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Safety aspects of the use of ammonia and an ammonia-hydrogen mixture

Katarzyna Stolecka-Antczak*, Andrzej Rusin

Silesian University of Technology, Department of Power Engineering and Turbomachinery, Konarskiego 18, 44-100 Gliwice, Poland
*Corresponding author email: katarzyna.stolecka-antczak@polsl.pl

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Abstract

Ammonia is a well-established raw material in the chemical industry. Among others, it is used to produce fertilisers, synthetic fibres or drugs. Ammonia is also used as a refrigerant, and it is viewed as a future fuel in the power industry. Technologies for its combustion in a mixture with hydrogen or methane (natural gas) are also being developed. The widespread use of ammonia involves the need for its transportation and storage. Both these processes are affected by its physical and chemical properties because ammonia is a toxic and flammable substance. The paper presents an analysis of the hazards posed by an uncontrolled leakage of ammonia and its mixture with hydrogen from a 10 m^3 tank. The range of hazard zones due to the toxic effects of ammonia is a few dozen metres. In the case of ammonia-hydrogen mixtures, a fire hazard appears, and the range of hazard zones due to thermal radiation is several metres. The total risk presented by an uncontrolled release of ammonia-hydrogen mixtures stored in 10 m^3 tanks is 10^{-7} and occurs in an area of up to 40 m.

Keywords: Ammonia; Hazards; Ammonia-hydrogen mixture; Risk

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1. Introduction

Many industries, including power generation, chemicals, petrochemicals and agriculture produce, store and transport numerous hazardous substances. If mishandled or if there is a failure of the installation components, their properties can pose a significant hazard to people and the environment.

Ammonia, produced globally on a large scale, belongs to such hazardous substances. It is a key product in the production of fertilisers, and it is also used in the food industry and refrigeration. Research is also being pursued on the use of ammonia as a fuel for engines and gas turbines. Indeed, considering its properties, ammonia is viewed as an alternative and emission-free future fuel that does not emit carbon dioxide during combustion. It can therefore be a good substitute for hydrocarbon fuels. The research on the use of ammonia as a potential fuel or as an additive to other fuels is currently the subject of many scientific publications.

In [1], the authors present a comprehensive review of ammonia as a fuel. The paper presents the principles of combustion reactions of ammonia and discusses their implications for practical energy applications, thereby supporting the development of ammonia as a carbon-free emission fuel. In [2], the authors review the use of ammonia as an alternative fuel in internal combustion engines, with a perspective on the future of possible applications and practical solutions to related problems. The main

goal of the authors of [3] was to show potential alternative fuels within their applications and present prospective alternative routes for their production. Chemicals like hydrogen and ammonia were tested as an alternative fuel for utilisation technologies. The objective in [4] was to show the prospects and challenges of ammonia as a fuel and suggest significant topics that could benefit from additional studies. The authors of [5] systematically summarise the fundamentals of ammonia-based combustion and its practical application. The comparative study presented in [6], based on a selected design, summarises the operating principles, together with the advantages and disadvantages of direct ammonia fuel cells. A model of a direct ammonia-fed solid oxide fuel cell was presented in [7]. The effect of selected parameters on the cell's efficiency was also studied in the paper. In [8], the authors provide a review of ammonia applications in various fields, including energy systems, fuel cells, gas turbines and propulsion technologies. These studies discuss the use of ammonia as a fuel in industrial applications, highlighting its potential as well as the challenges associated with its various application methods. The presented analyses indicate that ammonia combustion is a challenging process, primarily due to its low reactivity, which results in a low laminar burning velocity. Other limitations also include high ignition energy and combustion instability. To improve the process characteristics, mixtures of ammonia with flammable gases, such as methane and hydrogen, are used. The process of co-combustion of ammonia with other fuels is therefore also the subject of extensive research and analysis. In [9], the structure of hydrogen and ammonia flames and their combustion mechanisms were investigated. A comprehensive summary and review of research progress on the combustion characteristics and reaction mechanisms of ammonia and ammonia-hydrogen mixtures is presented in [10]. In [11], a numerical analysis of the combustion of ammonia-hydrocarbon mixtures under high-pressure dual-fuel conditions in large engines is presented. It was shown that mixtures of methanol and ammonia improve ignition stability and reduce the risk of excessive post-injection after burning. Experimental studies on an engine fuelled with diesel and 40% ammonia by energy fraction are presented in [12]. In subsequent stages, ammonia was gradually replaced with hydrogen, up to 28% by energy fraction. The effect of adding hydrogen to ammonia on combustion characteristics and stability was investigated. Experimental studies of the non-premixed gas turbine model combustor using ammonia fuel or fuel mixtures were presented in [13].

The widespread use of ammonia, which is a toxic and flammable substance, also involves an increasing number of accidents triggered by its uncontrolled release [14]. The spread of an ammonia cloud depends on the physical properties of the compound, the source of the release and the atmospheric environment. Ammonia vapours are flammable in the air, but they catch fire with difficulty, among others, due to their high minimum ignition energy. An explosion of ammonia vapours, on the other hand, occurs primarily in confined spaces (buildings). The hazard related to the spreading of a cloud of ammonia, in addition to the fire hazard, results from its toxicity. In the range of lower concentrations, the gas causes irritation, whereas at higher concentrations, it can cause death [15]. Recent studies based on computational fluid dynamics (CFD) have investigated ammonia leakage and dispersion in confined and semi-confined industrial environments. In [16], the authors analysed ammonia dispersion in closed industrial engine rooms with forced ventilation and demonstrated that ventilation configuration and obstacle distribution strongly influence ammonia accumulation and hazardous zones. An analysis of the effects of ammonia release is also presented in [17]. The authors analyse the hazard zones for various ammonia release scenarios and the influence of selected conditions on the range of impact. The fire-related and toxic consequences of a failure of major equipment and facilities intended for ammonia production are worked out in [18]. In the article [19], based on existing calculation formulas of the probability of explosion risk, analyses were performed in which the influence of the position and height of spatial obstacles and the time of the flammable refrigerant leak on the distribution of the flammable refrigerant concentration was fully considered.

In the paper [20], a comprehensive review of existing research on hydrogen and ammonia diffusion processes and explosion mechanisms is made. Prevention and detection techniques are discussed, providing theoretical support for the effective development of an energy system using hydrogen and ammonia. In the paper [21], a series of experiments conducted within a standard 20-L spherical combustion chamber to investigate the explosion characteristics of ammonia/hydrogen/air premixed gases under diverse conditions, spanning from low to high pressures (0.02–0.3 MPa) and from fuel-lean to fuel-rich scenarios (0.7–1.5), are discussed. The results reveal that the explosion characteristics are pronouncedly enhanced by hydrogen addition. Hydrogen addition remarkably increases the hydrodynamic instability and thermal-diffusion instability of the reactive mixture, enhances the thermal-mass diffusion on the flame front, and promotes flame deformation and cellular structure formation, resulting in an improvement of the flame propagation and compression effect on the unburnt reactant before the flame. In turn, the influence of variable ammonia concentrations on the self-ignition of hydrogen under high pressure during accidental release into a pipe is presented in [22]. Among other things, it was noted that as NH_3 concentration increases, the intensity and speed of shock-wave propagation within the pipe decrease. In [23], the authors analysed the explosion behaviour of ammonia, hydrogen and air mixtures in a closed duct. Both experiments and CFD modelling were used to reproduce, among other aspects, flame dynamics. The study focused on explosion parameters such as flame structure, flame propagation velocity, and overpressure. It was shown that when the ammonia blending ratio in the fuel ranged from 25% to 50%, the premixed flame underwent four distinct stages. It was also found that increasing the ammonia blending ratio in the fuel can significantly alter the explosion behaviour. The work [24] explores the flame evolution and pressure dynamics of NH_3/H_2/air mixtures in a closed duct. Flame instability parameters were calculated, and the NH_3/H_2/air flames on flame propagation were also assessed. Numerical studies of the characteristics of the ammonia, hydrogen and air flame in a micro combustor are presented in [25]. The effects of adding N_2 and CO_2 on the explosion behaviour of premixed hydrogen/ammonia/air mixtures were investigated in [26]

through experiments and analysis of reaction kinetics. The effects of N_2 and CO_2 on the evolution of the flame structure, flame front speed, pressure and reaction kinetics were analysed. The results indicated that increasing the hydrogen fraction in the fuel can accelerate the premixed flame, leading to higher flame front speeds and increased pressure. Conversely, the addition of N_2 and CO_2 has a suppressive effect.

