Supporting Information

Textile Functionalization by Porous Protein Crystal Conjugation and Guest Molecule Loading

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CJ Protein Sequence:
MKEYTLDKAHTDVGFKIKHLQISNVKGNFKDYSAVIDFDPASAEFKKLDVTIKIASVNTENQTRDNHLQQDDFFKAKKYPD
MTFTMKKYEKIDNEKGKMTGTLTIAGVSKDIVLDAEIGGVAKGKDGKEKIGFSLNGKIKRSDFKFATSTSTITLSDDINLNIEV
EANEKEGGSHHHHHH

CJ DNA Sequence:
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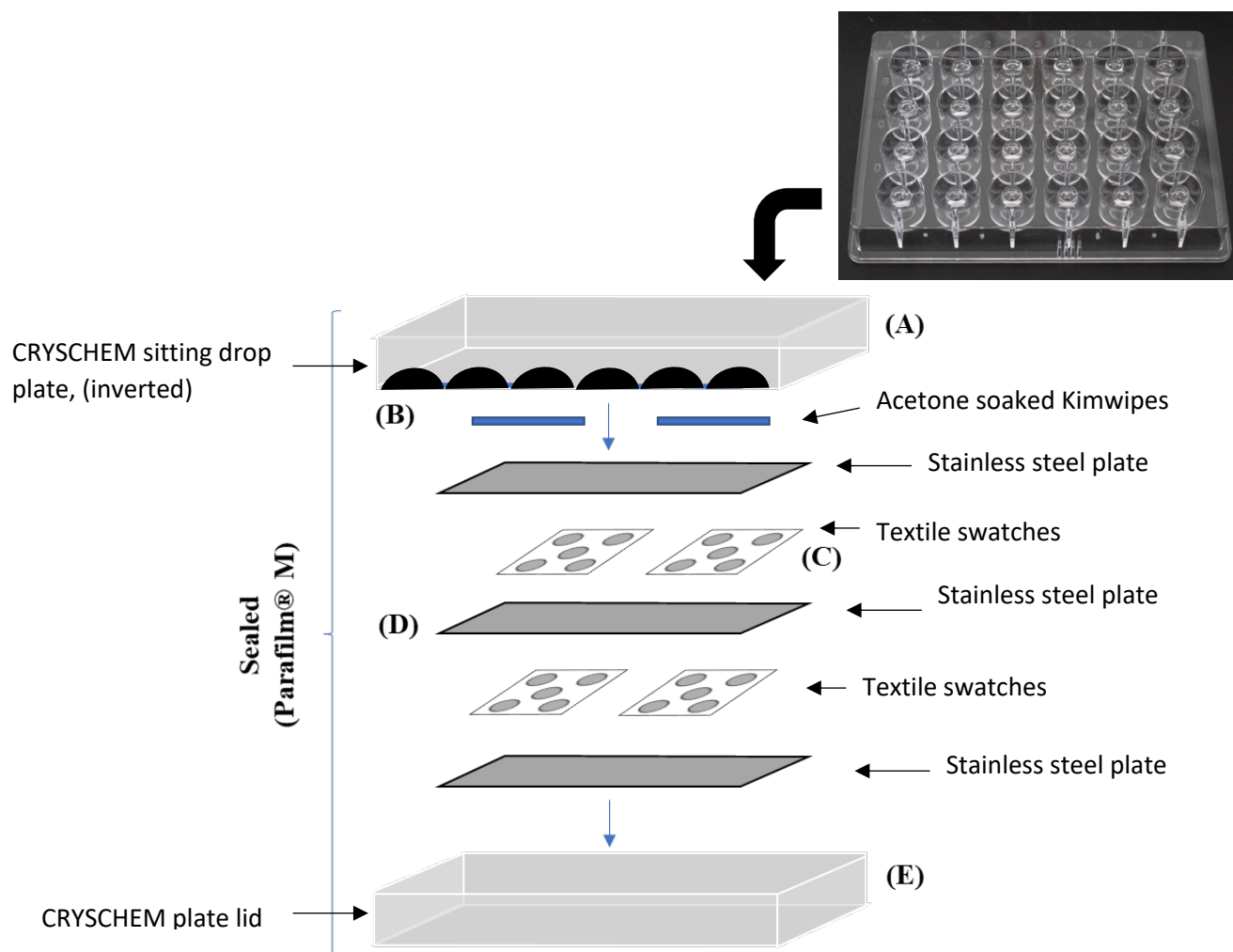TCGAGCACCACCACCACCACCACTGAGATCCGGCTG

