting point for still more ambitious efforts. The algorithm to be described in this section ("use at your own risk" and "as is") is openly available and the source code is open to the public as well, along the lines of the GNU open source software principles.

This algorithm was offered to both ASTM Committee G4 (Compatibility and Sensitivity of Materials in Oxygen Enriched Atmospheres) and ASTM E 27 (Hazard Potential of Chemicals) in the early 2000s and to NFPA 53M (Fire Hazards in Oxygen-Enriched Atmospheres) in the later 2000s. So far none has expressed any interest in pursuit of the approach. Since then ASTM's Governing body has indicated opposition to its Committees distributing software thwarting even distribution of G4's own baby steps: G4Math12.exe an algorithm for computing subtle but nuisance parameters.

This tutorial was prepared to promote and support the prototype materials and document the reasoning of empirical fire-limit analysis. A permanent way to archive copies of the prototype and source code (Visual Basic 4.0 language) and software is being pursued. All are donated to the public domain. And efforts are being made to get them available for a copy fee on Amazon .com, Googlebooks or the like.

FLLAME 1.1

The prototype software was titled FLLAME (Fire Limits for Linearly Afflicted Minds Every (FLLAME) to signify the difficulty in doing manual ternary analysis (and provide a keyword that should be easy to search for on the Internet). A few pages of description will overview the software, however, it has a detailed help file and other operations tools coded into itself. Its primary screen is shown in Figure 25.

The earliest code merely sought to convert and expand on manual ternary scale readings and was drafted in Microsoft 19k Extended Basic using a command line in the

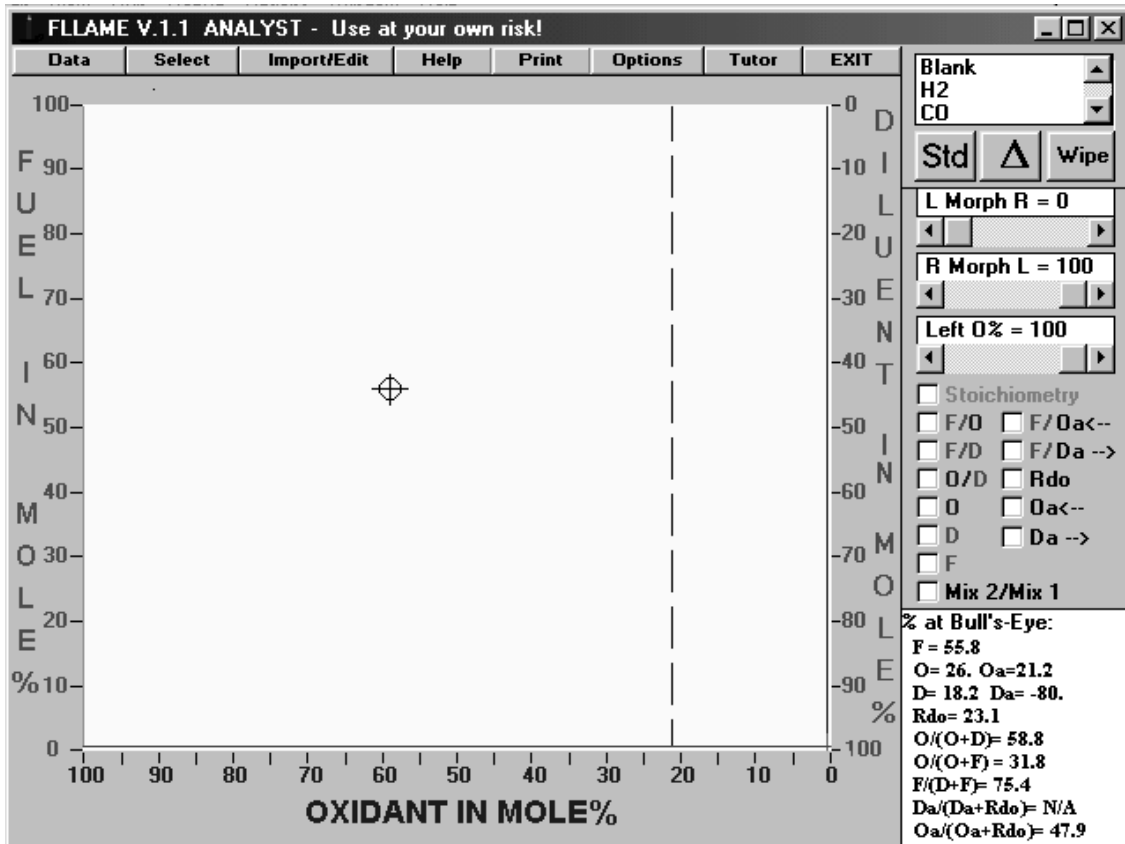


Figure 25—Main operating screen in FLLAME 1.1.

early 1990s. When Visual Basic was introduced for Windows 3.0, the first graphical user code was begun and continued through Version 3.0 by 1997 during the writer’s term of employment. After retirement in 1999, the writer sought and received release of any claims from his former employer (which retained the right to use the existing code itself.). In the 2000s 64 bit PCs were introduced and Visual Basic 16 bit code was not operational on them. Therefore as part of this archive effort, the code was imported into Visual Basic Version 4.0, which is 32 bit code, and will run on 64-bit Windows systems in “compatibility mode”.

This conversion was not seamless and required adjustments to the code but the look and feel was not altered. It is believed to operate essentially as the 16-bit version did with minor adjustments, repairs, upgrades and bug fixes.

