Electronic Supplementary Material (ESI) for Nanoscale.

Self-assembly of clay nanotubes on hair surface for medical and cosmetic formulations

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Supporting information

1. Human hair roughness

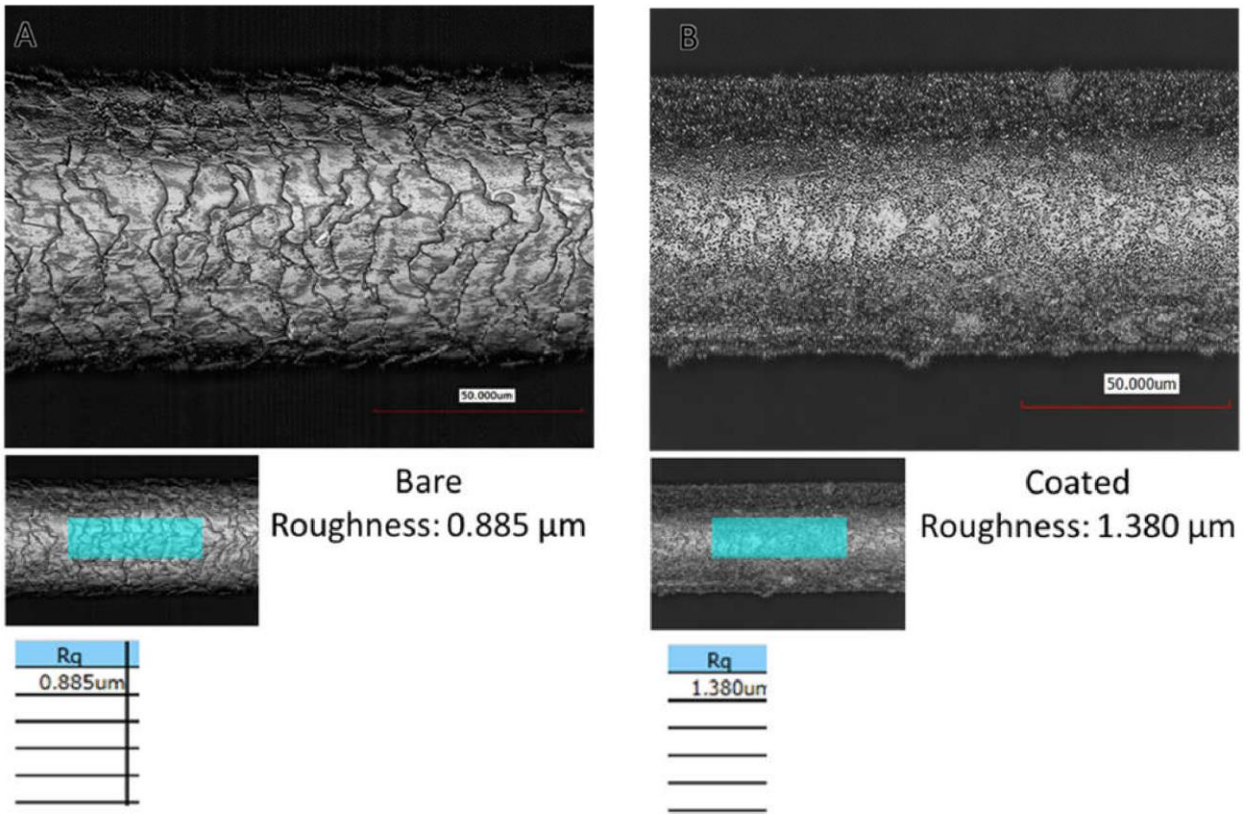
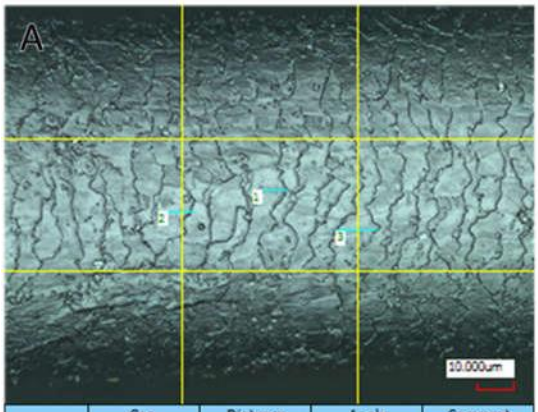


Figure S1. Visual and roughness measurement of bare (A) and coated (B) hair with the root mean square roughness (Rq) enlarged in inset.

2. Periodicity in coating by TEM, cuticle self-assembly



	Seg.	Distance	Angle	Comment
1	2-pt dist	9.227um		
2	2-pt dist	9.640um		
3	2-pt dist	10.883um		
4				
5				
6				
7				
8				
9				
10				
11				
12				
13				
14				
15				
16				
17				
18				
19				
20				
Total		29.749um	0.000°	
Max.		10.883um	0.000°	
Min.		9.227um	0.000°	
Ave.		9.916um	0.000°	
Std. DV		0.794um	0.000°	
3sigma		2.111um	0.000°	
No. of cou...				

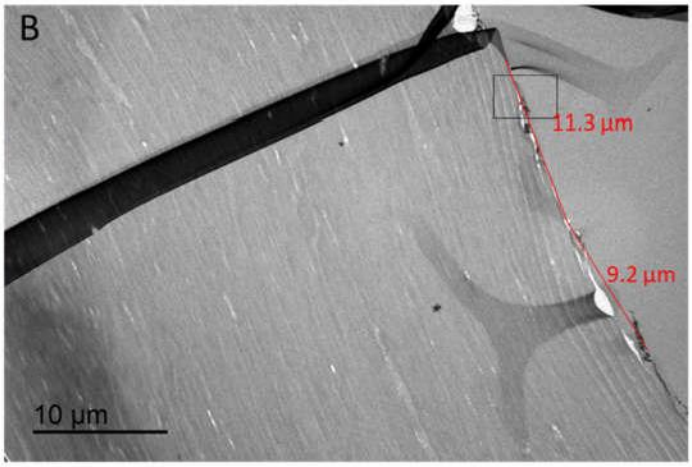


Figure S2: Profilometry measurements of pristine hair show cuticles are spaced out at an interval of ca. 10 µm away from each other (A). The TEM image (B) of longitudinal section of halloysite coated hair show dark clay deposits at a similar interval indicating the cuticle ends as sites for initiation of assembly of halloysite tubes.