Regardless of the application of ammonia and its mixtures with hydrocarbons, the safety of processes related to its production, storage and transport remains a significant issue. The authors of numerous publications note that due to its toxicity, flammability and the risk of explosions and leaks, ammonia requires a detailed hazard analysis and risk assessment. In [27], the authors presented an overview of current regulations and potential technologies proving the safety of the ammonia bunkering process. They also highlighted the critical issue of determining safety distances from the release site in order to minimise the consequences of hazardous events. The effect of ammonia on human health in the event of a release during a ship collision is discussed in [28]. The analyses included various release scenarios and types of storage tanks. The study showed, among other things, that semi-pressurised tanks may pose a higher toxic risk than refrigerated tanks, and that the use of water curtains and spill containment systems can significantly reduce the environmental impact of an accident.

This article presents an analysis of the hazards and risks associated with the uncontrolled release of ammonia and its mixtures with hydrogen, supplementing issues related to the safety of the processes using this type of fuel. Many studies indicate that adding hydrogen to ammonia improves combustion parameters, including flame propagation speed and reaction stability, making ammonia-hydrogen mixtures a promising direction for research into low-emission fuels and energy carriers. It should be emphasised, however, that both ammonia and hydrogen are hazardous substances, which implies the need for comprehensive process safety analyses as an integral part of research on their practical application. This study considers scenarios involving the release of ammonia in a gaseous state. The effect of the composition of the mixtures on the size of the hazard zones arising due to the toxic effects of ammonia, as well as to the impact of heat flux resulting from a fire of the mixtures, was studied. The novel aspect of this study is the integrated assessment of the consequences and risks related to the uncontrolled release of ammonia and mixtures of ammonia and hydrogen. The proposed approach combines modelling of the impact, including the determination of toxic and thermal hazard zones, with a quantitative risk assessment based on the probability of tank failure. The analysis allows for a comparison of safety levels related to the storage of ammonia and ammonia-hydrogen mixtures with varying proportions of the two gases.

2. Use of ammonia

Ammonia is used primarily as a fertiliser in agriculture. This is due to its rapid reaction with clay particles or organic matter and hydrogen ions. Other industries using ammonia include pharmaceuticals, food production and storage, metallurgical processes, pulp and paper production and textiles [29]. Ammonia is also used as a refrigerant, with benefits such as high efficiency, lower costs or no interaction with the ozone layer [30]. Moreover, ammonia has better heat transfer properties compared to other chemical refrigerants, which makes it possible to use devices with a smaller surface area [31].

There is also a chance of a wider use of ammonia in the power industry. It is expected that by applying the catalytic combustion method, it will be possible to burn ammonia at a lower temperature without emitting unnecessary pollutants. The high temperature of ammonia combustion favours the formation of nitrogen oxides, which are harmful to the environment.

Ammonia can be used as a fuel in various types of internal combustion engines, gas turbines and direct fuel cells, which is a significant advantage over other types of fuel. Using ammonia as a fuel, there will be no CO_2 or SO_x emissions. No soot will be produced either [30]. Fig. 1 presents the ways of using ammonia in industry for different types of generators. Ammonia can be used, for example, in spark-ignited ICEs, diesel ICEs with H_2, combustion turbines, ICEs powered with petrol or an ethanol mixture, transformed biogas generators and direct ammonia fuel cells [32].

Current research results indicate that ammonia has great potential to be used as a primary fuel in spark-ignition engines. The use of ammonia in an internal combustion engine may cause increased NO_x emissions compared to gasoline. The main problem with this use of ammonia is also the fact that its ignition energy is approximately 16 times higher compared to hydrocarbon fuels. The low combustion rate of ammonia makes combustion non-uniform at the low engine loads and/or high rpm values. Therefore, in order to ensure the combustion process stability, it is necessary to mix it, for example, with gasoline, diesel oil or hydrogen [30].

The use of ammonia as a fuel in gas turbines is still quite limited. Current studies concentrate primarily on ammonia-hydrogen and ammonia-natural gas mixtures. The use of ammonia as a fuel involves combustion instability and low combustion efficiency – current research takes account of the use of a swirler and a flame holder, for example [33], which could stabilise combustion and increase its efficiency, as well as reduce nitrogen oxide emissions. In [34], the authors show the possibility of adding ammonia to the gas turbine.

Ammonia can also be used in direct fuel cells, which generate electricity based on electrochemical reactions. Depending on the applied electrolytic materials or the operating conditions, these devices are divided into several types [35], including alkaline fuel cells (AFC), alkaline membrane fuel cells (AMFC), phosphoric acid fuel cells (PAFC), molten carbonate fuel cells (MCFC), proton exchange membrane fuel cells (PEMFC), direct methanol fuel cells (DMFC) and solid oxide fuel cells (SOFC). Solid oxide fuel cells are considered as the most energy-effective, and the gas which is most often used in fuel cells to produce electricity is hydrogen. Research is currently being conducted on the use of ammonia, where it can be used directly as a fuel in AFC, AMFC and SOFC technologies. The use of ammonia ensures that the efficiency of the cells remains relatively high. In the case of fuel cells, the concept of reverse fuel cells (RFCs), currently in development, it is also worth noting.

They enable clean conversion of nitrogen and water to ammonia under the influence of electricity obtained from renewable sources [36].

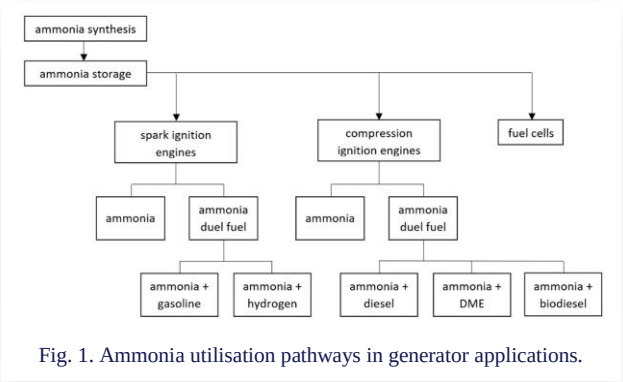


Fig. 1. Ammonia utilisation pathways in generator applications.