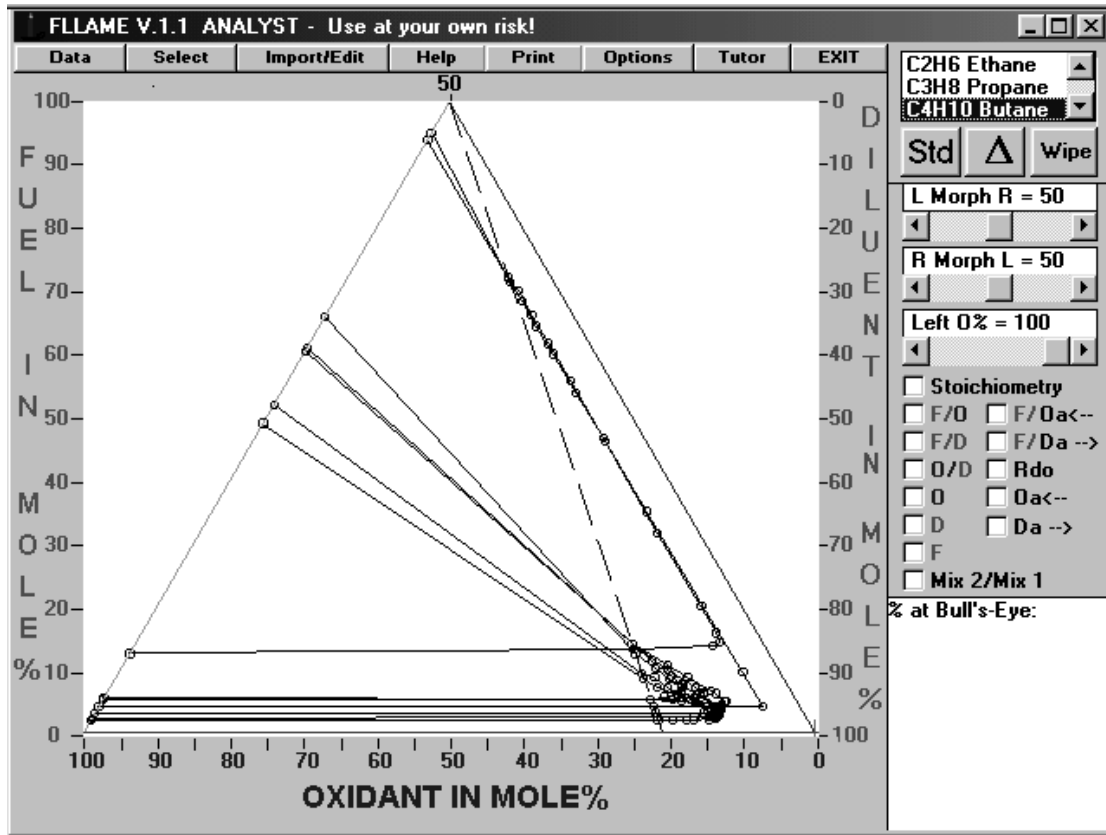


Figure 26—Main operating screen showing all data sets available in FLLAME 1.1.

When launched, introduction screens identify the software and warn about its free use “as is” and “at your own risk” as well as its other status. Then the “analysis” screen, Figure 25 , opens that is the heart of the software. On the right side of Figure 25 is a list box that cites a number of options (blank screen, hydrogen/N₂, carbon monoxide/N₂, methane/N₂, methane/N₂/CO₂, Methane/N₂/Ar, ethane/N₂, propane/N₂, butane/N₂) for which there are literature fire-limit data for ternary systems. Any number of these data can be plotted on to the analyst screen. Figure 26 shows the entire initial V.1.1 set overlaid in ternary format (other formats are possible).

When the cursor is moved onto the analysis area and clicked, the report box (Titled: % at Bulls-Eye) to the lower right reports on what the various compositions are in percentages relating to its location. The variables listed are those specified on an Options screen. The fixed cursor is drawn with a circle around it as a “bulls-eye”. If clicked

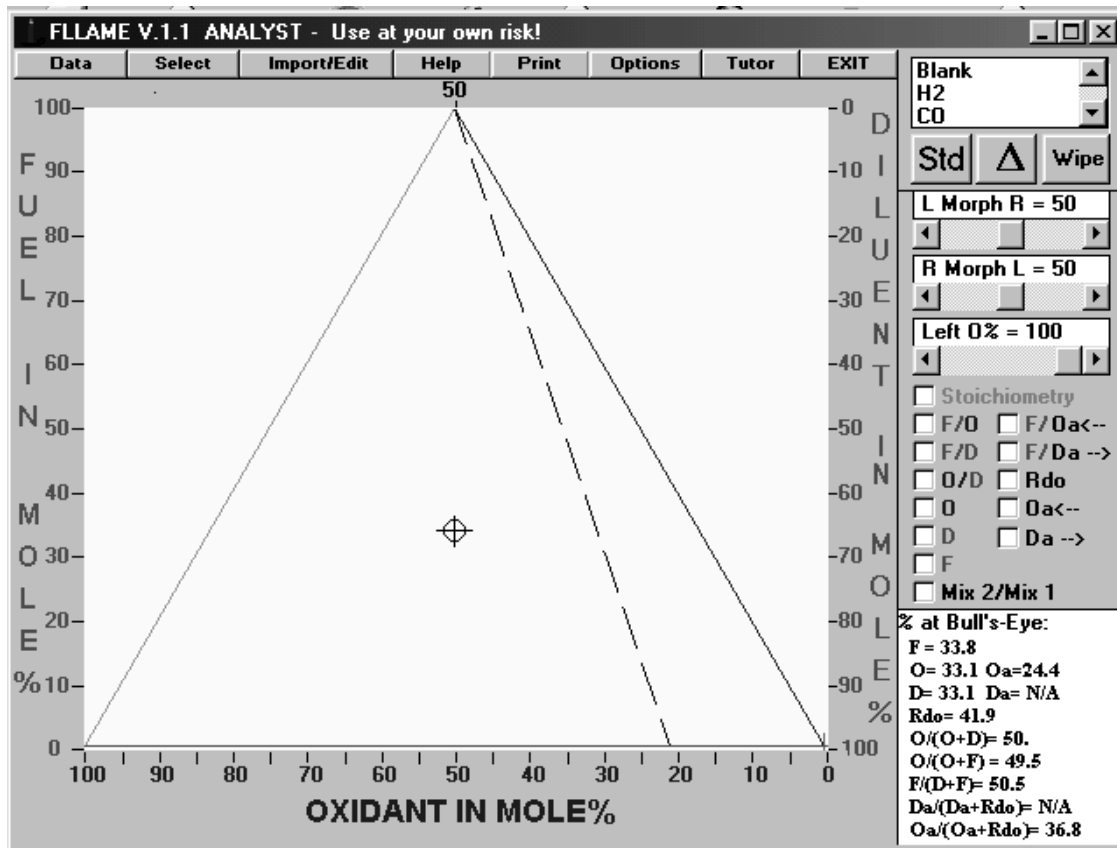


Figure 27—Main operating screen after cursor is clicked.

