Supplementary Information

Hydrogel-tissue adhesion by particle bridging: sensitivity to interfacial wetting and tissue composition

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1. Capillary transport calculation in tissue and coating porosities.

We describe the kinetics of capillary rise within the porosities of the liver lobules as well as within the porosities of the nanoparticle coating using the Lucas and Washburn law as follows:[1, 2]

$$X(t) = \left(\frac{\gamma R \cos\theta}{2\eta} \right)^{1/2} t^{1/2} \quad \text{(S1)}$$

where *X(t)* is the position of the moving fluid rising in the pores as a function of time *t*, γ is the surface tension of the liquid, R is the radius of the capillary tube, θ is the contact angle and η is the liquid viscosity.

From Equation (S1), we express the time t* for physiological fluids to rise through a medium of porosity Φ and form a layer of thickness h* wetting the surface of the medium as:

$$t^* = \left(\frac{h^*}{\Phi} \right)^2 \left(\frac{2\eta}{\gamma R \cos\theta} \right) \quad \text{(S2)}$$

In the case of the parenchyma, this calculation can be used to assess the time necessary for the free liquid contained in the liver lobule to rise to the interface through small vessels and form a layer of thickness h*. Considering blood (γ = 55.9 mN/m; η = 0.0035 N.s/m²)[3, 4] rising into hydrophilic pores (θ ~ 0°) and selecting published values for the sinusoid radius (8 µm),[5] and the porosity of the parenchyma (0.3),[6-8] one ends up with a characteristic time t* = 0.02 ms to wet the parenchyma surface with a fluid layer of thickness h* = 10 µm.

In the case of the nanoparticle coating, Equation (S2) can be employed to evaluate the time necessary for an interfacial liquid layer of thickness h* to be absorbed by capillarity through the nanoparticle coating porosities. In this case, the porosity Φ is evaluated thanks to the following equation:

$$\Phi = \frac{V_{coat} - V_{SiO2}}{V_{coat}} \quad \text{(S3)}$$

where V_{coat} is the total volume of the coating (including the "empty" porosities) and V_{SiO2} is the volume occupied by silica within the coating. For a given surface *S* of coating, the volume of the coating can be approximated to $V_{coat} = S \cdot < D_{agg} >$ where $< D_{agg} >$ is the average diameter of the silica aggregates (corresponding to the thickness of the coating). The volume occupied by silica V_{SiO2} is equal to the mass of the coating on the surface S divided by the density of amorphous silica d_{SiO2}. Equation S3 can thus be rewritten as:

$$\Phi = \frac{< D_{agg} > \cdot S - {m_c \cdot S}/{d_{SiO2}}}{< D_{agg} > \cdot S} = \frac{< D_{agg} > - {m_c}/{d_{SiO2}}}{< D_{agg} >} \quad \text{(S4)}$$

where m_c is the area density of the coating mentioned previously.

Taking $< D_{agg} >$ equal to 5 µm as measured from SEM images (Figure 1-D), m_c equal to 0.84 mg.cm^{-2} as measured experimentally, and d_{SiO2} equal to 2.3, we find a porosity fraction $\Phi = 0.27$ in the nanoparticle coating. Introducing this value in equation S4 and using porosity radius values of the order of 0.1-10 µm (as can be roughly estimated from SEM images – see Figure S3), the resulting time t^* necessary for an interfacial liquid layer of 10 µm-thickness to be absorbed by capillarity through the pores of coating is of the order of 0.002-0.2 ms.

2. Supplementary Figures

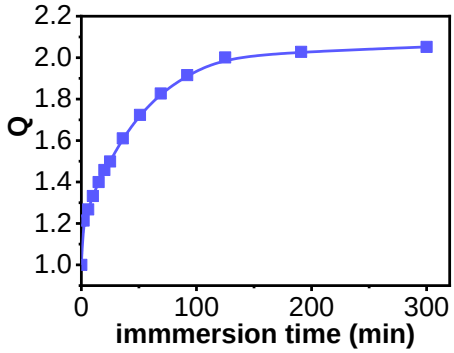

Figure S1: Swelling ratio of a 1 mm-thick PEG film as a function of immersion time in water.

