PhD thesis title

Optical diagnostics for carbonaceous solid fuels and flame retarded polymers in laminar and turbulent flows

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Brief summary

Combustion and fires involving solid materials are widespread in both engineering and everyday life. While being beneficial for clean energy production from biomass, they are hazardous in fires involving polymer-based products. Despite these different manifestations, the fundamental description of material, chemical reaction, and airflow interactions are similar. Previous experimental methods were too coarse to resolve these interactions at the relevant scales. In my dissertation, I developed state-of-the-art experimental methods to study solid material combustion using high-resolution optical diagnostics, including laser-based techniques. These methods allowed to observe combustion processes at the single particle level without disturbing them, providing critical insights in three areas:

First, by using a novel laser system to track the movement of individual biomass particles at 200,000 frames per second, my study achieved the highest combined spatial and temporal resolution of simultaneous particle and gas velocity measurements to date, revealing how differently shaped biomass particles such as grasses and walnut shells interact with a turbulent airflow. The results of these particle-turbulence interaction studies are critical to understanding the stabilization of biomass flames in power plants to decarbonize the energy sector.

Second, previous research has focused on the combustion of individual particles, but in power plants, millions of particles are injected into burners where they interact during combustion. Using advanced high-speed laser and camera systems, I tracked individual particles in these clouds while capturing the surrounding three-dimensional flame and found that as the number of particles increases, the individual particle flames merge into a single structure with a non-flammable core. This finding is critical for predicting and preventing unwanted reductions in reactivity of power plant burners and increasing their reliability as we transition from coal to sustainable biomass.

Third, I adapted and transferred optical methods used in solid fuel combustion to characterize flame retardant polymers under real burning conditions. This novel approach visualized selected chemical species during polymer combustion for the first time, providing a better understanding of flame retardant effectiveness and improving fire safety for critical applications such as insulation materials and electrical equipment.

Overall, the research contained in my dissertation demonstrates how cutting-edge optical diagnostics improves our understanding of solid-flow-chemistry interactions, leading to clean energy production and improved fire safety.

Scientific summary

The motivation for the research conducted in my dissertation is rooted in two critical challenges: mitigating anthropogenic climate change through innovative combustion technologies with carbon capture and storage, such as BECCS (Bioenergy with Carbon Capture and Storage) and improving fire safety in polymers through effective flame retardants. Both the combustion of flame-retarded polymers and the thermochemical conversion of pulverized solid fuels such as coal or biomass share phenomenological similarities at the meso- and microscale, where reactive solid particles interact with gaseous flows. Understanding these solid-flow-chemistry interactions is critical for accurately predicting and controlling thermochemical conversions in complex systems. To better illustrate selected aspects of solid combustion, Figure 1 shows the interaction triangle of the solid, the chemical reactions, and the surrounding flow field. Selected aspects of the solid phase include factors such as particle size, shape, and number density, which can vary significantly when transitioning from spherical coal particles to aspherical biomasses such as miscanthus. The flow field can be either laminar or turbulent, affecting heat and mass transfer at the gas-solid interface and the mixing of pyrolysis products with the surrounding oxidant. The presence of flame retardants adds another layer of complexity by altering chemical reactions and inhibiting combustion. By isolating and selectively studying these interactions, my research aimed to develop a more accurate understanding of each aspect, which is essential for predicting and controlling the thermochemical conversion of solid materials in practical applications.

Based on the current state of research, I identified three key areas investigated in my thesis: the interaction of aspherical biomass particles with turbulent flow, the effects of particle number density in solid fuel combustion, and the assessment of flame retardant effectiveness in polymers. In the following, I will share some highlights of the achievements I made employing minimally intrusive optical experimental methods to explore these phenomena with unprecedented detail. During my dissertation and based on the works resulting from it, I have published 16 peer-reviewed journal papers and 5 conference papers, including a review paper on optical diagnostics for particle-resolved solid fuel diagnostics.

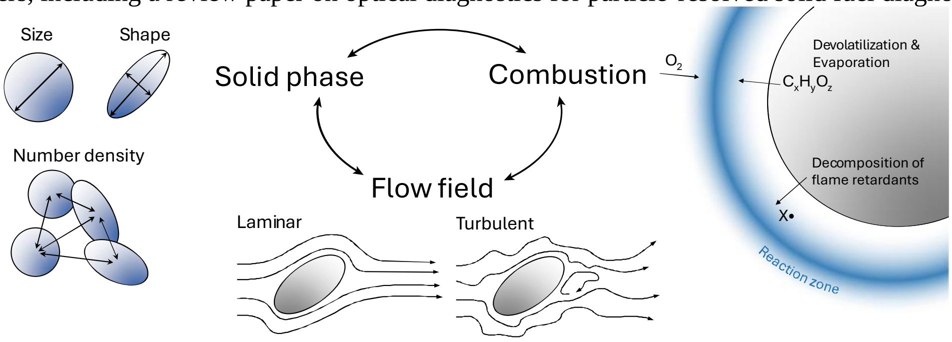

Figure 1. Interaction triangle of solid phase, combustion, and flow field present in solid material combustion phenomena.

