Supporting Information:

Pd Data Analysis and Quantification:

The JEOL JEM-2100F/Cs STEM was operated at an average current density of 50 pA using a 3 μs dwell time, giving a dose of 53 $e^-/Å^2$ at x800k magnification (4.2 Å/pixel), 13.2 $e^-/Å^2$ at x400k magnification (8.4 Å/pixel), and 3.3 $e^-/Å^2$ at x200k magnification (16.8 Å/pixel). Supporting Movie 1, Supporting Movie 2 and Supporting Movie 3 were thresholded to isolate the Pd particles from the background. A threshold value was chosen to give the best agreement by visual inspection between the black and white thresholded movie frames and the original grayscale frames. Quantitative particle area measurements (figures 2A and 2B) were then taken as the total area above the threshold value in the view area at each frame. Growth rates and growth areas were normalized to a dose of 1 $e^-/Å^2$ to directly compare growth rates at different magnifications (different doses). For the pure aqueous Pd salt solution, shown in figure 2A (x200k mag), the initial growth rate during nucleation is $2.36\pm0.06\text{x}10^{-3}$ $\mu m^4/e^-s$ (0-10 seconds), the second stage growth rate is $3.12\pm0.06\text{x}10^{-3}$ $\mu m^4/e^-s$ (10-50 seconds), and the final stage growth rate is $1.08\pm0.01\text{x}10^{-3}$ $\mu m^4/e^-s$ (50-120 seconds) after nucleation has ceased. For the Pd salt/Brij 56 templated mixture, shown in figure 2B (x800k mag), the average growth rate is $8.94\pm0.02\text{x}10^{-7}$ $\mu m^4/e^-s$. Growth rates were calculated by linear least squares fits to the experimental growth area *vs*. time plots in figures 2A and 2B. Error bars were calculated from plots of the residuals between the experimental data and least square fits.

Pd cluster migration distances and rates were calculated from visual inspection of Supporting Movie 4 raw data (no speed modification). Distance and time measurements were taken for six different Pd clusters (four of which appear in figure 6B-I, highlighted with colored circles) and are tabulated in Supporting Table 1 below. All migration distances

measured were relative to the ~25 nm diameter spherical shaped particle in the center of the view area (the stage was translated in x and y during the experiment to center this particle in the field of view).

Supporting Table 1: Migration distance, time, and rate for six different Pd clusters in Supporting Movie 4. All clusters are between 10 and 25 nm in diameter and the average migration rate is ~0.6 nm/s.

Cluster Diameter (nm)	Distance Traveled (nm)	Time (seconds)	Migration Rate (nm/s)
20-25	~60	120	0.5
15-20 (Green circle in Fig 4)	~22	36	0.61
15-18 (Yellow circle in Fig 4)	~35	32	1.09
12-15 (Blue circle in Fig 4)	~27	56	0.48
12-15 (Red circle in Fig 4)	~39	64	0.61

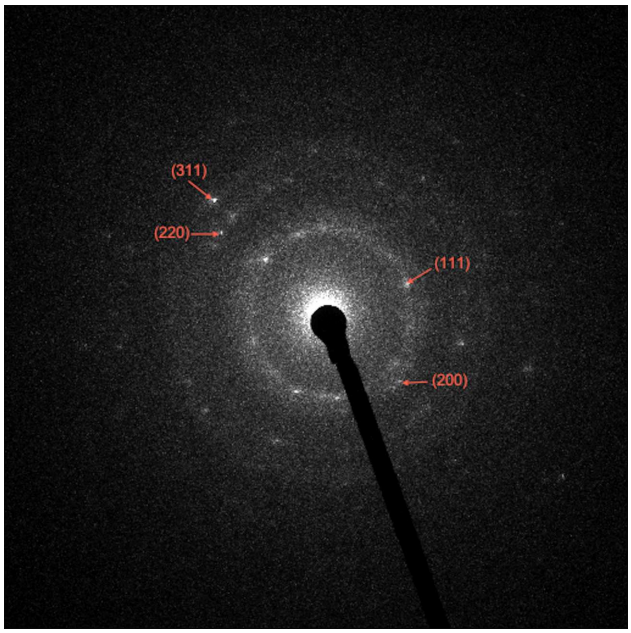
10-12	~14	28	0.5

Electron Beam Heat Effect Calculations:

