Molecular Insights and Origins of Internal Combustion Engine Deposits via OrbiSIMS

David Scurr

University of Nottingham

Abstract Content:

Instrument development in secondary ion mass spectrometry (SIMS) continues to yield more and more complex hyperspectral data-sets. These data-sets enable significant advancements in applications and research opportunities, however, they also possess an associated series of challenges to interpret them and unlock their full potential. This is exemplified in the introduction of Orbitrap SIMS (OrbiSIMS), where typical data-sets exhibit a 100 - 1000 fold increase in the number of secondary ions observed for any given sample as compared to comparable time of flight (ToF) data. As a materials analysis capability this has significantly impacted a multidisciplinary scope of academic and commercially focussed research areas from biology (e.g. in molecular analysis [1], cells [2] and tissues [3]) to engineering (e.g. 3D printing [4] and fuel deposit analysis [5]).

Carbonaceous fuel deposits are ubiquitous, being formed on surfaces in engines, fuel systems and on catalystsoperating at high temperatures for hydrocarbon transformations. In internal combustion engines, theirformation negatively affects worldwide vehicle emissions and fuel economy. Deposit composition and formation pathways are poorly understood due totheir insolubility and the intrinsic complexity of their layered carbonaceous matrix. Here, we apply OrbiSIMS to undersatand deposit chemistry and evidence common molecular distributions in deposit depth and in positions relative to the combustion chamber. We observe the productsof the growth of both planar and curved polycyclic aromatic hydrocarbons to form small fullerenes overtime in the engine and propose possible formation pathways which explain the molecular distributionsobserved. For the first time, OrbiSIMSresults were compared and validated with data from atmospheric pressure matrix assisted laser desorptionionization MS. The comprehensive characterization provided will help the development of a new generation of chemical additives to reduce deposits, and thus improve vehicle emissions and global air quality.

Reference:

[1]. Kotowska et. al., Nature Comms, 2020

[2]. Suvannapruk et. al., Anal. Chem, 2022

[3]. Starr et. al., PNAS, 2022

[4]. Trindade et. al., Commun. Mater., 2021

[5]. Edney et. al., ACS Appl. Mater. Interfaces., 2020

[6]. Edney et. al., Anal. Chem., 2022

Key words: OrbiSIMS, fuel deposits, polycyclic aromatic hydrocarbons