

2025 | 106

## Visualizing Ammonia Combustion: From Laminar Flames to Engine Application

Visualizations

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## ABSTRACT

Ammonia is one of the most promising future fuels for high-power combustion systems like large internal combustion engines and gas turbines. The number of publications and applications of ammonia combustion has increased rapidly in recent years. However, many phenomena of ammonia combustion remain unresolved and require further investigation. While fundamental experiments under laminar conditions show a very low reactivity of ammonia, the engine combustion of pure ammonia or ammonia with the addition of a small fraction of combustion promoters provides satisfactory combustion efficiency and stability. To gain a deeper understanding of the phenomena governing ammonia combustion, the LEC and FHNW have developed a methodology using a series of experimental setups to apply optical investigations ranging from quiescent, ambient conditions up to highly turbulent, engine-relevant conditions. The optical data furthermore provide an extraordinary wealth of information to develop and validate detailed simulation models.

This paper presents the holistic concept for a methodology that allows the experimental distinction of the effects relevant to ammonia combustion. Concepts for measuring the laminar burning velocity, turbulent burning velocity, turbulent jet ignition, and turbulence-flame interaction under engine-like thermodynamic and flow conditions are described. For each stage of the methodology, a specific optical test rig is designed or adapted to meet the relevant requirements. Selected results are presented, which underline the unique combustion behavior of ammonia and ammonia/hydrogen blends and demonstrate the applicability of ammonia in internal combustion engines. Thermodynamic instabilities are identified to have a decisive impact on the flame propagation of ammonia/air mixtures. The improvement of the combustion properties at high temperatures and pressures as well as high turbulence levels is reasoned with enhanced reactivity, strong turbulence-flame interaction, and the Lewis number effect. Future perspectives on the insights from optical data into ammonia combustion and the relevance of optical data for developing ammonia combustion models are derived.

## 1 INTRODUCTION

Ammonia is a promising energy carrier and fuel for large engines. It is carbon-free and can be produced from renewable energy, air, and water. Furthermore, ammonia is a suitable carrier of renewable energy due to its combination of high production efficiency, high volumetric energy density, and low energy demand for storage. Thus, green ammonia is an effective solution for transporting renewable energy over long distances, for example from regions with abundant solar and wind energy to regions with high energy demand [1].

The combustion characteristics of ammonia ( $\text{NH}_3$ ) pose challenges to its implementation as a primary fuel for large engine combustion systems. While  $\text{NH}_3$  use eliminates all carbon-based emissions from the conversion of the main fuel, nitrogen-based emissions gain further relevance. Unburned  $\text{NH}_3$ , nitrogen oxides ( $\text{NO}_x$ ), and nitrous oxide ( $\text{N}_2\text{O}$ ) are the subject of current research on  $\text{NH}_3$  combustion concepts and aftertreatment systems. Addressing the global warming potential of  $\text{N}_2\text{O}$ , Wüthrich et al. [2] assess the greenhouse gas reduction potential of pure  $\text{NH}_3$  combustion at high indicated mean effective pressures. With fuel/air mixtures that are close to stoichiometric, this potential is over 90% without the use of an exhaust gas aftertreatment system.

In premixed combustion, the low reactivity of  $\text{NH}_3$  at ambient conditions is well-documented in recent literature and indicated by its low laminar burning velocity, high ignition energy, high auto-ignition temperature, and narrow flammability limits [3–6]. Thus, a combustion promoter might be required to apply  $\text{NH}_3$  as a primary fuel in internal combustion engines (ICE). Hydrogen ( $\text{H}_2$ ) is a suitable enhancer since it can be produced from  $\text{NH}_3$  dissociation, so-called "cracking." The laminar burning velocity of  $\text{NH}_3$  and partially cracked  $\text{NH}_3$  was investigated in [7], where it was shown that laminar burning velocities equal to or higher than the one of  $\text{CH}_4$  are achieved with a 40%  $\text{NH}_3$  cracking ratio at the same air/fuel equivalence ratio  $\lambda$  under ambient conditions. Under engine-like conditions, Klawitter et al. [8] found that the required  $\text{NH}_3$  cracking ratio can be decreased to approximately 10%. Lower cracking ratios are required due to the favorable response of  $\text{NH}_3$ /air mixtures to engine-like conditions, i.e., high temperatures, high pressures, and high turbulence levels, as well as air/fuel equivalence ratios above unity.

Thus, combustion concepts for premixed  $\text{NH}_3$  combustion should focus on diluted conditions at high temperatures and under highly turbulent conditions. High temperatures and turbulence

levels during combustion may be achieved by utilizing high compression ratios, elevated intake temperatures, and ignition concepts such as turbulent jet ignition (TJI). The idea behind TJI is to locate the ignition source, usually a spark plug, inside a small additional combustion chamber, the prechamber (PC), which is connected via small nozzles to the main combustion chamber (MC). After ignition, the combustion inside the PC propels the hot, partially burned gases into the MC, which provides high ignition energy and additional turbulence to the premixed charge in the MC, enhancing the combustion speed and reducing unburned gas emissions [9–11].

Visualizing combustion is essential to characterizing new fuels. Schlieren imaging is useful for investigating flame propagation and flame front phenomena such as stretch effects or thermodiffusive instabilities.  $\text{OH}^*$  chemiluminescence in combination with schlieren imaging serves to distinguish between flame fronts and density gradients. The optical data provide an extraordinary wealth of information for the development and validation of detailed simulation models, which require optical investigations under various test conditions. Fundamental parameters such as laminar burning velocity (LBV) and turbulent burning velocity (TBV) are required inputs for simulation models [7, 8, 12]. Application-oriented investigations, e.g., on optical engines, enable assessment of the transferability of fundamental insights to engine conditions, understand phenomena in practical combustors, and identify the relevant factors for engine optimization [13, 14].  $\text{NH}_3$  is especially suitable for optical investigations due to its low flame speed and high flame thickness, thereby enabling the precise capture of flame propagation and the detailed resolution of the flame structure [15, 16].

Previous development of turbulent combustion models for computational fluid dynamics (CFD) of the ICE mainly focused on carbon-based fuels. To apply these models to non-carbon-based fuels such as  $\text{NH}_3$  or  $\text{H}_2$ , model adaptations are required to ensure the simulation quality is similar to that of carbon-based fuels. Fundamental experimental measurements of  $\text{NH}_3$  combustion at different complexity levels provide the basis for the development of high-fidelity turbulent combustion modeling approaches.

