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Flammability Properties of Hydrocarbon Fuels

Part 2 - The Importance of Volatile Components at Low Concentration on the Flammability of Liquid Fuels

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ABSTRACT

It has been demonstrated by a simplified treatment that vapor pressures of individual constituents play a more important role than concentration on the overall flammability properties of hydrocarbon mixtures. This treatment is based on the application of Raoult's and Dalton's laws governing vapor pressure and composition above a solution of two or more liquid hydrocarbons to Le Chatelier's rule governing the flammability of vapor mixtures. The most important conclusion demonstrated by the derived equations is that a very small amount of highly volatile contaminant in a relatively nonflammable fuel may make it flammable. The amounts needed can be predicted from the equations and the properties of the components. Although precise relationships have been derived only for relatively simple solutions of pure hydrocarbons, the concepts they imply are applicable to more complex mixtures such as jet and diesel fuels. The flammability properties included in this study are lower and upper flammability limits, flash points, and flammability indices of liquid solutions.

PROBLEM STATUS

This is an interim report; work on this problem is continuing.

AUTHORIZATION

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FLAMMABILITY PROPERTIES OF HYDROCARBON FUELS

Part 2 - The Importance of Volatile Components at Low Concentration on the Flammability of Liquid Fuels

INTRODUCTION

Users of large quantities of highly flammable solvents, fuels, and similar liquids and gases are usually alert for fire and explosion hazard in their use but are frequently unaware of similar dangers which may sometimes exist for supposedly less flammable mixtures. Because of improper preparation, contamination, or other reasons, such fuels can contain small quantities of highly volatile flammable components, and these may significantly influence the overall flammability properties of the mixture. A hypothetical but perfectly possible Navy situation will serve as an example to illustrate this problem. JP-5 is a jet fuel of low volatility, especially designed in accordance with military specifications as a safe fuel for jet operation and storage and handling on aircraft carriers. The fuel is relatively less flammable under most conditions than the more commonly used and more volatile JP-4. Because of its greater volatility, vapors of JP-4 form flammable mixtures in the vapor spaces of its containers at normal temperatures (roughly 0° to 70°F). Suppose that an almost empty storage tank which had initially been used for JP-4 is filled with JP-5 fuel. Important questions are raised. Would the flammability hazard of this tank be that of JP-5 and therefore be relatively safe? Or would it be more like JP-4 and therefore relatively hazardous? One might expect that its properties would fall somewhere in between the two, but the question is which of the two fuels would it be most likely to resemble. Since the tank consists chiefly of JP-5, one might guess that the properties of the JP-5 would predominate and that the fuel would probably be safe. Such a hazarded guess can be quite hazardous! Some properties of mixtures, density for example, usually are proportional to the quantities of each of the components. Flammability, on the other hand, occurs in the vapor phase and is therefore a function of the concentration and constitution of vapor in the air above the liquid. The evaporation tendency (vapor pressure) as well as the relative concentrations of each of the constituents are major factors in determining the vapor composition and, thus the flammability of the mixture in question. The question still remains, will the contaminated JP-5 be hazardous and, if so, to what degree? The answers to these questions are of importance to the Navy and to other users of flammable liquids. It is possible to shed some light on this subject by use of known theoretical principles. A detailed, formal NRL Report on the flammability of hydrocarbon solutions is in preparation and will include physicochemical concepts and mathematical equations and their derivations. The ideas on volatile components which will be discussed here are derived from that report, but the mathematical and physicochemical complexities inherent in this type of problem will be minimized or entirely avoided here to make this very important topic more generally readable. For those who are mathematically curious, but who do not wish to dig through the detailed NRL Report on which this work is based, equations have been inserted in most of the figures, which describe the curves that are shown.

SOME FLAMMABILITY CONCEPTS

What is a Flammable Liquid?

Numerous terms and concepts are employed in these types of discussions, and it should help to define some of these here. A dictionary will define a "flammable" liquid

as a liquid which is capable of being easily ignited, and which will continue to burn with unusual rapidity. "Heavier" (higher molecular weight) liquids of lower volatility, which ignite less readily and burn relatively slowly, are ordinarily referred to as "combustible" liquids. The distinction here is a matter of volatility, the tendency of a liquid to assume a vapor state or to evaporate. A useful definition is given by the Code of Federal Regulations (1) which defines a flammable liquid as any liquid that gives off flammable vapors. These are vapors which, when mixed with air, can be ignited by a suitable source such as a spark or flame at or below a temperature of 80°F. The term combustible liquid is used if the liquid gives off flammable vapors above 80°F but below 150°F. The temperatures which are specified are arbitrary of course, and we will not make a distinction here, but the definitions imply several important concepts. First of all the liquid must give off vapors; it must do so in sufficient quantity at ordinary temperatures to be able to be ignited by an appropriate ignition source after mixing with air, and it must continue to burn as long as both fuel and air are present. In fuels, these liquids are usually petroleum derived hydrocarbons.

What is Vapor Pressure?

