Supporting Information

Crystallization via Oriented Attachment of Nanoclusters with Short Range Order in Solution

Hao Su,[1,2] Paul H.H. Bomans,[1,2] Heiner Friedrich,[1,2,3] Yifei Xu[1,2,4] and Nico Sommerdijk[5]*

1. Laboratory of Materials and Interface Chemistry and Centre for Multiscale Electron Microscopy, Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, PO box 513, 5600 MB, Eindhoven, The Netherlands
2. Institute for Complex Molecular Systems, Eindhoven University of Technology, PO Box 513, 5600 MB, Eindhoven, The Netherlands
3. Laboratory of Physical Chemistry and Centre for Multiscale Electron Microscopy, Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, PO Box 513, 5600 MB, Eindhoven, The Netherlands
4. School of Chemistry, University of Leeds, Woodhouse Lane, LS2 9JT, Leeds, UK
5. Department of Biochemistry, Radboud Institute of Molecular Life Sciences, Radboud University Medical Center, Geert Grooteplein 6525 GA Nijmegen, The Netherlands

Supplementary Figures

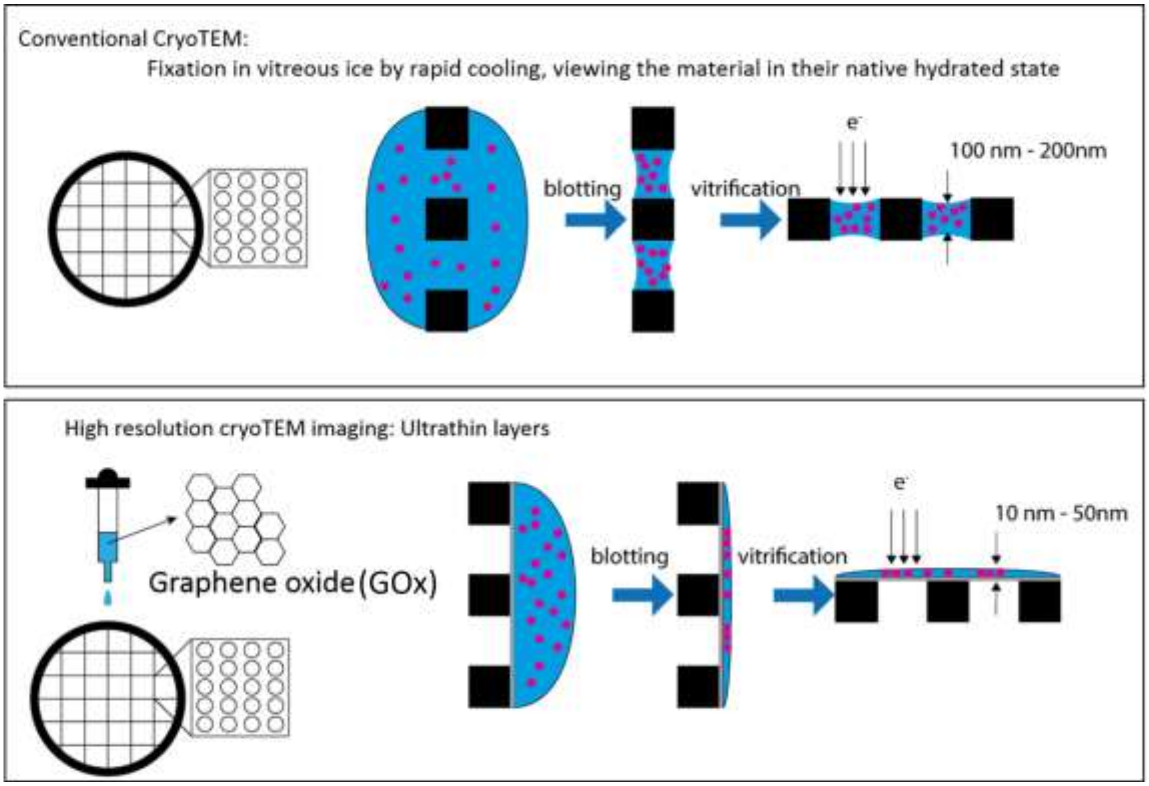

Figure S1 Scheme comparing conventional (top) and high-resolution (bottom) cryo-TEM sample preparation methods. For conventional cryoTEM observations, the solution sample is applied on TEM grid coated with a holey carbon film. The sample is then blotted and vitrified, generating 100-200 nm thick ice layers within the holes of the carbon film. This method does not introduce a direct contact between the sample particles and the carbon film, thus minimizes impact to the products. For high resolution studies, graphene oxide (GOx) layers are applied on top of the holey carbon film as mechanical support. This allows the liquid thickness to be reduced to a few tens of nanometers, resulting in improved contrast and resolution in cryoTEM, which is needed for reliably detecting small objects as investigated in this study. Although the cobalt complex/clusters might be directly contacting with the GOx layers in this case, they will unlikely react with the GOx and change their morphology. Furthermore, as shown in Figure S2, similar results can be observed without using the GOx method. It is noteworthy that the GOx method is only suitable for samples that are thinner than 50 nm.