3. EDX spectra of hair coated with pristine halloysite

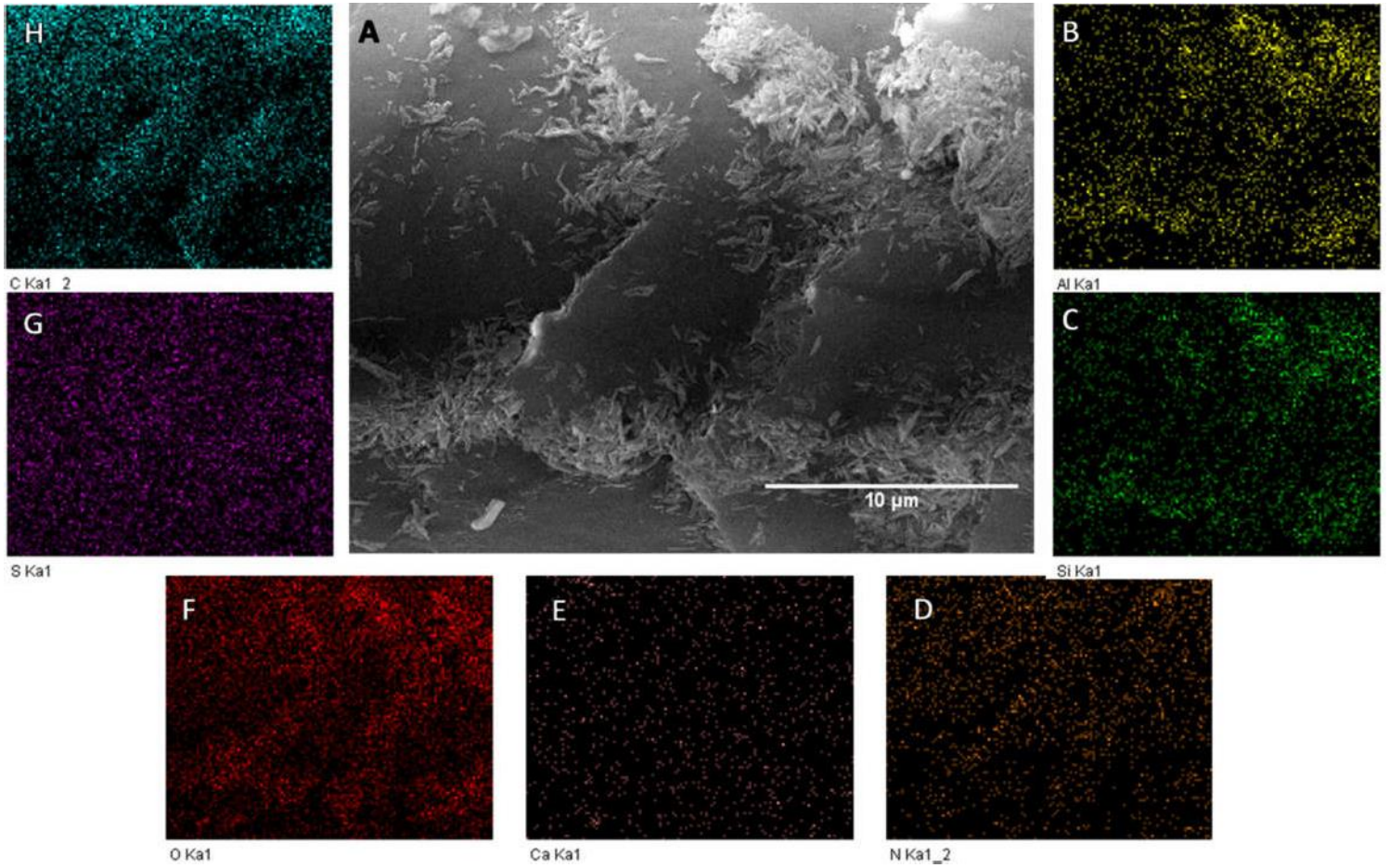


Figure S3: EDX spectra elemental maps of hair coated by pristine halloysite tubes. Original image (A) and corresponding Aluminium (B), Silica (C), Nitrogen (D), Calcium (E), Oxygen (F), Sulphur (G) and Carbon (H) maps. The Si and Al maps (B &C) are imprints of the halloysite deposit sites on the hair surface.

Table S1. Weight and atomic percentages of the elements on the coated hair fragment from Figure S3

Element	Weight%	Atomic%
N	20.13	24.58
O	60.85	65.04
Al	1.56	0.99
Si	1.47	0.90
S	15.71	8.38
Ca	0.28	0.12

4. Effect of 1-hour wash on hair coated with ODTMS-modified hydrophobic halloysite

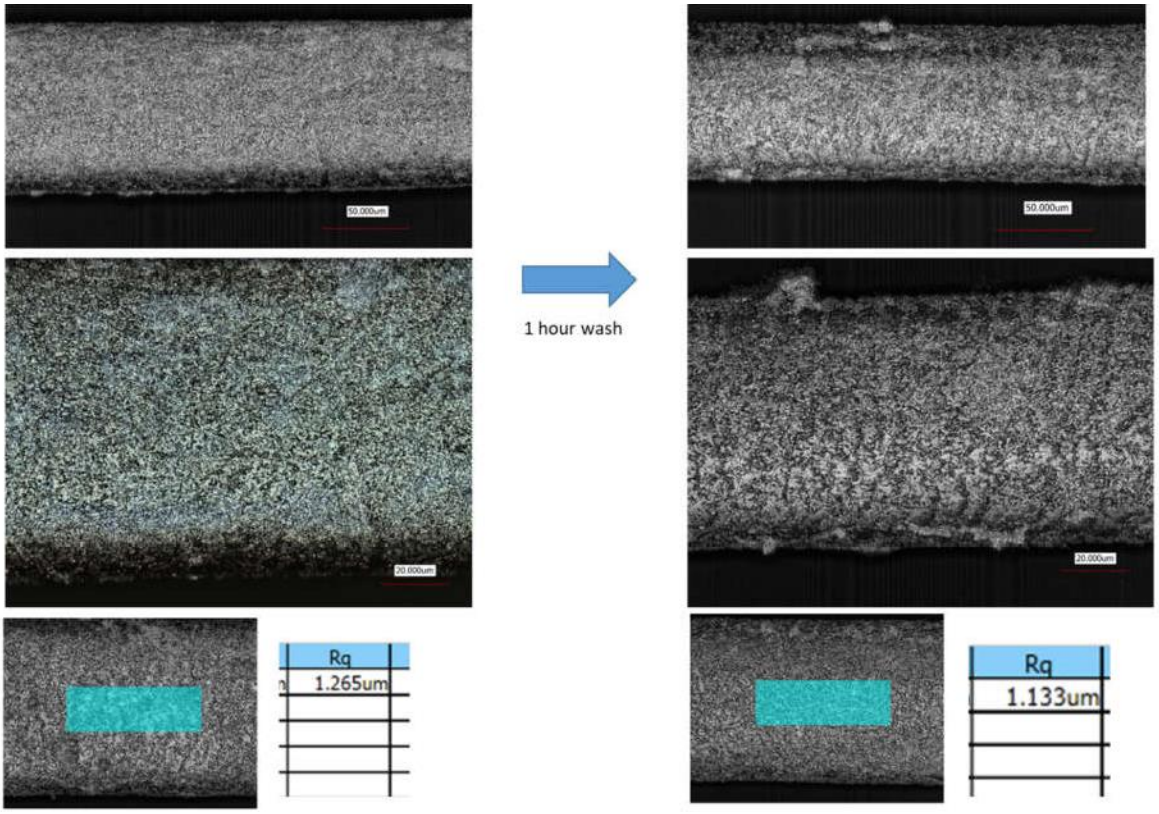


Figure S4. Coating of ODTMS-halloysite CA 57° on hair (left) is intact after 1-hour rigorous wash (right) as indicated by optical pictures and roughness measurement (Rq is root mean square roughness in inset)

