

SUPPORTING INFORMATION

Mimicking Natural Human Hair Pigmentation with Synthetic Melanin

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Figure S1. Blond, human hair purchased from a second vendor (Emosa) was dyed using polydopamine formulations (companion to **Figure 1**, main text). A) Photographs of hair before (1) and after dyeing at room temperature for 2 h using 3% NH_4OH (2), 0.05 N NaOH (3), 10 mM $\text{CuSO}_4/1$ M H_2O_2 (4) and Tris buffer pH 8.5 (5). B), as for A), but the dyeing was conducted at 37-40 °C. C) Optical micrographs of individual hair fibers dyed at 37-40 °C (panel B).

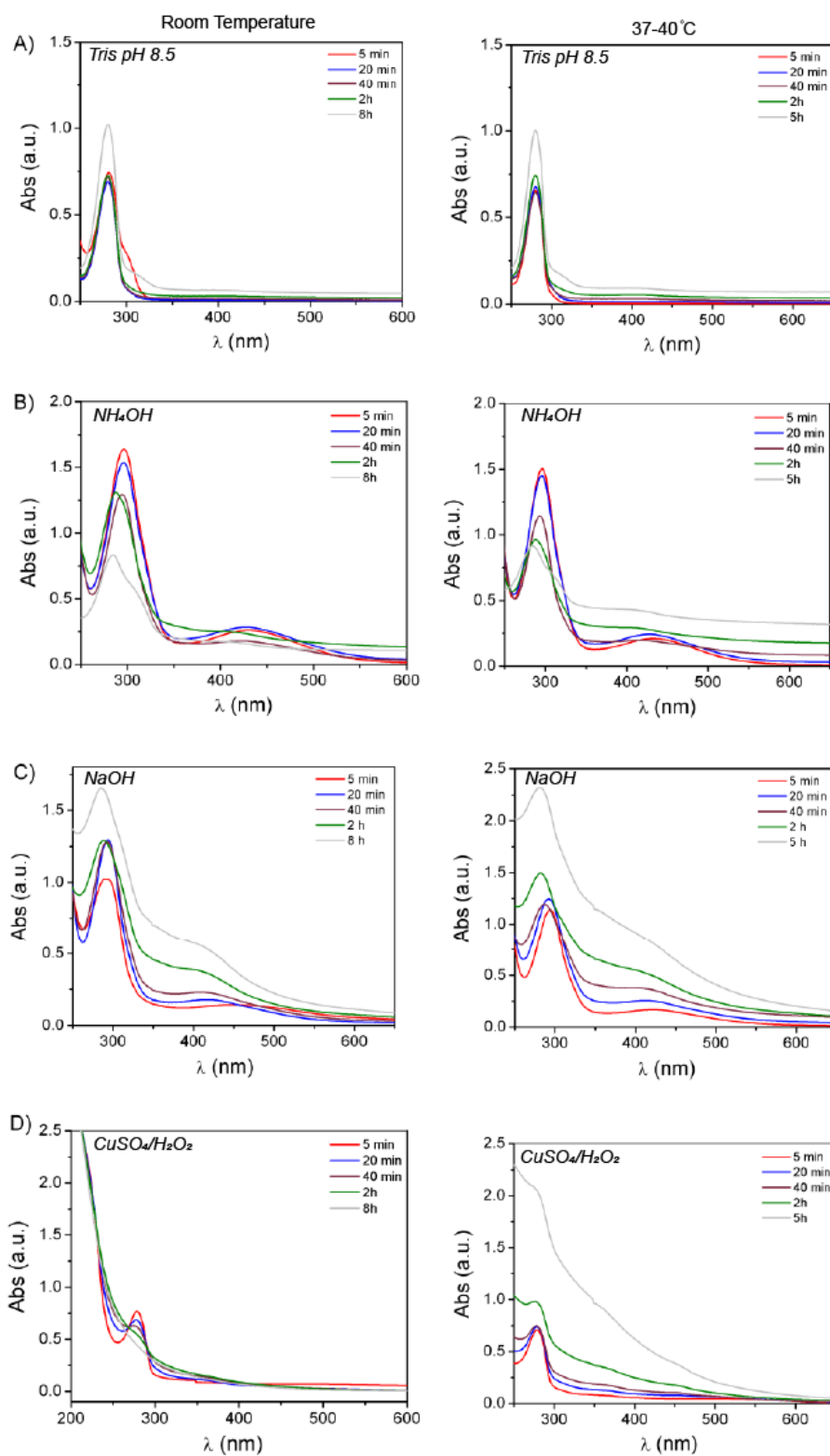


Figure S2. UV-Vis spectra of polydopamine solutions obtained at different reaction times using A) Tris buffer pH 8.5, B) 3% NH_4OH , C) 0.05 N NaOH , and D) 10 mM $\text{CuSO}_4/1$ M H_2O_2 . Reactions were carried out at room temperature (left panels) and at $T = 37\text{-}40$ °C (right panels).

