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Using Raman Microscopy to Study the Structure of Airplane Soot and Surrogates

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Air traffic is one of the main sources of man's impact on the atmosphere. Emission of soot particles by airplanes has a significant influence on the climate. Soot particles affect climate directly by the formation of longlived contrails and indirectly by acting as nuclei for cirrus clouds formation. There are some experiments focused on ice nucleation on soot, yet no definitive conclusion has been reached. Most likely one of the reasons behind these discrepancies resides in the different physico-chemical properties of soot particles produced in different conditions, e.g. with respect to fuel or combustion techniques.

The aim of our work is to investigate the influence of the combustion conditions on the structure of generated soot. We have used Raman microscopy (514 and 785 nm excitation) to study the spectral parameters of the first-order Raman band of various soot samples, collected from three different sources in the frame of the MERMOSE project (<u>http://mermose.onera.fr/</u>): PowerJet SaM-146 turbofan (four engine regimes), CAST generator (propane fuel, four different global equivalence ratios), and Kerosene laboratory flame. Figure 1 shows typical Raman spectra recorded for some of these samples.

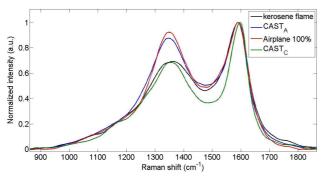


Figure 1. Raman spectra of different soot particles obtained with λ_0 =514 nm excitation.

To analyze the spectra a de-convolution is performed using the approach described by Sadezky *et al.* (2005). For each sample we calculated the contributions from five different bands: G band, corresponding to ideal graphite lattice (E_{2g} symmetry), D₁ band corresponding to a disordered graphite lattice (contribution from the edges, A_{1g} symmetry), D₂ band corresponding to a disordered graphite lattice (contribution from the surface, E_{2g} symmetry), D_3 band corresponding to amorphous carbon, and D_4 band corresponding to disordered graphite lattice (A_{1g} symmetry), polyenes or ionic impurities. Table 1 summarizes these results.

Table 1. Spectra de-convolution. Band positions (cm⁻¹), integrated areas and peak area ratios relative to G band.

Band	Parameter	CAST _A	CAST _C	Kerosene	SaM-146
G	Position	1585	1585	1585	1580
	Area	4.3×10^{5}	2.3×10^{6}	1.1×10^{6}	2.0×10^5
D ₁	Position	1345	1350	1350	1350
	Area	9.9×10^5	2.5×10^{6}	7.8×10^5	3.5×10^5
	A_d/A_g	2.3	1.1	0.71	1.7
D ₂	Position	1608	1608	1608	1600
	Area	1.1×10^{5}	1.1×10^{6}	1.2×10^{5}	1.2×10^{5}
	A_d/A_g	0.25	0.47	0.11	0.57
D ₃	Position	1520	1510	1500	1505
	Area	1.5×10^{6}	5.8×10^{6}	2.1×10^{6}	4.7×10^5
	A_d/A_g	3.5	2.5	1.9	2.3
D ₄	Position	1230	1280	1280	1280
	Area	8.6x10 ⁵	5.3×10^{6}	1.8×10^{6}	3.2×10^5
	A_d/A_g	2.0	2.25	1.65	1.55

CAST_A (equivalence ratio ~1) and airplane soot samples exhibit similar Raman spectra. Both present an important contribution of D_1 and D_3 bands. On the other hand the contribution of D_2 band is larger in the case of airplane soot. This might indicate that the size of the graphite crystallites is smaller in the case of airplane soot. Moreover, similarity of structure can promote CAST soot as a good candidate for airplane soot surrogate, though a comparison of other physico-chemical properties, like surface chemical composition, should be performed as well to confirm this suitability. Influence of the structural and physico-chemical properties of soot particles on their potential to act as ice nuclei will be investigated in the near future.

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