Supplementary Materials

Dual-frequency metastructured-hydrogel film for high-contrast ultrasound imaging

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Supplementary Table1. Parameters of the equipment used in this work.

Center Frequency	Band-width	Elements	Voltage	Pulse repetition frequency (PRF)	Pulse cycle	Sampling rate	Gain
2 MHz/6 MHz	20 %	128	70V	4000	1-6 (tunable)	4×10^8	1

Here, “Bandwidth” refers to the -6 dB bandwidth.

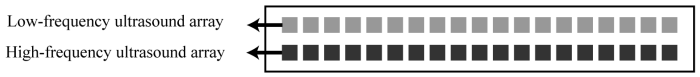
Supplementary Table 2. Comparison of existing metagel contrast agent and dual-frequency-specific metagel proposed in this work.

	Resolution	**SNR**	**Contrast (to Zinc tablet)**
Existing metagel contrast agent (Ref.) (Non-frequency-specific)	0.15 mm	20	1
This work (Dual-frequency-specific)	0.15 mm	50	4

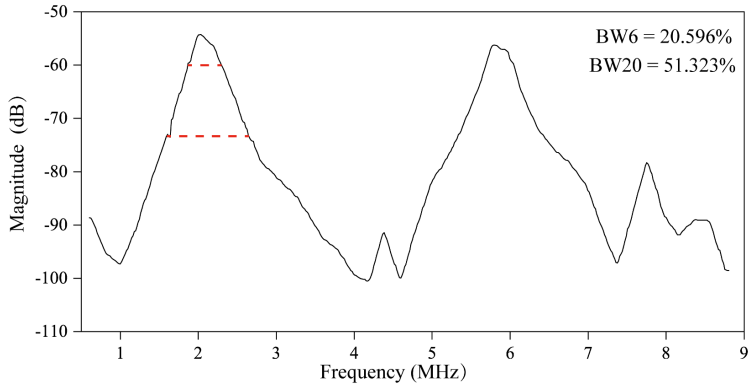
In particular, the resolution of ultrasound line-scan imaging depends on the spacing between the ultrasonic transducer elements.

Supplementary Table 3. Performance comparison of metagel film with various ultrasound contrast agents.

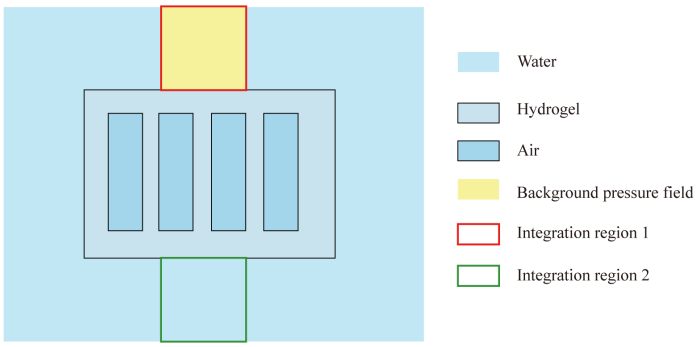
Type	**Material Components**	**Potential harm**	**Target tissue imaging**	**Signal-to-noise ratio (dB)**	**Example**
Microbubble contrast agent	Inert gases (e.g., sulfur hexafluoride, perfluoropropane) + polymers (e.g., phospholipids, albumin, etc.)	Toxicity	No	<25	Ref. 9
Non-microbubble contrast agent	Liquid fluorocarbon-based contrast agent	Toxicity	No	<25	Ref. 10
Solid-state contrast agent	Hydrogel + High-reflectivity material (Zinc metal)	Mechanical damage	Yes	~45	Ref. 11
Metagel film contrast agent	Hydrogel + Air metastructure	Biocompatible	Yes	~50	This work



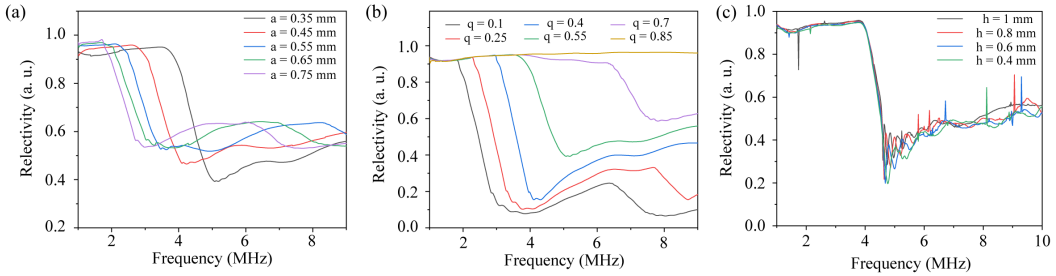
Supplementary Figure 1. Schematic of the dual-frequency ultrasound probe.



