# The Influence of Strong Ignition Sources on the Explosion and Decomposition Limits of Gases

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Explosion and decomposition limits of flammable and chemically unstable gases were determined experimentally in a closed autoclave with an ignition energy higher than the standard 10 J – 20 J. The ignition source was a lightning arc caused by an exploding wire igniter as described in EN 1839 B. With a newly developed ignition system a graded ignition energy between 3 J and 1000 J was generated. Different types of gases were studied with this ignition system: methane as a typical fuel gas and reference gas for some standards for explosion limit determination, the refrigerant R32 (difluoromethane) as a mildly flammable gas with low burning velocity and high minimum ignition energy compared with methane as well as the chemical unstable gases acetylene, nitrous oxide and ethylene oxide, which can decompose explosively in the absence of air or other oxidizers. It was found that the influence of strong ignition sources on explosion and decomposition limits can be very different for different systems. In case of methane only the upper explosion limit was influenced significantly by the ignition energy, whereas the lower explosion limit was constant. In a standard test vessel with an inner volume of 14 $dm^3$ it was difficult to quantify the upper explosion limit of methane exactly with the strong ignition source, because the explosion pressure did not increase abruptly near the explosion limit, but steadily over a large concentration range. Probably a larger explosion vessel is more appropriate in this case. In case of R32 however, it was the lower explosion limit that was influenced significantly by the ignition energy and not the upper explosion limit. A particularly strong dependency from the ignition energy was found for the decomposition limits of the chemically unstable gases in nitrogen. Here special regard is necessary in practical applications, if uncommonly strong ignition sources cannot be excluded.

## 1. Introduction

Avoiding explosive gas mixtures in industrial processes is the most effective and most preferred measure of explosion protection. For determining the explosive range of fuel gases, explosion limits are determined experimentally. For this purpose, ignition tests are carried out, in which defined gas mixtures of fuel gas and air or another oxidizing component are prepared and ignited with an ignition source in a test apparatus. In the common international standards for explosion limit determination of gases (i.e. EN 1839, EN ISO 10156, ASTM E681 or ASTM 918) ignition sources with ignition energy in the range between 10 J and 20 J are used, usually a high-voltage spark or a fusing wire. This level of ignition energy is considered as sufficiently high for explosion limit determination, since firstly the energy released by most of the ignition sources present in practice, i.e. for example mechanically generated or electrical sparks, static electricity or smoking, is usually much lower (Crowl, 2003). Secondly, the dependency of explosion limits from the ignition energy is low, if the ignition energy is far above the minimum ignition energy of the fuel gas, which is lower than 1 mJ for usual fuel gases like hydrocarbons or hydrogen (Bartknecht, 1993). However, special regard might be necessary in evaluating explosion scenarios, in which strong ignition sources, like open fires, jet flames or high-voltage current must be considered. Especially the explosion ranges of fuel gases difficult to ignite, i.e. fuel gases with high minimum ignition energies, might be influenced significantly by the impact of strong ignition sources, so that the appropriateness of explosion limits determined according to the common standards must be put into question in these scenarios. Examples for explosive reactions that can only be triggered by ignition sources with ignition energies that are much higher than the minimum ignition energy of usual fuel gases are

combustion of mildly flammable refrigerants (JSRAE, 2017) and explosive decomposition of chemically unstable gases like ethylene oxide or acetylene in absence of air (Britton, 1990). In previous works the influence of the ignition energy on explosion limits have been studied more occasionally (Cashdollar et al., 2000; Mynarz et al., 2017). Mainly the usual hydrocarbons were studied in this context and for simulating strong ignition sources pyrotechnic squibs were used. A graded and accurate adjustment of the ignition energy is not possible with pyrotechnic squibs. Also, the minimum amount of energy is limited to about 50 J (Safekinex, 2002a). Using completely different types of ignition sources for the lower and the higher energy range complicates the comparability of the results. Moreover, another chemical reaction is introduced into the system by using chemical igniters potentially influencing the explosive gas reaction. In this work a newly developed ignition system was used for studying the influence of the ignition energy on the explosion and decomposition limits of different substances determined in a closed autoclave. The ignition system is based on the so-called "exploding wire igniter" described in EN 1839 (2017). It allows generating electric arcs with defined energies between 3 J and 1000 J that are well comparable to electrical switching and short-circuiting sparks, which frequently occur in switching contacts at higher currents. The ignition energy can be provided accurately, graded and with good reproducibility. Using this ignition system, the standard test method according to EN 1839 (2017) should be modified first. Particularly the ignition criterion must be adjusted if the usual ignition source is substituted by a much stronger ignition source. Then the influence of the ignition energy provided by an electric arc on the explosion limits of different types of gases should be studied systematically. The results obtained with highly flammable gases with comparatively low minimum ignition energy and high burning velocity should be compared to the results obtained with mildly flammable refrigerants of class A2L according to the standard ANSI/ASHRAE 34 "Designation and Safety Classification of Refrigerants" with a much higher minimum ignition energy and much lower burning velocity. Finally, the influence of the ignition energy on the decomposition limits of chemically unstable gases in absence of oxidizing components should be studied. Conclusions on the impact of strong ignition sources for evaluating explosion scenarios could be derived depending on the type of fuel gas involved.