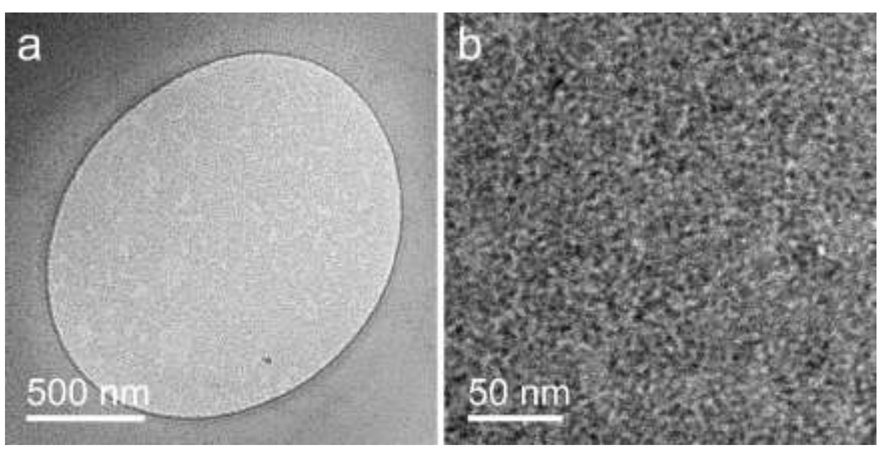

Figure S2 CryoTEM images of t=0 min sample applied on lacey carbon film cryoTEM grid (conventional method), in (a) lower and (b) higher magnifications, respectively. It can be seen that the ~0.8 nm size complexes are also visible when using this method, but the resolution and contrast are much lower in this case.

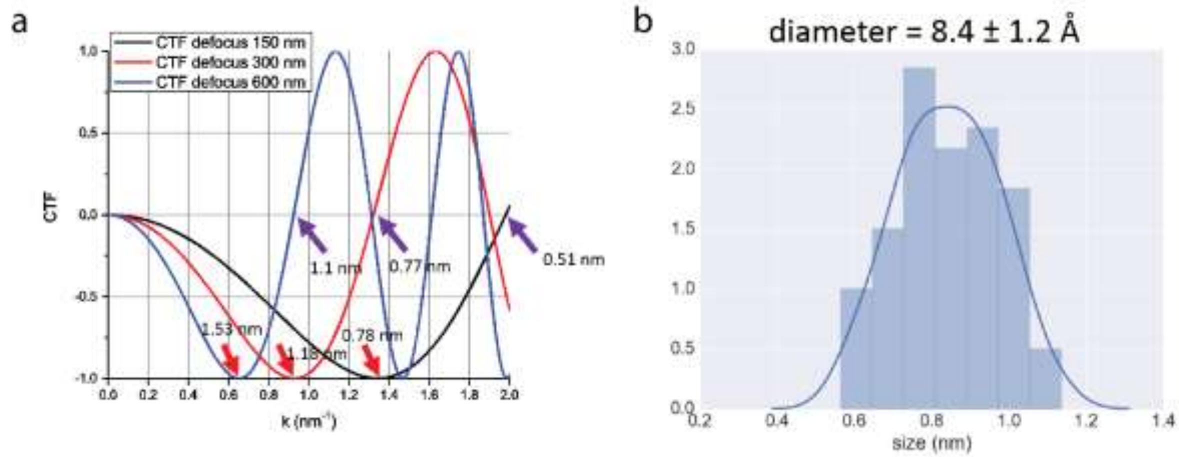

Figure S3 (a) Contrast transfer function (CTF) of cryoTitan at defocus -150 nm, -300 nm, and -600 nm. Purple arrows indicate the point resolution at a certain nominal defocus. Red arrows show the diameter of objects that has the highest contrast at the given defocus value. Imaging of the complex was done at -150 nm nominal defocus, where the frequency corresponding to the object with 0.8 nm diameter having the highest phase transfer. (b) Particle size distribution of the 0 min sample calculated from manually measuring around 80 clearly visible objects in the cryoTEM images.

