

# Critical Investigation of Metal-Organic-Frameworks to Improve the Silicon Anode of Lithium-ion Batteries

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**Table S1.** Results of peak fittings to XPS spectra shown in Figure 2 and 5 for MOF-74, MOF-199, 0.5Si@MOF and 05Si@MOF-c.

<b>Co-MOF-74</b>					
<b>Peaks in Figure 2d (Co2p)</b>			<b>Peaks in Figure 2e (C 1s)</b>		
<b>Name</b>	<b>Position</b>	<b>%Area</b>	<b>Name</b>	<b>Position</b>	<b>%Area</b>
Co (0)-1	778.06	30.4	C-C, C-H	284.8	13.94
Co (0)-2	781.06	4.32	C-O	286	24.12
Co (0)-3	783.06	3.07	C=O	288.8	9.63
Co LMM	776.38	5.29	COOH	289.89	3.96
Co (II)-1	779.96	25.30	C1s- sp2	284.5	45.21
Co (II)-2	782.06	14.47	C1s- sp2 satellite	290.91	3.14
Co (II)-3	785.16	0.79			
Co (II)-4	785.31	16.35			
<b>Cu-MOF-199</b>					
<b>Peaks in Figure 2f (Cu2p<sub>3/2</sub>)</b>			<b>Peaks in Figure 2g (C 1s)</b>		
<b>Name</b>	<b>Position</b>	<b>%Area</b>	<b>Name</b>	<b>Position</b>	<b>%Area</b>
Cu(II) Oxide-1	932.96	65.49	C-C, C-H	284.80	59.42
Cu(II) Oxide-2	934.84	19.61	C-O	286.27	14.50
Cu(II) Oxide-3	939.91	7.14	C=O	288.50	16.89
Cu(II) Oxide-4	944.16	7.76	COOH	289.88	9.19
<b>0.5Si@MOF</b>					
<b>Peaks in Figure 5a (Co2p)</b>			<b>Peaks in Figure 5b (C 1s)</b>		
<b>Name</b>	<b>Position</b>	<b>%Area</b>	<b>Name</b>	<b>Position</b>	<b>%Area</b>
Co (II)-1	778.59	33.15	C-C, C-H	284.8	45.84
Co (II)-2	780.69	25.23	C-O	286.22	20.11
Co (II)-3	783.16	8.37	C=O	288.03	22.2
Co (II)-4	784.53	33.26	COOH	289.79	11.85
<b>Peaks in Figure 5c (Si 2p &amp; Co 3s)</b>					
<b>Name</b>	<b>Position</b>	<b>%Area</b>			
Si 2p <sub>3/2</sub> (0)	98.53	6.12			
Si 2p <sub>1/2</sub> (0)	99.13	3.06			
Si 2p (IV)	102.63	7.7			
Co 3s-1	102.65	39.06			
Co 3s-2	107.09	34.32			
Co 3s-3	111.78	9.74			
<b>0.5Si@MOF-c</b>					
<b>Peaks in Figure 5a (Co2p)</b>			<b>Peaks in Figure 5b (C 1s)</b>		
<b>Name</b>	<b>Position</b>	<b>%Area</b>	<b>Name</b>	<b>Position</b>	<b>%Area</b>
Co (II)-1	781.12	34.17	C-C, C-H	284.8	52.67
Co (II)-2	783.22	25.63	C-O	286.39	20.91
Co (II)-3	785.72	11.92	C=O	288.47	22.95
Co (II)-4	787.55	28.28	COOH	290.52	3.47
<b>Peaks in Figure 5c (Si 2p &amp; Co 3s)</b>					