## 2. Experimental methods

### 2.1 Test set-up

The test set-up was in accordance with EN 1839 – bomb method. The ignitions were carried out in a closed spherical autoclave with an inner volume of 14 dm$^3$ made of stainless steel. It has connections for filling, purging and evacuating. For homogenizing the gas mixtures, a pc-fan was installed inside the autoclave. All test substances used in the ignition tests had a purity of minimum 99.5 %. The compressed air used in the tests was dry and free from oil. In Figure 1 a schematic diagram of the test set-up is shown.
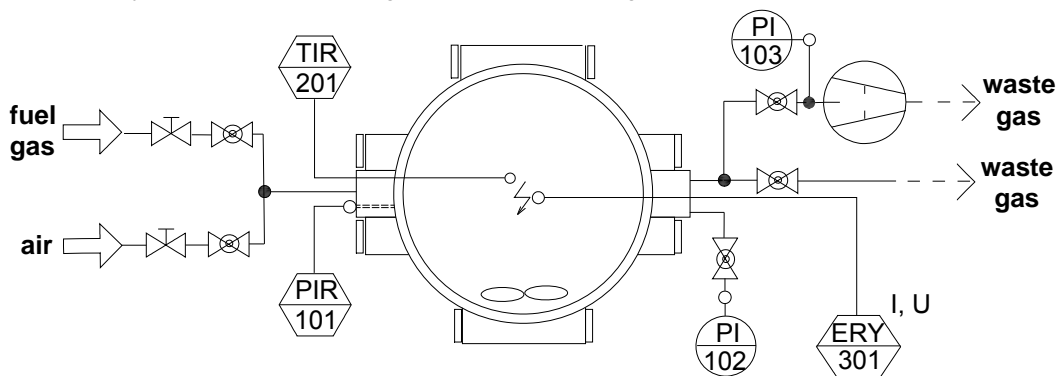


Figure 1: Schematic diagram of the test set-up

### 2.2 Measuring instruments

Pressure was measured by piezoresistive pressure sensors (company: Keller, model: PA-10L). The resolution of the pressure transducers is 10$^{-4}$ of full scale (FS). For preparing the gas mixtures, a pressure sensor with a measuring range up to 2 bar and a measuring uncertainty better than 0.2 % of full scale (FS) was used (PI 102 in Figure 1). For measuring the explosion pressures, a pressure sensor with a measuring range up to 10 bar and a measuring uncertainty better than 0.5 % of full scale (FS) was used (PIR 101 in Figure 1). Temperature was measured by a 1.5 mm coated thermocouple of type K installed in the centre of the autoclave (TIR 201 in Figure 1). The pressure and temperature signals were connected to an A/D-converter (company: Jet Systemtechnik GmbH, model: MCL-USB, 16 channels, 16 Bit A/D, sampling frequency: 500 kHz) and a pc for data recording and evaluating. The measuring frequency was set to 20,000 Hz within the

first 10 ms for measuring the ignition energy and then switched to 2,000 Hz for a measuring period of 4 s. For measuring the vacuum pressure, a Pirani gauge (company: Edwards, type: APG-M, PI 103 in Figure 1) with a measuring range from $1*10^{-3}$ mbar and a relative measuring uncertainty of 15 % was used.

