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PO Box 117 221 00 Lund +46 46-222 00 00

Relating refractory soot mass spectra with nanostructure and combustion conditions

V.B. Malmborg¹, A. Eriksson^{1,2}, S. Török³, K. Kling⁴, M. Novakovic⁵, S. Shamun⁵, M. Shen⁵, M. Tuner⁵, P-E. Bengtsson³, and J. Pagels¹

¹Division of Ergonomics and Aerosol Technology, Lund University, Box 118, SE-22100, Lund, Sweden

²Division of Nuclear Physics, Lund University, Box 118, SE-22100, Lund, Sweden

³Department of Combustion Physics, Lund University, Box 118, SE-22100, Lund, Sweden

⁴National Research Centre for the Working Environment, 2100 Copenhagen, Denmark

⁵Division of Combustion Engines, Lund University, Box 118, SE-221 00, Lund, Sweden

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Presenting author email: Vilhelm.malmborg@design.lth.se

The soot particle aerosol mass spectrometer (SP-AMS) can detect infrared-light absorbing refractory aerosol particles. Vaporization occurs when a focused beam of particles enters a cavity ring down laser (Nd:YAG 1064nm) and the two beams overlap. Refractory mass spectra of soot have previously been used mostly to characterise the ratio of organic to refractory black carbon in soot particles. However, recent studies highlight the information contained in the carbon cluster (C_x^+) distribution of soot particle refractory mass spectra (Malmborg et al., 2017; Onasch et al., 2015).

Soot mass spectra show strikingly different C_x^+ distributions depending on the source and combustion condition. Soot in fuel-rich flames at high temperatures and with well-organized nanostructures, typically traditional diesel combustion soot, have mass spectra completely dominated by a few carbon cluster ions with low carbon number ($C_1^+-C_5^+$) of which C_3^+ has the strongest signal. On the other hand, soot with less organized nanostructure, for example soot from advanced diesel combustion, show a wider distribution of carbon clusters. Soot from these latter types of combustion processes have elevated signals at $C_{10}^+-C_{15}^+$ and $C_{30}^+-C_{70}^+$. The highest signal intensities from carbon clusters with large carbon numbers (i.e., $C_{30}^+-C_{70}^+$) are found at carbon numbers of stable fullerenes (e.g., C_{36} , C_{44} , C_{50} , C_{60} , C_{70})

Refractory mass spectra of soot produced in a large number of different combustion processes were collected with the SP-AMS (Aerodyne Research Inc.). The soot nanostructure was analysed using high-resolution transmission electron microscopy (HR-TEM) for a selection of the soot samples. A thermodenuder (Aerodyne Research Inc.) was used, for a subset of the soot samples, to evaporate semi-volatile components.

The C_x^+ distribution of mini-CAST (Jing Ltd.) OP1 soot with well-organized nanostructures and long lamellas was confined to low carbon numbers. Such distributions were also present for soot from conventional diesel and biodiesel (rape seed methyl esters) combustion in a heavy duty diesel engine without exhaust gas recirculation (EGR). With increased nitrogen flow and decreased oxidation airflow, the C_x^+ distribution of mini-CAST OP5, OP6 and OP7 soot had increasing signal from large carbon clusters. The mini-CAST soot at OP5-OP7 have decreasing degree of ordered nanostructures and much shorter lamellas than OP1 soot. Similar types of C_x^+ distributions were found in low temperature diesel combustion at very high EGR (Figure 1).

Fuel composition (i.e., diesel vs biodiesel) influenced the soot nanostructure. However, The C_x^+ distribution of soot mass spectra from the two fuels was similar, although combustion with conventional diesel had higher fullerene signal. This could indicate that combustion temperatures (which are reduced by EGR) are more important in determining the soot properties that will form large carbon clusters, than fuel composition.

A first analysis between the tortuosity and fullerene signal did not show a correlation. However, it should be noted that short lamellas and highly disordered nanostructure may influence the determination of the tortuosity. Our analysis shows that a higher degree of disorder and shorter lamellas are associated with larger carbon clusters and a wider C_x^+ distribution. With the large set of refractory mass spectra collected from different soot, our aim is to determine the relationship between soot nanostructure and refractory mass spectra, and the combustion conditions that form the various soot properties.

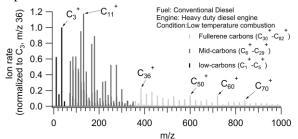


Figure 1. SP-AMS refractory mass spectrum of soot from low temperature diesel combustion. The spectrum has been normalized to the C_3^+ intensity.

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