

Organic Matter in Cosmic Dust

Scott A. Sandford¹, Cécile Engrand², and Alessandra Rotundi³

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Organics are a significant component of most cosmic dust, as revealed from actual samples of extraterrestrial dust in the Earth's stratosphere, in Antarctic ice and snow, in near-Earth orbit, and in asteroids and comets. Cosmic dust contains a diverse population of organic materials that owe their origins to a variety of chemical processes occurring in many different environments. The presence of isotopic enrichments of D and ¹⁵N suggests that many of these organic materials have an interstellar or protosolar heritage. The study of these samples is of considerable importance because they are the best preserved materials of the early Solar System available.

KEYWORDS: cosmic dust; organics; carbonaceous matter; hydrocarbons; microanalysis

INTRODUCTION

Dust is seen in a wide variety of astrophysical environments that include stellar outflows, the diffuse interstellar medium, dense interstellar clouds, and disks surrounding newly forming stars. Closer to home, our own Sun is surrounded by a cloud of dust (the Zodiacal Dust Cloud), formed by dust from comets and asteroids. Our knowledge of the nature of the dust in these environments is based on telescopic observations, spacecraft in situ observations, and laboratory studies of collected dust particles. The dust in all these environments consists of a number of different components, the primary ones being minerals and organic materials. In some very cold environments, condensed gases, in the form of mixed molecular ices, may also be present.

In this article, we offer a brief discussion of organics outside the Solar System, largely for context, and then restrict ourselves to discussing laboratory studies of organics in interplanetary dust particles (IDPs) collected in the Earth's stratosphere, in unmelted micrometeorites (MMs), and in cometary particles returned by the *Stardust* mission and studied in situ by the *Rosetta* mission.

The study of organics in extraterrestrial materials is an inherently difficult activity. Most extraterrestrial materials contain only a few weight percent organics, which are usually finely dispersed and amorphous. The organic populations present are often diverse, so that no individual compound or class of compounds is very abundant (e.g.

the abundances of amino acids in meteorites are typically reported in ppm). These difficulties are exacerbated for organics in cosmic dust where the samples are small—identifying a multitude of different organic compounds in the few percent of organic material in a nanogram sized dust particle is not a trivial matter! As a result, our knowledge of organics in cosmic dust is often restricted to the determination of overall organic abundance and basic characterization of the classes of compounds present. Finally, because we live

on a planet that is rife with complex organic compounds, the study of extraterrestrial organics requires constant vigilance against possible terrestrial contamination.

ORGANICS IN THE INTERSTELLAR MEDIUM

The study of the organic component of cosmic dust and gases found outside our own Solar System is restricted to telescope-based spectroscopic observations. These provide general information about the nature of organics found in stellar outflows, in the diffuse interstellar medium, and in the cold, dense clouds in which new stars and planetary systems form. These observations demonstrate that hydrogenated amorphous carbon, polycyclic aromatic hydrocarbons, and related materials are some of the dominant forms of complex carbon in space (e.g. Dartois and Muñoz-Caro 2007). Aliphatic hydrocarbons are also observed in some environments, including in the diffuse interstellar medium (e.g. Sandford et al. 1991). In the very low temperatures (10–100 K) of dense interstellar clouds, all these organic materials are expected to be largely condensed out onto refractory dust particles along with more volatile species like H₂O, CH₃OH, NH₃, CH₄, CO, CO₂, and so on (Boogert et al. 2015). Laboratory experiments have demonstrated that energetic processing by ultraviolet light, electrons, and cosmic ions modify the chemical composition of the ices and produce additional organic molecules, including organic refractory materials that can survive higher temperatures (25 °C and above) and can be found in objects like meteorites, in Antarctic MMs, and in IDPs (e.g. Dworkin et al. 2001). Subsequent irradiation of these “ice residues” can then alter the original organics and generate new ones.

The organics originally incorporated into our Solar System reflect a long and varied history involving a wide variety of environments and chemical processes. This initial complexity was then compounded by subsequent chemical processing in the protosolar nebula, asteroids, and comets.

