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Supplementary Materials for

Microcellular sensing media with ternary transparency states for fast and intuitive identification of unknown liquids

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Supplementary Figures

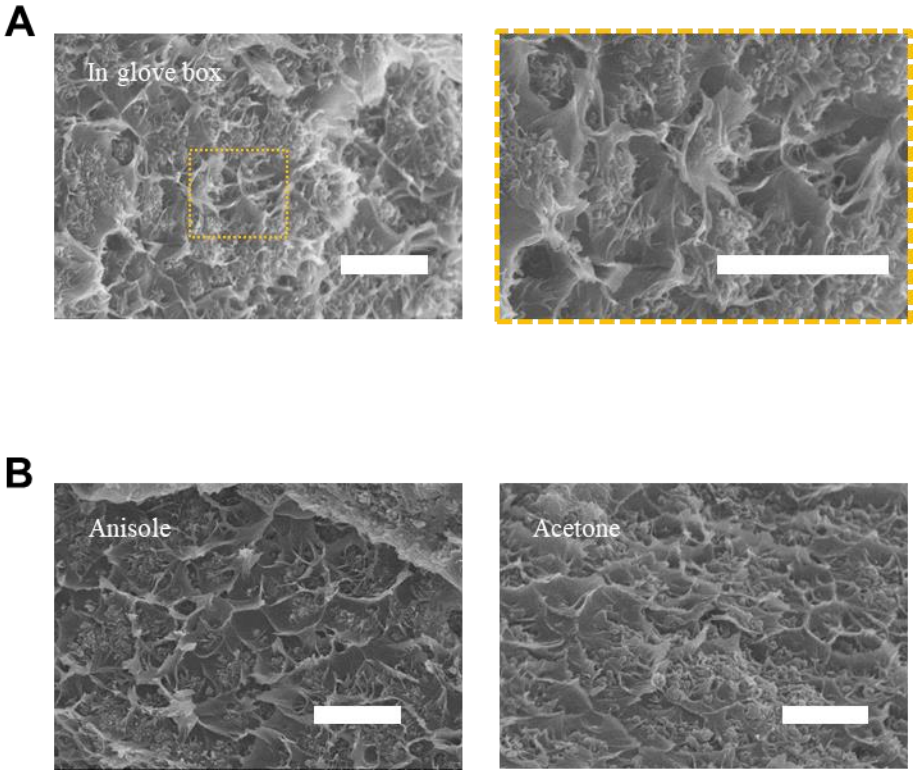

Fig. S1. Humidity and solvent-independent interphase void formation. Cross-section SEM images of the scattering media prepared (A) in glove box and (B) from different solvent. Scale bars denote 10um. The polymer that is used is PMMA.

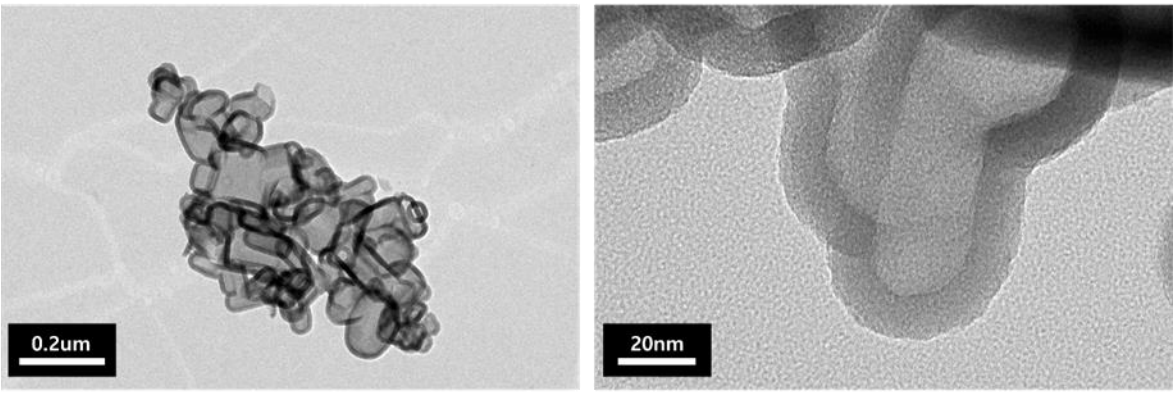

Fig. S2. TEM images of the hollow particles. The shell thickness is around 20 nm with a small variation (<20%). Their average diameter is 300 nm.

