

## Supporting Information

### **Bioinspired Polymeric Pigments to Mimic Natural Hair Coloring**

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## Materials

Dopamine hydrochloride was purchased from Inno-chem Co., Ltd. Tyrosine hydroxylase was purchased from Sigma-Aldrich (China). And iron chloride hexahydrate ( $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ), iron sulfate heptahydrate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) and Copper sulfate pentahydrate ( $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ) were purchased from Aladdin Co., Ltd. To test our method's hair dyeing ability, grey hair samples were purchased from Narcissus Salon. BALB/c mice were purchased from Institutional Animal Care and Shanghai Slaccas Laboratory Animals Co., Ltd.

## Experimental section

### 1. Hair dyeing.

Hair samples were cut into approximately 3 cm long and put into the centrifuge tube. 2 mL of dopamine hydrochloride (5 mg/mL) was added into the centrifuge tube in order to completely cover the hair samples, and then added 60  $\mu\text{L}$  tyrosine hydroxylase (7 mg/mL) into it. At this time metal chelators were added (10mM  $\text{FeCl}_3$ , 10mM  $\text{FeSO}_4$  and 10mM  $\text{CuSO}_4$ ). The reaction was carried at room temperature. After five minutes, all the hair samples were washed by water five times.

### 2. Characterization of polydopamine.

The solution above was diluted 20 times and UV-visible (UV-vis) absorption spectra was recorded by an ultraviolet spectrophotometer from Shanghai Jingke. 10  $\mu\text{L}$  of the diluted solution dripped onto a nitrocellulose-covered copper grid and then air-dried. TEM imaged by a JEOL JEM-2100F instrument at 200 kV equipped with a Gatan 894 Ultrascan 1 k CCD camera. 10  $\mu\text{L}$  of the diluted solution dripped onto a silicon wafer substrate. After air dried, the SEM imaged by scanning electron microscope from Zeiss.

### 3. SEM of hair samples.

Dyed hair samples were soaked in water for 2 minutes, afterwards washed with running water, and then air-dried at room temperature. The hair samples were cut into 5-7 mm and adhered onto the SEM sample stage with conductive tapes. After lightly pressing, the SEM images was taking by scanning electron microscope from Zeiss.

### 4. Color analysis

The color intensity was analyzed by a previous method using ImageJ [1]. One fixed size area in the digital image was randomly selected and calculated its RGB value ( $I = I_R + I_G + I_B$ ) through ImageJ. Grey hair acted as control experiment background ( $I_0$ ). The darkness ( $D$ ) was calculated as  $D = (I_0 - I) / I_0$ . The RGB color ratio was analyzed using ImageJ through RGB Measure Plugins in order to differentiate the colors from different conditions

### 5. Animal experiments.

All procedures were approved by the Tongji University Animal Studies Committee. Overall care of the animals was consistent with the Guide for the Care and Use of Laboratory Animals from the National Research Council and the USDA Animal Care Resource Guide. Female mice freely breed for 1 week, and then they were shaved on their back to expose the fluff below. And 1 mL of the solution was dripped onto their back and then smeared evenly with clear gloves. Five minutes later, the photographs were taken.

1. UV-vis spectra of polydopamine assemblies.

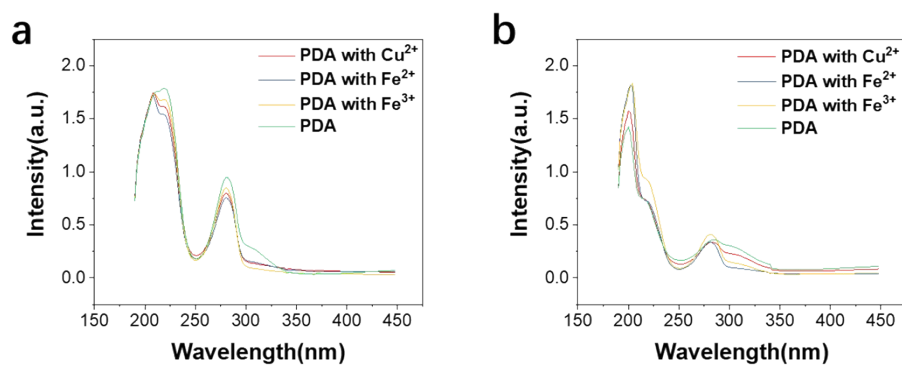


Fig. S1 The UV-vis spectra of dopamine before (a) and after (b) oxidation.

2. TEM images of polydopamine nanoparticles.

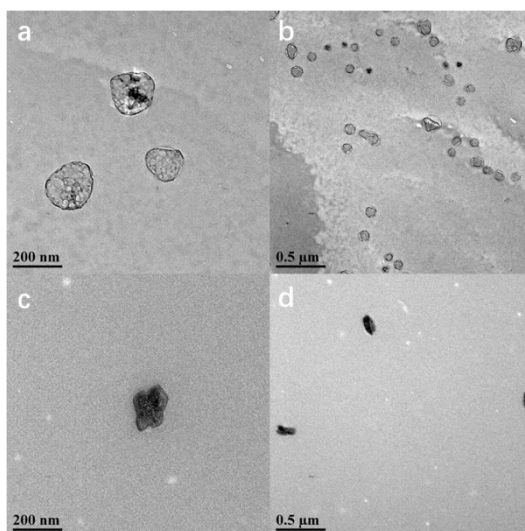


Fig. S2 The TEM images of polydopamine nanoparticles with  $\text{Fe}^{2+}$  chelating (a,b) and  $\text{Cu}^{2+}$  chelating (c,d).