Name	Position	%Area
Co 3s-1	103.21	48
Co 3s-2	107.65	39.76
Co 3s-3	112.2	12.25

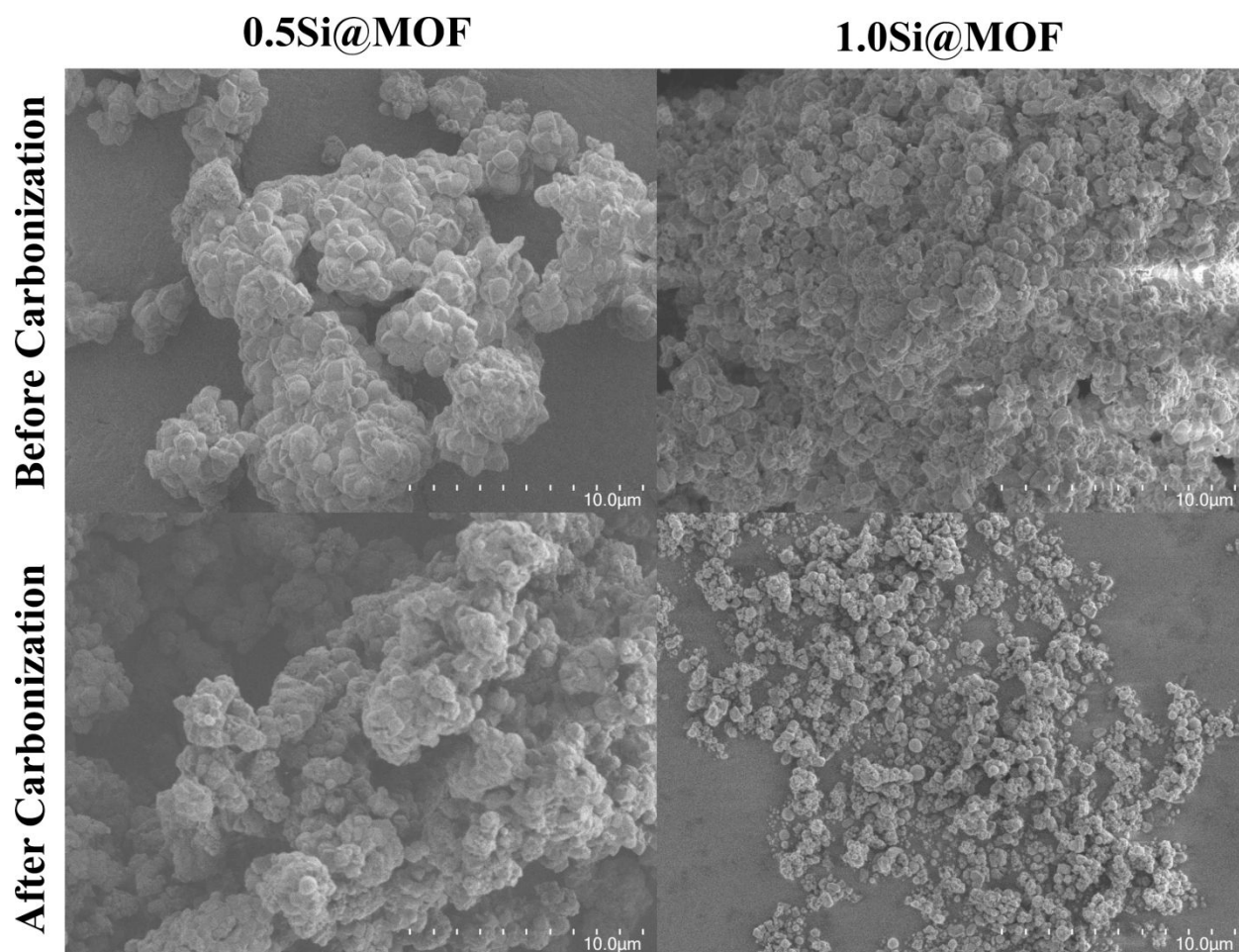


Figure S1. SEM images of 0.5Si@MOF and 1.0Si@MOF samples before and after carbonization.

