Properties of graphene-like materials derived from fuel-rich flames of diverse hydrocarbons

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Abstract

Soot constitutes the carbon particulate matter, in form of particles and aggregates, that can be produced in combustion by a step-wise mechanism starting from organic precursors, mainly hydrocarbons, and passing through polycyclic aromatic hydrocarbons (PAH) formed in high-temperature partially oxidative (fuel-rich) conditions. Combustion-formed PAH from low- to high-mass are assumed as the fundamental molecules to which graphene is extrapolated upon the decrease to molecular size, whereas soot structure is mainly based on two-dimensional graphene layers grouped, cross-linked and/or layered each other in a turbostratic way [1]. The properties of PAH and soot as topology and morphology, size, sp^2/sp^3 ratio and hydrogen content strongly depend on the burning conditions [1-3]. Thus, in line of principle the bottom-up process leading through PAH intermediates to soot with different graphenic character can be controlled by a suitable change of the combustion conditions.
In this work soot has been synthesized in the well-controlled combustion conditions of premixed laminar fuel-rich flames and its properties have been modulated changing the residence time into the flame and the precursors. Premixed flames burning methane (M), ethylene (E) and benzene (B) have been used to produce soot in a temperature range (T_{max} [1700-1770K]) appropriate for obtaining high soot yields [2].
Hydrocarbon gas provides both carbon species necessary for materials growth and the high temperature to its decomposition, thus no external sources of energy are required. Indeed, a premixed flame in partially oxidative condition represents a carbon-rich chemically reactive environment able to generate graphenic nanostructures during short residence times in a continuous single-step inexpensive process. The maximum soot yield spans from a minimum value of 1% to about 3.5% in dependence on the fuel. In particular, methane (M) exhibits the lowest sooting tendency, whereas benzene (B) shows the highest one [2]. Soot has been deposited on a quartz plate inserted horizontally at selected positions of the flame for the minimum time required both to collect a sufficient material for the further characterization and to limit the thermal degradation of the deposited sample at flame temperature [3]. Soot, after dichloromethane extraction of low MW aromatic species [3], has been characterized by UV-Visible spectroscopy and Raman spectroscopy with an excitation wavelength of 514nm and the correlations between the Tauc band gap, E_g, and the I(D)/I(G) ratio, reported in Figure 1, have been obtained.
Size exclusion chromatography performed on soot sampled has shown that this material is mainly composed by carbon aggregates with a band gap ranging from 0.4 to 0.1 with a minor contribution (<20%) of less organized carbon structures with a bang gap around 1 [3]. From HR-TEM analysis it has been observed that the graphene layers, grouped in number of 2-3, have a layer length much below 1 nm [1, 2]. For this layer size it has been shown that I(D)/I(G) increases as the graphite crystallite size increases [4]. Thus, in this work the band gap decrease accompanied by the increase the I(D)/I(G) ratio, testifies an increase in the order extent at higher residence times in the flame, and particularly in the benzene flame.
Overall, the well-controlled combustion conditions of a premixed flame have shown to be suitable to produce different carbon particulate matters burning different fuels in a narrow temperature range. A wide range of band gaps, i.e. electronic properties, can be easily obtained, showing the great flexibility of the flame reactor to produce carbonaceous materials with tailored properties by selecting the appropriate combustion conditions.

References

[1] Alfè M.,Apicella B., Barbella R., Rouzaud J.-N., Tregrossi A., Ciajolo A., Proceedings of the Combustion Institute, **32** (2009) 697.
[2] Russo C., Alfè M., Rouzaud J.-N., Stanzione F., Tregrossi A., Ciajolo A., Proceedings of the Combustion Institute, **34** (2013) 1885.
[3] Russo C, Stanzione F., Ciajolo A., Tregrossi A., Proceeding of Combustion Institute, **34** (2013) 3661.
[4] Ferrari, A. C., Robertson, J. Phys. Rev. B **61** (2000) 14095.

Figures

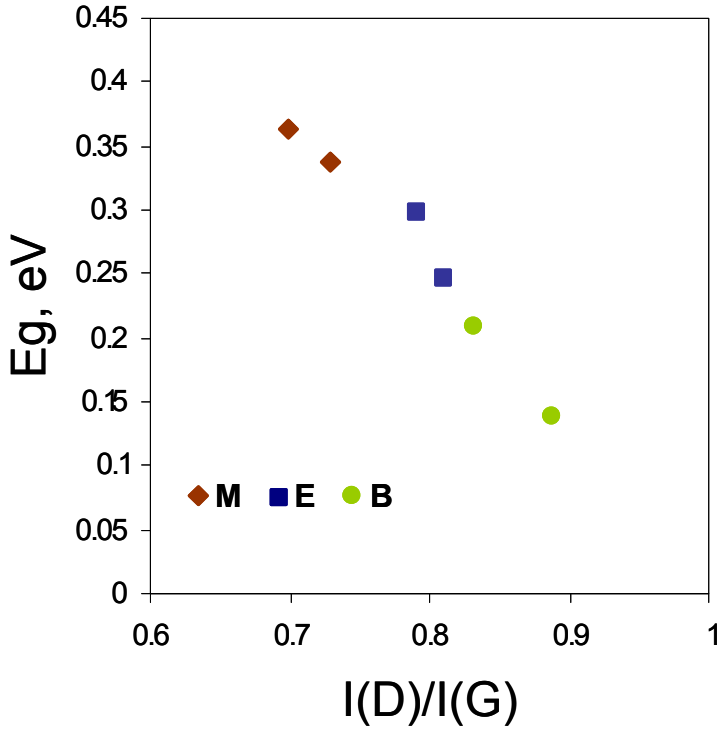

Figure 1 Correlation between Tauc band gap, E_g, and I(D)/I(G) ratio of soot for methane (M), ethylene (E) and benzene (B) flames.