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Explosion protection in biogas and hybrid power plants

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Abstract

Renewable energies became more and more important in the last years. The production of biogas using agricultural waste and the use of wind and solar energy in combination with water electrolysis is one way to substitute natural gas. Therefore the number of biogas plants is growing very fast in Germany. In the meantime, the operation of such plants is responsible for a significant number of accidents. New safety regulations on biogas plants and a short statistical summary of accidents in Germany are presented in the first part of this presentation. The main focus of the paper is the principle hazards arising from the substances and materials in biogas and hybrid power plants. Primarily, these are the hazards of fire and explosion induced by flammable methane gas. However, further hazards are the dangers of asphyxiation and poisoning by gases such as carbon dioxide, hydrogen sulphide and ammonia. Furthermore, hydrogen is produced by water electrolysis in hybrid power plants and mixed with bio methane in some cases. In order to prevent explosions when handling biogas and hydrogen it is necessary to know the explosion limits of gas and gas mixtures in mixture with air. However, biogas from agricultural plants can vary significantly in its composition. Therefore, for each gas composition the explosion limits would have to be determined. This would require a considerable amount of time and effort. Due to this fact, the explosion limits of biogas are frequently referred to only by the methane fraction of the gas mixture in the safety-relevant literature. In reality as biogas can consist of methane, carbon dioxide and further residual gases the explosion limits are generally over or underestimated. A calculation method for explosion limits was developed by means of explosion diagrams to avoid such errors. In a last topic methods are shown for the calculation of gas spreading in case of leakages in gas buffers for risk evaluation and land use planning. For this purpose the German directive VDI 3783 was evaluated.

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Keywords: Biogas; Hybrid Power Plants; Explosion Protection; Safety Characteristics; Technical Regulations

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1. Introduction

Biogas is an important source of renewable energy. It consists mainly of methane and carbon dioxide, with low levels of hydrogen sulphide and other gases. That means that the toxicological aspects and the flammability of biogas have to be taken into account. The number of biogas plants and the mean electric power per plant has increased significantly during recent years, see Fig. 1. In total an electric power output of about 3500 MW was installed in Germany in 2013. Part of this was made up by a considerable number of small biogas plants. However, such simple small plants in farms are often sources of accidents because they do not meet the criteria of the German Hazardous Incident Ordinance (Störfallverordnung) which requires monitoring by the authorities. Nevertheless, these plants should also be checked to avoid pollution and the risk of explosion. According to the European Directive 1999/92/EC (ATEX 137) and the German hazardous substances ordinance (Gefahrstoffverordnung) and the German Ordinance on Industrial Safety and Health (Betriebssicherheitsverordnung) operators of biogas plants must meet explosion prevention measures, e.g. the avoidance of uncontrolled explosive atmospheres (primary explosion prevention), the avoidance of ignition sources (secondary explosion prevention) and/or constructive explosion protection. Therefore, an explosion protection document must be provided and ATEX zones must be defined.

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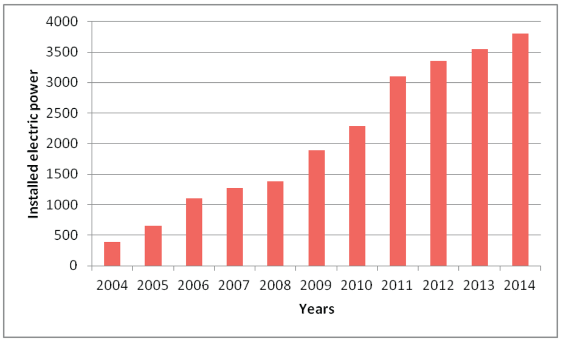

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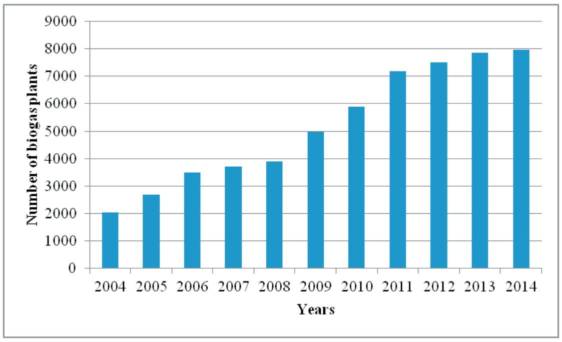

Fig. 1. (a) Number of biogas plants in Germany; (b) total installed electrical power in MW – 2014 Forecast [1].

That's why characteristics for primary explosion protection, the explosion limits and explosion ranges of biogas are in focus of this paper.

In addition to biogas, the water electrolysis has been considered as a component of power-to-gas technology in hybrid power plants. This technology has been developed to utilize excess electrical energy from wind turbines and solar power. A safety problem of electrolysis is impurities in product gases H_2 and O_2. Therefore BAM investigated the pressure dependence of explosion limits of hydrogen in oxygen.

Explosion (flammability) limits of hydrogen-methane mixtures at different initial pressure were of interest for the mixtures of hydrogen with bio methane. It was shown, that Le Chatelier's mixing rule fails in a number of cases for calculation of explosion limits on mixtures of hydrogen and methane.