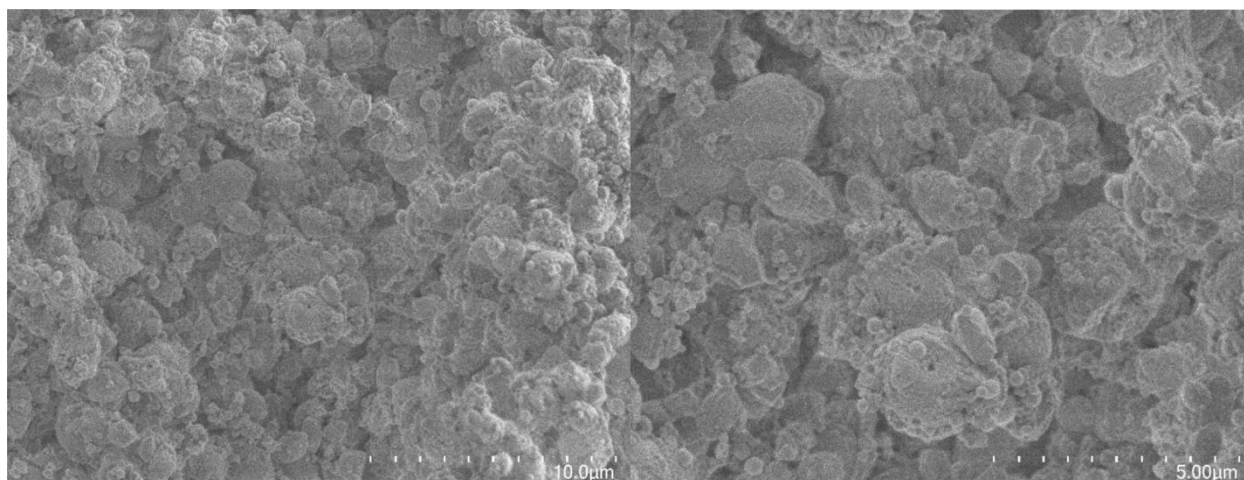


Figure S2. SEM images of the unsuccessful 1.5Si@MOF sample prior to carbonization. Notice the severe agglomeration of particles of different irregular morphology due to a large fraction of unencapsulated nano Si.

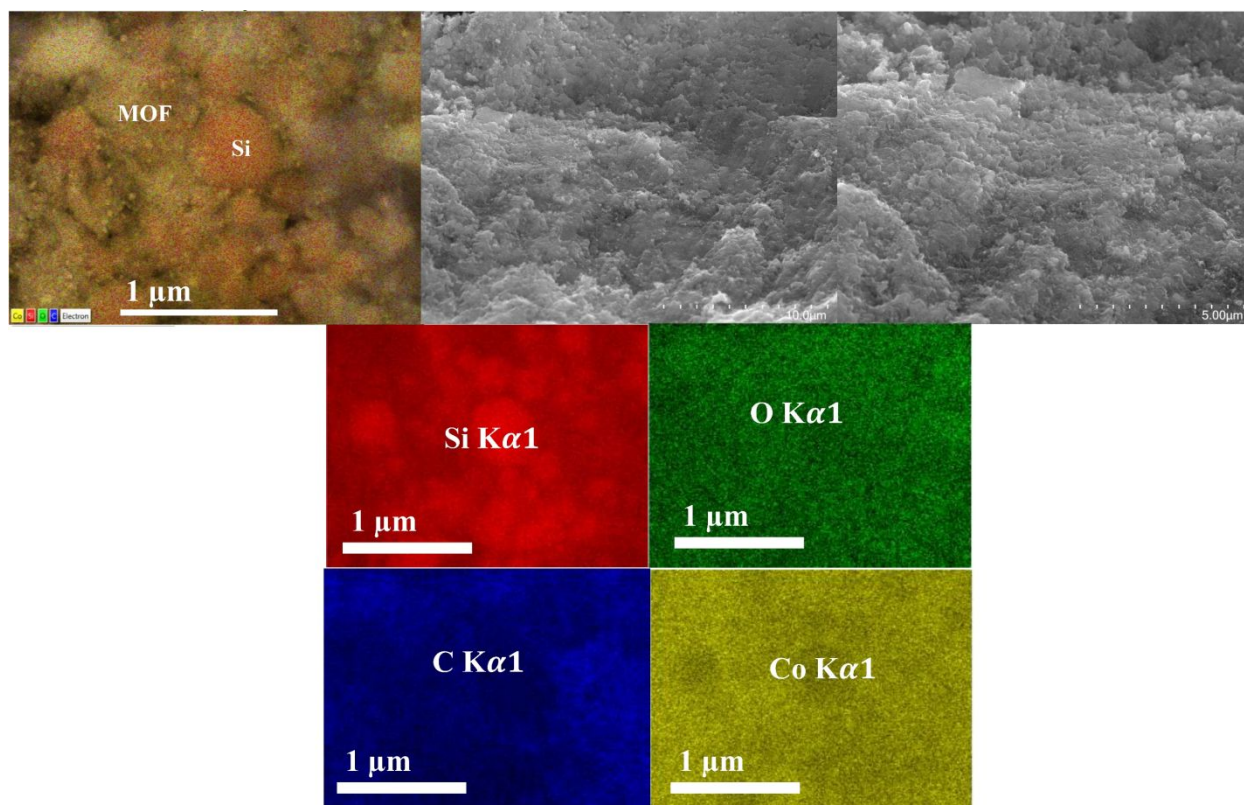


Figure S3. EDX map and SEM images of the bulk active material within an uncycled 0.5Si@MOF-c electrode.

