Supporting information

Cilia-Mimetic Hairy Surfaces Based on End-Immobilized Nanocellulose Colloidal Rods

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Figure S1. QCM-D plot of CNC/CNC-SH adsorption on gold surface at concentrations 0.1 mg ml⁻¹ and 1 mg ml⁻¹. In both plots (temp 23°C, flow rate 100 μ l min⁻¹), the CNC-SH/CNC dispersion was introduced at 7 minutes time point, followed by MQ at 37 minutes time point, as indicated by arrows in the plots. Frequency change upon adsorption (a) and Dissipation response after adsorption (b).



Figure S2. QCM-D plot of adsorption involving PS particles (0.01 wt vol⁻¹) onto a hairy layer of CNC-SH on gold surface. In all plots (temp 23°C, flow rate 100µl min⁻¹), the CNC-SH dispersion was introduced at 7 minutes time point, followed by MQ at 17 minutes time point and then PS particles at 28 minutes time point, as indicated by arrows in the plots. Frequency-dissipation vs time plot for 50nm (a), 200nm (b) and 1µm (c) PS particles.



Figure S3. Results of XPS analysis of CNC-SH hairy surface exposed to 0.01 wt vol⁻¹ % of 50 nm, 200 nm and 1 μ m PS particles. Ratio of relative atomic percentage of oxygen and carbon O/C (a), relative atomic percentage of Au (b). The data presented here is average of at least three measurements at three different locations on each sample.

Selective adsorption of polystyrene nanoparticles on the hairy surface. Cilia in combination with a mucus gel layer keep the lung airways clean from debris due to reduced adsorption. Even if not fully corresponding to the complex biological case, we next explored whether the hairy surface allow size selective adsorption/rejection of nanoparticles. By performing such studies using QCM-D, we can obviously provide beating of the hairs due to the vibrating resonator. Adsorption of polystyrene particles (PS) with three different sizes 50 nm, 200 nm and 1 µm on CNC-SH modified Au surfaces were studied. Surprisingly, the QCM-D resolution did not suffice to investigate the differences in adsorption (Supporting information Figures S2a, S2b and S2c). By contrast, XPS allowed observing

differences between the CNC-SH SAM subjected to adsorption with nanoparticles of different sizes. The relative atomic percentages of oxygen, carbon and gold corresponding to different PS particle adsorbed surfaces are presented in supporting information (Table T1). The ratio of relative atomic percentage of oxygen and carbon (O/C) of CNC-SH SAMs exposed to PS particles with different sizes are shown in Figure S3 a. Adsorption involving 1µm PS particle does not alter the O/C value relative to that of CNC-SH modified surface, while both 50 nm and 200 nm PS particles resulted in a decrease in the value of the same. Since the O/C value for PS particles (0.043) is much smaller when compared to that for CNC (0.21), adsorption of PS particles would result in a decrease in the O/C ratio. Hence we speculate that the altered O/C ratio upon exposure to smaller particles could be due to binding of these smaller particles. The relative atomic percentage of Au increases in case of exposure experiments involving 50 nm and 200 nm PS particles, while that for 1µm particles remains unaltered. One possible explanation for the increase in Au atomic percentage could be desorption of CNC-SH upon exposure to smaller particles (50 nm and 200 nm). In future we intend to use fluorescently labeled particles of varying sizes to further understand the size-selective particle binding property of the colloidal hairy surface and check the ability of the colloidal hairy surface to mimic an important aspect of biological cilia.

Table T1. Results of XPS analysis of surfaces after QCM-D study involving adsorption of PS particles (0.01 wt vol⁻¹) onto a hairy layer of CNC-SH on gold surface. The elemental relative atomic percentages of carbon, gold and oxygen corresponding to CNC-SH SAMs exposed to 50 nm, 200 nm and 1 μ m PS particles is shown in the table. The data presented here is average of at least three measurements at three different locations on each sample.

Sample	rel At% C	rel At% Au	rel At% O
CNC-SH SAM	54.9 ± 1.3	27.5 ± 2	13.3 ± 0.9
CNC-SH SAM + 50 nm PS	52.2 ± 3.1	33.3 ± 3.1	10.7 ± 0.7
CNC-SH SAM + 200 nm PS	50.2 ± 2.5	35 ± 0.8	10.1 ± 0.6
CNC-SH SAM + 1 µm PS	56.3 ± 0.2	28.0 ± 0.9	13.1 ± 0.4

Table T2. Zeta potential values of 50 nm, 200 nm and 1 μm PS particles PS particles measured using Zetasizer Nano ZS90, Malvern instruments Ltd, UK.

Particle	Zeta potential (mV)	
50 nm PS	-47.4 ± 11.1	
200 nm PS	-46.7 ± 17.0	
1 μm PS	-42.4 ± 9.1	