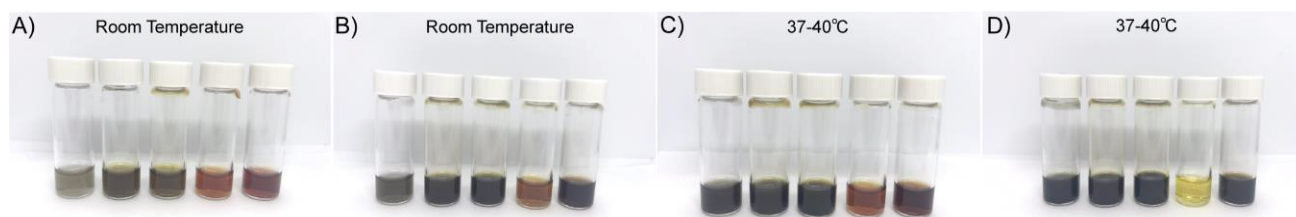


Figure S3. Color of polydopamine solutions obtained using different reaction conditions. Left to right in each panel: Tris buffer pH 8.5, 3% NH₄OH, 0.05 N NaOH, 10 mM CuSO₄/1 M H₂O₂ and 10 mM CuSO₄/15 mM H₂O₂). A, B) Reactions were carried out at room temperature. C, D) 37-40 °C. (A, C) Imaged after 20 mins. (B, D) Imaged after 2 hrs.

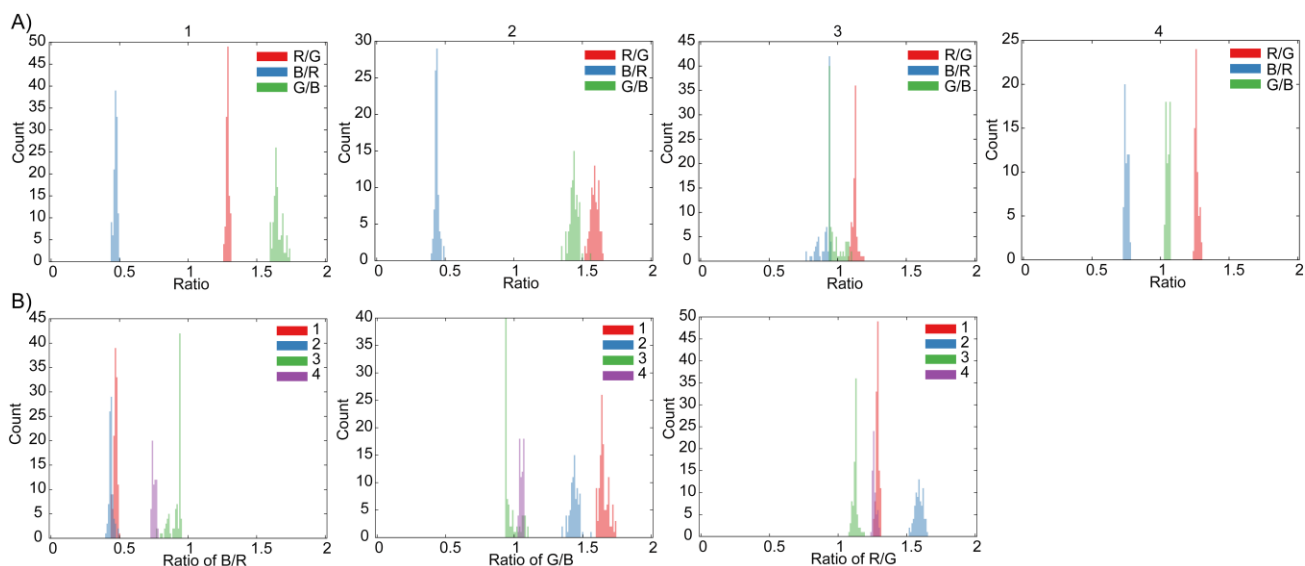


Figure S4. A) RGB color ratios. 1) Untreated blond hair. 2) Hair dyed with polydopamine using 3% NH_4OH at room temperature. Hair dyed with polydopamine using 3) 3% NH_4OH and 4) 6% NH_4OH at 37-40 °C. B) Comparison between RGB color ratios of hair samples 1-4. The photographs of samples 1-4 are shown in **Figure 3A** of the main text.



