Supplemental Material (Appendix)

Observation of Weibull, Lognormal, and Gamma distributions in electrodeposited Cu and Cu-Ag particles

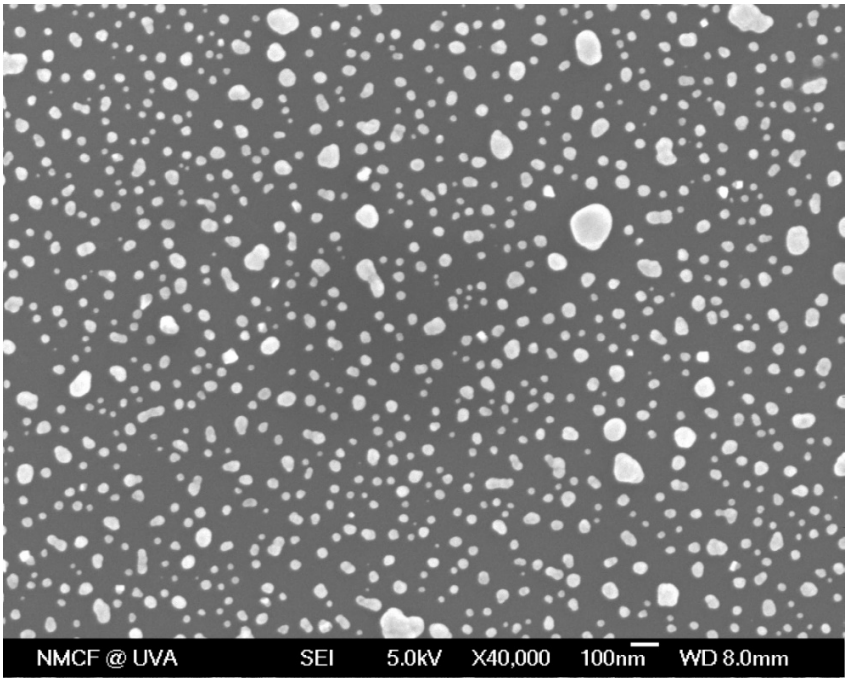
Yunkai Sun[a*], Giovanni Zangari[a]

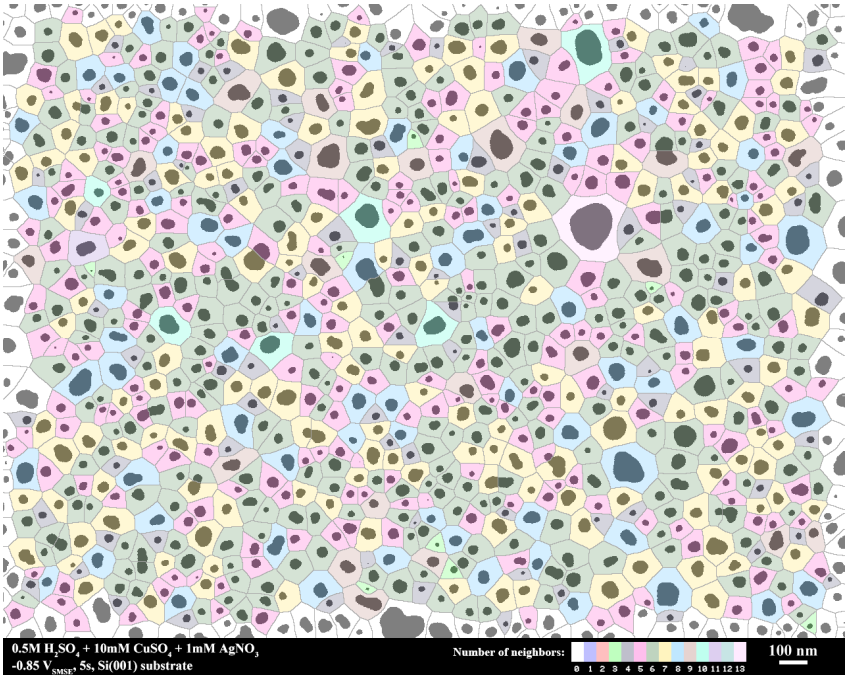
a. Department of Materials Science and Engineering, University of Virginia, Charlottesville, VA, 22904, USA

* Corresponding authors. Yúnkāi Sūn, Email: ys3ej@virginia.edu; Giovanni Zangari, Email: gz3e@virginia.edu

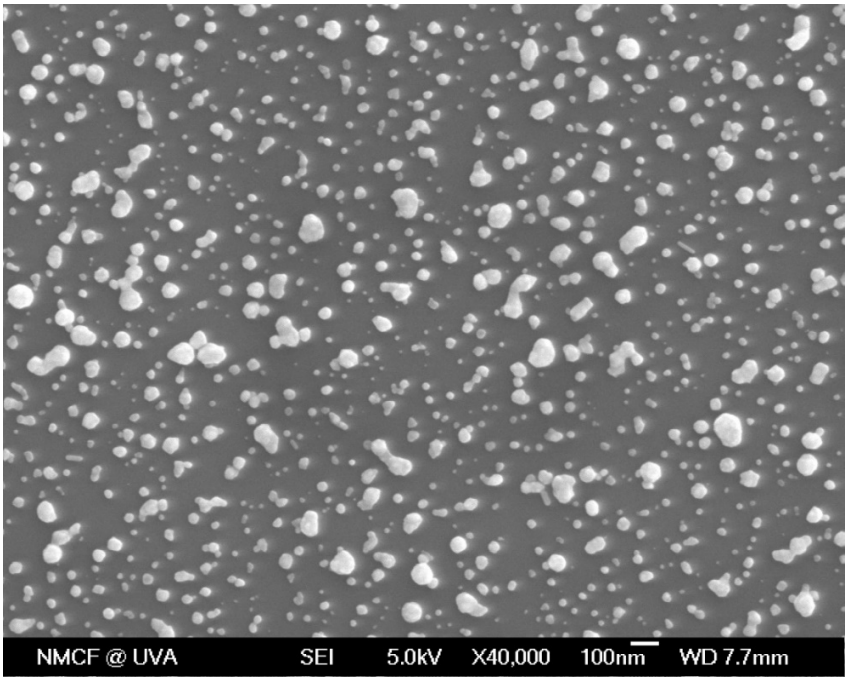
Appendix-A SEM images and Voronoi cells of the grains

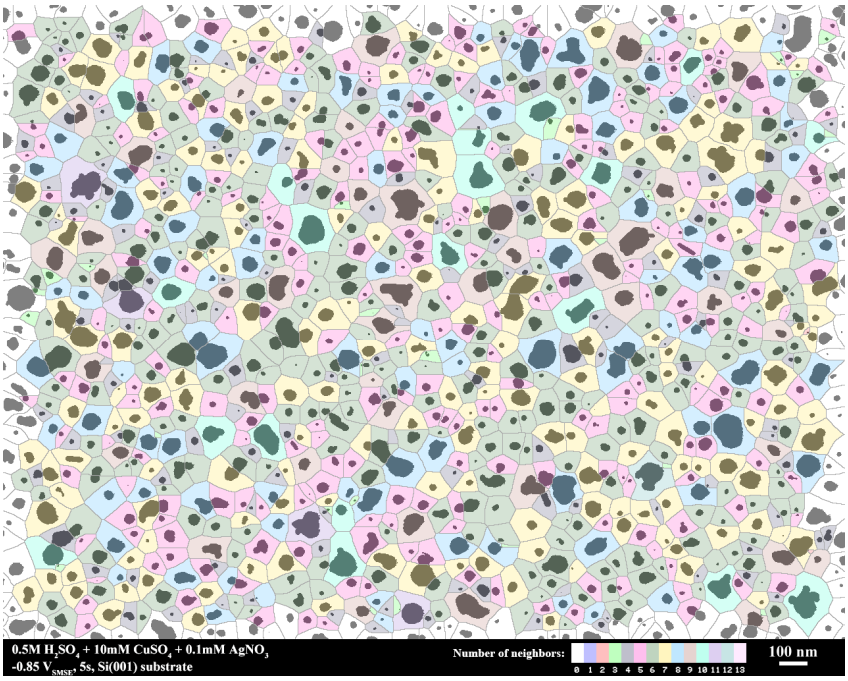
1mM Ag(I) + 10mM Cu(II), -0.85 V_{SHE}, 5s



