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공학석사 학위논문

**슐리렌 및 미산란법을 이용한 고압
암모니아의 스프레이 특성에 대한
실험적 연구**

**An Experimental Study on High-pressure
Spray Characteristics of Ammonia by
Using Schlieren & Mie-scattering Method**

2018년 2월

서울대학교 대학원

기계항공공학부

박 현 호

슐리렌 및 미산란법을 이용한 고압 암모니아의 스프레이 특성에 대한 실험적 연구

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지도교수 송 한 호

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Abstract

An Experimental Study on High-pressure Spray Characteristics of Ammonia by Using Schlieren & Mie-scattering Method

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Ammonia is one of the most produced compounds in the world and has been used in various fields. In addition, many research efforts have been made on hydrogen carriers, direct fuel cells, and the use of fuel in internal combustion engines. However, research on the ammonia spray characteristics as the basis for using ammonia in internal combustion engines has not been conducted. In order to use ammonia as a fuel, the shape of ammonia spray under various chamber ambient pressure conditions at a specific temperature was

photographed by a schlieren and mie-scattering technique using a high-speed camera, and its characteristics were analyzed based on the results. Furthermore, the widely used fuels were sprayed under chamber ambient pressure conditions such as ammonia to compare the spray of ammonia. Experimental results show that the penetration length of the ammonia spray decreases when the chamber atmosphere pressure increases or the background temperature increases. When the chamber ambient pressure was much higher than the vapor pressure at a given temperature, the ammonia spray was flash-boiling and observed that each plume was injected as if it were coming out of one hole. Compared to methane spray, the ammonia spray shows a wider cone angle under the same conditions. Also, despite the fact that ammonia is a liquid in the injector when injected to CVCC, ammonia spray shows spray characteristics similar to methane fuels. Compared with the iso-octane spray experiment, ammonia showed faster evaporation than iso-octane under all pressure conditions.

keywords : Ammonia(암모니아), Spray characteristic(스프레이 특성), Schlieren(슐리렌 기법), Mie-scattering(미산란 기법)

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1. Introduction

1.1 Research background

Globally, regulations are being tightened on the main products of burning traditional fossil fuels. The United States enacted the Corporate Average Fuel Economy (CAFE) standards for CO₂ emissions by 2021, which targets CO₂ emissions of 108g per kilometer, about 23% less than in 2016. In the case of the European Union, emissions standards were set for the same period, aiming at CO₂ emissions of 95 g/km. In addition to emission regulations for transportation, emission regulations for power plants that use large amounts of fuel are also increasing day by day.

As regulations on fossil fuels and emissions from fossil fuels have become more restrictive, various efforts have been made to improve emissions management and fuel combustion efficiency to meet regulatory standards. In recent years, there have been many efforts to use renewable energy to fundamentally reduce these emissions. As can be seen in Figure 1.1, it can be seen that the research and development efforts on renewable energy have steadily increased over the past decade, and renewable energy consumption has also increased exponentially in recent years. (Figure 1.2)

However, the amount of renewable energy varies greatly between regions, as the energy intensity to obtain energy differs depending on the region.(Figure 1.3) Therefore, research on the means of

transporting energy from a region where sufficient renewable energy can be obtained to a region where it is not sufficient has been required, and research using hydrogen has been actively carried out. However, hydrogen is not easy to transport and store, and research has been conducted to develop various hydrogen carriers. Ammonia has also been developed through the use of ammonia-borane as a hydrogen storage material.[1]

However, because it is inefficient to change the ammonia back to hydrogen, research is being conducted to develop an energy conversion devices using ammonia as a fuel directly.

1.2 Previous research

Traditionally, ammonia has been produced industrially using high pressure (150–200 bar) nitrogen and hydrogen through the Haber-Bosch process. Much of the ammonia produced has been used to make fertilizers, as well as industrial refrigerants and household cleaners. Nowadays, research into new fields has been carried out using the characteristics of ammonia. Ammonia contains 1.5 moles of hydrogen molecules per mole of ammonia, representing 17.8% when measured in terms of hydrogen mass fraction. These high hydrogen mass fractions have been studied as excellent hydrogen carriers. [1,2]

In the field of fuel cells, research has also been conducted to develop an energy conversion device using ammonia. By using ammonia that does not generate carbon when supplying hydrogen instead of using fuel from methane or methanol, which can lead to

carbon poisoning in the anode when the SOFC is operated, it is possible to obtain satisfactory results when operating the fuel cell at high temperature.[3,4]

In addition, studies for use as fuel for internal combustion engines (ICEs) have been conducted for a long time. During World War II, an internal combustion engine vehicle powered by ammonia was developed in Belgium. Because of the war, diesel could not be used, so the buses were operated using fuel mixed with coal gas.[5] These results led to the SI and CI engine experiments using ammonia in the 1960s.[6] Since then, research on engines using ammonia has been rarely conducted. Recently, studies have been conducted on fuels that mixed ammonia with conventional diesel and gasoline to reduce carbon oxide emissions. Ammonia has significantly lower flame propagation speeds and narrow flammability limits than gasoline fuels. Also, In comparison with diesel fuel, since ammonia has a relatively high self-ignition temperature and high evaporation heat during fuel injection, it is difficult to study an engine using ammonia as a sole fuel. Therefore, ammonia is mixed with a conventional fuel at a certain ratio and was used in the study. [7,8]