The property of a substance to volatilize or evaporate depends on its vapor pressure. Some materials, such as methane, ether, and aviation gasoline, are called "volatile," since they have relatively high vapor pressures at ordinary temperatures; other liquids, such as lubricating oil and mercury, are considered to be "nonvolatile," since their vapor pressures are relatively low at ambient temperatures. According to the kinetic theory the molecules of a liquid are in constant motion; the higher the temperature, the faster is this agitation. If a given surface molecule has sufficient kinetic energy, it breaks through the interface into the free space above it and thus enters into the vapor or gas phase. There is a continuous flight of molecules from the surface of the liquid to the vapor state, and simultaneously the reverse process is taking place where vapor molecules return to the surface of the liquid at a rate depending on the vapor concentration and are condensed back to the liquid phase. In a closed container, eventually a condition of dynamic equilibrium is established between the liquid and its vapor when the rate of escape is exactly equal to the rate of condensation of the vapor. The vapor is then said to be "saturated." The pressure exerted by vapor which is in equilibrium with the liquid is known as the "vapor pressure." The equilibrium between a liquid and its vapor depends on the temperature. Vapor pressure varies exponentially (i.e., at an ever increasing rate) with temperature, rising rapidly with relatively small temperature increases. It is measured in the usual pressure units, such as mm of Hg, psi absolute, and atmospheres. The temperature at which the vapor pressure is equal to 1 atmosphere (or at which vapor and liquid may exist in equilibrium in all proportions) is called the boiling point of the liquid. The vapor pressure of a pure liquid is a constant quantity at any given temperature and is independent of the amounts of liquid and vapor present. Typical vapor pressure-temperature curves for two pure hydrocarbons are shown in Fig. 1.

Before we get too far involved in this discussion it is necessary to set up some boundary limits. We will confine the subject to liquid and vapor fuels (but not droplets or mists) in air at atmospheric pressure. The question of the nature and energy of ignition sources will not be included, since it will be assumed from the safety standpoint that, if there is a flammable fuel-air mixture, an appropriate ignition source is likely to exist. In the case of evaporation of fuel vapors from liquid fuels it will be assumed that these vapors are at equilibrium with the liquids at a given temperature. Finally, it will be understood in this discussion that fuel vapor-air mixtures are uniform throughout.

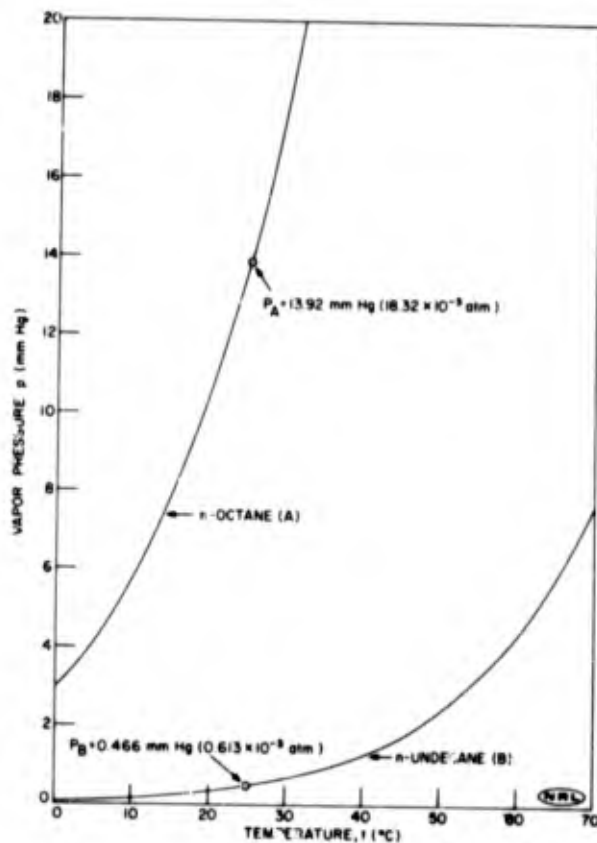


Fig. 1 - Vapor pressure vs temperature for two pure liquid hydrocarbons: n-octane (A) and n-undecane (B)

What are Flammability Limits?

The well known concept of the "fire triangle" states that to have a fire, "three sides of the triangle" are necessary, a fuel, air (oxygen), and an ignition source (energy). The definitions of a flammable liquid, which have been given earlier, imply the fire triangle, with the additional requirement that the fuel give off flammable vapor. This is because a fuel must be in the vapor state to burn. There is, however, still another aspect of flammability which we must consider. It can be seen intuitively that the relative amounts of fuel vapor and air must also be important. A fuel mixture with no oxygen cannot burn, and this is likewise the case for a mixture of air but no fuel. Somewhere between these two extremes there are fuel-air mixtures which are flammable. The question of how much fuel vapor (or conversely how much air) is necessary to form a flammable mixture is of prime significance. If, for example, the fuel is of low volatility (for example, lubricating oil at ordinary temperatures), one can foresee that there will not be sufficient fuel vapor above the liquid in a semienclosed space, such as a storage tank, to be ignited. The fuel-air mixture is said to be too "fuel-lean." Less obvious is the "fuel-rich" case, where a highly volatile fuel (for example aviation gasoline at ordinary temperatures) might produce sufficient vapor to displace most of the air in the space and thus would also be nonflammable, but for the opposite reason. If, however, as is common, we have a fuel of the appropriate volatility (for example JP-4 jet fuel at ordinary temperatures), the fuel would give off sufficient vapor to fall within the limits of its flammability range and would thus form a flammable mixture with air. These concepts are illustrated for a

hypothetical fuel in Fig. 2, where oxygen concentration is plotted against fuel vapor concentration. The shaded area represents the flammable region, outside of which the mixtures are nonflammable. It can be seen that the flammable region widens with increasing oxygen concentration, as might be expected. The diagonal line starting at about 21% oxygen on the left represents mixtures of air and fuel, and this line crosses the flammable region at two concentrations, L and U. The former, L, represents the fuel concentration at the lower flammability limit in air, below which the concentration is too low to support combustion. The latter, U, is the concentration at the upper flammability limit in air, above which the fuel vapor is too rich to support combustion.

