# HIGH FLUENCE SYNCHROTRON RADIATION MICROPROBE EFFECTS ON STARDUST INTERSTELLAR DUST CANDIDATES.

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**Introduction:** Besides the cometary samples collected from comet 81P/Wild 2 [1], the Stardust NASA mission returned in 2006 the first interstellar dust samples. The interstellar tray collected particles for a total of 195 days during two exposures to the interstellar dust stream. Analysis of these candidates from both low density aerogel capture cells and Al impact foils is performed by the Stardust InterStellar Preliminary Examination (ISPE) collaboration [2]. The aerogel samples were analyzed *in situ* in their pico-keystones by synchrotron radiation focused microprobe beams at four international facilities : the ESRF, in Grenoble France, the ALS in Berkeley, CA, USA, the NSLS in Upton, NY, USA and the APS in Argonne, IL, USA.

**Experimental:** Following identification of impacts by the Stardust@home distributed microscopy search protocol, pico-keystoning [3] extracts mimimal thickness slabs of aerogel containing the IS particles for further analysis. The ISPE team has implemented a unique analysis protocol for rare samples involving sequential analyses, starting with the least destructive probes in order to preserve sample integrity and avoid contamination and alteration risks. Consequently, synchrotron radiation has been singled out as the first non-destructive technique to be applied to the ISPE samples as FTIR, XRF, and STXM analyses. Details of these synchrotron radiation analyses can be found in Westphal *et al.* [4].

**Results:** Tracks 30 and 34 of the IS collector containing 3 candidates hereafter dubbed Orion and Sirius (Tr. 30) and Hylabrook (Tr. 34) were analyzed at ESRF, ID13 and ID22NI nano-XRF/XRD beamlines, and ALS, 11.0.2 STXM beamline. Beam damage effects were noticed on both samples and a careful analysis of their irradiation history was then established. The purpose of this abstract is to present these facts, analyze potential damage mechanisms and propose new experimental protocols minimizing such effects in the future.

Prior to the ESRF last experiments, we imposed a simple exposure limit of the ISPE samples based on the following assumptions: IS particles have an average residence time in the InterStellar Medium (ISM) of 100 My velocities between 10 and 20 km/s. They were exposed to the quasi-isotropic diffuse cosmic X-rays background in the 3 – 300 keV energy range, most likely produced by active galactic nuclei (AGN) components. This background was measured by detectors on board the HEAO 1 spacecraft and fitted by a simple analytical expression by Gruber *et al.* [5]. Conservatively, we take the X-ray flux integrated over time (or fluence) and neglect solar system irradiation from any other other origins. For the 30 My interval we chose, fluence is about $(4 - 6)\times10^5$ $J/cm^3$ depending on the grain's composition and effective absorption. Using the ESRF beam energy of 17 keV and the ID22NI nanoprobe focused beamspot of about 150 x 190 nm (vert. x horiz.), this yields a fluence of about $3\times10^{19}$ $ph/cm^2$ hereafter called the Astrophysical Limit (AL). Here we conservatively consider that all energy absorbed or scattered by a Ø 1 μm particle is contained within the particle. At 17 keV, the main interaction process is photo-ionization yielding photoelectrons of energy between 0 and 16.5 keV followed by inelastic (Compton) and elastic (Rayleigh) scattering. Following the ionization, deexcitation proceeds by the fluorescence and Auger electron channels, as a function of the atomic number Z of the sample. Ranges of electrons in matter at these energies using the CSDA (Continuously Slowing Down Approximation) are on the order of 1-2 μm in silicates, as a function of average Z, A and density of the sample so we can conclude that most of the absorbed energy (with the exception of a small fraction re-emitted by fluorescence or scattered) will remain in the sample. The irradiation of Track 30 was performed at lower fluences ≤ AL by STXM and XRF at ALS, ID13 ESRF and yielded low-Z composition maps identifying the grains as Mg-rich (Sirius) and Al-rich (Orion). As the ID13 maps taken at 45° angle of incidence, the 2 particles appeared as one. The next analyses were done at ID22NI at normal incidence (≈ 150 nm resolution) which resolved the two particles and also yielded diffraction data featuring a spinel microphase and another so far unidentified phase. Because of low flux from monochromator problems, the pink beam was used instead, featuring a roughly 1.5% energy bandwidth or one full undulator harmonic at 17 keV which caused beam monitoring errors and irradiation at over 50 times more than the AL. Short term irradiation with a 0.2 sec/point dwell time produced no damage but the following two exposures of 8 sec dwell time, yielded 50-60 times larger doses than the AL and produced a horizontal (scan direction) smearing of Sirius. Orion (probably $Al_2O_3$) appears unchanged and its composition is stable while Sirius (MgO or $Fo_{90}$), quite unexpectedly, has lost some of its mass, including some Mn and K.