Figure S1. CDI treatment incubation in "sandwich" apparatus: **(A)** 24-well sitting drop crystallization plate. **(B)** Kimwipes® saturated with acetone to prevent drying. **(C)** Fabric swatches activated with CDI at 5 evenly distributed locations. **(D)** Stainless steel plate to provide weight and prevent corrosion of plastic crystallization plate. **(E)** Crystallization plate cover.
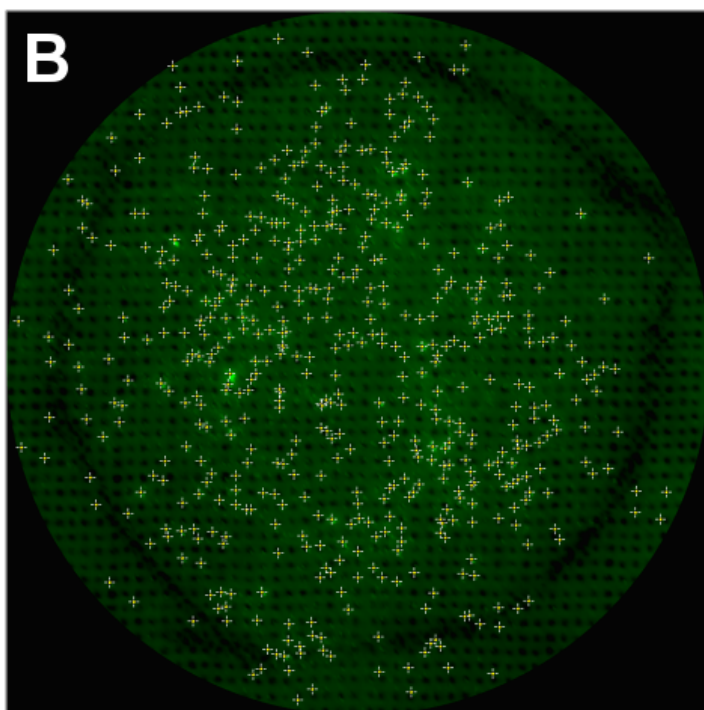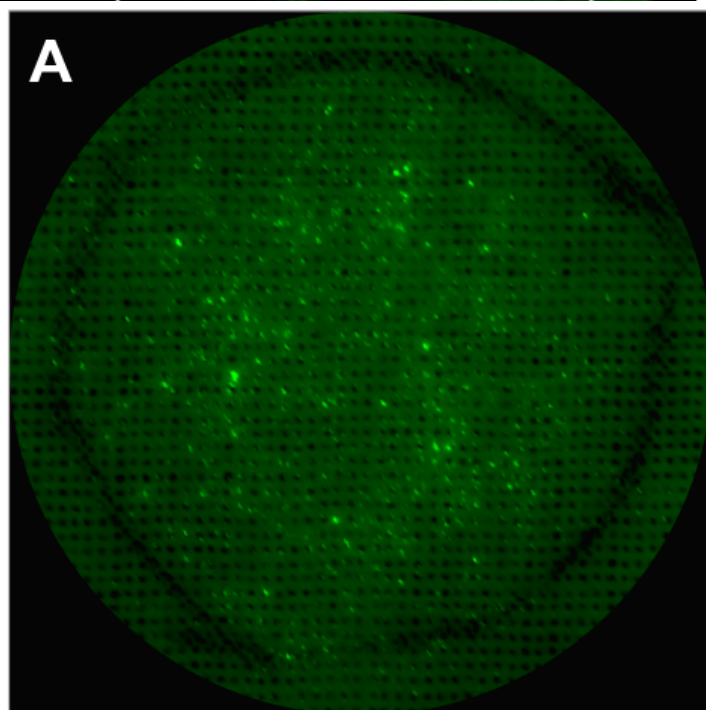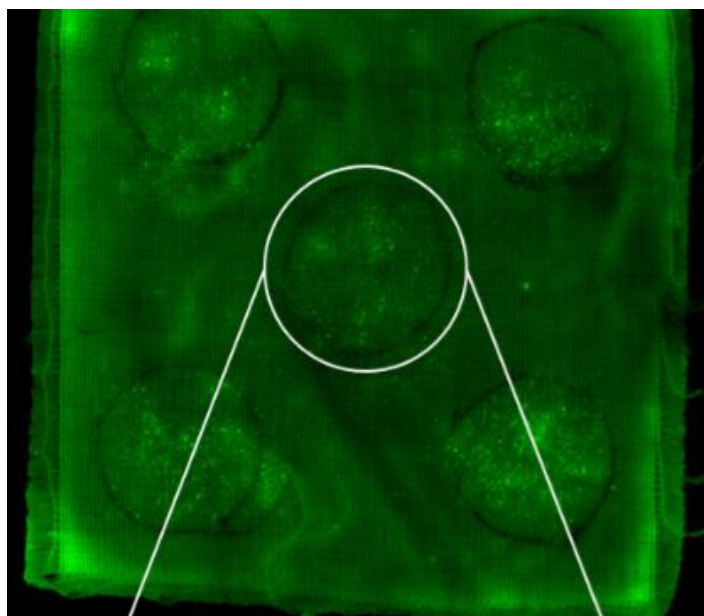
Figure S2. (A) Representative Typhoon image showing labeled HEWL crystals. **(B)** Detection and quantification of crystal puncta in Fiji. Consistent imaging and puncta detection parameters were used after each accelerated laundering interval.