Figure S5. Photographs of hair before (1) and after dyeing with polydopamine using 3% NH_4OH (2), 0.05 N NaOH (3), 10 mM $\text{CuSO}_4/1 \text{ M H}_2\text{O}_2$ (4) and Tris buffer pH 8.5 (5). Dyeing was carried out for 2 h at A) room temperature and B) 37-40 °C. The colors of the resulting polydopamine solutions in which the hair was suspended, are shown in vials. C) Optical microscopy images of an untreated blond hair fiber (sample 1 in panels A-B) and D) hair dyed using 10 mM $\text{CuSO}_4/1 \text{ M H}_2\text{O}_2$ at 37-40 °C (Sample 4 in panel B).

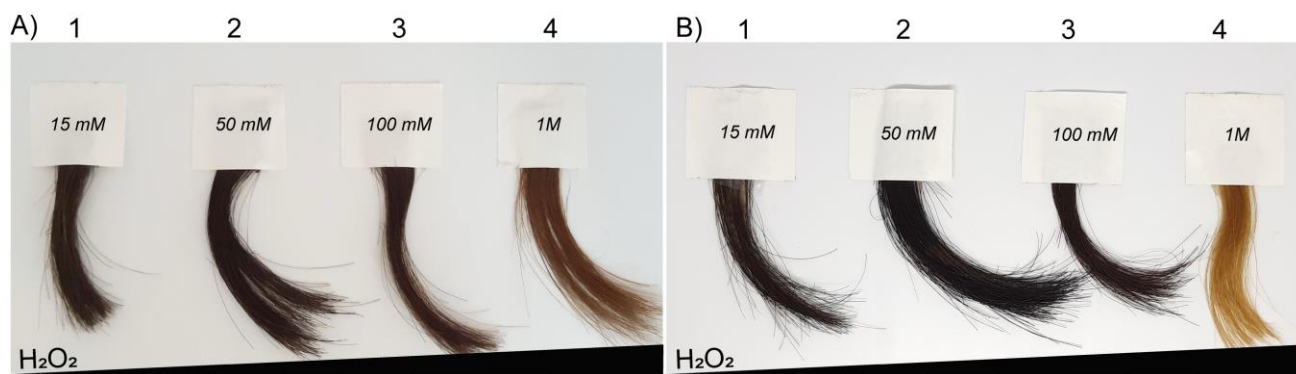


Figure S6. Photographs of hair dyed with polydopamine and CuSO₄/H₂O₂ using increasing H₂O₂ concentrations: 15 mM (1), 50 mM (2), 100 mM (3) and 1 M H₂O₂ (4). Reactions were carried out at A) room temperature and B) 37-40 °C.