1.3 Ammonia fuel

Ammonia is one of the most produced compounds in the world. Ammonia is a molecule composed of one nitrogen atom and three hydrogen atoms. Because there is no carbon atom in the molecule, ammonia does not generate any carbon oxides (carbon monoxide,

carbon dioxide, etc.) when completely burned. In addition, the vapor pressure of ammonia at room temperature is as low as about 7 bar, so it is easy to pressurize and store, and the infrastructure for carrying is already well-equipped. Due to the above-mentioned advantage of production and storage, transportation and the not producing any oxides of carbon, the ammonia is used as the fuel for the energy conversion device through the combustion, although the heating value of the fuel is only about 40% of that of gasoline and diesel.[9]

However, when ammonia is used as fuel for energy conversion devices, there is a problem to be solved because of the fuel characteristics of ammonia. The self-ignition temperature of ammonia is much higher than that of other fuels for compression ignition engines, so self-ignition is not easy and requires higher temperature and pressure in the cylinder during injection. In addition, the high latent heat of vaporization of ammonia can also be a problem. As the injected ammonia evaporates, the temperature of the cylinder or chamber is lowered, which also prevents spontaneous combustion of the ammonia fuel.

Even when used in spark ignition engines, there are some problems due to fuel properties. First, ammonia has significantly lower flame propagation velocity than gasoline used in current spark ignition engines. The flame propagation rate of known ammonia is about 1/6 of the flame propagation rate of gasoline, so that the burning period of the fuel becomes longer. This causes more problems when controlling various timings within the engine where

the rpm changes. And because ammonia has a narrow range of flammability limits, it is difficult for the fuel mixed with air to burn. In order to be used as fuel for the engine, it is necessary to overcome the limitations described above. The fuel characteristics of ammonia are compared with other fuels and summarized in Table 1.1 [10]

1.4 Research object

Several attempts have been made to test with ammonia, but no spray characterization experiments with ammonia have been performed. It is important to understand the spraying characteristics of this fuel in order to use ammonia in energy conversion devices such as engines for transitional fuel such as ammonia. The atomization and evaporation that occur when the ammonia fuel is injected into the energy conversion device affects the mixing of the fuel and the air, and may further affect the overall combustion process and ultimately the efficiency of the energy conversion device.

In this study, the geometric spray pattern of ammonia fuel according to the vapor pressure at a specific temperature is photographed using schlieren method and mie-scattering method among many optical imaging techniques. And then, we study the spray characteristics using image processing technology and compare how they differ from existing conventional fuels under the same experimental conditions. The data and trends obtained in this experiment can be used as a basic experiment for experiments using ammonia as a fuel in the future.

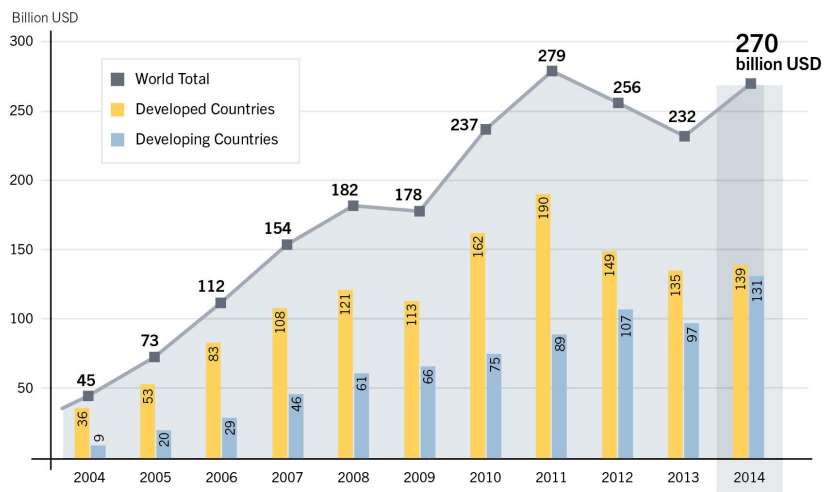


Figure 1.1 Global New Investment in Renewable Power and Fuels[11]