1 NASA Ames Research Center
Moffett Field, CA, USA
E-mail: Scott.A.Sandford@nasa.gov

2 Centre de Sciences Nucléaires et de Sciences de la Matière-CSNSM
Bat. 104, 91405 Orsay-Campus, France
E-mail: Cecile.Engrand@csnsm.in2p3.fr

3 University of Naples “Parthenope”, Naples, Italy and
INAF-Istituto di Astrofisica e Planetologia Spaziali, Rome, Italy
E-mail: rotundi@uniparthenope.it

STRATOSPHERIC INTERPLANETARY DUST PARTICLES (IDPs)

Dust particles released from asteroids and comets migrate through the Solar System under the influence of both gravitational and nongravitational forces that can ultimately lead them into collision with the Earth's atmosphere. Materials that survive deposition into the upper atmosphere drop to the Earth's surface on timescales of minutes to weeks, depending on their masses and densities. It is, therefore, possible to collect dust-sized particles as they settle through the stratosphere using specialized aircraft or balloon-borne instruments (Brownlee 2016 this issue; Rietmeijer et al. 2016; Taylor et al. 2016 this issue).

Given the extremely small size of extraterrestrial dust collected in the stratosphere (particles typically have masses of ~1 ng), most of what we know about them derives from microanalytical techniques, including infrared and Raman spectroscopy; X-ray absorption near edge spectroscopy (XANES); scanning transmission X-ray microscopy (STXM); laser-desorption laser-ionization mass spectrometry (L^2MS); scanning electron microscopy and transmission electron microscopy (SEM and TEM); and (nano) secondary ion mass spectrometry (SIMS and NanoSIMS).

Chondritic porous interplanetary dust particles (CP IDPs), considered the least altered samples of the solar nebula dust, often contain carbonaceous material and, in many cases, show significantly higher bulk carbon abundances than do carbonaceous chondrites (e.g. Thomas et al. 1993; Fig. 1). The carbon in stratospheric IDPs shows a wide range of morphologies, including poorly graphitized carbon, coatings on mineral grains, and discrete “nanoglobules” (e.g. Keller et al. 2004; Matrajt et al. 2013 and references therein). High-resolution X-ray imaging and spectroscopic analyses on ultramicrotome sections of CP IDPs show many have an ~100 nm thick coating of organic matter that often exists on the surface of individual submicron mineral grains within IDP aggregates. These organic layers may have acted like a “glue” that increased the efficiency of dust particle aggregation in the solar nebula (Flynn et al. 2013).

Much of the carbonaceous material in IDPs consists of amorphous aromatic materials, but aliphatic moieties and C=O groups are also present (Flynn et al. 2013). The population of aromatic species includes a wide variety of polycyclic aromatic hydrocarbons (PAHs) and their alkylated derivatives, some having masses >500 atomic mass units (amu) (Clemett et al. 1993). The PAH population sometimes shows a two-lobed mass distribution, with peaks centered near ~250 amu and ~400 amu. The higher mass part of the distribution may be due to thermal alteration (sintering) produced during atmospheric entry heating.

Isotopic studies show that many IDPs exhibit extreme H and N isotopic heterogeneity on the submicron scale. The most typical variations are excesses in D and ^{15}N , which are generally attributed to organics (e.g. Keller et al. 2004 and references therein). The presence of such anomalies demonstrates that the materials have not been terrestrially contaminated. Furthermore, because D and ^{15}N enrichments are expected from many types of astrochemical processes, particularly those from very cold environments such as dense interstellar clouds—the environments where new stars and planetary systems form (Sandford et al. 2001)—their presence suggests that their chemical organic carriers (or at least their precursors) have origins in the interstellar medium and/or outer portions of the protosolar disk. If true, then these materials could potentially predate the Solar System.

ANTARCTIC MICROMETEORITES

Micrometeorites (MMs) are IDPs collected at the Earth's surface, most efficiently at the polar ice caps. They range in size between 20 μm and 2,000 μm , the smaller fraction being the more abundant. Antarctic micrometeorites are mostly related to a primitive class of meteorites called the carbonaceous chondrites, which represent only a few percent of the meteorites collected on Earth. Chondritic micrometeorites contain a few weight percent of insoluble organic matter (IOM), which tends to have a primitive and very disorganized structure (Engrand and Maurette 1998).