again in a different position the existing fixed cursor bulls-eye is removed and a new cursor bulls-eye is shown and new report is given. In Figure 27, there is a reference gas (air) shown and the cursor is near the centroid of the figure. It reports: O= 33.1%, F=33.8%, and D=33.1% consistent with being near the centroid. It also cites data for added oxygen (24.4%), reference air (41.9%), the relation of oxygen to the oxygen diluent fraction (50%), the relation of oxygen to the oxygen/fuel fraction (49.5%), the relation of fuel to the fuel/diluent fraction (50.5%), and the relation of added oxygen to the added oxygen plus air fraction (36.8%). Added-diluent is not present to the left of the air line and so added diluent and fractional diluent data are not applicable (N/A).

If any of eleven option buttons on the right are selected, a constant composition (iso-composition) metric will be shown on the analysis area for each of them when the mouse is clicked. If any of these same constant metrics are selected on the options

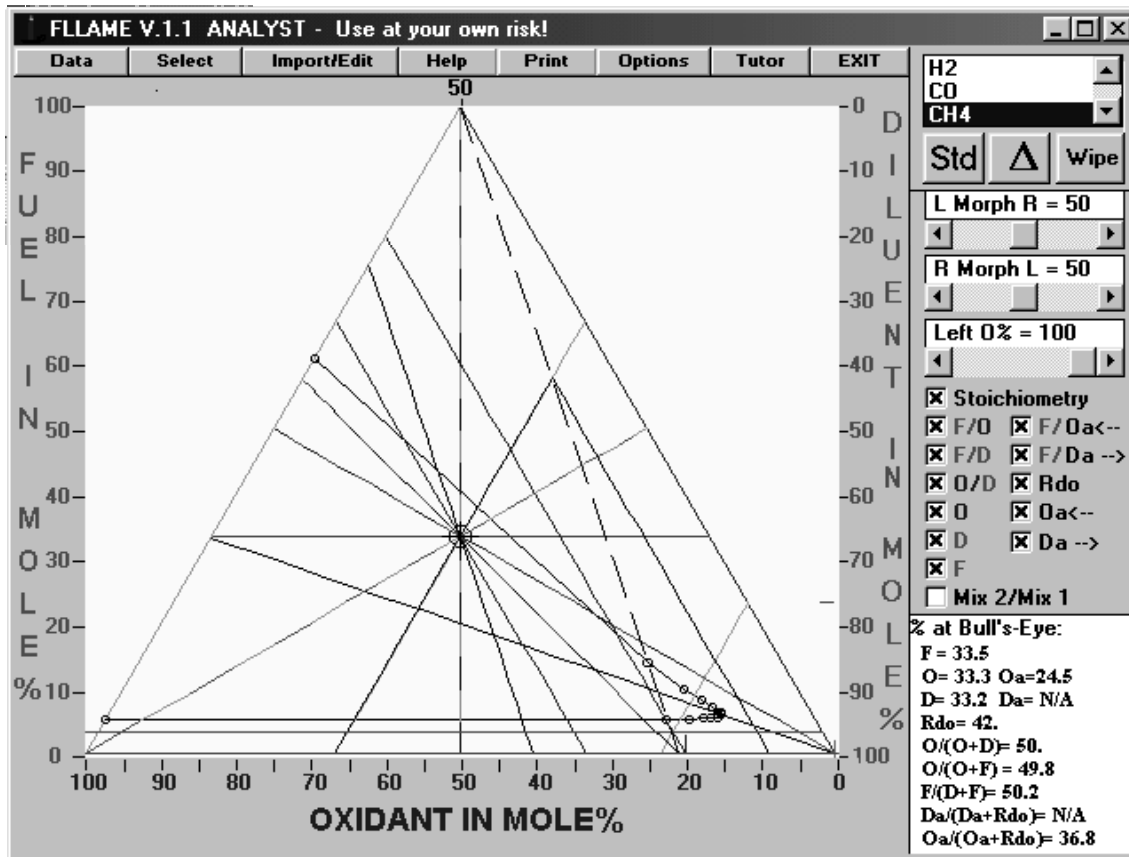


Figure 28—Main operating screen showing stoichiometry and iso-composition curves.

screen, a temporary guideline metric will track the cursor as it moves on the screen to aid placement and analysis. Figure 28 exhibits the main screen with the fire data for oxygen/methane/nitrogen shown along with standard stoichiometry and eleven iso-composition curves. Most curves are color coded to help identify them). The color coding is shown on digital copies of this text.

When a tracking metric contacts the fire region the mouse can be clicked and it will draw the metric and report on the various properties for it. Figure 29 shows how the MinO would be established as 11.5% oxygen. And as has been reviewed in earlier text for the case where an O+D stream feeds into a fuel stream, then the minimum O/D composition that will allow combustion is shown with the metric to the fuel apex as $12.2\% = \frac{O_2}{(O_2+N_2)}$.