A consequence of the need to increase the combustion efficiency of ammonia is that it is often mixed with other agents that upgrade the fuel, such as methane, propane, petrol, kerosene or hydrogen. Mixing ammonia with hydrogen strengthens the combustion flame. In addition, hydrogen can be produced from ammonia, which contributes to the practical benefits of transporting this mixture. Such mixtures have been tested, among others, in spark-ignition engines and gas turbines. The high reactivity of hydrogen contributes to an increase in laminar combustion velocity compared to pure ammonia. On the other hand, mixing ammonia with other hydrocarbon fuels generally leads to a higher flame speed and to an increase in the intensity of radiation and heat release rate. It also results in lower CO_2 emissions [30].

3. Effects of an uncontrolled release of ammonia

The use of ammonia presents various challenges due to its properties. This raw material is used directly or indirectly in a number of industrial processes, and its use requires storage and/or transportation to sites of its final utilisation.

Two methods are generally used for ammonia storage. One, with no refrigeration, involves increasing pressure while maintaining the ambient temperature, e.g. to 0.99 MPa at 25°C. The other method, with refrigeration, involves maintaining atmospheric pressure while lowering the temperature to approximately –33°C. Large volumes of ammonia are most often stored using refrigerated tanks, which minimises the potential leakage rate in the case of failure (compared to pressure tanks). Ammonia tanks should have an appropriate structure and be equipped with, among other things, a system of protection against an excessive increase or drop in pressure.

Ammonia is considered a toxic and flammable substance. Exposure to ammonia can therefore occur under conditions of industrial use or as a result of an accidental leakage during storage or transport. In recent years, a number of incidents involving ammonia spills have occurred around the world. For example, in the USA in 2010, an ammonia release led to the poisoning of more than 120 people [31]. One of the worst accidents involving ammonia occurred in 1992. A cloud of released ammonia caused 129 deaths and 1150 injuries as a result of inhaling high concentrations of the substance [28]. Ammonia tanks, installations and pipelines are therefore potentially dangerous elements of the industrial infrastructure which can fail, for example, due to corrosion or an accidental impact [37]. A leak can pose a fire hazard, or it can have a toxic effect. Depending on the location of the damage, the storage/transport conditions and the type of installation, ammonia may be released in a gaseous or liquid form. A release of liquid ammonia into the environment leads to the immediate formation of a cloud of gaseous ammonia (Fig. 2). This is due to its low boiling point.

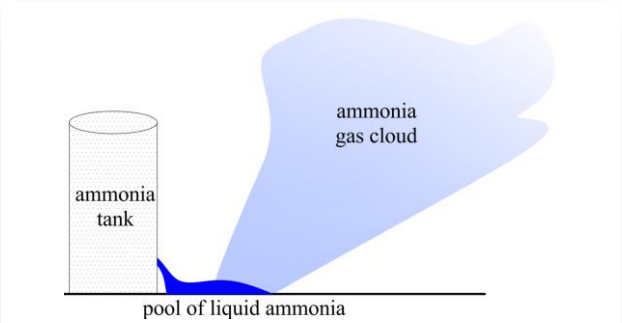


Fig. 2. Release of ammonia.

3.1. Ammonia toxicity

Ammonia is a colourless substance that is lighter than air. It can be smelt in the air at very low concentrations as it has a characteristic pungent and acrid odour. If released into the atmosphere, the gas rises in the air. Contact with ammonia can cause skin burns and changes in the respiratory tract. If inhaled, ammonia can even cause blindness. Ammonia poisoning through the alimentary tract causes disorder in intestinal peristalsis, and in extreme cases, it can even lead to death. The greatest risk of ammonia poisoning is for people working in industries where ammonia is used in large quantities. The gas is also released during the decomposition of matter, for example, in animal faeces.

At concentrations of 5 ppm, ammonia becomes noticeable by smell [38]. Concentration levels of 100 ppm cause violent irritation of the respiratory tract, bronchi and eyes (especially conjunctivitis). Immediate irritation to the eyes, nose and throat will occur for concentrations of 400–700 ppm. Pulmonary oedema, cough and laryngospasm will occur at concentrations higher than 1500 ppm. A concentration higher than 4650 ppm means a lethal dose [39].

As for ammonia interaction with the soil, toxicity is non-existent as the substance evaporates from the surface. The ammonia remaining in the soil is a source of nitrogen fertiliser. Part of the ammonia undergoes photolytic decomposition in the air, and the rest returns to the soil surface through dry or wet deposition.

The toxicity of ammonia in the aqueous environment, where it is very harmful to many aquatic organisms, is a different matter.

3.2. Ammonia explosivity and flammability

The problem of ammonia explosivity arises in confined spaces when very specific conditions are met. For the phenomenon of explosion to occur, the following have to coincide: an oxidiser,

an external source of ignition (explosion trigger), the mixing of ammonia with air within the limits of explosiveness and a spatial limitation, for example in the form of the walls of the room.

The occurrence of an explosion depends on whether the flammable ammonia-air mixture will reach the appropriate temperature due to external energy (minimum ignition energy). This value is about 40 mJ and is significantly higher than the ignition energy of hydrogen, which totals 0.02 mJ. The range of explosivity limits of ammonia is the concentration of 15 to 28% (with oxygen: 14 to 79%), and it changes under the influence of hydrogen contained, for example, in water. When water enters the surface of liquid ammonia, there follows a reaction of intense heat release from the process of ammonia combining with water (exothermic reaction) and due to the evaporation of significant amounts of ammonia, which produces an ignitable and explosive mixture. In addition, the presence of other substances, such as chlorine, bromine, iodine, mercury, sodium and calcium, favours the formation of explosive mixtures [30].

Ammonia is considered a substance which is flash resistant and slow burning. The prerequisite that triggers the combustion process of ammonia in the liquid form is the supply of a sufficient amount of heat (energy) that enables the formation of ammonia-air vapours above the surface of the liquid. The vapours must be in the concentration range between the lower and the upper flammability limit, i.e. 15–28% in the air.

In the event of damage to tanks storing ammonia or its mixtures in the gaseous form, the leaking gas may ignite, and this may result in a jet fire. Another consequence of damage to ammonia storage tanks is the BLEVE phenomenon, i.e. an explosion of rapidly expanding vapours of a boiling liquid, and the formation of a fireball. This phenomenon usually occurs due to the tank being heated by an external flame. As a result, the pressure inside the tank rises considerably, and the tank ruptures violently. The contents of the tank are ejected abruptly through the cracks, a cloud is formed, and a violent ignition follows. The hazards to humans and the environment related to the event of fire are the direct impact of the resulting flame and the heat flux generated by the fire. A heat flux of 4–5 kW/m^2 will not cause injury to humans over a long exposure time. A heat flux of 12.5 kW/m^2 will cause first-degree burns in humans, and a value above 25 kW/m^2 will cause significant injury within 10 seconds. The lethal heat flux for humans is 35–37.5 kW/m^2 [40].

The conditions of flammability and explosivity change when other gases, such as hydrogen, are added to ammonia. This is analysed in the sections below.

4. Methods of calculation

The technical risk related to a failure of a system of transport and storage of dangerous substances can be understood as the product of the probability of the occurrence of a hazardous event and its consequences. Denoting the risk as *R*, the following can be written [41]:

$$R = \sum C_i P_i. \tag{1}$$

where: P_i – probability or frequency of occurrence of a hazardous event *i*, C_i – consequences of the occurrence of event *i*.

Depending on the event under consideration, the consequences can be expressed in monetary units or in the number of potential casualties. In order to determine the risk value according to the above formula, it is necessary to identify possible hazard scenarios, estimate the probabilities of the occurrence of the hazards and assess the consequences [42].

