Supporting Information

Magneto-Optical Nanoparticles for Cyclic Magnetomotive Photoacoustic Imaging

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Figure S1. Photoacoustic signals generated by MNP-PPy (red) and MNP (blue) nanoparticles of the same mass concentration ($60 \mu\text{g ml}^{-1}$) with tunable narrow-band laser excitation from 676 nm to 804 nm. The signal strength is proportional to the absorption profiles of the two nanoparticles.

Figure S2. TEM images of MNP-PPy nanoparticles prepared at different pH values: (a-d) pH 1, 4, 6, and 7, respectively. (a) Significant MNP size reduction because of etching by the strong acidic condition. (b-d) Insufficient PPy encapsulation due to slow polymerization rate at these pH values.

Figure S3. Photoacoustic signal produced by MNP-PPy core-shell nanoparticles and AuNRs of the same mass concentration ($30 \mu\text{g ml}^{-1}$) in polyvinyl alcohol phantoms under the same experimental conditions.

Figure S4. Photostability measured by photoacoustic imaging. Photoacoustic signals from MNP-PPy and AuNR (both at $30 \mu\text{g ml}^{-1}$) are irradiated with a pulsed laser (laser wavelength 800 nm, fluence 5 mJ/cm^2 , pulse repetition rate 20 Hz). Both samples are stable during this 5-min exposure at modest intensity.

Figure S5. Ultraviolet–visible absorption spectra of MNP-PPy hybrid nanoparticles before (black solid line) and after (red dash line) 10 min oscillating magnetic field (50 Hz, 0.7 T) treatment.

Photoacoustic spectral measurement method

Photoacoustic (PA) spectrometry was used to verify photoacoustic amplitudes of materials at a specific wavelength. The photoacoustic signal p , generated by thermal expansion due to laser-induced temperature increase, is laser wavelength dependent and can be expressed as Equation S1:

$$p(\lambda) = \Gamma \mu_a(\lambda) \Phi(\lambda) \quad (\text{S1})$$

where λ is the laser wavelength, Γ is Gruneisen parameter, μ_a is the optical absorption coefficient, and Φ is the laser fluence. Therefore, by measuring photoacoustic signal amplitude and the fluence at different wavelengths, the absorption coefficient can be obtained as a function of wavelength. That is (Equation S2),

$$\mu_a(\lambda) = \frac{p(\lambda)}{\Gamma \Phi(\lambda)} \propto \frac{p(\lambda)}{\Phi(\lambda)} \quad (\text{S2})$$

As a result, photoacoustic (PA) spectrometry also verifies the absorbability of particles suspended in solution, which is hard to directly measure through UV-Vis absorption spectrometer due to scattering effects.

Photoacoustic spectrometry setup: A 532-nm pumped wavelength tunable OPO laser (Surelite OPO plus, Continuum, Santa Clara, CA, USA) delivered laser pulses sweeping from 670 nm to 800 nm on a 5-ml sample inside a transparent transfer pipette immersed in a water tank. A home-made wideband PVDF transducer (50 kHz- 30 MHz bandwidth by 1/e level, 6 mm aperture and 7 mm focus depth) was used to detect photoacoustic signals. Gold nanorods were first tested to validate the system, which produced an absorption profile well-matched with the UV-vis spectrum. MNPs and MNP-PPy nanoparticles at the same mass ($60 \mu\text{g ml}^{-1}$) concentration were then measured using the same photoacoustic spectrometry procedure, with the results shown in **Figure S1**. At 800 nm, MNP-PPy nanoparticles efficiently generated photoacoustic signals following NIR pulsed laser irradiation, which is 10 times higher than for pure MNPs. The increase in photoacoustic amplitudes for all irradiating wavelengths demonstrates the PPy (NIR absorber) shell encapsulation, thus enhancing the photoacoustic response of hybrid nanoparticles.