





# Calculate Flammability Limits Using Process Safety Office® SuperChems Expert™

An ioMosaic Corporation White Paper

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# IOMOSAIC CORPORATION

# Calculate Flammability Limits Using Process Safety Office<sup>®</sup> SuperChems Expert<sup>TM</sup>

Process Safety and Risk Management Practices

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## **1** Introduction

This paper presents a general method for the estimation of flammability envelopes for chemical mixtures containing gases, liquids, and/or solids based on chemical equilibrium. The impact of mixture initial temperature, the presence of diluents and elevated system pressures are implicitly accounted for. The performance of this method is tested against much of the experimental data reported in the literature for systems containing a wide range of chemicals from CHNO compounds to compounds containing sulfur, phosphorus, silicon and halogens.

This method presents a very useful and accurate approach for assessing the flammability envelopes of mixtures where no experimental data is available, and to guide experimental flammability testing work. <sup>1</sup>

## 2 Flammability Limits

Figure 1 summarizes thermochemical estimates of flame temperature for a mixture of methane and oxygen at 1 bar and 25 C. We observe from Figure 1 that both the lower and upper flammability limits occur at a temperature of around 1500 K. We also observe from Figure 1 that the lower flammability limit (LFL) and the upper flammability limit (UFL) do not change significantly over a 500 degrees window. The calculated LFL varies from 3 % at 1000 K to 4.8 % at 1500 K. The calculated UFL varies from 60 % at 1000 K to 68 % at 1500 K.

At the reported literature flammability limits (LFL of 5 % and UFL of 60 %), the methane-oxygen system has a threshold theoretical flame temperature of 1500 K. At this temperature, the combustion reaction is able to generate enough heat to produce a self-sustaining (propagating) reaction.

Many other systems of interest have a similar behavior. Table 1 summarizes estimates of calculated theoretical flame temperatures at both LFL and UFL limits for a variety of chemicals. Most calculated flame temperatures are between the range of 1000 and 1500 K. For most organic chemicals, the flammability limits in air can be approximately related the the stoichiometric limits in mole or volume percent:

$$LFL_{25} \simeq 0.545 \text{ CST}_{25}$$
(1)  

$$UFL_{25} \simeq 4.8 \sqrt{CST_{25}} = 6.5 \sqrt{LFL_{25}}$$
(2)

The stoichiometric limit can be obtained by finding the composition in air that yields the highest theoretical or adiabatic flame temperature. For the combustion of methane in air, the above equations yield:

$$LFL_{25} = 5\%$$
(3)

$$\text{CST}_{25} \simeq \frac{5}{0.545} = 9.17\%$$
 (4)

$$\text{UFL}_{25} \simeq 6.5 \ \sqrt{5} = 14.5\%$$
 (5)

<sup>&</sup>lt;sup>1</sup>This paper is an updated version of what appeared in [1].

### 2 FLAMMABILITY LIMITS

Compound	LFL(vol %)	UFL (vol %)	$T_{LFL}$ (K)	$T_{UFL}$ (K)
Carbon monoxide	12.50	74.00	1394	1268
Methane	5.00	15.00	1481	1774
Acetylene	2.50	100.00	1268	2831*
Ethylene	2.70	36.00	1370	1216
Ethane	3.00	12.50	1534	1399
Propylene	2.00	11.00	1431	1444
1-Butene	1.60	9.30	1479	1254
1-Hexene	1.20	9.20	1583	1091
CnHn (Mw=200)	0.50	4.70	1543	1000
Hydrogen sulfide	4.30	45.00	1045	1383
Ammonia	15.00	28.00	1632	1825
Ethanol	3.30	19.00	1492	1041
Acetone	2.60	12.80	1541	1229
Dimethyl ether	3.40	27.00	1565	980
Ethyle acetate	2.20	11.40	1571	1047
Acetaldehyde	4.00	36.00	1552	977
Trichlorosilane	7.00	75.00	1260	915
Methyldichlorosilane	3.40	54.00	1543	847
Methyltrichlorosilane	5.10		1602	

Table 1: Calculated theoretical flame temperatures for a veriety of chemical compounds. Combustion occurs in air at an initial temperature of 25 C and an initial pressure of 1 bar.