The hazards related to the use of ammonia are due to its physicochemical properties, especially its toxicity and flammability. Ammonia is characterised by a narrow range of flammability and a high temperature of autoignition, which results in its low flammability. In the gaseous form, it can quickly disperse in atmospheric air because its density is lower than the air density. This feature reduces the risk of a possible fire and explosion. However, ammonia is also classified as a toxic substance. Even a small leak of ammonia in a residential area can cause very serious consequences.

The paper presents an analysis of the risks associated with the uncontrolled release of ammonia and its mixtures with hydrogen from the storage tanks of these gases. The considerations involved different sizes of damage. Both the size of the tank and the size of the hole through which a gas is released affect the amount of gas released into the atmosphere, as well as the duration of the leakage. Due to the mentioned properties of ammonia and hydrogen, hazard scenarios related to the toxic effects of the gases and fire of the mixture of these gases were considered.

The risk of occurrence of a hazardous event, for example, in the form of an ammonia tank rupture or puncture, is related to a potential release of ammonia into the atmosphere. This, in turn, can lead to the formation of an ammonia cloud with concentrations hazardous to humans and the environment. Among others, the hazard level will depend on the atmospheric conditions, the size of the damage, the gas parameters and the duration of the leakage.

The analyses of the consequences of such scenarios were conducted using the PHAST v6.7 software package [42]. The program makes use of the unified dispersion model (UDM) [42], which makes it possible to calculate the quantities describing the outflow and transport of the created cloud in the atmosphere, taking account of all the cloud stages, such as the release, the touchdown, the pool formation or evaporation, for example.

Another scenario considered in the risk analysis is the hazardous effects of heat radiation resulting from a fire in the ammonia-hydrogen mixture. A jet fire is possible with the release of flammable gas at a high pressure. The quantities related to this type of fire were calculated using the PHAST v6.7 software [42] with implementation of the Chamberlain model. The model assumes the flame to be a spatial solid in the form of a truncated cone (cone frustum) with dimensions depending, among other things, on the wind speed.

Another type of fire that can occur due to the unsealing of tanks with flammable substances is a fireball, which may be due to the BLEVE phenomenon. This phenomenon is an effect of a sudden and complete rupture of a tank.

For each of the above mentioned scenarios, the probability of its occurrence is estimated. In the article, this probability will be calculated by using the event tree. In the next step, the calculated probability of each scenario and the corresponding proba-

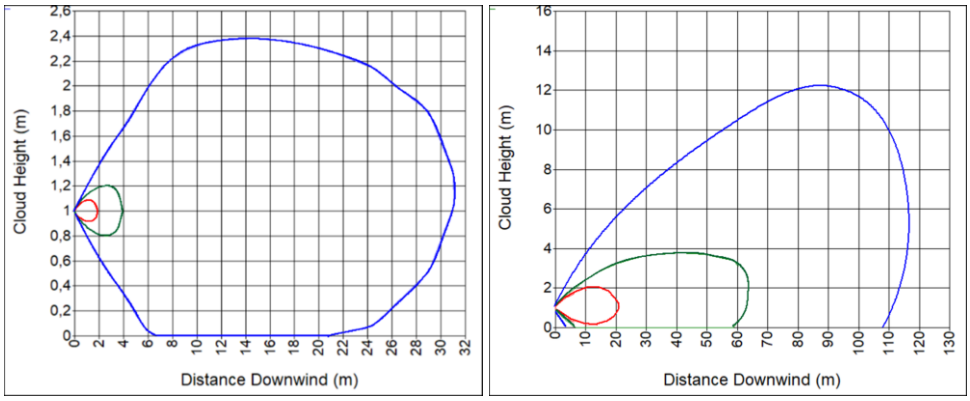
bilities of death of people within the range of negative effects of radiation from a potential fire or toxic properties of gases are presented in risk diagrams. The risk level is also calculated according to Eq. (1).

5. Results of analysis of hazards presented by an uncontrolled release of ammonia and an ammonia-hydrogen mixture

The figures below show the range of clouds with an elevated concentration of ammonia formed when the tank with a capacity of 10 m^3 becomes unsealed. The gaseous form of ammonia was considered. A 10 m^3 tank holds 44.3 kg of ammonia. In the calculations, the air temperature, the wind speed and the atmospheric stability class are assumed at 20°C, 2 m/s and D, respectively. The relative humidity is assumed to be 85 %. The ammonia was released in an open, undeveloped area. The different cloud ranges correspond to different ammonia concentrations of 1000, 10 000 and 25 000 ppm. These are the concentrations that cause severe breathing difficulty, eye and skin irritation in humans (1000 ppm – blue), death after several minutes (10 000 ppm – green) and immediate human death (25 000 ppm – red).

The ranges of a cloud with elevated concentrations of ammonia released due to damage to a tank containing ammonia in the gaseous form with a pressure of 0.5 MPa and temperature of 20°C are shown in Fig. 3. An analysis was conducted of the outflow of the gas through damage with an equivalent diameter of 0.5 cm and 5 cm located at a height of 1 metre above the ground. The hole with the smaller diameter could be the result of corrosion processes in the tank walls. The bigger hole could correspond to damage due to a mechanical effect, e.g. an impact against the tank. The cloud ranges shown in the figure from the side view of the cloud show the concentration of ammonia at a specific time during the process of the outflow from the tank.

Analysing the above graphs, it can be noticed that the most dangerous zone with a lethal level of ammonia concentration for humans will not touch the ground surface. However, it can be seen from the side view that it will cover a range of about 2 and 20 metres for the hole diameters of 0.5 cm and 5 cm, respectively. The zone with the lethal concentration for humans after several minutes of exposure, i.e. 10 000 ppm, will reach a range of about 60 metres on the ground surface only for the larger diameter of the damage (5 cm). The second factor with a significant impact on the death hazard is the time of exposure to the toxic effects of ammonia. This time depends, among other things, on the volume of the released gas. In the case of the tank with a capacity of 10 m^3, the zone with the highest concentration of 25 000 ppm will keep for about 30 seconds, while in the case of the smaller tank, e.g. with a volume of 1 m^3, this time is 5s.



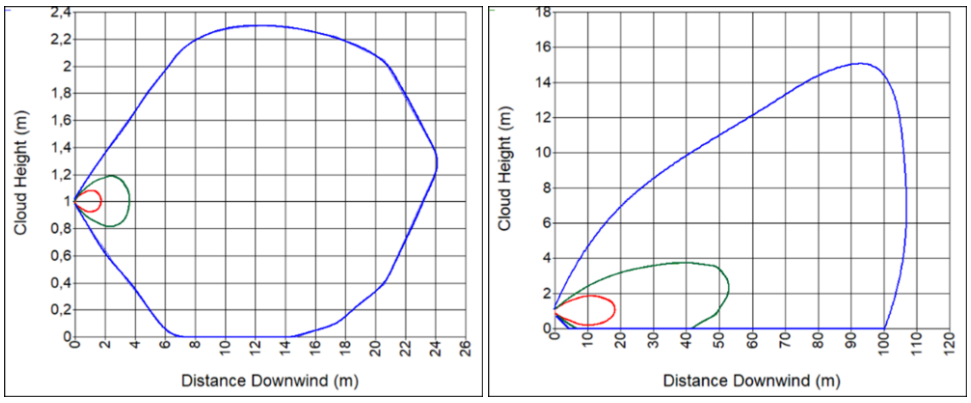
a) b)

Fig. 3. Range of the cloud with an elevated concentration of ammonia (side view, blue – 1000 ppm; green – 10 000 ppm; red – 25 000 ppm): a) hole (damage) with the diameter of 0.5 cm, tank volume: 10 m^3 (time: 14.7 s); b) hole (damage) with the diameter of 5 cm, tank volume: 10 m^3 (time: 23.9 s).

