Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

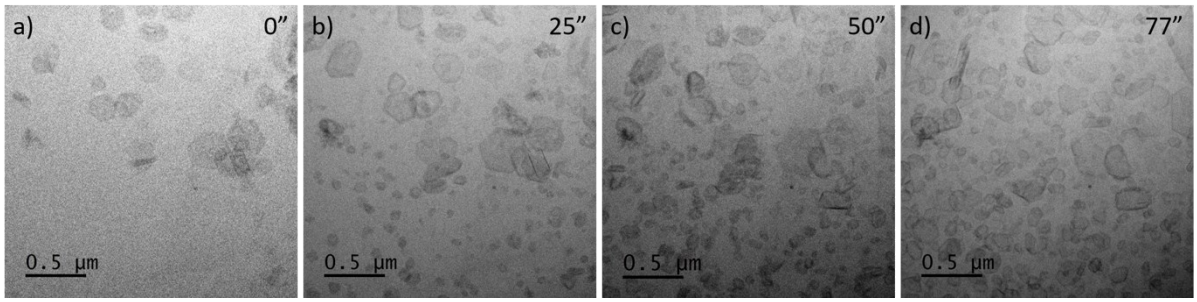

Figure S1: a) First frame of the S6 video showing hexagonal crystals, frames at b) 25 s, c) 50 s and d) 77 s showing the development of the existing hexagonal features and nucleation events in areas where no features were observed initially. Dose rate: 201 $e^-/Å^2/s$.

Upon further irradiation with the electron beam the existing hexagonal features continued to grow (figure 2) while nuclei began to appear and grow into hexagonal crystals also. The existing crystals and nuclei underwent similar processes with a linear growth trend upon continued irradiation however the existing crystals tended to degrade after ~40 s of illumination. This was not observed for all existing crystals as it would depend on their stability, local interacting environment and propagation of radiolysis products in its vicinity.

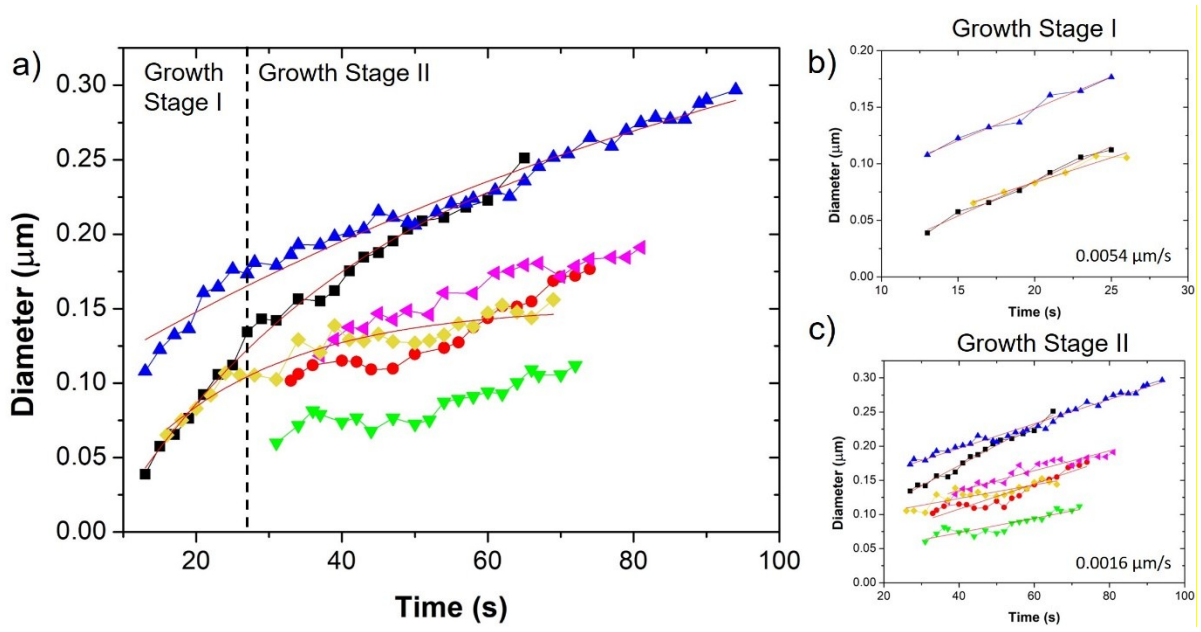

Figure S2: Graphs showing time dependent sizing measurements of six particles. a) The particles measured from their inception follows an exponential trend (blue, black and yellow) exhibiting a higher rate of growth. A tangent made along the exponential fit identified two stages of growth which enabled quantification of the rate from the slope of a linear fit. b) At the first stage of growth quantified the rate was 0.0054 μm/s, and c) for growth stage two a slower rate was calculated of 0.0016 μm/s, approximately five times slower.