\* implies self decomposition

The idea of using a threshold theoretical flame temperature to estimate flammability limits is not new (See Stull [2]). Recently, Melhem and Shanley [3] demonstrated that calculated adiabatic reaction temperature (CART) is an effective screening tool in evaluating the reactivity hazard potential of many classes of chemical compositions including CH, CHO, nitro, organic nitro, nitrogen compounds (other than nitro and organic nitro compounds), nitrates, redox compositions, etc.

Many combustion reactions leading to the formation of carbon dioxide and water have "freeze-out" temperatures on the order of 1400 K. This "freeze-out" limit is most likely related to the minimum temperature required for carbon monoxide to propagate a self-sustaining flame which is around 1400 K. The concept of a "freeze-out" temperature can be used to estimate flammability limits for multicomponent mixtures w/wo diluents and can be used to assess the impact of initial mixture temperature and/or pressure on flammability limits.

Figure 2 illustrates the use of constant theoretical flame temperature to estimate the flammability envelope for a hydrocarbon mixture where water is the diluent. As shown by Figure 2, the selection of a lower temperature limit, such as 1200 K, results in a wider flammability envelope, a conservative estimate for hazard prediction. With few exceptions, the majority of reported experimental flammability limits in air correspond to adiabatic flame temperatures in the range of 1000 to 1500 K.

#### 2 FLAMMABILITY LIMITS



Figure 1: Computed theoretical flame temperature for the system methane-oxygen at 1 bar and 298 K using Process Safety Office SuperChems

Figure 2: Impact of temperature limit on flammability envelope



Most compositions yielding adiabatic temperatures lower than these values will not continue to react even if initiated at higher temperatures [4].

The estimation of theoretical flame temperature for vapor and multiphase systems is easily accomplished using direct minimization of the Gibbs free energy. Using a threshold theoretical flame temperature value, of say 1000 K, we can estimate the flammability limits of many vapor and multiphase systems of interest. The 1000 K criterion provides a slightly conservative estimate for the LFL and will provide good guidance for performing empirical flammability testing.

The use of direct minimization of the Gibbs free energy provides many advantages over existing empirical and semi-empirical methods including but not limited to:

- Estimation of flammability limits where the mixture contains inerts and/or a mixture of fuels.
- Estimation of flammability limits where the initial temperature is different from 25 C.
- Estimation of flammability limits where the initial pressure is different from 1 bar.

## 3 Multiphase Chemical and Physical Equilibria

An algorithm for direct minimization of the Gibbs free energy is described in [5]. This algorithm was incorporated into Process Safety Office SuperChems [6] and can be used to perform a wide variety of simultaneous multiphase physical and chemical equilibrium estimates including:

- Constant temperature envelopes, such as the ones presented in this paper.
- Constant volume estimates.
- Hugoniot combustion curves and identification of upper and lower Chapman-Jouguet points.
- Limiting oxygen concentration (LOC).

Key differences between our algorithm and other previously published chemical equilibrium codes include the use of a modified cubic equation of state for both the liquid and vapor phases [7, 8]. This equation of state provides analytic derivatives and the ability to perform direct minimization of the Gibbs free energy at high pressure and for highly non-ideal systems and systems with strong solution effects. A sample truncated output is illustrated in Figure 12 for methane combustion.

#### 4 COMPOSITION EFFECTS

Figure 3: Estimation of ethane flammability limits using Process Safety Office SuperChems at a constant theoretical flame temperature of 1400 K



## 4 Composition Effects

Le Chatelier [9] proposed a simple formula for the estimation of mixture flammability limits. This formula applies to both the lower and the upper flammability limits:

$$LFL_{mix} = \frac{100}{\sum_{i}^{C} \frac{X_{i}}{LFL_{i}}}$$

$$UFL_{mix} = \frac{100}{\sum_{i}^{C} \frac{X_{i}}{UFL_{i}}}$$
(6)
(7)

where  $X_i$  is the vapor mole fraction, and both flammability limits are specified in volume percent. This simple formula may not provide accurate estimates for mixtures containing inerts or mixtures containing fuels such as acetone and ether leading to "cool flames".