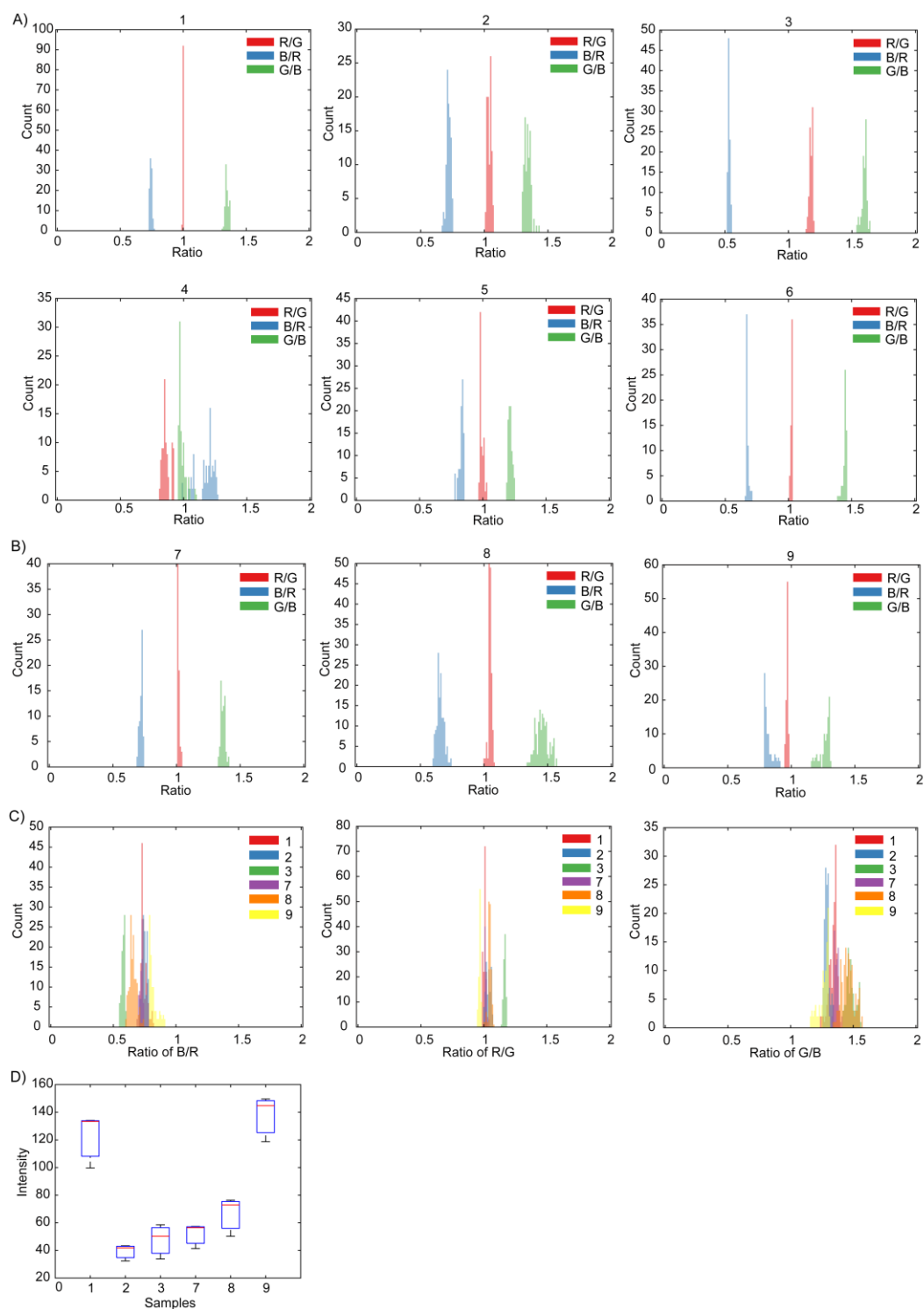


Figure S7. A) RGB color analysis of hair samples (1 to 6) highlights different RGB components for the untreated blond hair sample (1) and for hair dyed with polydopamine solutions. Reaction conditions: 10 mM CuSO_4 /1 M H_2O_2 at room temperature (2) and at 37-40 °C (3) as well as 3% NH_4OH /50 mM, 100 mM or 1 M H_2O_2 at 37-40 °C (4,5,6). B) RGB color analysis of hair dyed using

3% NH_4OH /50 mM, 100 mM or 1 M H_2O_2 at room temperature (7,8,9). C) RGB color ratios and D) color intensities of hair samples 7-9 are compared to those of untreated blond hair (1) and hair dyed using 10 mM CuSO_4 /1M H_2O_2 , either at room temperature (2) or at 37-40 °C (3). Photographs of hair samples 1-9 are shown in **Figure 4** of the main text.

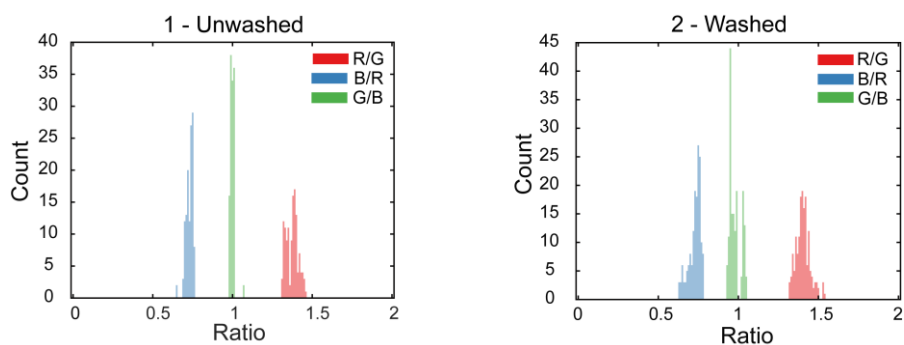


Figure S8. RGB color ratios of hair dyed with polydopamine using 6% NH_4OH for 2 at 37-40 °C before (1) and after (2) 18 washes with a 10% shampoo solution. A photograph is shown in the main text, **Figure 3A**.

