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研究課題名（和文） Experimental and numerical study on the interactions between liquid ammonia flashing spray and flame properties for carbon-free technologies.

研究課題名（英文） Experimental and numerical study on the interactions between liquid ammonia flashing spray and flame properties for carbon-free technologies.

研究代表者

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研究成果の概要（和文）：この研究プロジェクトでは、液体アンモニア噴霧の特性を調査し、噴霧の数値モデリングの検証のために実験結果のデータセットを得た。噴射条件（温度、形状）の変化が噴霧パターンに及ぼす影響を明らかにした。液体アンモニアのフラッシュボイリング（飽和圧力以下の環境下で液体を吐出すると急激に蒸発し、微粒化すること）へ移行する条件を検討した。また、燃焼実験を行い、噴射条件の変化による影響、噴霧パターン、火炎安定性を関連付け、産業用噴射ノズルの設計に必要な知見を得ることができました。本研究プロジェクトの成果は、国際会議および査読付き雑誌論文でも発信されました。

研究成果の学術的意義や社会的意義
The present research is expected to contribute to the development of accurate numerical models necessary for the design of efficient, low-emission industrial burners. It also provides valuable information on geometry which can be used as guidelines for the design of nozzles for liquid ammonia.

研究成果の概要（英文）：During this research project, the characteristics of liquid ammonia sprays were investigated, and a dataset of experimental results was obtained for the validation of the numerical modeling of the spray. The effects of the change in the injection conditions (temperature and geometry) on the spray patterns were clarified. The conditions leading to the transition to flash-boiling (sudden evaporation occurring when a liquid is discharged in an environment below its saturation pressure, leading to fine atomization) for liquid ammonia were investigated. Combustion experiments were also performed to relate the effect of the change in the injection conditions, the spray patterns, and the flame stabilization, providing some insights for the design of injection nozzles for industrial applications.
The results of this research project were also communicated at an international conference and in a peer-review journal (Fuel, IF: 6.6) paper.

研究分野： Combustion - Thermal Engineering

キーワード： liquid ammonia　spray combustion　flash-boiling spray　spray modeling　spray characteristics　model validation

１．研究開始当初の背景

In recent years, the use of ammonia as a carbon-free fuel [1,2] has been investigated as a means to cut carbon emissions in power generation applications, with, for example, the demonstration of gas turbines fuelled by gaseous ammonia. However, the use of gaseous ammonia requires vaporizing systems, which limits the operational flexibility of the gas turbine, hindering the development of such applications. Thus, direct injection of liquid ammonia is under consideration [3,4].

Yet, liquid ammonia spray combustion remains merely investigated. In addition, and due to its thermophysical properties, ammonia might go through flash-boiling evaporation when directly injected into the combustor. Flash-boiling is known to strongly affect spray characteristics and it is thus crucial to clarify the characteristics of ammonia spray to enable the development of industrial applications. Moreover, liquid ammonia combustion modeling remains challenging due to the lack of data to confirm its validity, and both experimental and numerical work is necessary.

２．研究の目的

This present research project thus aims at the observation of the ammonia spray flame through both experiment and numerical simulation. The configuration selected in this study is simple to ease the comparison between experimental and numerical results and assess the validity of the models employed. The spray characteristics will be first investigated and modeled in the cold flow case, with for objective to establish the injection conditions in which flash-boiling occurs and their effect on the spray alone. Then, the interactions between the flame and the spray will be analyzed in the combustion case.

３．研究の方法

Liquid ammonia is supplied from a 50 kg heated cylinder, at a pressure close to 1.4 MPa, as represented in Fig. 1a. The liquid ammonia then flows through a co-axial line surrounded by a coolant which temperature can be regulated between 293 K and 253 K, giving temperature at injection between 270 K and 293 K. The flow rate is controlled by a Coriolis mass flow controller. Pressure is checked along the supply line using pressure gauges and a pressure transducer. Thermocouples are also positioned along the line to confirm the temperature at various points up to the nozzle. The nozzles employed in the study are represented in Fig. 1b. The flow goes first through a cylindrical section of 5 mm, then through a reduction section following a 120º angle before reaching the orifice of the nozzle, of diameter *D* and length *L*, and being released in an atmospheric environment. The effect of the orifice geometry on the spray shape was investigated, and the diameter and aspect ratio of the orifice varied in the ranges 0.1 mm ~ 0.21 mm and 2.5 ~ 10, respectively, as summarized in Table 1. Aluminum nozzles were employed in the preliminary stages of the project, as well as commercial pressure swirl atomizer nozzles. They were replaced by stainless-steel nozzles obtained by electro-erosion for the main study of the spray patterns. Glass nozzles were also realized to observe the inner flow, and the presence of two-phase flow corresponding to internal flash boiling. The spray patterns were observed by backlit imaging and the spray angles were extracted from those images.