In the last part of this paper information are given about possible leakages with resulting gas spreads. Here the risk of explosion was compared with the risk of poisoning using the explosion limits giving by this paper and toxicity thresholds.

2. Accidents on Biogas Plants and new regulations for explosion protection

The growing number of biogas plants and the diversity of technical rules in German federal states have led to a increasing number of accidents at these plants. Table 1 shows examples of some accidents in the years from 2005 - 2008.

Table 1. Examples of accidents in biogas plants in Germany 2005 – 2008 [2].

Date	Part of plant	City	Type of accident	Death/Injured people
21.05.2008	fermenter	Kogel	fire	0/0
05.02.2008	fermenter	Bad Sassendorf	release of manure	0/0
15.01.2008	cesspool	Henglarn	release of manure	0/0
16.12.2007	fermenter	Riedlingen	explosion	0/0
13.07.2007	cesspool	Eglingen	release of manure	0/0
14.06.2007	silo	Binsheim	explosion	0/2
12.04.2007	biogas plant	Herrieden	explosion	0/2
04.04.2007	electric generator	Reken	fire	0/0
27.03.2007	gas buffer	St. Magarethen	explosion	0/0
24.03.2007	biogas plant	Bertlich	release of gas	0/0
12.03.2007	biogas plant	Babst	explosion	0/0
01.12.2006	cesspool	Natzungen	release of manure	0/0
07.04.2006	building	Volkmarst	fire	0/0
29.02.2006	power house	Garmhausen	fire	0/0
13.02.2006	power house	Ziertheim	fire	0/0
29.01.2006	fermenter	Lelbach	release	0/0
21.01.2006	power house	Göttingen	explosion	0/0
01.01.2006	pipes	Schettau	release	0/0
08.11.2005	biogas plant	Rhadereistedt	release of gas, poisoning	4/13
22.10.2005	silo	Soltau	fire	0/0
01.03.2005	biogas plant	Nusbaum	explosion	0/0

Due to this large number of accidents, the Commission for Plant Safety (KAS) of the German Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety (BMU) has focussed on biogas plants. Between 2001 and 2006 KAS examined 115 biogas plants. Significant defects were found in 80% of the 115 plants checked, with the majority of the defects being in the field of gas explosion protection [3]. Examples included:

- Wrong classification of ATEX zones
- Absent or faulty safety flares
- Non-adequate safety distances between the gas reservoir and power station
- Unacceptable air systems and explosion protected electric equipment
- The absence of emergency planning for fire brigades and missing information for employees

A number of new rules and guidelines were created in Germany for biogas plants, a part of them in the field of explosion protection. The KAS published special instructions for biogas plants [4]. Currently a new technical rule to the German hazardous substances ordinance (Gefahrstoffverordnung) is prepared for biogas. Furthermore, the German regulations for explosion protection (Ex-RL) have got a new section for biogas [5].

3. Experimental

The explosion limits at atmospheric conditions were determined with an apparatus in accordance with DIN 51649-1. Since 2004 this standard has been replaced by the EN 1839 “Tube method”. Both standards recommend almost identical test equipment. The new European standard recommends a little larger inner diameter for the ignition vessel (80 mm instead of 60 mm by DIN). Nevertheless, the larger diameter does not affect the results for the mixtures of methane and carbon dioxide reported here.

In Fig. 2 a tube apparatus can be seen for determination of the flammability limits according to EN 1839 (DIN 51649-1), as it is used at BAM. The gas mixture is metered by thermal mass flow controllers at different nominal volume flow rates. For the corresponding gas, the mass flow controller must be calibrated by a precision gas meter. The gas is piped through a mixing vessel into the ignition vessel. After purging the ignition vessel, the gas is ignited by using high voltage sparks and a visual flame observation is made. The criterion for an ignition is flame detachment and spreading from the electrodes through the ignition vessel in the test mixture. The explosion regions of gas mixtures have been determined by varying the percentages of flammable gas, inert gas and air, respectively. The increment in the determination of the flammability limits was 0.2 mol% methane. For the determination of limiting oxygen concentration a step size of 0.5 mol% air was used.

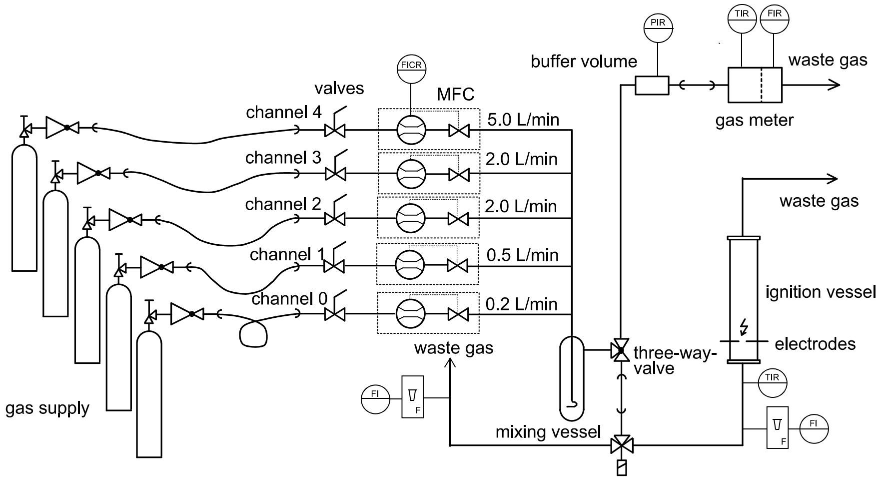

Fig. 2. BAM apparatus according to DIN 51649 and EN 1839 “Tube method”.