Ultra-high repetition rate diagnostics of biomass-laden turbulent jets

In the first part of my thesis, I investigated the interaction between solid biomass fuel particles and the surrounding turbulent flow field in a non-reactive round jet. To achieve the necessary spatial and temporal resolutions, a pulsed fiber laser system was adapted and integrated into a time-resolved particle image velocimetry (TR-PIV) system and was initially used to characterize a single-phase turbulent round jet. This setup allowed for unprecedented temporal dynamic ranges and provided valuable insights into turbulent scales, spectra, and space-time correlations at repetition rates exceeding 500 kHz. In particular, the study identified deviations from Taylor's frozen flow hypothesis, revealing critical aspects of turbulent shear flows and underlining the need for simultaneous temporal and spatial resolution of turbulent flows for their thorough characterization [1]. Highlighting the significance of this work, the study was selected by the editors of Experiments in Fluids for presentation at their webinar series.
Based on the ultra-high repetition rate of the fiber laser system, a single cavity laser suitable for both dual-pulse and time-resolved PIV measurements was developed, combining the strengths of time-resolved PIV with high temporal resolution and dynamic range and conventional dual-pulse PIV with high spatial resolution and dynamic range. By incorporating acousto-optic and electro-optic pulse picking systems, single pulses could be selectively directed into the measurement volume, resulting in an extremely flexible laser system for multi-phase velocimetry. This improved diagnostic setup was then applied to study the particle-turbulence interactions of single Miscanthus particles in a turbulent jet as highlighted in Figure 2(a) [2]. Using a multi-parameter measurement system consisting of a two-phase velocimetry system and simultaneous diffuse back-illumination (DBI) for particle sizing, I was able to observe and analyze the interactions of differently shaped biomass particles and their surrounding turbulent flow field at a repetition rate of 200 kHz and a pixel resolution of 10 µm, as highlighted in Figure 2(a) for a representative aspherical biomass particle. Because particle size and relative velocity were measured simultaneously, the particle Reynolds number could be measured in a time-resolved manner, revealing how the flow around an individual particle can detach and modify the surrounding turbulent flow field, allowing a particle-dependent analysis of turbulence modulation in particle-laden flows.

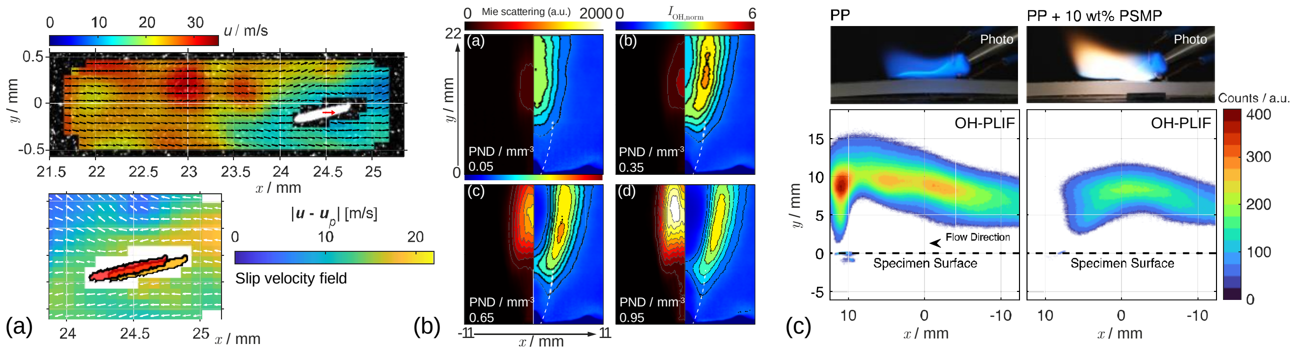

Figure 2. (a) Two-phase velocimetry of an aspherical biomass particle including the computed slip velocity field. (b) Effect of increasing PND on particle group combustion for a fixed atmosphere containing 20 vol% of oxygen. (c) Planar OH-PLIF of a polypropylene (PP) slab interacting with an external premixed methane flame. For reference, photos showing the combustion luminosity are shown.

Group combustion of solid fuels in laminar oxy-fuel environments

The second part of my thesis focuses on the effect of particle number density (PND) on the combustion behavior of coal particles within a laminar flat flame burner. By varying the surrounding atmosphere between air and CO_2-rich oxy-fuel conditions, the study aimed to understand how these environments affect pulverized fuel flames, considering the higher heat capacity of CO_2 and the lower diffusivity of oxygen in CO_2. Multi-parameter three-dimensional optical diagnostics were used to derive conditioned statistical data and to evaluate the measurement errors associated with particle number density using a newly developed imaging simulation tool as reported in [3].