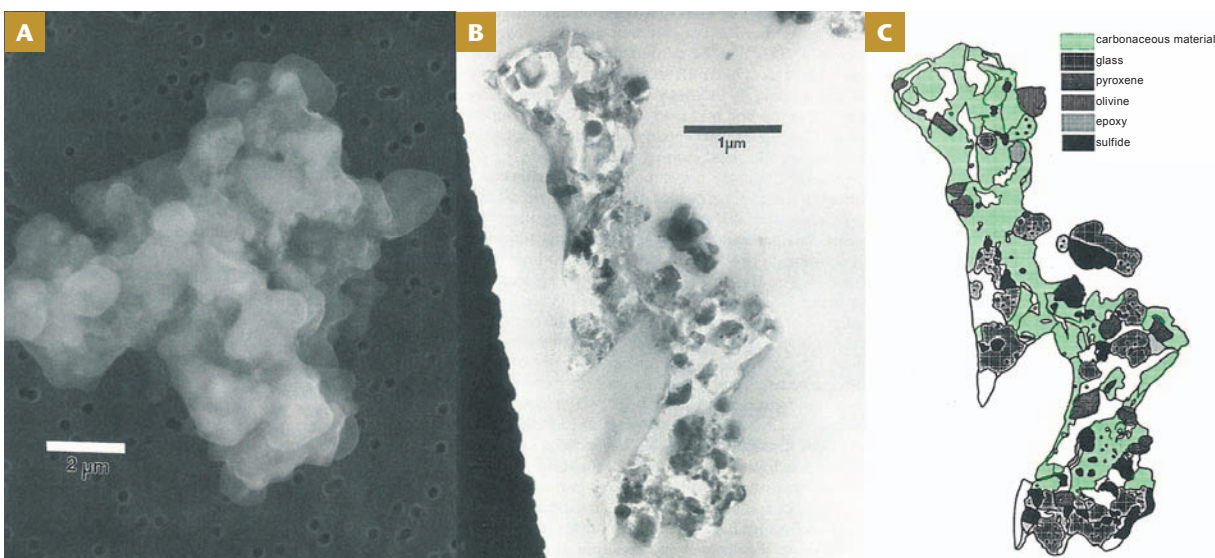


FIGURE 1 (A) Scanning electron microscope image of a stratospheric interplanetary dust particle (IDP). (B) High magnification image, taken using a transmission electron microscope, showing that the IDP particle shown in (A) is an aggregate of smaller grains. (C) Compositional map corresponding to the TEM

image in (B). Note that carbonaceous material, indicated by the light green regions, is present at the 40–50% level. By comparison, carbonaceous chondrites (meteorites rich in carbon) typically only contain 3–5% carbon. IMAGES IN B AND C MODIFIED AFTER THOMAS ET AL. (1993)

Micrometeorites collected from blue ice at Cap Prud'homme (Antarctica) contain soluble organic molecules such as PAHs (Clemett et al. 1998; Fig. 2) and amino acids (Matrajt et al. 2004 and references therein).

Ultracarbonaceous micrometeorites have been found in the Dome Fuji collection from Antarctica (Nakamura et al. 2005) and in the CONCORDIA micrometeorite collection made at Dome C (Fig. 3) (Duprat et al. 2010). These particles have carbon contents up to 10 times that of the most C-rich carbonaceous chondrites, i.e. they are up to 85% organic matter by volume (65 weight percent of C) (Dartois et al. 2013 and references therein). Such concentrations are comparable with the most C-rich stratospheric IDPs (Thomas et al. 1993; Keller et al. 2004) and are compatible with the "CHON" grains (grains rich in C, H, O, and N) detected in comet Halley by the *Giotto* and *Vega* spacecraft.

The organic matter in ultracarbonaceous MMs show extreme D/H ratios and is significantly enriched in nitrogen compared to meteorites, with nitrogen abundances reaching ~20 atomic percent in the organic matter (Duprat et al. 2010). The organic material is low in oxygen and CH₂/CH₃. Overall, ultracarbonaceous MMs have many mineralogical, isotopic, and spectroscopic similarities with cometary grains. Their organic matter could have been produced by the irradiation of N- and C-containing ices in dense clouds, or in the colder portions of the protosolar nebula, or possibly at the surface of comets.