The example (proof of concept) software contains many more features: a graphi-

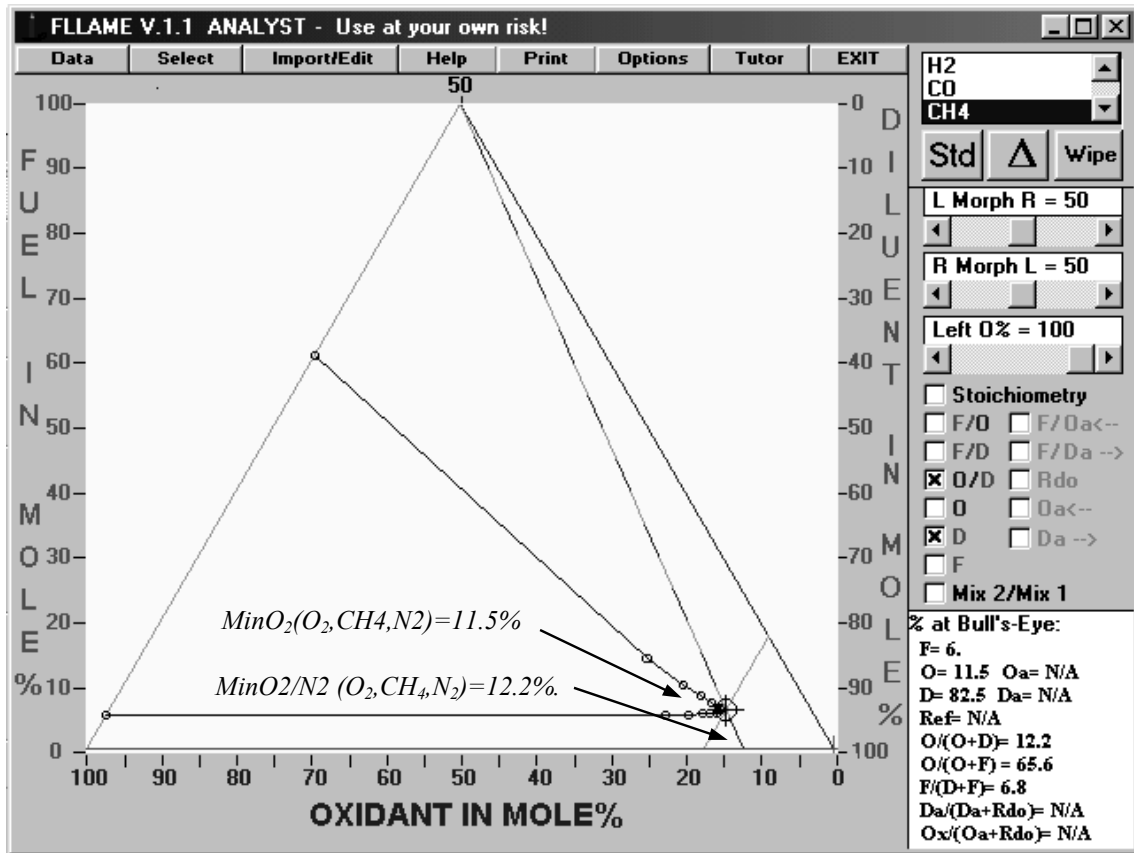


Figure 29—Main operating screen showing two analysis for $MinO_2(O_2,CH_4,N_2)=11.5\%$ and $MinO_2/N_2 (O_2,CH_4,N_2)=12.2\%$.

cal illustration of the gas mixtures at any location, the ability to vary reference gas mixture from 0-100%, the ability to morph and zoom to many different geometrical perspectives, and the ability to import new data.

However, many more powerful enhancements are still possible: the ability to designate reference mixture of (G1,G2), and (G3,G2) systems, The ability to designate arbitrary (M1, M2, M3) systems within a parent G1,G2,G3) system and more.

And if course, the ability to address gas mixtures of four or more gases would also be desirable, and is possible. There does not appear to be any published analysis of four component (quaternary) systems, and they are much more complex. The final section of this text will introduce quaternary analysis superficially without going into great detail.

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Quaternary System Analysis

Quaternary analysis, extends the same approaches as for binary and ternary analysis into the third dimension. One can generically define it as the fire behavior of four arbitrary gases (G1,G2,G3,G4) but instead of producing a fire map that is on a line or a triangle, the standard system is in the shape of a tetrahedron (a pyramid), Figure 30.

The quaternary system includes four “pure” chemicals (G1,G2,G3,G4) or mixtures of chemicals (M1,M2,M3,M4), six parent binary boundary relationships among them [(G1,G2), (G1,G3), (G1,G4), (G2,G3), G2,G4), (G3,G4)], four parent ternary boundary systems [(G1,G2,G3), (G1,G2,G4), (G1,G3,G4), (G2,G3,G4) and the combined interior (contained) parent quaternary system, The amount of testing required to map the three-dimensional fire region of such a system could be huge, and the writer has never actually seen a quaternary system depicted in print, but will finesse one for the sake of introduction in this tutorial.

Within a quaternary system, every binary system is based on any two points, every ternary system based on any three points and they are analyzed with the same set of metrics examined previously. Figure 31 exhibits piping schematics and abbreviated metrics for a binary system Part A, and ternary systems Parts B1 and B2. However, in a quaternary system, new, yet analogous, metrics are required.

Figure 31, Part A, exhibits a binary system that is analyzed with a moving-point-metric that seeks the amount of Gas2 to just enable combustion. Part B1 exhibits three