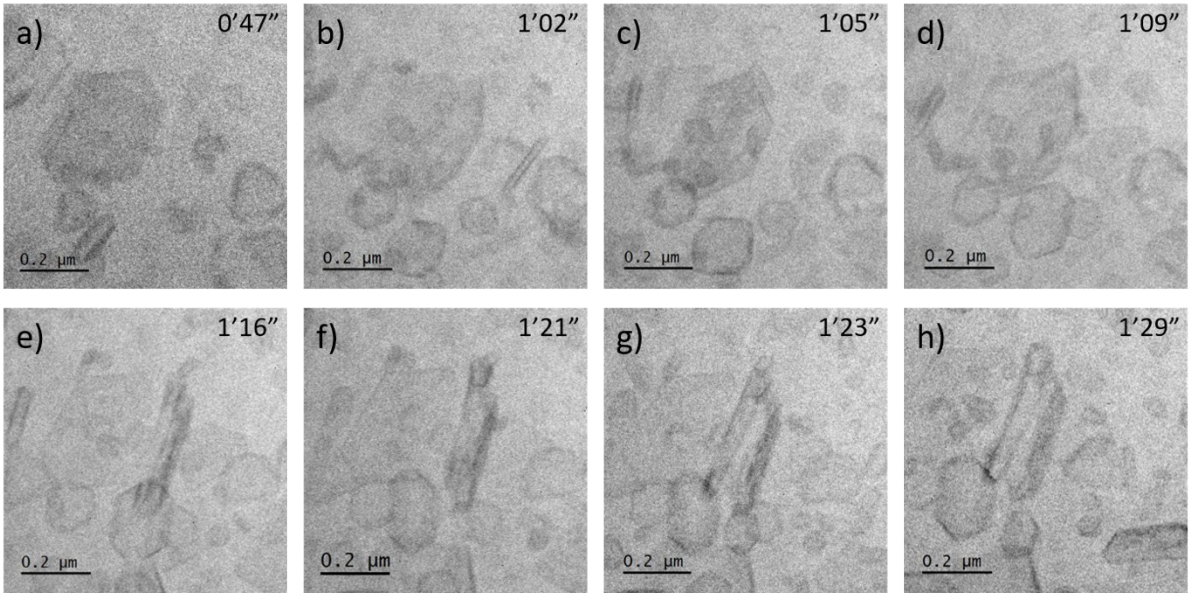

Figure S3: Sequence of images over a period of 42 s showing the dynamics of a hexagonal shaped crystal initially appearing as one crystal, revealing itself as three similarly shaped stacked crystals undergoing subsequent delamination. Dose rate: 201 $e^-/Å^2/s$.

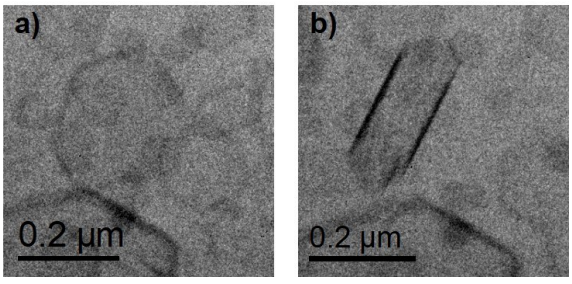

Figure S4: a) A hexagonal shaped crystal of FFA in planar view measuring ca. 301 µm in diameter at 1’27’’ and b) the same crystal rotated at 1’34’’ displaying a longitudinal view with a thickness of ca. 101 µm.

A particular example of the available area between the two windows of the liquid cell in figure S4 shows a crystal of hexagonal shape measuring approximately 300 nm in diameter, rotating to its side. This would suggest that at that position in the liquid cell the available area between the windows is at least 300 nm.