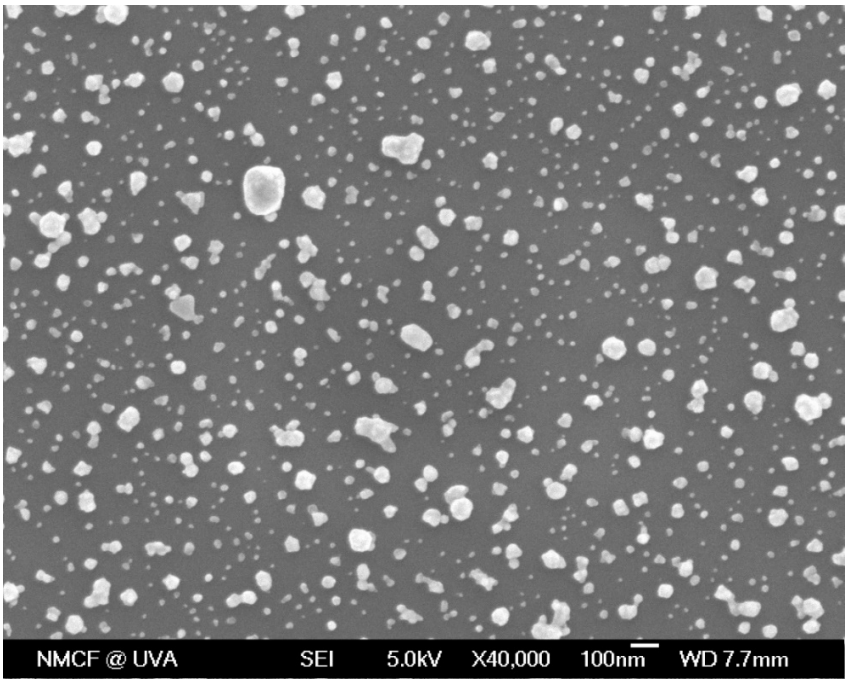


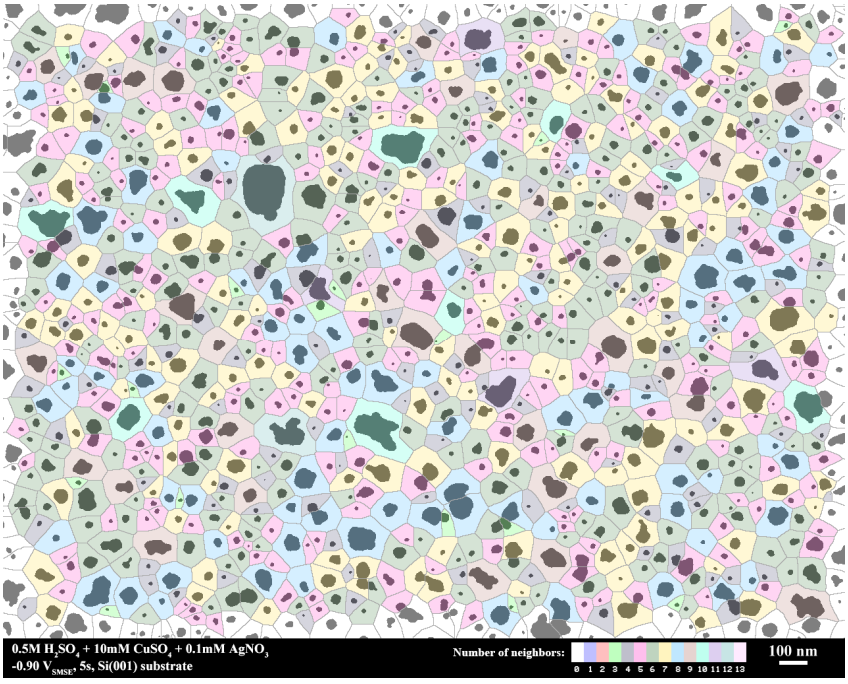
0.1mM Ag(I) + 10mM Cu(II), -0.85 V_{SHE}, 5s


NMCF @ UVA
SEI
5.0kV
X40,000
100nm
WD 7.7mm

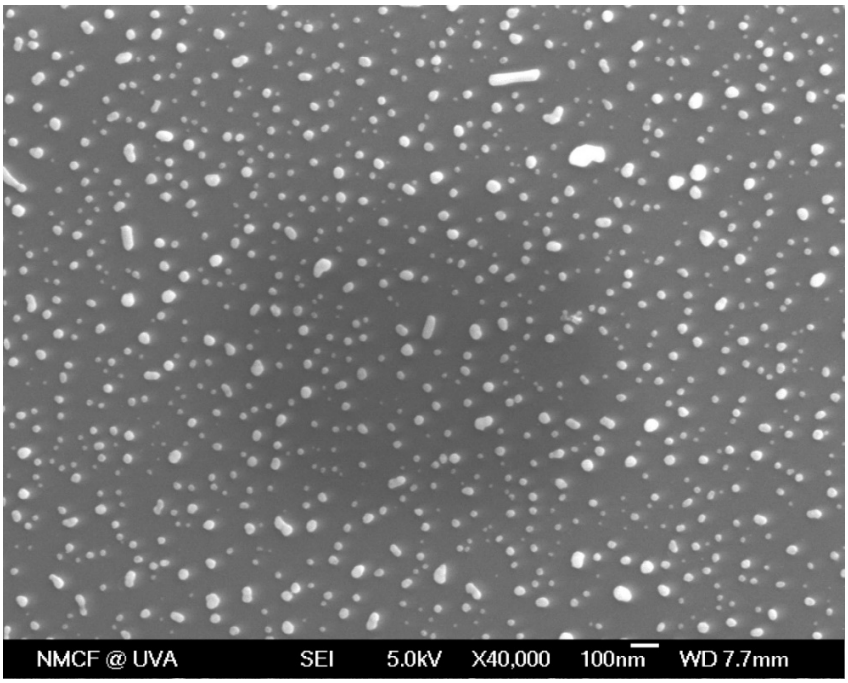

0.5M H2SO4 + 10mM CuSO4 + 0.1mM AgNO3
-0.85 VSMSE, 5s, Si(001) substrate
Number of neighbors:
0 1 2 3 4 5 6 7 8 9 10 11 12 13
100 nm

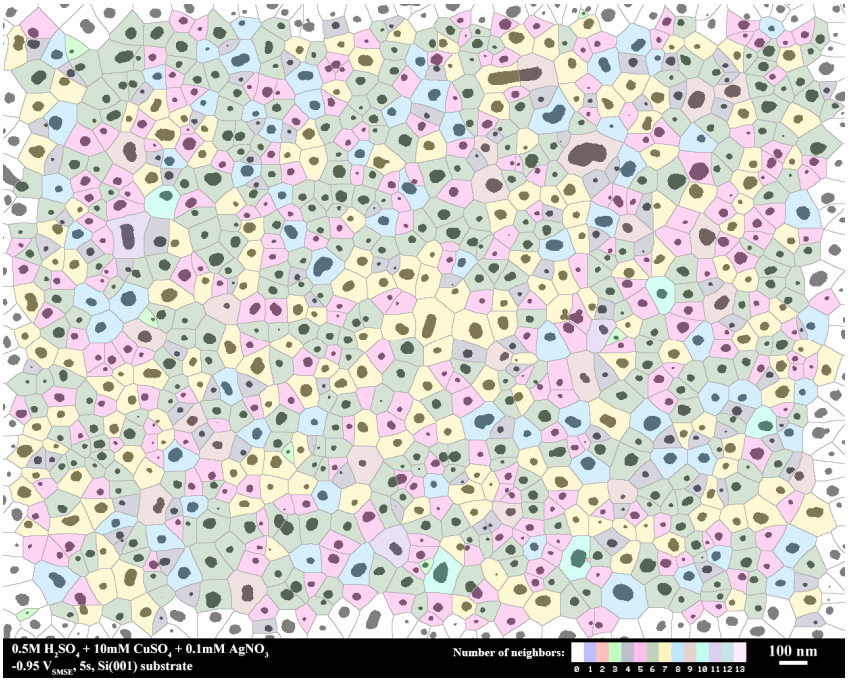
0.1mM Ag(I) + 10mM Cu(II), -0.90 V_{SHE}, 5s