The explosion limits at higher initial pressures and with hydrogen-oxygen mixtures were measured according to the bomb method of EN 1839, using 3 dm^3 and 6 dm^3 vessels made of stainless steel, see Fig. 3. The gas mixture is ignited by an exploding wire with ignition energies between 10 J and 20 J. Criterion of an explosion is a pressure rise of more than 5% after ignition.

4. Safety related properties of biogas

4.1. Chemical composition

Biogas is produced by fermentation of biodegradable waste or other organic materials. It consists mainly of methane and carbon dioxide, with low levels of hydrogen sulphide and other gases. The composition of biogas varies strongly depending on the introduced organic materials and on the biological process. Raw biogas usually consists of the following components [6]:

- Methane (50 mol% to 80 mol%)
- Carbon dioxide (20 mol% to 50 mol%)
- Water vapor (0 mol% to 12 mol%)
- Nitrogen (0 mol% to 5 mol%)
- Oxygen (0 mol% to 2 mol%)
- Hydrogen sulfide (0.01 mol% to 0.4 mol% (100 to 4000 ppm(v)), and traces of ammonia, hydrogen and higher hydrocarbons.

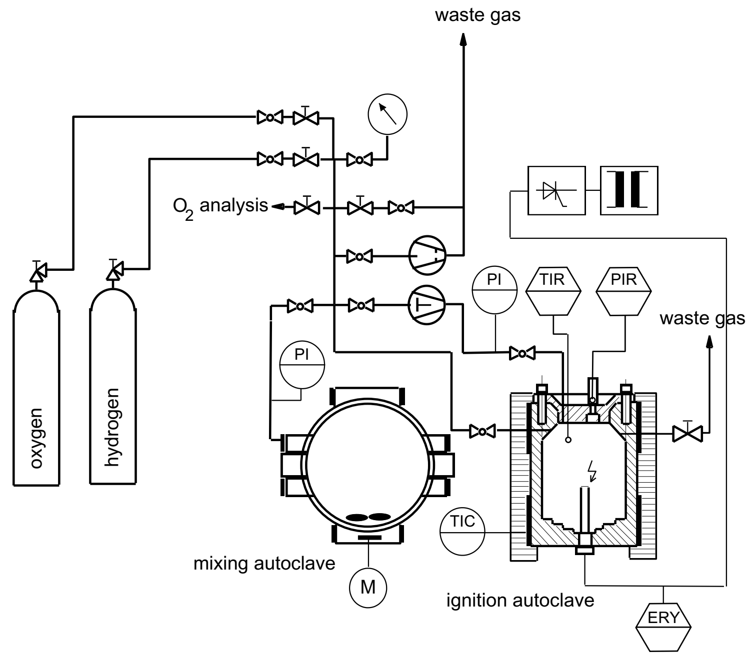

Fig.3. BAM apparatus according to EN 1839 “Bomb method”.

Other sources stated maximum H_2S amounts up to 2 mol%. From the information on accidents at biogas plants presented in Table 1, a number of hazards related to uncontrolled releases of gases and liquids can be grouped into the following categories:

- fire and explosion
- poisoning
- asphyxiation
- damage of the environment by the release of liquid manure

At this juncture the danger of poisoning, particularly by hydrogen sulphide, should be mentioned. This toxic gas was responsible for the 4 deaths and 13 injuries during the Rhadereistedt incident in 2005. In Table 2 the toxic effects of H_2S to humans are shown depending on the gas concentration and exposure time to adverse effects.

Biogas which contain significant amounts of hydrogen sulphide have to be regarded as toxic gas and have to classify according to the Regulation (EC) No 1272/2008 and the German hazardous substances ordinance. The toxic effects are summarized in Table 2.

Table 2. Toxic effect of hydrogen sulphide [6].

Gas concentration	**Exposure time**
< 100 ppm	dangerously after several hours
100 ppm ≤ x < 500 ppm	dangerously after 30 min to 1 h
~ 500 ppm	life-threatening after 30 min
~ 1000 ppm	life-threatening in a few minutes
~ 5000 ppm	lethal in a few seconds

4.2. Safety characteristics

The safety characteristics of hazardous materials are the basis of primary explosion protection and of an explosion protection document. In the case of biogas plants these are usually the explosion limits of biogas-air mixtures. The explosion limits of pure substances can be found in data books or databases. Safety characteristics are, however, not pure physicochemical properties like the mass or the density, but depend also on the experimental test methods, therefore giving rise to a diversity of data found in the literature. For explosion protection documents, only characteristics should be used, which were determined in standardised test procedures. If possible, the applicability of these data for biogas plants should be examined by experts.