Recent analyses by combined STXM, μ XANES, SEM, and TEM have shown the presence of two distinct phases of carbonaceous matter in ultracarbonaceous MMs that have different N contents: the N-rich phase (atomic N/C up to 0.2) shows a smooth texture and is devoid of minerals; the N-poor phase is associated to fine-grained minerals (Enggrand et al. 2015). This suggests that ultracarbonaceous MMs contain organics that formed by different mechanisms and/or in different locations in the protoplanetary disk. The soluble organic content of ultracarbonaceous MMs has yet to be characterized.

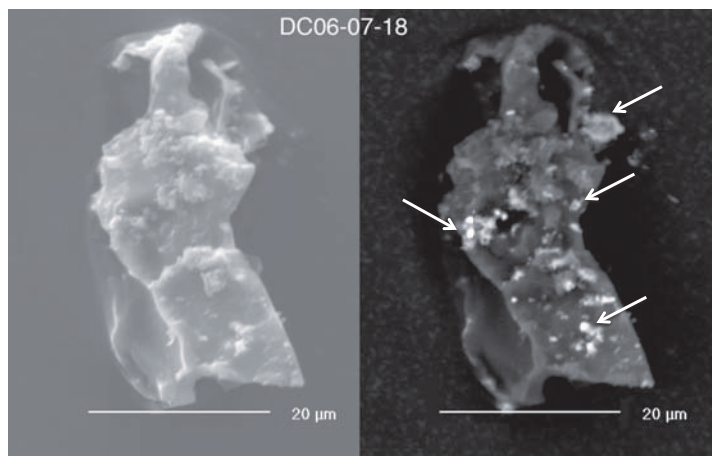


FIGURE 3 Two images of the same fragment of an ultracarbonaceous Antarctic micrometeorite that has been placed on carbon tape. (LEFT) Secondary electron (SE) micrograph. (RIGHT) Back scattered electron (BSE) micrograph showing that the particle is essentially made of organic matter (dark grey), with small mineral inclusions (showing up in lighter grey shades) that are mostly silicates and Fe-Ni sulfides (some of them are arrowed).

COMETARY ORGANICS SEEN BY THE STARDUST AND ROSETTA MISSIONS

Dust samples were collected from comet 82P/Wild 2 in 2004 when the *Stardust* spacecraft flew through the comet's coma at a relative velocity of ~6.1 km s⁻¹. The samples were subsequently returned to Earth in 2006 and have since been available for laboratory study.

Stardust had a passive dust impact collector made of aerogel, an extremely low density Si-O-based material that contained some residual carbon. Despite the use of low-density aerogel, impacting particles suffered varying degrees of damage during impact and, unfortunately, organic materials generally survived impact less well than mineral grains. In some particles, much of the original organic material was mobilized and injected into the surrounding aerogel, as evidenced by infrared spectra of "halos" of -OH, aromatic -CH, aliphatic -CH₂- and -CH₃, and C=O groups that were seen surrounding the physical tracks (Sandford et al. 2006). Halos of organics are not seen around tracks when samples of carbonaceous meteorites are fired into aerogel, suggesting that the halos in the *Stardust* aerogel are due to organic materials that are more labile than those typically seen in meteorites. Comparisons between the infrared spectra of the organics in Solar System IDPs and in Wild 2 particles with those from the diffuse interstellar medium suggest that both Solar System-derived materials have much higher CH₂ to CH₃ ratios than the interstellar organics (Flynn et al. 2003). In the case of the Wild 2 comet particles, it is difficult to assess the extent to which this material was altered from its original form in the particle.

The diffuse dispersion of organics in *Stardust*'s non-track-containing aerogel is demonstrated by the detection of amines and amino acids that have nonterrestrial isotope ratios (Elsila et al. 2009). Whether these compounds were volatilized from impacting particles or simply arrived at the collector in the gas phase is unknown.

Fortunately, there are cases where organics in the particles survived largely intact and unaltered because they were protected during impact by surrounding minerals (Fig. 4; Matrajt et al. 2013). In these cases, organics are distributed throughout the terminal particle or as individual concentrations within them (Rotundi et al. 2014).