This paper presents a novel holistic methodology for characterizing the premixed combustion behavior of future fuels, specifically  $\text{NH}_3$ . With this methodology, relevant processes of  $\text{NH}_3$  combustion are isolated at different levels of complexity and investigated experimentally under clearly defined boundary conditions. This allows

the optical measurement data to be used in the further development and validation of simulation models. Developed by LEC GmbH (LEC) and the University of Applied Sciences and Arts Northwestern Switzerland (FHNW), the approach is presented in Chapter 2, which describes the optical test rigs and optical measurement techniques involved. It was developed out of a thorough survey of current literature and research results of LEC and FHNW, which are outlined in Chapters 3.1 and 3.2. Based on this, future perspectives on the insights from optical data into NH<sub>3</sub> combustion are derived in Chapter 3.3. The relevance of optical data for developing NH<sub>3</sub> combustion models is elaborated in Chapter 3.4.

## 2 METHODOLOGY

This chapter outlines the methods developed for the optical investigation of NH<sub>3</sub> as a fuel for large engines. This approach provides a holistic methodology that deepens the understanding of NH<sub>3</sub> and NH<sub>3</sub>-H<sub>2</sub> blend combustion under various conditions and establishes a comprehensive database for enhancing and developing simulation models. The ultimate goal is to facilitate the rapid and effective adoption of NH<sub>3</sub> as a primary fuel for large bore engines.

### 2.1 Holistic Flame Visualization Concept

To understand the combustion behavior of NH<sub>3</sub>, it is critical to experimentally identify the factors that influence combustion. Consequently, the present

approach aims to measure and visualize fundamental parameters, i.e., LBV and TBV, as well as engine-relevant processes, i.e., TJI, under idealized conditions and realistic, engine-like conditions. Figure 1 presents the optical test rigs involved and classifies them according to their test conditions; the entire range from fundamental to engine-like conditions is covered. The test rigs are introduced in the following section.

This methodology focuses on premixed, spark-ignited combustion. Future optical investigation of high-pressure jet-guided NH<sub>3</sub> combustion that is in planning is not covered here.

### 2.2 Optical Test Rigs

All test rigs used in this methodology are briefly described below, and their specific purposes and unique features are highlighted.

#### Optical Quiescent CVCC

An optical constant volume combustion chamber (CVCC) is currently being developed and built at the LEC as part of a joint research project. It can be operated as a quiescent CVCC to investigate laminar flames. The precise determination of the LBV from outwardly propagating spherical flames is achieved by applying the constant pressure method. The results regarding laminar flames are an essential input for the validation of reaction mechanisms and combustion simulations and serve as a reference for turbulent flame results.

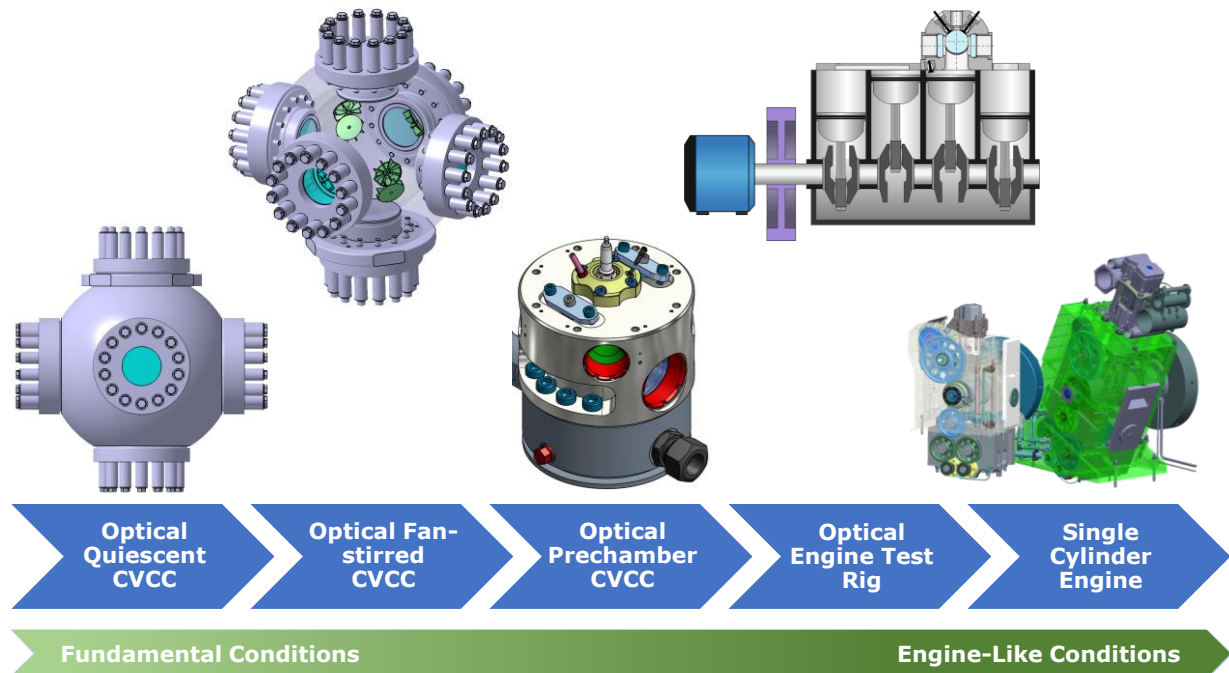


Figure 1: Overview of research test rigs implemented in the methodology for visualising premixed ammonia combustion under various test conditions: laminar and quiescent conditions, idealized turbulent conditions, idealized prechamber and turbulent jet conditions, engine-like conditions

The laminar flame investigations can be performed within a limited range of initial temperatures and pressures due to experimental as well as flame-induced limitations.

### Optical Fan-stirred CVCC

The optical CVCC currently being developed and built at the LEC also allows the investigation of turbulent flames. To this end, six fans are arranged symmetrically within the inner sphere to create nearly homogeneous isotropic turbulence in the center of the chamber while inducing minimal mean flow. The fan-stirred CVCC (FS CVCC) enables measurements of the turbulent flame propagation under idealized conditions. The TBV is the central parameter to be determined. Furthermore, the flame propagation and the flame structure can be analyzed with adequate measurement techniques. The relevance of the CVCC results to ICE applications is assured by matching the velocity and length scale ratios of the turbulence and flame scales to ICE operation conditions. This results in comparable combustion regimes and turbulence-flame interaction.