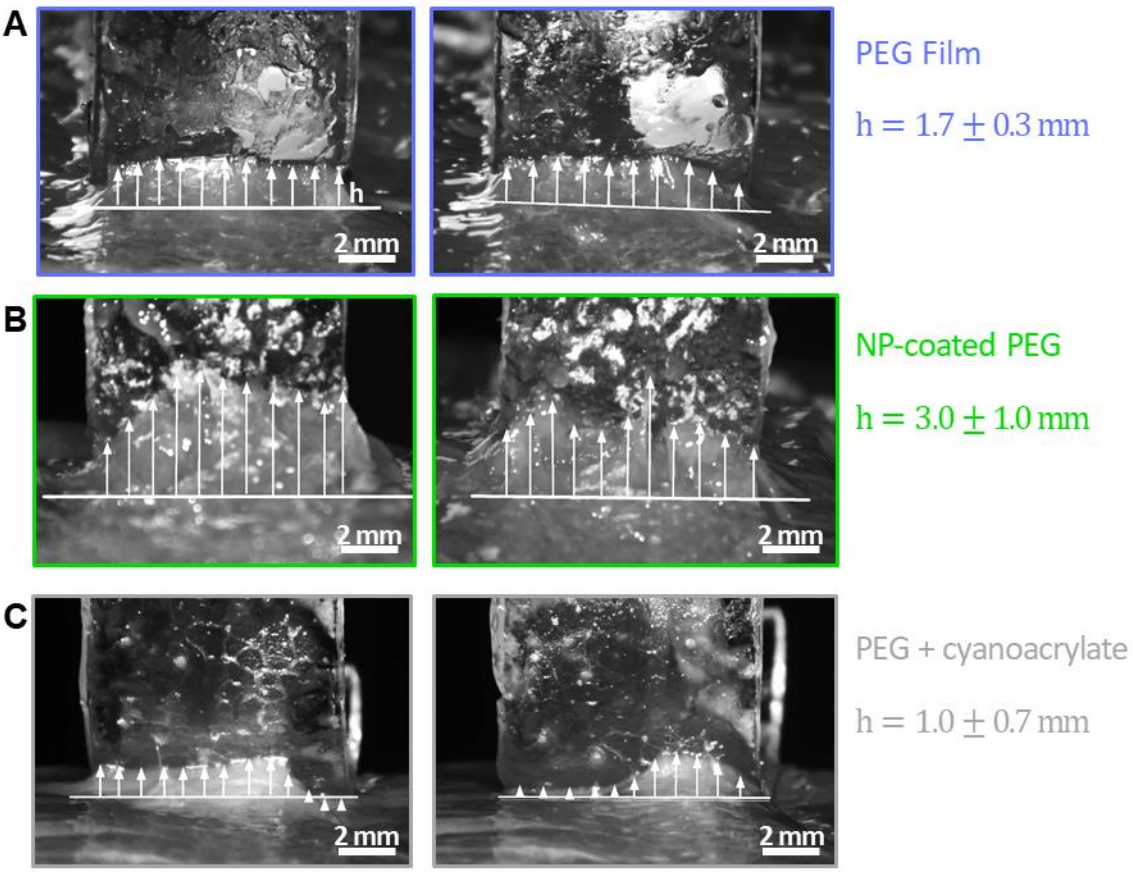

Figure S2: Measurement of the height h of the detachment line above the plane of the film-tissue interface during steady-state peeling in the case of a bare PEG film (**A**), an NP-coated PEG film (**B**) and a PEG film covered with medical grade cyanoacrylate (**C**). Measurements of h were performed on at least 50 different locations (represented by the white arrows) randomly selected over at least 4 different snapshots of the peeling front of each system.