The presence of inerts such as nitrogen, carbon dioxide, etc. presents a medium which absorbs reaction energy. If enough inerts are present, the reaction cannot provide enough energy to produce a self sustaining propagation.

Figure 3 illustrates the use of constant temperature limit of 1400 K to calculate the flammability envelope for ethane with two different diluents, carbon dioxide and nitrogen. The data compares favorably to values reported by the Zabetakis [10].

Please note that once a flammability envelope is constructed, the limiting oxygen concentration (LOC) is also estimated. The estimated flammability envelope for ethane would have been slightly

wider had a lower threshold flame temperature been used.

## **5** Temperature Effects

An increase in initial temperature of a flammable mixture will enlarge its flammability envelope. Less energy is spent by the combustion reaction to heat the reactants to the threshold temperature required for the reaction to produce a self sustaining flame. Higher initial temperatures will result in wider flammability envelopes. Zabetakis [10] reports the following empirical relations to assess the impact of temperature on flammability limits:

$$UFL(T) = UFL(298K) + \frac{0.75}{\Delta H_c} (T - 298)$$
(8)

$$LFL(T) = LFL(298K) - \frac{0.75}{\Delta H_c} (T - 298)$$
 (9)

where T is the initial mixture temperature in Kelvins, LFL and UFL are in volume percent, and  $\Delta H_c$  is the heat of combustion in kcal/mol. These relationships appear to be valid for straight chain hydrocarbons only.

Figure 4 illustrates the use of the chemical equilibrium algorithm to estimate the impact of initial temperature and water vapor on the flammability of a gas mixture which contains carbon monoxide and other hydrocarbons. The flammability limits for pure carbon monoxide at 25 C and 1 bar are shown in Table 1.

## 6 Elevated Pressure Effects

The impact of system pressure on the lower flammability limit is small while its impact on the upper flammability limit is significant. The limits of flammability of natural gas (85 to 95 % methane, 15 to 5 % ethane) are shown in Figure 5. A similar dependence of flammability limits on pressure is expected for other flammable gases. Zabetakis reports the following log-linear fit of the natural gas data:

$$LFL = 4.9 - 0.71 \log P$$

$$UFL = 14.1 + 20.4 \log P$$
(10)
(11)

where LFL and UFL are both in volume percent and P is the initial system pressure in atmospheres. These relationships may not apply directly to other systems.

Figure 6 illustrates how Gibbs free energy minimization can be used to estimate the impact of pressure on flammability limits. Figure 6 clearly shows that the LFL of natural gas does not vary with pressure while the UFL does. It is also important to note that the equilibrium estimates show a similar trend to the experimentally observed UFL change with pressure. A smaller change is

Figure 4: Impact of initial temperature and water content on flammability limits for gas mixture containing carbon monoxide and hydrocarbons. Envelope is calculated at a threshold flame temperature of 1200 K using Process Safety Office SuperChems



Figure 5: Effect of pressure on the flammability limits of natural gas at 301 K. Taken from Zabetakis, 1965.



Figure 6: Calculated flammability limits at a threshold flame temperature of 1000 K using Process Safety Office SuperChems with non-ideal gas effects assuming a natural gas composition of 85 % methane and 15 % ethane



predicted between 100 and 200 bars than from 1 to 100 bars. This trend is clearly shown in the experimental data illustrated in Figure 5.

Figures 7 and 8 also illustrate equilibrium estimates of flammability limits for propane at a flame temperature of 1000 K. Good agreement is shown between the model predictions and the actual data reported in Zabetakis [10]. The equilibrium model predictions are slightly conservative at limiting flame temperature values of 1000 K. This should be adequate for most hazard assessment studies. It is interesting to note, that the trend of change of both LFL and UFL are also well predicted by the equilibrium code for propane at 7.91 and 14.8 bars.

