

## Supportive Information

### Multifunctional MRI/PET Nanobeacons Derived from the In situ Self-Assembly of Translational Polymers and Clinical Cargo through Coalescent Intermolecular Forces

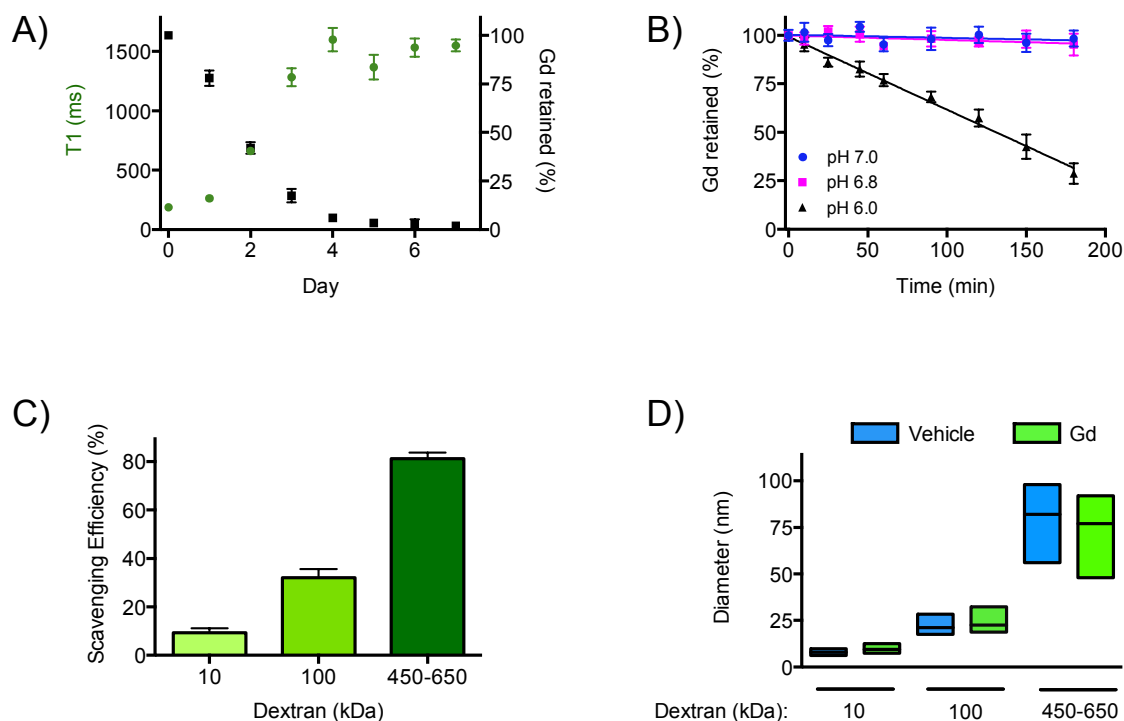
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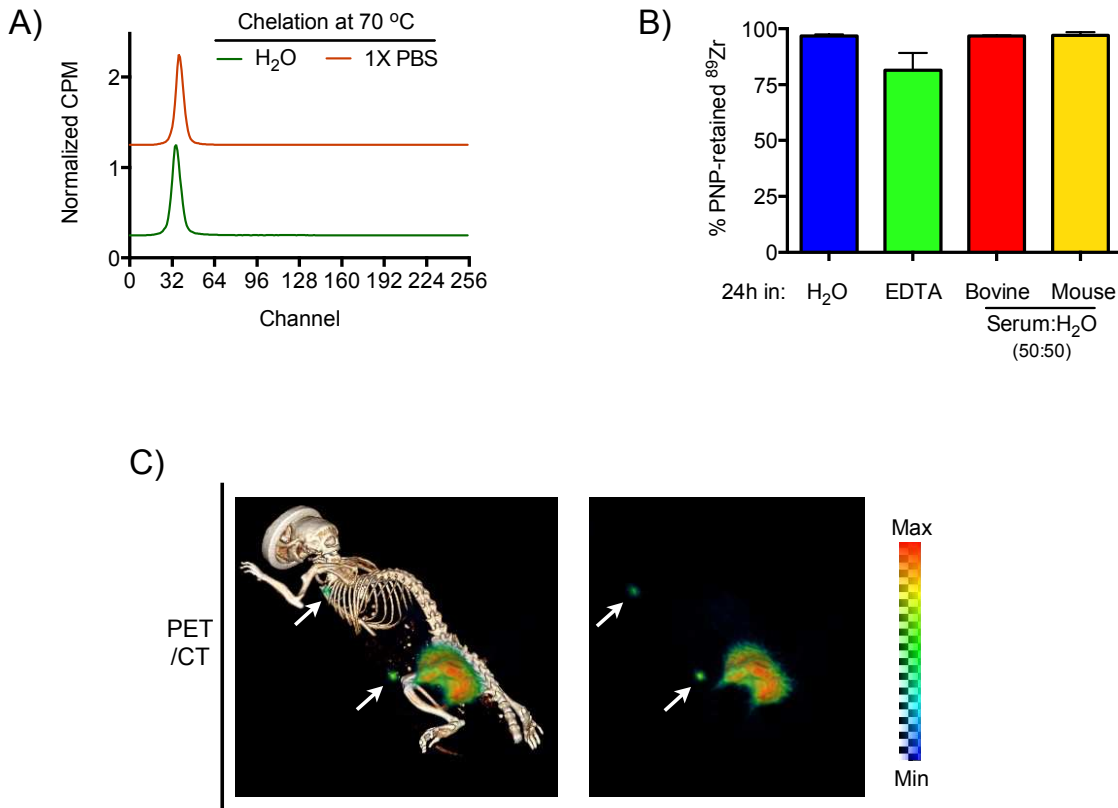
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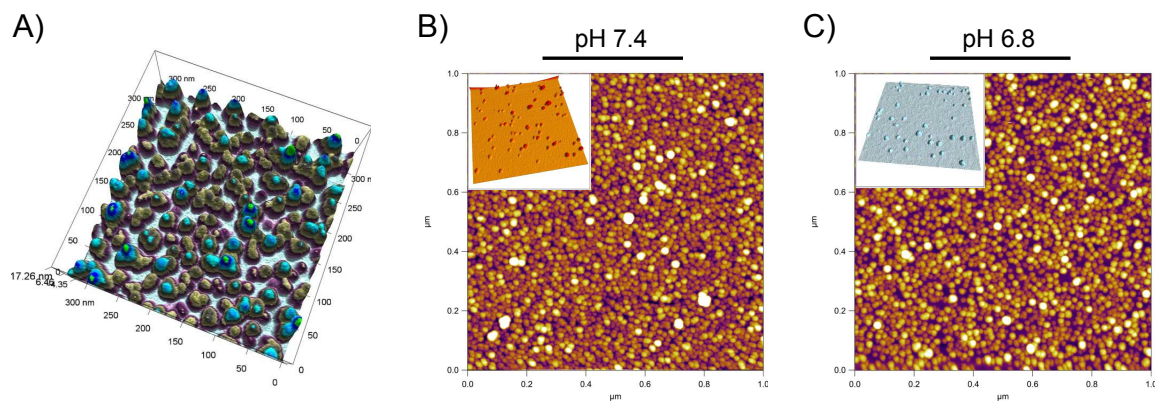
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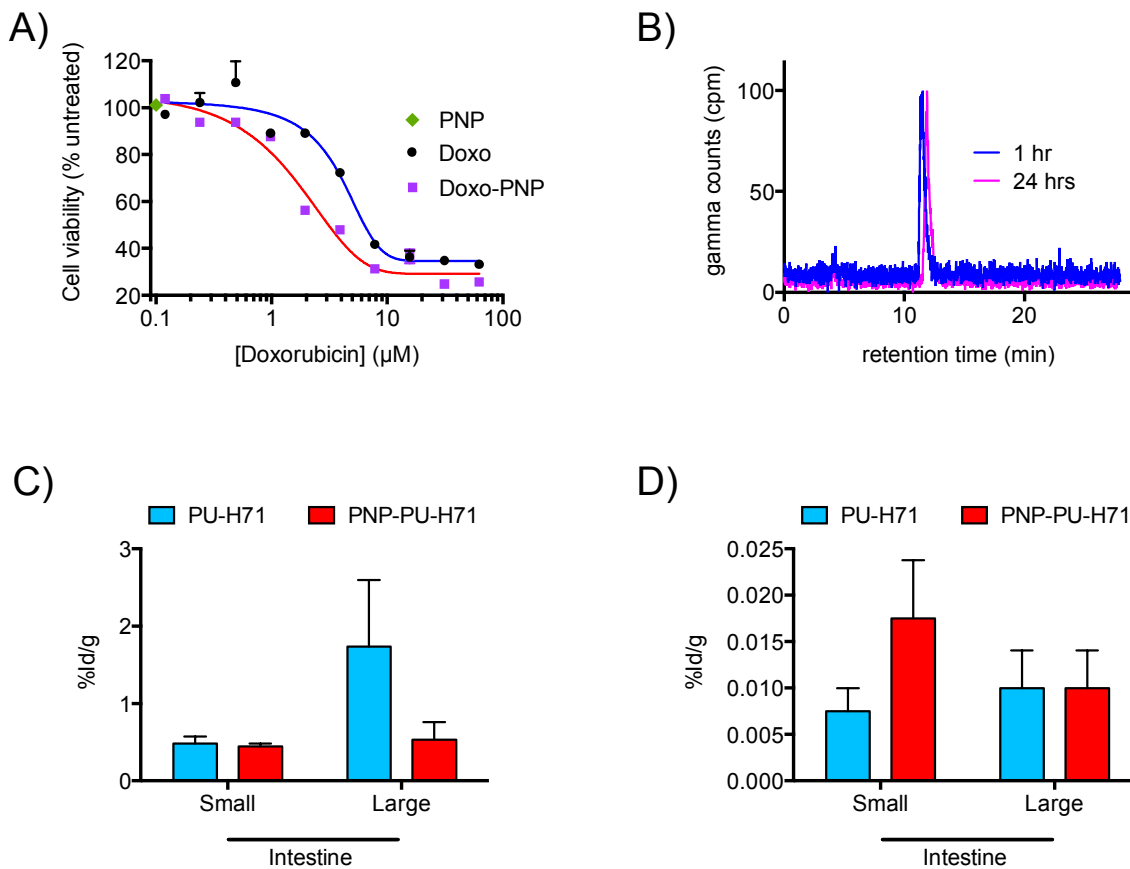
**Figure S1.** Retention of gadolinium (Gd) by carbohydrate PNPs. **A)** The PNPs that chelated Gd at room temperature slowly released the tracer in serum, indicated by increases in the PNP solution's T1 signal. **B)** When the Gd chelation at room temperature, the PNPs unloaded gadolinium once the pH reached 6.0. **C)** PNP of high-molecular-weight dextran chelated Gd more efficiently, **D)** whereas Gd chelation did not affect the nanoparticles size and distribution. Means  $\pm$  SEM.