The database CHEMSAFE [8] published by DECHEMA, BAM and PTB (Physikalisch Technische Bundesanstalt) provides information on the safety characteristics of chemicals. This information is annually evaluated by experts of both BAM and PTB to ensure it is up-to-date. CHEMSAFE contains safety characteristics for more than 3000 flammable substances and approximately 2800 mixtures of gases, liquids and dusts. The temperature and pressure influence on safety properties are also taken into account, with explosion limits reported up to 400 °C and 10 MPa.

The most important safety characteristics of biogas components are summarized in Table 3.

Table 3. Safety characteristics of biogas components [6, 7].

Characteristic	**Unit**	**Methane**	**CO_2**	**H_2S**	**NH_3**	**H_2**
Lower explosion limit (LEL)	vol% (mol%)	4.4	n.f.	3.9	15.4	4.0
Upper explosion limit (UEL)	vol% (mol%)	17.0	n.f.	50.2	33.6	77.0
Auto ignition temperature	° C	595	-	270	630	560
Minimum ignition Energy	mJ	0.29	-	n.k.	14	0.017
MESG	mm	1.14		0.83	3.18	0.29
Explosion group		2A	-	2B	2A	2C
Temperature class		T1	-	T3	T1	T1
Acute Toxicity (LC50)	ppm(V)	n.t.	> 8 vol%*)	712	7338	n.t.
Gas density	rel. to Air	0.55	1.53	1.19	0,60	0.07

(n.f. = non flammable, n.t. = non toxic, n.k. = not known *) hazardous to health

Many of the characteristics of biogas are influenced by its main components. For example the flammable characteristics are similar to those of methane, except for the upper explosion limit (UEL). The toxicity of biogas is strongly influenced by the percentage of H_2S.

4.3. Explosion ranges of biogas

For the explosion protection in biogas plants, it is often necessary to know the exact value of explosion limits of the process gases in air. The composition of biogases from agricultural plants varies significantly, so for each composition the explosion limits should be determined experimentally. However, this would need a considerable amount of time and effort. A method is presented here which shows that the explosion limits of biogases with known compositions can be assessed with the help of two explosion diagrams [8]. In addition to the methane and carbon dioxide content of the biogas the influence of water vapor content in the system is also considered.

The upper (UEL, fuel-rich) and lower explosion limits (LEL, fuel-lean) are referred to as the mole fractions (concentration) of the biogas in air at which flame propagation after spark ignition no longer occurs (the last non-ignition point). The range between the lower and upper explosion limits is known as explosion range. The area restricted by the lower and upper explosion limit curves in a fuel-air-inert gas system is called the explosion region. Mixtures with a composition in the explosion region are considered explosive and dangerous.

The explosion limits for pure substances, such as methane, can be taken from data books and databases. For more complex mixtures such as "biogas" this data is not readily available, so, in practice the explosion limits of methane (see Table 3) are frequently used. At the lower explosion limit this is acceptable, because methane has a lower LEL than biogas. At the upper explosion limit, such as by air inlet into biogas, this can lead to dangerous conditions. In this case the UEL of biogas can be higher than the UEL of methane, so that the acceptable air content must be more restricted.

If the composition is known, the explosion limits of biogas can be estimated with the help of so-called "explosion diagrams". The explosion ranges can be represented in different types of explosion diagrams, the most commonly used are triangular or Cartesian coordinates. A collection of such explosion diagrams of fuel-inert gas-air mixtures, as measured according to the German standard DIN 51649 have been published [9].

The explosion regions of ternary mixtures can be represented most clearly in the so-called "triangular diagrams". Here each side corresponds to a binary mixture and the three corners correspond to the respective pure substances. The composition of a mixture can be read directly from the diagram. The explosion diagram of methane/carbon dioxide/air in Fig. 4 is an example of this type of diagram. The LEL and UEL of methane in air are read off on the left side of the diagram. The values are LEL = 4.3 mol% and UEL = 16.3 mol% in this case. The air fraction at the apex of the explosion region is used to determine the limiting oxygen concentration (LOC). The air fraction is found with 65 mol%. This corresponds to an oxygen concentration of 13.6 mol%.

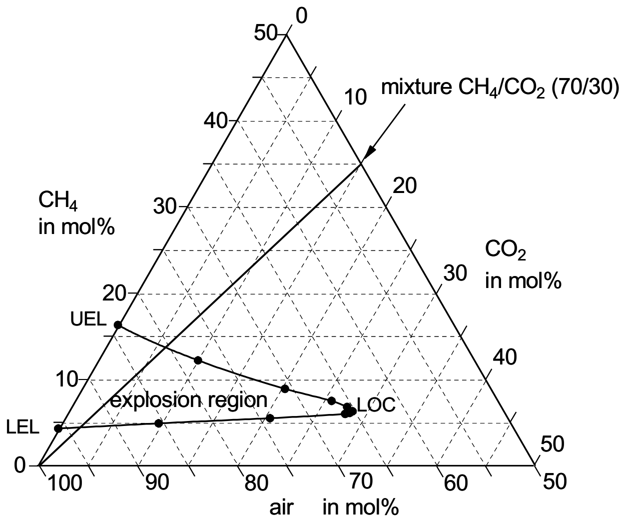

Fig. 4. Explosion region of methane-carbon dioxide-air mixture (acc. to DIN 51649,20 °C, atmospheric pressure).