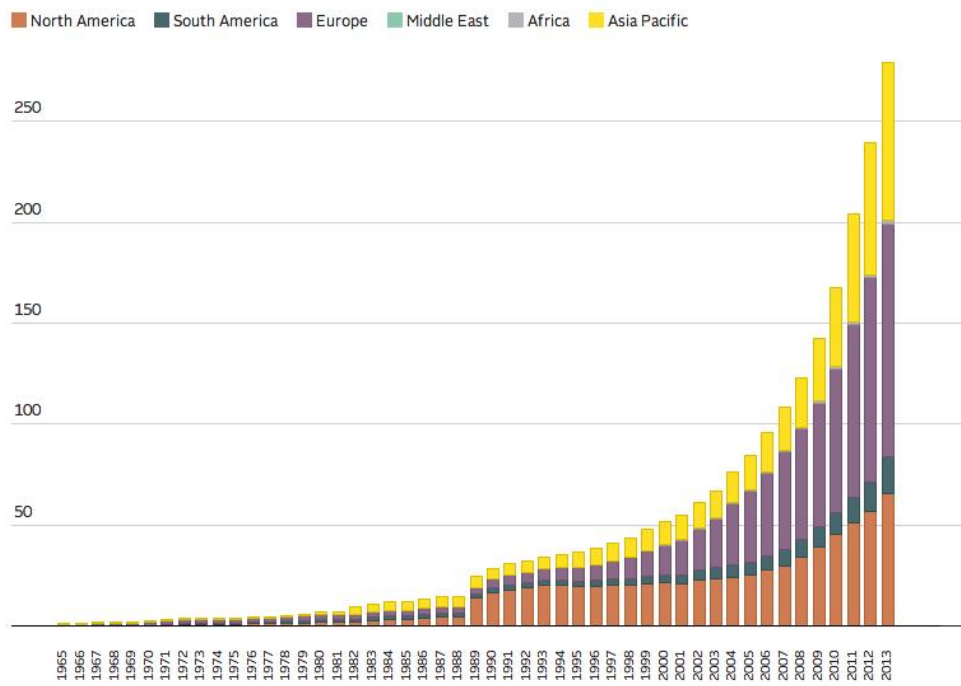


Figure 1.2 Global renewable energy consumption[12]

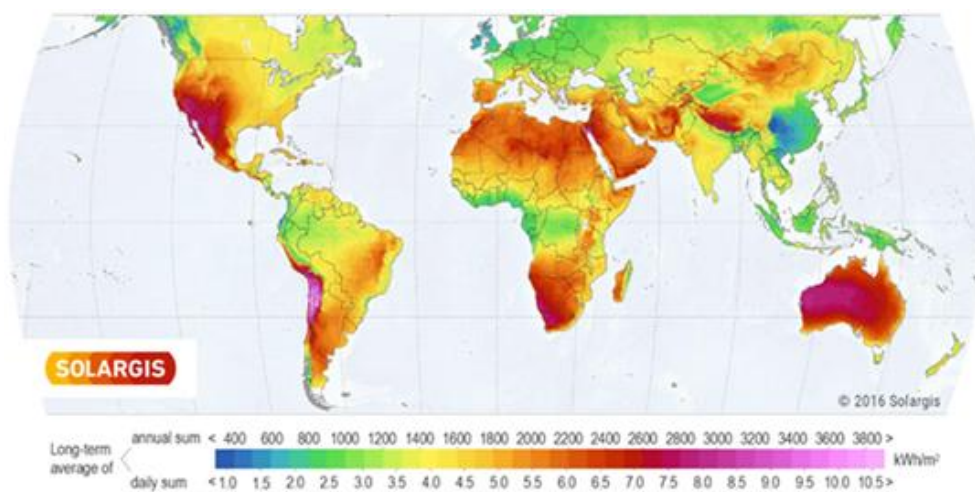


Figure 1.3 World map of solar direct normal irradiation[13]

Table 1.1 Characteristics of ammonia fuel compared to conventional fuel

Property	Gasoline	Diesel	Natural gas	H ₂	NH ₃
Flammability limit, (volumes % in air)	1.4 - 7.6	0.6-5.5	5-15	4-75	16-25
Auto ignition Temp(°C)	300	230	450	571	651
Storage temp.(°C)	25	25	25	25	25
Storage Press.(kPa)	101.3	101.3	25000	35000	1000
Density(kg/m ³)	698	840	187.2	25	602.8
LHV(MJ/kg)	42.5	43	38.1	120.1	18.8
Values to match energy of volumetric of Gasoline					
Fuel Volume(L)	1	0.82	4.16	9.88	2.62
Fuel Weight(kg)	0.698	0.689	0.779	0.247	1.578

2. Experimental setup and methodology

The purpose of this experiment is to record and photograph sprayed images of pure ammonia and iso-octane and methane using a high-speed camera. And then photographed images are analyzed by Matlab program, The result images were analyzed qualitatively for the effect of various variables on each spray profile.

2.1 System rig setup

First, when constructing a system for providing ammonia fuel to the injector due to the corrosiveness of ammonia, it is important to select the material in contact with ammonia. Ammonia actually corrodes copper and rubber, so copper and rubber should not be used to construct the system. Also, ammonia is harmful to the human body, so be careful when sealing the system. Figure 2.1 shows the apparatus used in this experiment. The setup consisted of a gas booster fuel pressurization system (Figure 2.2), a static combustion chamber (Figure 2.3) and an optical measurement setup.

2.1.1 Ammonia fuel pressurization system

The ammonia from the gas cabinet is pressurized through an air-operated gas booster, and the gas booster is equipped with a needle valve at the rear end to prevent damage to the line due to rapid vaporization as ammonia travels along the line. The pressurized ammonia enters the bladder accumulator. The bladder accumulator is

used to keep the pressure of the ammonia entering the accumulator constant by using nitrogen. It also helps to prevent waste of fuel between experiments by storing ammonia after the experiment. A Bosch 6-hole GDI injector was used to inject pressurized fuel. The constant volume combustion chamber is in the form of a hexahedron and is designed to observe the spraying of the spray through the optical measurement method using a quartz window on the four sides. In addition, all surfaces of the CVCC are surrounded by a heating jacket, which increases the temperature inside the CVCC and the temperature of the injector tip. So that fuel injection experiments can be performed under different temperature conditions.

