Report on the Short Term Scientific Mission (STSM) conducted on the frame of the COST Action CM - 1404

Chemical characterization of soot nanoparticles in nucleation flames

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Home institution: Universität Bielefeld, Facultät für Chemie - Physikalische Chemie I

The current Short Term Scientific Mission STSM) was a continuation of the collaboration between Laboratoire de PhysicoChimie des Processus de Combustion et de l'Atmosphère (PC2A) in the Université de Lille 1 and Physikalische Chemistry 1 laboratory (PC1) in Universität Bielefeld. A previous STSM had been granted.

Motivation

Several approaches have been used to explain the soot formation process in combustion systems. In general, the whole process has been described in five steps: Formation of molecular precursors, soot nucleation, surface growth, coagulation – agglomeration and oxidation. Although, the steps of soot formation process are generally accepted within the scientific community, there are some open questions related to the soot nucleation process [1,2].

Some studies have been developed in PC2A – University of Lille in order to obtain new insights into the nucleation process. They have proposed the existence of a threshold equivalence ratio for which the duration of the laser-induced incandescence signal, which is linked to the size of the laser-heated primary particles, does not undergo any

increase with height above burner (HAB). This behavior, which was observed in low pressure flames [3] and at atmospheric pressure [4] means that the particles do not grow further along the reaction time or HAB. This threshold can be found in any premixed flame. The Lille group has called this type of flames "nucleation flames".

The nucleation flames are very attractive because the nucleation process is the major one. Thus, studying the chemical features of the soot nanoparticles formed in these flames can lead to a better understanding of the transition process between the gas phase and the solid phase.

As it was established in the last STMS with the participation of Dr. Pascale Desgroux and Christopher Betrancourt using the helium ion microscope (HIM) available in Bielefeld, small particles have been also detected with this instrument in the nucleation flames. However, there is currently no information on the chemical features of these particles.

Main goal

The main goal of the STSM was to obtain samples in order to analyze the chemical composition of soot nanoparticles in nucleation flames by Time-of-Flight Secondary Ion Mass Spectrometry (TOF-SIMS) available at Lille and by X-ray photoelectron spectroscopy (XPS) and polarization modulation - infrared reflection absorption spectroscopy (PM-IRRAS) available at Bielefeld. The objective was to analyze the molecular composition by TOF-SIMS and to correlate this information with the chemical features obtain by XPS and PM-IRRAS.

Description of the work carried out during the STSM

The set-up used for the sampling procedure was developed by PC2A laboratory. The first days of the STMS were dedicated to establish the sampling procedure. The specific set-up was optimized in order to obtain samples for TOF-SIMS. The preliminary TOF – SIMS analyses were performed and the results obtain showed similar profiles of those observed in other flames.

Afterwards, an optimization was required of the set-up, sampling time and vacuum used to obtain the samples for the PM-IRRAS and XPS. The PM-IRRAS has the main advantage

that a low amount of the sample is required to obtain the spectra. Besides, it eliminates any interference that can be produced by the presence of water molecules.

The cases of study were two butane flames (low-sooting and sooting flames). The heights above the burner used to do the sampling were determined according to the previous results of the Laser Induce Incandescence (LII) measurements in Lille.

Description of the main results obtained

Typical PM-IRRAS and XPS spectra are shown in Figure 1 and Figure 2. The PM-IRRAS measurements performed here show signals corresponding to aromatic C-H (3050 cm^{-}), alkane asymmetric C-H (2960 cm^{-1}), alkane asymmetric C-H(2928 cm^{-1}), alkane symmetric C-H (2860 cm^{-1}), carbonyl C=O (1724 cm^{-1}), aromatic C=C (1595 cm^{-1}), unsaturated C-H (1462 cm^{-1}) and C-O stretch (1257 and 1093 cm^{-1}) and are in good agreement with earlier results [5,6].

The XPS carbon signal was deconvoluted with respect to C-O, C=O and carbon bonds. The carbon bonds were not separated into single or double carbon bonds, since this fit usually is highly dependent on the parameters used to do the deconvolution. The fitting obtained for the carbon signal is accurate and it showed a significant contribution of the oxygenated functional groups to the carbonaceous particles composition[7].