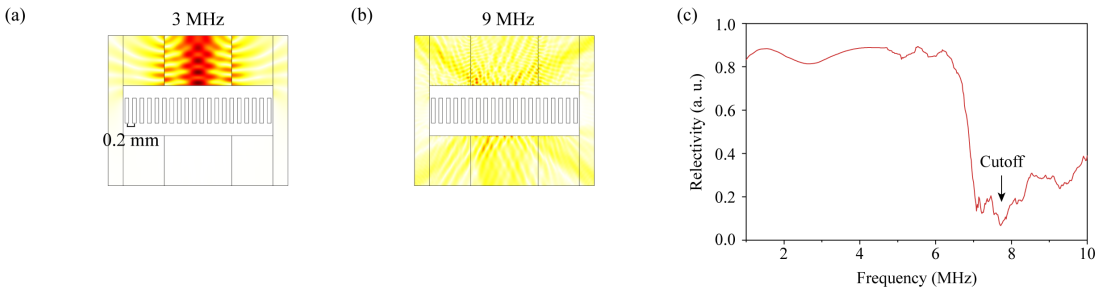
Supplementary Figure 2. Spectra of the dual-frequency ultrasound probe. The two red dashed lines represent bandwidth ranges that are 6 dB and 20 dB below the center frequency, denoted as BW6 and BW20, respectively.



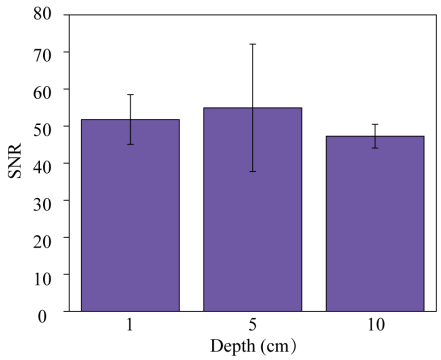
Supplementary Figure 3. Simulation setting for the ultrasonic reflection of the metagel.



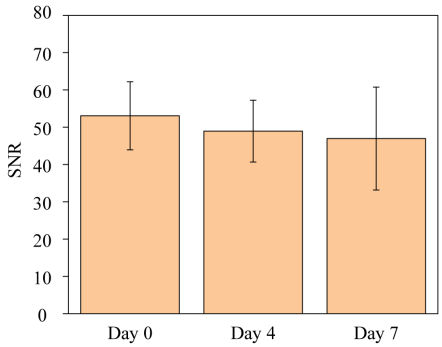
Supplementary Figure 4. Parameter implication on the ultrasonic reflection of the metagel. (a) *a*, (b) *q*, (c) *h*.



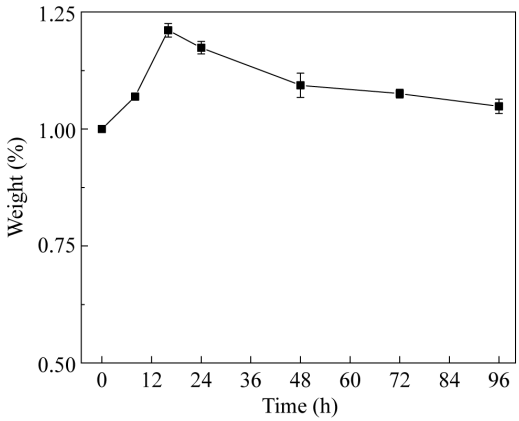
Supplementary Figure 5. Reflection condition of the metagel with a = 0.2 mm at (a) 3MHz, (b) 9MHz and (c) the spectrum.



Supplementary Figure 6. Metagel reflections at different depths. Error bars represent standard deviations (SD) based on three independent measurements (n = 3).



Supplementary Figure 7. Metagel reflections during the 7-days period. Error bars represent standard deviations (SD) based on three independent measurements (n = 3).



Supplementary Figure 8. Swelling ratio of the metagel. Error bars represent standard deviations (SD) based on three independent measurements (n = 3).