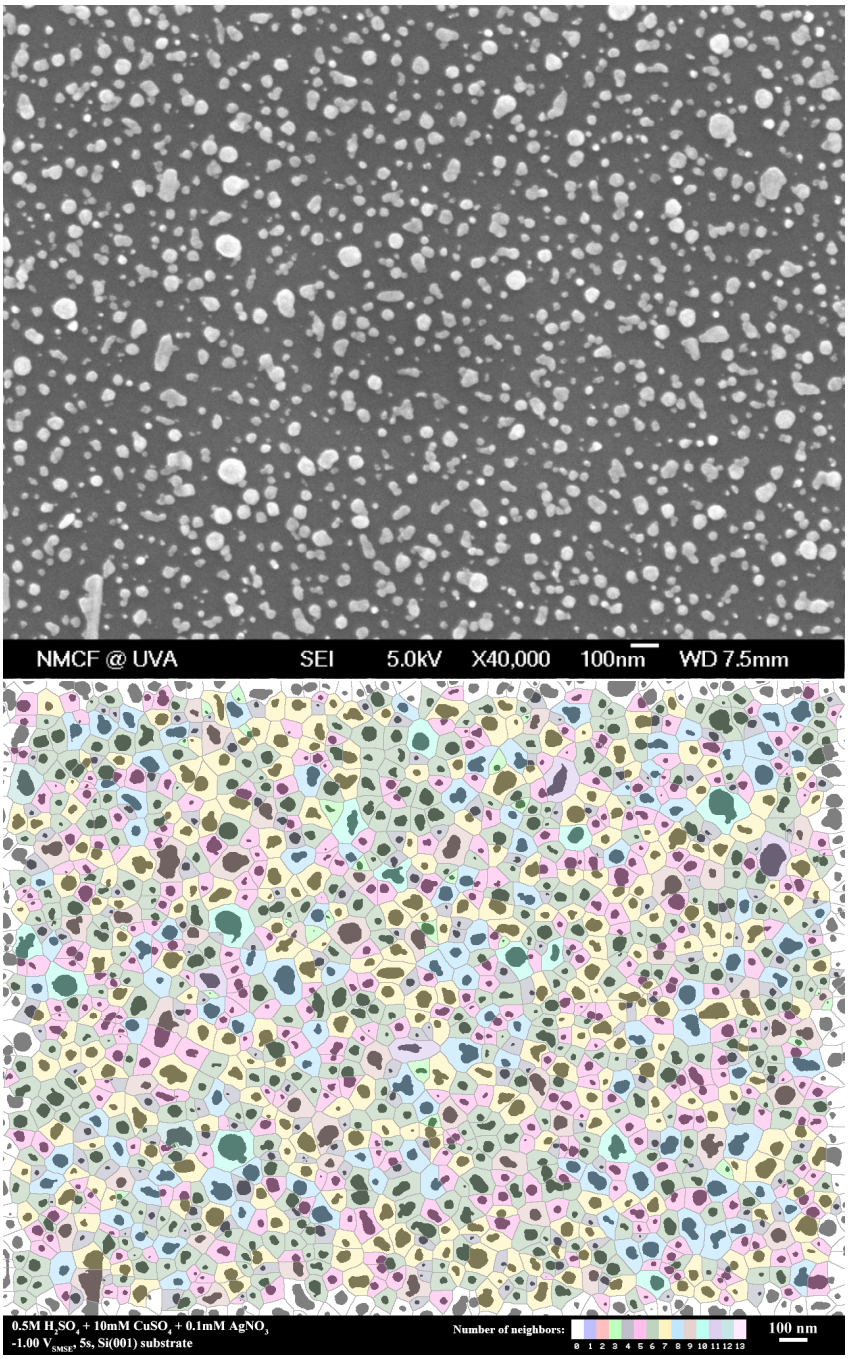


0.1mM Ag(I) + 10mM Cu(II), -0.95 V_{SHE}, 5s

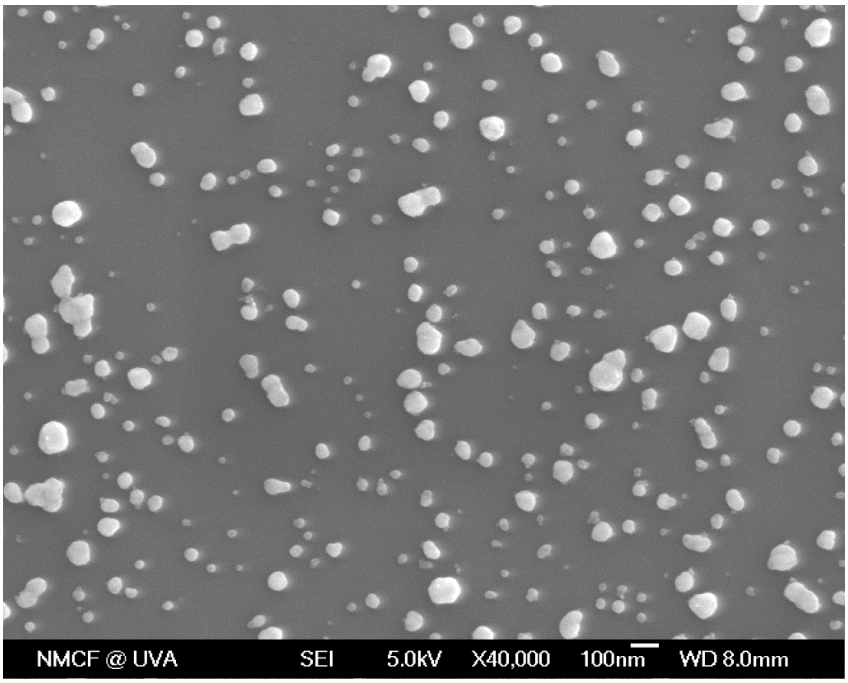


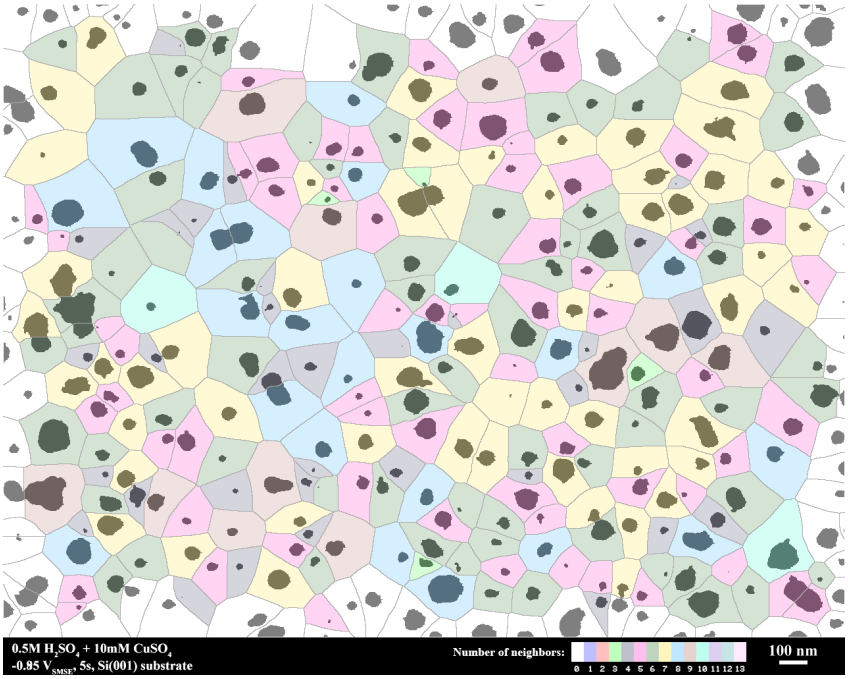


0.1mM Ag(I) + 10mM Cu(II), -1.00 V_{SHE}, 5s

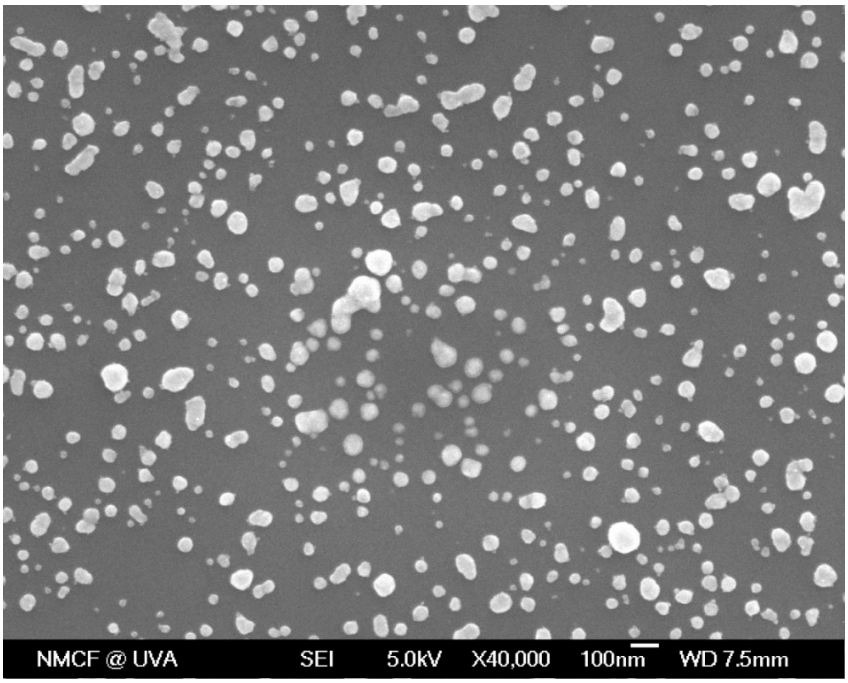


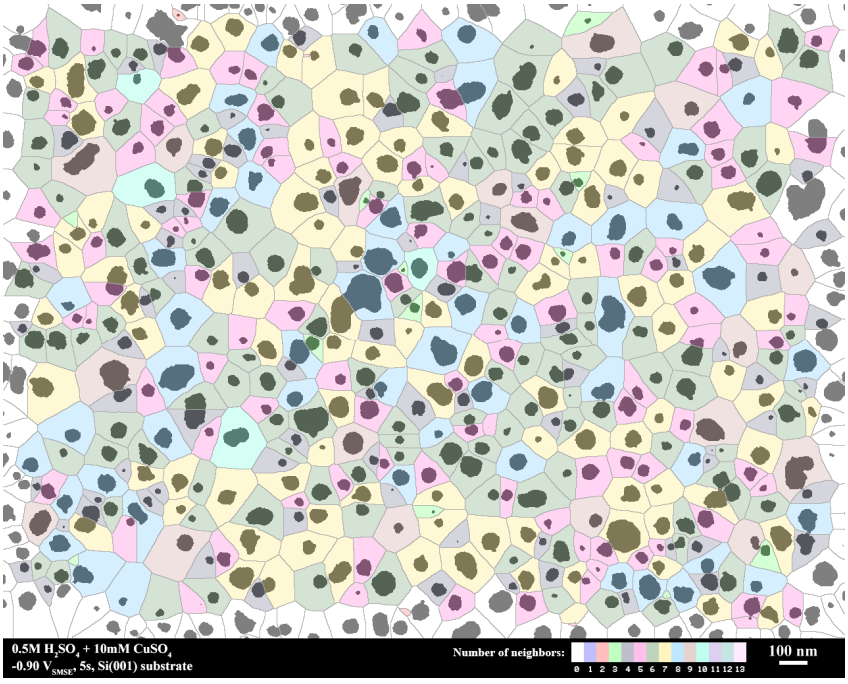
10mM Cu(II), -0.85 V_{SHE}, 5s



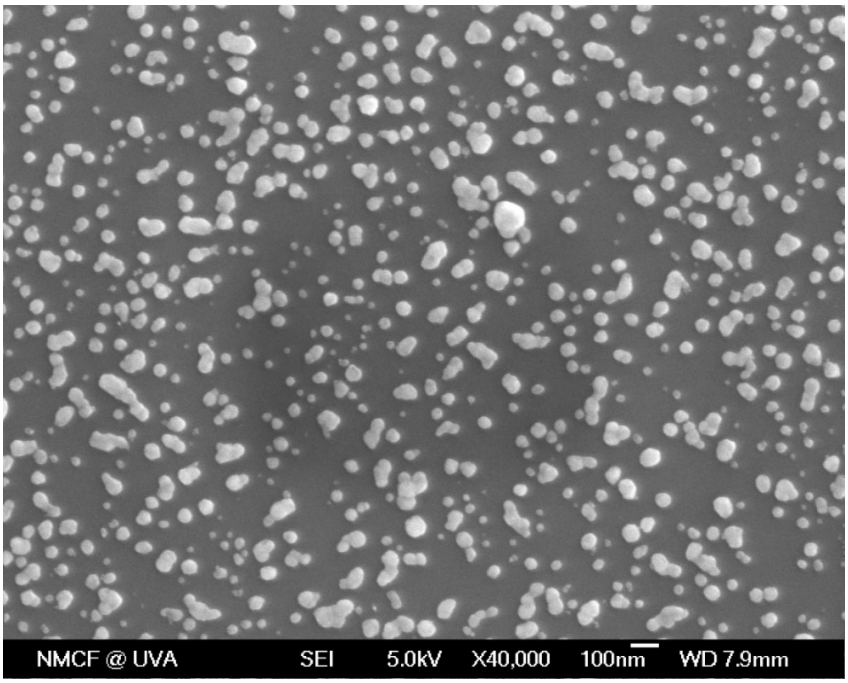


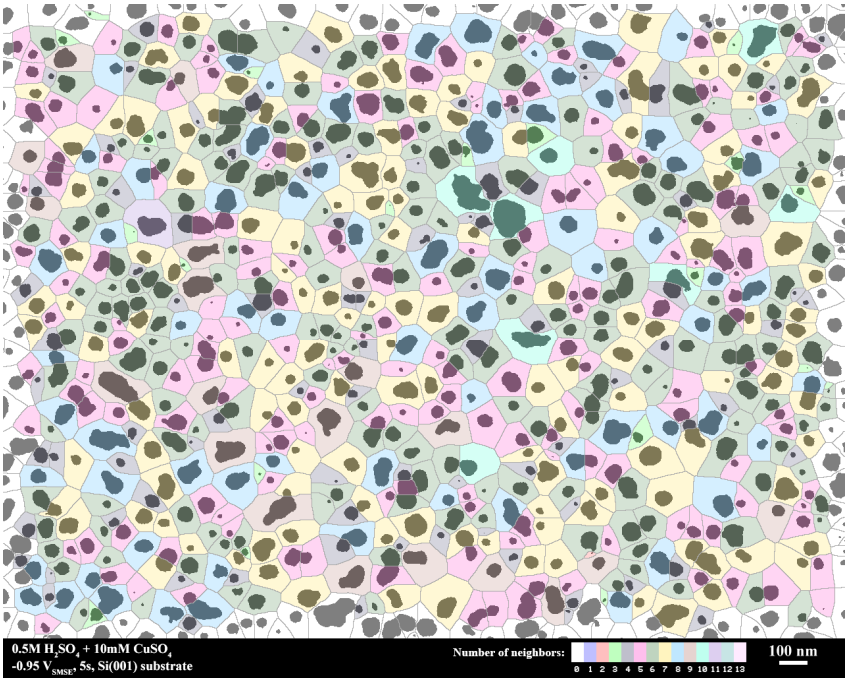
10mM Cu(II), -0.90 V_{SHE}, 5s



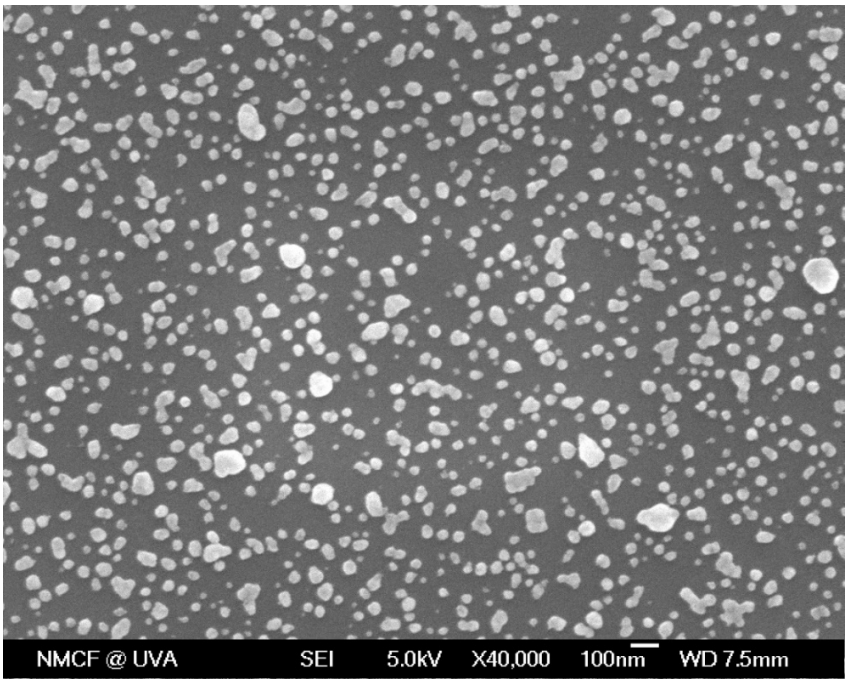