As already mentioned, apart from its widespread use in the chemical industry or agriculture, ammonia can be used as an alternative fuel. Research on the combustion process of this substance in engines has been pursued for many years. In addition to the possibility of using ammonia itself, the combustion of ammonia-hydrogen and ammonia-methane mixtures is also investigated in those works. The addition of hydrogen and methane improves the conditions of ammonia combustion. It will also have an impact on the safety of the use of these mixtures. By adding hydrogen in a mixture with ammonia, the toxicity of the mixture is reduced, which thus reduces the hazard zones in the event of an uncontrolled release of this type of mixture. The effect of hydrogen content in the mixture on the size of areas with high ammonia concentrations of 1000 ppm, 10 000 ppm and 25 000 ppm is shown in the figures below.

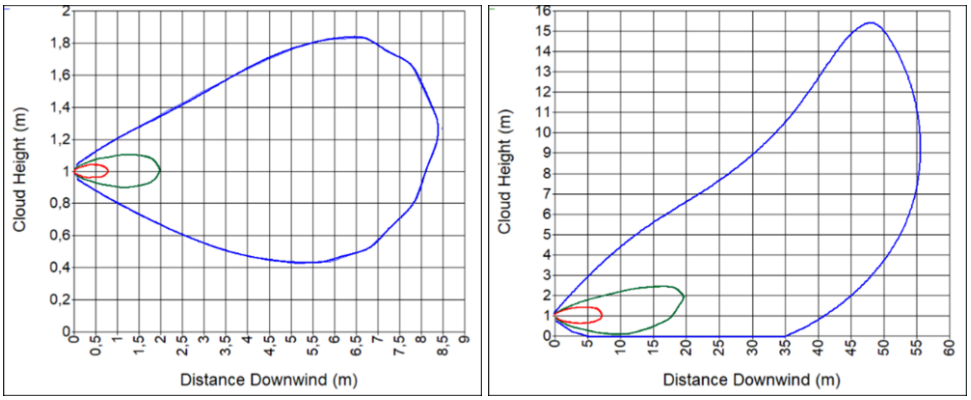
Fig. 4 and Fig. 5 show the size of these zones for an uncontrolled release of ammonia-hydrogen mixtures at the volume ratios of 80/20 and 20/80 from a 10 m^3 tank. Punctures with equivalent diameters of 0.5 cm and 5 cm were analysed. The mixture parameters are the pressure of 0.5 MPa and the temperature of 20°C. The wind speed is assumed at 2 m/s.

Analysing the graphs in Fig. 4, it can be seen that the zones related to the impact of the elevated concentration of ammonia contained in the mixture and causing immediate human death will not touch the ground surface. The range of about 2 and 20 metres will be reached for damage with the diameters of



a) b)

Fig. 4. Range of the cloud with an elevated concentration of ammonia for a gaseous 80/20 (v/v) ammonia-hydrogen mixture (side view, blue – 1000 ppm; green – 10 000 ppm; red – 25 000 ppm): a) hole (damage) with the diameter of 0.5 cm, tank volume: 10 m^3 for the time of 10.9 s; b) hole (damage) with the diameter of 5 cm, tank volume: 10 m^3 for the time of 21 s.



a) b)

Fig. 5. Range of the cloud with an elevated concentration of ammonia for a gaseous 20/80 (v/v) ammonia-hydrogen mixture (side view, blue – 1000 ppm; green – 10 000 ppm; red – 25 000 ppm): a) hole (damage) with the diameter of 0.5 cm, tank volume: 10 m^3 for the time of 2.8 s; b) hole (damage) with the diameter of 5 cm, tank volume: 10 m^3 for the time of 12.6 s.

0.5 cm and 5 cm, respectively, for different times. The zones with the concentration of 10 000 ppm already reach a distance exceeding 50 metres for the larger diameter of the tank damage. The zone with the highest concentration of 25 000 ppm disappears after 25 seconds if the mixture is released from the tank.

Figure 5 shows the range of the areas with an elevated concentration of ammonia at an uncontrolled release of the ammonia-hydrogen mixture with the volume ratio of 20/80. Like before, a tank volume of 10 m^3 was considered, as well as two sizes of damage with the diameters of 0.5 cm and 5 cm. The mixture parameters remain unchanged.

The hazard zones related to the concentration of ammonia, causing immediate death of humans, will occur in an area with a range of about 7 metres. The concentration of 10 000 ppm can reach a distance of about 20 metres from the failure site. High concentrations in the air last for 18 seconds.

The maximum ranges of hazard zones related to the toxic effects of ammonia and its mixture with hydrogen are presented in Tables 1 and 2.

Table 1. Maximum range of toxic hazard zones for varying portions of gases (hole (damage) with the diameter of 0.5 cm).

H_2/NH_3	1000 ppm	10 000 ppm	25 000 ppm
0/100	31.5 m	4.0 m	2.0 m
20/80	24.0 m	3.7 m	1.7 m
80/20	8.4 m	2.0 m	0.7 m

Table 2. Maximum range of toxic hazard zones for varying portions of gases (hole (damage) with the diameter of 5 cm).

H_2/NH_3	1000 ppm	10 000 ppm	25 000 ppm
0/100	116.0 m	64.0 m	21.0 m
20/80	108.5 m	53.5 m	18.0 m
80/20	55.5 m	20.0 m	7.5 m

As already mentioned, another (though less likely) consequence of a release of ammonia is a fire, provided that the conditions for it are favourable. A fire hazard applies primarily to gaseous ammonia and its mixtures with another flammable gas, such as hydrogen, because the ignition of liquid ammonia is difficult.

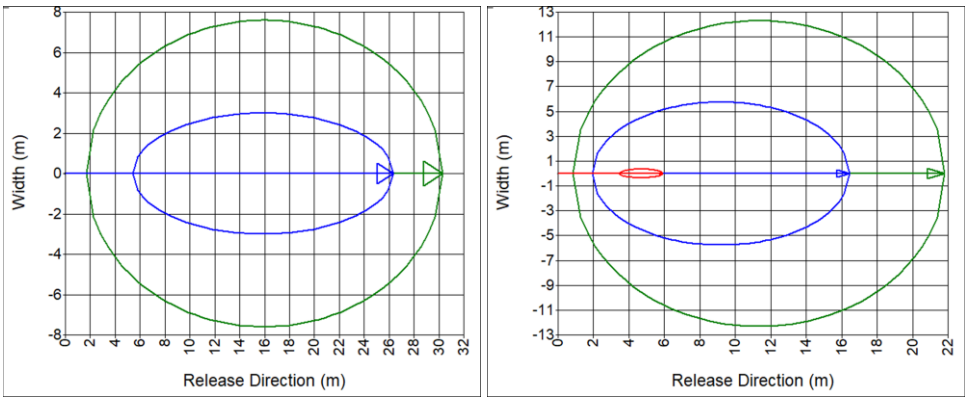
The physical properties of such a mixture differ from those of pure ammonia, which means that the quantities that decide the dimensions of the negative consequences of its uncontrolled release will also vary. Adding hydrogen to ammonia results, among other things, in a change in the lower and the upper flammability limits of the mixture. It also affects the heat of combustion, as shown in Table 3. It should be noted that the addition of ammonia to hydrogen raises the lower flammability limit and lowers the upper flammability limit, thus narrowing the range of concentrations at which ignition and explosion of the mixture are possible. Mixing ammonia with other flammable gases also affects the combustion process parameters, including the value of the heat flux generated during mixture combustion.

