Single-Particle Laser Ablation Time-of-Flight Mass Spectrometer (SPLAT-MS)

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Abstract

Atmospheric aerosols impact the Earth’s climate, air quality, and human health. To understand these impacts, it is vital to know the size and chemical composition of aerosols (Imre et. al., 2000). The SPLAT-MS is an innovative new instrument that can analyze both of these properties on single aerosol particles ranging in size from 80 to 500 nm. SPLAT-MS utilizes an aerodynamic focusing lens to focus incoming aerosols into a beam. Two optical detection stages can detect a single particle and determine its aerodynamic diameter through a time-of-flight calculation. Detection of a particle at both stages triggers an ablation laser to fire, and the ions created during the ablation process are collected and analyzed using a reflectron time-of-flight mass spectrometer. Thus, a mass spectrum is produced, and the composition of the aerosol particle is revealed.
Aerodynamic Focusing Lens

- The lens tube consists of six precision machined orifice lenses ranging from 5 mm inner diameter at the entrance to 3 mm inner diameter at the exit.
- The lens focuses particles into a narrow beam (~ 1 mm diameter) with nearly 100% particle transmission efficiency to the detector for particles with diameters between 70 and 500 nm.
FLUENT simulation of the aerodynamic lens for 100 nm diameter spheres with density 1 g/cm³ at typical lens inlet pressure (2.1 torr) and volumetric flow rate (97.3 sccm).
Detection and Sizing

• The final orifice of the lens controls supersonic gas expansion and particle acceleration.
  – During the gas expansion, smaller diameter particles accelerate to faster velocities than larger ones due to different inertias; Thus, a distribution of velocities is obtained.
• The particle-beam passes through two differentially pumped skimmers in order to remove excess gas.
  – The pressure in the main chamber is \( \sim 10^{-8} \) torr.
Optical Detection

- The particle-beam passes through a series two green laser beams (frequency-doubled Nd:Yag, 532nm), orthogonal to one another.

- Large ellipsoidal reflectors are used to collect over 40% of the scattered light and to image it into a small pinhole, where it is detected by a photomultiplier.
If a pre-set number of photons per microsecond are detected at the first laser beam, then a countdown clock is started. If this pre-set number is detected at the second laser beam before the countdown clock expires, then the elapsed time is recorded, providing the particle velocity.

The particle velocity is then used to determine aerodynamic diameter, and it also is used to trigger the ablation laser.
Velocity-Aerodynamic Diameter Calibration

$$D_{aero} = fxn(D_{geometric}, density, shapefactor)$$
Size Distribution Calibration

1 minute ambient size distribution obtained by SPLAT-MS superimposed on a calibration run against a mixture of PSL spheres prepared by combining 98nm, 204nm, 302nm, 404nm and 504nm at a ratio of 50:1:1:1:1.
Resolution of SPLAT-MS
Size Distribution of 163,000 Hit Particles

(60 hours of data collection)

TexAQS 2000
Time-of-Flight Mass Spectroscopy

- An argon-fluorine eximer laser operating at 193 nm ablates the detected particle.
  - The ablation process is intended to evaporate the particle and form molecular ions.
- A reflectron time-of-flight mass spectrometer is used for particle composition analysis.
Example Mass Spectrum
Sample Mass Spectra

Vanadium

Na⁺, Ca⁺, C⁺, Al⁺, Fe⁺, V⁺, VO⁺

Organic nitrate

N⁺, O⁺, C⁺, CH⁺, CH₂⁺, CH₃⁺, NH⁺, NO₂⁺

Organic amine

C⁺, CH⁺, CH₂⁺, CH₃⁺, H⁺, NH⁺

Biomass

1.45 nm
TexAQS 2000 Nitrate Plume
(Sept. 13 - Sept. 14)

NITRATE

VANADIUM

3000nm
20nm
Sulfate Containing Particles

TexAQS 2000
Sept 13-Sept 14, 2000
For the month of July, 2001, SPLAT-MS was deployed in Queens, NY for the PM (particulate matter) 2.5 Technology Assessment and Characterization Study (PMTACS).

The purposes of PMTACS-NY is to collect, analyze, and interpret data on the chemical and physical composition of PM to:

- Measure the temporal and spatial distribution of the PM 2.5 and other pollutants.
- Support health effects and exposure research.
- Evaluate new measurement technologies and establish their potential for routine monitoring.
- Monitor the effectiveness of new emission control technologies.
SPLAT-MS Queens Team

Pictured Left to Right: Cynthia Randles (GCEP SURE), Dr. Alla Zelenyuk, Dr. Dan Imre, Logan Chieffo (ERULF)
PMTACS-NY Field Study

Above: View of site at Queens College, NY

Left: View of trailers, sample intakes, and tower

Right: Instrument Tower
PMTACS-NY Field Study

Views of Manhattan from Queens College Site

High PM

Low PM

Sample Intakes

SPLAT-MS in Queens
References


• PMTACS-NY Field Study web-site: http://www.asrc.cestm.albany.edu/pmtacsny/
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