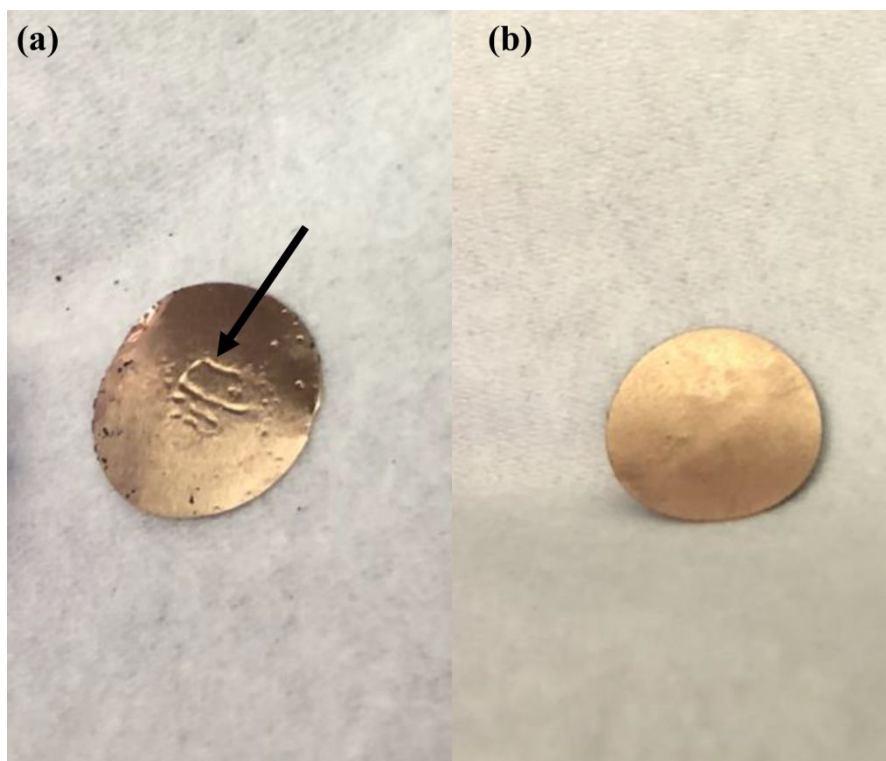


Figure S4. Postmortem images of the Cu-side for a) Si-rich electrode and b) SP-Si-MOF (MOF-74) electrode. Notice the wetting pattern on the Si-rich baseline electrode.

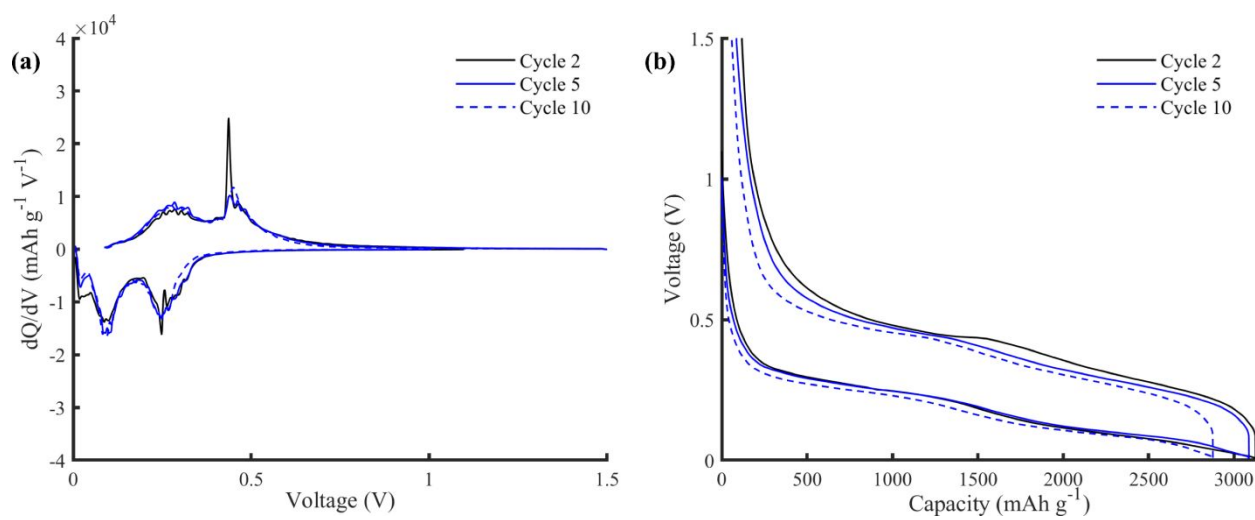


Figure S5. (a)  $dQ/dV$  plot and (b) potential profile for the SP-Si-cushion.