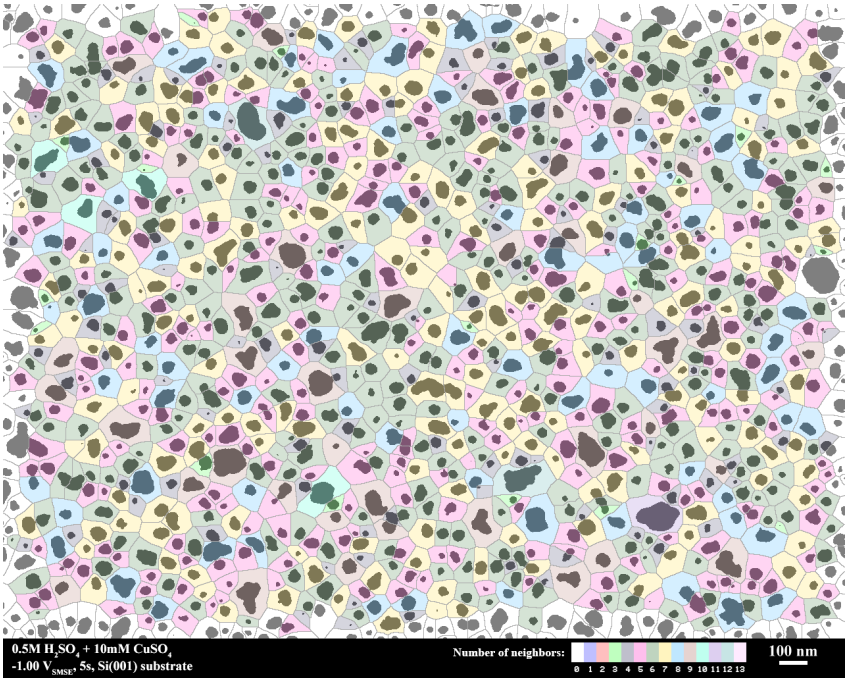
10mM Cu(II), -0.95 V_{SHE}, 5s





10mM Cu(II), -1.00 V_{SHE}, 5s





Appendix-B Comments on the Generalized Gamma distribution, the Lognormal distribution, the Weibull distribution, and the Gamma distribution

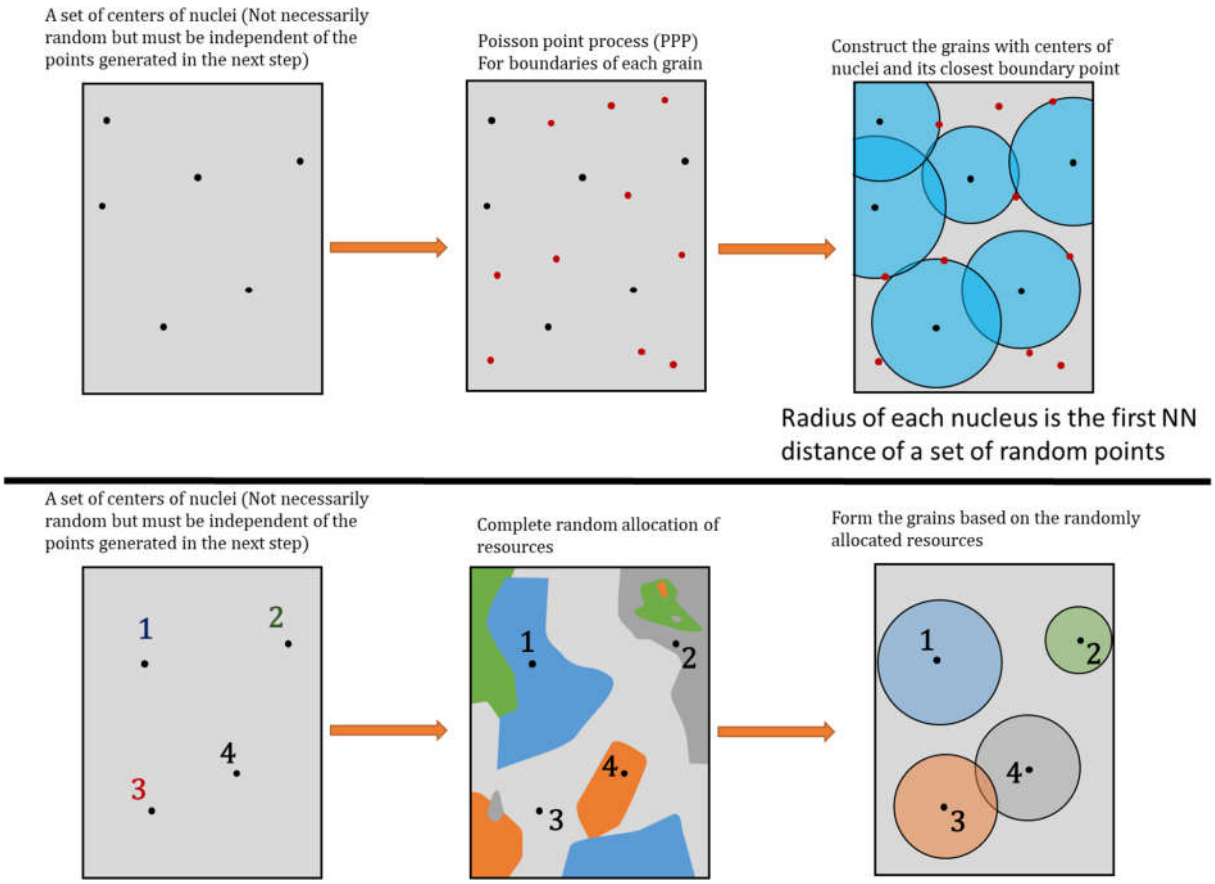


Fig. B1 Generation of Weibull distribution. Top figure indicates the random radius scenario, which should fulfill $k = 2$. Bottom figures indicate the random allocation of resources scenario, which should also fulfill $k = 2$.

