# Numerical analysis for nitrogen dilution on flammability limits of hydrocarbon mixtures

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## ARTICLE INFO

## ABSTRACT

There are numerous flammability literature data available for pure substances. For fuel mixtures, however, these are not always available. Especially, for fuel mixture storage, operation, and transportation, inert gas inerting and blanketing have been widely applied in chemical process industries while the related data are even more scarce. Lower and upper flammability limits of hydrocarbon mixtures in air with additional nitrogen were measured in this research. Typically, fuel mixture lower flammability limits almost keep constant at different fractions of added nitrogen; and approximately upper flammability limits of fuel mixture vary linearly with the addition of nitrogen except those mixtures containing ethylene. Numerical analysis of effect from inert gas dilution on fuel mixture lower and upper flammability limits was conducted by introducing the parameter of inert gas dilution coefficient, through which fuel mixture flammability properties, e.g., lower flammability limit, upper flammability limit, and minimum inerting concentration, can be quantitatively characterized with the modified Le Chatelier's mixing rule.

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## 1. Introduction

As a result of high safety requirements imposed on process plants, the larger flexibility to facilitate variety in feedstock, the design of new processes making use of intensified conditions and in general for hazard analysis, accurate prediction of explosion limits of mixtures of flammable substances is highly desirable. The evaluation of flammability limits is not absolute, but rather depends on experimental conditions. There are no definite parameters to quantitatively characterize the flammability limits. In practice, the limits of flammability of a particular system of air-fuel are affected by a variety of factors, e.g., temperature, pressure, oxygen concentration, inert gas addition, size and shape of equipment (Babrauskas, 2003).

There are a large variety of cases where internal gas explosion may occur, which are caused by uncontrolled leaks, or simply by accidental purging with air without inerting systems or tank inert blanketing system fails. The subsequent explosion can result in significant consequences. The most famous internal tank explosion case is the 1996 TWA Flight 800 disaster in which the center fuel tank exploded shortly after takeoff and resulted in the deaths of 230 people (Friedrich, 2002). Due to the non-ozone-depleting, non-toxic and non-pyrolytic properties, some inert gas agents, mostly including nitrogen is classified as a clean fire-suppressing agent of interest for tank inert blanketing. To control fire and explosion, inert additives are sometimes added to mixtures in order to narrow their flammable ranges or to render the mixture entirely non-flammable. Besnard (Besnard, 1996) provided some excellent examples of the influence of inert gases on the flammability limits, where different inert gas inactivating capacities to reduce the flammable ranges of fuel-air mixtures are systematically investigated. For most hydrocarbon gases, nitrogen in the amount of

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40–50 vol % must be added to a fuel/air mixture to make it non-flammable (Zabetakis, 1965).

In this research, nitrogen dilution effect on the lower flammability limit (LFL) and upper flammability limit (UFL) of low-carbon hydrocarbons (methane, ethane, propane, n-butane, ethylene, and propylene) and their binary mixtures were measured at ambient conditions. Le Chetalier's mixing rule is extensively used because of its simplicity and effectiveness to estimate the flammability limits of fuel mixtures; its application, however, focuses on fuel mixture in air without additional inert component introduced (Kondo et al., 2006). In the case of additional nitrogen added to hydrocarbon mixtures, numerical data analysis was conducted to extend Le Chatelier's mixing rule application, which included some proposed empirical equations, e.g., LFL and UFL quantitative characterization with addition of inert gas nitrogen, and minimum inerting concentration (MIC) at different fuel mixture compositions.

## 2. Experimental method

The flammability limit data for fuel/air mixtures are measured through a flammability apparatus developed by Wong (Wong, 2006) at Texas A&M University. The flammability apparatus (Fig. 1) used in this research is a closed stainless steel (SS 316) cylindrical vessel with an internal diameter of 10.22 cm and 100 cm length. At the central line of the reaction vessel, there are five evenly-separated temperature sensors consisting of NTC thermistors (Thermometrics, FP07DB104N with fast response time 0.1 s in still air), which can monitor a self-sustained flame propagation when fuel/air premixed mixtures combust upwardly. Thermistor 1 is located at a distance of 15 cm from the ignition source, which is 5 cm off the bottom surface of reaction vessel. The greatest distance from the ignition source to the farthest thermistor number 5 is 75 cm.