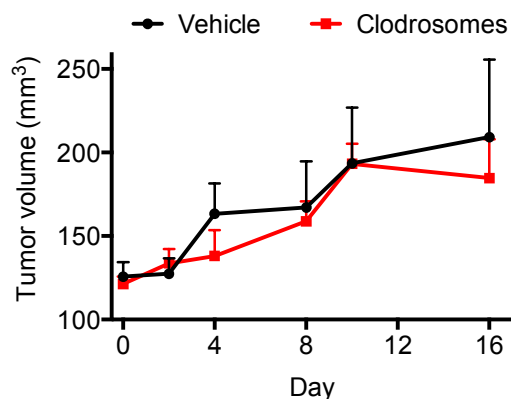
**Figure S2.** Chelation and imaging with <sup>89</sup>Zr-PNPs. **A)** The PNPs effectively chelated <sup>89</sup>Zr after 30-min heating at 70 °C, as shown by the radio-ITLC chromatograms. **B)** After chelation at 70 °C, the stability of the PNP – <sup>89</sup>Zr association was evaluated, which showed that the PNPs retained the radiotracer under EDTA challenge, and in fetal bovine and mouse sera. Means  $\pm$  SEM. **C)** CT/PET (left) and PET only (right) of draining lymph nodes (white arrows) after peritumoral administration of <sup>89</sup>Zr-PNPs, revealing proximal and distal drainage through the afferent lymphatics.