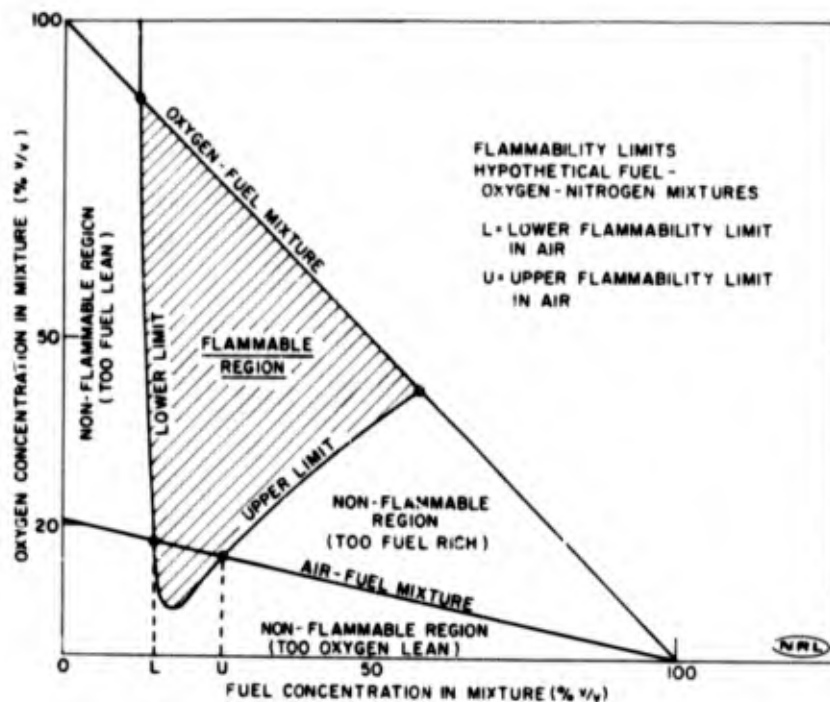


Fig. 2 - Flammability limits of hypothetical fuel-oxygen-nitrogen mixtures

The definition of flammability limits from Coward and Jones (2) is presented here by way of summary. "A flammable mixture of gases, such as methane and air, may be diluted with one of its constituents until it is no longer flammable. The dilution limit of flammability, or simply the 'limit of flammability' is the borderline composition; a slight change in one direction produces a flammable mixture, in the other, a nonflammable mixture. There are clearly two limits of flammability, a lower and a higher, for each pair of so-called combustible gases and supporters of combustion. The lower limit corresponds to the minimum amount of combustible gas, and the higher or upper limit to the maximum amount of combustible gas capable of conferring flammability to the mixture."

Flammability limits depend on the nature (heat of combustion, etc.) of the fuel, the composition of the oxidizing atmosphere (air or atmospheres containing other oxygen concentrations, and nitrogen or other diluents), temperature, pressure, and on other factors. Unless otherwise stated, flammability limits will be assumed to be at atmospheric pressure in air at ordinary temperatures (in the neighborhood of 25°C).

FLAMMABILITY LIMITS OF MIXTURES OF TWO OR MORE FUEL VAPORS

We have considered the meaning and definition of flammability limits for a given fuel, and the question now arises concerning the properties of mixtures of two or more fuels. Before we consider liquid fuels, let us first consider the less complex subject of vapor mixtures. Suppose that we know the flammability limits of a given fuel vapor to which we add a second fuel vapor whose limits are also known. Assuming that we also know the relative amounts of each, what would be the flammability limits of the mixture? The answer to this question was given by Le Chatelier (3) in 1891. He developed a simple additive relationship from experimental observations of lower flammability limits of various fuel mixtures. The rule states that if a flammable mixture is formed from two flammable gases, then the sum of the ratios of the actual concentration to the concentration at the lower flammability limit for each gas is equal to unity. Le Chatelier's rule is illustrated in Fig. 3 for binary vapor mixtures of n-octane and n-undecane in air. It is seen that a plot of n-octane concentration vs n-undecane concentration gives a straight line separating the flammable from the nonflammable region near the lower flammability limit. The mathematical equations and the meanings of the symbols used are given in the figure, as is the case for those figures which will follow. Plots of binary mixtures of n-octane (and sometimes other n-alkanes) in n-undecane will be used for illustrative purposes. In Fig. 3 the lower flammability limits for pure n-octane (0.892%) and pure n-undecane (0.656%), which are the extremes of the line, are shown. Le Chatelier's rule has been found applicable to many gases and vapors including hydrocarbons; it has been extended also to upper flammability limits and can be applied to three or more flammable gases (2).

Two very interesting corollaries are implied by Le Chatelier's rule (2), and these can be demonstrated mathematically from Le Chatelier's equation, or graphically as in Figs. 3 and 4.

The first corollary is concerned with a single mixture of two or more vapors or gases in air, each of which is at a fraction of its individual lower flammability limit concentration. Each gas, therefore, if it were the only one present in air at that concentration, would be nonflammable. The corollary states that if the sum of the fractions is equal to unity, then the mixture of gases is a lower limit flammable mixture. For example, a mixture of n-octane and n-undecane in air which contains one-quarter of the amount of n-octane ($1/4 \times 0.892 = 0.233\%$) and three quarters of the amount of n-undecane ($3/4 \times 0.656 = 0.492\%$) which are individually necessary to form a lower limit mixture, will be a lower limit mixture, since the sum of the fractions ($1/4 + 3/4$) is equal to unity. This is indicated as a triangle on the lower limit line in the two figures.