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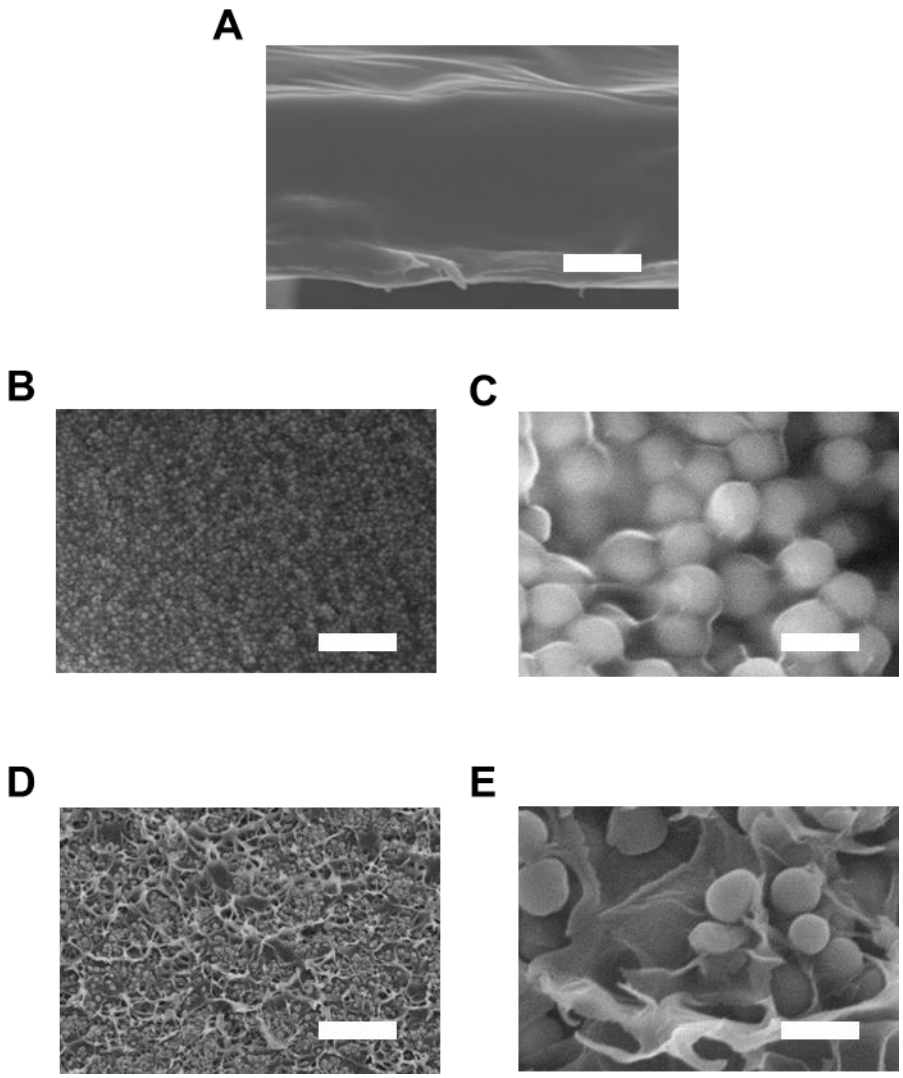


Fig. S3. Internal structure of various light-scattering media. Cross-section SEM images of the scattering media composed of (A) only polymer matrix, (B, C) only solid silica particles, and (D, E) solid silica particles with inter-phase voids. PMMA is used for the polymer matrix. Scale bar of (A) denotes 20 um. Scale bars of (B, D) denote 10um. Scale bars of (C, E) denote 1um.