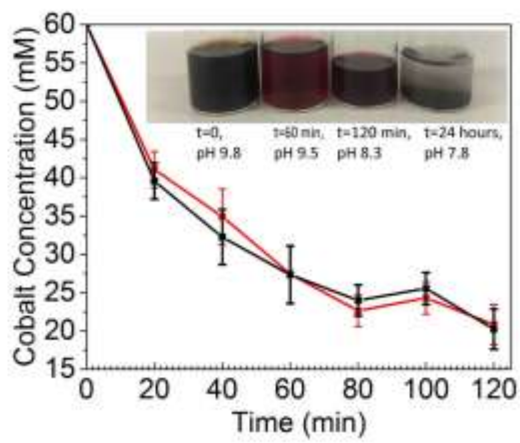

Figure S4 The color change of the solution (top) and concentration of the cobalt (II) ions obtained from UV-Vis absorption spectra by analyzing the signals at 525 nm (red) and 375 nm (black) using Lambert-Beer's law. Original UV-Vis spectra and calibration curve are shown in Figure S5.

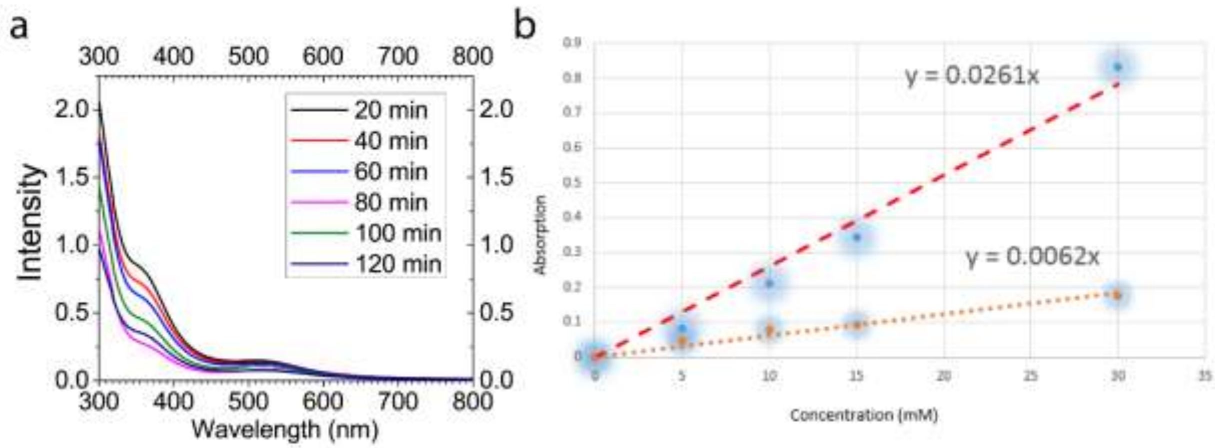

Figure S5 (a) UV-Vis absorption spectra at different time points. (b) Calibration curve for analysis of cobalt (II) concentration from UV-Vis signals at 525 nm (top) and 375 nm (bottom).

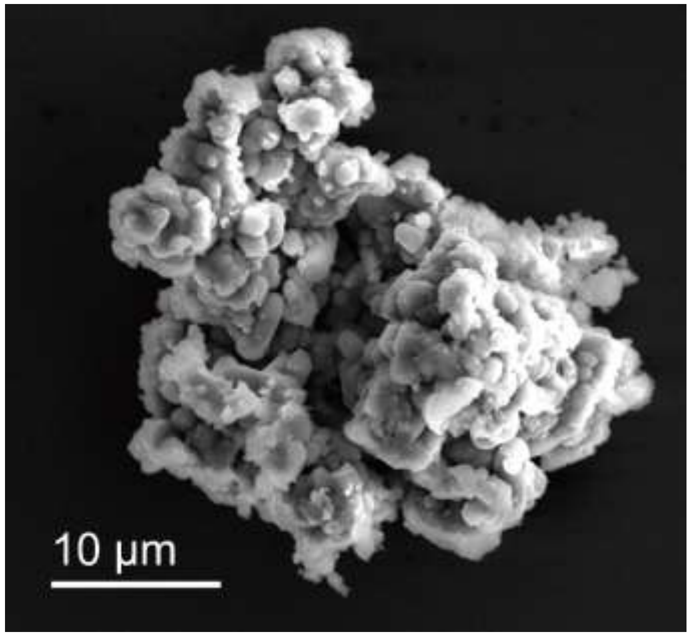

Figure S6 Scanning electron microscopy (SEM) image of the product collected after 24 hours of reaction.