The second corollary is concerned with two or more individual flammable mixtures of vapors or gases in air when mixed together to form a single more complex mixture. The corollary states that two or more lower limit flammable mixtures, if mixed in any proportions, give rise to new mixtures which are also at their lower limits and are flammable. For example, suppose that we have two separate flammable mixtures, the first a mixture of n-octane in air at its lower limit (0.892%) and the second a mixture of n-undecane in air also at its lower limit (0.656%), and that the two mixtures are mixed in the proportion of 1:2 respectively to form a single new mixture. The resulting mixture will contain 0.297% n-octane and 0.437% n-undecane, and this is also a flammable mixture, as is shown by the squares in Figs. 3 and 4.

By algebraic transformation, Coward, Carpenter, and Payman (4) derived a relationship for the overall lower flammability limit of the combined flammable gases (this will be referred to as the "fuel mixture") as a function of the concentrations and lower flammability limits of the individual components. This relationship, which is illustrated for a binary mixture in Fig. 4, states that the reciprocal of the lower flammability limit of

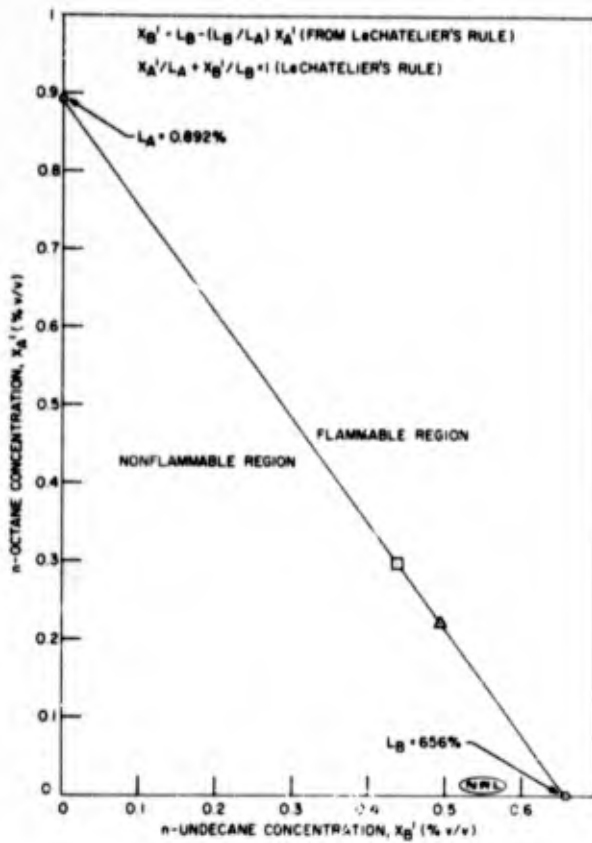


Fig. 3 - Le Chatlier's rule illustrated by the plot of the lower flammability limits of a binary vapor mixture of pure n-octane (A) and n-undecane (B) in air

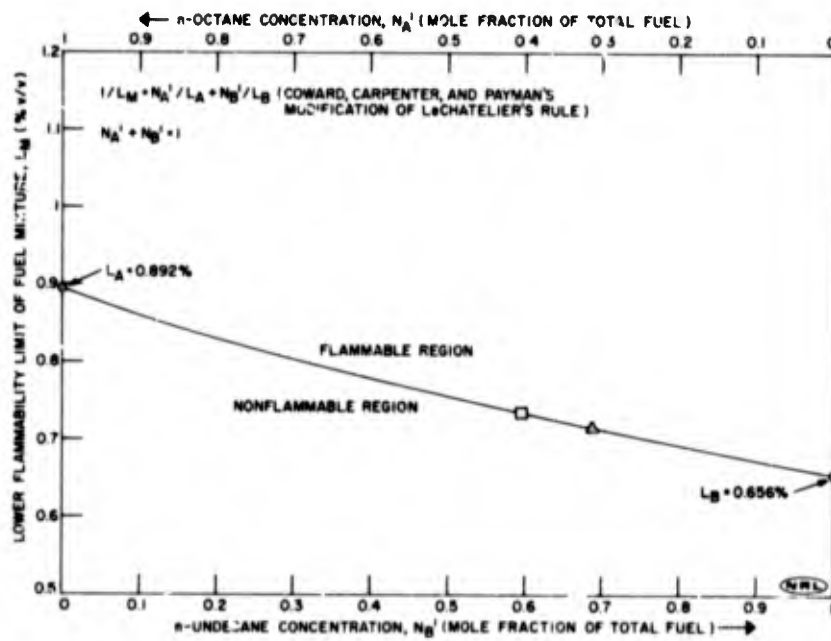


Fig. 4 - Theoretical lower flammability limits of binary fuel vapor mixtures of n-octane and n-undecane in air

the mixture is equal to the sum of the ratios of the proportions of each fuel in the flammable mixture (expressed as mole fraction of total air-free fuel vapor) to its concentration at the lower flammability limit. In the figure the lower flammability limit of the mixture (percent by volume) is plotted against the vapor concentration (mole fraction of total hydrocarbon vapors) of n-undecane to the right (or n-octane to the left). The plot gives a gently curved line dividing the flammable from the nonflammable region and terminates at the lower limits for the pure hydrocarbons. This relationship can also be extended to upper flammability limits and to mixtures of more than two components (2). The figure suggests (and it can be shown from the equations) that the relative influence of each flammable vapor component on the lower flammability limits of the total mixture is a function of its concentration in the total fuel vapor.