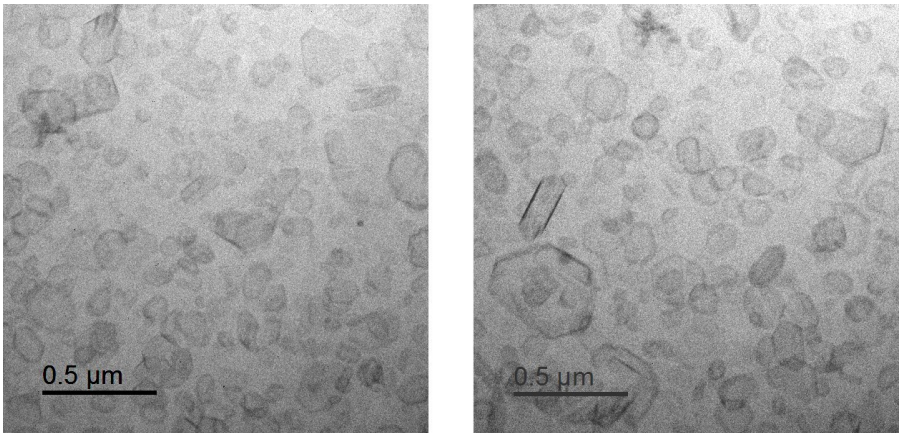

Figure S5: a) The final frame in the same area of observation from 0 s, b) the stage was re-positioned to view an area that was not under illumination showing larger features. Dose rate: 201 $e^{-}/Å^{2}/s$.

After approx. 1 min of irradiation after which many smaller crystals had grown and filled the area, at 1’20” the stage was moved to reveal an area that was not under complete illumination. The new area in view contained a mixture of much larger crystals of hexagonal and truncated triangular structures (figure 7a and 7b). The existence of particles in this new area was expected, however, their increased size and sparsity compared to the area under continuous illumination from the onset of observation to 1’20”, is suspected to be due to the depletion of reagents to the area despite the system being under constant flow. It is theorised that the usage of the reagents to nucleate and grow the crystals appear much faster than the supply of reagents to the area.

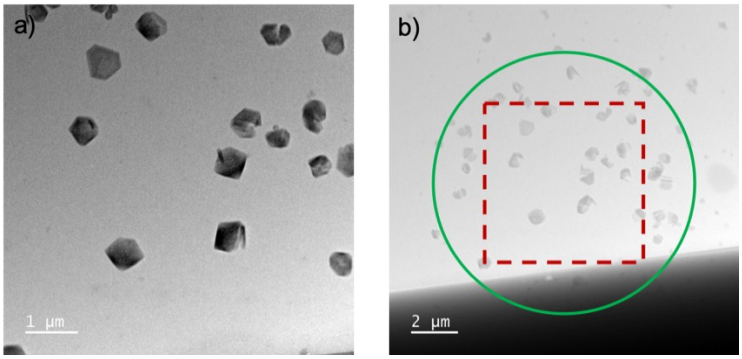

Figure S6: a) Final frame taken from a video showing particle growth acquire at 9.2kx magnification, b) magnification was reduced to 4.1kx to reveal additional particles that had grown outside the field of view but inside the diameter of the electron beam, other particles have also grown which were not in the field of view nor within the diameter of the electron beam.

Video S7 FFA Particle Growth: Video acquired using GMS3 coupled with the Gatan OneView camera. The camera was operating in in situ mode with a frame rate of 20 fps in 2k x 2k view. The video shows the development of FFA nuclei and crystals over a period of 1'34''. Frames from this video were used for figure 2. Dose rate: 201 $e^-/Å^2/s$.

Video S8 Particle Delamination: Video acquired using GMS3 coupled with the Gatan OneView camera. The camera was operating in in situ mode with a frame rate of 20 fps in 2k x 2k view. The video shows in more detail the delamination and appearance of needle-shaped particles that are hexagonal particles on their side. Frames from this video was used for figure 3. Dose rate: 201 $e^-/Å^2/s$.

Video S9 Particle Dissolution: Video acquired using GMS3 coupled with the Gatan OneView camera. The camera was operating in in situ mode with a frame rate of 20 fps in 2k x 2k view. This video shows the dissolution of a formed hexagonal molecular crystal degrading in gas phase under constant irradiation. Frames from this video was used for figure 5. Dose rate 488 $e^-/Å^2/s$.