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Quantitative laser diagnostics of gas-phase potassium species in biomass combustion and gasification

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Abstract
Thermochemical energy conversion processes, such as combustion and gasification, are applied worldwide for generation of electricity, heat and synthesis of chemicals. Today, these processes are mostly run on non-renewable, fossil fuels and constitute a major source of carbon dioxide emissions. A promising renewable energy source with low net carbon dioxide emissions is biomass, in particular rest products from agriculture and forestry. However, biomass usually contains high amounts of volatile inorganic compounds, such as chlorine, potassium (K) and phosphorus (P), which lead to ash-related operational issues, including deposit build-up, slagging and corrosion. Therefore, efficient utilization of biomass requires knowledge of the chemistry and fate of the inorganic compounds during thermochemical conversion. Due to the reactive, high-temperature environments in those processes, gaseous compounds are preferably measured in situ using optical techniques.

This thesis mainly deals with the development of a laser-based technique for simultaneous in situ detection and quantification of the main gaseous K species in biomass combustion and gasification: atomic K, potassium hydroxide (KOH) and potassium chloride (KCl). The novel method combines photofragmentation (PF) with tunable diode laser absorption spectroscopy (TDLAS) and achieves sub-ppm detection limits for all three K species for a path length of 2 cm and a time resolution of 20 ms. Recording the ns-µs PF signal decay due to fragment recombination allows probing the K reaction kinetics. Together with TDLAS sensors for water, methane and gas temperature, the PF-TDLAS system was employed to characterize biomass reactors from laboratory- to pilot-scale. The results were compared to predictions by numerical models. In addition, PF-TDLAS was employed for quantitative wide-field imaging of K species in a laboratory flame during KCl salt and biomass conversion. Finally, in situ detection of phosphorus pentoxide (P_4O_{10}) with a time resolution of 140 ms was demonstrated using broadband infrared absorption spectroscopy. Absorption line strengths of P_4O_{10} at temperatures relevant for combustion were determined for the first time. The techniques presented in this thesis can provide unique experimental data for validation and further development of numerical models and advance the understanding of K species chemistry during solid fuel conversion, which is needed to facilitate the utilization of biomass in the energy system.

Keywords
Thermochemical conversion, pyrolysis, phosphorus, single pellet, entrained-flow, in situ, spectroscopy, photofragmentation, imaging, numerical modelling.

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