Some basic statements can already be made by using these key data. In case of leakages and the accidental release of biogas into the surrounding atmosphere an explosive mixture can only be formed if the lower explosion limit of methane is exceeded. Furthermore, it is only possible to obtain an explosive mixture in case of ingress of air into a biogas vessel, if the LOC is exceeded. The LOC of 13.6 mol% refers to a dry biogas, in which the water vapor portion is negligible. For the digesters, in which the biogas contains major portion of water vapor at temperatures up to 50 °C, the LOC drops down to 12.4 mol%. This value is determined from the methane-water vapor-air explosion diagram [9].

The straight line shown in Figure 4 corresponds to a biogas with a methane content of 70 mol% and a carbon dioxide content of 30 mol%. During addition of air to this mixture the composition of the mixture moves from the right side along the straight line through the explosion region to the lower left corner which corresponds to pure air. The flammability limits (explosion range) of this type of biogas (70/30) can be obtained from the intersection points of the line with the explosion region.

4.4. Calculation of explosion limits

In good approximation the lower percentages of other fuel gases which are present in biogas can be added to the methane portion for the calculation procedure. The flammability limits can be determined from the explosion region in Fig. 4 for dry biogas with negligible portions of water vapor and oxygen. For better representation of the intersection points with the explosion region a diagram with Cartesian coordinates is often used instead of a triangular diagram. This representation has the advantage that the axes are scalable, and the explosion region can be displayed in optimum size. Moreover, most graphic and spreadsheet programs do not deliver triangle coordinates by default. A disadvantage of this Cartesian type diagrams is, however, that the representation of the axis for the third component is missing. Therefore, the portion of air in a mixture can be gained by subtracting the portions of methane and carbon dioxide from 100 mol%. In Fig. 5 two biogas compositions with methane/carbon dioxide (70/30) mol% / mol% and (50/50) mol% / mol% are represented. The flammability limits of biogases are calculated with the help of this diagram by using the mole fractions of methane and carbon dioxide according to equation (1) and (2).

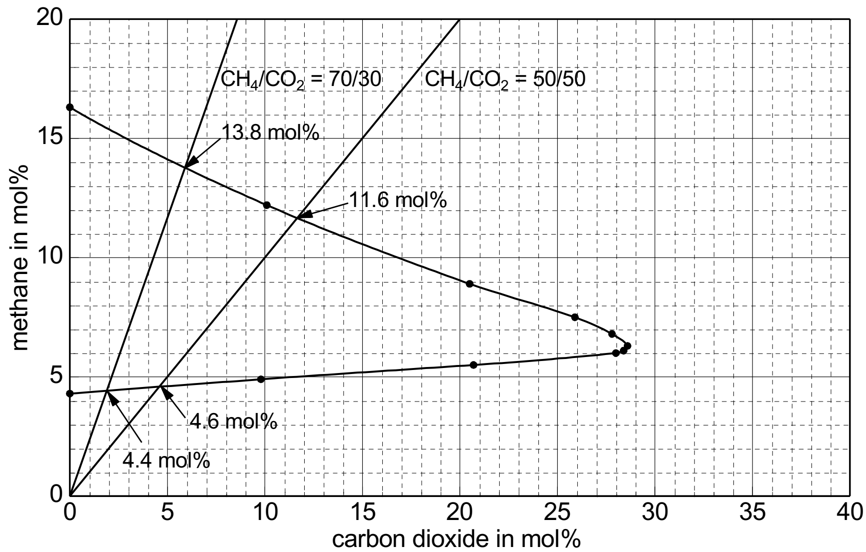

Fig. 5. Explosion region from Figure 4 in Cartesian coordinates.

$$\mathrm{LEL}_{Biogas} = \left(1 + \frac{x_{CO2}}{x_{CH4}}\right) \cdot \mathrm{LEL}_{CH4,CO2} \tag{1}$$

$$\mathrm{UEL}_{Biogas} = \left(1 + \frac{x_{CO2}}{x_{CH4}}\right) \mathrm{UEL}_{CH4,CO2} \tag{2}$$

where:

LEL_{Biogas}, UEL_{Biogas}	Lower/Upper explosion (flammability) limit of biogas
x_{CO2}, x_{CH4}	fractions of CO_2 and CH_4 respectively
$\mathrm{LEL}_{CH4,\ CO2}$, $\mathrm{UEL}_{CH4,\ CO2}$	methane fraction at the intersection point with the boundary curves

These equations resulted in the following lower explosion limits:

LELBiogas 70/30 = (1 + 30/70) 4.4 mol% = 6.3 mol%

LELBiogas 50/50 = (1 + 50/50) 4.6 mol% = 9.2 mol%

and the upper explosion limits for the sample mixtures

UELBiogas 70/30 = (1 + 30/70) 13.8 mol% = 19.7 mol%
UELBiogas 50/50 = (1 + 50/50) 11.6 mol% = 23.2 mol%

If these calculations are carried out for several mixtures, a diagram can be constructed as shown in Fig. 6. From this diagram the flammability limits of a biogas can be determined depending on the methane concentration.