2.1.2 Optic system

For the analysis of spray profile, mie-scattering method and schlieren method were used. The liquid particles of the spray were observed using mie-scattering, which is generally a way of scattering light through particles larger than the wavelength of light. The schlieren method, which obtains images through the difference of the density of the medium, was used to simultaneously observe the gas phase spray on the evaporated ammonia from the injector as well as the liquid. When the signal from the generator is transmitted to the injector driver and the high-speed camera at the same time by connecting to the pulse generator, the camera starts shooting at a predetermined shutter speed, and the injector injects fuel after the internal mechanical delay. The pictures taken by the camera are sent

to a computer, which stores the pictures and processes them for analysis using Matlab programs.

2.2 Experimental condition

The experiment proceeds by injecting a total of three types of fuel (ammonia, gasoline, and methane) into the CVCC. The experimental conditions commonly used during three experiments are shown in Table 2.1. First, experiments using only ammonia proceed at 25 ° C and 50 ° C, respectively. Experiments are carried out using the mie-scattering and schlieren techniques under four conditions (0.1, 0.5, 1, and 2 times) of the pressure in the combustion chamber based on the vapor pressure at each temperature. Since the vapor pressure of ammonia is 10 bar at 25 degrees Celsius and 20 bar at 50 degrees Celsius, the experiment under 25°C is performed at 1, 5, 10, and 20 bar, and the experiment under 50°C is performed at 2, 10, 20 and 40 bar. Ammonia test conditions are shown in Table 2.2.

After the experiment to find out the characteristics of the ammonia injection, Additional experiments were performed for comparing the two fuels (gasoline and methane), which are currently commercially available. Since the gasoline sold in the market is a mixture, the experiment was conducted using iso-octane, which is a substitute fuel for gasoline. Experiments were carried out at 25°C and chamber background pressure conditions such as the ammonia spray test at 25°C. Because gasoline is present in liquid state under experimental conditions, it is photographed using only mie-scattering method unlike

ammonia.

In addition, the spray test using methane was carried out at 25 °C in the same manner as the previous two experiments. Experiments were carried out under the same pressure as the ammonia test at the same temperature. Because methane is present as a gas under experimental conditions, it is difficult to obtain the shape of a spray using the mie-scattering method. Therefore, methane spray experiments were carried out using the schlieren technique. Information on methane and gasoline experimental conditions is shown in Table 2.3.

In summary, the ammonia injection experiments were carried out for a total of 16 sets, taking into account the temperature, the measuring method and the pressure conditions, and four sets of gasoline and methane were measured. Each experiment set was repeated three times to minimize shot-to-shot variations. The macroscopic characteristics and morphology of the spray obtained from the repetitive results show qualitatively how the ammonia spray behaves when the ratio of the internal pressure to the vapor pressure is varied. In addition, the injection duration, the camera exposure time, and the shutter speed of the injector, which are thought to affect the experiment, were fixed.

2.3 Image processing

The raw-image of the spray obtained through the experimental procedure is not suitable for direct analysis. It is difficult to analyze

directly because it may contain various noise such as the gas filled to achieve the predetermined chamber background pressure. To effectively remove the noise from the obtained spray shape, several image processing techniques were applied for accurate analysis. A series of processing was performed using the Matlab program.[14] As shown in Figure 2.4, the image processing process is as follows. The background is removed by the threshold value difference between the first image in time and the other image. After that, the remaining image of the spray part is binarized to obtain a black and white image.

2.4 Spray characteristic

The parameters obtained from the spray injection data obtained through image processing are defined as shown in Figure 2.5. There is a spray penetration S , defined as the distance from the origin of the spray to the farthest point in the spray axis direction. It is known that this spray phenomenon is affected by the injection pressure and the chamber background. Next, the spray cone angle θ is determined by the angle formed by the line that is formed by the point farthest from the axis from the origin and the spray axis direction. This factor is known to be influenced by the density of the fuel and the density of the chamber background gas. We measured the angle of the spray cone at a point with a sufficiently large penetration value from the injector nozzle, since an error that can not be accurately measured may be caused near the tip of the injector when the spray is not fully-developed when this is measured.

However, due to the problem of injector installation, the angle of both sides of the spray axis may be slightly different. To overcome this, we obtain the angle of both sides of the spray axis, add them, and define this as the final spray cone angle. Once the above two factors are determined, factors such as spray tip speed can be obtained. In addition, the spray projected area and spray volume are also frequently considered in spray characterization.

Also, when the background pressure of the chamber in which the fuel is injected is much lower than the steam pressure at the corresponding temperature of the fuel, the spray appears to evaporate at the same time as the injection without being sprayed in the form of a general cone. The characteristic of these sprays is called flash-boiling.[15] When the fuel is injected in the above-mentioned superheated state, the flash-boiling is initiated by boiling within the fuel droplet, forming a vapor bubble. Once boiling occurs, the degree of evaporation inside the droplet becomes larger than the support of the droplet surface, and the internal bubble grows. As the growing bubble grows, the droplet bursts. When this flash-boiling occurs, the atomization of the fuel is accelerated, and the degree of spreading into the chamber and the cylinder becomes large so that the fuel mixes well with the internal gas. In conclusion, flash-boiling can help self-ignition if the shape of the cylinder and chamber is well controlled. Because vapor pressure of ammonia is much higher than 1 atm under the temperature conditions of the experiment (Table 1.1), it can be easily seen when the fuel is injected if the background pressure inside the chamber is adjusted.