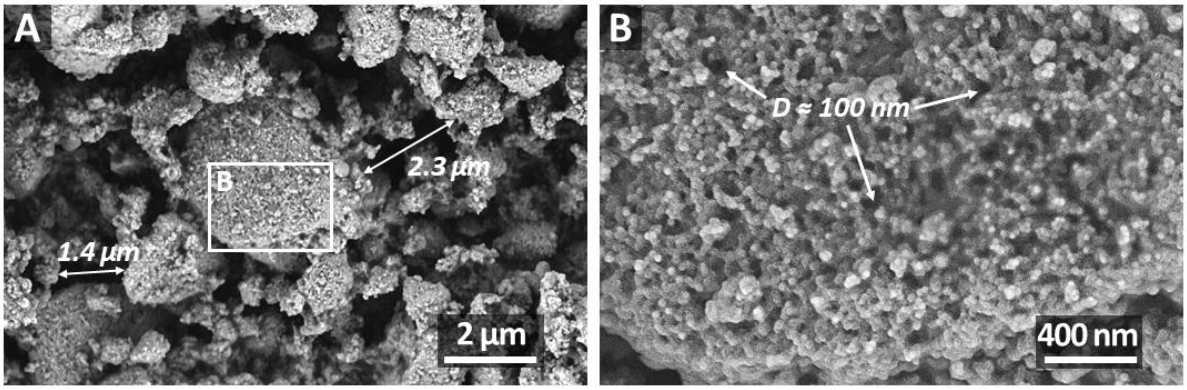

Figure S3: SEM images of the NP coating on which the size of pores is displayed. In particular, the image (A) show microscopic inter-aggregate porosities while image (B) displays intra-aggregate porosities in the submicron range. From these images the coating porosities is roughly estimated to be of the order of 0.1 to 10 µm.

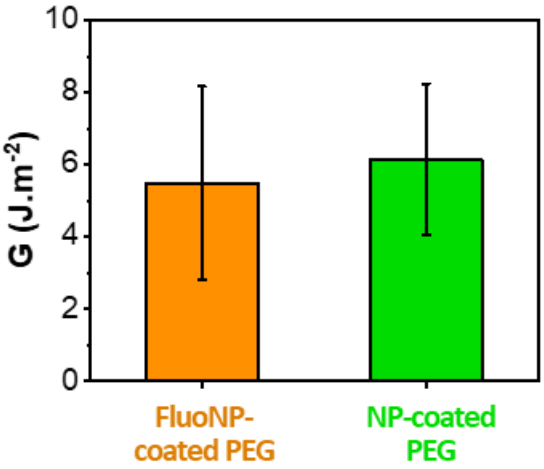

Figure S4: Comparison of the adhesion energy obtained with a PEG film coated with a fluorescent NP-coating (orange) and with a non-fluorescent NP-coating (green) on two samples of the same freshly dissected liver (i.e. same level of tissue hydration *H* in both cases).