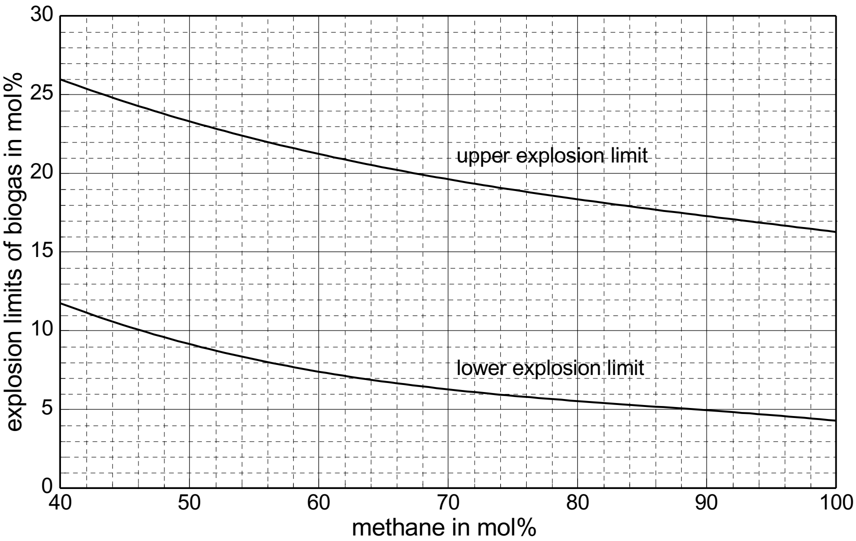

Fig. 6. Explosion limits of biogas at known methane content.

5. Power to gas – Water electrolysis

It is energetically advantageous to use electrolysers that can operate at high pressures, e.g. high pressure electrolysers with PEM or with alkaline electrolyte. At first, electric energy for compression of the hydrogen can be saved, on the other hand, the electrical resistance of the electrolyte is lower at high pressures. The reason is the smaller volume of the gas bubbles in the liquid electrolyte at high pressures. Due to the higher diffusion of the product gas through the membrane a contamination of the hydrogen with minor amounts of oxygen and vice versa is observed [11]. To ensure a safe process, the pressure dependence of the explosion limits of hydrogen-oxygen mixtures is of interest, see Fig. 7.

At atmospheric pressure, the slightest impurities of hydrogen in oxygen (LEL) are possible. The LEL increases with higher initial pressures. At the UEL the possible amount of oxygen (impurities of oxygen in H_2) increases from 4.8 mol% (100 kPa) to 5.8 mol% (1000 kPa) and decreases back to 5 mol% at 15000 kPa.

6. Hybrid power plants – Mixtures of hydrogen and biomethane

When hydrogen is mixed with natural gas or biomethane, the explosive limits of such hydrogen-methane mixtures are of interest. In many cases, if the explosion limits of the components are known, the explosion limits of the mixtures are calculated using Le Chatelier's approach

$$EL_G = \frac{100}{\sum \frac{x_i \cdot 100}{EL_i}} \tag{3}$$

where:

EL_G Explosion limit of the mixture of flammable gases [mol%]
EL_i Explosion Limit of flammable gas I [mol%]
x_i molecular fraction of flammable gas i

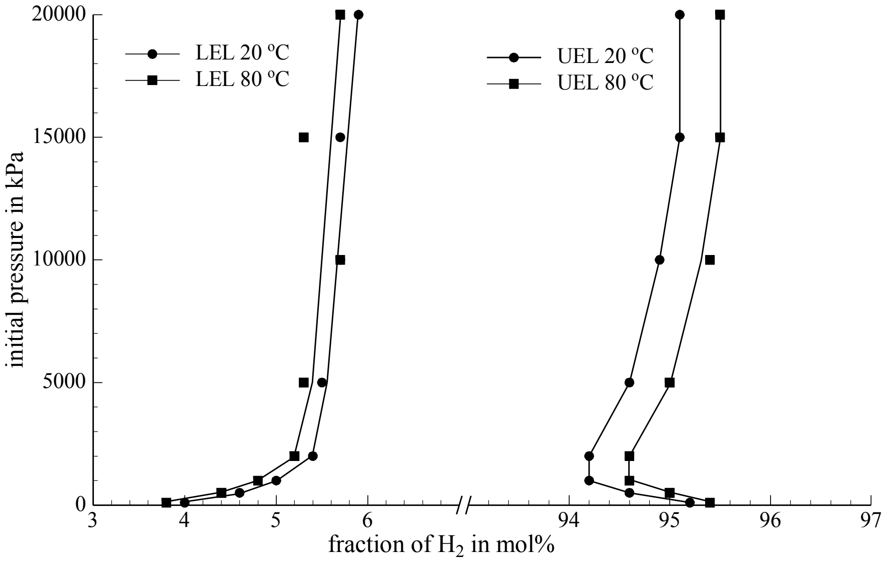

Fig. 7. Explosion limits of hydrogen in oxygen at different initial pressures

This approach is based on the assumption that the combustion enthalpies can be added and the flame temperatures and molar heat capacities at the explosion limits do not change in the mixture. In general, these conditions are fulfilled at the LEL better than at the UEL. Furthermore, the combustion kinetics may not be influenced significantly by the other components. This is often fulfilled with chemically similar fuels.