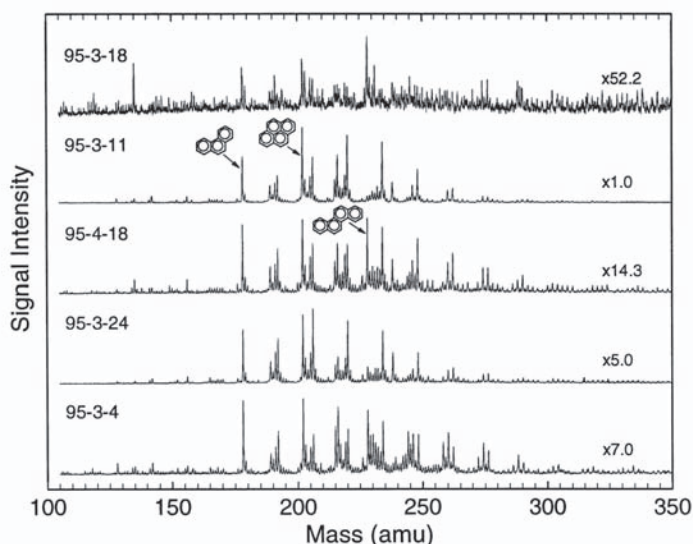


FIGURE 2 Five spectra of polycyclic aromatic hydrocarbons (PAHs) detected in Cap Prud'homme (Antarctic) micrometeorites using the L²MS technique in which an initial laser is used to vaporize material from the sample and a second laser ionizes selective molecular species before they are passed into a mass spectrometer. In this case, the ionization laser is tuned to give good yields for PAHs, which here consist of fused benzene rings. Example PAH structures are shown (moving from left to right) for peaks consistent with the PAHs phenanthrene (178 amu), pyrene (202 amu), and chrysene (228 amu). IMAGE REPRODUCED WITH PERMISSION OF SPRINGER FROM CLEMETT ET AL. (1998)

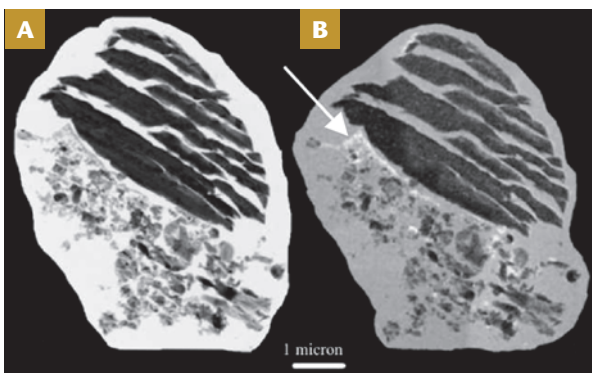


FIGURE 4 Energy-filtered transmission electron microscope images of a microtome slice of a *Stardust* particle. (LEFT) An image obtained using zero-loss filtering to provide contrast improvement. (RIGHT) An image made using energy passbands above and below the 285 eV carbon edge: in such images, the presence of carbon is indicated by light areas. The sulfide at the upper right of the images (arrow) is carbon free, but regions of carbon are clearly seen in the fine-grained chondritic material below the sulfide grain. IMAGE REPRODUCED WITH PERMISSION OF THE AMERICAN ASTRONOMICAL SOCIETY FROM MATRAJT ET AL. (2013)

Given the small amounts of organics in terminal particles, most of what we know is based on use of the same micro-analytical techniques used to study stratospheric IDPs, including XANES (Sandford et al. 2006), infrared and Raman spectroscopy (Rotundi et al. 2014), SEM (Rotundi et al. 2014), L²MS (Clemett et al. 2010), and SIMS (McKeegan et al. 2006). Many of the cometary organics are similar to those found in other extraterrestrial materials. For example, Raman data suggests that the *Stardust* samples contain material similar to the IOM that dominates the organics in meteorites (Sandford et al. 2006; Rotundi et al. 2014). *Stardust* samples also contain similar three-dimensional organic structures, such as the “nanoglobules” (De Gregorio et al. 2010) that can be found in carbonaceous meteorites and IDPs (Matrajt et al. 2013 and references therein).

However, analyses of *Stardust* particles also show these samples are more diverse than those found in other extraterrestrial materials; they contain additional types of organics that are absent, or considerably less abundant, in meteorites and IDPs. The XANES, infrared, L²MS, and Raman data all indicate that more labile, aromatic-poor materials are also sometimes present. The diversity of organic material present is demonstrated by nanoSIMS measurements, which show that the C/N ratios can vary enormously across a single particle (Sandford et al. 2006).