HOW TO PREDICT THE FLAMMABILITY PROPERTIES OF LIQUID SOLUTIONS

We now redirect our attention to liquid fuels but this time to solutions containing two or more components. The question arises as to how to predict the overall flammability properties of a given solution from the properties and concentrations of the individual constituents. The answer to this question is based on physical chemistry principles by which it is possible to calculate the equilibrium composition at a given temperature above a liquid hydrocarbon solution from the individual vapor pressures of the components and their relative concentrations in the liquid solution. Once we know the vapor phase composition above the liquid we can predict the overall flammability properties by use of Le Chatelier's rule, as has been shown. Vapor composition above a liquid solution is dependent on the vapor pressures and the concentrations in the solution of the individual constituents or, in other words, on the partial vapor pressures of each component. The partial vapor pressure is the vapor pressure which the individual pure component would exhibit, if it were the only vapor present in the volume occupied by the vapor mixture. The keys to the overall flammability properties of a liquid solution are the partial vapor pressures and the individual flammability properties of the components. Since vapor pressure is a colligative property and depends on the number of molecules present, it is therefore equal to the vapor pressure of the pure component multiplied by its concentration (mole fraction) in the liquid solution (Raoult's Law). The total vapor pressure of the solution is equal to the sum of the individual partial vapor pressures (Dalton's Law). Raoult's and Dalton's laws have been found to be applicable to liquid hydrocarbon solutions which approximately resemble "ideal" solutions. These concepts are illustrated for a binary solution in Fig. 5, which is chosen as a "textbook" example. In the figure the vapor pressures of the individual pure components are not far apart. However, in the case of a binary liquid mixture of two components of widely different vapor pressures, the general shape of the graph is distorted from that of the textbook case and is shown in Fig. 6 for a solution of n-octane and n-undecane. In both figures the diagonal lines for the partial pressures of each component, p'_A and p'_B , are shown, and the total vapor pressure p_M obtained by adding p'_A plus p'_B at each concentration is indicated by the upper line. Where the vapor pressures of the pure components are fairly close (Fig. 5) the total vapor pressure line p_M is the sum of the contributions of two oppositely sloped but relatively equivalent diagonal lines. However in the other case, where the pure vapor pressures differ considerably, the contribution of the lower vapor pressure constituent is relatively low, and p_M is approximately equivalent to the partial pressure of the more volatile component p'_A . This is an important point, that the total vapor pressure of a liquid solution of volatile and relatively nonvolatile components is approximately equivalent to the partial pressure of the more volatile constituents. This has an important bearing on the vapor composition and the flammability properties of liquid solutions.

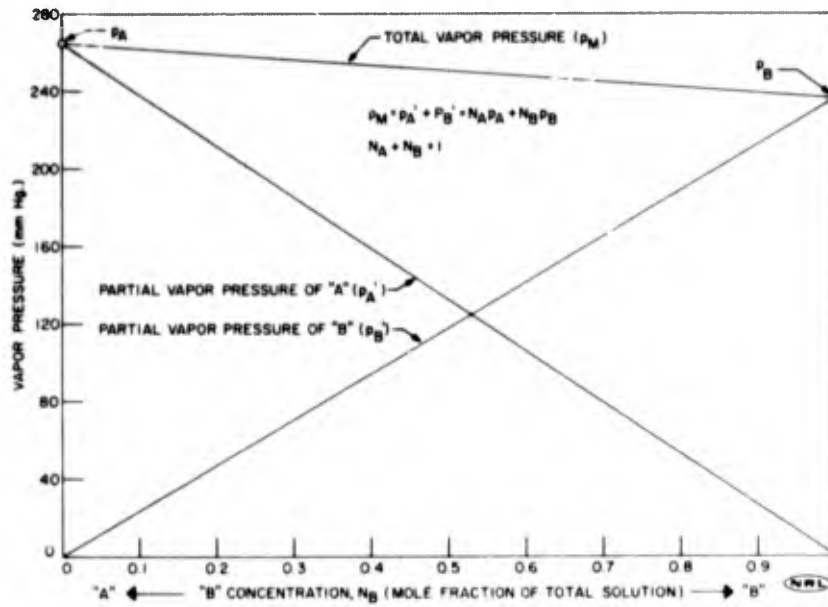


Fig. 5 - Vapor pressure vs concentration of ideal binary liquid solutions at constant temperature

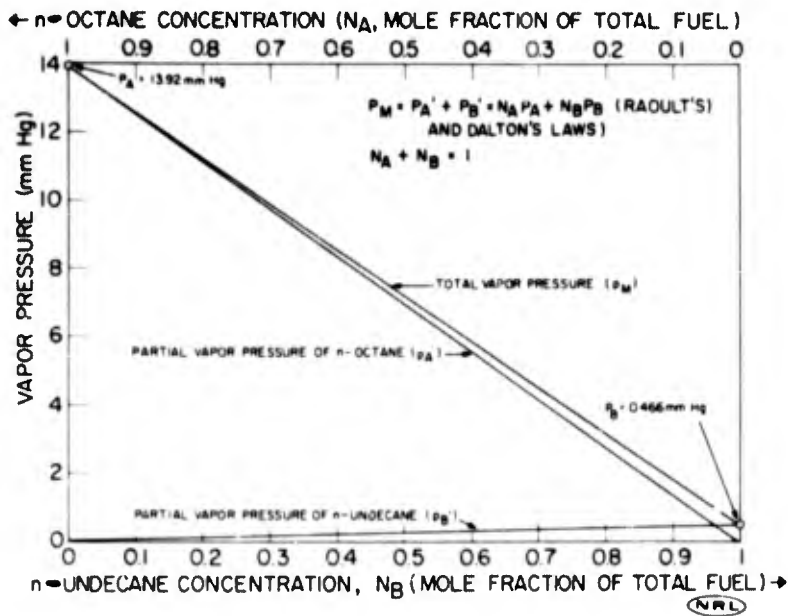


Fig. 6 - Vapor pressure vs concentration of binary liquid solutions of n-octane (A) and n-undecane (B) at 25°C

LOWER FLAMMABILITY LIMITS OF LIQUID SOLUTIONS

By combining the laws of Le Chatelier, Raoult, and Dalton, we can determine the composition and flammability limits above a liquid solution at a given temperature. This is illustrated for a binary mixture of n-octane and n-undecane in Fig. 7. The linear relationship for vapor mixtures (Fig. 4) obviously does not apply to liquid solutions. The lower flammability limit of the mixture, starting with n-undecane (0.656%, at the right side of Fig. 7), rises rapidly with increasing n-octane concentration (as we proceed to the left) and quickly approaches that of the more volatile constituent n-octane, while still at relatively low concentrations of n-octane. For still more volatile fuel components, such as n-hexane (also in n-undecane) the rise is even more rapid, and the curve approaches an almost constant level close to the lower limit of the hexane. The farther apart the volatility of the components of a mixture, the more will the total properties of the mixture approach that of the more volatile constituent.