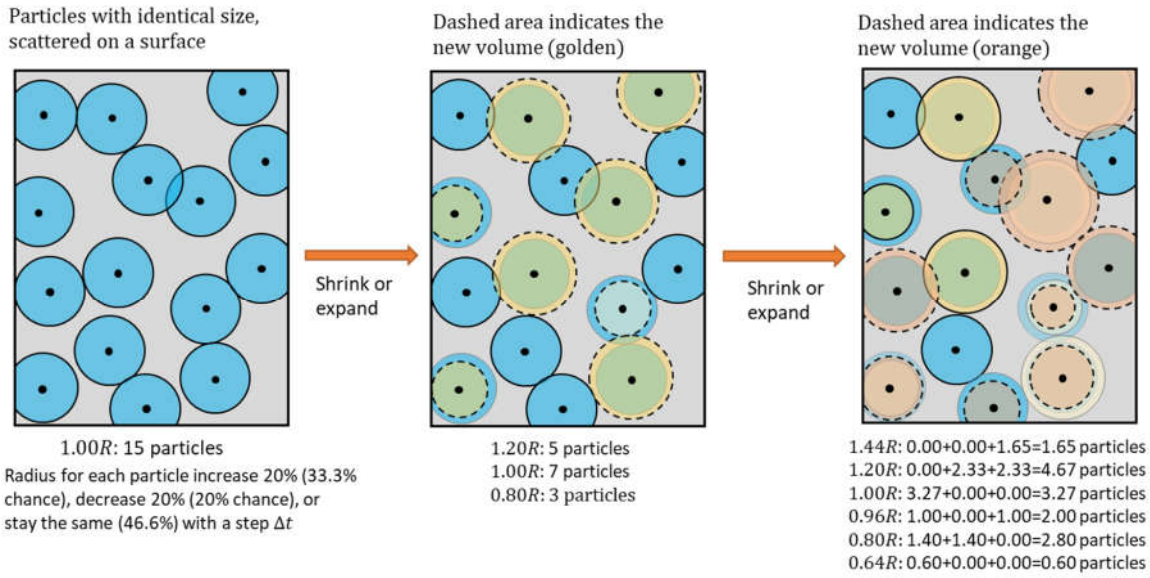


Fig. B2 Illustration of the law of proportionate with a hypothetical scenario of 2D overlapping circles, and random but time-independent growth rate. At the long-time limit, the distribution of these circles should fulfill the Lognormal distribution, with moments related to the randomness of the grow/shrink operation. At each step, $\langle \ln T \rangle = 0.333 \times \ln 1.2 + 0.2 \times \ln 0.8 = 0.016 > 0$. Therefore, the average radius of the particles (and thus the total area of the particles) will gradually increase. The dispersion of each growth step is $\langle (\ln T - \langle \ln T \rangle)^2 \rangle = 0.021$. When the step n is large, this system should fulfill lognormal distribution with $\mu = n\langle \ln T \rangle = 0.016n$ and $\sigma^2 = n\langle (\ln T - \langle \ln T \rangle)^2 \rangle = 0.021n$. Details in **Appendix-A**.

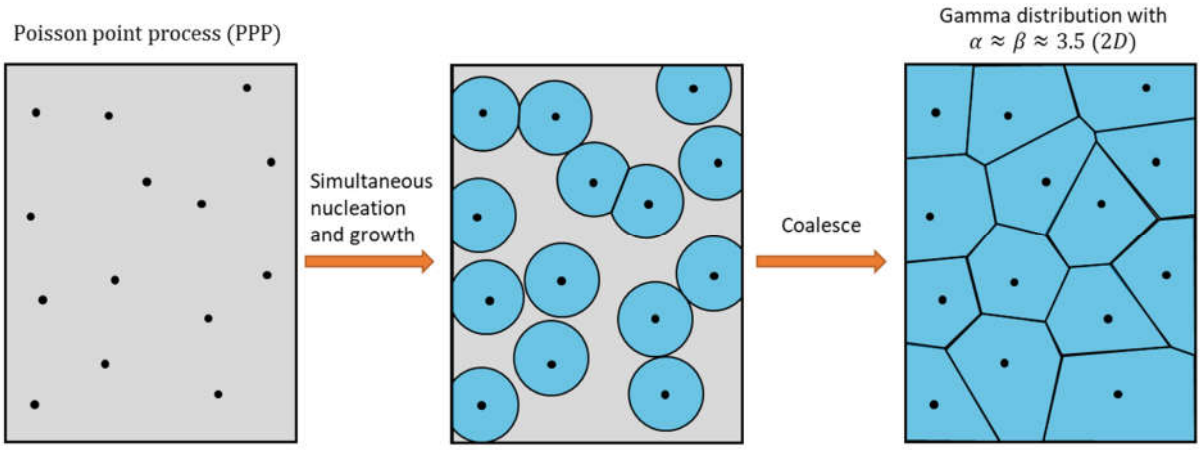


Fig. B3 Illustration on one of the scenarios where Gamma distribution could represent the final grain size distribution. The empirical results for a 2D complete spatially random nuclei is a Gamma distribution of the area of the cells with $\alpha \approx \beta \approx 3.5$.

Appendix-C Comments on the Generalized Gamma distribution, the Lognormal distribution, the Weibull distribution, and the Gamma distribution

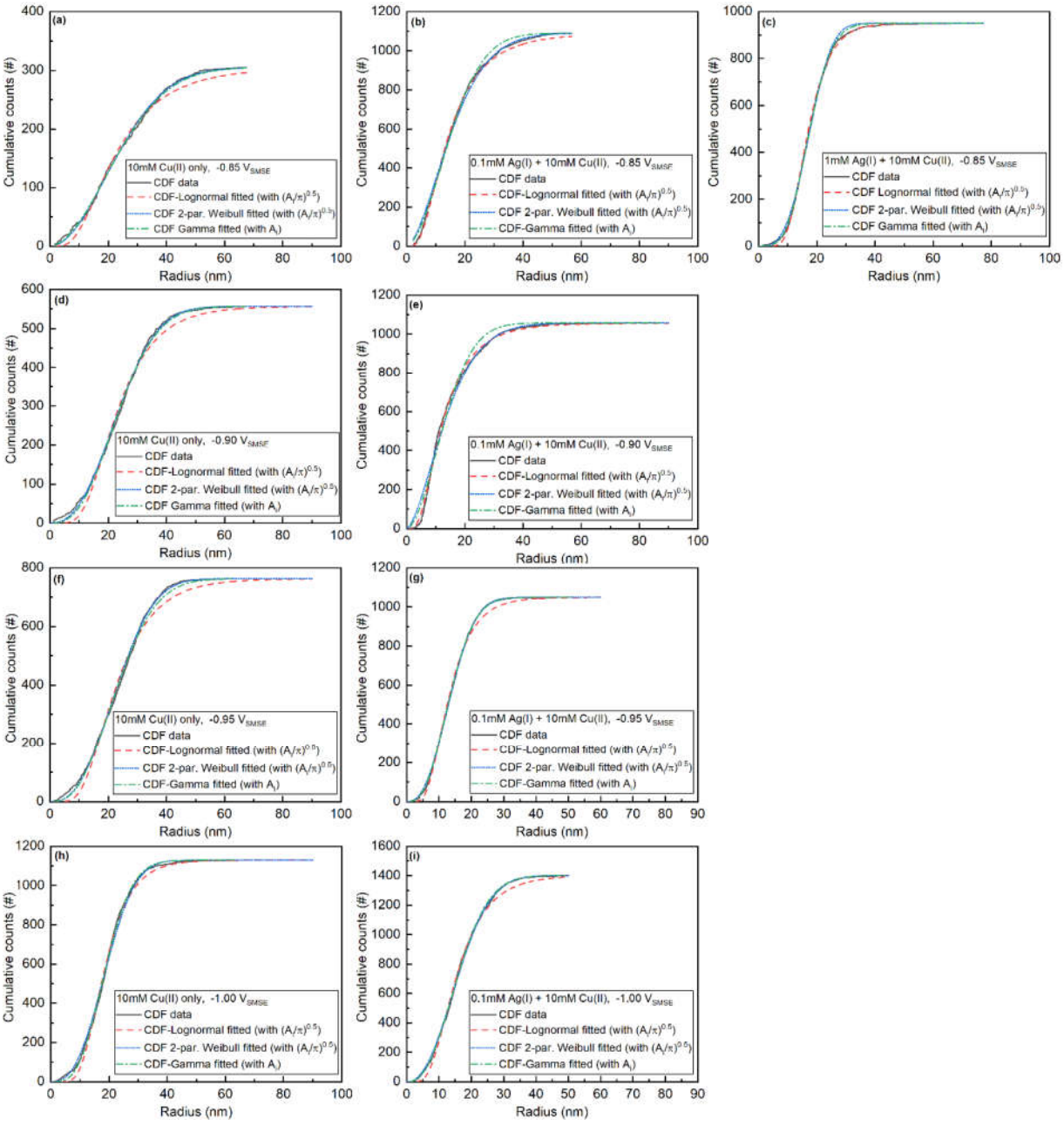


Fig.C CDF profile of the grain size distributions for the SEM images in **Fig.4**.