## 2.3 Ignition system

The ignition source used in the tests was an exploding wire igniter as described in EN 1839 (2017). A 0.12 mm nichrome wire was used, which was soldered over the tips of two isolated brass electrodes in a distance of (5 – 8) mm. For ignition tests with an ignition energy of 1000 J, the distance was increased to (10 – 15) mm. The location for the ignition was in the centre of the vessel. With a newly developed ignition device, a portion of the energy stored in 450 V capacitors is switched to the electrodes via a choke coil by means of an electronic control circuit and two high-performance-thyristors. After about 0.3 ms the nichrome wire melts, evaporates and through the metal vapor, an electric arc burns between the electrodes with a burning voltage of (70 – 120) V and a current intensity of up to 1,600 A. The burning duration can be adjusted up to 10 ms. The arc is short-circuited and quenched by the delayed activation of another thyristor. The ignition energy is adjusted roughly by selecting the capacity of the energy store (battery of capacitors) and fine-tuned by adjusting the burning duration of the arc (time from activating the first thyristor until activating the second thyristor). Ignition energy from about 3 J to 1000 J can be provided by this ignition system. The ignition energy induced into the gas system (ERY 301 in Figure 1) is calculated from the measured current and voltage development during the ignition process.

## 2.4 Test procedure

In each test the test autoclave was evacuated first until the pressure was less than 1 mbar. Then the test gas mixture was prepared directly in the test autoclave according to the partial pressure method, i.e. each component of the gas mixture was filled according to its partial pressure. It was assumed that the partial pressure fraction of each component in the final mixture is equal to its molar fraction (ideal gas). After the last component was filled, the mixtures were homogenized by the installed pc-fan for minimum 3 min. Then the pc-fan was switched off and after a waiting time of another 30 s the igniter was triggered in the quiescent gas mixture. Simultaneously measure recording was started. The explosion pressure was determined after each test. An explosion was detected, if the explosion pressure exceeded a certain threshold value that was dependent from the ignition energy.

## 3. Test results

### 3.1 Definition of ignition criteria

According to EN 1839 a test gas mixture is defined as explosive if the measured overpressure in an ignition test is equal to or greater than 5 % of the initial pressure. This criterion shall comply with the criterion of a self-propagating combustion, which is the typical visual criterion for detecting explosions in open glass apparatuses (tube-method). In the closed system, the ignition source itself contributes to the overpressure by the heat input in each test. The extent of this contribution is dependent from the strength of the ignition source and from the vessel volume. Therefore, the overpressure caused by triggering the ignition source in the test autoclave without any fuel gases is determined in dependence from the ignition energy. This overpressure is subtracted in each ignition test. Apart from that, it must be considered that by the impact of the ignition source a part of the combustible in the direct vicinity of the ignition source is always converted without any self-sustaining flame propagation. The pressure rise caused thereby is also dependent from the vessel volume and from the strength of the ignition source. Therefore, the pressure rise threshold for detecting explosions must be dependent from the ignition energy. The ignition criteria used in this work are summarized in Table 1. For tests with an ignition energy of 15 J, the pressure rise threshold of 5% was used for detecting explosions as given in EN 1839 (2017). For the tests with an ignition energy of 1000 J the pressure rise threshold was increased to 50 % in accordance with standard EN 14034-3 (2011) for determining explosion limits of dusts, where a similarly strong ignition source is used. For validating the ignition criteria, pre-tests were carried out in a cylindrical windowed autoclave with an inner volume of 11 $dm^3$ recording the flame appearance by a video camera. The window allows comparing the pressure-threshold criterion with the visual criterion of a self-sustaining flame-propagation. In Figure 2 video sequences of ignitions with different ignition energy were carried out. In the first test (top row) a gas mixture with 15.4 mol% methane in air was ignited with an ignition energy of about 15 J. In the second test (bottom row) a gas mixture with 25.0 mol% methane in air was ignited with an ignition energy of about 1000 J. The pressure rise measured was very similar in both cases. But in the first case, a clear flame can be observed detaching from the ignition source, propagating upwards to the vessel wall, finally sustaining for more than 1.4 s in the upper part of the vessel and even beginning to propagate downwards. However, in the second case a much bigger flame kernel can be observed appearing

soon after triggering the ignition source and extinguishing within 0.5 s without showing a clear upward propagation or a clear movement at all. It was concluded that a pressure rise threshold as high as 50 % for ignitions with an ignition energy of 1000 J as taken from the standards for determining explosion limits of dusts is reasonable. Also, it was observed that with higher ignition energy, the transition from "no flame propagation" to "flame propagation" becomes very unsharp, especially near the upper explosion limit of methane. So finally, the definition of a clear visual ignition criterion would not be possible.