## 7 Reduced Pressure Effects

The flammability hazard potential is thought to disappear or to be greatly reduced at pressures lower than 0.065 bara (50 mmHg). This is shown for methane in Figure 9 where the lean and rich limits are reported to converge until a low pressure value of 100 mmHg below which flames will not propagate.

Figure 10 illustrates detailed equilibrium calculations at pressures below atmospheric for methane combustion. These estimates do not show such a convergence and support the opinion [11] that the measured convergence behavior is due to wall quenching effects by the tube in which the experiments were conducted.

Figure 7: Calculated effects of pressure on the flammability limits of propane at 298 K using Process Safety Office SuperChems



Figure 8: Calculated flammability limits at a threshold temperature of 1000 K using Process Safety Office SuperChems with non-ideal gas effects for propane





Figure 9: Measured flammability limits for methane at reduced pressures [12]

Figure 10: Calculated flame temperature for methane at reduced pressures using Process Safety Office SuperChems



Figure 11: Calculated flash point using Process Safety Office SuperChems for ethanol-water vs. Measured NFPA-325 [13] Data



It is well known that flame propagation will only occur if the rate of heat generated by the combustion reaction exceeds the rate of heat loss by radiation (majority of heat loss), convection, and conduction to the test apparatus or tube walls. Smaller tubes have larger surface to volume ratios which results in larger heat loss. Unless there is evidence to the contrary, systems that operate under reduced pressures or partial vacuum where air might be ingested due to leaks should use the flammability limits reported at 1 bar for potential hazard evaluation. The atmospheric flammability data can easily be corrected for elevated temperatures.

## 8 Flash Points for Nonideal Mixtures

The method developed to establish flammability limits can also be used to estimate flash points for highly non-ideal mixtures. Figure 11 shows Process Safety Office SuperChems estimates of the flash point for an ethanol-water mixture at different mass fractions. A flammability threshold flame temperature of 1400 K was used. The calculated flash points compare well with the measured values.

For a specific ratio of ethanol to water, Process Safety Office SuperChems calculates the bubble point temperature at which the vapor composition in equilibrium with the liquid yields a flame propagation temperature equal to 1400 K when mixed with air. A partial pressure is calculated by trial and error such that the coincident bubble point temperature and the ratio of air to vapor in equilibrium with the liquid yield the limiting flame temperature of 1400 K for a total pressure of 1 atm. The air atmosphere can also be substituted with a different oxidant such as pure oxy-

gen for example. As indicated earlier, the impact of inerts or other chemically active species is automatically accounted for by the Gibbs free energy minimization.

## 9 Conclusions

This paper presents a general method for the estimation of flammability limits for vapor and multiphase systems. The method can also be used to calculate flash points for ideal and non-ideal mixtures as well as limiting oxygen concentrations. The effects of pressure, temperature, inerts, and composition are shown to be well predicted for a variety of systems. This method has proven over time to be a very useful tool for fire and explosion hazard assessment studies and for providing guidance to flammability testing. Figure 12: Typical computer output from Process Safety Office SuperChems multiphase equilibrium estimator

Final	system	temperature	(K)				 •••		 	298
Final	system	pressure (Pa	ı) <b></b>	• • • •	• • • •	• • •	 •••	••••	 	1E+05

Overall material balance:

Compound		Initial	Change	Final	Fraction
CARBON-REF	(S)	0.00000	0.67625	0.67625	1.00000
METHANE	(V)	1.00000	-0.83813	0.16187	0.08094
OXYGEN	(V)	1.00000	-1.00000	0.00000	0.00000
NITROGEN	(V)	0.00000	0.00000	0.00000	0.00000
CARBON MONOXIDE	(V)	0.00000	-0.00000	0.00000	0.00000
CARBON DIOXIDE	(V)	0.00000	0.16188	0.16188	0.08094
HYDROGEN	(V)	0.00000	0.00001	0.00001	0.00001
WATER	(V)	0.00000	1.67624	1.67624	0.83812
NITRIC OXIDE	(V)	0.00000	-0.00000	0.00000	0.00000
NITROGEN DIOXIDE	(V)	0.00000	-0.00000	0.00000	0.00000
NITROUS OXIDE	(V)	0.00000	-0.00000	0.00000	0.00000
HYDROXYL	(V)	0.00000	-0.00000	0.00000	0.00000
MONOATOMIC OXYGEN	(V)	0.00000	-0.00000	0.00000	0.00000
MONOATOMIC HYDROGEN	(V)	0.00000	-0.00000	0.00000	0.00000
METHYL	(V)	0.00000	-0.00000	0.00000	0.00000
HYDROGEN ION +	(V)	0.00000	0.00000	0.00000	0.00000
HYDROGEN ION -	(V)	0.00000	0.00000	0.00000	0.00000
Totals:		2.00000	0.67625	2.67625	