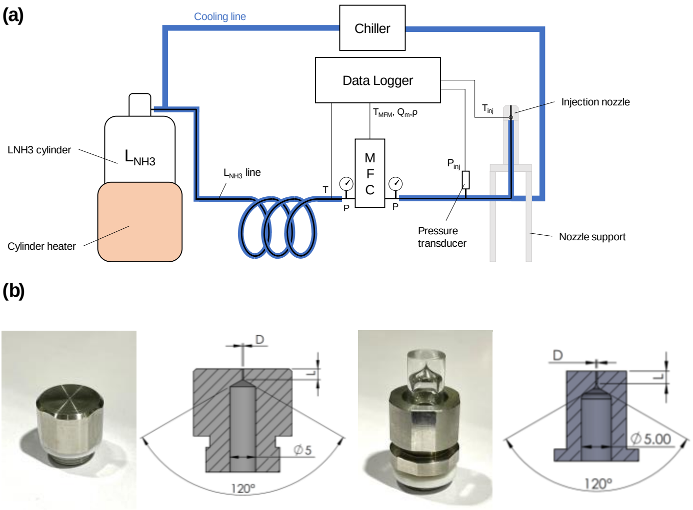

Figure 1. Liquid ammonia supply (a) and nozzle geometry (b)

Table 1. Nozzle orifice size

Nozzle number	D	L/D
#1	0.1	2.5
#2	0.15	2.5
#3	0.21	2.5
#4	0.1	5
#5	0.15	5
#6	0.21	5
#7	0.13*	10
#8	0.15	10
#9	0.21	10

** For L/D = 10, minimum manufacturable diameter was 0.13 mm*

The combustion experiments were realized using the burner represented in Fig.2. The liquid ammonia spray is surrounded by a simple coflow of oxygen and nitrogen. The velocity of the coflow varied between 0 and 1 m/s and the oxygen fraction, β, was between 0.4 and 1. The velocity profile of the coflow was checked by hot-wire measurement, for later comparison with numerical simulations.

Numerical simulations were done using OpenFOAM software [5], employing the Eulerian-Lagragian approach as implemented in the sprayFoam solver. In addition, to investigate the flow inside the nozzle, and in the direct vicinity of the outlet, two-phase flow modeling was considered. For that purpose, the volume-of-fluid approach (VOF) as implemented in the compressibleInterFoam solver was selected. Nonetheless, to account for flash boiling evaporation, modifications of the existing solver are necessary. The numerical study could not be completed during the duration of the project and consequently not introduced in the present report.