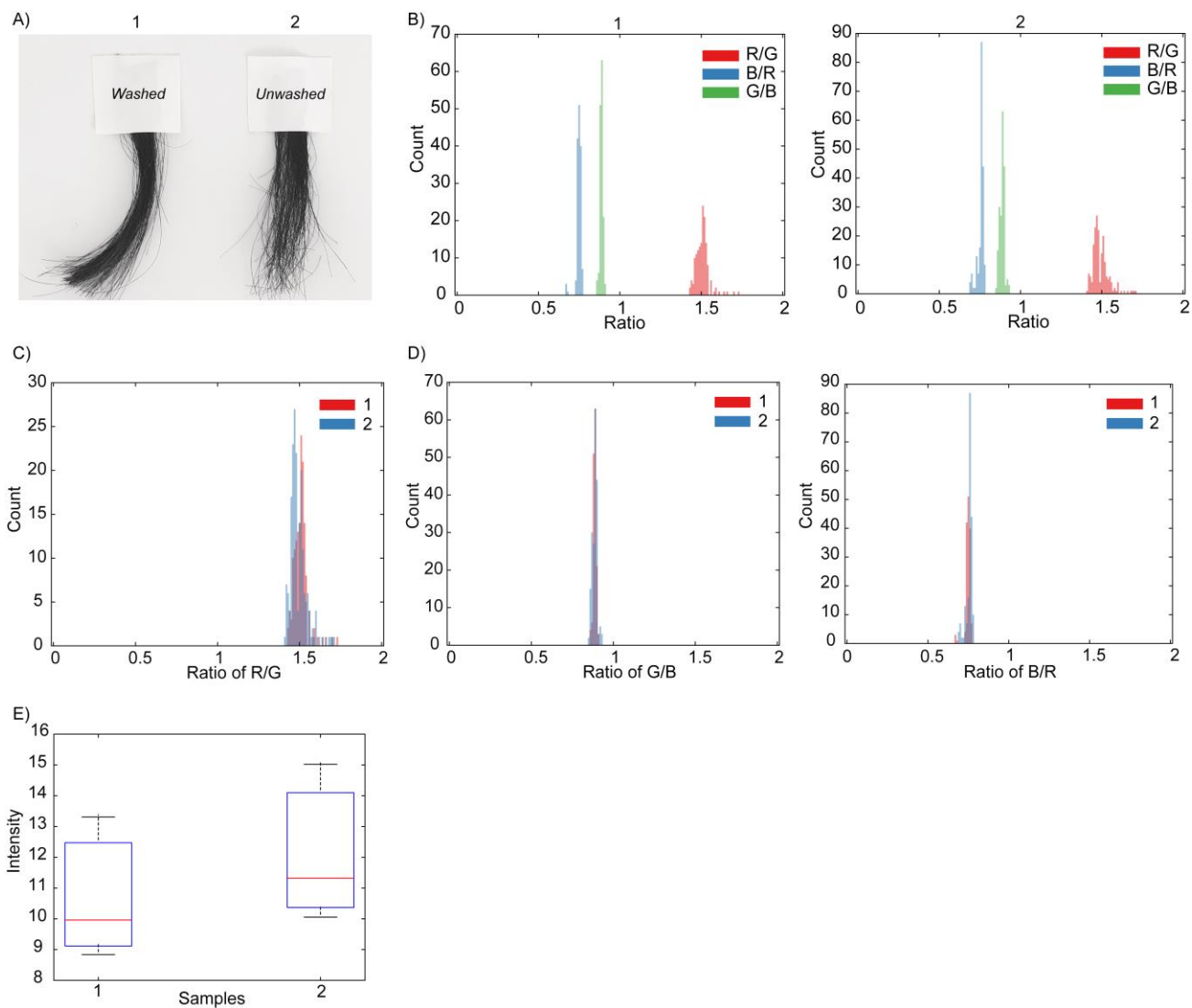


Figure S9. A) Photograph, B) RGB color analysis and C) comparison between RGB color ratios and D) color intensities of hair samples dyed with a commercially available hair dye before and after 18 washes with 10% shampoo.