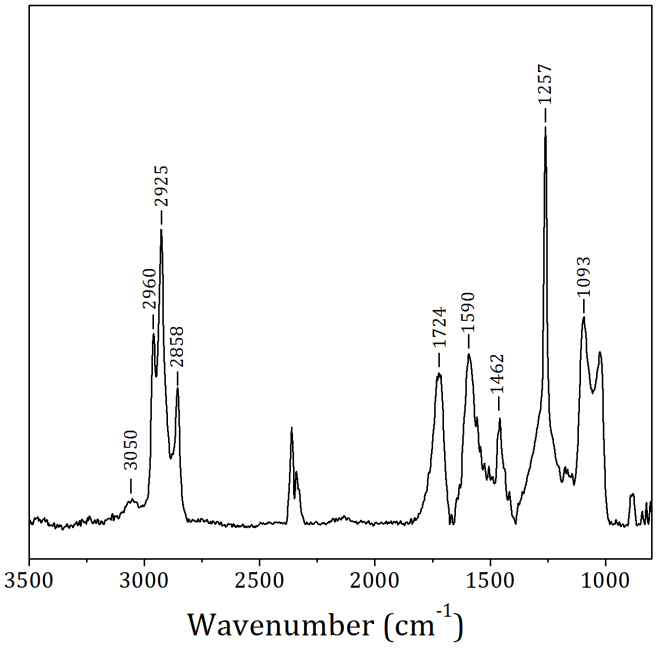

Figure 1. Typical PM-IRRAS spectrum obtain for carbon nanoparticles formed in flames.

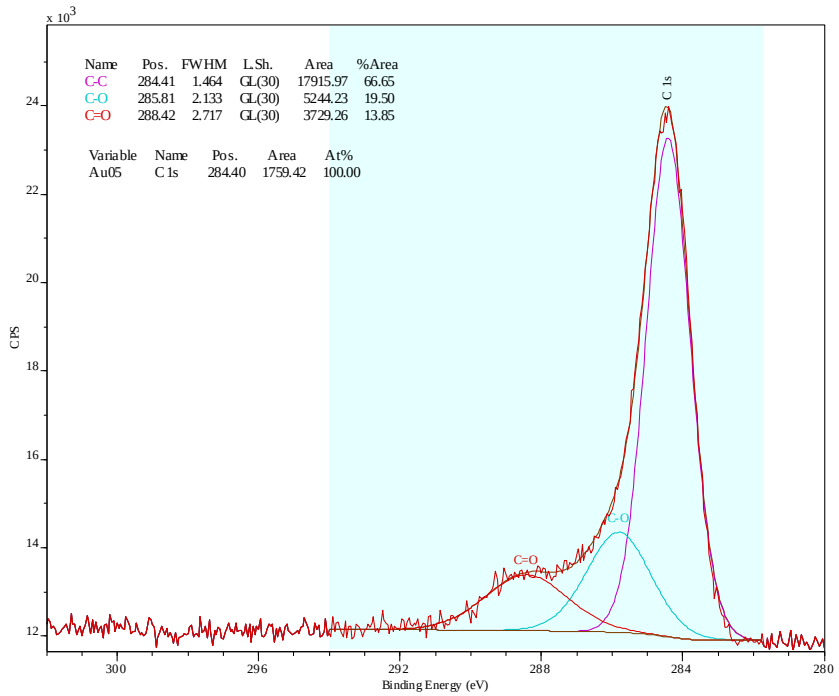

Figure 2. Typical XPS Carbon signal spectrum obtain for carbon nanoparticles formed in flames.

The results obtained in this STSM are the first insight regarding the particles formed in the nucleation flame. Understanding the chemical composition will increase the knowledge of the species associated to the nucleation process.

People involved

PC2A – University of Lille

- Christopher Betrancourt (PhD Student)
- Pascale Desgroux (Researcher)
- Alessandro Faccinetto (Researcher)

PC1 – University of Bielefeld

- Dr. Maurin Salamanca Guzmán (AvH Fellow)
- Lena Ruwe (PhD Student)
- Prof. Katharina Kohse-Höinghaus

EP – University of Bielefeld

- Daniel Emmrich (PhD student)
- PD Dr. André Beyer (Senior Researcher)
- Prof. Armin Gölzhäuser

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