### Optical Prechamber CVCC

An optical prechamber CVCC (OPC) at FHNW is a constant volume combustion chamber designed for detailed investigations of TJI [17]. The OPC is composed of two volumes: the PC (3.4 cm<sup>3</sup>) and the MC (187 cm<sup>3</sup>). The two chambers are connected by an interchangeable nozzle whose maximum orifice diameter is 4 mm. The OPC is currently being redesigned for compatibility with new renewable fuels such as NH<sub>3</sub> and methanol; its internal geometry is practically unvaried in order to benefit from the numerous simulations available [18, 19]. The OPC allows the optical and thermodynamic investigation of the TJI process, including the flame propagation inside the PC, the turbulent jet ignition process in the MC, and the subsequent flame propagation in the MC, and it provides data for validating high-fidelity LES.

### Optical Engine Test Rig

The Flex-OeCoS is an optical test rig developed to study combustion under engine-like conditions. Flame propagation is investigated using a specially designed cylinder head which features two opposed windows, each with a diameter of 60 mm. These windows provide optical access through which the initial stages of flame development may be observed. Attached to one cylinder of a four-cylinder engine block, the Flex-OeCoS is equipped with an electrically driven crankshaft and a fully variable valvetrain. This configuration permits the precise control of thermodynamic conditions and

turbulence levels, replicating the varying thermodynamic conditions of actual engine operation. The cylinder fitted with the optical head has a 1990 cm<sup>3</sup> displacement and a compression ratio of 13.8:1. Thanks to the possibility of conditioning the intake air pressure and temperature as well as the engine speed, investigation of a variety of engine-like pressure and temperature trajectories and turbulence levels during ignition and combustion is possible. The optical cylinder head can accommodate different inserts so that a spark plug can be mounted directly in the main chamber to study turbulent flame propagation or an actively fueled prechamber installed to investigate TJI processes. A more detailed description and characterization of the test rig is found in [2, 13, 14, 20].

### Single-cylinder Engine

The LEC has several large bore single-cylinder engines (SCE) with up to 900 kW output power that are retrofitted to be fueled by NH<sub>3</sub> and H<sub>2</sub> [21, 22]. Comprehensive databases of the combustion progress under real engine conditions are obtained from in-cylinder pressure indication.

SCEs are the most application-oriented research test rigs used with the present methodology. Compared to multicylinder engines, they allow improved access to the combustion chamber for sensors such as temperature and pressure probes or optical access for endoscopic or tomographic investigations. Optical fibers can be applied to the combustion chamber, in the cylinder head gasket, or as part of the spark plug without influencing the geometry of the combustion chamber significantly [23, 24]. However, optical accessibility remains limited due to geometrical and mechanical constraints.

## **2.3 Optical Measurement Techniques**

This section introduces the main optical measurement techniques of the methodology. High-speed schlieren imaging serves as the primary method for capturing flame propagation by visualizing density gradients. This provides clear insights into flame dynamics and behavior. In addition, simultaneous chemiluminescence imaging is employed to gain deeper insight into the reaction zone; it is particularly valuable for the study of TJI as it highlights regions with active chemical reactions through light emission from excited species. Particle image velocimetry (PIV) measurements are also critical as they characterize the flow conditions within the combustion chambers of the various test rigs by tracking the movement of seeded particles in the flow. This tracking provides the detailed velocity field data essential to validating and refining numerical simulations.

Together, these measurement techniques contribute to a comprehensive understanding of  $\text{NH}_3$  combustion processes.

### High-speed Schlieren Imaging

Schlieren imaging captures subtle changes in refractive index variations, enabling the visualization of density changes within a transparent medium. The setup typically starts with a light source shaped into a point source using a diaphragm. This light is then focused through a planoconvex lens to generate collimated (parallel) beams. The collimated light passes through the test area, where density gradients induce a slight refraction due to changes in the refractive index. After traversing the area of interest, a second planoconvex lens refocuses the light onto a high-speed camera, which records the resulting image. A knife edge or secondary iris diaphragm is placed at the focal point to block diverging beams, accentuating the refractive variations to enhance sensitivity [25, 26].

### High-speed Chemiluminescence Imaging

Flame monitoring using the emitted light is widely used due to its simplicity and non-intrusiveness. Spectroscopic techniques are often used to track the de-excitation of excited radicals usually produced within the flame front.  $\text{OH}^*$  and  $\text{CH}^*$  can be used to follow combustion parameters such as flame structure, strain rate, and equivalence ratio [27]. The filtering of flame emission within the spectral range of chemically excited radicals is a powerful technique to qualitatively track the occurrence and location of specific radicals. The setup typically features a narrow bandpass filter to isolate the primary emission from the targeted excited species. A high-speed camera, often paired with an image intensifier or an intensified camera, enhances sensitivity and accurately captures detailed emissions [28].

Due to their strong correlation with the heat release rate and their effectiveness in visualizing ignition delay and flame propagation mechanisms,  $\text{OH}^*$  radicals are frequently utilized in combustion diagnostics [27, 29]. In the context of  $\text{NH}_3$  combustion, the exploration of additional radicals such as  $\text{NH}^*$  could provide valuable insight and open up new possibilities for understanding combustion dynamics.

### Simultaneous Schlieren and Chemiluminescence Imaging for the Study of TJI

Chemiluminescence from specific radicals occurs in the near UV spectral range, enabling simultaneous schlieren and  $\text{OH}^*$

chemiluminescence imaging on the same visualization plane. To this end, dichroic mirrors are used that reflect near UV light, e.g., 310 nm for  $\text{OH}^*$  detection, while transmitting visible wavelengths for schlieren visualization. This setup allows differentiation between regions with density gradients alone and areas where chemical reactions are present, e.g., the flame front. Figure 2 shows simultaneous high-speed imaging using  $\text{OH}^*$  chemiluminescence and schlieren imaging in the MC and schlieren imaging in the PC of the OPC test rig.