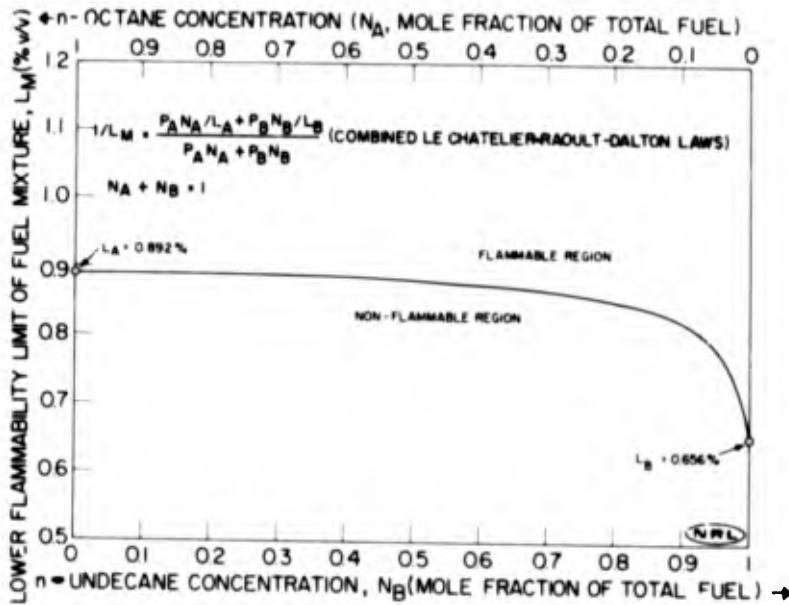


Fig. 7 - Theoretical lower flammability limits of binary liquid solutions of n-octane and n-undecane in air

FLASH POINT OF LIQUID SOLUTIONS

It is often convenient and useful to express the flammability limits of a liquid fuel by the temperatures at which its vapors become flammable. We will confine this discussion to the lower limit of flammability. The lower flash point, commonly referred to simply as the "flash point," is the minimum temperature at which sufficient vapor is released by a liquid to form a flammable vapor-air mixture at 1 atmosphere pressure. In other words, the flash point is that temperature at which the vapor pressure (in atmospheres) of the fuel is equivalent to the concentration at the lower flammability limit (mole fraction). Both vapor pressures and lower flammability limits are temperature dependent, and the flash point temperature may be obtained by the intersection of the plots of vapor pressure and lower flammability curves, each plotted against temperature (5). This is shown at "A" for a typical pure liquid fuel in Fig. 8. The application of these concepts to

solutions of two or more liquid fuels is somewhat complex, more so than our earlier discussion of flammability limits of solutions. This is because the lower flammability limit discussion assumed a constant temperature (conveniently at 25°C), whereas the concept of flash point adds temperature as a new variable. Nevertheless, by application of appropriate physicochemical principles involving the two temperature dependent functions (vapor pressure and lower flammability limits) to the combined laws of Le Chatelier, Raoult, and Dalton, equations can be derived to describe the flash point of hydrocarbon solutions. For these purposes, the linear temperature dependence of the lower flammability limit which has been described previously (5) was used. A form of the Antoine equation was used to describe the variation of vapor pressure of the fuels with temperature. Both of these temperature functions have been found to be applicable to hydrocarbon fuels. By means of the combined equations it is possible to calculate the flash point of solutions of one or more pure hydrocarbon components from the flash points and the concentrations of each constituent. Examples of this for three binary solutions are given in Fig. 9. The curves also illustrate the marked importance of the more volatile components to the flammability properties of the mixture.

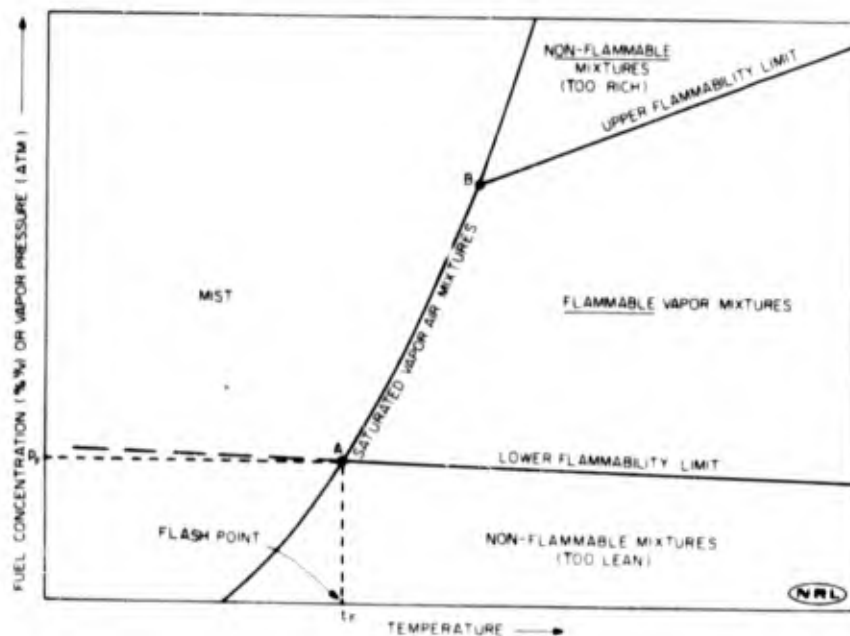


Fig. 8 - Vapor pressure and flammability limits vs temperature for a typical hydrocarbon fuel-air mixture