*Table 1: Ignition criteria used in this work*

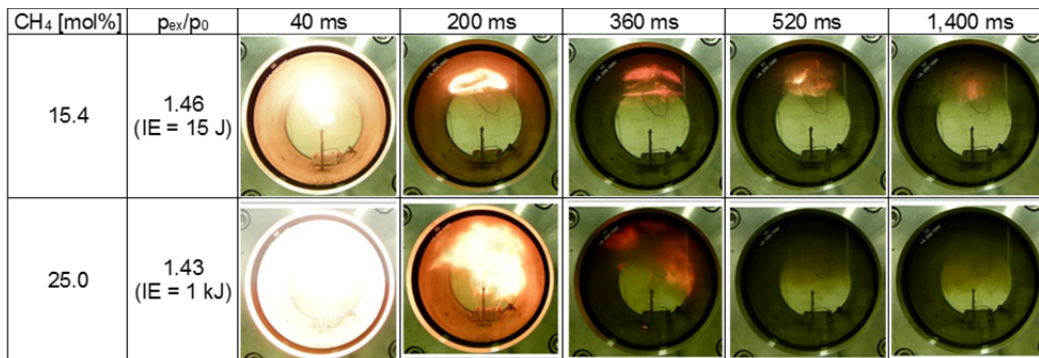| Ignition energy | Normalized overpressure caused by the ignition source ($p_{IS}/p_0$) | Explosion criterion (pressure rise threshold) |
|---|---|---|
| (10 – 20) J | 1,010 | $p_{ex}/p_0 \geq (1{,}05 + p_{IS}/p_0 - 1)$ |
| (75 – 125) J | 1,016 | $p_{ex}/p_0 \geq (1{,}15 + p_{IS}/p_0 - 1)$ |
| (900 – 1100) J | 1,135 | $p_{ex}/p_0 \geq (1{,}50 + p_{IS}/p_0 - 1)$ |



*Figure 2: Video sequence of ignitions in a windowed autoclave with an ignition energy of 15 J (top row) and 1000 J (bottom row) with similar explosion pressure ratio*

### 3.2 Influence of the ignition energy on explosion limits

In Figure 3 the explosion pressure ratio ($p_{ex}/p_0$) in dependence from fuel gas fraction is shown for ignitions of methane/air- and R32-/air-mixtures (i.e. difluoromethane/air-mixtures) near the explosion limits with ignition energy of 15 J, 100 J and 1000 J.
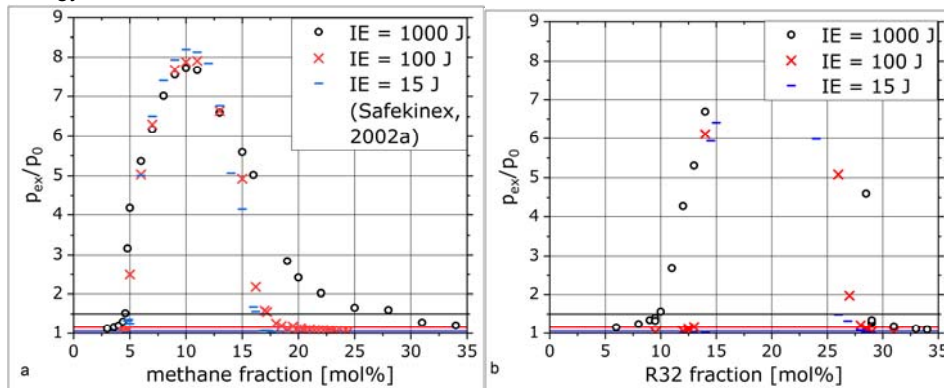


*Figure 3: explosion pressure ratios ($p_{ex}/p_0$) measured in ignitions of methane/air- (a) and R32/air-mixtures (b) with different ignition energies in dependence from the fuel gas fraction in air*