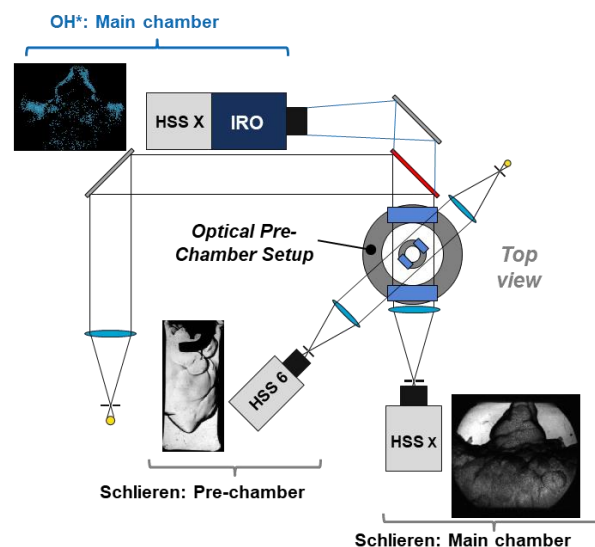


Figure 2: Optical setup for schlieren imaging in the prechamber and simultaneous schlieren and chemoluminescence imaging in the main chamber

The setup described above is particularly relevant to studying TJI in the investigation of gaseous premixed combustion. Dependant on the operating conditions, the jet originating from the PC is not always reactive. Since the turbulent jet originating from the PC has a different temperature and, consequently, a different density than the MC fuel, schlieren imaging detects intensity changes when the jet mixes in the MC. Nonetheless, it is not immediately clear whether the observed schlieren activity is solely due to density gradients or the establishment of a flame front. Simultaneous  $\text{OH}^*$  chemiluminescence imaging enables the detection of chemical reactions occurring on the flame front, thereby permitting the precise quantification of jet quenching, mixing, and reignition phenomena in the MC.

### Particle Image Velocimetry

PIV is a laser-based optical measurement technique for visualizing fluid flow. Seeding particles are introduced into the flow and illuminated by a double-pulsed high-speed laser sheet. The light scattered from the seeding

particles is captured in image pairs by a high-speed camera. The motion of the particles is determined from the image pairs using cross-correlation algorithms. High-speed PIV (HS-PIV) is a variant of PIV that is applied to rapidly changing flows such as in-cylinder flows in an ICE. For further details on the PIV technique, see [7, 20, 30].

HS-PIV investigations that characterize the flow field within the optical engine test rig have already been performed at FHNW [20]. The LEC took HS-PIV measurements in an optically accessible prechamber connected to a rapid compression machine despite severe spatial restrictions [30]. Future PIV investigations will also evaluate the mean flow, turbulence level, homogeneity, and isotropy of the turbulence inside the newly designed FS CVCC.

### 3 OPTICAL RESULTS AND FUTURE PERSPECTIVES ON AMMONIA COMBUSTION

This chapter provides the basis for the planned experimental measurement campaign introduced in the methodology section, including the measurement data that will enable precise modeling of  $\text{NH}_3$  flames. Section 3.1 presents a preliminary investigation of  $\text{NH}_3$  combustion, establishing the basic understanding necessary for exploring effects that will be examined in detail in future studies. The planned research also focuses on TJI of  $\text{NH}_3/\text{H}_2$  flames for which OPC measurement data is currently unavailable. Section 3.2 introduces a basic optical interpretation of TJI combustion by comparing the combustion of methane ( $\text{CH}_4$ ) and  $\text{H}_2$ ; this provides a baseline for comparison in future  $\text{NH}_3$  investigations. Insights derived from optical data into the distinctive combustion behavior of  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$  blends are discussed in Section 3.3. This preliminary analysis deepens the understanding of the fundamental phenomena governing  $\text{NH}_3$  combustion and lays the foundation for the development of  $\text{NH}_3$  combustion models discussed in Section 3.4.

#### 3.1 First Optical Results on Ammonia Combustion

This section presents selected results obtained from the test rigs at the LEC and FHNW that were specifically converted for  $\text{NH}_3$  combustion investigations. It highlights the initial findings of the visualization methodology.

##### Quiescent Conditions

Initial tests of the laminar burning velocity were performed on a rapid compression machine (RCM) available at the LEC. The RCM was repurposed as a CVCC by fixing the piston in the maximum

position. High-speed schlieren imaging visualized the flame front according to the described method. The RCM was retrofitted to  $\text{NH}_3$ -compatible components. A detailed description of the test conditions and results regarding the laminar burning velocity have been reported in [7]. Outwardly propagating spherical flames were investigated at nearly ambient conditions. The low reactivity of  $\text{NH}_3$  under ambient and laminar conditions was quantified through LBV measurements [7, 12].

The optical data gives insight into the flame propagation of  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$  blends undetectable by other measurement technologies. Figure 3 depicts a partially cracked  $\text{NH}_3$ /air flame at a cracking ratio of 40%, a fuel/air equivalence ratio of 1.11, and an initial temperature and pressure of 298 K and 5.0 bar, respectively. Starting from the critical flame radius, the flame surface becomes increasingly unstable and forms wrinkles. The flame front is obviously wrinkled when the flame radius is greater than 15 mm but starts to affect the flame surface area and thus the flame speed starting at approximately 12 mm.

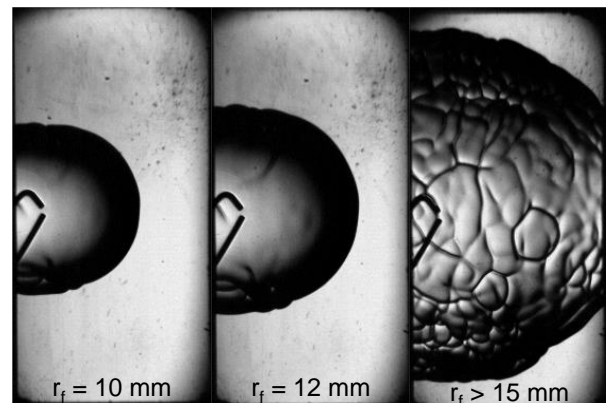


Figure 3: Schlieren images of partially cracked  $\text{NH}_3$  flame at different radii,  $\gamma = 40\%$ ,  $\lambda = 1.11$ ,  $T_0 = 298 \text{ K}$  and  $p_0 = 5.0 \text{ bar}$ ; onset of instabilities visible; reprinted from [7]

The observed thermodiffusive instabilities are caused by nonequal diffusion of heat and mass into the flame front, which is indicated by an effective Lewis number below unity [31]. These instabilities are enhanced by high pressures, low air/fuel equivalence ratios, and high  $\text{NH}_3$  cracking ratios, which yield a higher  $\text{H}_2$  content [12]. The detection of instabilities is essential to the determination of LBV. Once a flame front becomes unstable, local stretching enlarges and accelerates the flame. The flame speed cannot be extrapolated to an unstretched flame since the stretch rate is not simply a function of the flame radius in the unstable flame case. Thus, the LBV of the unstable flame cannot be determined by the detected contour. The optical monitoring of flame front stability is one major advantage of the image-based constant

pressure method over the pressure-based constant volume method [32].