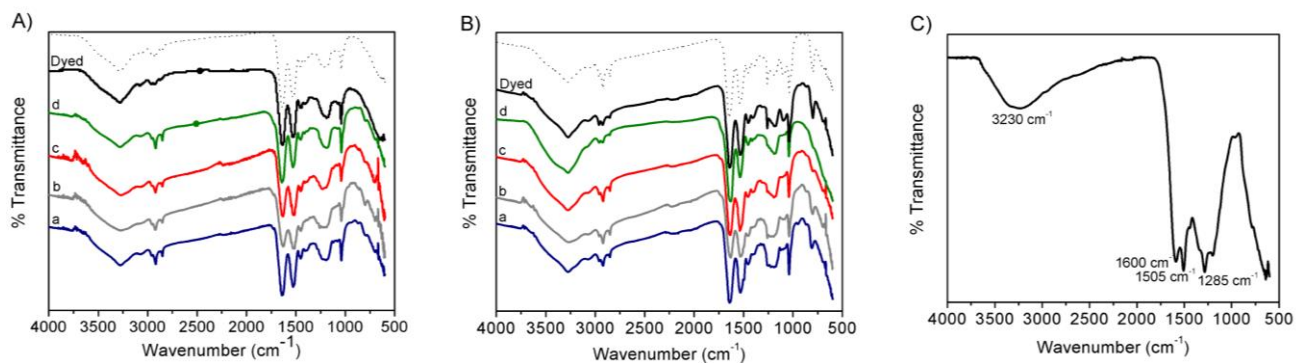


Figure S10. A) FTIR-ATR spectra of hair before (dashed line) and after (solid lines) dyeing with polydopamine formulations (a= Tris buffer pH 8.5, b= 3% NH_4OH , c= 0.05 N NaOH , d= 10 mM $\text{CuSO}_4/1$ M H_2O_2) or with a dark brown, commercial dye (Dyed). B) Hair dyed using the same protocols as A), but derived from a second vendor (Emosa) were also analyzed and showed very similar results. C) FTIR-ATR spectra of lyophilized polydopamine nanoparticles.