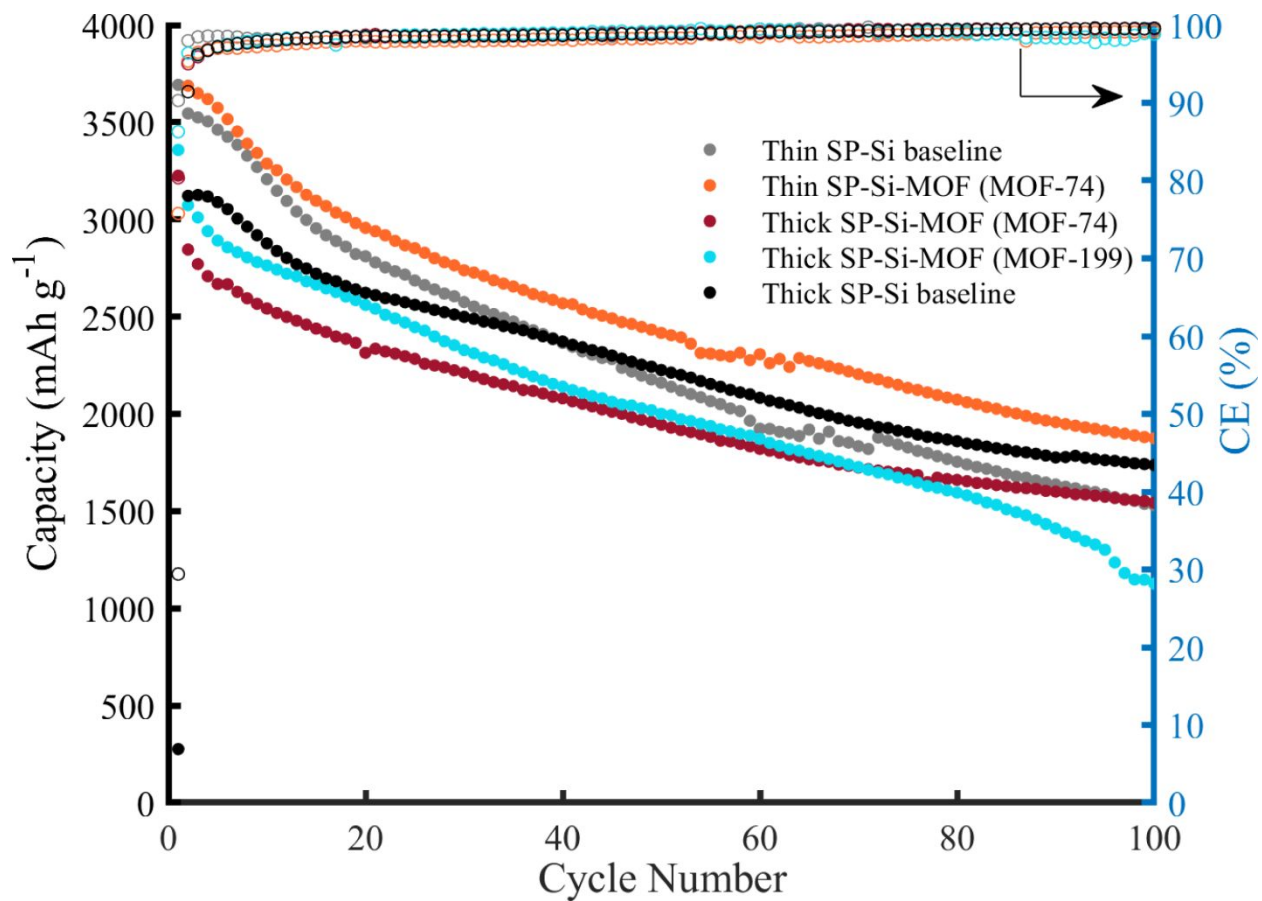


Figure S6. Active material discharge capacity for sandwich configuration electrodes of varied thickness and MOF type.