The comparison of the RCM results to literature data shows generally good agreement. However, using the RCM as a CVCC limits the precise measurement of the laminar burning velocity especially in the case of flames with high stretch sensitivity. The optical access limits the evaluable flame radius to 15 mm, the proximity of the ignition location to the cylinder head influences the induced flow of the burned gas, and the small chamber volume causes an early pressure rise. These limitations have been addressed by the development of a new CVCC at the LEC. In this new CVCC, a centrally ignited flame propagates in a spherical chamber. Both the CVCC chamber volume and the optical access are more than twice as large as those of the RCM.

### Engine-like Conditions

Turbulent flame propagation of NH<sub>3</sub> and NH<sub>3</sub>/H<sub>2</sub> blends has been investigated under engine-like conditions on the Flex-OeCoS optical engine test rig at FHNW. Details of the experimental setup and the results of the apparent turbulent flame

propagation can be found in [8, 12]. The improved combustion properties of NH<sub>3</sub> at high temperatures and pressures as well as high turbulence levels have been quantified.

Figure 4 depicts the turbulent flame front evolution of CH<sub>4</sub>/air, NH<sub>3</sub>/air, and partially cracked NH<sub>3</sub>/air (7% cracking ratio) mixtures. When turbulent flame structures are compared at similar propagation stages, the CH<sub>4</sub> mixture recorded at -8°CA and the two NH<sub>3</sub> mixtures at -4°CA, the flame wrinkling seems more pronounced in the NH<sub>3</sub> cases. This increased wrinkling appears visible due to longer flame "fingers" and darker flame fronts, implying more corrugated flame structures as a result of a Lewis number below unity.

### 3.2 Optical Data for the Characterization of Turbulent Jet Ignition

Data on the TJI of NH<sub>3</sub>/H<sub>2</sub> flames in the OPC is currently unavailable. Nevertheless, the introduction of a basic optical interpretation of TJI combustion by comparing the combustion of CH<sub>4</sub>/air and H<sub>2</sub>/air mixtures provides a basis for comparison in future NH<sub>3</sub> investigations.

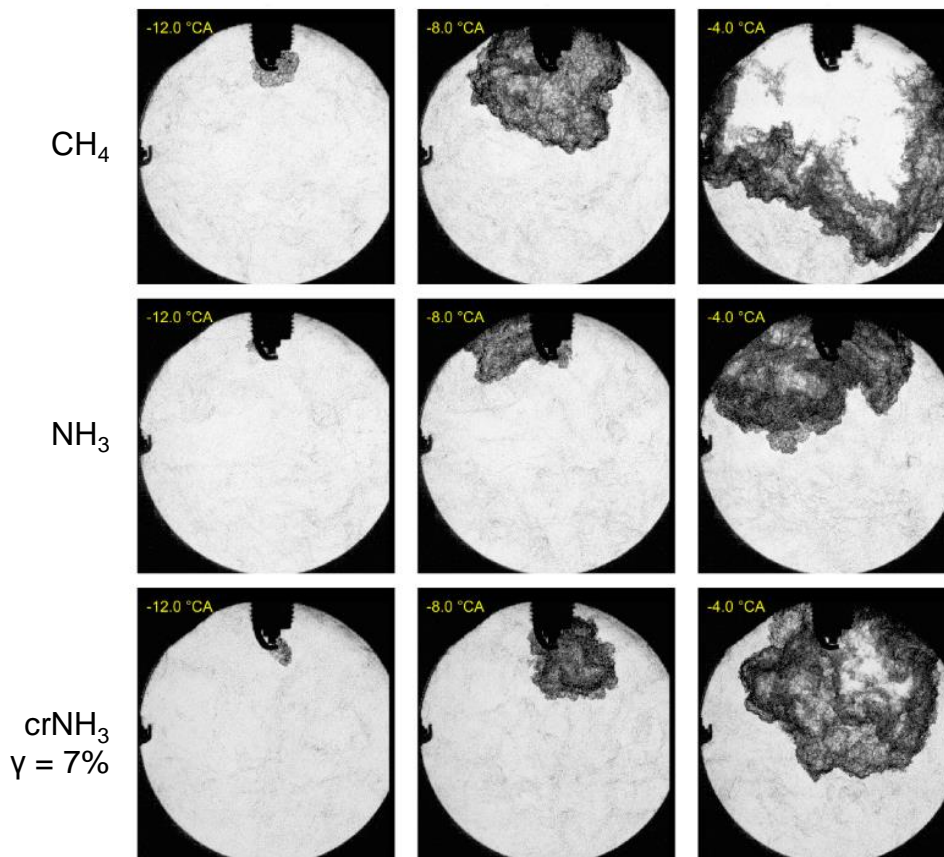


Figure 4: Schlieren images of CH<sub>4</sub> (top row) NH<sub>3</sub> (middle row) and partially cracked NH<sub>3</sub> ( $\gamma = 7\%$ , bottom row) at  $\lambda = 1.25$ ,  $n = 600 \text{ min}^{-1}$ ,  $p_c = 70 \text{ bar}$ ,  $T_{in}(\text{CH}_4) = 323 \text{ K}$ ,  $T_{in}(\text{NH}_3) = T_{in}(\text{crNH}_3) = 373 \text{ K}$

As mentioned above, the jet originating from the PC is not always reactive. Quenching of the turbulent reacting jet entering the MC can happen according to two distinct mechanisms. The first is thermal quenching, which occurs when nozzle wall heat losses exceed the heat released by the flame, preventing the flame from sustaining itself. The second mechanism, which is known as hydrodynamic quenching, is caused by the intense mixing of combustion products from the PC with the cold, unburned mixture in the MC, which rapidly dilutes reactive species and hinders flame propagation [17]. If quenching occurs, reignition may take place after an ignition delay provided that the reactivity of the mixed jet and the fresh charge is sufficiently high. As described in Section 2.3, the combination of schlieren and OH\* chemiluminescence imaging is utilized to distinguish between areas in which mixing alone occurs and areas in which flame propagation is present.