Table S1. HEWL crystal retention using CDI-only conjugation.

Time (min.)	R1	R2	R3	R4	R5	
0	191	177	175	178	180	
15	121	134	107	116	121	
30	91	95	89	66	75	Total Count
45	68	78	55	45	48	
60	52	52	42	15	23	

Ave.	Std. Dev.	Time	R1	R2	R3	R4	R5	
100	0	0	100	100	100	100	100	
66.5	5.6	15	63.4	75.7	61.1	65.2	67.2	
46.2	6.8	30	47.6	53.7	50.9	37.1	41.7	Norm. Percent
32.6	7.6	45	35.6	44.1	31.4	25.3	26.7	
20.4	9.2	60	27.2	29.4	24.0	8.4	12.8	

Table S2. HEWL crystal retention using CDI+AAD+GA conjugation.

Time (min.)	R1	R2	R3	R4	R5	
0	476	517	365	335	517	
15	319	380	249	216	376	
30	283	319	190	180	314	Total Count
45	216	286	156	152	282	
60	182	266	150	123	207	

Ave.	Std. Dev.	Time	R1	R2	R3	R4	R5	
100	0	0	100	100	100	100	100	
66.5	5.6	15	67.0	73.5	68.2	64.5	72.7	
46.2	6.8	30	59.5	61.7	52.1	53.7	60.7	Norm. Percent
32.6	7.6	45	45.4	55.3	42.7	45.4	54.5	
20.4	9.2	60	38.2	51.5	41.1	36.7	40.0	

Table S3. CJ-LPC retention using CDI+AAD+OA conjugation.

Ave.	Std. Dev.	Time (min.)	R1	R2	R3	R4	R5	
		0	139	127	142	159	151	
		15	75	76	72	69	107	
		30	48	66	69	54	79	Total Count
		45	50	85	61	54	72	
		60	42	53	53	43	64	
100	0	0	100	100	100	100	100	
66.5	5.6	15	54.0	59.8	50.7	43.4	70.9	
46.2	6.8	30	34.5	52.0	48.6	34.0	52.3	Norm. Percent
32.6	7.6	45	36.0	66.9	43.0	34.0	47.7	
20.4	9.2	60	30.2	41.7	37.3	27.0	42.4	

*Red data point indicates an outlier that was removed from the average and standard deviation in Figure 3.