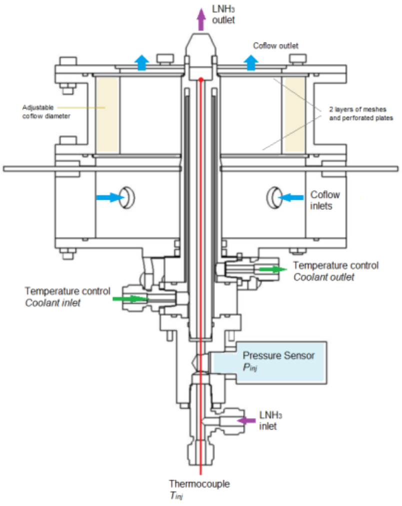

Figure 2. Burner geometry

４．研究成果

（1）非燃焼研究

The effects of both injection temperature and orifice geometry on liquid ammonia spray were investigated. The change in diameter, *D*, had a limited effect on the spray patterns and is not developed here, but the effects of the aspect ratio, *L/D*, and the degree of superheat are presented in Fig. 3. The images were taken for fixed injection velocity conditions, U_{inj} = 30 m/s, and *D* = 0.21 mm. The degree of superheat, *Rp*, is varied between 8.5, 5.2, and 3.5 corresponding to temperatures of 293 K, 280 K, and 270 K and Reynolds numbers close to 28 000, 25 800, and 23 600, respectively. For the smallest values of *L/D*, in Figs. 3a and 3b, the ammonia goes out in a straight column, with a diameter close to that of the orifices. In addition, though break-up remains mostly mechanical in those two *L/D* cases, bubbles appear in the column for the case *L/D* = 5, for the largest degree of superheat. This corresponds to the onset of flash boiling. When the aspect ratio is further increased to 10, as in Fig 3c., a different type of spray pattern can be observed. The spray presents an overall bowl-like shape, with, depending on the degree of superheat, an inner core of liquid jet, breaking up in ligament and droplets of dimensions comparable to the orifice diameter, surrounded by a cone of fine droplets which for most cannot be resolved with the present resolution (40 ~ 50 μm). This fine mist, with droplets several orders lower than the orifice diameter, is characteristic of flash-boiling atomization. This change in the behavior of the spray with increasing *L/D* is consistent with the observations done in previous work as reviewed by Sher et al [6]. In their review, they associated larger *L/D* with the reattachment of the liquid to the orifice wall and greater heterogeneous nucleation at the contact between the liquid and the wall.

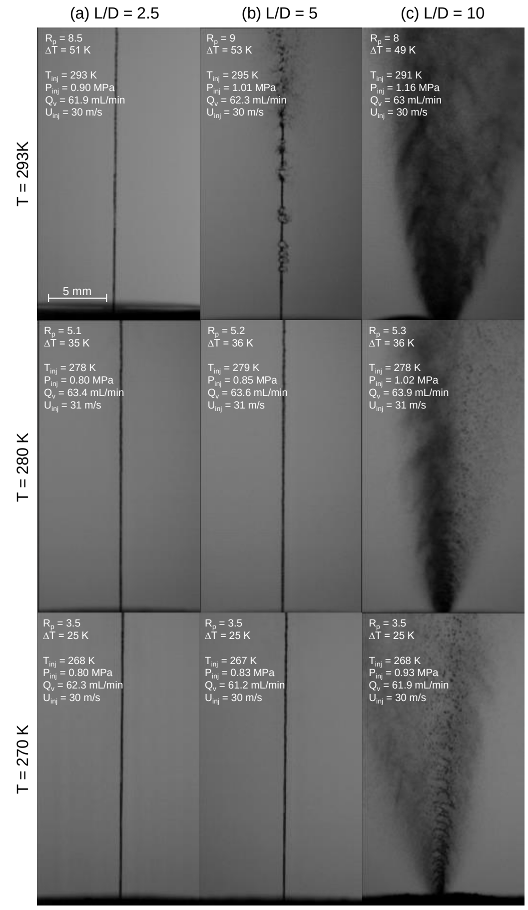

Figure 3. Liquid ammonia spray backlit images

Looking at the effect of the degree of superheat, whereas no major change is observed for *L/D* = 2.5, a small variation of the degree of superheat (20 K) strongly affects the shape of the spray in the case *L/D* = 5 and *L/D* = 10. In those cases, the sprays vary from purely mechanical break-up to the onset of partial flashing (*L/D* = 5) and from partially to fully flashing spray (*L/D* = 10).

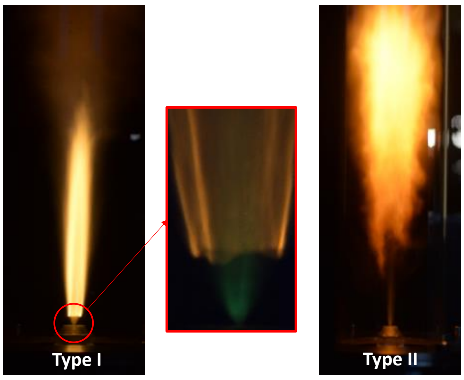

Figure 4. Observation of the spray flame

The evolution of the spray angle with the degree of superheat and for the various geometry investigated was obtained using the backlit imaging and will be used for numerical modeling validation.