FLAMMABILITY INDEX ("EXPLOSIVENESS") OF LIQUID SOLUTIONS

Another useful flammability property is the flammability index (5) or "explosiveness" which is a measure of potential flammability hazard of a fuel. It represents the fraction, or ratio, of the actual concentration of fuel vapor above a liquid to that at its lower flammability limit. If the concentration is equal to that at the lower limit, then the flammability index is equal to unity. If the value is unity or above, the mixture is flammable, and if less than unity, it is nonflammable. For a given pure hydrocarbon at a specific temperature one can estimate the vapor concentration above the liquid from its vapor pressure. The flammability index can then be calculated from the vapor concentration

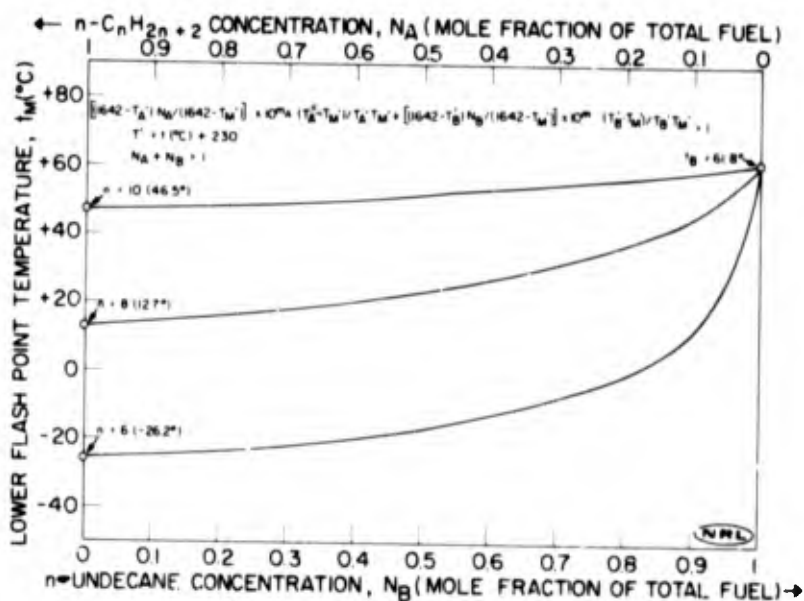


Fig. 9 - Theoretical lower flash point temperatures of binary liquid solutions of n-alkanes and n-undecane in air

and the lower flammability limit. The flammability index can also be determined experimentally, as is done, for example, for JP-5 jet fuels in the "explosiveness test" (6) which is performed at an arbitrary temperature of 125°F. By means of the equations which we have employed for flammability limits, it is possible to calculate flammability indices for solutions of hydrocarbons. As shown in Fig. 10 the resulting plot of flammability index vs concentration is linear. The horizontal line at $E = 1$ represents the critical value above which the vapors are flammable. In the example, a binary solution of n-octane in n-undecane, the graph crosses the line at a concentration of 6 mole-percent of n-octane. This means that n-undecane, which is not flammable at 125°F, can be made flammable by adding as little as 6 mole-percent of n-octane. Plots of other mixtures of n-alkanes in n-undecane are shown in Fig. 11. It is seen that with higher vapor pressures (lower carbon numbers, C_n) a smaller concentration of the n-alkane is required to make the final solution flammable, so that for n-hexane only 1 mole-percent is sufficient. In Fig. 12 a plot of concentrations of the volatile component vs n-undecane is given. This curve shows that for still more volatile fuels, such as n-pentane and below, extremely small concentrations will yield flammable mixtures. The relative importance of vapor pressure over that of concentration on flammability is clearly demonstrated.

SUMMARY AND CONCLUSIONS

It has been demonstrated by application of Raoult's and Dalton's laws governing vapor pressure and composition above a solution of two or more liquid hydrocarbons to Le Chatelier's rule governing the flammability of vapor mixtures, that vapor pressures of individual constituents play a more important role than concentration on the overall flammability properties of the solution. Although precise relationships have been derived only for relatively simple solutions of pure hydrocarbons, the concepts they imply are applicable to more complex mixtures such as jet fuels, diesel fuels, and the like. The most important conclusion demonstrated by these equations is that a very small amount of highly volatile contaminant in a relatively nonflammable fuel may make it flammable.