Table S4. CJ-LPC retention using CDI+AAD+GA conjugation.

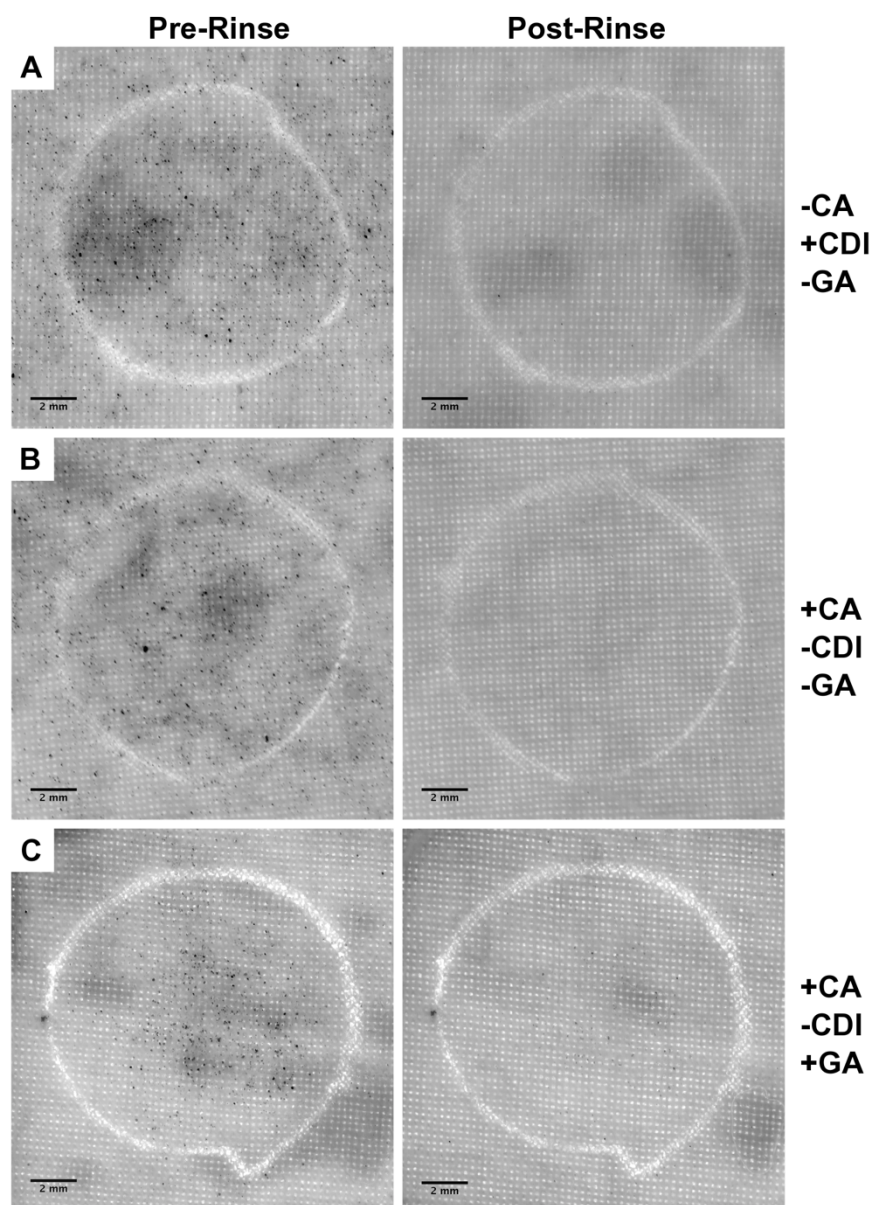| Ave.	Std. Dev.	Time (min.)	R1	R2	R3	R4	R5	
		0	659	861	497	393	685	
		15	525	633	335	295	524	
		30	433	543	276	236	445	Total Count
		45	332	433	216	198	365	
		60	293	379	202	163	309	
100	0	0	100	100	100	100	100	
66.5	5.6	15	79.7	73.5	67.4	75.1	76.5	
46.2	6.8	30	65.7	63.1	55.5	60.1	65.0	Norm. Percent
32.6	7.6	45	50.4	50.3	43.5	50.4	53.3	
20.4	9.2	60	44.5	44.0	40.6	41.5	45.1	