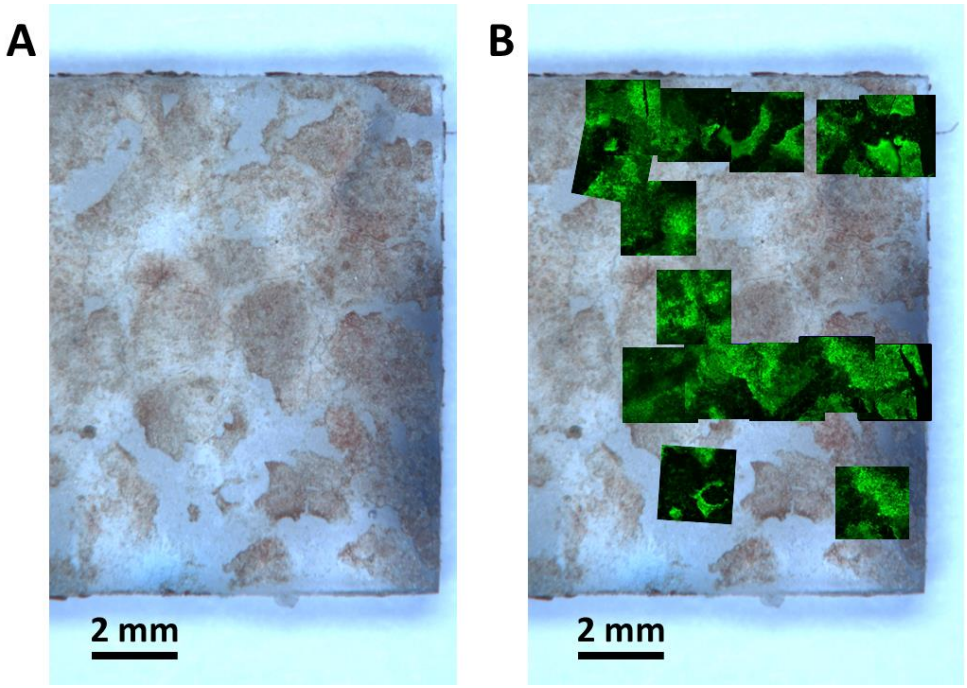

Figure S5: (**A**) Photograph a PEG film coated with a fluorescent NP-coating after peeling in the adhesive regime (v_{abs}/v_{free} >1) on a freshly dissected liver sample. (**B**) Same photograph displaying superimposed fluorescent micrographs taken in some areas. These observations show that the areas where liver tissue was transferred correspond to areas where the fluorescent NP-coating is still present on the PEG surface after peeling.

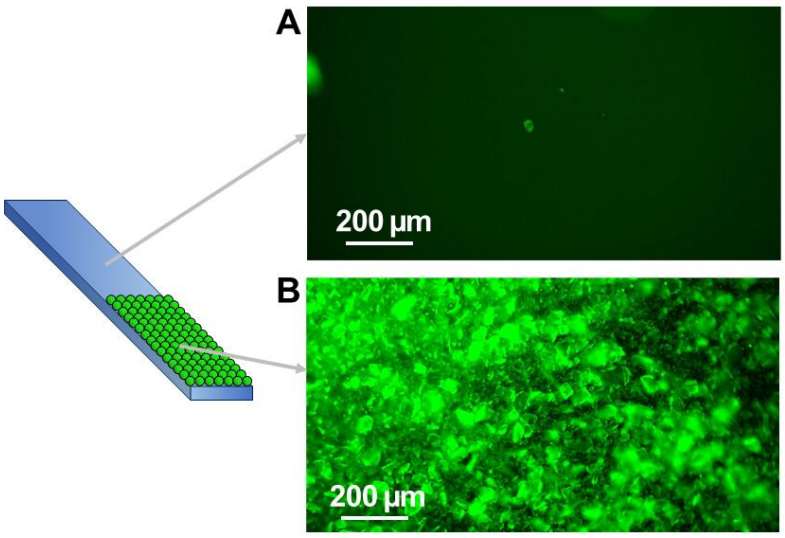

Figure S6: Fluorescent micrographs of an uncoated (A) and NP-coated (B) PEG film before peeling.

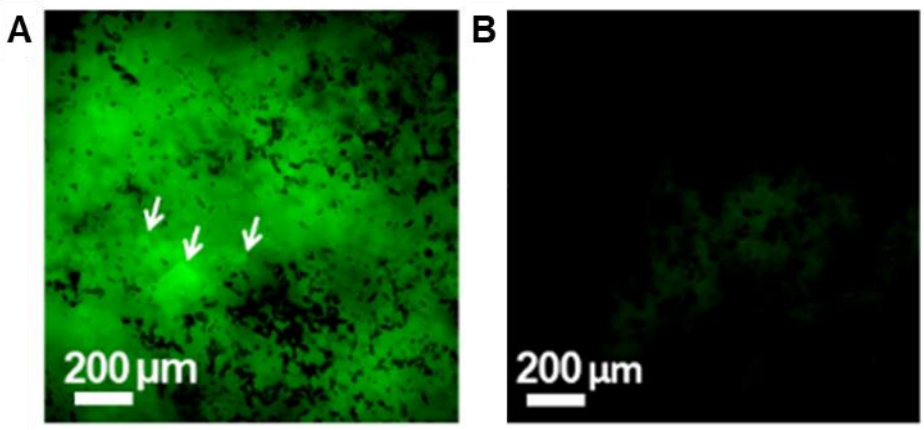

Figure S7: (A) Fluorescent micrograph taken after a peeling experiment on a liver surface which was in contact with a NP-coated PEG film. (B) A fluorescent micrograph of a pristine liver surface is shown for comparison.

3. Supplementary References

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