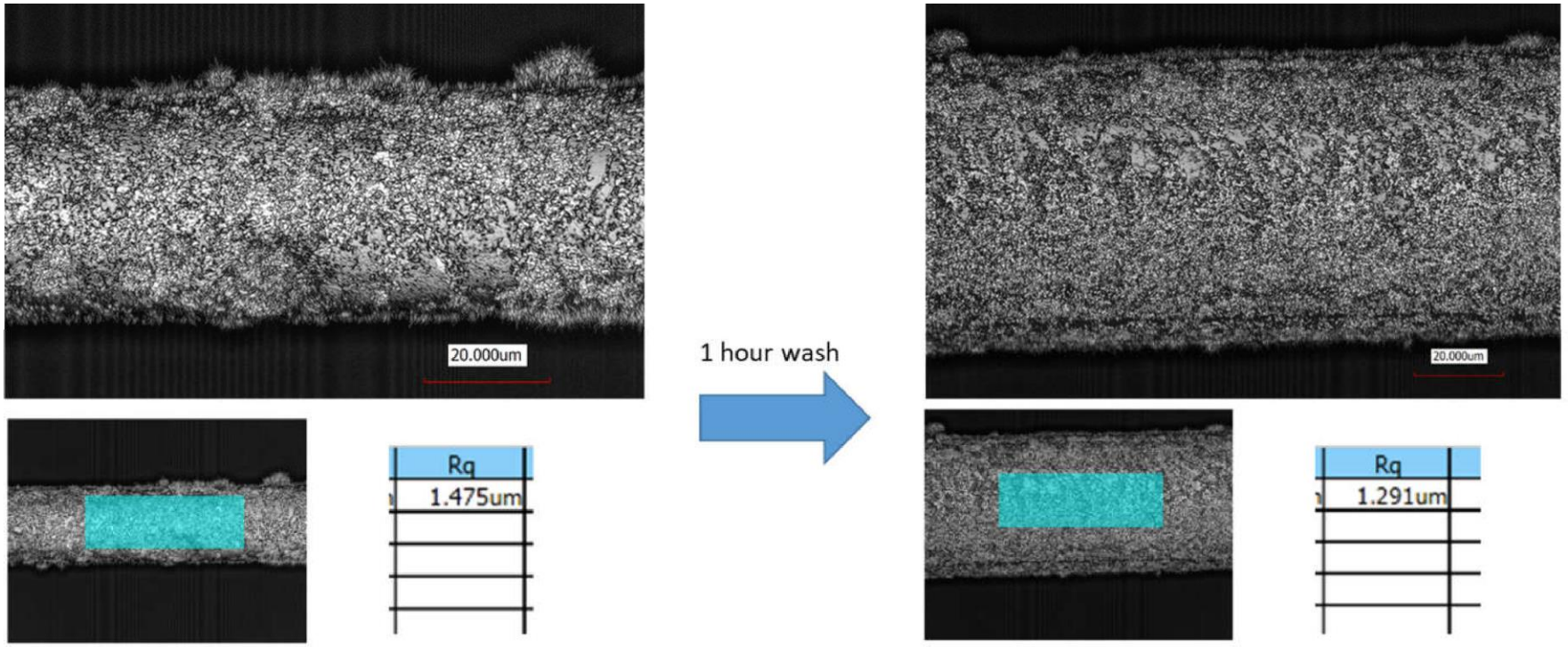
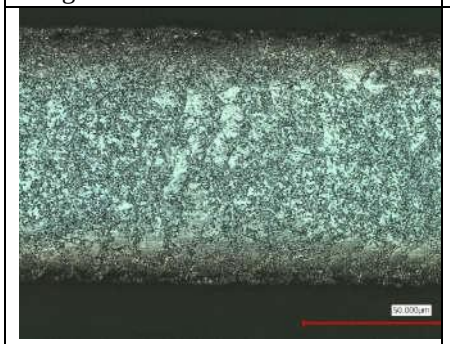
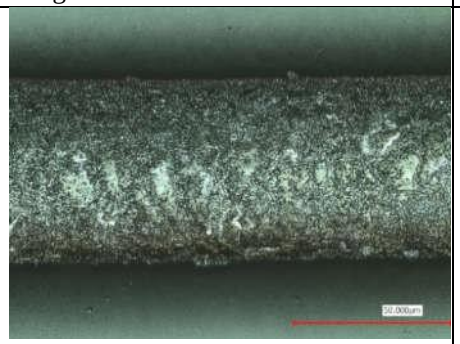
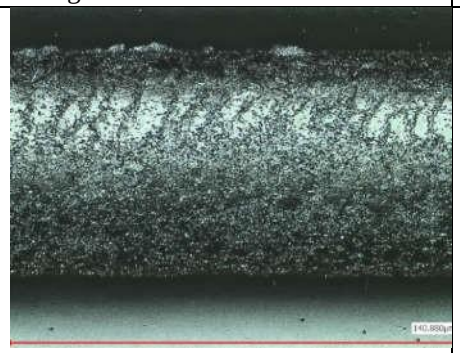
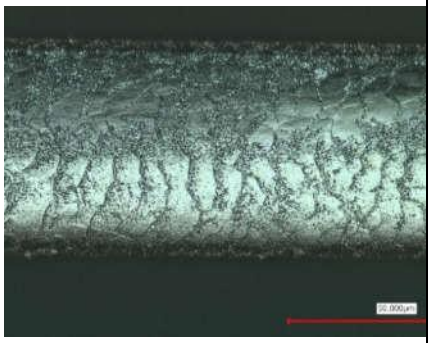
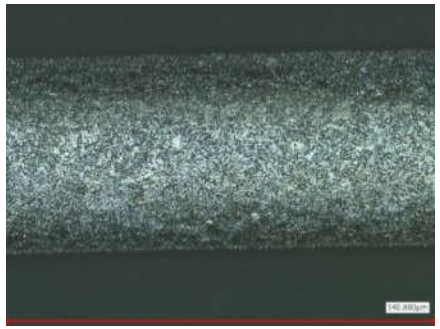
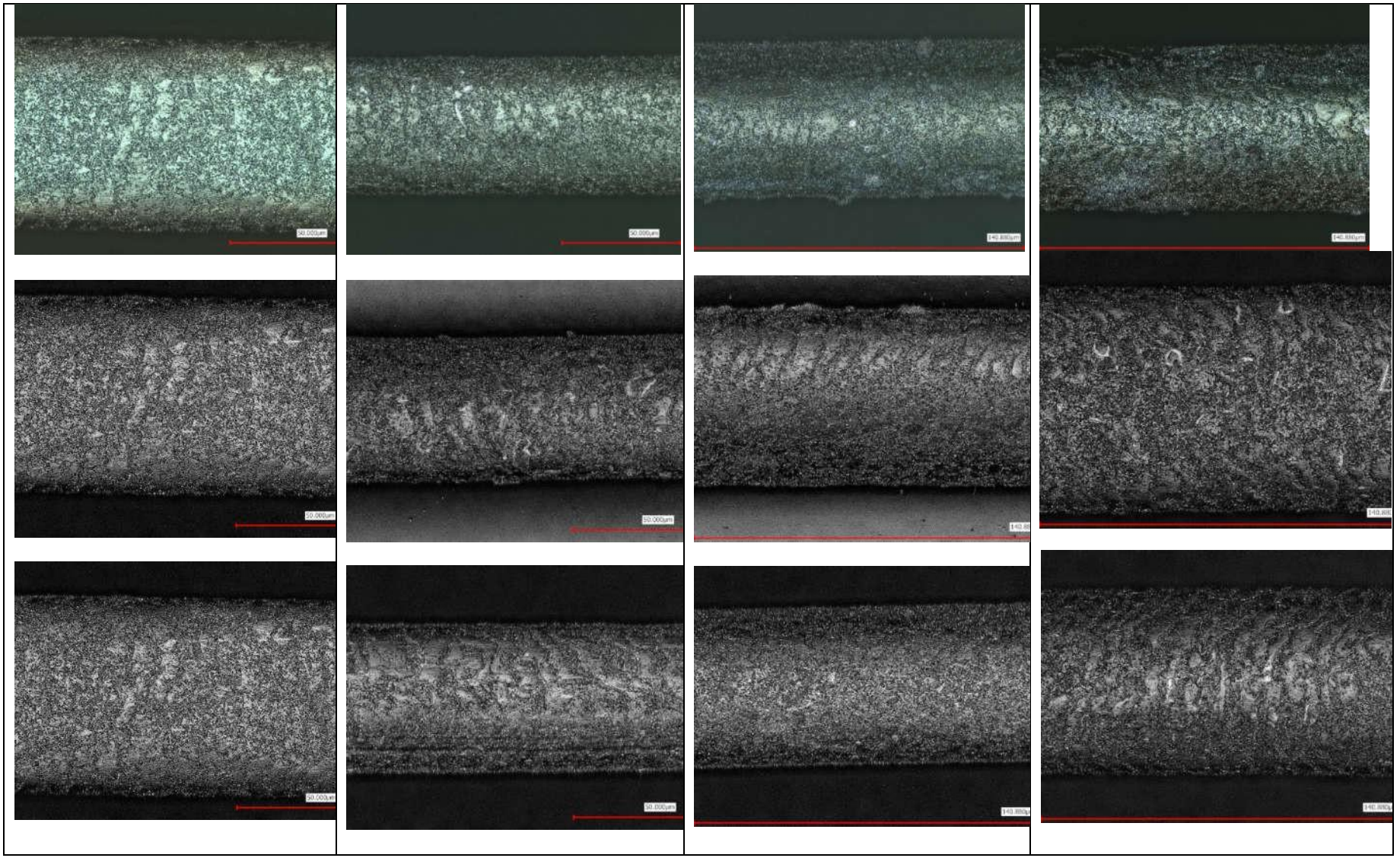