Isotopic analyses demonstrate that some of the organics contain isotopic anomalies in the form of enrichments of D and ¹⁵N relative to terrestrial H and N (McKeegan et al. 2006). As with the stratospheric IDPs, the presence of such anomalies demonstrates that the materials are not contaminants but of interstellar and/or protosolar heritage.

Although the European Space Agency’s *Rosetta* mission to comet 67P/Churyumov–Gerasimenko (hereafter “67P”) will not return samples to Earth, it is equipped with several instruments designed to study dust in the comet’s coma. These include GIADA (grain impact analyser and dust accumulator), which measures the number, mass, momentum, and velocity distribution of dust particles in the near-comet environment, and COSIMA (cometary secondary ion mass analyzer), which images dust collected on metallic targets and measures the elemental and isotopic composition of key elements.

GIADA collected compact particles (0.03–1 mm in size) and fluffy aggregates (0.2–2.5 mm in size) of submicron grains that had a bulk density constrained in the range $(1.9 \pm 1.1) \times 10^3 \text{ kg m}^{-3}$ and up to 1 kg m^{-3} , respectively (Rotundi et al. 2015). The fluffy aggregates are probably similar to the aggregates collected by COSIMA (FIG. 5) and are probably related to cosmic dust particles collected on Earth (Schulz et al. 2015).

The organic content of these particles as detected by COSIMA is currently under investigation. The Cometary Sampling and Composition experiment on *Rosetta*’s lander *Philae* did detect a suite of 16 molecules of which all but one (H₂O) contained one or more C atoms (Goesmann et al. 2015). Thus, it is likely that organic species are common in the cometary grains from this comet.

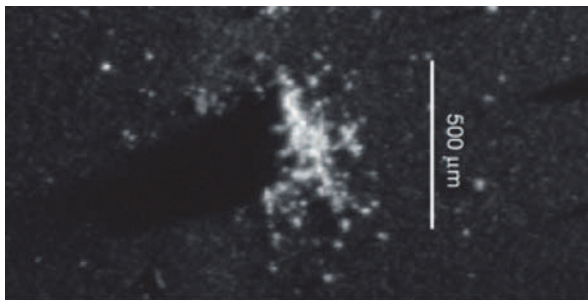


FIGURE 5 An agglomerate dust particle from comet 67P/Churyumov–Gerasimenko, as collected by COSIMA (cometary secondary ion mass analyzer). The image was obtained using grazing illumination from the right. The length of the shadows indicates that the altitude above the substrate reaches about 100 μm . REPRODUCED WITH PERMISSION OF NATURE PUBLISHING GROUP FROM SCHULZ ET AL. (2015)

SUMMARY

Cosmic dust particles contain organics in a variety of morphological forms. The populations of organic compounds present in these particles are complex and appear to vary from particle to particle, implying these materials have had complex histories. The presence of isotopic enrichments in D and ¹⁵N in some of these materials suggest this history extends back to the interstellar dust cloud from which our Solar System formed, i.e. some of these materials may well be older than the Solar System itself. These organics are presumably the result of universal processes that occur whenever and wherever new stars and planetary systems form: thus, we can speculate that similar materials commonly fall onto planetary surfaces elsewhere. If such materials played an important role in the origin of life on Earth, they are expected to be available to play a similar role on other planets throughout the universe.

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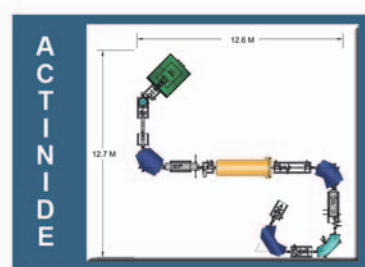
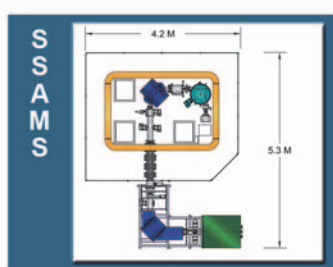
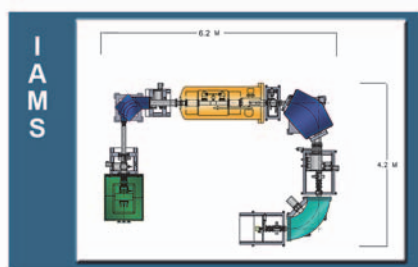
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