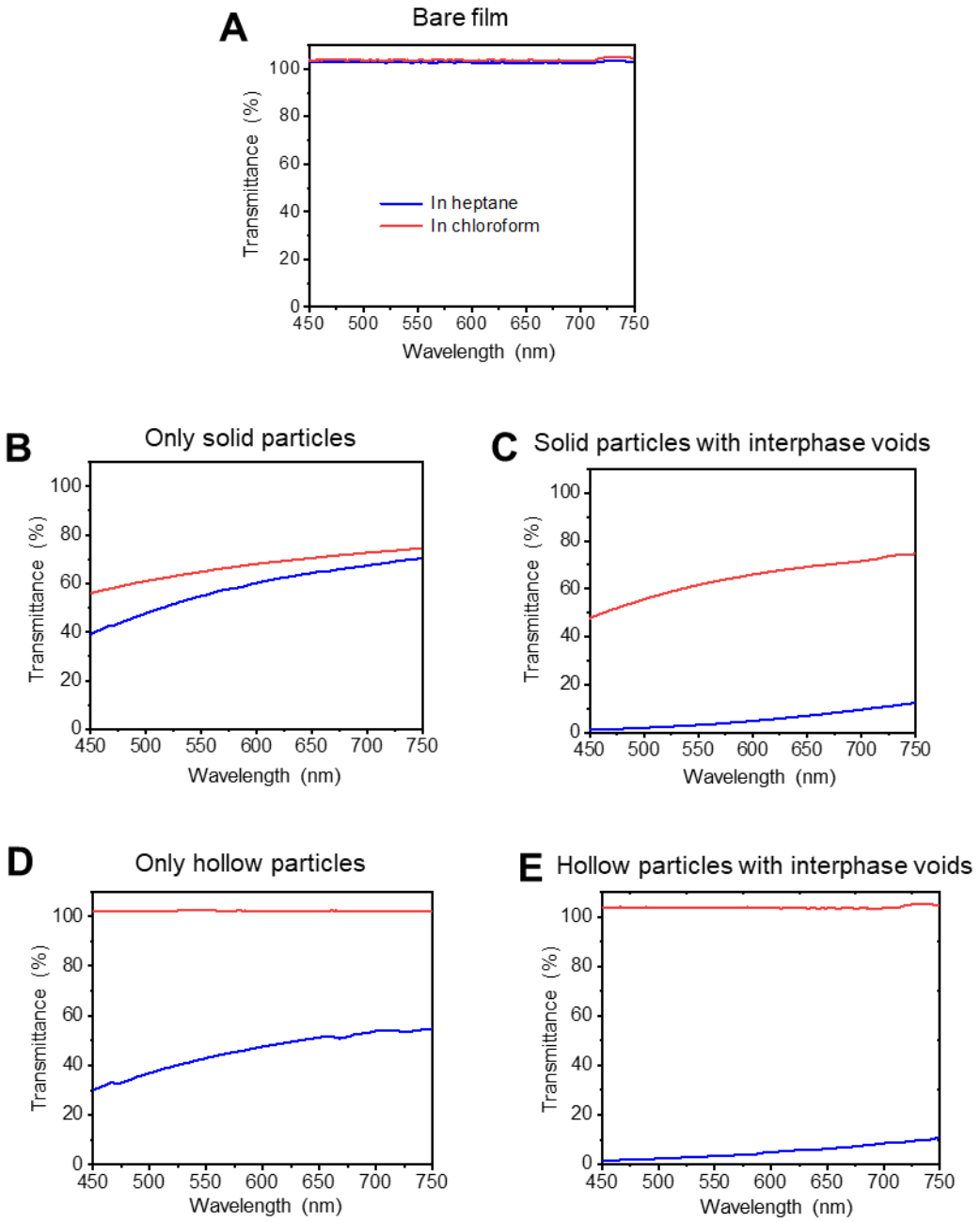


Fig. S4. Transmittance window of various light scattering mediums. Transmittance graph of various types of the light scattering media after immersing in heptane and chloroform. PMMA is used for the polymer matrix.