A flame propagating 75 cm distance or over detected by thermistors is defined as continuous flame propagation (Zhao et al., 2009). The procedure to determine the lower flammability limit is according to the one developed by Wierzba et al (Wierzba et al., 1988). that the probability of continuous flame propagation can vary from 0 to 100% when the fuel is within a certain concentration range near the lower flammability limits. At each fuel concentration, 10 tests are performed, and the number of times of the continuous flame propagation is recorded; therefore, the probability of continuous flame propagation is obtained at this selected concentration. Then, the probability of continuous flame propagation is plotted against fuel concentration, and by regression a linear line is obtained, on which a concentration with a 50% probability of continuous flame propagation is identified as the lower flammability limit of the measured fuel/air mixture. The same approach is applied for upper flammability limit experimental estimation. Fig. 2 is an example, which illustrates the procedure to determine the lower flammability limit of methane in air. The experimentally determined lower flammability limits of pure methane are compared with some literature data from previous research using different experimental apparatus and detection criteria, and the data are shown in Table 1 (Zhao et al., 2010).

## 3. Experimental results

The flammability limits (LFLs and UFLs) with varied concentrations of additional nitrogen were measured for pure hydrocarbons and some of their binary mixtures in air at room temperature and ambient atmospheric pressure. As an example, Fig. 3 illustrates methane flammability property changes with dilution of nitrogen.
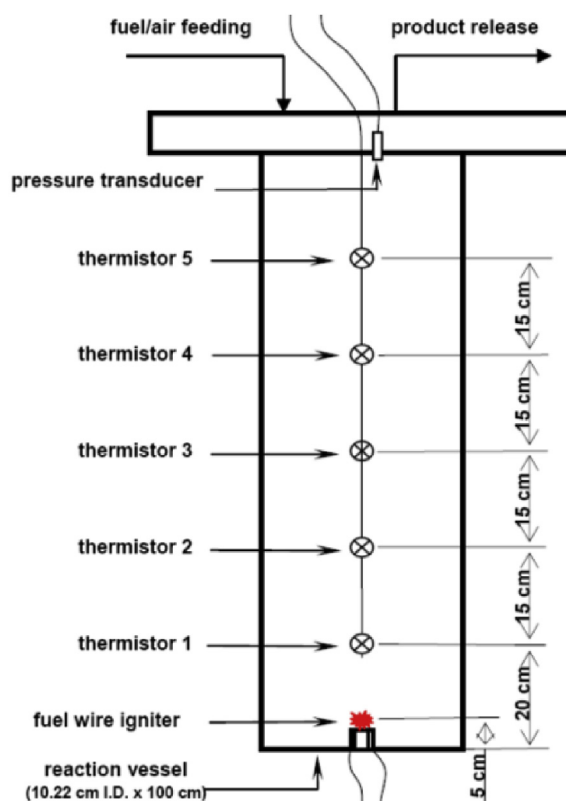


**Fig. 1.** Schematic diagram of the flammability apparatus.

Approximately, the lower flammability limit of methane remains constant with addition of nitrogen; the upper flammability limit, however, decreases dramatically. These two values become closer with continuous addition of nitrogen, and finally merge at the MIC point, beyond which fire or explosion is impossible. The region enclosed by the LFL and UFL curves is called the flammable zone. Outside of this region, fuel mixture is non-flammable. Specifically, a small zone close to the point, MIC, is set aside and defined as the flammability nose, where the flammability properties become deviant since fuel content turns leaner at LFL and richer at UFL till they are equal. Figs. 4–8 show the flammability properties of
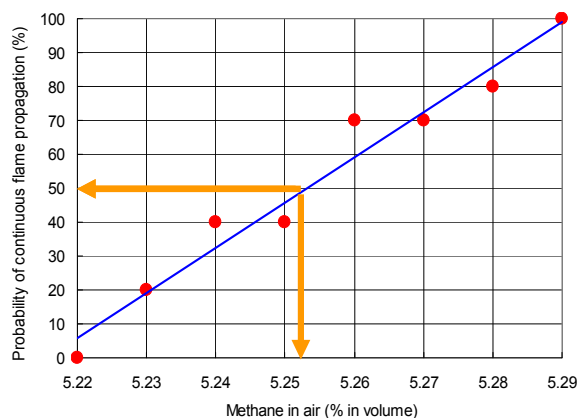


**Fig. 2.** Determination of the lower flammability limit of methane in air.