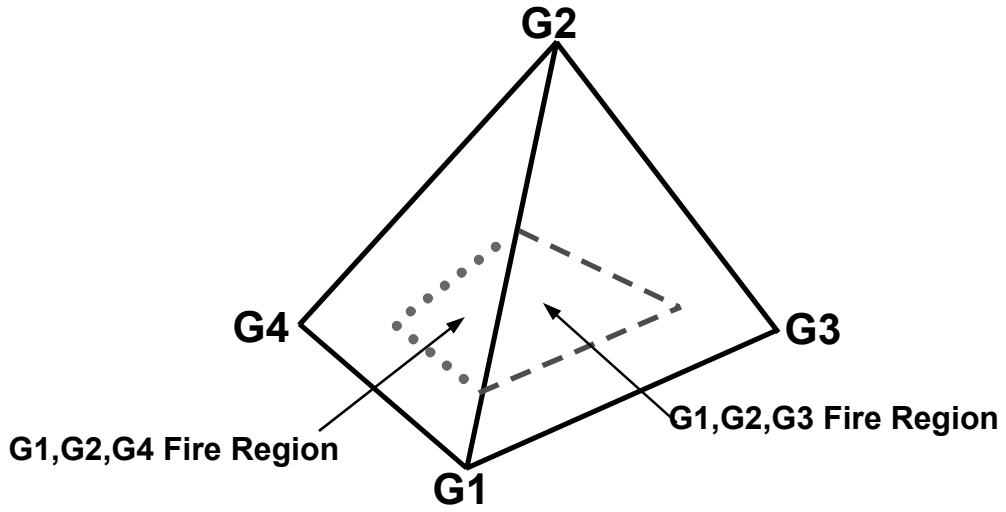


Figure 30—Quaternary coordinate system

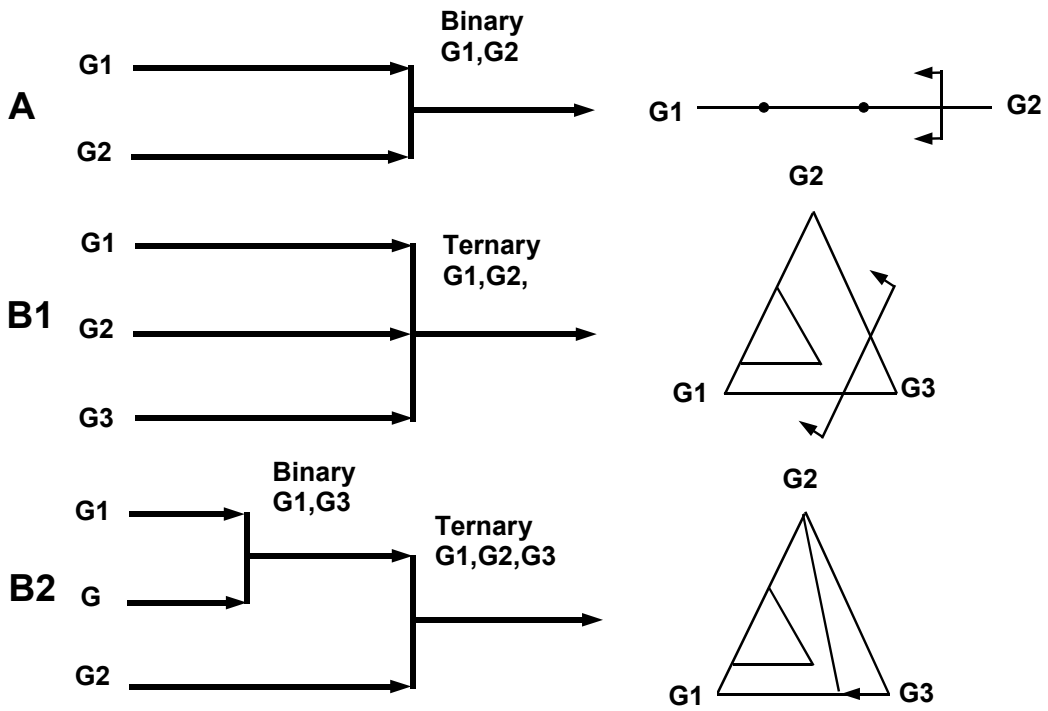


Figure 31—Binary and ternary coordinate systems.

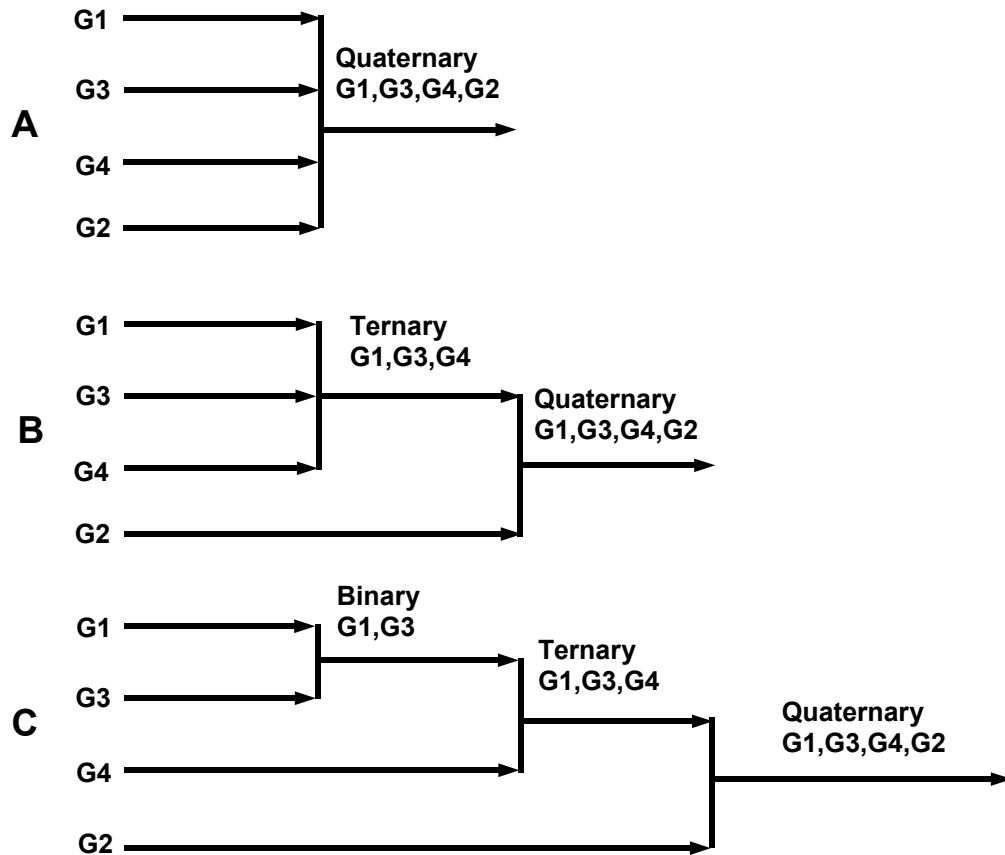


Figure 32—Quaternary coordinate system

mixed gases and how a moving line metric seeks the amount of Gas3 that just enables combustion. Part B2 exhibits a binary system feeding into a ternary system that is analyzed with a moving point to which a pivoting-line-metric is attached to analyze the amount of Gas3 in the binary feed stream that will just enable combustion.