The figures show example hazard zones arising due to the ignition of ammonia, hydrogen and ammonia-hydrogen mixtures. The analysis concerns effects of a jet fire and a fireball.

Table 3. Lower and upper limits of mixture flammability.

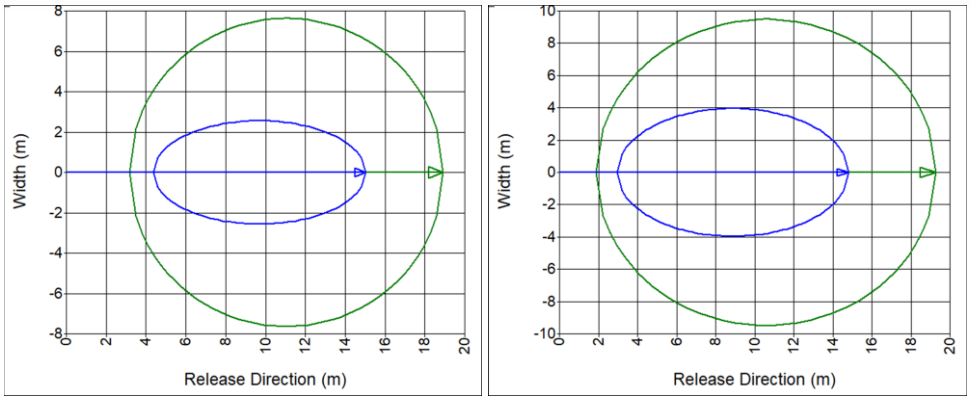
H_2/NH_3	LFL	UFL	Heat of combustion
100/0	4.00 %	75.00 %	241820 kJ/kmol
90/10	4.32 %	62.50 %	242794 kJ/kmol
80/20	4.71 %	53.57 %	243976 kJ/kmol
70/30	5.16 %	46.88 %	245441 kJ/kmol
60/40	5.71 %	41.67 %	247306 kJ/kmol
50/50	6.40 %	37.50 %	249759 kJ/kmol
40/60	7.27 %	34.10 %	253130 kJ/kmol
30/70	8.42 %	31.25 %	258054 kJ/kmol
20/80	10.00 %	28.85 %	265923 kJ/kmol
10/90	12.30 %	26.79 %	280511 kJ/kmol
0/100	15.00 %	28.00 %	316830 kJ/kmol

A jet fire (Fig. 6 and Fig. 7) is a fire resulting from the leakage and ignition of a jet of a pressurised substance flowing out of a hole (damage) formed in the tank wall. The hazard zones shown in the figure refer to the fire-generated heat flux of 4, 12.5 and 37.5 kW/m^2 (green, blue and red, respectively). Exceeding



a) b)

Fig. 6. Heat flux generated by a jet fire of ammonia and hydrogen; hole (damage) with the diameter of 5 cm, tank volume: 10 m^3 (green – 4 kW/m^2, blue – 12.5 kW/m^2, red – 37.5 kW/m^2): a) ammonia (mass: 44.3 kg, mass flow at the hole outlet 1.82 kg/s); b) hydrogen (mass: 4.95 kg, mass flow at the hole outlet 0.62 kg/s).



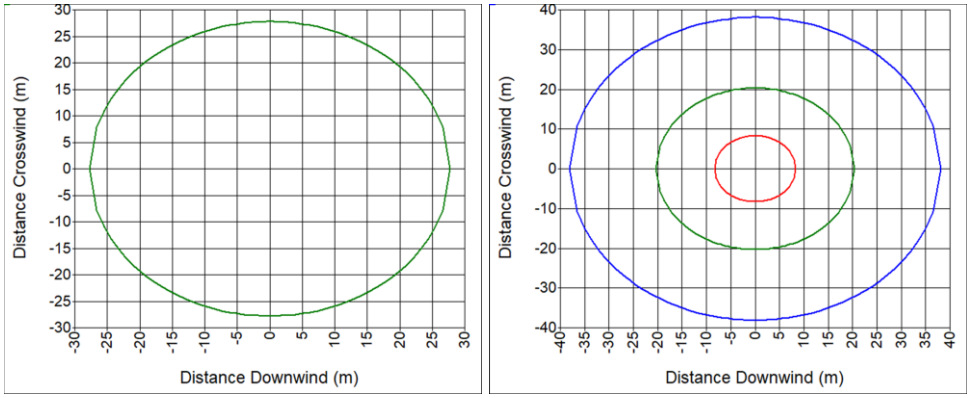
a) b)

Fig. 7. Heat flux generated by a jet fire of ammonia and hydrogen; hole (damage) with the diameter of 5 cm, tank volume: 10 m^3 (green – 4 kW/m^2, blue – 12.5 kW/m^2, red – 37.5 kW/m^2):
a) ammonia-hydrogen mixture (80/20) mass: 35.73 kg, mass flow at the hole outlet 1.64 kg/s;
b) ammonia-hydrogen mixture (20/80) mass: 12.36 kg, mass flow at the hole outlet 0.98 kg/s.

the value of 4 kW/m^2 causes noticeable effects for humans; the value of 12.5 kW/m^2 causes first-degree burns, and the value of 37.5 kW/m^2 causes immediate death. The areas indicated by the red line, which correspond to human deaths, occurred only in the case of a hydrogen release. The presented results relate to the leakage and ignition of ammonia, hydrogen and an ammonia-hydrogen gaseous mixture with the pressure of 0.5 MPa and temperature of 20°C, released from tanks with the volume of 10 m^3. The analysis concerns the larger diameter of the damage, i.e. 5 cm. Two values of the ammonia-hydrogen mixture volumetric ratio are assumed for the considerations: 20/80 and 80/20. In the case of the smaller diameter of the damage, the hazard zones will not occur.

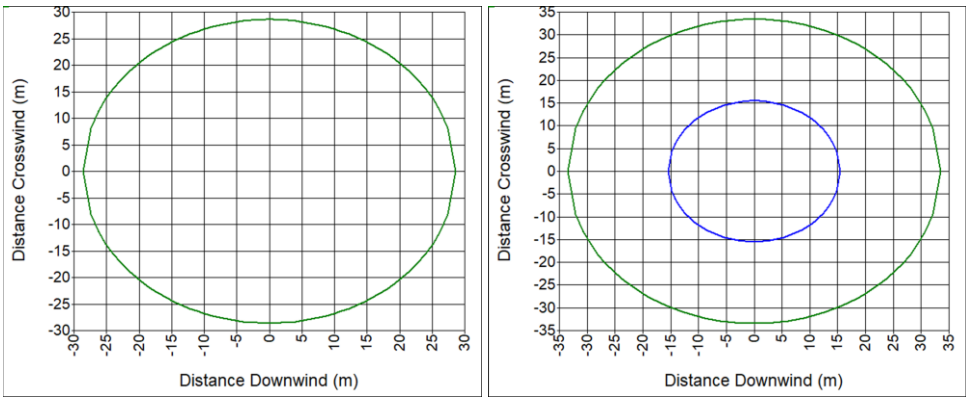
The ranges of the radiation zones shown in the above figures depend, among other things, on the amount of the stored gas and the mass flow through the hole. For pure ammonia, the mass flow is 1.82 kg/s, whereas for pure hydrogen it totals 0.62 kg/s. Figures 8 and 9 show the range of the areas of thermal radiation due to the formation of a fireball. The ranges are shown for the failure of the 10 m^3 tank and two different compositions of the ammonia-hydrogen mixture. Like for the jet fire, the zones illustrate the range of the fireball-generated heat flux values of 4, 12.5 and 37.5 kW/m^2.