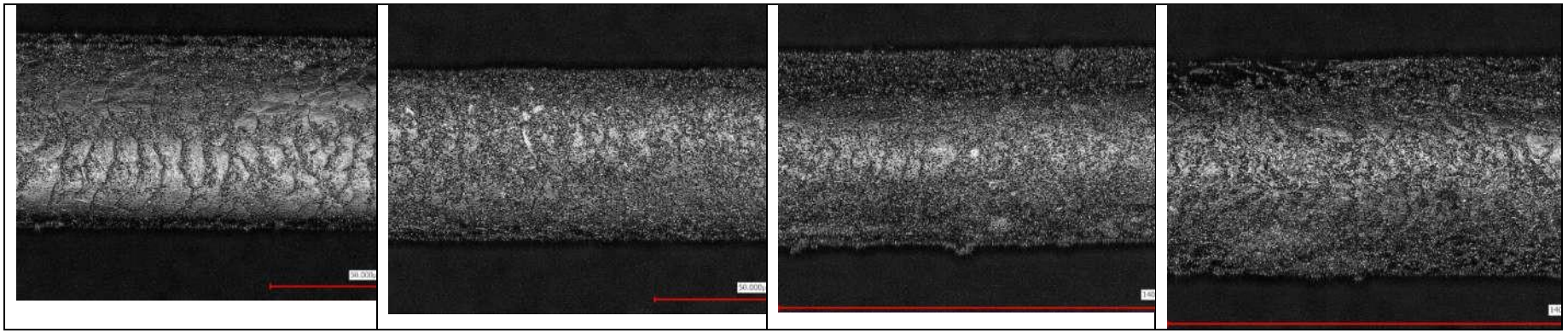
Figure S5. Coating of ODTMS-halloysite CA 116° on hair (left) is intact after 1-hour rigorous wash (right) as indicated by optical pictures and roughness measurement (Rq is root mean square roughness in inset)

5. Dispersion concentration dependent coating of pristine halloysite on young Caucasian hair

Table S2: Confocal images of hair coated with pristine halloysite at dispersion concentrations from 2 mg/ml to 20 mg/ml

2 mg/ml	5 mg/ml	10 mg/ml	20 mg/ml
			
			





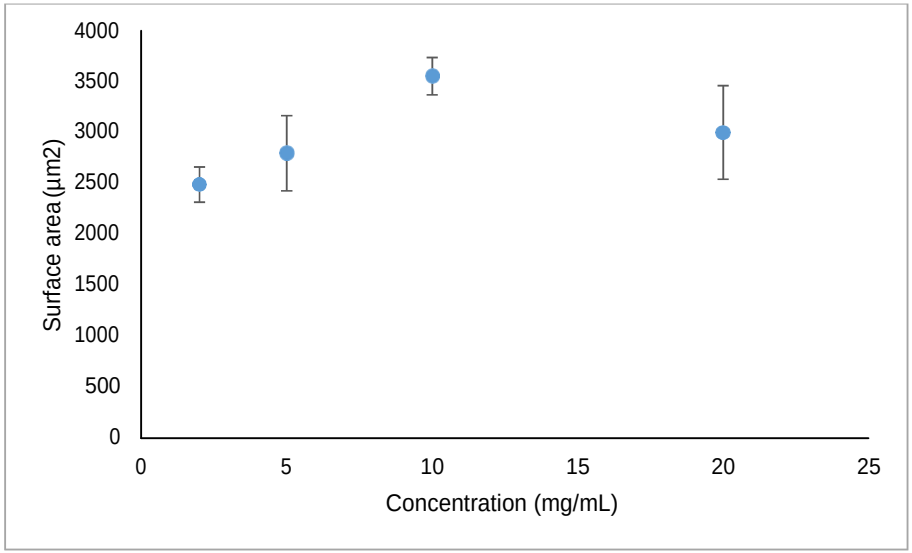


Figure S6: Profilometry surface area measurements of coated hair samples varying with dispersion concentration.