Figure 32 exhibits three analogous systems that specifically apply to quaternary mixtures. Part A relates to four gases that are directly combined where each one may independently affect the combined fire limit. Part B exhibits a ternary system, any one gas of which might affect the fire limit of the quaternary system that it feeds into. Finally Part C exhibits a binary system either gas of which might affect the fire limit of a ternary and quaternary systems that it feeds into in sequence. Examples of each of these three scenarios will illustrate some of the many ways these analysis are performed after a quaternary system is first constructed from a ternary system

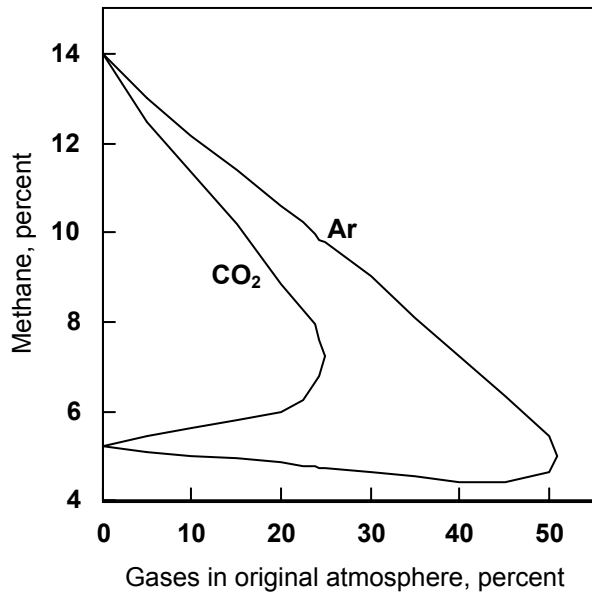


Figure 33—Methane, air fire limit data with CO₂ and Argon Diluents.

Visualizing Quaternary Three-Dimensional Systems

Perhaps quaternary system figures are not common (if indeed there are any), in the literature is because the analysis of even the less-complex ternary data has been disdained so by nearly everyone. However ternary data can be reconfigured to approximate meaningful (but not robust) quaternary systems.

Two-dimensional illustrations of three-dimensional quaternary systems must therefore be finessed. First consider the data of Figure 31 based upon Coward and Jones [1], their page 49. In their Figure 24, rectangular fire-limit data are shown for five three-component mixtures of methane and air combined with five different diluent gases. Among them carbon dioxide and argon give the greatest range of results and are replicated in Figure 33.

These data from Figure 33 can be transformed into two separate ternary diagrams each with a triangular shape. The binary air/methane axis will be the same in each of them and can “hinge” the two figures together so they can be pivoted open to 60 degrees. When viewed along the (methane,air) axis, a quaternary figure (that appears triangular, results that looks as shown in Figure 34.

How will the fire limits look where there are mixtures of the two diluents pre-

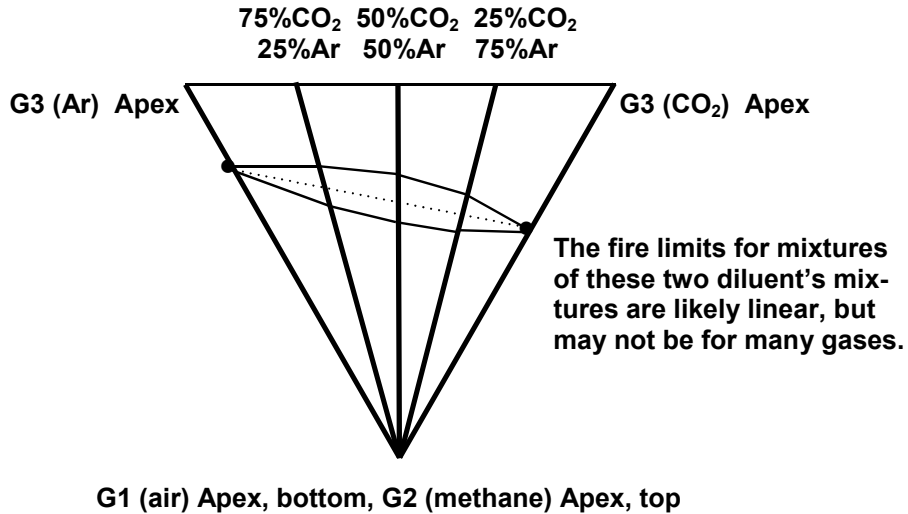


Figure 34—Simplistic quaternary coordinate system

sent between the parent ternaries? There are no published data that indicate if the shape of the curves between the two faces of the quaternary will be the concave, convex or linear as is illustrated on the figure. In the case of carbon dioxide and argon which tend to affect the limit principally with extremes of heat capacity alone, the intermediate mixtures are perhaps likely to be linear combinations, as shown by the dotted line, but for many gases that will not be the case.

A second real, complete, and somewhat more complex quaternary system can be exhibited for a number of gases if one of the apex mixtures is taken as a mixture of two of the other gases. For example, consider a standard (oxygen, methane, nitrogen) ternary diagram Part A of Figure 35 based upon Zabetakis, [2]. The binary system air, methane is indicated in its interior and it can be used to divide the parent ternary into two sub-ternaries: (oxygen, methane, air) and (air, methane, nitrogen). Both of these can be transformed (morphed, zoomed) to produce full-size standard ternaries as shown in Figure 35, Parts B and C, respectively. A fourth ternary diagram for oxygen, air, nitrogen would display no fire regions.

This ternary and its sub-ternaries can now be assembled into the tetrahedron of a quaternary system. The common (Air, CH₄) binary system of Parts B and C can be positioned (hinged) together, as can the (N₂, CH₄) binary systems of Parts A and C, and the (O₂, CH₄) binary systems of Parts A and B. This resulting quaternary, shown in Figure 37, defines interior fire regions and can be analyzed in many ways with a wide range of