The explosion ranges for mixtures of hydrogen and methane have been determined experimentally in BAM. Fig. 8 shows the lower explosion limits of mixtures. In contrast to the fuel gas–inert gas-air diagrams both fuel gases, hydrogen and methane, have been illustrated in the triangle diagram. In Fig. 8 (LEL) the explosive mixtures are limited to the right by the plotted lines. In Fig. 9 (UEL) the plotted lines limit the explosive mixtures to the left. The straight lines represent the calculated values according to Le Chatelier's law.

The figures 8 and 9 show that there are significant deviations from Le Chatelier's rule for mixtures of hydrogen and methane. The lower explosion limits of mixtures at atmospheric pressure can still be calculated in good agreement. At initial pressures of 1000 kPa bar and 10000 kPa, the differences to the measured values are already significant higher (see Fig. 8). However, the calculated values are conservative according to safety related aspects (smaller LELs). At the upper explosion limits significant deviations are found, already at atmospheric pressure. In addition, the calculated values are within the explosion range and can lead to dangerous situations (see Fig. 9).

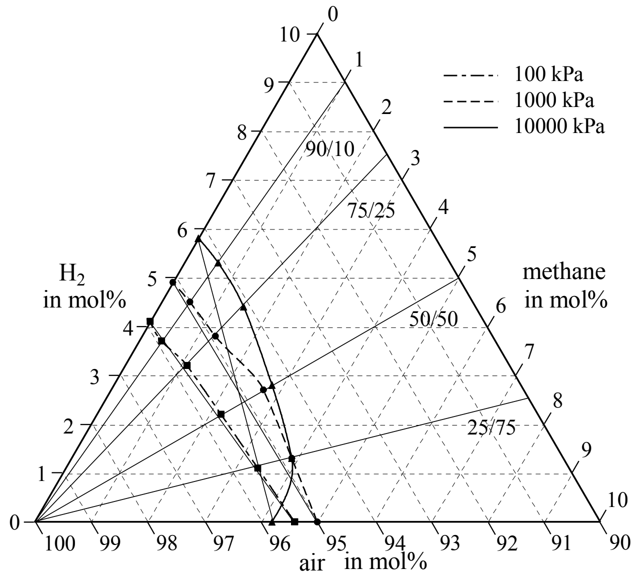

Fig. 8. Lower explosion limits of hydrogen-methane mixtures at 20 °C and pressures of 100, 1000 and 10000 kPa.

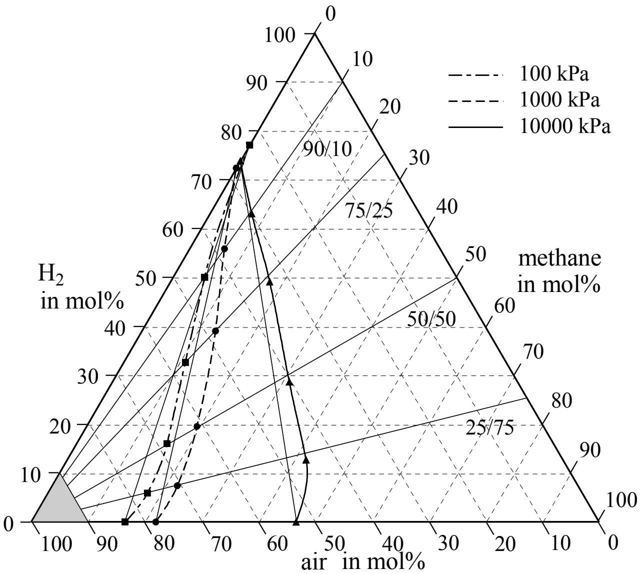

Fig. 9. Upper explosion limits of hydrogen-methane mixtures at 20 °C and pressures of 100, 1000 and 10000 kPa.

7. Calculation of safety distances for biogas plants in Germany

According to requirements of European Seveso-II-Directive the effects of gas releases were considered. In this case dangers of explosion were compared with dangers due to poisoning.

Therefore, a scenario of a crack has been considered, which may be occur in the plastic foil of the fermenter. For different crack lengths a release of a mass flow was assumed and the gas dispersion has been calculated following the method according to VDI 3783. The boundary conditions, the safety characteristics and material properties of biogas be used in this calculation can be found in Table 4.

Table 4. Parameter for calculation of gas spreading acc. to VDI 3783.

Parameter	Quantity
Composition of biogas	
Methane	74 mol%
Carbon dioxide	25 mol%
Hydrogen sulfide	1 mol%
Temperature	20 °C
Operating overpressure	5 mbar
Coefficient of discharge	1
Duration of release	10 min
Wind speed	3 m/s
Geometry of source	horizontal line
High of release	6 m
High of point of impact	2 m
Roughness of soil	0,5 m
LEL of biogas acc. to Fig. 6	6 mol%
ERPG-2-Threshold of H_2S	30 ppm

In Fig. 10 the concentration of hydrogen sulfide for different crack length is shown as a function of distance. The ERPG-2-Threshhold of 30 ppm H_2S is marked as dotted line. Fig. 11 shows the concentration of biogas and the LEL, respectively. It is clearly seen, that the ERPG-2-Threshold requires a larger safety distance of about 180 m for a 4 m crack. For biogas with significant quantities of H_2S the danger of poisoning has priority with respect to the risk of explosion of the gas cloud in case of damage of fermenters and gas buffers.

