Free nanoparticle studies using synchrotron radiation

J. Brian A. Mitchell

Institut de Physique de Rennes, UMR No. 6251 du CNRS, Université de Rennes I, 35042 Rennes, France
.
Presenting author email: mitchell@univ-rennes1.fr

The advent of lasers has undoubtedly had an enormous impact on the study of aerosol nanoparticles. Visible and even near ultra-violet light however, has its limitations as an *in-situ* characterising tool, as the wavelength is perhaps much greater than the species being studied, there is always a question of the index of refraction of the target, particularly if it is hot and it is not applicable to optically dense targets such as electrical arcs. X-rays offer a unique complementary tool for nanoparticle characterisation as the wavelength (typically 1 Å) is smaller than the target particle size, the index of refaction is fixed at Unity and they can penetrate even opaque solid matter. To be able to benefit from the special properties of x-rays, one must turn to the use of a high intensity source, such as a synchrotron radiation facility. Fortunately, there are now, more than 100 hundred such machines, in the World today and being user facilities, one can have relatively easy access to them, free of charge and indeed in many cases, with financing from the centres themselves or (under special circumstances) via European Support.

To be able to perform experiments at centres such as the European Synchrotron Radiation Facility, (ESRF) (France), Synchrotron Soleil (France), MAX I to IV (Sweden), ASTRID I and II (Denmark), BESSY II (Germany), to name a few, one must have (a) a reasonably compact, self-contained apparatus, able to be SAFELY operated by remote control (this feature generally being handled by the facility), (b) a good idea which can only be exploited using synchrotron radiation (i.e. requiring high x-ray flux), (c) perserverence to try again if your proposal is not accepted the first time and (d) a hardworking team as the experiments are generally run 24 hours per day over a period of at most up to 4 or 5 days.

In this talk, I shall mention a few techniques which have been used for nanoparticle characterisation. The first is Small Angle X-Ray Scattering (SAXS) in which a fine beam of intense x-rays is passed through a target and the nanostructure of the target (in our case nanoparticles) is investigated by measuring the angular scattering of the x-rays. The information collected allows the particle size, state of aggregation, surface character and particle number density to be determined as a function of particle formation conditions such as for example, residence time in a flame, the chemical origin and modification of the particles (flame additives [1], surface material for ablated particles [2], parent material for condensing particles [3,4]).

Another technique which is begun to be used is X-Ray Photoelectron spectroscopy of free nanoparticles in which a gas driven jet of prepared particles, focused by an aerodynamic lens, is crossed with soft-x-rays to generate electrons whose kinetic energy spectra are then measured. This provides direct information on the particle's chemical composition (e.g. state of oxidation) [5].

Molecular processes concerned with nanoparticle formation in flames are studied using Threshold Ionisation Mass Spectrometry [6,7].

My earliest studies involved x-ray induced ionisation for nanoparticle mapping in a flame and this is a technique which turned out to be surprisingly easy to perform due to an intense ionisation process which occurs during aggregated nanoparticle–x-ray interaction which is still not really explained [8].

The applications of synchrotron radiation to aerosol physics and chemistry have already been demonstrated therefore, and the future will depend upon the imagination of the researchers who have questions to answer which only synchrotron radiation can provide. Indeed the future is very exciting with the development of free electron laser sources (FELs) such as the LCLS in the US, X-FEL and FLASH in Germany, FERMI in Italy, and SACLA in Japan. These machines offer a billion times more X-Ray intensity that a 3rd Generation synchrotron (such as the ESRF or Soleil) and so one can imagine doing in-situ, single particle imaging with these apparatus. At the moment, access to FEL's is very limited but this should become easier in the future as the number of machines increases. Again what it will take, is a good idea !

References :

[1], J.B.A. Mitchell, J-L. LeGarrec, G. Saidani, F. Lefeuvre and S. di Stasio, Energy and Fuels, **27** (2013) 4891

[2] G.D Forster et al. Phys. Rev. Lett. (2015) In press

[3] E. Carvou, J.L. Le Garrec, J. Pérez, J. Praquin, M. Djeddi and J.B.A. Mitchell, AIP Advances **3** (2013) 032139

[4] S. Popescu, E. Jerby, Y. Meir, Z. Barkay, D. Ashkenazi, J.B.A. Mitchell, J.L. LeGarrec and T. Narayanan, J. Appl. Phys. **118** (2015) 023302

[5] O. Sublemontier, et al. J. Phys. Chem. Lett. **5** (2014) 3399

[6] Y. Li, F. Qi, Acc. Chem. Res. **43** (2010) 68

[7] N. Hansen et al. J. Vis Exp 87, (2014) 51369

[8] J. B. A. Mitchell et al. Combustion and Flame **131** (2002) 308

[9] Aquila et al. Structural Dynamics, **2** (2015) 041701

[10] N. D. Loh, C. Y. Hampton, A. V. Martin, D. Starodub, R. G. Sierra, A. Barty, A. Aquila, J. Schulz, L. Lomb, J. Steinbrener, et al., Nature **486**, (2012) 513