Overall equilibrium balance:

Compound		Fraction Fugacity		Fugacity Coefficient		
CARBON-REF	(S)	1.00000	1.0000E+00			
METHANE	(V)	0.08094	7.9996E-02	Phi=	1.0015E+00	
OXYGEN	(V)	0.00000	4.9580E-11	Phi=	1.0047E+00	
NITROGEN	(V)	0.00000	1.3899E-09	Phi=	1.0060E+00	
CARBON MONOXIDE	(V)	0.00000	4.9673E-11	Phi=	1.0066E+00	
CARBON DIOXIDE	(V)	0.08094	7.9554E-02	Phi=	9.9591E-01	
HYDROGEN	(V)	0.00001	6.7612E-06	Phi=	1.0133E+00	
WATER	(V)	0.83812	8.1481E-01	Phi=	9.8507E-01	
NITRIC OXIDE	(V)	0.00000	4.9654E-11	Phi=	1.0062E+00	
NITROGEN DIOXIDE	(V)	0.00000	4.8665E-11	Phi=	9.8621E-01	
NITROUS OXIDE	(V)	0.00000	4.9114E-11	Phi=	9.9531E-01	
HYDROXYL	(V)	0.00000	5.0032E-11	Phi=	1.0139E+00	
MONOATOMIC OXYGEN	(V)	0.00000	5.0032E-11	Phi=	1.0139E+00	
MONOATOMIC HYDROGEN	(V)	0.00000	5.0032E-11	Phi=	1.0139E+00	
METHYL	(V)	0.00000	5.0032E-11	Phi=	1.0139E+00	
HYDROGEN ION +	(V)	0.00000	5.0032E-10	Phi=	1.0139E+00	
HYDROGEN ION -	(V)	0.00000	5.0032E-10	Phi=	1.0139E+00	

Figure 13: Typical computer output from Process Safety Office SuperChems multiphase equilibrium estimator (continued)

```
Calculated Reaction Stoichiometry:
Atom Matrix (Input):
```

0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	4.0000	1.0000	0.0000	0.0000	0.0000
2.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	2.0000	0.0000	0.0000
1.0000	0.0000	1.0000	0.0000	0.0000	0.0000
2.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	2.0000	0.0000	0.0000	0.0000	0.0000
1.0000	2.0000	0.0000	0.0000	0.0000	0.0000
1.0000	0.0000	0.0000	1.0000	0.0000	0.0000
2.0000	0.0000	0.0000	1.0000	0.0000	0.0000
1.0000	0.0000	0.0000	2.0000	0.0000	0.0000
1.0000	1.0000	0.0000	0.0000	0.0000	0.0000
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	3.0000	1.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	1.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	-1.0000

```
Rank of the atom matrix ......
Number of independent reactions .....
```

```
+0.5 CH4 <---> +0.5 C + H2
+0.5 CH4 +0.5 O2 <---> +0.5 C + H2O
+0.5 O2 +0.5 N2 <---> + NO
+ O2 +0.5 N2 <---> + NO2
+0.5 O2 + N2 <---> + N2O
+0.25 CH4 +0.5 O2 <---> +0.25 C + OH
+0.5 O2 <---> + O
+0.25 CH4 <---> +0.25 C + H
+0.25 C +0.75 CH4 <---> + CH3
+ C +0.5 O2 <---> + CO
```

1

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## About the Author



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

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

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

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