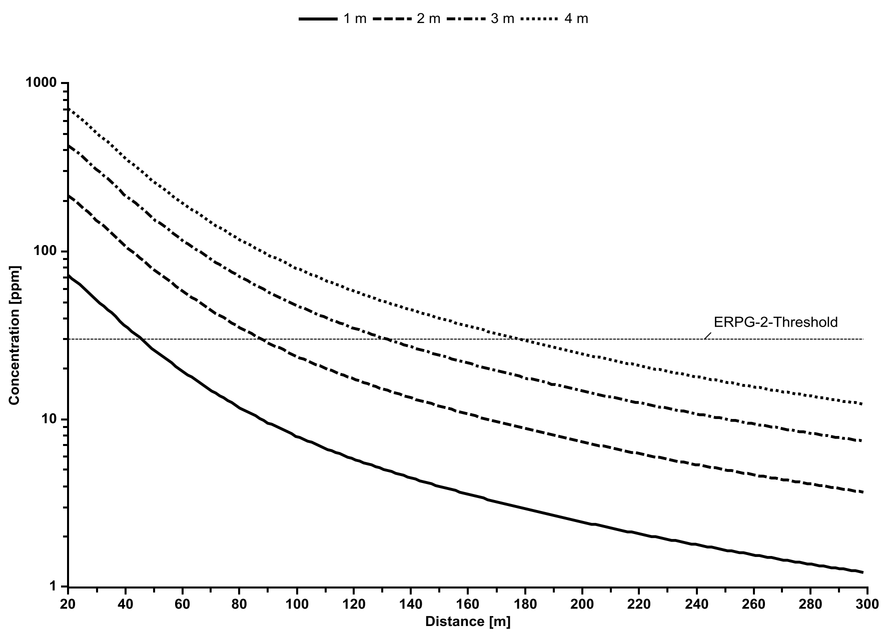

Fig. 10. Concentration of H_2S against distance for different lengths of cracks.

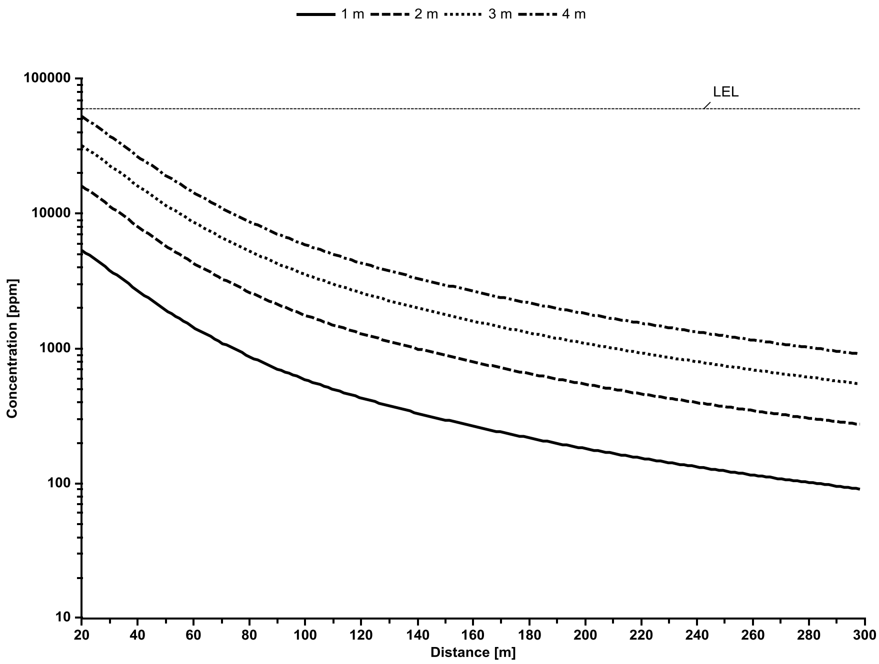

Fig. 11. Concentration of biogas against distance for different lengths of cracks.

8. Summary

To improve the safety data base for the operation of biogas plants, the explosion regions of mixtures of methane, hydrogen and carbon dioxide have been determined experimentally in the German Federal Institute for Materials Research and Testing. These appliances have been used in accordance with EN 1839 in a modified form. With the help of the measured explosion diagrams it is possible to determine exactly the explosion potential of raw gas in biogas plants at various process steps.

In addition, the water electrolysis has been considered for safety-related systems with power-to-gas technology. Based on the pressure dependence of the explosion limits of hydrogen in oxygen maximum allowable impurities of the electrolysis gases can be determined using the diagram in Fig. 7.

From our investigations, using Le Chatelier's equation for the calculation of the explosion limits of mixtures leads to significant errors, particularly at UEL. Therefore it is recommended to take experimental values for H_2/CH_4 mixtures.

According to the European Seveso-II-Directive the effects of gas releases were considered. For biogas with significant quantities of H_2S the danger of poisoning has priority with respect to the risk of explosion of the gas cloud in case of damage of fermenters and gas buffers.

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