Appendix-D: Statistical characteristics of the observed features

Particle size (in radius)	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	24.39	13.01	0.49	-0.13
-0.90V 10mM Cu(II)	23.88	10.81	0.43	0.67
-0.95V 10mM Cu(II)	23.23	10.05	0.20	-0.06
-1.00V 10mM Cu(II)	19.24	8.01	0.79	2.13
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	16.42	9.93	1.12	1.10
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	14.75	9.45	1.77	5.29
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	13.76	5.98	0.71	1.42
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	16.45	7.74	0.71	0.75
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	17.94	7.15	1.48	7.03

Voronoi cell size (in radius)	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	76.46	21.06	0.12	-0.13
-0.90V 10mM Cu(II)	56.40	16.31	0.06	0.19
-0.95V 10mM Cu(II)	48.18	13.22	0.13	0.01
-1.00V 10mM Cu(II)	39.62	9.79	0.53	0.83
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	39.82	12.44	0.51	0.65
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	40.53	12.17	1.21	3.09
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	41.02	9.67	0.49	0.74
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	35.30	9.74	0.55	0.66
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	43.35	10.47	1.00	3.91

1st NN distance	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis

-0.85V 10mM Cu(II)	87.01	28.43	0.50	0.96
-0.90V 10mM Cu(II)	68.24	19.89	0.42	0.17
-0.95V 10mM Cu(II)	60.73	15.38	0.57	0.12
-1.00V 10mM Cu(II)	51.92	11.74	0.16	0.16
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	50.05	15.87	0.45	0.20
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	50.85	14.64	0.60	0.44
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	51.92	14.58	0.40	-0.09
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	45.87	12.90	0.48	0.39
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	58.13	14.43	0.75	1.23

2nd NN distance	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	114.98	31.97	0.74	0.66
-0.90V 10mM Cu(II)	88.58	21.79	0.46	0.03
-0.95V 10mM Cu(II)	77.99	17.13	0.33	0.15
-1.00V 10mM Cu(II)	64.89	12.38	0.26	-0.11
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	66.01	15.78	0.28	-0.28
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	65.90	15.96	0.51	0.16
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	67.21	14.36	0.28	-0.30
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	59.33	13.01	0.32	0.19
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	71.96	14.88	0.49	0.83

5th NN distance	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis

-0.85V 10mM Cu(II)	180.03	31.88	0.11	0.21
-0.90V 10mM Cu(II)	137.45	23.46	0.28	-0.31
-0.95V 10mM Cu(II)	118.93	19.40	0.12	-0.46
-1.00V 10mM Cu(II)	97.52	13.57	0.11	-0.18
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	101.14	14.78	0.11	-0.14
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	100.85	17.02	0.13	-0.12
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	99.72	14.12	0.27	0.16
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	88.71	12.19	0.11	0.21
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	105.15	15.43	0.43	0.98

10th NN distance	Mean (nm)	Standard deviation (nm)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	256.05	29.50	0.55	0.79
-0.90V 10mM Cu(II)	191.04	23.26	0.14	-0.48
-0.95V 10mM Cu(II)	165.90	19.02	-0.12	0.50
-1.00V 10mM Cu(II)	137.27	12.41	0.09	0.36
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	140.43	14.29	0.07	-0.31
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	140.38	17.06	0.24	-0.22
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	139.50	14.20	0.39	-0.24
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	123.68	12.66	-0.02	-0.14
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	147.09	16.46	0.34	0.13

Coordination number	Mean (#)	Standard deviation (#)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	6.004	7.287	0.079	-1.068
-0.90V 10mM Cu(II)	5.919	10.638	-0.007	-1.184
-0.95V 10mM Cu(II)	5.953	11.589	0.064	-1.133
-1.00V 10mM Cu(II)	5.956	12.832	0.141	-0.793
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	5.989	14.326	0.141	-1.160
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	5.945	13.323	0.116	-1.029
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	5.951	11.965	0.065	-0.958
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	5.952	15.083	0.065	-1.020
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	5.984	10.611	0.115	-0.616

Voronoi cell occupancy	Mean ($\%_{area}$)	Standard deviation ($\%_{area}$)	Skewness	Kurtosis
-0.85V 10mM Cu(II)	11.81	9.15	1.07	0.91
-0.90V 10mM Cu(II)	19.14	11.20	0.58	0.14
-0.95V 10mM Cu(II)	24.75	13.45	0.40	-0.05
-1.00V 10mM Cu(II)	24.37	11.82	0.35	0.01
-0.85V 0.1mM Ag(I) + 10mM Cu(II)	16.55	11.66	0.92	0.89
-0.90V 0.1mM Ag(I) + 10mM Cu(II)	13.32	9.61	1.17	1.45
-0.95V 0.1mM Ag(I) + 10mM Cu(II)	11.53	6.29	0.79	0.99
-1.00V 0.1mM Ag(I) + 10mM Cu(II)	21.92	11.47	0.37	-0.37
-0.85V 1.0mM Ag(I) + 10mM Cu(II)	17.13	6.75	0.31	0.12

Appendix-E Summary of fitting results

The cumulative distribution function (CDF) of the Lognormal distribution for feature R, with two fitting parameters (γ_L, k_L):

$$F(R) = \frac{N_{tot}}{2}\left(1 + \mathrm{erf}\left(\frac{k_L \ln(R/\gamma_L R_0)}{\sqrt{2}}\right)\right)$$

The CDF of the Weibull distribution for feature R, with two fitting parameters (γ_W, k_W):

$$F(R) = N_{tot}\left(1 - \exp\left(-\left(\frac{R}{\gamma_W R_0}\right)^{k_W}\right)\right)$$

The CDF of the Gamma distribution for feature A_R, with two fitting parameters (α', γ_G):

$$F(A_R) = \frac{N^{tot}}{\Gamma(\alpha')}\left(\frac{\gamma_G}{A_0}\right)^{\alpha'} \int_o^{A_R} A^{\alpha'-1} \exp\left(-\frac{\gamma_G A}{A_0}\right) dA$$

Note that R_0 and A_0 are the average value of the feature R. Using average radius R as an example, R_0 is defined as:

$$R_0 = \frac{\sum_i R_i}{N^{tot}}$$

Where N^{tot} is total number of particles identified in the SEM image.

E1. Particle size

Table-E1a Fitting results of the particle size distribution of the deposits in **Fig.9**

Particle size	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.12987	1.97089	0.90767	1.69584	0.90760	0.92953
10mM Cu(II), -0.90V	1.12542	2.43890	0.95418	2.18174	1.42000	1.43376
10mM Cu(II), -0.95V	1.12792	2.54006	0.95873	2.15880	1.35661	1.41381
10mM Cu(II), -1.00V	1.11847	2.65641	0.95424	2.54298	1.94835	1.87777
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.10658	1.63926	0.85323	1.55664	0.96770	0.86407
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.08407	1.55333	0.82237	1.60110	1.09406	0.87531
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.12282	2.47259	0.93677	2.17052	1.45187	1.43831
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.12401	2.24908	0.92584	2.03861	1.31336	1.28070
10mM Cu(II) + 1mM Ag(I), -0.85V	1.08417	3.16925	0.93590	2.75953	2.34142	2.18333

Table-E1b Goodness of fitting (reflected by the value of reduced chi-square). Best-fitted results are highlighted in the table.

Particle size (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	76.82417	**17.54486**	**17.23666**
10mM Cu(II), -0.90V	314.40398	**33.70238**	**59.57201**
10mM Cu(II), -0.95V	678.58269	**85.73903**	**145.33532**
10mM Cu(II), -1.00V	704.44851	**175.32914**	**184.43372**
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**214.48689**	**179.32499**	337.10703
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**439.99152**	882.90216	1968.32421

10mM Cu(II) + 0.1mM Ag(I), -0.95V	400.91358	**39.46859**	**35.19554**
10mM Cu(II) + 0.1mM Ag(I), -1.00V	785.37822	**187.71873**	**194.52263**
10mM Cu(II) + 1mM Ag(I), -0.85V	**82.86434**	205.1378	**61.38359**

E2. Voronoi cell size

Table E2a Voronoi cell size fitted parameters

Voronoi cell size	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.05289	4.09502	0.94885	3.61167	3.44035	3.47088
10mM Cu(II), -0.90V	1.05154	4.05645	0.93161	3.62056	3.30820	3.33436
10mM Cu(II), -0.95V	1.05133	4.21264	0.93442	3.76194	3.61725	3.63806
10mM Cu(II), -1.00V	1.04224	4.75414	0.94301	4.15507	4.56086	4.51131
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.03378	3.80001	0.91514	3.32303	3.08137	3.00337
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.01579	4.03190	0.91605	3.44566	3.62628	3.39955
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.03866	4.91986	0.94825	4.34650	5.00855	4.91622
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.03940	4.14152	0.93085	3.73009	3.60812	3.54397
10mM Cu(II) + 1mM Ag(I), -0.85V	1.03547	5.00922	0.94511	4.17150	5.14593	5.02626