Figure S3. Control samples for protein crystal conjugation to cotton fabric. *Left:* pre-H$_2$O rinse; *Right:* post DI H$_2$O rinse, note: samples were not subjected to any laundering time. GA cross-linked, quenched, and NHS-fluorescein trace-labeled HEWL crystals were used in all samples. **(A)** Cotton not activated with citric acid. **(B)** Cotton not treated (conjugated) with CDI. **(C)** Cotton activated with citric acid and treated (conjugated) with GA.
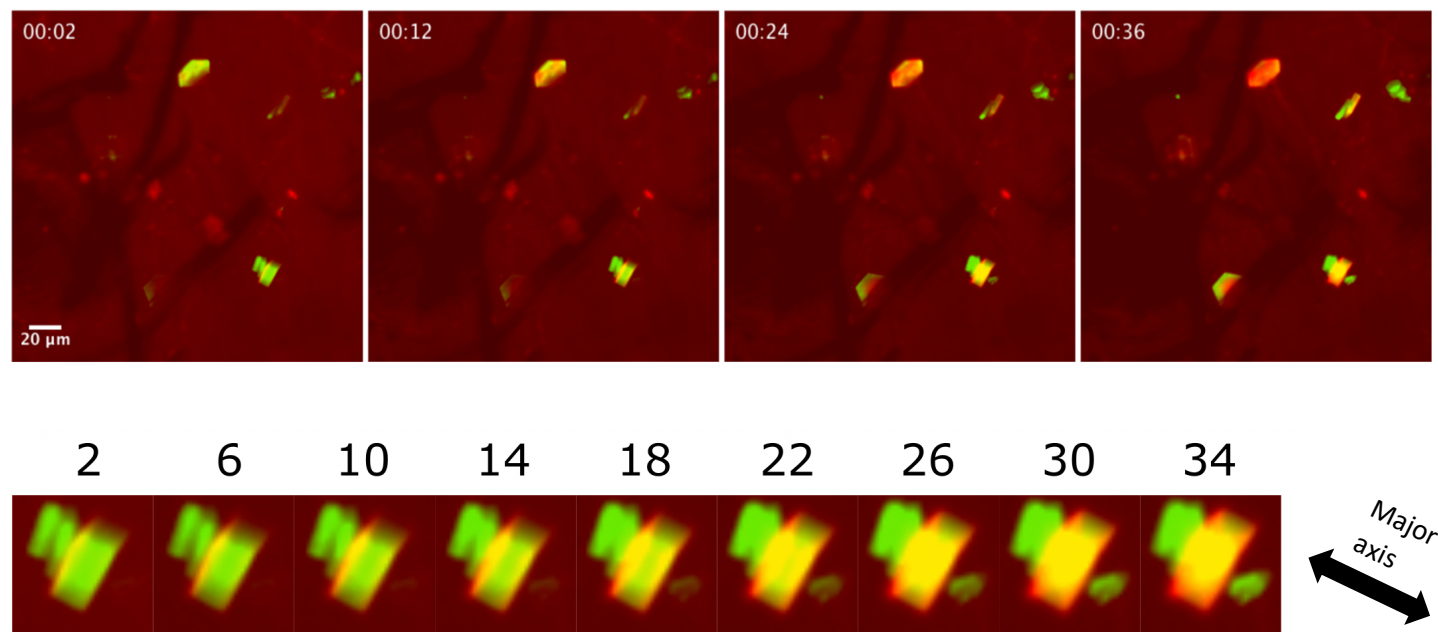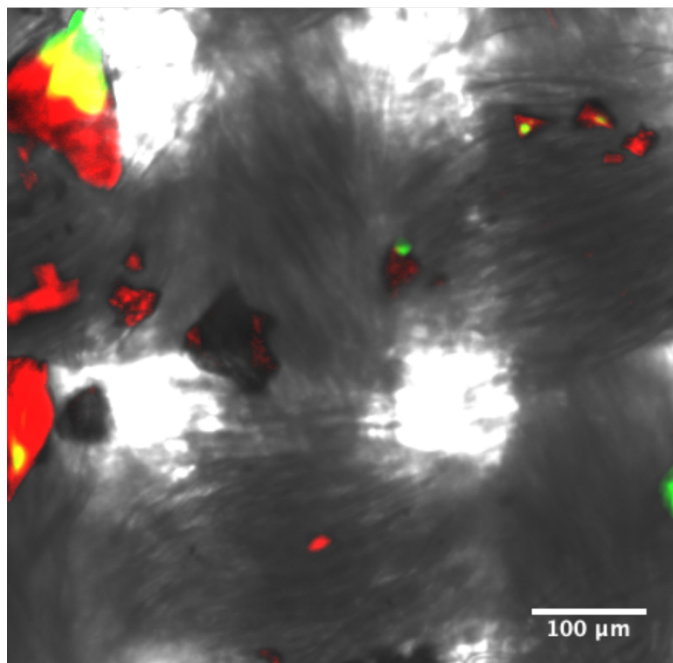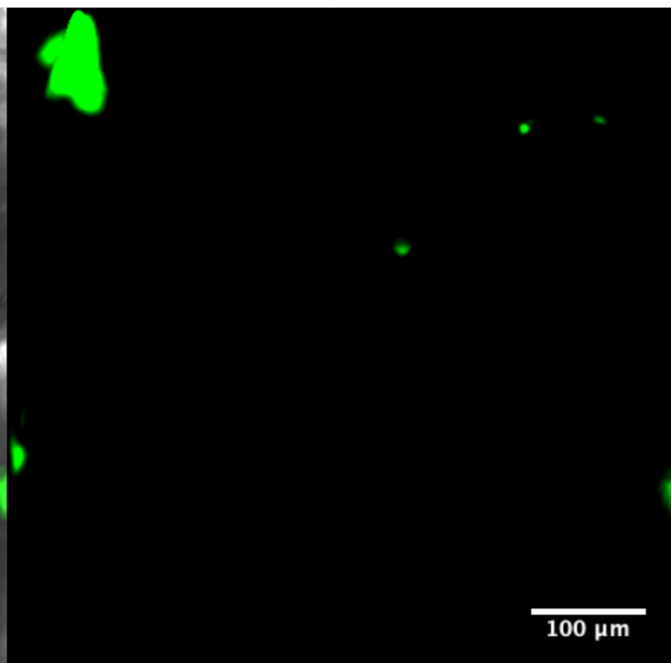
Figure S4. Top row: time-lapse images of sulforhodamine-labeled P450 (red) loading into GA cross-linked CJ-LPCs (green) conjugated to cotton fabric using a CDI+AAD+GA conjugation scheme over the course of 36 min. These are selected snapshots from the Supplementary Video 2: PorousCrystalEnzymeLoading.avi.