（2）燃焼研究

The flame stabilization range was studied for a selected set of injection conditions (P_{inj}, T_{inj}), keeping the velocity of the liquid droplet at the outlet of the nozzle constant, while varying the coflow velocity and oxygen fraction. The injection pressure was maintained constant (P_{inj} = 1.1 MPa, and two temperature conditions were considered (cooling – T_{inj} = 270 K and non-cooling – T_{inj} = 293 K). Experiments were done with an aluminum nozzle (A2024) of diameter D = 0.21 mm and an aspect ratio L/D = 10 and for stainless steel nozzle (SUS304) of diameter D = 0.21 mm and 0.15 mm and L/D = 10. Those nozzles correspond to the cases with partial to fully-flashing conditions. Depending on the coflow conditions, several flame patterns and stabilization regimes were observed as illustrated in Fig. 4. Two flame types were essentially distinguished and denoted as flame type I and type II. Type I correspond to a highly luminous flame, stabilizing close to the spray outlet, with limited fluctuation in the flame base position. The base of the flame is relatively smooth with few wrinkling. Type II corresponds to a flame of much lower luminous intensity, presenting a highly turbulent front, with a more complex structure, stabilizing much higher above the nozzle, and spreading very close to the glass liner.

The stabilization limits were obtained as follows: the O_2 flow rate was fixed to a pre-defined value, and the flame was ignited. For those initial low coflow velocity and high oxygen fraction, β, the flame stabilized as Type I, or low-lift position. Then, the flow rate of N_2 was then gradually increased, leading to a simultaneous increase of the coflow velocity, U_{co}, and a decrease of the oxygen fraction, β. The flame eventually transits to Type II or gets directly blown out. From type II, if keeping the increase in the N_2 flow rate, the flame extinguishes. If decreasing the N_2 coflow from Type II, the flame will transit back to Type I, for a lower U_{co} and higher β than the reverse transition from Type I to Type II. This presents a similarity with the hysteresis phenomenon observed in gaseous jet flames [7], for the liftoff and re-attachment transition. Here too, the presence of the flame, and the hot burnt gas, is expected to have a positive effect on the flow field around the spray and at the flame base, and help maintain the lower lifted flame (Type I) for higher U_{co} /lower β. This process is repeated for different initial O_2 flow rates to obtain the stabilization domain of the flame in Fig. 5.

The transitions are plotted in Fig. 5 for several injection conditions. The two left graphs correspond both to the cooling case (T_{inj} ~ 270 K), for the same nozzle inner geometry (D = 0.21, L/D =10) but made of aluminum for the upper one ① and stainless steel for the lower one ②. It can be observed that the stabilization range is slightly extended for the aluminum nozzle. For the same coflow velocity range, the transition Type I → Type II occurs for values of β between 0.35 and 0.4, whereas they are in the range β = 0.4 ~ 0.45 for the stainless steel nozzle. This is consistent with the backlit imaging observations made for both nozzles. Both nozzles presented partially flashing sprays but with an earlier onset of flashing for the aluminum nozzle (closer to fully-flash). The liquid core in the cooled aluminum nozzle case is smaller than in the stainless steel one, and the atomization in small droplets is expected to be better, which might explain the larger stabilization range.

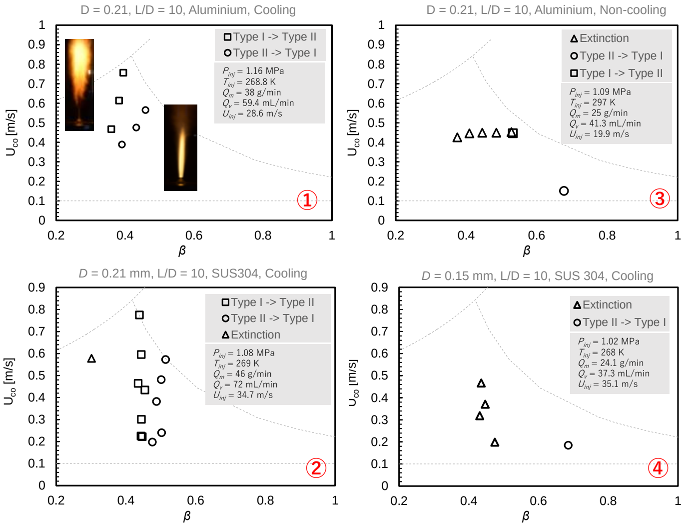

Figure 5. Stabilization domain of the spray flame

The two upper graphs both correspond to the aluminum nozzle, with ① and without cooling ③. Contrary to what is expected from the higher degree of superheat, the flame stabilization domain was reduced in the non-cooling case.