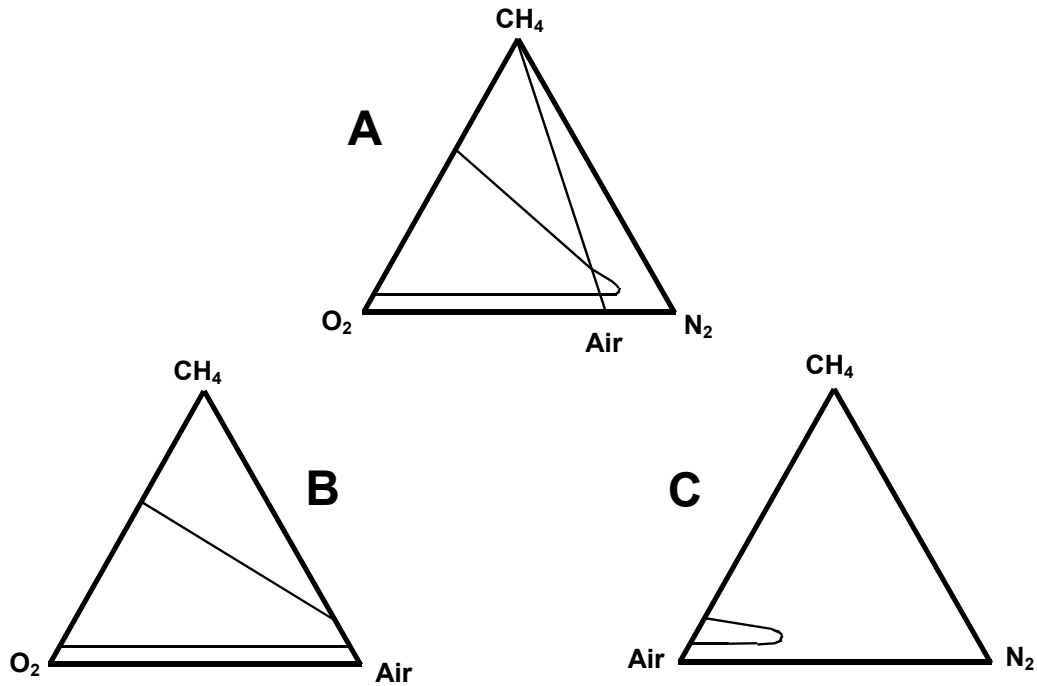


Figure 35— (O_2 , CH_4 , N_2) ternary and, (O_2 , CH_4 , Air), and (Air, CH_4 , N_2) sub-ternary diagrams.

metrics. Just three examples will be covered here to illustrate the analysis for the ternary piping scenarios of Figure 32 parts A, B, and C.

Figure 36 shows how the oxygen/methane/nitrogen ternary is transformed (morphed) and assembled into three equilateral ternaries that form a full-sized quaternary. The point of view is chosen to allow clarity when looking *into* the quaternary volume, looking in to see the G3, or N_2 , apex. In this case the chemical and thermodynamic behavior of the intermediate (air, oxygen) mixtures between the left and right faces will be linear like mixtures of oxygen and nitrogen would be, because they *are* mixtures of oxygen and nitrogen. And the line on the surface between the noses of the two parent ternaries should be straight as shown.

Consider first the scenario of Figure 32, Part A, for which the fire limits related to G3 will be sought. Notice the points on Figure 36 at a, b, and c which are equidistant from the apex G3. The binary system from the G3 apex to point a are all the mixtures of N_2 with the mixtures of N_2 and fuel at point a. That is, it is all N_2 and fuel mixtures at or above the N_2 concentration at point a. Similarly, the binary system from the G3 apex to point b are all the mixtures of N_2 at or above the mixture of N_2 and O_2 at point b. And

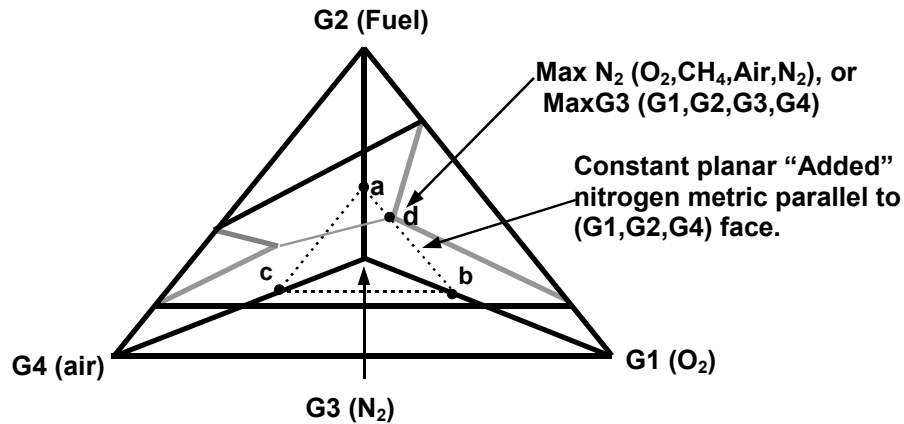


Figure 36—Quaternary system (O_2 , CH_4 , Air, N_2) and, $MaxG_3$ (G_1, G_2, G_3, G_4) metric.

again similarly, the binary system from the G_3 apex to point c are all the mixtures of “added” N_2 at or above the mixture of N_2 and Air (which contains non-added nitrogen) at point c.

Finally, the dotted triangle formed by connecting points a, b, and c therefore represents all the mixture of the mixture at point a with that of b, and all the mixtures of point b with that at c, and all the mixtures of point c with that of point a. Consequently all of the interior points of the triangle formed by point a, b, and c represent all possible mixtures with the same constant amount of nitrogen. The points contained in the sub-tetrahedron formed with the pure-nitrogen apex, G_3 , represents all mixtures with nitrogen equal or greater than the common value at points a, b, and c.

This triangular metric, equidistant from G_3 and parallel to the (G_1, G_2, G_4) ternary side, at all points can be moved away from G_3 until it contacts any point on the three dimensional fire region which it first does at point d. That metric establishes the maximum amount of nitrogen below which fire is a possibility, the $Max N_2$ (O_2, CH_4 , Air, N_2).

In the case of Quaternary system scenarios as in Figure 32 Part A, metrics for Min or Max apex gases are planes parallel to the face opposite the apex for the gas of interest. There are eight of them in all.