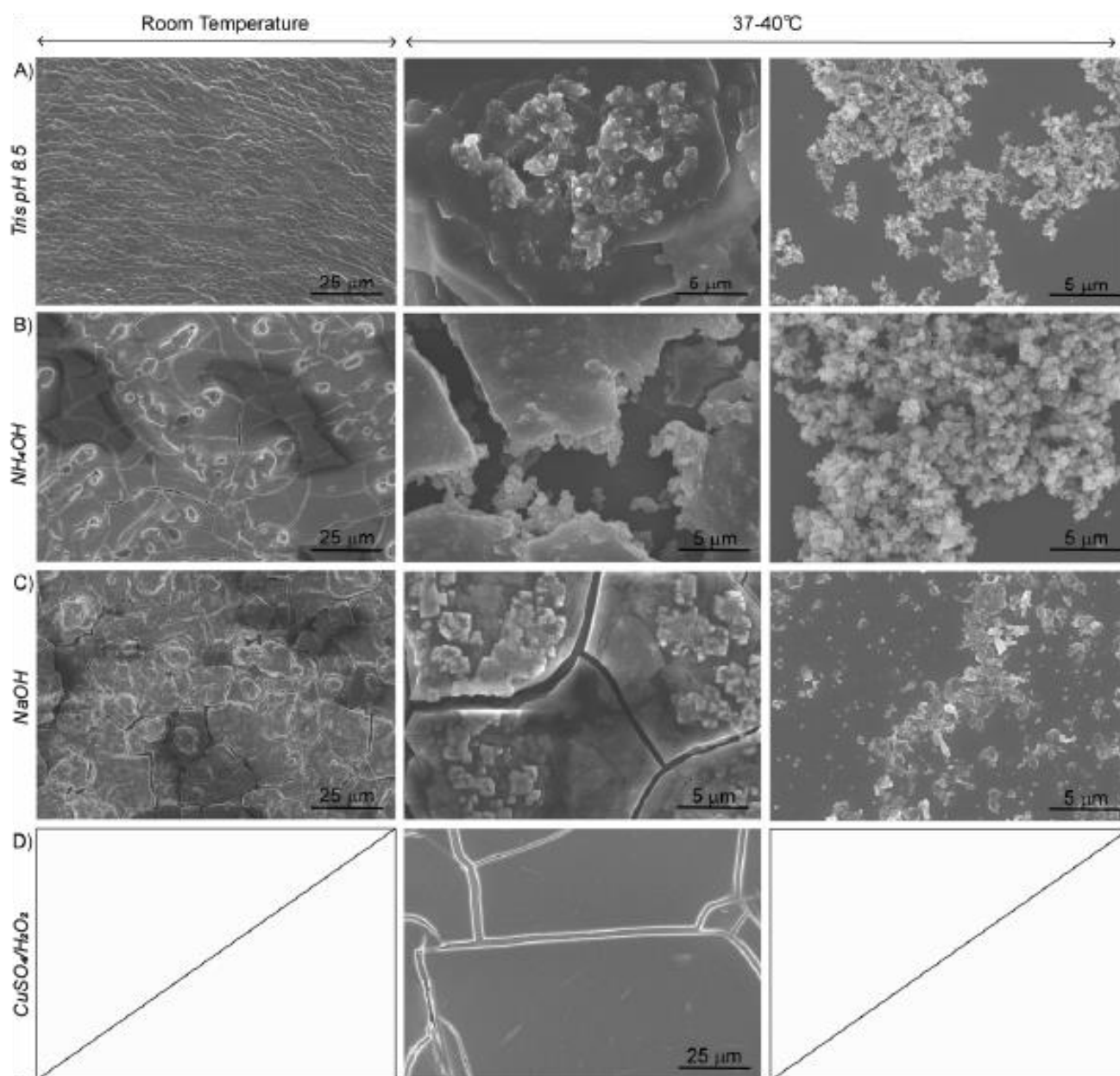


Figure S11. SEM images of polydopamine solutions obtained after a 2 h reaction at room temperature (left images) and at 37-40 °C (center and right images). Reaction conditions: A) Tris buffer pH 8.5, B) 3% NH₄OH, C) 0.05 N NaOH, and D) 10 mM CuSO₄/1 M H₂O₂. Purified polydopamine nanoparticles obtained after 2 h oxidation at 37-40 °C using alkaline conditions A-C are shown in the right panel. Reaction solutions obtained using D) CuSO₄/ H₂O₂ did not result in nanoparticle formation, one example of SEM image is reported in the center panel (D, 37-40 °C).