Figure 5 presents two experiments conducted in the OPC. The top sequence depicts results with CH<sub>4</sub>/air mixtures while the bottom sequence shows results with H<sub>2</sub>/air mixtures. In both cases, the OPC is filled to 10 bar and 100°C. After filling, sufficient time is allowed before ignition to ensure initial quiescent conditions. CH<sub>4</sub> and H<sub>2</sub> are injected to achieve air/fuel equivalence ratios of 2.0 and 2.2, respectively, which results in comparable amounts of fuel energy for both experiments.

The OH\* recordings in the MC are superimposed onto the schlieren recordings, with magenta used to highlight OH\* in the CH<sub>4</sub> experiment and light blue for the H<sub>2</sub> experiment.

Despite the initial quiescent conditions, the first frame of the H<sub>2</sub> case shows notable flame wrinkling. This wrinkling is attributed to thermodynamic instabilities arising from the Lewis number being below unity [33].

In the second frame, high schlieren intensity is observed with CH<sub>4</sub> and H<sub>2</sub>; however, no OH\* activity is detected. This indicates that the hot jet originating from the PC is quenched and mixes with the unburned charge in the MC. In the third frame, OH\* activity becomes visible in both cases. With H<sub>2</sub>, the OH\* activity is concentrated near the nozzle, while with CH<sub>4</sub>, it appears on the wall opposite the nozzle. This activity leads to the ignition of the charge in the MC and the subsequent flame propagation.

The TJI optical methodology has proven effective in detecting distinct combustion features in similar cases with different fuels such as CH<sub>4</sub> and H<sub>2</sub>. It will be employed to investigate the TJI behavior of NH<sub>3</sub>/air flames, including jet reactivity, jet mixing, and ignition delay under various conditions.

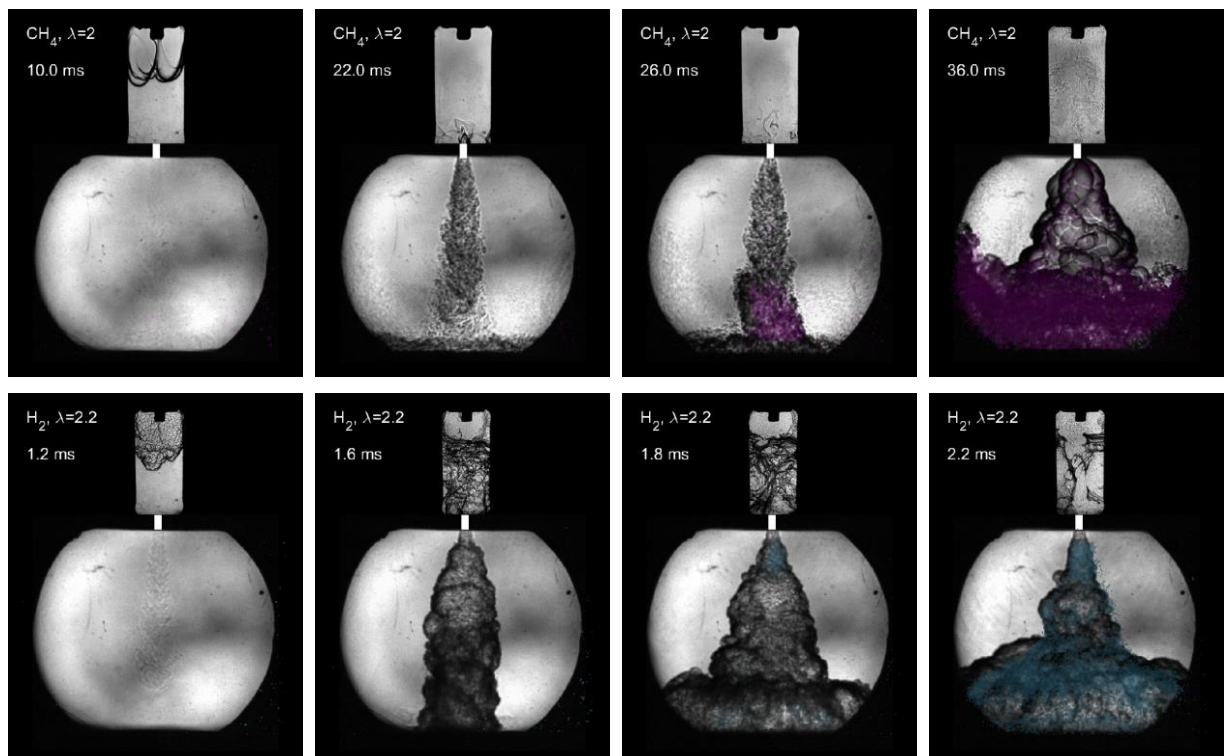


Figure 5: TJI process in the OPC, CH<sub>4</sub>/air mixtures (top) and H<sub>2</sub>/air mixtures (bottom)

### 3.3 Insights into Ammonia Combustion from Optical Data

A comprehensive optical dataset ranging from laminar flames at ambient conditions to highly turbulent flames under engine-like conditions offers a unique opportunity to obtain a consistent chain of knowledge transfer. Insights gained at a fundamental level, e.g., on the FS CVCC and OPC, can be evaluated for their validity and significance in practical combustors.

Under turbulent conditions, combustion regime characterization is essential to assessment of the turbulence-flame interaction. This requires the characterization of the flow field in the combustion chamber, which can be achieved through CFD simulations or optical methods such as PIV investigations. The turbulence scales in combination with flame scales determined experimentally or numerically allow the assessment of the premixed turbulent combustion regimes. With regard to the current optical engine test rig, characteristic measurement points of CH<sub>4</sub>, NH<sub>3</sub>, and partially cracked NH<sub>3</sub> are depicted in the classic Borghi-Peters regime diagram in Figure 6 [12]. Higher Karlovitz numbers and thus stronger turbulence-flame interaction are apparent with NH<sub>3</sub> and partially cracked NH<sub>3</sub> flames as compared to CH<sub>4</sub> flames [8].