The primary objective was to investigate the transition from single particle to particle group combustion and to analyze the combined effects of PND, gas-phase oxygen content, and diluent composition. Using comprehensive diagnostic techniques such as DBI, scanning OH planar laser-induced fluorescence (OH-PLIF), and Mie scattering, volatile flame structures and soot formation were observed under various boundary conditions. The results showed that increasing the oxygen content in the gas phase effectively suppresses soot formation, especially at high PND. In addition, it was found that an increase in PND and a decrease in gas-phase oxygen concentration led to non-flammability in the fuel-rich core of the flame, as highlighted in Figure 2(b). The reason for this is that as the number of particles increases, they draw more heat from their environment, lowering the gas temperature and creating competition for oxygen in the dense particle cloud. These trends were consistent in both air and oxy-fuel environments, providing critical insight into optimizing combustion processes for retrofitting existing power plants and designing future systems. This work represents a significant advance in understanding the complex interplay between particle density and combustion behavior in different atmospheric conditions.

Optical diagnostics of flame retarded polymers

The third part of my thesis investigated combustion inhibition in polymers, specifically the effect of flame retardants on polypropylene using advanced optical diagnostics. By adapting diagnostic methods originally developed for solid fuel combustion, I was able to apply OH-PLIF to study the combustion inhibition behavior of flame-retarded polypropylene (PP) particles in laminar environments. This was a significant achievement as it demonstrated the potential of OH-PLIF to identify the gas-phase activity of flame retardants [4].
Building on this foundation, the optical diagnostic setup was expanded to include DBI for simultaneous combustion and particle size measurement. This multi-parameter measurement system allowed the determination of the normalized OH-LIF intensity and the dimensionless stand-off distance of diffusion flames surrounding PP particles treated with four different flame retardants, each with distinct modes of action. The study revealed a reduction in OH-LIF signal intensity within the flame for gas-phase active flame retardants, confirming the effectiveness of using laser-induced fluorescence for flame inhibition studies [5].
Additionally, the methodology was extended to larger polymer samples by studying the interaction of rod-shaped specimens with a premixed methane Bunsen flame as illustrated in Figure 2(c). This demonstrated the applicability of optical diagnostics in studying polymer combustion and flame inhibition in realistic fire scenarios. Combining optical diagnostics with established pyrolysis fragment analysis methods provided a comprehensive understanding of the chemical and combustion dynamics of flame-retardant polymers, highlighting the versatility and importance of this experimental approach to future studies and the development of tailor-made flame inhibitor formulations.

Conclusion

In summary, my dissertation demonstrates significant advances in the field of optical diagnostics for the study of solid material combustion and its inhibition. By developing and applying state-of-the-art, high-resolution, minimally intrusive optical techniques, I have provided critical insights into the interactions between solid particles, chemical reactions, and the surrounding flow field in both laminar and turbulent environments. These findings improve our understanding of solid fuel combustion processes, aiding in the development of cleaner energy production methods, and improve the effectiveness of flame retardants in polymers, contributing to greater fire safety.
Beyond my thesis work, I have continued to advance these methods by using novel data processing algorithms, such as optical flow methods, to generate high-density velocity field data. This advancement overcomes the limitations of particle image velocimetry (PIV), which relies on interrogation windows that limit spatial resolution, and allows for more comprehensive analysis and visualization of flow dynamics. Overall, the innovative methods and comprehensive data generated by this research pave the way for future studies and the development of advanced technologies to address pressing environmental and fire safety challenges. I remain committed to further improving and applying these methods to various aspects of fluid mechanics and combustion science for energy conversion and fire safety.

Selected publications

[1] Geschwindner C, Westrup K, Dreizler A, Böhm B. Ultra-high-speed time-resolved PIV of turbulent flows using a continuously pulsing fiber laser. Exp. Fluids 2022;63(4).
[2] Geschwindner C, Westrup K, Dreizler A, Böhm B. Pulse picking of a fiber laser enables velocimetry of biomass-laden jets at low and ultra-high repetition rates. Proc. Combust. Inst. 2023;39(1):1325–35.
[3] Li T, Geschwindner C, Dreizler A, Böhm B. An experimental study of coal particle group combustion in conventional and oxy-fuel atmospheres using multi-parameter optical diagnostics. Proc. Combust. Inst. 2023;39(3):3259–69.
[4] Geschwindner C, Goedderz D, Li T, Köser J, Fasel C, Riedel R et al. Investigation of flame retarded polypropylene by high-speed planar laser-induced fluorescence of OH radicals combined with a thermal decomposition analysis. Exp. Fluids 2020;61(2).
[5] Geschwindner C, Goedderz D, Li T, Bender J, Böhm B, Dreizler A. The effects of various flame retardants on the combustion of polypropylene: Combining optical diagnostics and pyrolysis fragment analysis. Polymer Degradation and Stability 2023;211:110321.