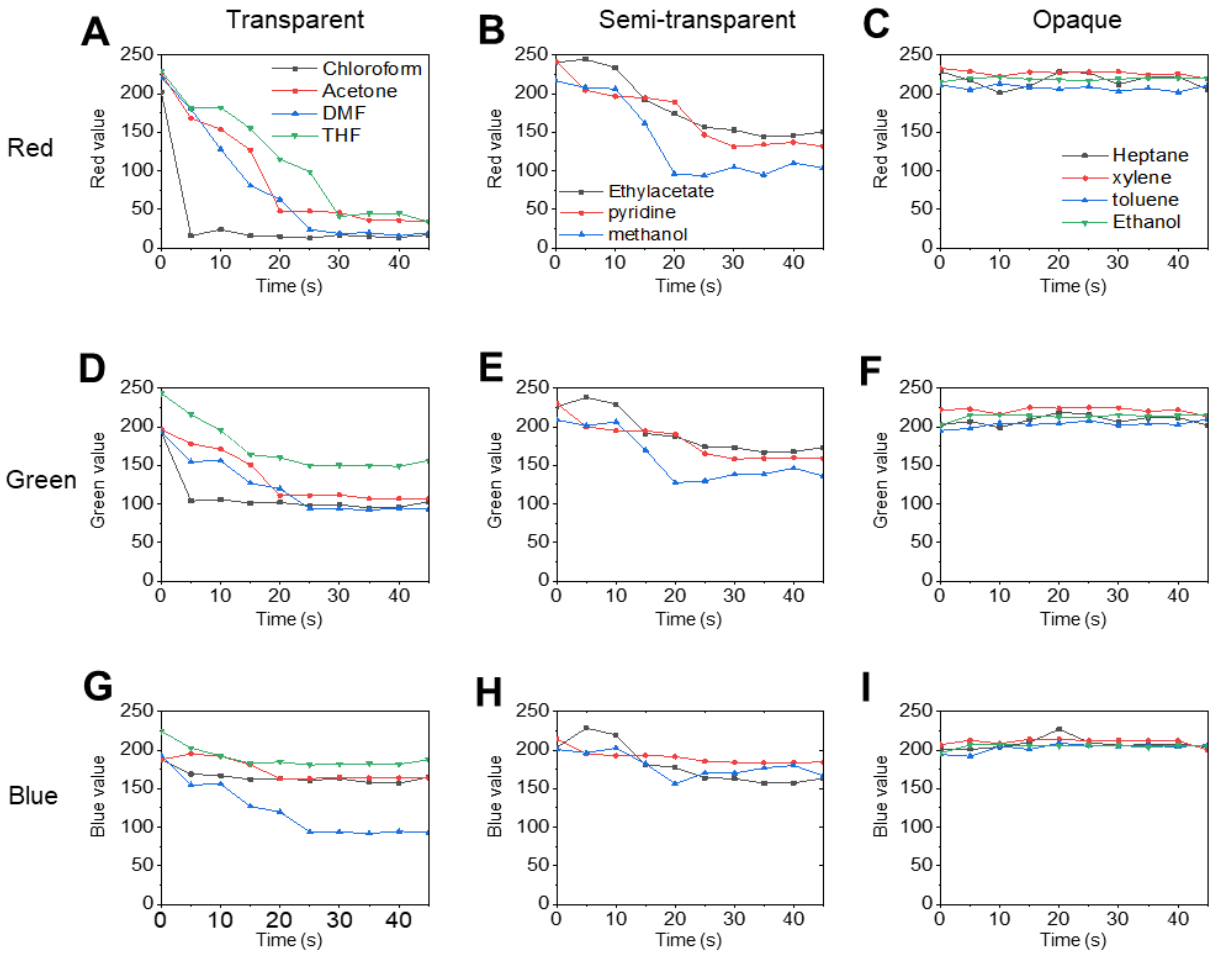


Fig. S5. RGB value changes of the scattering medium films after immersing in different organic liquid. Red (A to C), green (D to E), and blue (G to I) value changes of the scattering media as a function of exposure time. All RGB values are extracted from the optical images of the scattering media captured by a smartphone with blue background.