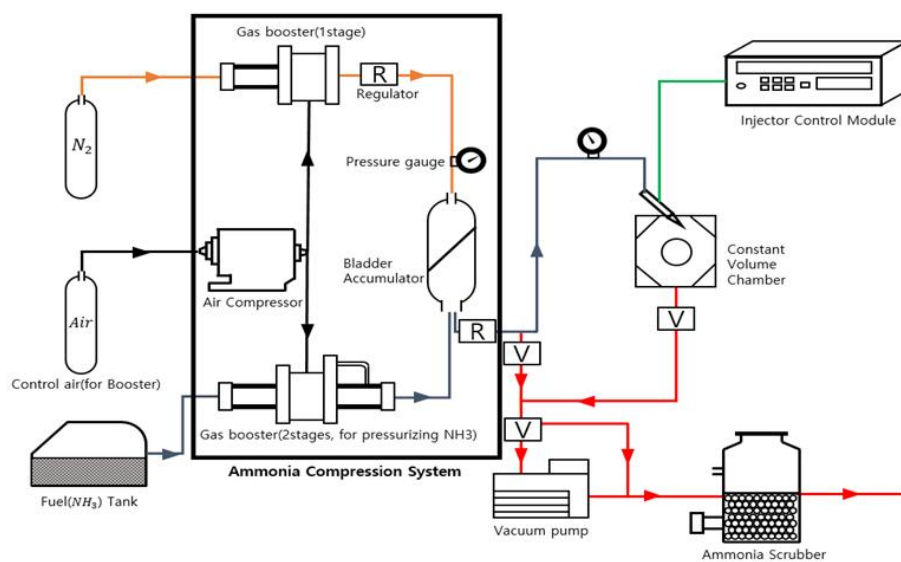


Figure 2.1 Apparatus used in this experiment

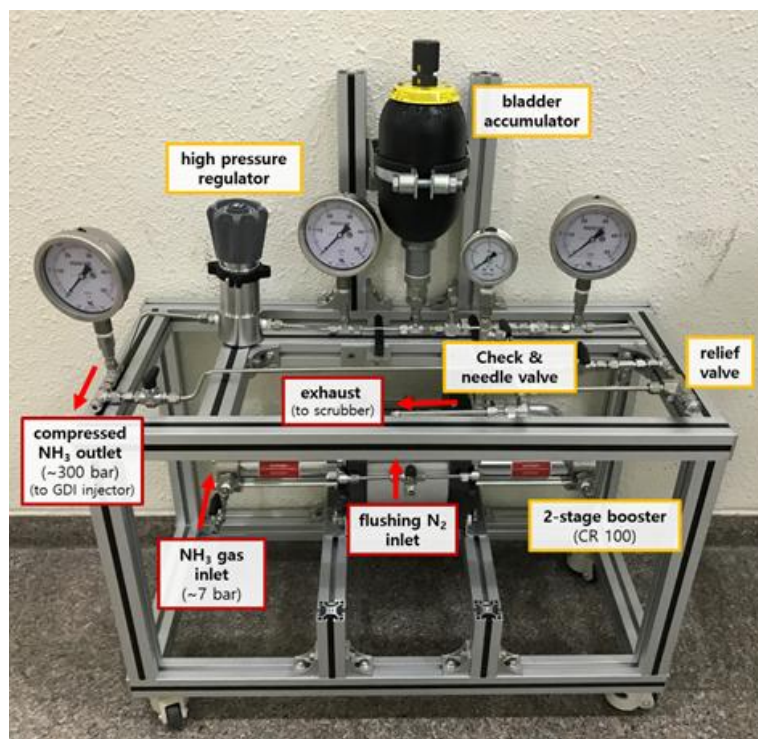


Figure 2.2 Ammonia pressurizing system



Figure 2.3 Constant volume combustion chamber

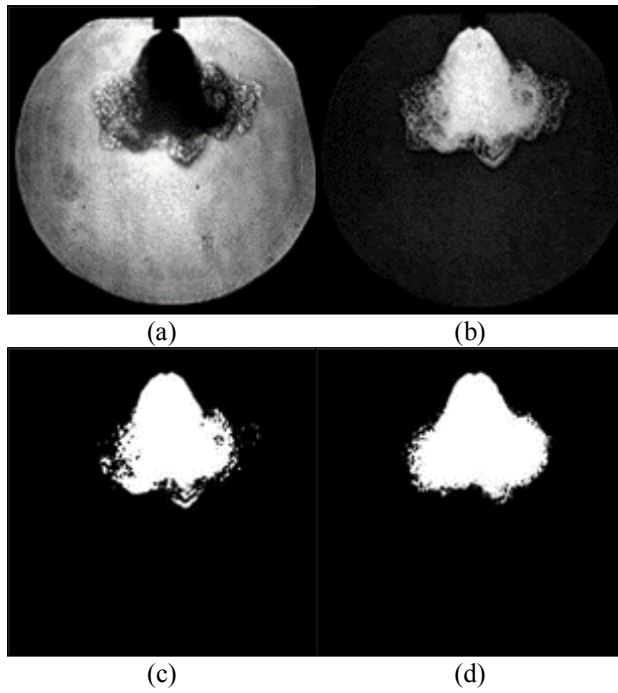


Figure 2.4 image processing of a spray photograph. (a) raw image (b) background subtraction (c) binarization (d) after morphological operations