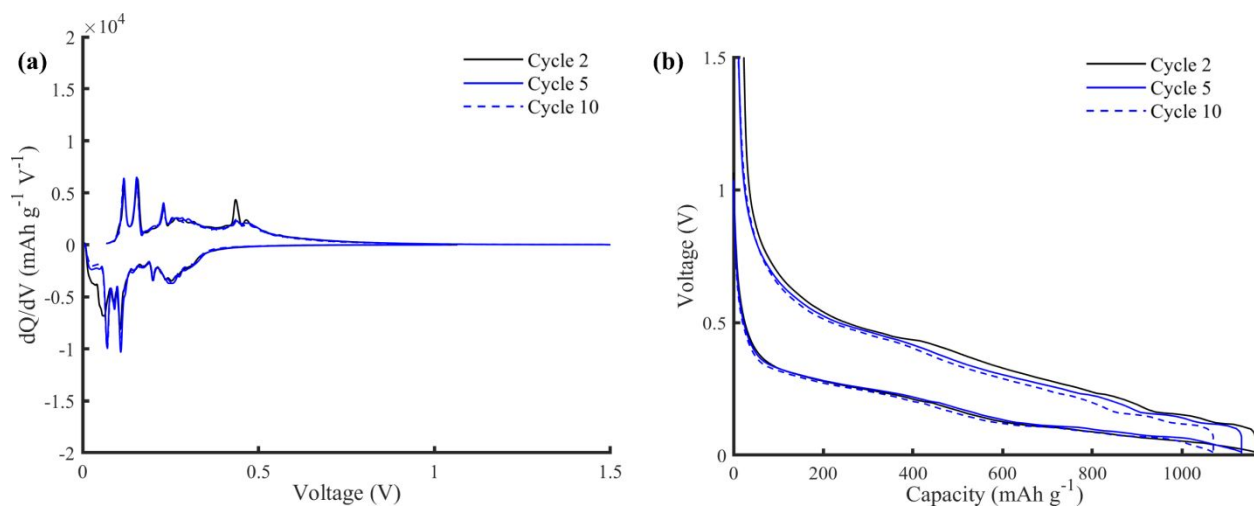


Figure S7. (a)  $dQ/dV$  plot and (b) potential profile for the 20Si-10CMC baseline.