Table 4 and Table 5 present the maximum ranges of hazard zones associated with the effects of the heat flux generated by fires of ammonia and its mixture with hydrogen. Table 4 refers



a) b)

Fig. 8. Heat flux generated by a fireball for the ammonia-hydrogen mixture; tank volume: 10 m^3 (green – 4 kW/m^2, blue – 12.5 kW/m^2, red – 37.5 kW/m^2): a) ammonia (mass: 44.3 kg); b) hydrogen (mass: 4.95 kg).



a) b)

Fig. 9. Heat flux generated by a fireball for the ammonia-hydrogen mixture; tank volume: 10 m^3 (green – 4 kW/m^2, blue – 12.5 kW/m^2, red – 37.5 kW/m^2): a) ammonia-hydrogen (80/20), mass: 35.73 kg; b) ammonia-hydrogen mixture (20/80) mass: 12.36 kg.

Table 4. Maximum range of radiation hazard zones for varying portions of gases (hole (damage) with the diameter of 5 cm).

H_2/NH_3	4 kW/m^2	12.5 kW/m^2	37.5 kW/m^2
0/100	30.0 m	26.0 m	–
20/80	19.0 m	14.8 m	–
80/20	19.2 m	15.0 m	–
100/0	21.8 m	16.3 m	6.0 m

Table 5. Maximum range of radiation hazard zones for varying portions of gases (hole (damage) with the diameter of 5 cm).

H_2/NH_3	4 kW/m^2	12.5 kW/m^2	37.5 kW/m^2
0/100	27.5 m	-	–
20/80	28.0 m	-	–
80/20	34.0 m	15.0 m	–
100/0	37.5 m	20.0 m	7.5 m

to the case of a jet fire, while Table 5 refers to the case of a fireball.

If pure ammonia is ignited, the maximum radiation due to the fire totals 4 kW/m^2. With the addition of hydrogen, higher values of radiation will appear, including the zone posing a death hazard. The range of radiation depends on the proportion of hydrogen in the mixture, but it does not exceed several metres.

6. Risk of the occurrence of undesirable effects of an uncontrolled release of ammonia and an ammonia-hydrogen mixture

The following is an estimation of the risk arising due to the failure of an example storage tank of gaseous ammonia and of the ammonia-hydrogen 80/20, 50/50 and 20/80 mixture. The analysed tank storing gas with a pressure of 0.5 MPa and a temperature of 20°C has a volume of 10 m^3; the equivalent diameter of the damage is 5 cm.

6.1. Estimation of the probability of occurrence of dangerous events triggered by tank damage

An uncontrolled release of ammonia or an ammonia-hydrogen mixture can cause a jet fire, a fireball, the movement of a cloud with a toxic concentration, or the gas can disperse in the atmosphere without causing negative effects. The event tree in Fig. 10 presents various possible scenarios for subsequent events triggered by damage to a tank containing hazardous substances [43]. The probability of the occurrence of the triggering event in the form of damage to the gas tank was assumed at 2×10^{-6} according to [44]. The probability of further events is assumed based on the estimations given, for example, in [43].

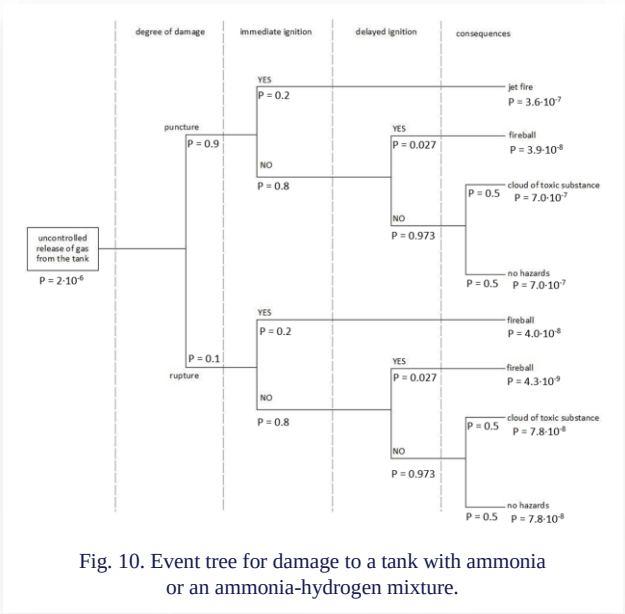


Fig. 10. Event tree for damage to a tank with ammonia or an ammonia-hydrogen mixture.

6.2. Assessment of the consequences of a release of ammonia

The probability of human injuries due to the impact of the negative effects of an accident caused by an uncontrolled release of ammonia and ammonia-hydrogen mixture can be estimated using probit functions. The negative factors affecting humans and the environment in the event of an ammonia release are those analysed above: heat flux, in the case of a fire, and the impact of toxic concentrations, if the released gas cloud does not ignite. For these cases, the probit function can take the form presented in [45]. Based on the analyses presented above of areas with elevated thermal radiation, caused by a fire of ammonia and an ammonia-hydrogen mixture, and areas with elevated ammonia concentrations in the released gas cloud, it is possible to determine the probability of death of humans being within the range of the impact of elevated thermal radiation or elevated toxic gas concentration. The results of such analyses are presented in the figure below.

The graphs shown in the figures below were obtained using probit functions. Different values of the ammonia-hydrogen mixture volumetric ratio and different exposure times were assumed for the considerations. Analysing the graphs presented in Fig. 11, it can be seen that the maximum area will occupy a range of about 30 metres for 5 minutes of exposure. The next figures show the probabilities of death due to the toxic effects of released ammonia. Analysing the graphs presented in Fig. 12, it can be noticed that the concentration of ammonia for the assumed times of exposure does not result in deaths at the level of 100%. The highest probability of death is about 55% and occurs in the area with a radius of about 15 m from the site of the release of pure ammonia. This probability decreases with an increasing proportion of hydrogen in the mixture.

6.3. Estimation of the risk of death due to damage to a tank containing ammonia and an ammonia-hydrogen mixture

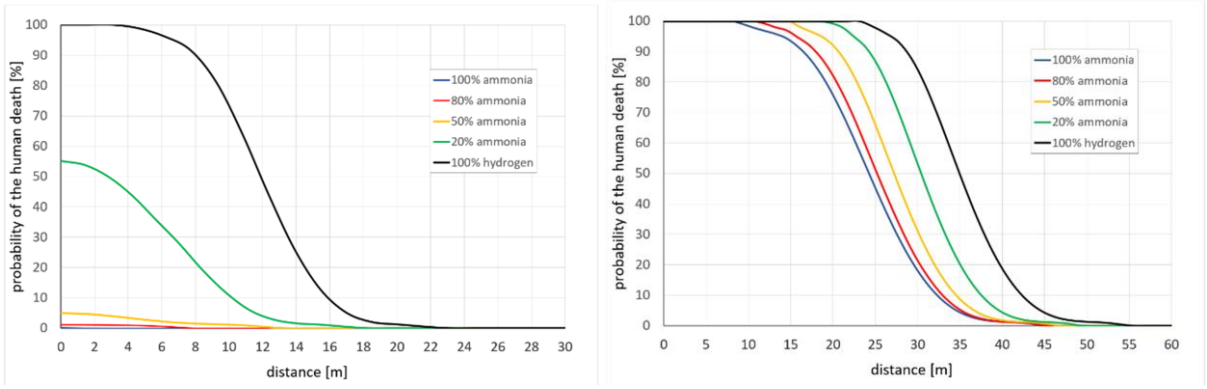
The total risk of death due to the release of ammonia, hydrogen and the ammonia-hydrogen mixtures with the proportions of 80/20, 50/50 and 20/80 resulting from the occurrence of undesirable events following damage to the storage tank is shown in Fig. 13. The calculations were carried out for a tank with a capacity of 10 m^3. The graph shows the risk for the longer analysed time of exposure to negative factors, i.e. 5 minutes.

