Influence of zeolite topology in Catalytic Fast Pyrolysis of Biomass: a Kerr-gated Raman study using model compounds

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Biomass offers a route to obtain chemical products normally derived from crude oil, from a more sustainable and carbon neutral source (since biomass absorbs CO_2 for photosynthesis). The oxygen content of biomass, however, poses some problems, causing bio-oils to be more acidic and unstable than their crude oil derived counterparts. One method for the removal of oxygenates is through Catalytic Fast Pyrolysis of biomass (CFP), where biomass is pyrolyzed using fast heating rates and the resulting vapours are upgraded over zeolite catalysts.

Mechanistic studies are few in this area but some point towards the idea of a hydrocarbon pool mechanism, where hydrocarbons build up in zeolite pores to react with further pyrolysis vapours, undergoing decarbonylation, decarboxylation and dehydrogenation to produce aromatic species, olefins, CO and CO_2, as well as undesirable coke which builds up. During this upgrading process, zeolites deactivate rapidly through coking, making frequent regeneration necessary and affecting process efficiency. Through understanding these chemical transformations, we can gain insight as to how reaction efficiency can be improved.

Raman Spectroscopy is a useful tool for mechanistic studies, but in many cases catalyst defects or emissive hydrocarbon species present can cause intense fluorescence, preventing Raman signals from being detected. To avoid fluorescence, UV or near-IR probe wavelengths can be used but these often result in sample damage and low signal intensity respectively. This work uses a visible wavelength source (400 nm) with a Kerr-gated spectrometer that allows Raman signals to be separated from fluorescence due to their different lifetimes.

In this work, we study the interaction of oxygenated hydrocarbons with zeolites by operando Kerr-gated Raman Spectroscopy and identify reaction intermediates whilst measuring catalytic activity through mass spectrometry (MS).

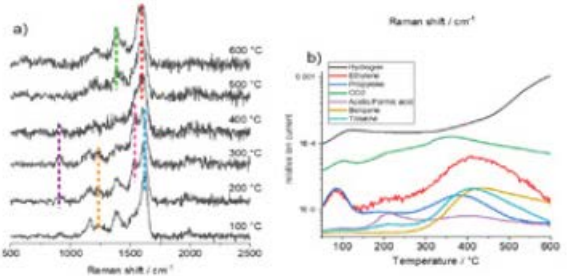

Figure a) Raman spectra collected during temperature ramp experiment of furan on H-ZSM-5 Si/Al 40 b) MS data simultaneously measured

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