



Flammability of gases in focus of European and US standards



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ABSTRACT

The presentation will discuss the difference between EU and US standards for the determination of explosion (flammability) limits and limiting oxygen concentration. Small differences observed in measured values can be traced back to the different test apparatuses and criteria. The discrepancies can be much greater in the case of limiting oxygen concentration because of the high amount of inert gases and the corresponding low laminar burning velocities. The paper describes some examples and the influence of the chosen criteria on the results. The European and US standards use the criteria of flame propagation in open test vessels and of pressure rise in closed ones. The examples discussed show that flame propagation is still possible at very small pressure rise values, as observed much below the pressure rise criterion of usual standards. However, flame propagation in a process plant can cause an accident or explosion and must be avoided. Therefore, the flame propagation criterion is recommended to be used in chemical safety engineering. The European safety database CHEMSAFE contains expert-evaluated safety data for cases where the determination method and criteria are known. Flammability characteristics based on the pressure rise criterion may suffice in certain cases, e.g. for explosion protection in closed vessels without any connecting pipes.

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1. Introduction

Flammability of gases is a fundamental material property that entails legal classifications in various instances and requires appropriate labelling. Explosion protection measures are necessary only when flammable gases are present, e.g. in processes, storage and use. In European technical legislation, a gas or a gas mixture is regarded as “flammable” if under atmospheric conditions an explosion range (explosion limits) in a mixture with air exists. Therefore determination methods for flammability and explosion limits are standardized in many countries. Nevertheless, explosion limits are not the type of independent physicochemical material characteristics such as boiling temperature or density of a substance. As most other safety characteristics they are influenced by the test apparatus and the determination procedure applied. The evaluation and standardization of determination methods for flammability limits and limiting oxygen concentrations are particularly important for chemical safety engineering.

2. Terminology: “flammability limits” vs. “explosion limits”

The expression “explosion limit” often is called “flammability limit” in the literature. But “flammable” is an ambiguous term: it means combustible in air on the one hand and explosively reacting without any further addition of air or another oxidizer on the other. Typical examples are *flammable* gases, e.g. pure hydrogen and *flammability* (explosion) ranges that characterize hydrogen-air mixtures. In the European ATEX directives and standards of explosion prevention the term “explosion” is used for explosively reacting mixtures to avoid misunderstanding.

The actual situation in standardization and scientific literature is characterized by the fact that most of the US authors use the term “flammability limit”, while European standardization calls the same material safety characteristic “explosion limit”. Nowadays there are discussions in international standardization committees about a different meaning in the sense of more violent reactions if the “explosion limit” is reached, compared to slower reactions at the “flammability limit”. An informal IEC paper (IEC, 2003) used the direction of flame propagation as a criterion. Bureau of Mines measurements on hydrogen-air mixtures are often cited, where an upward flame propagation was observed at 4 mol% hydrogen but downward propagation does not begin before 9 mol% hydrogen (Coward and Jones, 1952). Therefore, 4 mol% is proposed as the

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lower flammability limit (LFL) and 9 mol% as the lower explosion limit (LEL). Nevertheless, such definition is not helpful in the field of explosion protection. There is actually no international standard available for the criterion “downward propagation” on the one hand and it can never be known where a potential ignition source is located in a process plant on the other. Therefore, only the more conservative upward propagation can be the relevant criterion for explosion protection.

To avoid misunderstanding it must be pointed out that “flammability limit” in the US and “explosion limit” in Europe represent the same material property, determined by similar standard test procedures using the same criterion “upward propagation” of flames.

3. Comparison of European and US standards for determination of explosion limits

With the creation of European directives in the field of explosion prevention, which apply uniformly to all Member States, it became necessary to develop new European standards for the determination of explosion characteristics. The European Technical Committee 305 “Potentially explosive atmospheres – Explosion prevention and protection” (CEN/TC305) is heading this project. Working Group 1 is responsible for the development of new standards for the determination of safety characteristics of gases, vapors and dusts.

In this field a new European draft prEN 1839:2015 “Determination of explosion limits and of the limiting oxygen concentration of gases and vapors” is under development. It is a merged revised version of EN 1839:2012 (EN, 1839; 2012) (explosion limits) and EN 14756:2006 (EN 14756, 2006) (limiting oxygen concentration). Parallel to the European standards, corresponding US standards are available (ASTM E681 (ASTM E681-09, 2015), ASTM E2079 (ASTM E2079, 2013)).