This regime diagram assumes equal thermal and mass diffusivities, which is not generally valid for NH<sub>3</sub> and NH<sub>3</sub>/H<sub>2</sub> flames. The reassessment of regime diagrams for NH<sub>3</sub>-based combustion may be facilitated by highly resolved optical data that exposes the actual combustion regime of the flame.

Optical investigations of turbulent flame propagation at engine conditions, e.g., on the current optical engine test rig, illustrate the improved combustion properties of NH<sub>3</sub> at high temperatures and pressures and high turbulence levels. In addition, the optical data exposes the Lewis number effect with lean fuel/air mixtures and its significance in engine conditions. Direct measurements of the flame speed are usually not possible without optical data. The determination of the flame speed by in-cylinder pressure indication continues to improve but still relies on models developed for hydrocarbon fuels; these have to be reassessed for NH<sub>3</sub>-based combustion [12].

### 3.4 Development of Ammonia Combustion Models

The combination of experimental investigations at different complexity levels and their associated numerical simulations builds the foundation for the development of high-fidelity turbulent combustion modeling approaches. The fundamental research on canonical flames along with direct numerical simulation (DNS) provides detailed insight into the underlying flame dynamics of NH<sub>3</sub> and NH<sub>3</sub>/H<sub>2</sub> flames. With a focus on ICE applications, investigations under the controlled turbulent conditions of a fan-stirred CVCC make it possible to mimic relevant combustion conditions characterized by the apparent flame regimes; however, pressure and temperature levels differ from engine conditions. Although limited to a very low number of flow conditions, these measurements in combination with DNS not only allow valuable investigations of flame propagation but also facilitate the evaluation of the flame regime

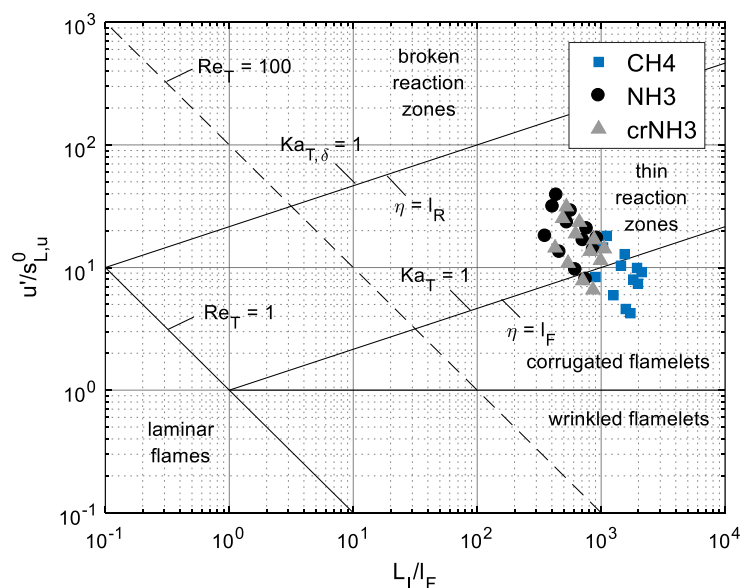


Figure 6: Borghi-Peters diagram of premixed turbulent combustion for measurement points at engine conditions with CH<sub>4</sub>/air, NH<sub>3</sub>/air, and partially cracked NH<sub>3</sub>/air mixtures; reprinted from [12].

assumptions. The knowledge of the apparent flame regime is a prerequisite for numerical simulation of turbulent combustion since the choice of the combustion modeling approach strictly depends on the flame conditions.

While conventional ICE operated with carbon-based fuels mainly exhibit flames assigned to wrinkled or corrugated flamelets (one exception being TJI due to high turbulence levels or highly diluted mixtures), the low reactivity of  $\text{NH}_3$ , which leads to high Karlovitz numbers, shifts the flame regime towards the broken reaction zone in the regime diagram (cf. Figure 6). This effect is even intensified if a high turbulence combustion concept such as TJI is applied. Yet since the transitions between the regimes are gradual rather than abrupt, the use of optical measurements along with DNS helps to identify the actual flame regime and builds the foundation for the development of turbulence combustion models appropriate for  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$  applications.

#### Non-unity Lewis Number and Preferential Diffusion

While most carbon-based fuels exhibit unity Lewis number behavior under engine-like conditions, the combustion of  $\text{NH}_3$  and especially  $\text{NH}_3/\text{H}_2$  mixtures are characterized by thermodiffusive instabilities due to their pronounced mass diffusivity. These thermodiffusive instabilities significantly increase the flame speed and thus the flame consumption speed [34]. Furthermore, thermodiffusive instabilities lead to higher flame stretches in turbulent flames than in laminar ones, influencing early flame kernel development in the ICE [35]. Since the theoretical formalism of several turbulent combustion models such as the coherent flame model (CFM) assumes a unity Lewis number, comprehensive model adaptations are required so they may be used for fuels with a non-unity Lewis number. Geometry-based models such as the G-equation model rely on the determination of turbulent flame speed, thus accounting for thermodiffusive instabilities. Flamelet-based models such as the flamelet generated manifold (FGM) approach usually assume a unity Lewis number and equal diffusivity of different species in flamelet precalculation. Yet several studies have shown that the consideration of preferential diffusion and flame stretch effects in the flamelet formulation – essential for non-unity Lewis number flames – increases numerical simulation accuracy in terms of flame propagation and structure [36–38].

High-quality optical measurements of turbulent flames provide valuable insight into the flame front, which usually takes on a “finger-shaped” form enhanced by thermodiffusive instability. These

measurements are thus one of the main sources of knowledge and data for developing and validating advanced turbulent combustion models.