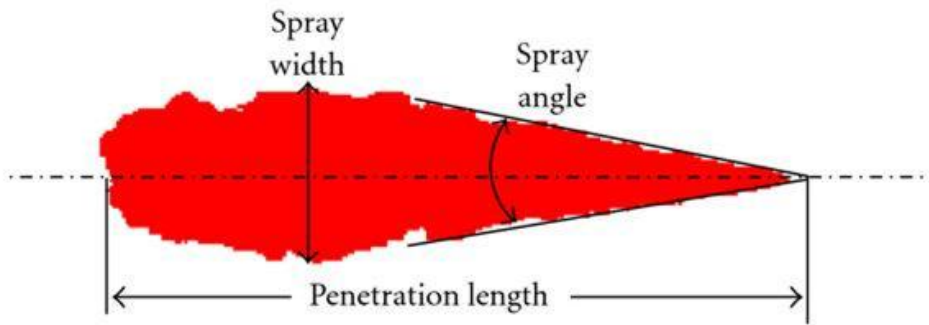


Figure 2.5 Spray characteristics obtained by spray experiment[16]

Table 2.1 General experimental condition

	Conditions of experiment
Frame rate	10000 fps
Exposure time	1/950000 second
Resolution	512 x 512 pixels
Fuel	Ammonia / Iso-octane / Methane
Injection duration	5 millisecond
Injection pressure	100 bar
Injector type	Bosch HDEV-5-2LS high pressure injector
Number of hole	6
Nozzle size	0.170 mm

Table 2.2 Temperature & pressure condition of NH₃ experiment

	Conditions of experiment
Chamber temperature	25, 50 °C
Chamber pressure	1, 5, 10, 20 bar at 25 °C
	2, 10, 20, 40 bar at 50 °C

Table 2.3 Temperature & pressure condition of conventional fuel spray experiment

	Conditions of experiment
Chamber temperature	25°C
Chamber pressure	1, 5, 10, 20 bar at 25°C

3. Result

3.1 Experiment of ammonia spraying characteristics

The vapor pressure of ammonia is about 10 bar at 25 degrees and about 20 bar at 50 degrees. In the experiment, the chamber pressure for the vapor pressure at that temperature was used as a factor. The Schlieren results are shown in the Figure 3.1 below, showing how the spray behavior of the sprayer varies with temperature as the value of factor is changed to 0.1, 0.5, 1, 2. All the photographs shown in the figure were shot at 2ms after the start of spraying. As a result, in the case of spraying of ammonia, the penetration becomes shorter as the background pressure in the chamber increases at the same temperature. Also, the penetration becomes shorter as the temperature increases for all pressure conditions. However, if the P_c / P_v values are the same, the shape of the spray is not significantly affected by the temperature.

When P_c / P_v value is 0.1 in four sets of experiments with P_c / P_v values, the spraying pattern is significantly different from the other three cases. The spray is observed as though it is ejected from one nozzle rather than six nozzles. This phenomenon can be explained by the flash-boiling that occurs when the fuel is injected at a significantly lower background pressure than the vapor pressure of the fuel, as the interaction between the spray plums from each nozzle occurs. Since the flash-boiling phenomenon also correlates to the

temperature of the fuel itself, this phenomenon is observed closer to the nozzle in experiments where the fuel temperature is higher.

Figure 3.2 shows the ammonia spray shot at the same temperature and pressure conditions taken with the mie-scattering technique, with the schlieren photograph taken earlier. By spraying the spray in two different ways, it is possible to observe atomization by spraying ammonia into liquid and gas. The results of the Mie-scattering measurement are affected by the background temperature and pressure as well as the results obtained using the Schlieren method. Also, it can be seen that as the background pressure increases, the schlieren image and the mie-scattering image show similar shapes. This is due to the fact that the degree of atomization of ammonia is low as measured at a higher background pressure than the vapor pressure at the corresponding temperature.

3.2 Comparison experiment with conventional fuels

The experiment was carried out with two fuels currently used under the same background pressure conditions as the previously analyzed ammonia spray test. This explains how the spray characteristics of the two fuels differ from ammonia sprays. The background temperature of the chamber was set at 25 degrees Celsius and the experiment was conducted using methane and iso-octane which is used as simulated fuel for gasoline.

3.2.1 Methane spray experiment

Methane is the main source of natural gas and is always present

in the gas under the temperature conditions which the experiment is conducted. Therefore, the injection of methane fuel was observed using Schlieren method. The measured methane spray is shown in Figure 3.3.