BAM has performed tests (Schroeder and Daubitz, 2004) to compare US and European standards for a number of years with 5 test substances, 4 gases and a vapor, ethanol, chosen for the tests. The gases, hydrogen, ethylene, methane and ammonia, had different laminar burning velocities. It was believed that differences in the results due to test apparatuses and test procedures could be checked by measurements on the same test substances. 40 mol% of nitrogen was added to the gas-air mixtures to increase sensitivity. Altogether 4 test methods were used (for parameters see Table 1). The parameters of apparatuses and test methods are the same as in the latest versions of the standards (e.g. prEN,

1839:2015 and ASTM E681-09), with the only exception of ammonia. Nowadays in European and US standards, special procedures are available for so-called “difficult-to-ignite” substances such as ammonia and halogenated refrigerants. Tests to compare these test methods should be carried out in the future. Three of the four test methods (DIN 51649 (DIN 51649, 1986), EN 1839-T (tube method), ASTM E681) are so-called “open vessel methods” that apply upward flame propagation as the criterion for a reaction. Another method was the “closed vessel method” EN 1839-B (bomb method) that used the criterion of pressure rise.

The following conclusions were drawn from the experiments (see Table 1 and Figs. 1–3):

- The procedures according to DIN 51649 and EN 1839-T provide identical results in almost all cases. The reason lies in the very similar test apparatuses of the methods.
- The ASTM method yields similar explosion ranges in many cases. This can be explained by the use of a 5-dm³ flask in connection with the sensitive visual criterion. However, a major disadvantage of this method is the undefined step size in connection with the definition of the explosion limit (mean value between ignition and non-ignition point). This can easily result in unsafe explosion data if the concentration steps are not specified clearly.
- The closed vessel method EN 1839-B shows the strongest deviations. LELs are higher compared to the other methods and a clearly lower ethylene UEL was observed. The reason might be the pressure threshold criterion used that is obviously less sensitive than the optical criterion. Ammonia was an exception with a significantly higher UEL. The large quenching distance of ammonia may play a role, thus ammonia reactions in the 14-dm³ sphere are preferred opposed to smaller volumes.

In general, the deviations of difficult-to-ignite gases, e.g. ammonia and nitrogen mixtures, are the greatest. Such reactions are strongly affected by apparatus parameters.

4. Comparison of the flame propagation criterion and pressure rise criterion

Determination methods for explosion limits at elevated pressures need another criterion for a reaction. Usually a measured pressure rise (p_{ex}/p_0) in a closed ignition vessel is used. ASTM E918-83 (2011) uses a pressure rise $p_{ex}/p_0 > 7\%$, EN 1839-B a pressure rise $p_{ex}/p_0 > 5\%$ as the criterion. De Smedt & Berghmans (De Smedt

Table 1
Key characteristics of the most commonly used standard test methods for explosion limit determination.

	DIN 51649-1 withdrawn in 2004	EN 1839-T:2004	EN 1839-B:2004	ASTM E681-01
Explosion vessel	vertical glass cylinder, open $\varnothing = 60$ mm, H = 300 mm	vertical glass cylinder, open $\varnothing = 80$ mm, H = 300 mm	closed sphere or cylinder H/D = 1 to 1.5, V > 5 dm ³	flask, spherical V = 5 dm ³ (V = 12 dm ³)
Ignition source	induction sparks typical 0.5 s, min. 0.2 s	induction sparks typical 0.2 s, max. 0.5 s	induction sparks or exploding wire, E = 10 J–20 J	induction sparks or exploding wire
Criterion for ignition	visual, flame detachment	visual, flame detachment >100 mm or aureole H > 240 mm	pressure increase $\geq 5\%$ + ignition pressure in air	visual, flame propagation up to 13 mm to the wall (horizontal or vertical)
Increment	0.1–0.2 mol%, depending on the test substance fraction	10% (rel.) to 0.2 mol% (abs.), depending on the test substance fraction	10% (rel.) to 0.2 mol% (abs.), depending on the test substance fraction	freely selectable, to be specified in report if x > 10% (rel.) of the test substance fraction
Number of repetition tests	5	4	4	1
Explosion limit	last non-ignition point	last non-ignition point	last non-ignition point	mean value from non-ignition and ignition point

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