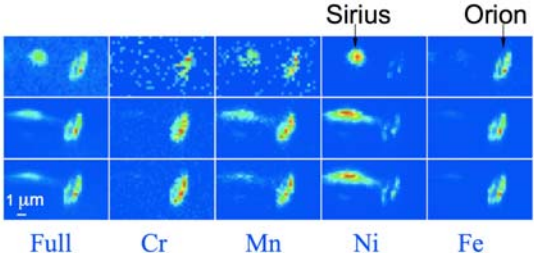

*Figure 1. XRF maps for three scans on Track 30's Sirius and Orion: upper, at short dwell time per pixel (0.2 s), lower two, at long dwell time (8 s), showing lateral dispersion of Sirius.*

Hylabrook/Track 34 (a particle of MgO composition) was analyzed by STXM before and after the ESRF ID13 measurement, the fluence of wisphich was roughly 50 times higher than the AL (, due to a large number of repeats (160) for a diffraction angular scan but using a single exposure fluence below the $3x10^{19}$ $ph/cm^2$ limit.

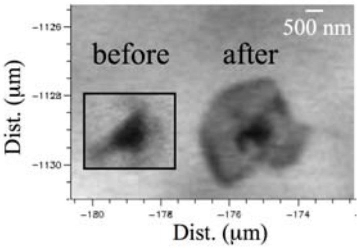

*Figure 2. STXM density maps for two scans on Track 34's Hylabrook, left: before the multiple exposure on ID13, right: after, showing radial smearing and a center loss.*

This raises the questions of damage as a function of the composition and fluence vs. flux or whether multiple shorter exposures are preferable to long ones, a situation known in biochemistry as the "recovery effect" [6] but which produces here an unwanted effect.

XANES spectroscopy, performed in STXM mode is a very useful tool for assessing the chemical changes undergone by a sample. Here XANES performed at the Mg K edge before and after the ID13 exposure, shows little mineralogical changes.

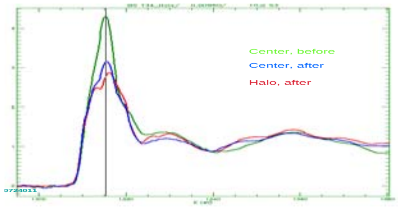

*Figure 3. Mg K-edge XANES on Hylabrook*

**Damage mechanisms:** After analyzing effects of heating, charging and ion displacement [7, 8], we can safely dismiss heating and ion displacement in favor of charging of our particles, subsequent to photo and Auger electron emission. At thermal equilibrium, the temperature rise due to beam heating of our particles is less than 1 K, thus negligible. Similar to $e^{-}$beam charging, photon irradiation followed by electron emission on a Ø 1µm particle impacted by our $3x10^{10}$ ph/s beam will add up to 100 V in 25 ms, producing spark discharges in the aerogel. Overall structural damage of the mineral lattice will occur in a more reduced way due to the absence of the electrostatic mirror charge or to lower relaxation effects. Finally, ion displacement following photoionization is reduced as air around the particle swiftly restores the electric neutrality.

**Outlook:** To address potential beam damage we propose to enforce the following prerequisites for all future analyses on high energy (10-17 keV) synchrotron beams:

- Use of a He shower for x-ray microprobe analyses;
- Establish 10-17 keV flux/fluence limits in a He shower based on the increased optical density of aerogel as measured by STXM;
- Use FIB and TEM at NCEM, Berkeley to establish benchmark samples of, *e.g.*, $MgSiO_3$, forsterite and muscovite, which show measurable effects to low-intensity exposures to 10-17 keV $e^{-}$ beams [8];
- Establish automatic shutter protocols on ID13 and ID22NI on beam-loss or scan-stop events;
- Enforce setting up hardware/software limits on any high-energy beamlines preventing sample irradiation above the established flux/fluence limits.

**References:** [1] Brownlee *et al.*, (2006), *Science* **314**, 1711-1715. [2] Westphal *et al.*, *LPS XLI*, #2050, (2010). [3] Westphal *et al.*, *LPS XL*, #1786, (2009). [4] Westphal *et al.*, *AIP Conf. Proc.* **1221**, 131, (2010). [5] D. E. Gruber *et al.* ApJ 520, 124-129 (1999). [6] P. Oger *et al.*, Spectrochim. Acta Part B 63, 512–517, (2008). [7] R.F. Egerton *et al.* Micron 35, 399-409 (2004). [8] L. Lemelle *et al.*, Geochim. Cosmochim. Acta 67, 1901–1910 (2003).