Compared with the ammonia injection at the same background pressure, the ammonia injection shows a wider cone angle than the methane injection. It can be seen that methane also shows the same tendency as ammonia has decreased spray penetration with increasing pressure. However, because methane is a gaseous fuel present in the gas at 100 bar of the injection pressure, no phase change occurs and flash-boiling does not occur like the ammonia spray at 1 bar of the chamber pressure. It is interesting that the ammonia spray behavior at 1, 5, 10, and 20 bar looks similar to the spray form of methane, despite the injection of pressurized liquid fuel in the case of ammonia.

3.2.2 Iso-octane spray experiment

Experiments were conducted in the same manner using iso-octane under the same experimental conditions as methane. Contrary to methane, iso-octane is a liquid in the temperature and pressure conditions under which the experiment is conducted, so the shape of the spray was measured using the mie-scattering method.

Compared with the spray of ammonia and iso-octane, the spray penetration length of iso-octane was higher than that of ammonia at each pressure. On the other hand, as shown in figure 3.4, iso-octane does not have a single plume as in the ammonia experiment, even though the background pressure is low. At a background pressure of

25 degrees Celsius, the vapor pressure of iso-octane is much lower than that of ammonia, so in iso-octane spray experiments, flash-boiling does not occur. Comparing the spray images of the two fuels obtained with mie-scattering, the size of the white highlight area is larger for iso-octane than area of ammonia spray. This shows that the evaporation rate of the fuel after the injection of ammonia is larger than the evaporation rate of the iso-octane after the injection.