Table E2b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

Voronoi cell size (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	**12.5026**	**13.48914**	20.97627
10mM Cu(II), -0.90V	**4.06918**	22.23003	78.47933
10mM Cu(II), -0.95V	**3.07649**	41.92228	81.85295
10mM Cu(II), -1.00V	**0.50762**	153.71785	91.80609
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**1.21227**	300.57755	68.38255
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**0.26581**	979.38649	244.08844
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**0.20933**	463.80098	31.60001
10mM Cu(II) + 0.1mM Ag(I), -1.00V	**0.39089**	602.76994	79.01999
10mM Cu(II) + 1mM Ag(I), -0.85V	**0.25940**	248.90517	16.12252

E3. 1st nearest neighbor distance

Table E3a 1st nearest neighbor distance fitted parameters

1st nearest neighbor distance	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.07442	3.79652	0.96007	3.31323	3.19153	3.03407
10mM Cu(II), -0.90V	1.08401	3.89880	0.96985	3.36270	3.17277	3.13083
10mM Cu(II), -0.95V	1.07092	4.30908	0.96850	3.82405	4.00653	3.91415
10mM Cu(II), -1.00V	1.07188	5.07042	0.98463	4.46269	5.25076	5.22543
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.09141	3.55359	0.96632	3.08794	2.69432	2.66291
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.08170	3.89419	0.96704	3.37268	3.19299	3.13916
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.08255	3.93216	0.96950	3.45077	3.28200	3.24035
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.08349	4.01786	0.97187	3.47295	3.34836	3.31379
10mM Cu(II) + 1mM Ag(I), -0.85V	1.06616	4.57076	0.96897	4.03372	4.46957	4.33880

Table E3b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

1st nearest neighbor distance (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	**6.95421**	30.83670	**13.04475**
10mM Cu(II), -0.90V	46.43307	50.47370	**17.13337**
10mM Cu(II), -0.95V	**56.33470**	362.67955	**145.20994**
10mM Cu(II), -1.00V	**76.50000**	383.51553	**69.34010**
10mM Cu(II) + 0.1mM Ag(I), -0.85V	182.92772	161.42956	**40.17723**
10mM Cu(II) + 0.1mM Ag(I), -0.90V	123.35001	239.17917	**73.73904**
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**72.13299**	378.45089	**122.48618**
10mM Cu(II) + 0.1mM Ag(I), -1.00V	297.70254	330.83611	**105.23790**
10mM Cu(II) + 1mM Ag(I), -0.85V	**63.78937**	569.24604	213.37346

E4. 2nd nearest neighbor distance

Table E4a 2nd nearest neighbor distance fitted parameters

2nd nearest neighbor distance	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.07091	4.10578	0.96333	3.53895	3.56725	3.45576
10mM Cu(II), -0.90V	1.07267	4.48601	0.99625	3.95179	4.2292	4.16438
10mM Cu(II), -0.95V	1.06925	5.21588	0.98386	4.49580	5.38779	5.35998
10mM Cu(II), -1.00V	1.06311	5.94565	0.98822	5.11797	6.88030	6.86881
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.07748	4.53512	0.97870	3.99441	4.25542	4.24055
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.06908	4.66717	0.97454	4.08097	4.52443	4.44175
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.06948	5.09897	0.98215	4.46090	5.27564	5.25186
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.06944	5.16101	0.98331	4.50187	5.37672	5.34460
10mM Cu(II) + 1mM Ag(I), -0.85V	1.06522	5.45188	0.98364	4.76395	6.00192	5.95536

Table E4b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

2nd nearest neighbor distance (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	**6.51241**	18.83942	**6.31931**
10mM Cu(II), -0.90V	**16.01587**	109.6247	**32.22873**
10mM Cu(II), -0.95V	69.83068	83.54715	**27.12900**
10mM Cu(II), -1.00V	157.68637	207.43057	**57.21822**
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**83.11630**	371.20762	**101.73362**
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**42.24740**	386.37557	**91.34833**
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**52.64047**	352.10098	**59.70631**
10mM Cu(II) + 0.1mM Ag(I), -1.00V	**120.10369**	517.14598	**84.19707**
10mM Cu(II) + 1mM Ag(I), -0.85V	**48.88195**	225.06924	**29.13834**

E5. 5th nearest neighbor distance

Table E5a 5th nearest neighbor distance fitted parameters

5th nearest neighbor distance	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.06169	6.51987	0.99348	5.65419	8.36297	8.36532
10mM Cu(II), -0.90V	1.05837	6.30849	0.98678	5.49028	7.86605	7.83635
10mM Cu(II), -0.95V	1.05900	6.63327	0.99174	5.83908	8.72582	8.74774
10mM Cu(II), -1.00V	1.04415	8.59413	0.99452	7.07614	12.78786	13.21044
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.05051	7.85100	0.99459	6.79256	11.81234	11.82001
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.06002	6.65224	0.99259	5.73625	8.50450	8.53542
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.04824	7.92292	0.99238	6.95991	12.39202	12.35253
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.0463	8.56217	0.99524	7.51823	14.36227	14.34471
10mM Cu(II) + 1mM Ag(I), -0.85V	1.04563	8.02095	0.99076	6.97739	12.58901	12.48448

Table E5b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

5th nearest neighbor distance (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	11.25316	**4.16373**	**5.16774**
10mM Cu(II), -0.90V	**31.95991**	87.94815	**34.50869**
10mM Cu(II), -0.95V	**41.86595**	120.98114	**25.90731**
10mM Cu(II), -1.00V	**54.51746**	201.79084	**35.25198**
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**90.51654**	168.75814	**34.97838**
10mM Cu(II) + 0.1mM Ag(I), -0.90V	143.7591	116.78033	**63.31640**
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**33.14437**	260.67084	**35.23964**
10mM Cu(II) + 0.1mM Ag(I), -1.00V	**94.16697**	508.74290	**72.12255**
10mM Cu(II) + 1mM Ag(I), -0.85V	**22.55905**	259.41360	**25.99566**

E6. 10th nearest neighbor distance

Table E6a 10th nearest neighbor distance fitted parameters

10th nearest neighbor distance	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.03068	10.45879	0.98944	9.34230	22.34817	21.97758
10mM Cu(II), -0.90V	1.04347	8.80726	0.99322	7.84437	15.60722	15.56108
10mM Cu(II), -0.95V	1.04063	10.63479	0.99934	9.18780	21.31828	21.40040
10mM Cu(II), -1.00V	1.03116	13.31070	0.99838	11.58346	33.76866	33.77081
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.03735	11.0321	0.99733	9.52835	22.99701	23.02839
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.04159	9.21163	0.99365	8.02439	16.44656	16.39098
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.03097	11.04403	0.99155	9.72763	24.19991	23.93456
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.03809	11.06642	0.99823	9.59049	23.23152	23.29686
10mM Cu(II) + 1mM Ag(I), -0.85V	1.03884	9.90710	0.99396	8.58148	18.82291	18.75860

Table E6b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

10th nearest neighbor distance (R^2)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	**9.29948**	30.50684	**11.79879**
10mM Cu(II), -0.90V	**17.96297**	85.68206	**23.27756**
10mM Cu(II), -0.95V	48.33878	41.26070	**25.96019**
10mM Cu(II), -1.00V	**60.78702**	176.62761	**42.71459**
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**65.63527**	174.73055	**35.95656**
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**13.53730**	210.42230	**11.95216**
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**76.29233**	553.88154	**23.93456**
10mM Cu(II) + 0.1mM Ag(I), -1.00V	**108.68298**	267.53559	**46.63068**
10mM Cu(II) + 1mM Ag(I), -0.85V	**37.51865**	193.01249	**33.57950**

E7. Coordination number

Table E7a Coordination number fitted parameters

Coordination number	Weibull (#)		Lognormal (#)	
	γ_W	k_W	γ_L	k_L
10mM Cu(II), -0.85V	1.05243	4.52299	0.98847	3.73148
10mM Cu(II), -0.90V	1.07828	4.24416	1.00623	3.32568
10mM Cu(II), -0.95V	1.05863	4.50610	0.99549	3.90793
10mM Cu(II), -1.00V	1.04041	4.86615	0.98445	4.33032
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.04150	4.41768	0.98169	3.89484
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.04143	4.45299	0.97718	3.90686
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.04136	5.30097	0.99009	4.65095
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.04731	4.62329	0.98578	3.96212
10mM Cu(II) + 1mM Ag(I), -0.85V	1.03349	5.41587	0.98603	4.82462

Table E7b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

Coordination number (#)	Lognormal	Weibull
10mM Cu(II), -0.85V	**2.00561E-4**	3.74048E-4
10mM Cu(II), -0.90V	5.89064E-4	**2.81596E-4**
10mM Cu(II), -0.95V	**8.89230E-5**	1.96721E-4
10mM Cu(II), -1.00V	**2.38815E-5**	3.74904E-4
10mM Cu(II) + 0.1mM Ag(I), -0.85V	**4.97329E-5**	3.00103E-4
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**2.61589E-5**	4.38461E-4
10mM Cu(II) + 0.1mM Ag(I), -0.95V	**7.72067E-5**	2.87891E-4
10mM Cu(II) + 0.1mM Ag(I), -1.00V	**8.13826E-5**	2.43556E-4
10mM Cu(II) + 1mM Ag(I), -0.85V	**7.37872E-5**	3.64700E-4

E8. Voronoi cell occupancy

Table E8a Voronoi cell occupancy fitted parameters

Voronoi cell occupancy	Weibull (R)		Lognormal (R)		Gamma (R^2)	
	γ_W	k_W	γ_L	k_L	γ_G	α'
10mM Cu(II), -0.85V	1.09200	1.25809	0.77794	1.10347	1.38815	1.43862
10mM Cu(II), -0.90V	1.14749	1.7438	0.89458	1.49191	2.35974	2.48281
10mM Cu(II), -0.95V	1.15610	1.88019	0.91857	1.62022	2.71266	2.86440
10mM Cu(II), -1.00V	1.14272	2.23379	0.94268	1.94420	3.83393	3.99241
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1.12282	1.36463	0.81687	1.18282	1.56321	1.64387
10mM Cu(II) + 0.1mM Ag(I), -0.90V	1.08159	1.36545	0.78319	1.20198	1.68412	1.69500
10mM Cu(II) + 0.1mM Ag(I), -0.95V	1.12544	1.92729	0.89914	1.68549	3.00344	3.07514
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1.14935	1.90802	0.91346	1.64796	2.83528	2.96879
10mM Cu(II) + 1mM Ag(I), -0.85V	1.11416	2.85079	0.95775	2.48311	2.67300	2.72143

Table E8b Goodness of fitting (reflected by the value of reduced chi-square). Best fitted results are highlighted in the table.