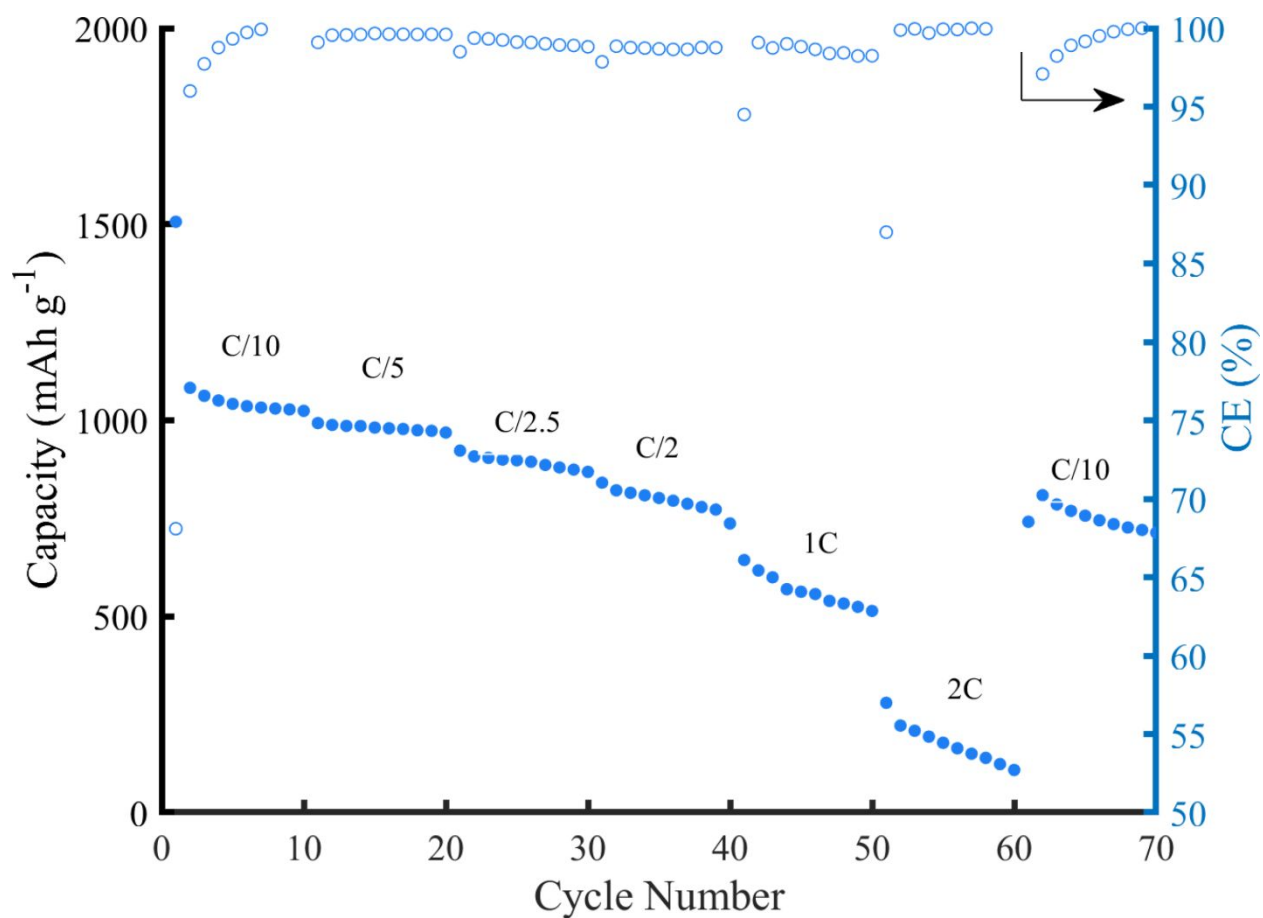


Figure S8. Active material discharge capacity during rate capability test on the best-performing 0.5Si@MOF-c electrode.

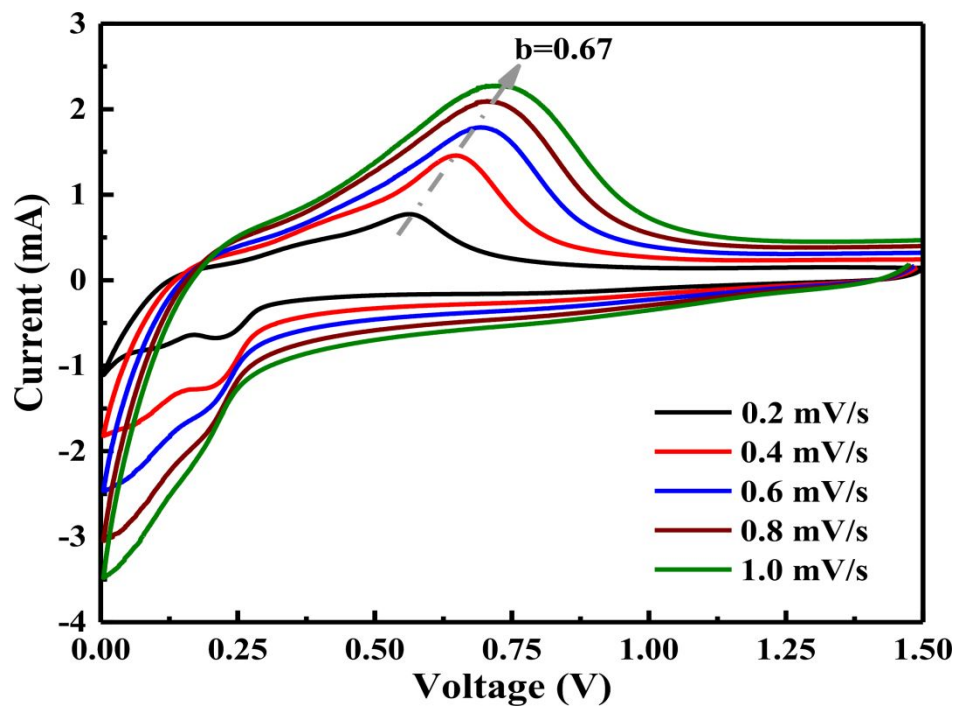


Figure S9. CVs at various scan rates on a 0.5Si@MOF-c electrode. Notice the high current, consistent with the high active material loading used in this study.