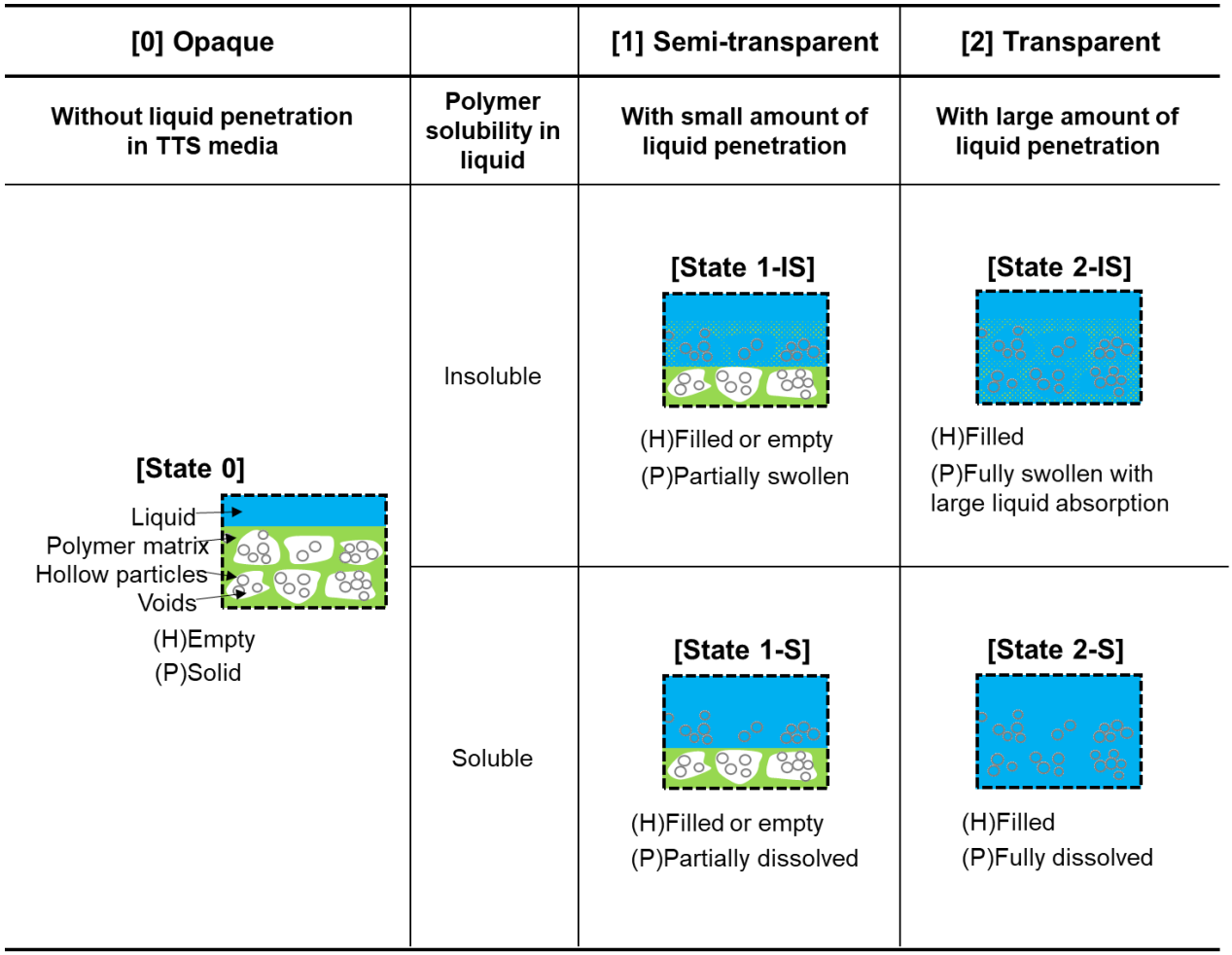


Fig. S6. The proposed mechanism of three different transparency states (opaque, semi-transparent, and transparent) of the scattering medium after being immersed in organic liquid. The