The total risk level is of the order of 10^{-7} and similar for the analysed compositions of the mixtures. However, the range of the risk varies. The risk due to the release of pure ammonia is the highest, but it occurs in a small area about 10 metres from the tank. The risk posed by the release of the mixture with hydrogen is slightly lower but extends over a larger area.

Figure 14 shows risk diagrams with selected scenarios of uncontrolled release of ammonia and its mixture with hydrogen. For each scenario, both related to toxic hazards and fire hazards, the probability of occurrence and the probability of causing human death are given. The following figure refers to different distances from the accident site and gas release, i.e. risk at a distance of 20 m and 40 m from the gas leak site.

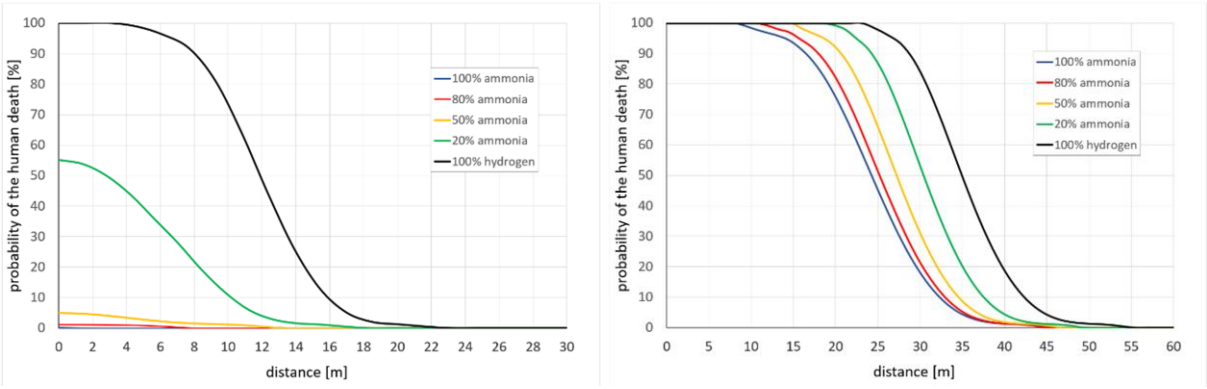
7. Summary

The intensive search for new fuels that can effectively replace hydrocarbon fuels, the combustion of which is considered a ma-



a) b)

Fig.11. Probability of death from exposure to heat radiation generated by a fireball for different compositions of the ammonia-hydrogen mixture and for different exposure times (tank volume 10 m^3): a) 0.5 minutes of exposure; b) 5 minutes of exposure.



a) b)

Fig.12. Probability of death due to the impact of toxic concentrations of ammonia for different compositions of the ammonia-hydrogen mixture and for different exposure times (tank volume 10 m^3): a) 0.5 minutes of exposure; b) 5 minutes of exposure.

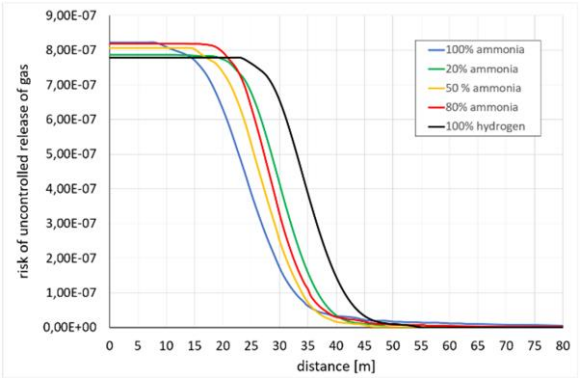


Fig. 13. Risk of death due to the impact of toxic concentrations and heat flux for released ammonia and the ammonia-hydrogen mixture.

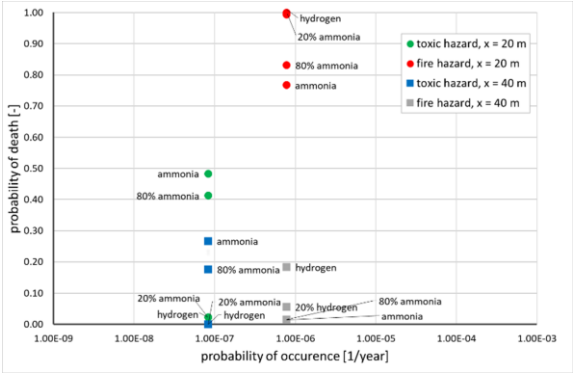


Fig. 14. Risk diagram at 20 meters and 40 metres for ammonia and ammonia-hydrogen mixture leak scenarios.

jor cause of climate changes, indicates that hydrogen fuels could be the solution. In addition to pure hydrogen, the use of ammonia as a stand-alone fuel or as an additive to other gases, such as hydrogen, is also being considered. The paper presents an analysis of the safety of storing such fuels, with a special emphasis on the toxic hazard posed by ammonia, the fire hazard posed by hydrogen and the mutual contribution of these hazards in mixtures of the two gases. Two sizes of damage and different ammonia-to-hydrogen ratios in the mixture were analysed in detail. At a release of gaseous ammonia, the zones with a concentration of 10 000 ppm, i.e. those that can cause death to people, reach a distance of several metres for a small diameter of the damage

and about 60 metres if the damage diameter is large. The capacity of the tank itself, i.e. the amount of the stored gas, affects the time of duration of high concentrations, which is about a few minutes for the 10 m^3 tank.

Ammonia-hydrogen mixtures present a smaller toxic hazard, but the fire hazard becomes more real. A 20% share of hydrogen in the mixture reduces the toxic zone range by several per cent. An 80% share of hydrogen reduces the hazard zones by more than three times. The addition of hydrogen results in the appearance of hazard zones due to the potential impact of thermal radiation from fires of the released mixture. The range of the zones for the analysed volumes of the gases totals several metres. The total risk determined for the effects of toxic concentrations and the fire-generated heat flux has values of the order of 10^{-7}. This level of risk is considered acceptable. Depending on the composition of the mixture, it extends over an area of 30 to 40 m.

The above conclusions concern the leakage of the analysed gases from a 10 m^3 tank under the assumed atmospheric conditions. Leakages from tanks of smaller volume will generally pose a hazard to a relatively smaller area, while tanks of larger volume or diameter of damage may increase both the range and duration of the hazard. Similarly, different meteorological conditions will influence the dispersion of the analysed gases and, consequently, the expected ranges of the hazard zones and levels of risk. Therefore, quantitative results should not be directly applied to other scenarios without calculating the relevant input parameters again, although the general trends observed for different compositions of the ammonia-hydrogen mixture remain valid. It was indeed the aim of the study to compare risk levels for different gas mixture compositions.

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