6. Loading natural dye, Lawsone into Halloysite lumen.

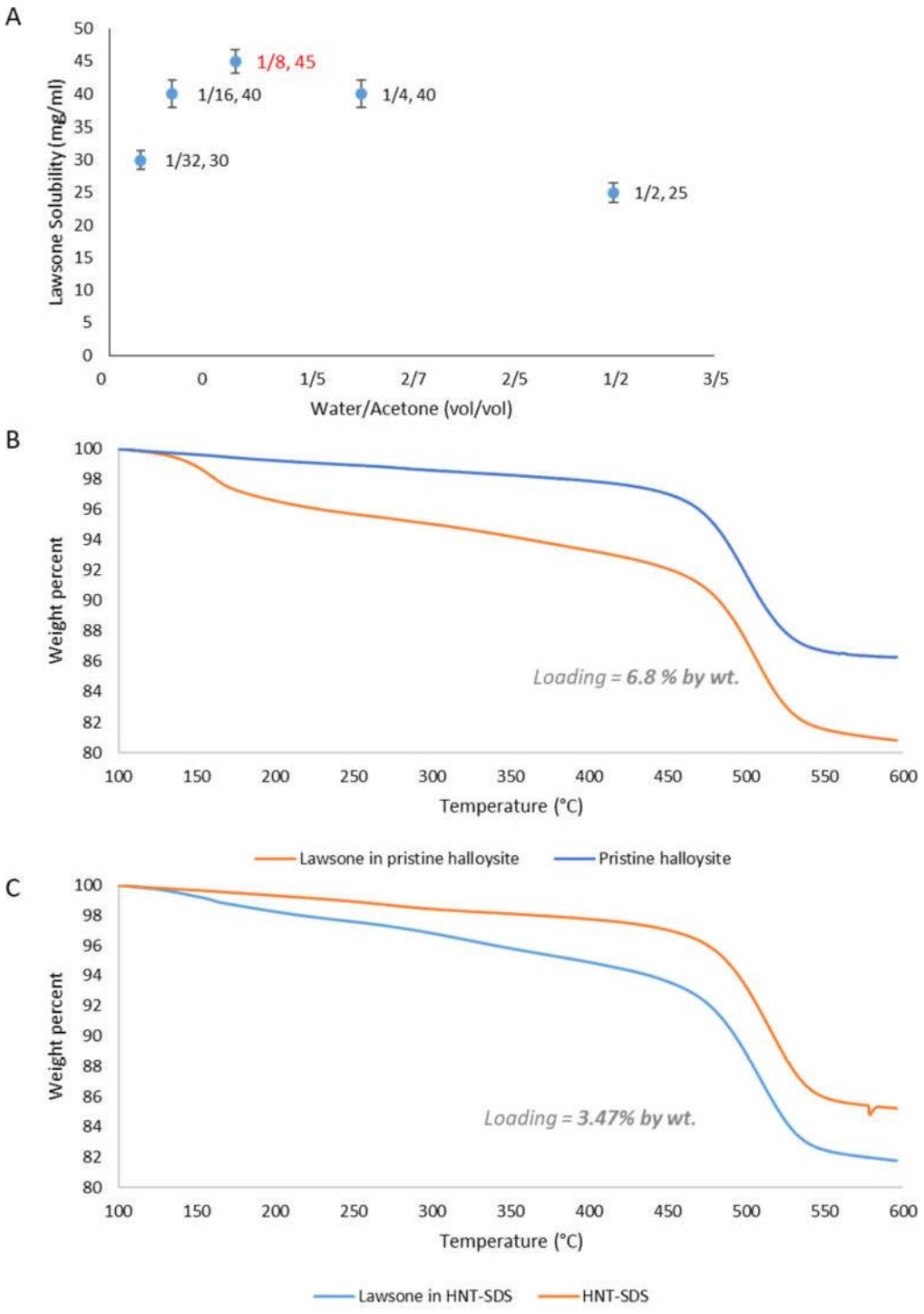


Figure S7: Optimisation of lawsone solubility (A). Lawsone was found to have increased solubility in a mixture of water to acetone, with the 1:8 mixture bearing solubility 2.25 times over pure acetone. TGA spectra of lawsone loaded in pristine (B) and HNT-SDS(C).

7. Hydrophobising lumen of Halloysite

Halloysites and Sodium Dodecyl sulfate (SDS) is mixed into a dispersion through ultrasonication with HNT:SDS weight ratio at 1:1. The concentration of both the components is kept at 2 mg/ml below the critical micellar concentration (CMC) of 2.364 mg/ml. The mixture is left stirring for 48 hours. After the stirring, the precipitate after centrifugation @ 1000 rpm, 3 min was separated and analysed. The separated part showed normal zeta potential and hence was discarded as it showed no loading. The following results are of the supernatant collected and centrifuged @ 5000 rpm, 20 min. The higher zeta potential indicates neutralization of positive lumen and Thermogravimetric analysis (TGA) shows a loading of SDS at 1.2% by wt.