3. SEM images of polydopamine nanoparticles.

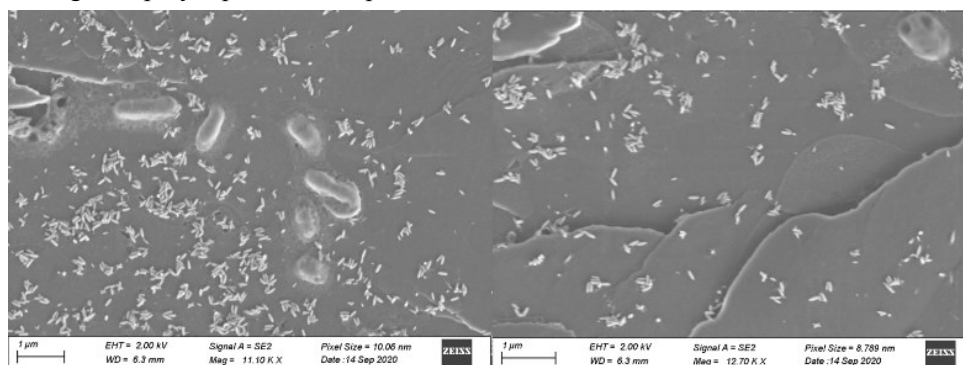
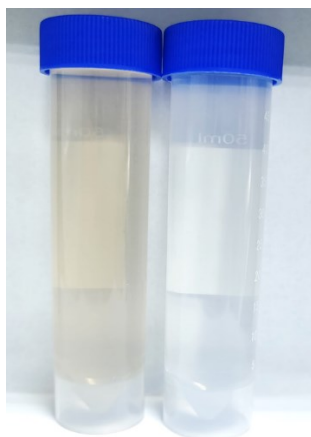


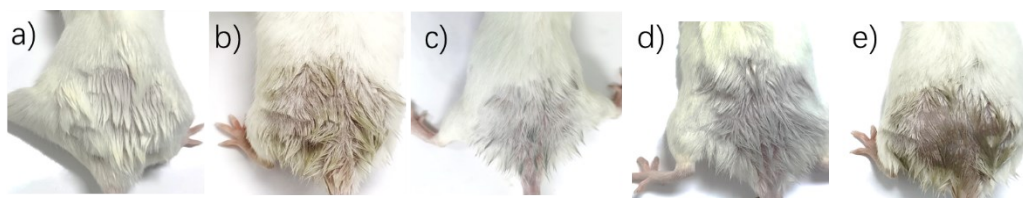
Fig. S3 The SEM images of polydopamine nanoparticles with  $\text{Fe}^{3+}$  chelating.

4. Soiled solution resulting from washing dyed hair.



**Fig. S4** Soiled solution resulting from washing commercial dyeing hair (left) and polydopamine dyeing hair (right).

5. Images of mice after hair dyeing.



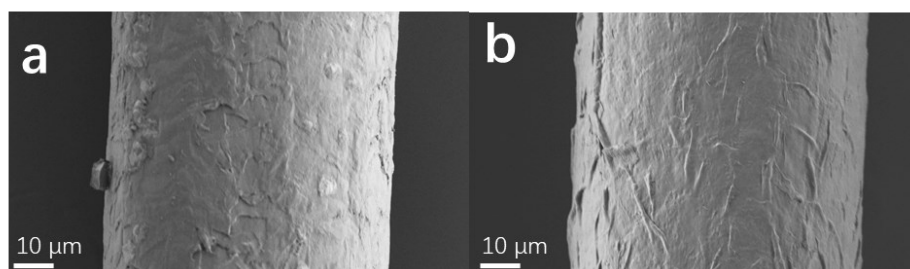
**Fig. S5** Photographs of dyeing mice hair with water (a),  $\text{Fe}^{3+}$  chelating (b),  $\text{Fe}^{2+}$  chelating (c),  $\text{Cu}^{2+}$  chelating (d) and pure polydopamine (e).

6. Images of mice after skin dyeing.



**Fig. S6** Images of BALB/c mice before (left) and after (after) treated with polydopamine assemblies. Middle image shows the dropping process of polydopamine assemblies onto mice backside.

7. Photographs of hair treated by alkali and oxidation environment.



**Fig. S7** Photographs of hair samples treated by (a) 0.01 M NaOH and (b) 3% H<sub>2</sub>O<sub>2</sub>.

### **Reference**

[1] Z.F. Gao, X.Y. Wang, J.B. Gao, F. Xia, Rapid preparation of polydopamine coating as a multifunctional hair dye, *RSC Advances*, 9 (2019) 20492-20496.