In Figure 32 Part B there is a three-gas mixture feeding into a fourth gas. Figure 37 illustrates one instance of this analysis again involving the effect of nitrogen, G_3 , on the fire limit. Once again points, b and c, are shown that represent mixtures of G_3 with either G_1 or G_4 , containing the same amount of G_3 , (N_2). Points on the dotted line be-

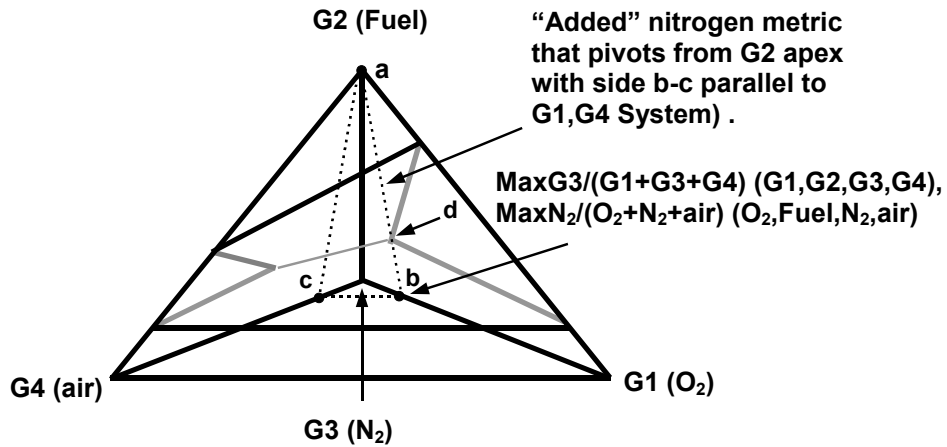


Figure 37—Quaternary system (O_2 , CH_4 , Air, N_2) and, $MaxG3/(G1+G3+G4)$ ($G1,G2,G3,G4$) Metric.

tween b and c represent all mixtures of those mixtures and have the same G_3 , N_2 fraction. The triangle defined by b, c and the G_3 apex represents all the mixtures of G_3 with G_1 and/or G_4 that can form and be fed into the G_2 stream with G_3 concentrations equal to that of b and c or up to pure G_3 . The surface defined with the dotted lines between b and a and between c and a, define all the mixtures that can be formed with the concentration of G_3 in b and c or up to pure G_3 or pure G_2 gas. This metric (triangle a,b,c) pivots at the G_2 apex away from the G_3 apex and at point of first contact (point d) with the fire region defines the maximum amount of G_3 in the ternary stream that will just allow combustion in the quaternary stream. In this case, $Max G3/(G1,G4)$ ($G1,G2,G3,G4$) is established upon first contact (d) when the binary $G3/G1$ mixture is at point b.

And this brings us to the final example: that of the scenario Part C of Figure 32. In this case the gas G_3 forms a binary mixture with gas G_1 , that mixture of which forms a binary mixture with gas G_4 , that ternary mixture of which forms a mixture with G_2 . What is the maximum G_3 that will just allow combustion?

Figure 38 exhibits the metric used for the analysis. Again there is a point b shown representing a binary mixture of G_3 with G_1 in the first step. In this case, the next gas added to the mixture is G_4 and so there is a dotted line shown between point b and point c. This dotted line from b to c, represents all of the possible mixtures that can then mix into the G_2 stream that contain nitrogen at or above the amount at point b. The dotted lines drawn from points b and c to point a, establish a planar surface that represents all the possible mixtures of the gases between points b and c with the G_2 gas. Clearly this plane, as shown, passes into the fire region at many points and therefore is

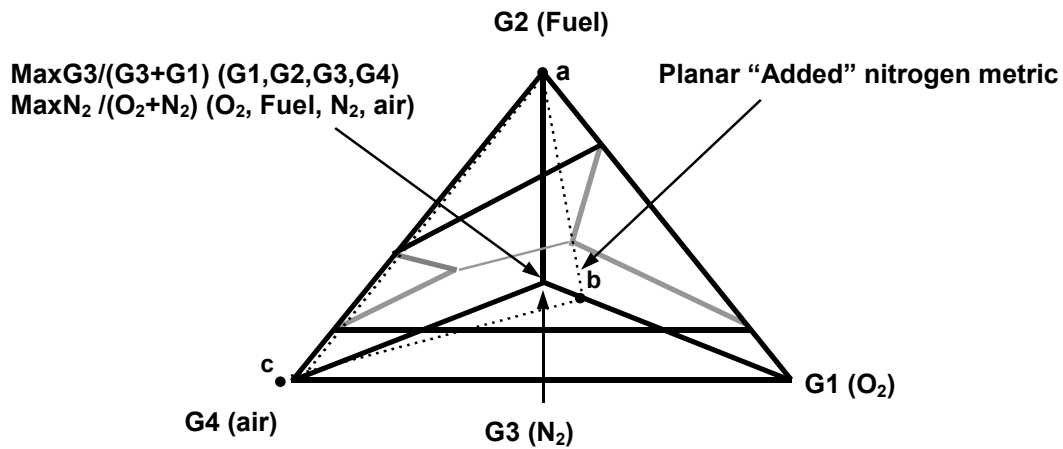


Figure 38—Quaternary system (O₂, CH₄, Air, N₂) and, $\text{MaxG3}/(\text{G1}+\text{G3})$ (G1,G2,G3,G4) Metric.

within the respective fire limits. Indeed, notice that even if the point b is moved back to the G3 apex representing pure nitrogen, the metric still passes through the ternary fire region on the (G2, G3, G4) face. *There is no upper limit* for nitrogen in the (G3,G1) binary stream that will just allow combustion in the Figure 32, Part C scenario. Combustion is possible for the entire range 0-100% nitrogen in the binary (G1,G3) stream.

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Closure

Fire limit analysis is crucial to both the safe design of many systems and to the nagging incident investigations of systems that may have been lacking in design. Some feel all accidents *can* be prevented but not all *will* be prevented.

These analyses include the binary and ternary practices examined in detail, and they do not end with quaternary systems that have been sampled. However, all higher-order systems are beyond the third dimension. Their names might go something like the sequence: penternary, hexternary, hep-ternary, oct-ternary and so forth. All could be useful provided they could be reduced to a simplified PC software.

These analyses are found occasionally in the pertinent literature [4-7] and are something that is smack in the bailiwicks of consensus Committees like ASTM G4, ASTM E-27, NFPA Committee 53M, and others. These committees do not seem to be working to standardize anything like this at present and are urged to begin. These analyses are presently attempted by only the hard-core of cognoscenti. But this tutorial hopes to make them practical to a bigger user base with the promotion and introduction of simplifying PC software.

This tutorial introduced the methodology and presented one prospective approach (a proof of concept) for a PC-based prototype program that might aid analysis and education, and contributes both to the public domain as a foundation for future effort. The prototype software and source code are freely offered for public use (as is, use at your own risk basis) in the fashion of the GNU Open source software license.

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