Voronoi cell occupancy ($\%_{area}$)	Lognormal	Weibull	Gamma
10mM Cu(II), -0.85V	83.01969	**17.52204**	**23.56213**
10mM Cu(II), -0.90V	443.36252	**96.96739**	197.33850
10mM Cu(II), -0.95V	822.81991	**145.32177**	363.57820
10mM Cu(II), -1.00V	1153.98342	**111.66994**	502.23432
10mM Cu(II) + 0.1mM Ag(I), -0.85V	1225.90952	**113.03177**	261.30803
10mM Cu(II) + 0.1mM Ag(I), -0.90V	**240.22976**	428.13348	**278.25982**
10mM Cu(II) + 0.1mM Ag(I), -0.95V	510.59526	**27.99956**	113.48462
10mM Cu(II) + 0.1mM Ag(I), -1.00V	1990.49278	**222.02641**	758.58013
10mM Cu(II) + 1mM Ag(I), -0.85V	286.26469	**44.38022**	**84.55129**

Appendix-F Mean and standard deviation of the Voronoi cell occupancy and the coordination number

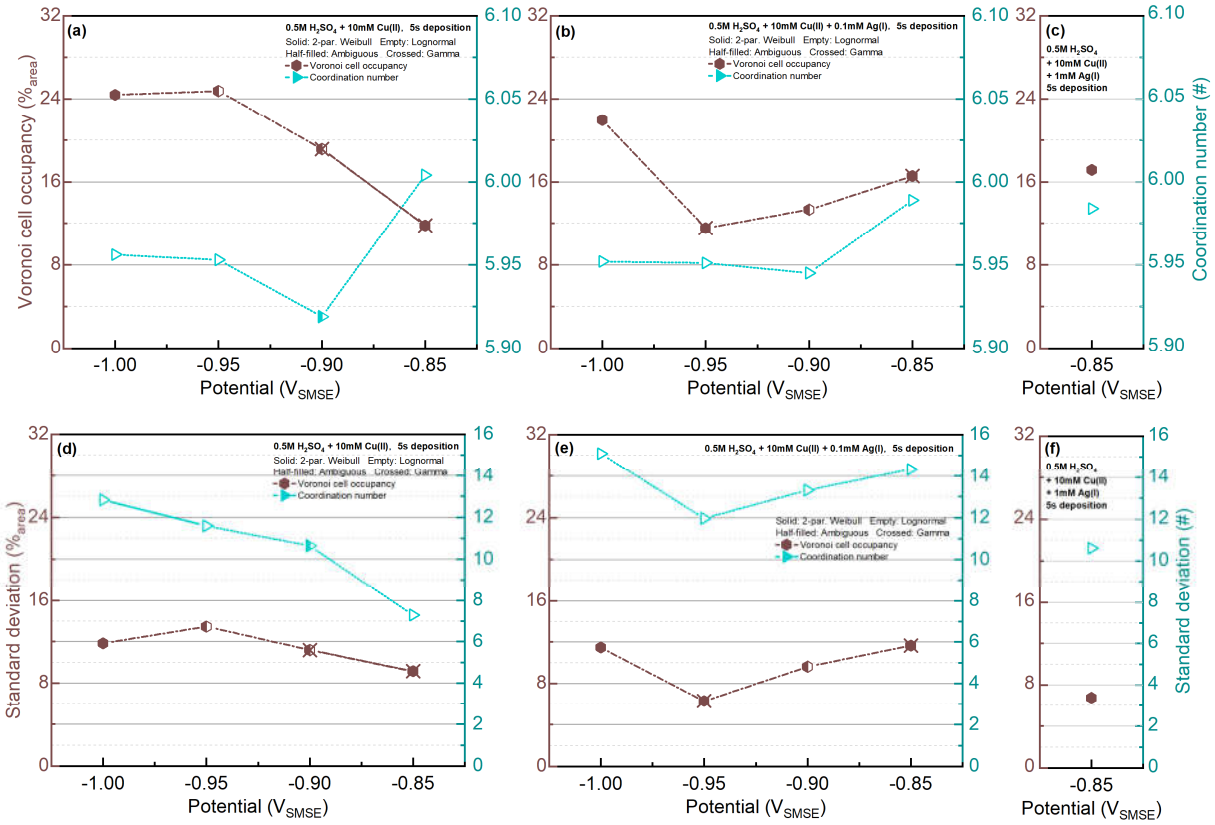


Fig.F1, Mean and standard deviation of Voronoi cell occupancy and coordination number.

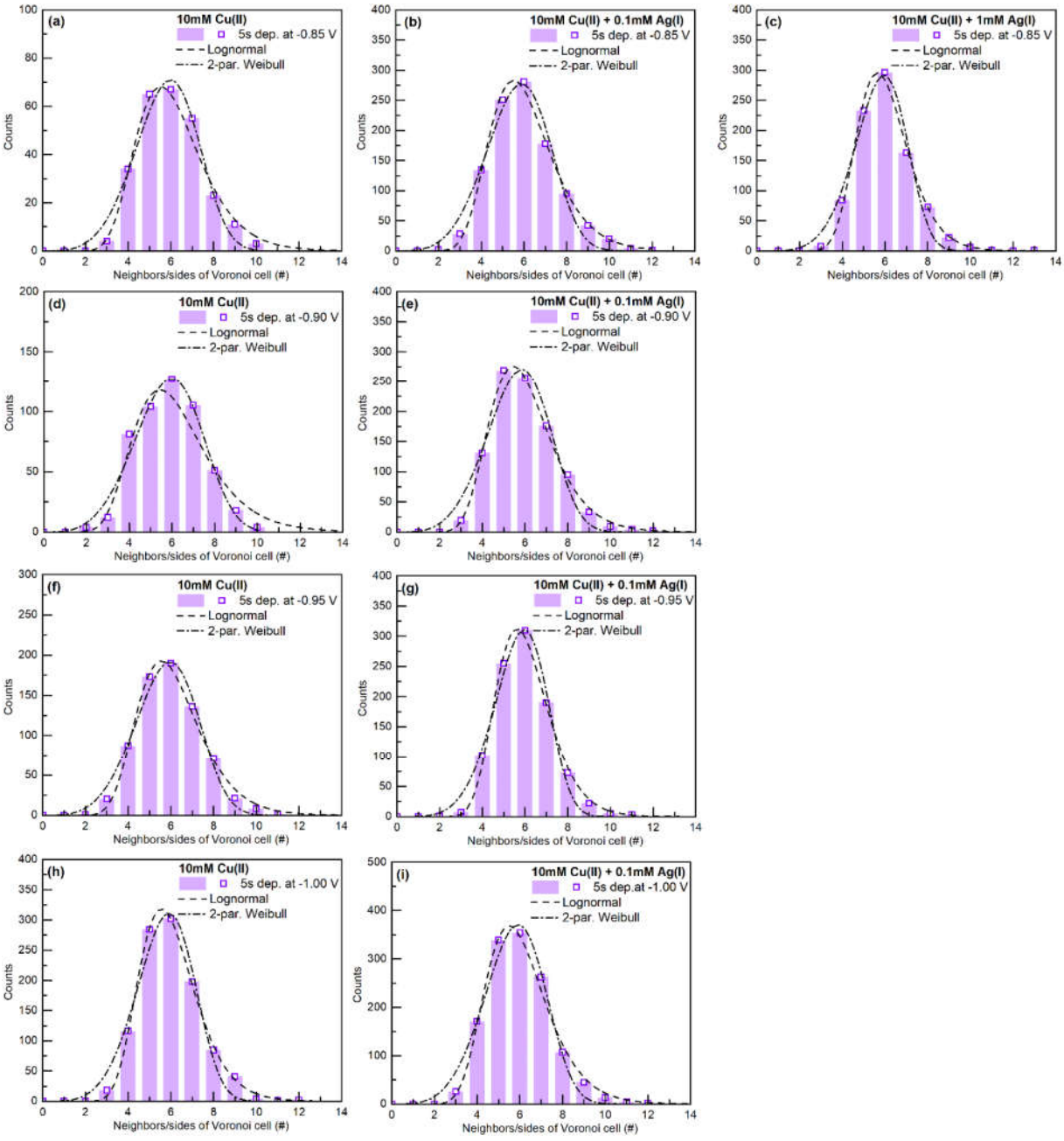


Fig.F2 Voronoi cell coordination number. Supporting electrolyte is 0.5M H_2SO_4. The fitting was conducted directly with PDF. Reference electrode of SMSE was used for the potentials.