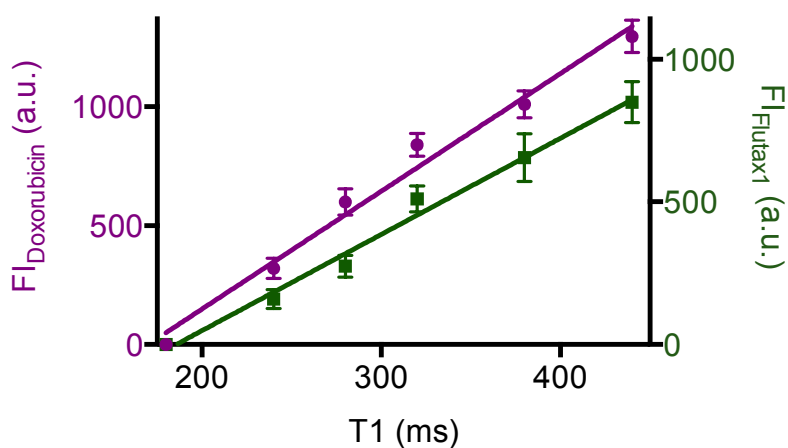
**Figure S3.** Atomic force microscopy performed at liquid state showed the structural stability of the PNPs after cargo loading and release. **A)** Distribution of the unloaded PNPs. Doxorubicin-loaded PNPs after incubation at **B)** pH 7.4 and **C)** pH 6.8. (**insets** show higher dilutions of the two samples, with the PNP being monodispersed).



**Figure S4. A)** Doxorubicin-loaded dextran nanophores cause more cell death than free doxorubicin in the androgen-receptor-negative human prostate cancer cell line PC3. (PNP: unloaded nanoparticles; Doxo: Doxorubicin; Doxo-PNP: doxorubicin-loaded PNPs) The unloaded PNP are not toxic to the cells. **B)** Serum stability of <sup>131</sup>I-PU-H71 determined through HPLC, after 1 and 24-hour incubation in mouse serum. Intestinal excretion of free and PNP-loaded <sup>131</sup>I-PU-H71 **C)** 4 h and **D)** 24 h post i.v. administration (%ID/g: % injected dose/tissue mass). Means ± SEM.



**Figure S5.** Tumor growth response after clodrosome-based therapy of athymic, male nude mice with LNCaP xenografts on their flanks ( $n_{\text{vehicle}}=n_{\text{Clodrosomes}}=5$ ). Means  $\pm$  SEM.



**Figure S6.** The loading of molecular cargos into PNP affects the nanoparticles' magnetic resonance signal. Gd-retaining PNP were co-loaded with Doxorubicin ( $\lambda_{\text{ex}} = 485 \text{ nm}$ ,  $\lambda_{\text{em}} = 590 \text{ nm}$ ) and Flutax1 ( $\lambda_{\text{ex}} = 495 \text{ nm}$ ,  $\lambda_{\text{em}} = 520 \text{ nm}$ ), which caused increased in the T1 relaxation times. Means  $\pm$  SEM.