No significant influence of the ignition energy on the lower explosion limit (LEL) of methane could be found. The LEL varies between 4.6 mol% and 4.8 mol%, which complies with the values given in EN 1839 (2017) for verification of the test apparatus with the reference substance methane. Independent from the ignition energy $p_{ex}/p_0$ increases strongly even if the LEL is exceeded only slightly. Near the upper explosion limit (UEL) of methane the situation is very different. As already observed in previous works (Safekinex, 2002b), near the UEL determined with the standard ignition energy of 15 J, there is a wider concentration range or plateau with "weak" explosions, i.e. explosions with explosion pressure ratios between about 1.05 and 2.00. This concentration range becomes much larger with increasing ignition energy what makes the applied explosion criterion of exceeding a certain explosion pressure threshold even more vague. The same was observed for a

visual explosion criterion before (see chapter 3.1). But no matter how an explosion is defined exactly, it is evident that the UEL of methane is much stronger dependent from the ignition energy than the LEL. Applying the ignition criteria defined in Table 1, the UEL increases from about 17 mol% to 20 mol% and up to about 30 mol% if the ignition energy is increased from 15 J to 100 J and up to 1000 J. Compared to the results with methane the concentration range with "weak" explosions near the upper explosion limit is much closer for R32. Unlike methane it was also found in a similar extent near the lower explosion limit of R32 and it was nearly the same for different ignition energies. Also, in contrast to the results with methane no significant influence of the ignition energy on the upper explosion limit, but a clear decrease of the lower explosion limit with increasing ignition energy was found, from 14.0 mol% at 15 J to 9.5 mol% at 1000 J applying the ignition criteria in Table 1.

### 3.3 Influence of the ignition energy on the decomposition limits of chemically unstable gases

Under the impact of an ignition source chemically unstable gases can decompose explosively in absence of air or other oxidizing components. The decomposition limits are defined as the maximum concentrations of the chemically unstable substances in mixture with an inert gas, at which the mixture cannot be ignited. The decomposition limits of the chemically unstable substances nitrous oxide, ethylene oxide and acetylene were determined at different levels of ignition energy with nitrogen as inert gas. The results are shown in Figure 4 and in Table 2. In most cases, a massive influence of the ignition energy on decomposition limits was found.
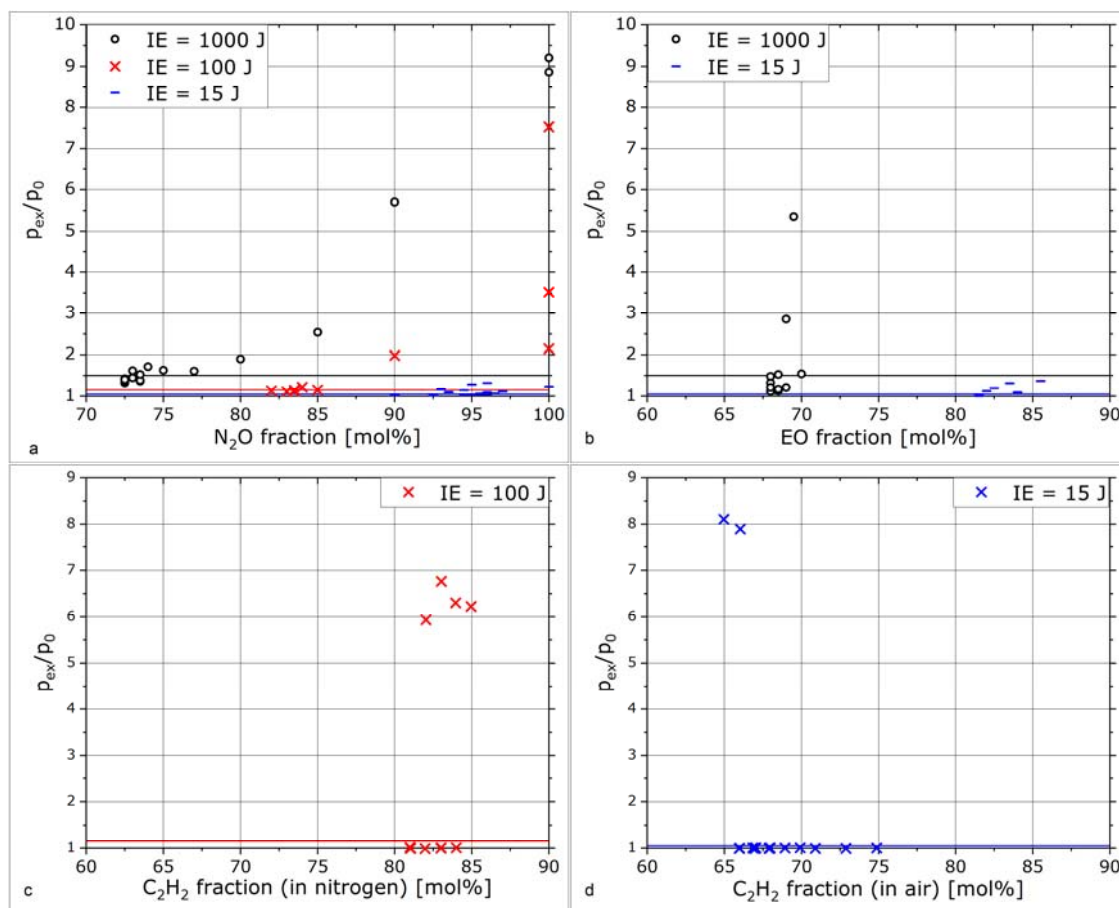