#### High Karlovitz Number

As previously mentioned, the use of low reactive fuels such as  $\text{NH}_3$  leads to a shift in the apparent flame regime towards the broken reaction zone. While flames at moderate Karlovitz numbers are characterized by short reaction time scales that result in laminar structures in the flame front known as “flamelets,” the increased impact of turbulent motion on the preheat and reaction zones due to the larger reaction time scales of low reactive fuels means there is an absence of a laminar flame structure. Since the use of flamelet-based turbulent combustion models is usually less computationally demanding than modeling approaches that do not rely on any flamelet assumptions such as the probability density functions (PDF) model, knowledge of the validity range of flamelet-based models is required. While DNS is limited in terms of resolving high turbulence levels due to computational costs, the use of optical measurements can shed light on the validity range of flamelet-based models. By analyzing the flame front, qualitative statements on the presence of flamelet structures can be made that are vital to the choice and validation of the modeling approach. Another cause of high Karlovitz numbers is turbulent jet ignition, which might be a prerequisite for using  $\text{NH}_3$  in large engines. The high levels of turbulence further increase the Karlovitz number values, leading to the previously mentioned effects. As combustion progresses, however, the Karlovitz number decreases, probably shifting the flame towards flamelet-like regimes requiring multi-regime combustion models which show accurate behavior over a wide range of flame regimes. In this context, the combination of advanced optical measurements and high-fidelity numerical simulations will also allow the development of enhanced turbulent combustion models for accurate simulation of  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$ -fueled ICE.

#### Synergies of Experimental-Numerical Methodology

The methodology introduced here relies on the deep interconnection between experimental optical efforts and numerical simulations. Simplified experimental configurations facilitate the creation of fundamental simulations for model development, whereas conditions that closely resemble engine environments require significant computational effort, necessitating the use of more cost-effective simulations. For each level of complexity in modeling  $\text{NH}_3$  combustion—from high-fidelity DNS to highly efficient Reynolds-averaged Navier-Stokes (RANS) simulations—there exists an

appropriate experiment that matches the boundary conditions and provides valuable validation data, see Figure 7.

This integration of experimental and numerical efforts, ranging from fundamental to engine-like conditions, fosters positive synergies that enhance the understanding of relevant mechanisms and facilitate the development of effective models for  $\text{NH}_3/\text{H}_2$  simulation.

#### 4 CONCLUSIONS AND OUTLOOK

Based on the results presented in this publication, a consistent experimental methodology for investigating the unique combustion behavior of  $\text{NH}_3$ -based flames is currently being implemented at the LEC and FHNW. This should lead to a deeper understanding of the phenomena governing  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$  combustion by isolating the relevant effects and provide a comprehensive database for simulation model enhancement and development. To this end, existing optical test rigs are utilized and new optical test rigs are set up to measure and visualize laminar burning velocity, turbulent burning velocity, and turbulent jet ignition under ideal conditions; as well as turbulent flame propagation under engine-like conditions.

##### Conclusions

Optical results at quiescent, close-to-ambient conditions highlight the relevance of thermodiffusive instabilities for  $\text{NH}_3$  and  $\text{NH}_3/\text{H}_2$  flames. Optical results from an optical engine test rig emphasize instabilities and strong turbulence-flame interaction. The comparison of TJI of  $\text{CH}_4/\text{air}$  and  $\text{H}_2/\text{air}$  mixtures shows the importance of the fuel properties on the quenching and re-ignition propensity of the fuel-air mixture in the MC. These effects are very important for understanding and modeling TJI processes in premixed  $\text{NH}_3/\text{air}$  mixtures.

The insights into  $\text{NH}_3$  combustion obtained from optical data are manifold. While LBV and TBV can be quantified by direct measurement, further insight

into the phenomena driving the flame propagation is possible. The combustion of  $\text{NH}_3$  is temperature-driven, i.e., it requires high temperatures for high reactivity. In addition, the strong turbulence-flame interaction is exposed by the characterization of the combustion regime. The qualitative difference in the flame front of  $\text{NH}_3$  and  $\text{CH}_4$  is apparent in optical data at engine-like conditions. Determination of the combustion regimes helps ensure the transferability of fundamental insights to engine conditions. Moreover, the optical data exposes the Lewis number effect with lean fuel/air mixtures and its significance under engine-like conditions.

The optical investigations at different complexity levels are combined with associated numerical simulations to develop high-fidelity turbulent combustion modeling approaches. As turbulent combustion models have been the focus of optimization for carbon-based fuels in the past, a thorough reassessment is required. In particular, non-unity Lewis number effects and preferential diffusion as well as high Karlovitz numbers determine  $\text{NH}_3$ -based combustion. These effects are not commonly covered by present combustion models.

##### Outlook

In future investigations that apply the methodology presented in this paper, the remaining test rigs will be set up for  $\text{NH}_3$  combustion and optical investigations will be performed that go far beyond the results presented here. The available optical measurement techniques may be extended by spectroscopic and tomographic investigations. Spectroscopy captures the relative intensity of all emissive species while tomography yields highly resolved images of the flame contour. Both will provide insight into turbulent combustion of  $\text{NH}_3$ -based fuels beyond the current level of knowledge. For example, spectroscopic investigations of the qualitative intensities of  $\text{OH}^*$  and  $\text{NH}^*$  chemiluminescence can give insight into the reaction pathways of  $\text{NH}_3$  oxidation and serve as a valuable input for reaction mechanism validation.

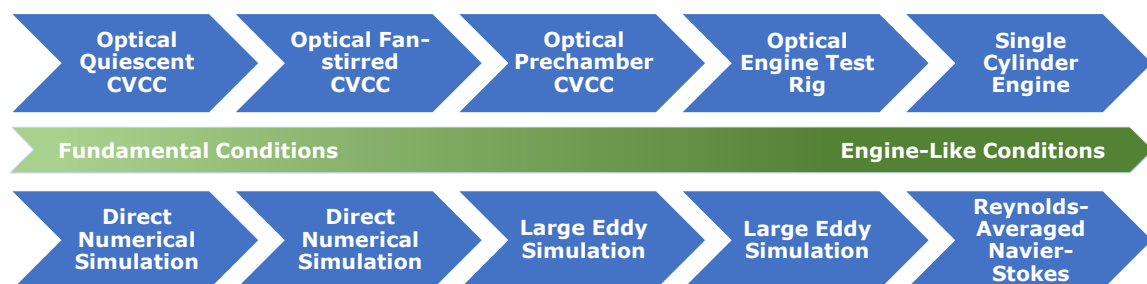


Figure 7: Corresponding investigative stages in experiments and simulations of turbulent premixed combustion

The high-quality optical measurements will be accompanied by DNS or LES for each of the test rigs. This will enable the investigation of effects undetectable in experiments and facilitate the precise modeling of all phenomena governing NH<sub>3</sub> combustion.

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