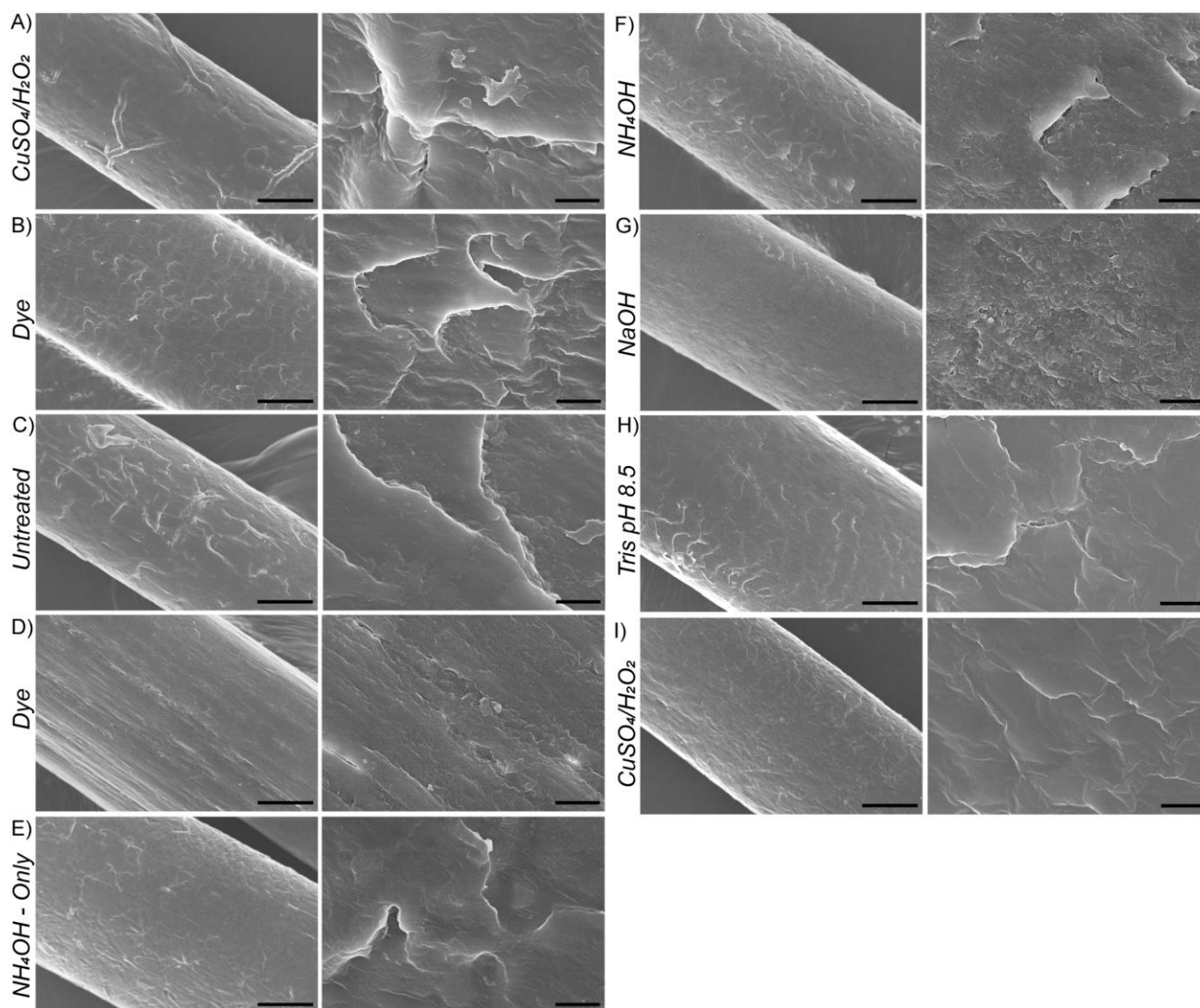


Figure S12. A) SEM images of hair purchased from Jerome Krause Fashion Hair and dyed using dopamine and 10 mM $\text{CuSO}_4/1\text{M H}_2\text{O}_2$ and B) hair dyed with a commercial dark brown hair dye. C) SEM images of hair samples purchased from Emosa and D) dyed with a commercial dark brown hair dye or E) treated with 3% NH_4OH . Polydopamine deposition was carried out at 37-40 °C for 2 h using F) 3% NH_4OH , G) 0.05 N NaOH , H) Tris buffer pH 8.5 and I) 10 mM $\text{CuSO}_4/1\text{ M H}_2\text{O}$. Scale bars 25 μm (left columns), and 2 μm (right columns).

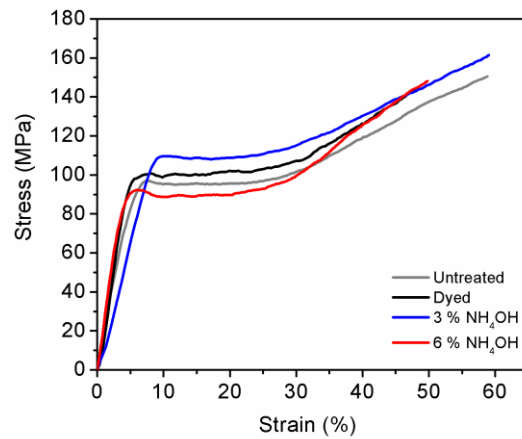


Figure S13. Shear and stress curves of hair before (untreated) and after polydopamine dyeing using 3% and 6% NH_4OH , as well as after dyeing with a commercial dark brown hair dye (Dyed). Results are reported as the average of five independent experiments.

Table S1. Mechanical properties of hair before and after dyeing using either 3% or 6% NH_4OH or using a commercially available hair dye. The average results are indicated in bold.

Conditions	Strain at Break (mm/mm)	Young's Modulus (GPa)
Untreated	0.795	3.073
	0.606	3.482
	0.746	1.716
	0.681	2.032
	0.524	3.448
	0.6704	2.7502
3% NH_4OH^*	0.644	1.92
	0.576	2.875
	0.815	1.743
	0.77	2.737
	0.83	1.841
	0.727	2.2232
6% NH_4OH^*	0.566	3.415
	0.617	3.156
	0.566	2.181
	0.614	2.869
	0.671	2.304
	0.5988	2.785
Dyed**	0.592	2.657
	0.461	4.566
	0.8	1.931
	0.653	2.107
		0.6265

* Hair dyed with polydopamine at 37-40°C for 2 h.

**Hair dyed using a commercial hair dye.