Differences in flow rate between both cases can be noticed (~ 20 mL/min) leading to a drop in the velocity of the jet (~ 9 m/s), and in mixing with the surrounding coflow. Spray patterns obtained from the backlight image under slightly higher injection pressure are introduced in Fig. 6 for discussion. It can be seen in Fig. 6 that the non-cooled spray is particularly dense and that entrainment from the surrounding is more limited. The drop in the flow rate might also affect the stabilization as developed in the comparison of ② and ④ below.

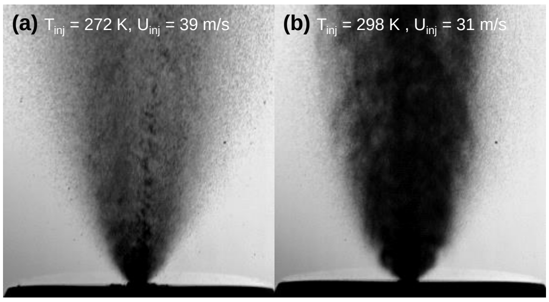

Figure 6. Injection temperature effect on spray pattern for #9-1-A at $P_{inj} \sim 1.5$ MPa: (a) $T_{inj} = 272$ K, cooling case; (b) $T_{inj} = 298$ K, non-cooling case.

The effect of the nozzle diameter, and flow rate reduction, is investigated by comparison of nozzles of $D = 0.21$ mm ② and $D = 0.15$ mm ④ in the same injection condition (P_{inj}, T_{inj}). Like in the non-cooling aluminum case ③, direct extinction from Type I flame is observed for $D = 0.15$ mm, and extinction occurs around $\beta = 0.45$, close to the value for which the transition Type I → II is observed for the nozzle $D = 0.21$ mm②. The injection velocity is the same for both cases, but the flow rate is smaller in case ④ and the spray appeared shorter than in case ② (shorter penetration length). Thus, the ammonia gas fraction, droplet density, size, and velocity distributions are expected to differ in both sprays at higher positions above the outlet of the nozzle, which corresponds to the region where the Type II flame stabilizes. This might explain to some extent the absence of Type II flame in that case.

In the four cases investigated, and except for case ③ the coflow velocity has a relatively minor effect on the transition limit when compared to the oxygen fraction, β, and the LNH_3 injection conditions. Overall, from the present results, it might be inferred that for the spray flame in coflow configuration, in addition to the coflow oxygen fraction, β, and the quality of atomization and flashing (size of droplet and spray broadening), mixing (mostly controlled by the spray in this case) is determinant in the flame stabilization.

(3) Main conclusions

- The effect of the orifice geometry and degree of superheat on the transition to flashing of ammonia jets were investigated.
- LNH_3 flames could be stabilized in a coflow of O_2/N_2 mixture with oxygen fraction above 0.4 ~ 0.5 for partially to fully-flashing spray. Stabilization was not possible for non-flashing liquid ammonia jet.
- For the same injection velocity and flow rate conditions, an earlier onset (triggered by surface or material effect) led to a broader stabilization range.
- Reduction in the flow rate and velocity led to a narrower stabilization range.
- Clarification is necessary in case ③ to distinguish between the effect of the reduction in injection flow rate and velocity and the effect of temperature and degree of superheat.

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５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　1件／うち国際共著　1件／うちオープンアクセス　0件）

１．著者名 Colson Sophie, Yamashita Hirofumi, Oku Kohei, Somarathne Kapuruge Don Kunkuma Amila, Kudo Taku, Hayakawa Akihiro, Kobayashi Hideaki	４．巻 348
２．論文標題 Study on the effect of injection temperature and nozzle geometry on the flashing transition of liquid ammonia spray	５．発行年 2023年
３．雑誌名 Fuel	６．最初と最後の頁 128612～128612
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.fuel.2023.128612	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計1件（うち招待講演　0件／うち国際学会　1件）

１．発表者名 Sophie Colson, Hirofumi Yamashita, Taku Kudo, Akihiro Hayakawa and Hideaki Kobayashi
２．発表標題 Study of the effects of injection temperature and nozzle geometry on liquid ammonia spray characteristics and flame stabilization (poster presentation)
３．学会等名 39th International Symposium on Combustion（国際学会）
４．発表年 2022年～2023年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関