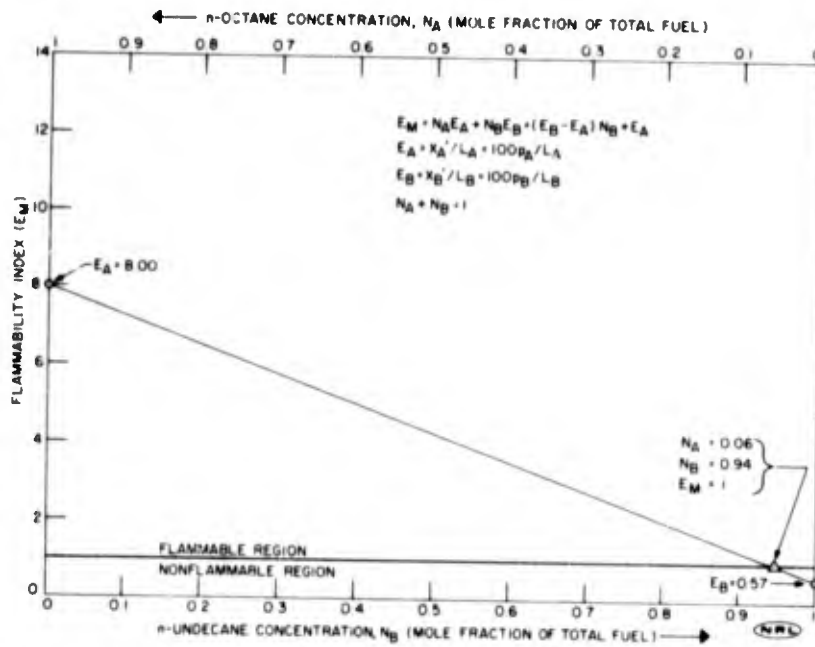


Fig. 10 - Flammability indices (explosiveness) of binary liquid solutions of n-octane and n-undecane in air at 125°F

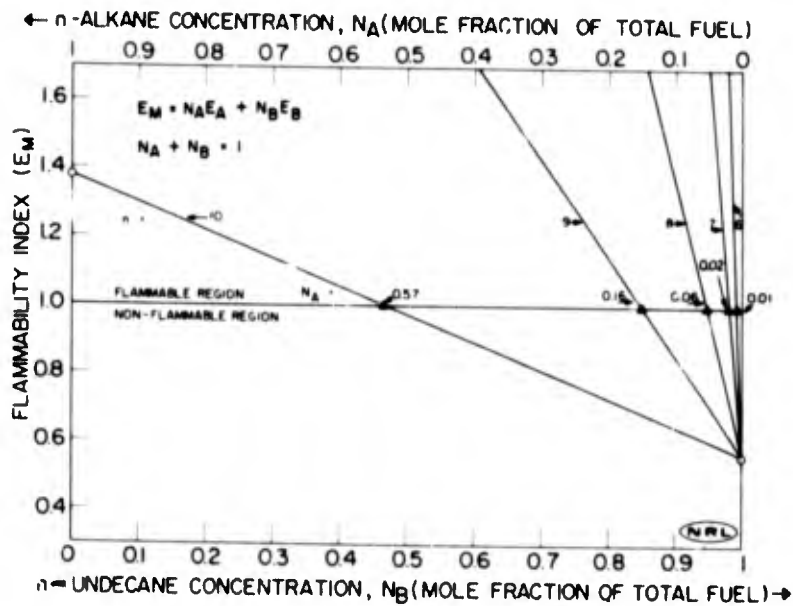


Fig. 11 - Flammability indices (explosiveness) of binary liquid solutions of five n-alkanes in n-undecane in air at 125°F

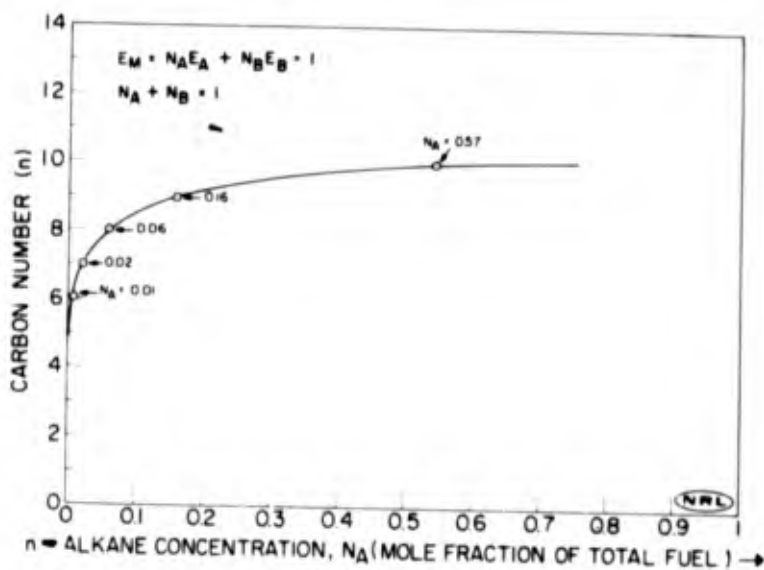


Fig. 12 - Minimum concentrations for flammability vs carbon number of binary liquid solutions of n-alkanes and n-undecane in air at 125°F

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		Dept. of the Navy (Office of Naval Research and Naval Ship Systems Command), Washington, D.C. 20360	
13. ABSTRACT			
<p>It has been demonstrated by a simplified treatment that vapor pressures of individual constituents play a more important role than concentration on the overall flammability properties of hydrocarbon mixtures. This treatment is based on the application of Raoult's and Dalton's laws governing vapor pressure and composition above a solution of two or more liquid hydrocarbons to Le Chatelier's rule governing the flammability of vapor mixtures. The most important conclusion demonstrated by the derived equations is that a very small amount of highly volatile contaminant in a relatively nonflammable fuel may make it flammable. The amounts needed can be predicted from the equations and the properties of the components. Although precise relationships have been derived only for relatively simple solutions of pure hydrocarbons, the concepts they imply are applicable to more complex mixtures such as jet and diesel fuels. The flammability properties included in this study are lower and upper flammability limits, flash points, and flammability indices of liquid solutions.</p>			

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Hydrocarbon fuels Flammability Flash point Flammability index Flammability limits n-alkanes Jet fuels Vapor pressures of fuel mixture Liquid fuel solutions Fuel vapor mixtures						