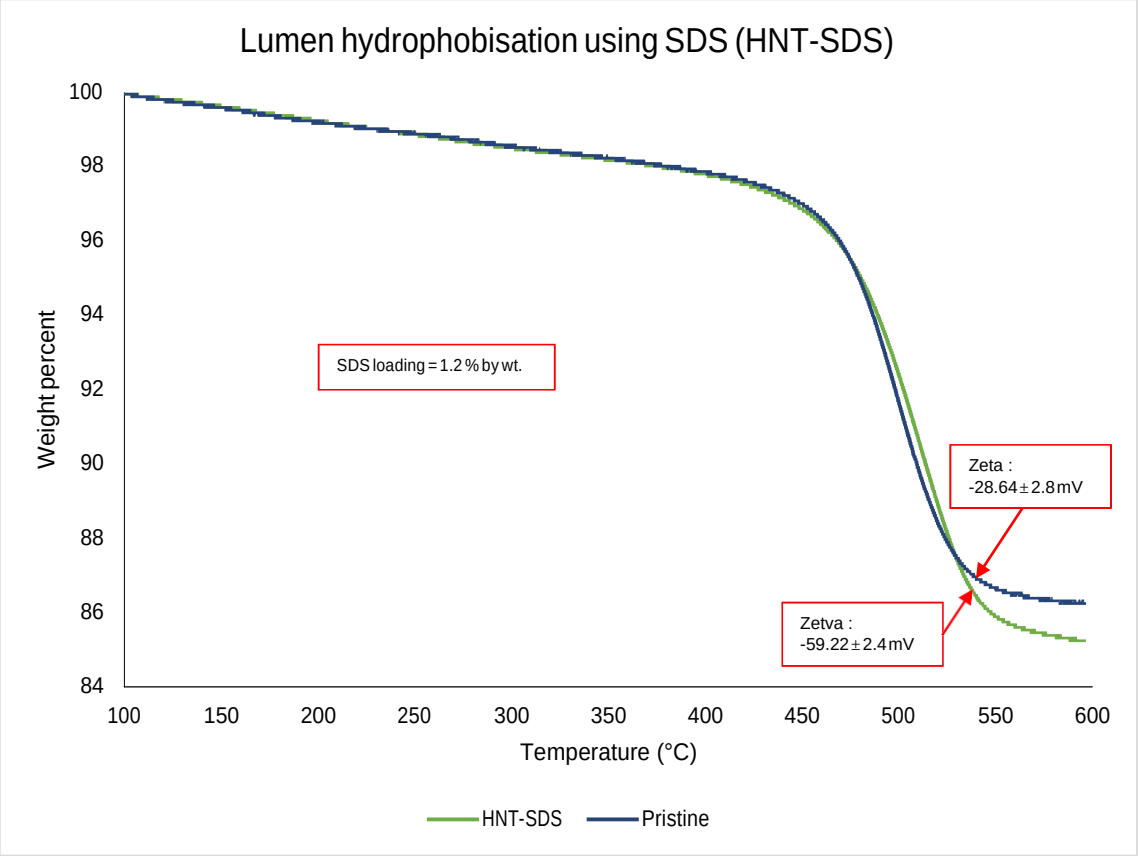


Figure S8: TGA spectra of HNT-SDS against pristine halloysite with zeta potential measurement labels showing increased negative charge on modified halloysite.

8. Halloysite coating on hair from different animal species: Dog, Cat & Horse

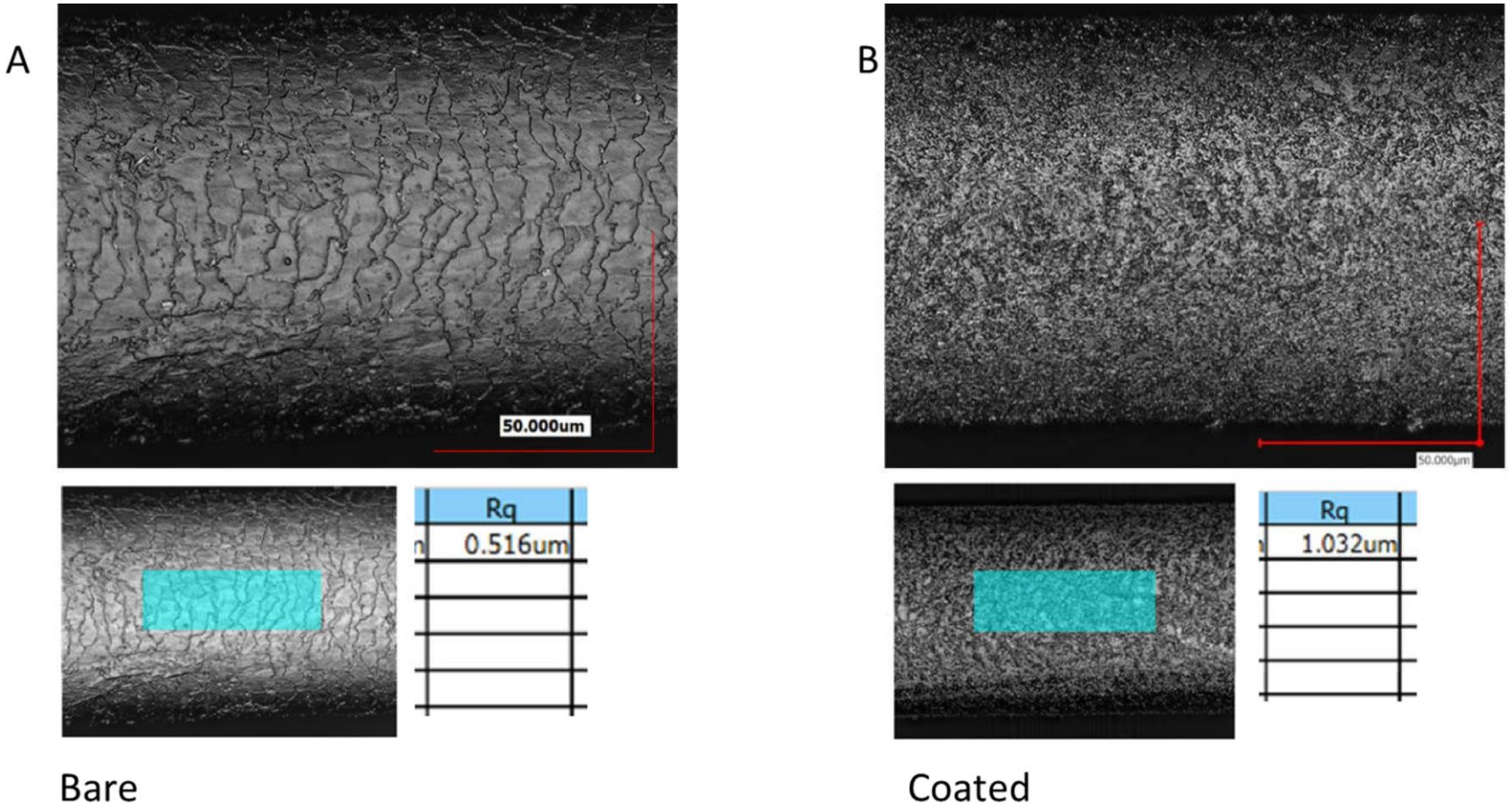


Figure S9: Visual and roughness measurement of bare (A) and coated (B) **Dog** hair with the root mean square roughness (Rq) enlarged in inset.