Figure 4: explosion pressure ratios ($p_{ex}/p_0$) in dependence from the mixture composition measured in ignitions with different ignition energies for binary mixtures of the chemically unstable gases nitrous oxide (a), ethylene oxide (b) and acetylene (c) with nitrogen respectively and for the binary mixture of acetylene and air (d)

Nitrous oxide for example is difficult to ignite with a standard ignition source of 15 J according to EN 1839. A decomposition limit of 92.5 mol% was found, but at atmospheric conditions the explosions are very weak. Even in pure nitrous oxide the explosion pressure was less than 1.3 bar at an ignition energy of 15 J. However, with increasing ignition energy, the decomposition limit decreases strongly, and also much more violent explosions are possible with explosion pressures up to nearly 10 bar. Pure acetylene could not be ignited at all with the standard ignition energy of 15 J in the closed vessel. If air was added an upper explosion

limit was found at 67 mol%. If the ignition energy was increased to about 100 J, explosive decomposition was possible in pure acetylene without air and a decomposition limit of 81 mol% in nitrogen was found. In contrast to all other tested substances not a single "weak explosion" was observed in the tests with acetylene. Either there was no pressure rise at all or an explosion pressure of minimum 6 bar was measured accompanied by soot formation. In pure ethylene oxide a clear explosive decomposition with an explosion pressure of several bar is already possible with a standard ignition energy of 15 J. With increasing ignition energy in the range between 15 J and 1000 J, the decomposition limit decreases clearly from 81.5 mol% to 68.0 mol%. The distinction between "ignition" and "no ignition" based on the explosion pressure is also very clear for ethylene oxide, irrespective of the ignition energy.

*Table 2: Decomposition limits of chemically unstable gases in nitrogen in dependence from the ignition energy*

| Ignition energy | 15 J | 100 J | 1000 J |
|---|---|---|---|
| ethylene oxide | 81.5 mol% | | 68 mol% |
| nitrous oxide | 92.5 mol% | 83.5 mol% | 72.5 mol% |
| acetylene | *UEL = 67 mol%** | 81 mol% | |

*no explosive decomposition could be triggered in pure acetylene or in acetylene/nitrogen mixtures in the closed autoclave. Instead the upper explosion limit in air was measured.

## 4. Conclusions

With stronger ignition sources, the explosion range of flammable substances can be larger than the explosion range determined with the usual ignition sources according to the international standards, which provide an ignition energy in the range between 10 J – 20 J. The impact of strong ignition sources on explosion limits can be very different. The lower explosion limit of methane for example is not influenced significantly by strong ignition sources, whereas the upper explosion limit is increased. However, in case of R32 (difluoromethane), which has a much higher minimum ignition energy and a much lower burning velocity than methane, it is the lower explosion limit that is shifted under the impact of a strong ignition source, whereas the upper explosion limit is nearly constant. However, in some cases like at the upper explosion limit of methane, it is difficult to quantify exactly the upper explosion limit with a standard apparatus and a strong ignition source, because within a larger concentration range the explosion pressure and the observable extent of the flame propagation increase only slightly at the upper explosion limit of methane and not abruptly. For ammonia this behaviour can also be observed, if the explosion limits are determined with the standard ignition energy of 10 J – 20 J, because of the formation of halos. Probably, in a larger test vessel the upper explosion limits can be determined much clearer in these cases. The influence of the ignition energy on the decomposition limits of chemically unstable gases seems to be particularly high. In case of acetylene or nitrous oxide it was not even possible to trigger a clear explosive decomposition with a considerable explosion pressure with a standard ignition energy of 10 J – 20 J. But if the ignition energy was increased to just 100 J violent explosions were observed. Consequently, special regard is necessary, if explosion scenarios are to be evaluated, in which stronger ignition sources cannot be excluded and chemically unstable gases are involved. Also, it is open to discussion, if it is appropriate to use stronger ignition sources for the determination of decomposition limits.

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