

Applications of Synchrotron-based VUV Photoionization Mass Spectrometry in Combustion

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Combustion is one of the earliest technologies handled by human beings and one that remains our primary source of energy, yet it embodies a complex suite of physical and chemical processes that are inadequately understood. Combustion chemistry includes both chemical thermodynamics and chemical kinetics. Experimental achievements in combustion chemistry mainly depend on the development of combustion diagnostics and serve as the foundation of theoretical progress in this area. The major objective of combustion diagnostics is to present comprehensive product identification and detailed concentration information of flame species, which can be used to develop kinetic models for simulation of practical combustion. However, conventional combustion diagnostic methods are challenged in distinguishing isomeric species, detecting reactive radicals, etc. Therefore to achieve deep insight into combustion chemistry, a diagnostic method with high detection sensitivity, isomeric selectivity and radical detectability is required.

This presentation reports recent applications of synchrotron-based vacuum ultraviolet photoionization mass spectrometry (SVUV-PIMS) in various research topics of combustion chemistry [1-2]. The wide tunability of synchrotron photon energy can facilitate the selective identification of isomeric intermediates and near-threshold detection of radicals to avoid fragmentation interference [3]; and the convenient combination of SVUV-PIMS with various laboratory-based combustion approaches demonstrates its universality in combustion studies. Recent experimental achievements have demonstrated the successful applications of this technique in premixed flame [3], oxidation in jet-stirred reactor [4], pyrolysis in flow reactor, coflow diffusion flames, catalytic oxidation, and analysis of polycyclic aromatic hydrocarbons (PAHs) and soot. Also we will provide some new results using the upgraded instruments located at Hefei Light Source (HLS) and Taiwan Light Source (TLS) (Fig. 1). Benefiting from the new design with short distance between the sampling position and the ionization region, a lot of “new” reactive species can be unambiguously detected, i.e., H, O, OH, etc.

In all applications, combustion intermediates including isomers and radicals can be distinguished unambiguously, which extends our knowledge of intermediate pools and provide more precise targets for quantum chemical calculations of significant reaction channels. The observed mass range covers both small combustion products and large ones such as PAHs with two to five carbonic rings, which present clues to understand molecular growth process from fuel to PAHs and consequently soot in fuel-rich hydrocarbon flames. Furthermore, quantitative analyses of chemical structure are available in most applications, e.g. concentration profiles of flame species versus position in premixed flames, or versus temperature in pyrolysis and oxidation, with objectives to validate current kinetic models and develop new kinetic models [5].

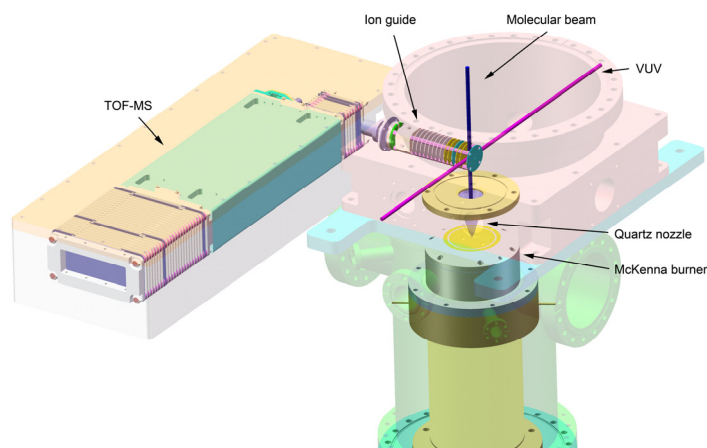


Fig. 1 Scheme of newly designed premixed flame apparatus using SVUV-PIMS.

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