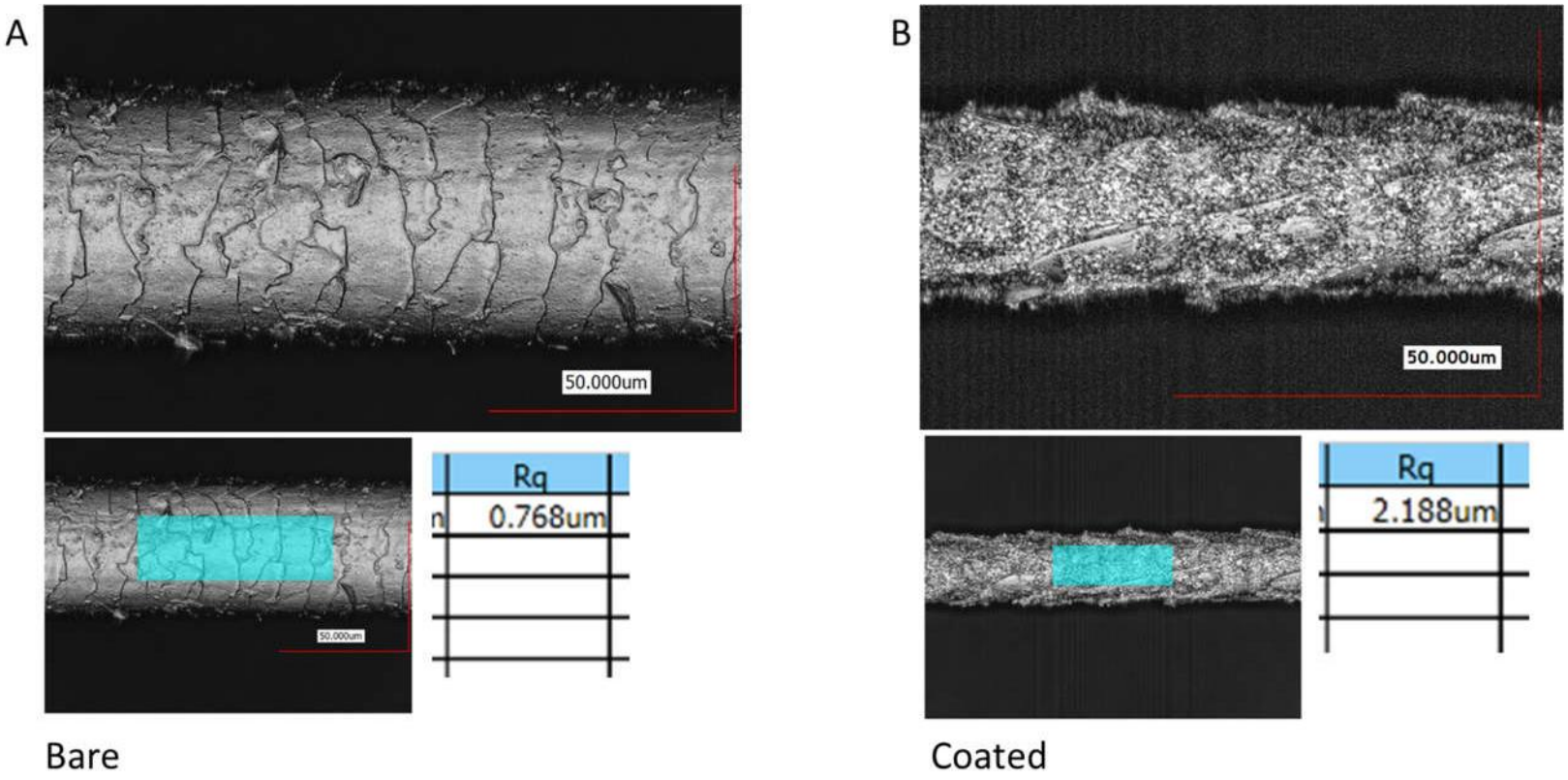


Figure S10: Visual and roughness measurement of bare (A) and coated (B) **Cat** hair with the root mean square roughness (Rq) enlarged in inset.

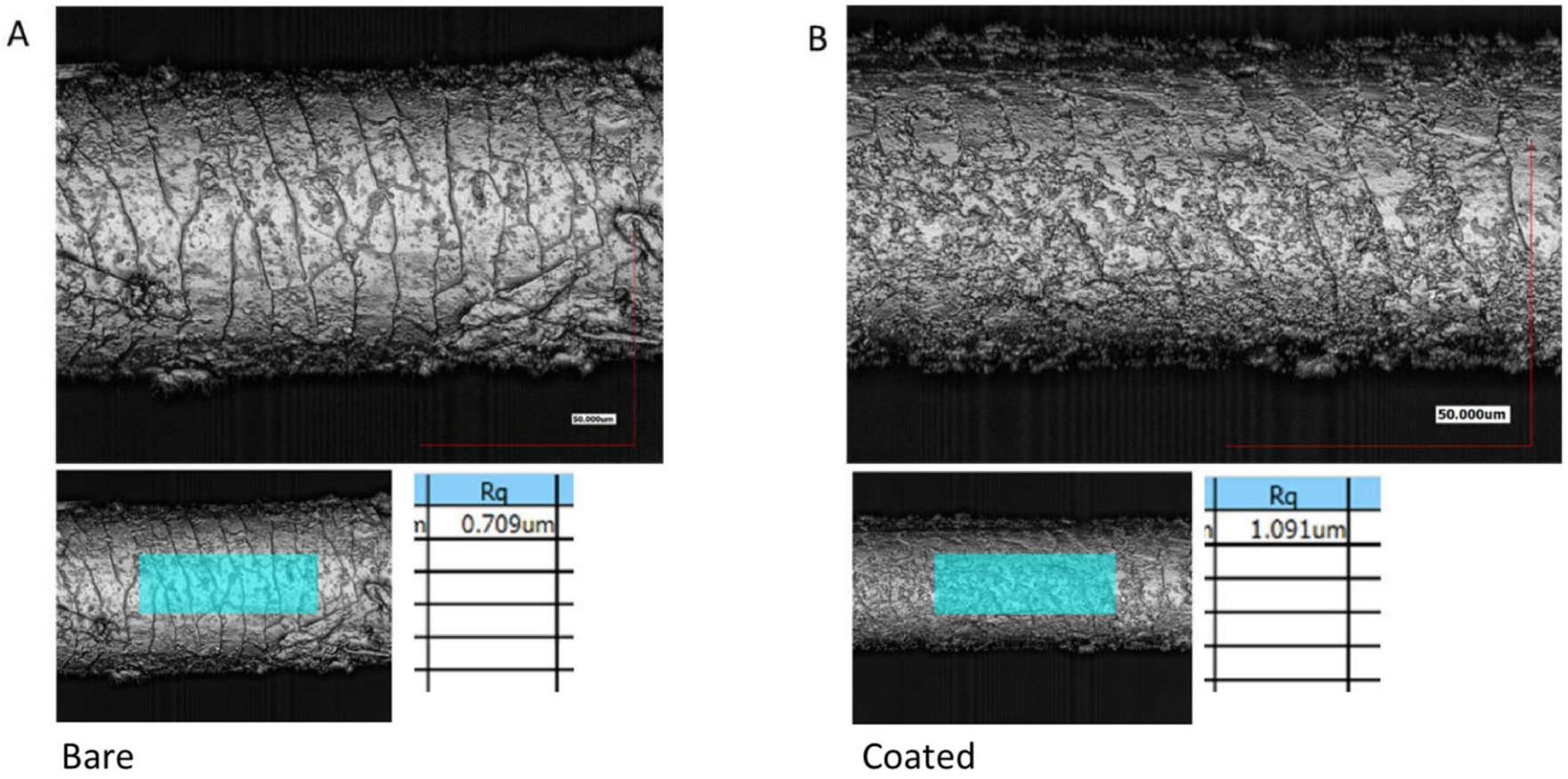


Figure S11: Visual and roughness measurement of bare (A) and coated (B) **Horse** hair with the root mean square roughness (Rq) enlarged in inset.