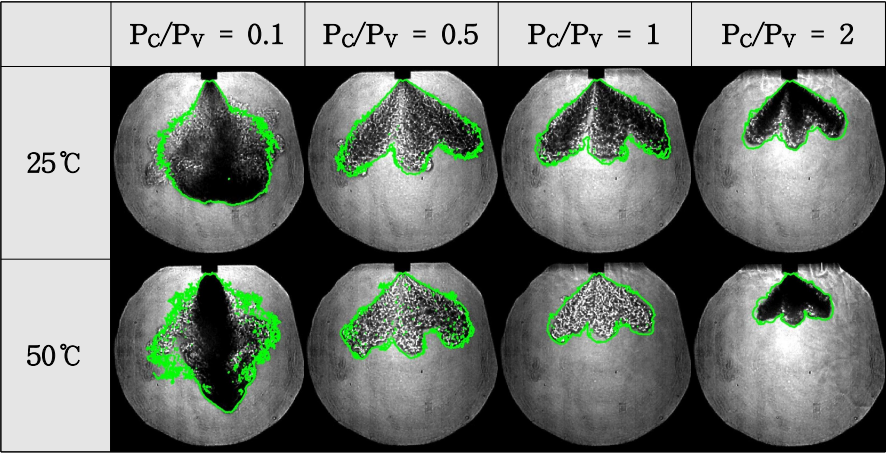


Figure 3.1 Ammonia spray taken by schlieren method

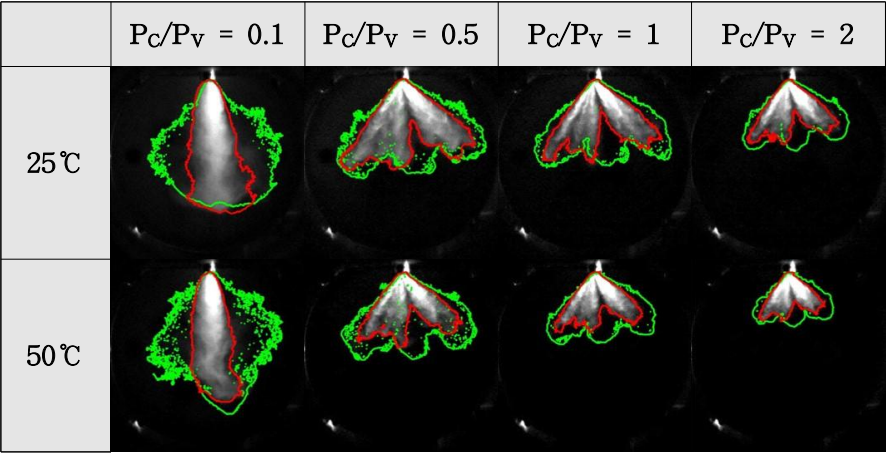


Figure 3.2 Comparison of ammonia spray by shooting method

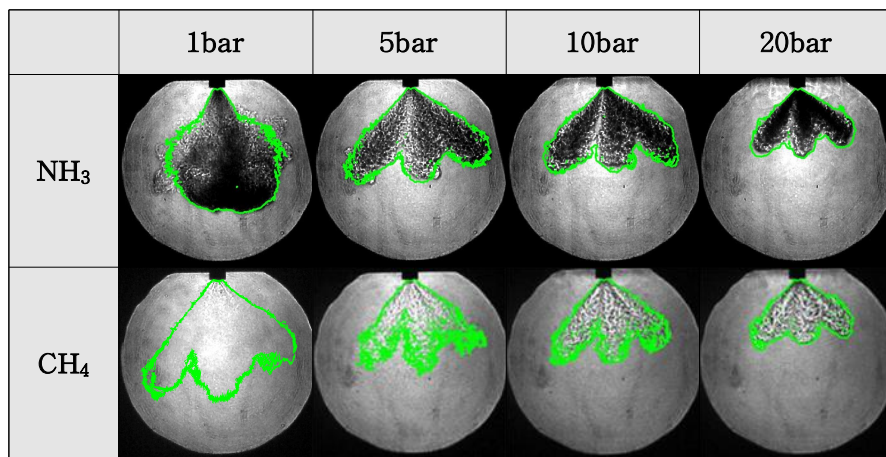


Figure 3.3 Spray shape comparison of methane and ammonia

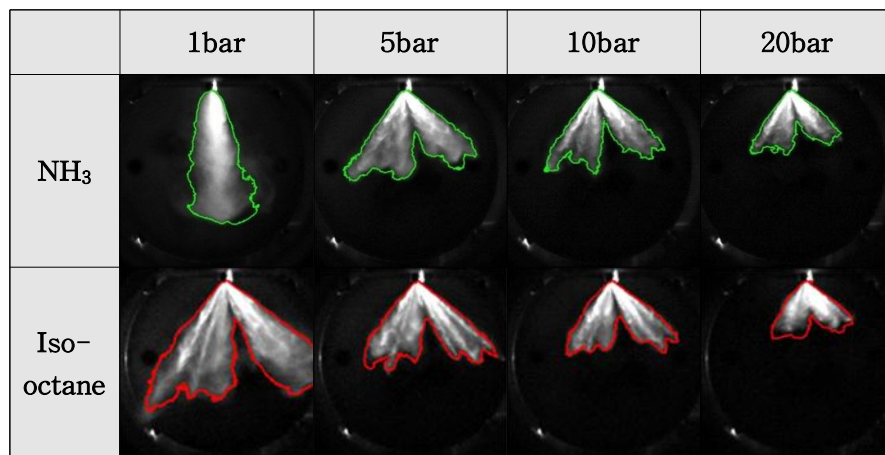


Figure 3.4 Spray shape comparison of iso-octane and ammonia

4. Conclusion

In this study, the characteristics of high-pressure ammonia spray were studied under various temperature and pressure conditions under non-reacting conditions. Here is the summary of the findings:

(1) The penetration length of the ammonia spray decreased when the chamber background pressure increased or the chamber background temperature increased. If the chamber background pressure rises above the vapor pressure of the ammonia, the shape of the liquid spray and the shape of the gas spray obtained through the schlieren coincide. And as the pressure ratio(P_C/P_V) decreased, the vaporization of the fuel increased.

(2) At the pressure condition of $P_C \ll P_V$, it was observed that the shape of the ammonia spray was injected as if the respective plums were merged as if they were coming out from one hole by flash-boiling. During flash-boiling, we could see vaporization from the top of the spray.

(3) Compared to the methane spray, the ammonia spray showed a wider spray cone angle. Also, despite the fact that ammonia is a liquid in the injector when injected to CVCC, ammonia spray shows spray characteristics similar to methane fuels. Compared with iso-octane spray experiments, ammonia was found to cause faster evaporation under all pressure conditions than iso-octane.

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요약

솔리렌 및 미산란법을 이용한 고압 암모니아의 스프레이 특성에 대한 실험적 연구

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박현호

암모니아는 세계에서 가장 많이 생산되는 화합물 중 하나로, 다양한 분야에서 이용되어 왔다. 연구적인 측면에서도 수소의 운반체, 연료전지의 직접적인 연료, 내연기관에서 연료의 사용 등으로 많은 노력이 있어 왔다. 그러나 내연기관 등에서 암모니아를 이용하기 위한 기반이 되는 암모니아 스프레이 분사특성에 대한 연구는 진행되지 않았다. 암모니아를 연료로 사용하기 위하여, 특정 온도에서 다양한 분위기압력 조건에서의 암모니아 스프레이의 형상을 고속카메라를 이용하여 솔리렌법과 미산란 기법으로 촬영하였고, 결과물을 토대로 그 특성을 분석하였다. 나아가 널리 사용되고 있는 연료를 암모니아와 같은 챔버 분위기 압력 조건에서 분사하여 암모니아의 스프레이를 비교하였다. 실험 결과, 챔버 분위기 압력이 증가하거나 배경 온도가 증가하게 되면, 암모니아 스프레이의

관통 거리가 줄어드는 것을 확인할 수 있었다. 정해진 온도에서의 증기 압력보다 챔버 분위기 압력이 매우 클 때, 암모니아 스프레이는 플래시 비등이 일어나 마치 하나의 구멍에서 나오는 것처럼 각각의 플럼이 합쳐져서 분사되는 것을 관측하였다. 메탄 스프레이에 비해 암모니아 스프레이는 같은 조건에서 더 넓은 스프레이 분무 각을 형성하는 것을 보여준다. 또한, 암모니아가 정적연소실에 분사될 때 액체상태로 분사되지만, 분사된 스프레이의 형태는 기체연료인 메탄의 연료와 비슷한 형태를 보이는 것을 알 수 있었다. 같은 온도 및 압력 조건에서 iso-octane 스프레이 실험과 비교하여 암모니아는 iso-octane에 비해 모든 압력조건에서 더 빠른 증발을 일으키는 것을 관찰 할 수 있었다.

주요어 : 암모니아, 스프레이 특성, 숄리렌 기법, 미산란 기법

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