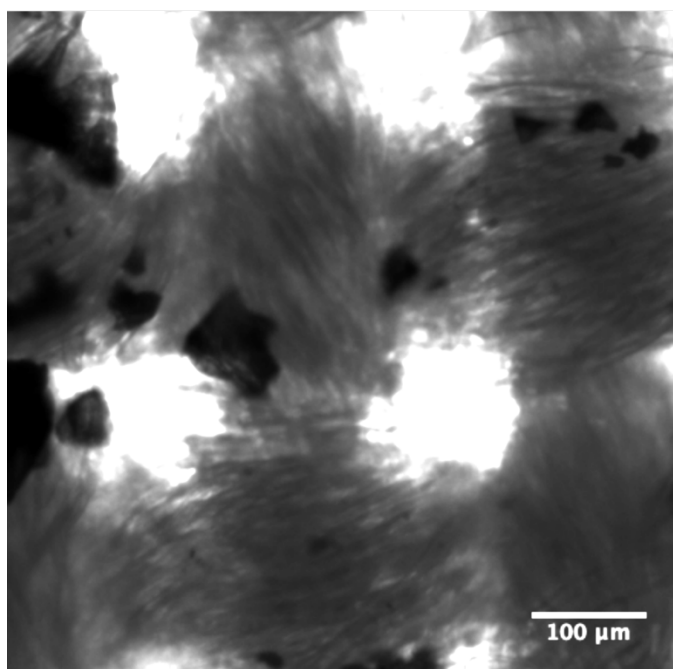Bottom row: zoomed-in view of crystal from lower-right. Anisotropic guest uptake occurs along the major axis.
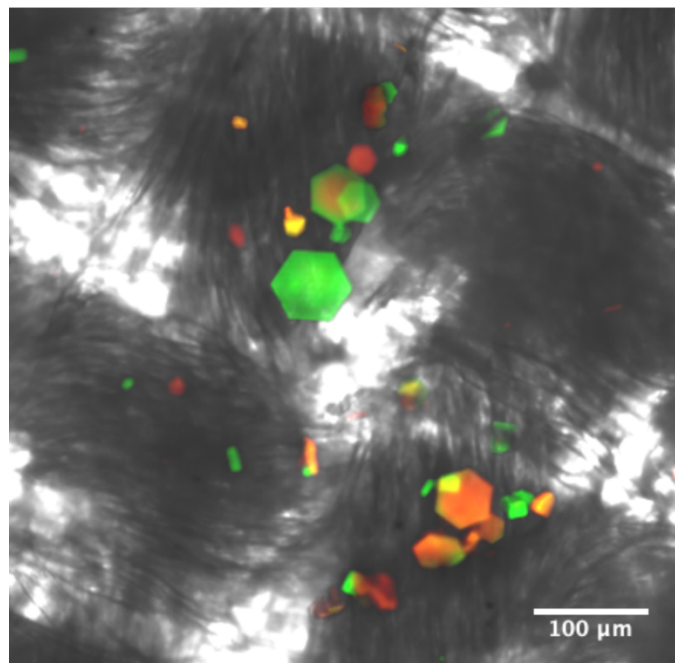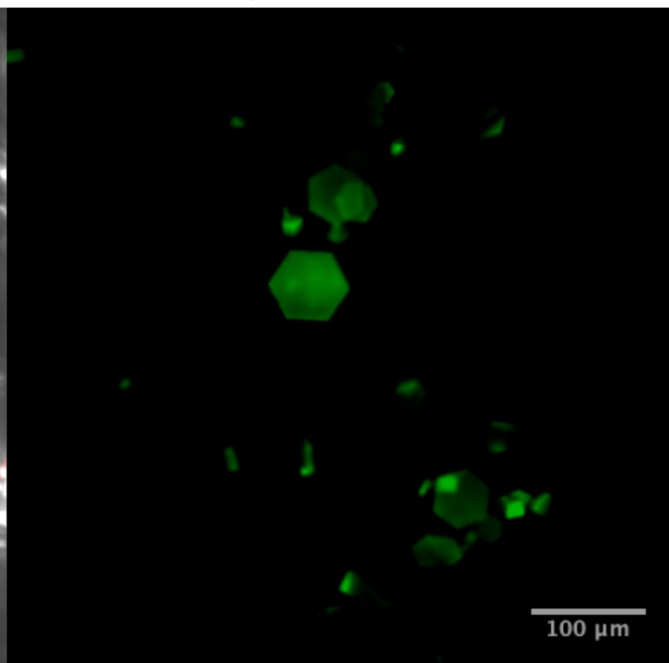
Figure S5. Variable labeling and loading of cotton-attached HEWL crystals. The merged channel **(A)** has several yellow regions where crystals were both trace-labeled via NHS-fluorescein and were able to uptake and retain non-reactive sulforhodamine 101 (TexasRed). **(B)** Relatively few crystals (or zones within crystals) were successfully labeled with NHS-fluorescein. **(C)** Irregular crystal domains are visible as darker regions in the DIC image. **(D)** Most, but not all, of the visible crystals were found to have taken up sulforhodamine 101 during a 24-hour incubation. We did not attempt to count puncta in this case because the boundaries between crystals or crystal fragments were not clear.
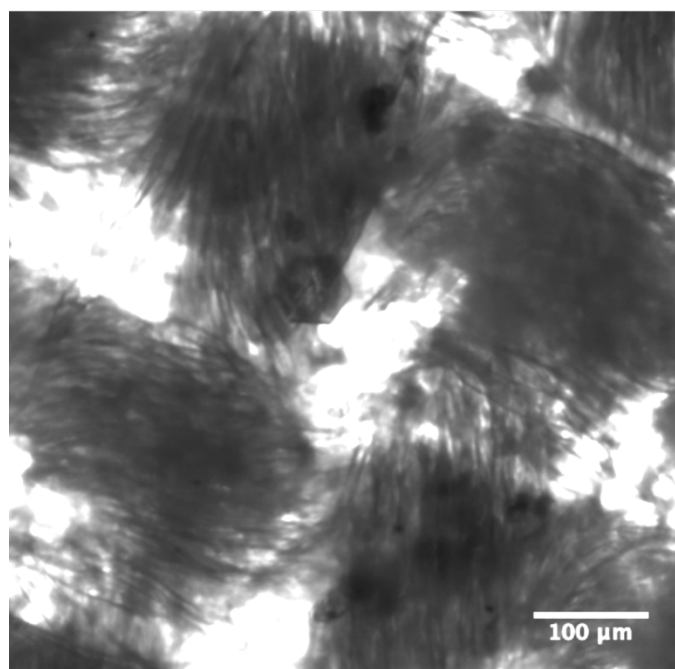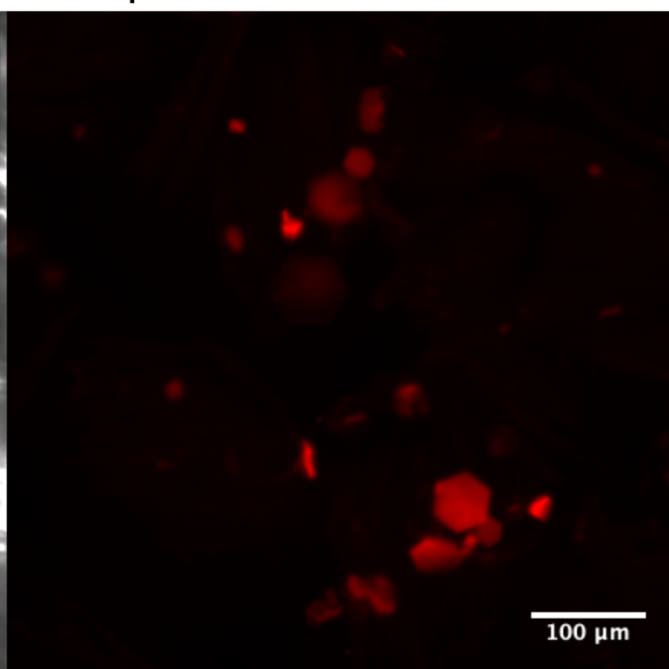
Figure S6. Variable labeling and loading of cotton-attached CJ crystals. The merged channel **(A)** has several yellow regions where NHS-fluorescein trace labeling and NHS-TexasRed (sulforhodamine 101) labeled p450 signals have comparable intensity. **(B)** Most, but not all, of the CJ crystals appear to have been successfully trace labeled with NHS-fluorescein. **(C)** CJ crystals are less visible than HEWL crystals in the DIC image, perhaps due to lower protein density. **(D)** Most, but not all, of the CJ crystals appear to have adsorbed TexasRed-labeled guest enzyme. Approximate puncta counting (including faint signals) yields 10 features that only appear in the green channel, 12 features that only appear in the red channel, and 17 features that appear on both channels.

Video S1. Time-lapse images textile swatch before accelerated laundering and after 15, 30, 45, and 60 minutes of accelerated laundering.

Video S2. Time-lapse (34 minutes, 2-minute interval) confocal microscope images showing TR-P450 enzyme (red) diffusing into cotton-attached CJ crystals (green).

Video S3. Z-stack video (5 layers, 5-micron focal plane difference) confocal microscope images showing TR-P450 enzyme (red) loaded within a subset of the cotton-attached CJ crystals (green).