9. Encapsulation of different color dyes

Blue dye: Indigo

Thermogravimetric analysis (TGA) of pristine halloysite loaded with Indigo:

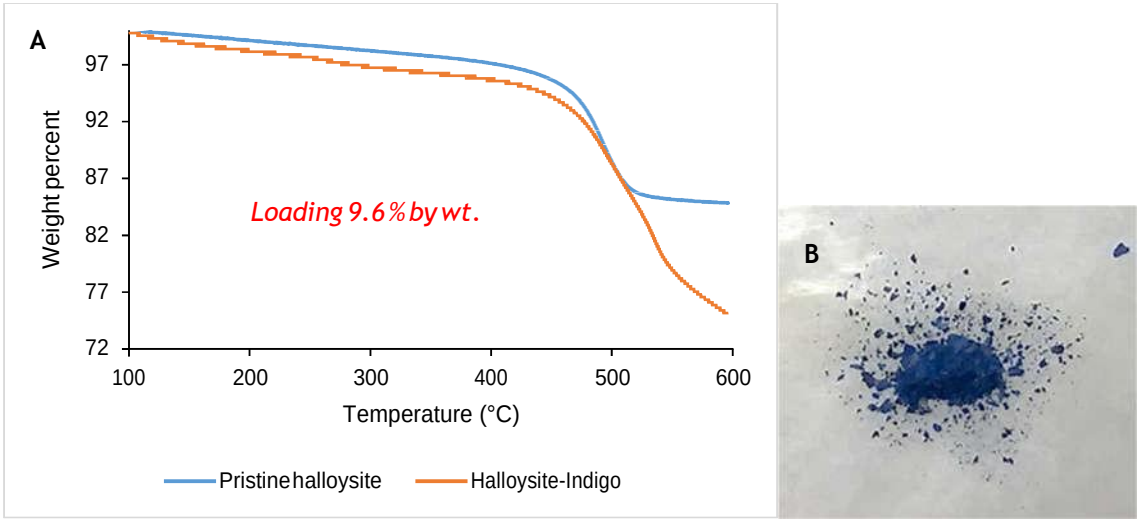


Figure S12. TGA spectra (A) and image of HNT-Indigo halloysite (B)

Red Dye: Carmine (from Cochineal)

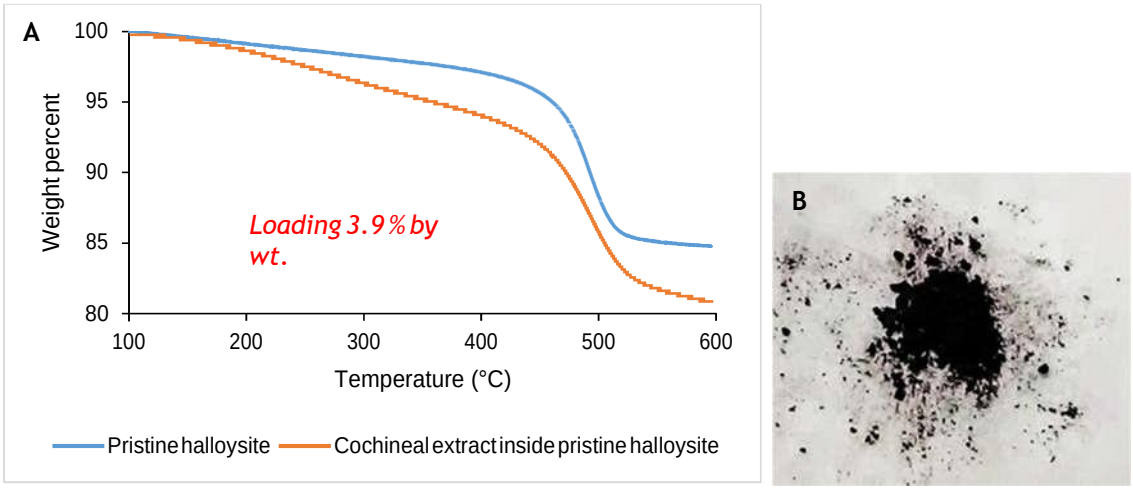


Figure S13. TGA spectra (A) and image of cochneal extract (carmine) in pristine halloysite (B)

Purple dye: Shikonin (from Alkanet plant)

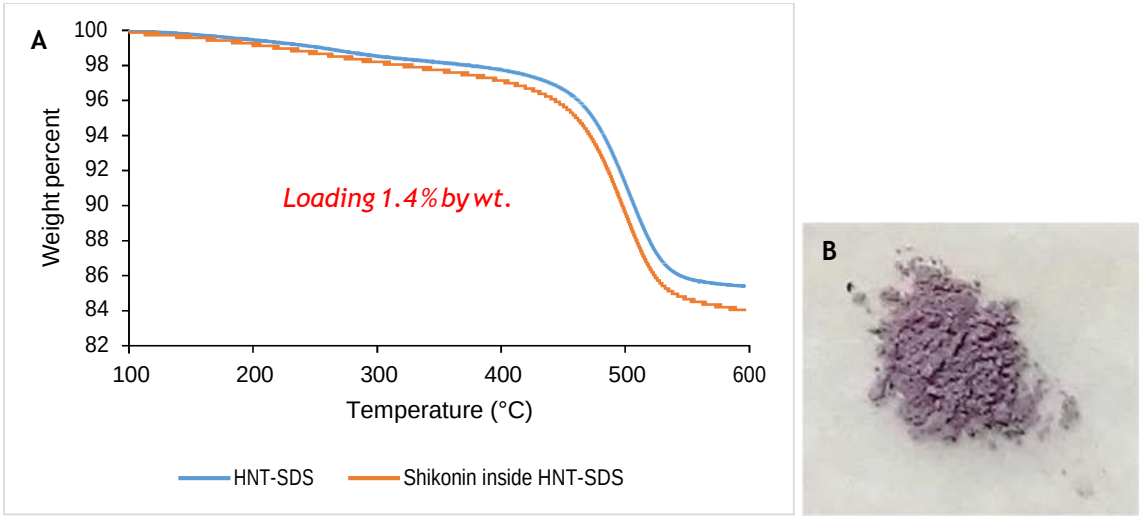


Figure S14. TGA spectra (A) and image of shikonin from extract of Alkanet root encapsulated in pristine halloysite (B)

Figure S15. Lawsone loaded HNT-SDS.