All heating calculations follow the equations and methodology described previously.[18] For the highest dose condition used in our experiments (x5M magnification, supporting movie 3), the electron beam was operated at an acceleration voltage of 200 keV, a current density of J_e=42,228 A/m^2, and a scan area of 1184 nm^2. For the lowest dose condition used (x200k magnification, supporting movie 1), the electron beam was operated at an acceleration voltage of 200 keV, a current density of J_e=67.6 A/m^2, and a scan area of 739,875 nm^2. For all dose conditions, we use an average energy loss of (dE/dX)=500 eV per electron (from EELS measurements of the Pd salt/Brij 56 paste samples in the liquid stage), a total energy dissipation of $\Delta E=2.5\times10^{-8}$ W, and a silicon nitride film thickness (h) of 100 nm. We calculate a heat transfer flux density of $J=2.11\times10^{14}$ W/m^3 and an electron beam induced rise in temperature of ΔT=1.87˚C for the highest dose condition. For the lowest dose condition, we calculate a heat transfer flux density of $J=3.38\times10^{11}$ W/m^3 and a rise in temperature of ΔT=1.36˚C induced by the e-beam.

Electron Diffraction Data of Pd Particles:

Electron diffraction has been performed on the exact same samples used during *in situ* growth experiments (sample from supporting movies 3 and 4), shown in supporting figure 1. Diffraction patterns of the particles grown in the paste sample show crystalline rings, with spacings that correspond to the palladium lattice constant. This diffraction data provides evidence that metallic Pd nanoparticles have been grown during *in situ* experiments.



Supporting Figure 1: Electron diffraction pattern of particles grown *in situ* by partial ascorbate reduction and e-beam induced reduction in the Brij 56 paste (same sample from supporting movies 3 and 4). Spacing of rings corresponds with the palladium lattice constant, verifying that metallic Pd was grown during *in situ* experiments.

Supporting Movie Captions:

Supporting Movie 1: *In situ* growth movie of e-beam induced reduction of the pure aqueous Pd salt solution (no surfactant template). Dark field movie on left, bright field movie on right. The movie was acquired at x200k magnification, and is ~80 seconds in real time. The movie has been adjusted to x4 speed for the first 8 seconds, and x8 speed for the last 6 seconds.

Supporting Movie 2: DF *in situ* growth movie of e-beam induced reduction of the surfactant templated Pd salt/Brij 56 mixture. The movie has been adjusted to x24 speed for the first 7 seconds, and x48 speed for the last 15 seconds, and is ~15 minutes in real time. The movie begins at x400k magnification, and after 9 seconds is increased to x800k magnification. The magnification is dropped back to x400k for the final few frames of the movie. When the beam is turned on initially (start of the movie) and when the magnification is adjusted there is a several frame delay for the scale bar to refresh and adjust to the appropriate size.

Supporting Movie 3: *In situ* movie of Pd sintering event in the partially ascorbate reduced Brij 56 templated mixture (DF left, BF right). The movie has been adjusted to x8 speed, and is ~2 minutes in real time. The movie begins at x1.5M and after 3 seconds, the magnification is increased to x5M. During the first 3 seconds of the movie the scale bar is displaying the wrong value (it has not updated for x1.5M mag). When the magnification is increased to x5M the scale refreshes to become the proper size.

Supporting Movie 4: *In situ* movie of Pd cluster migration and agglomeration in the partially ascorbate reduced Brij 56 templated mixture (DF left, BF right). The movie has been adjusted to x16 speed, and is ~7.5 minutes in real time. The movie begins at x1M magnification, and after 4 seconds the magnification is increased to x1.2M, then after another 7 seconds the magnification is increased to x2M. When the magnification is adjusted there is a slight delay (several frames) for the scale bar to adjust to the appropriate size.