transparency states are dependent on whether the hollow particles and porous polymer structure are empty or filled with a liquid, and whether the polymer matrix is swollen or dissolved.

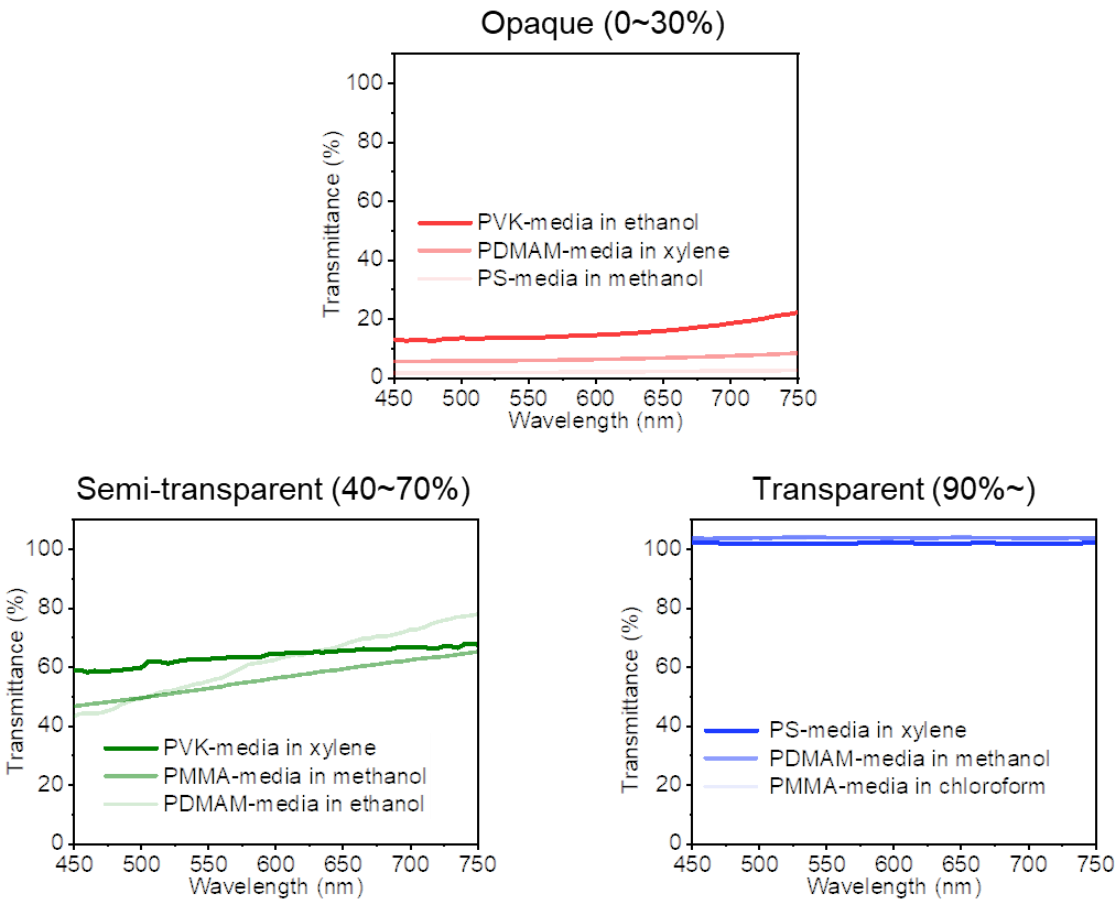
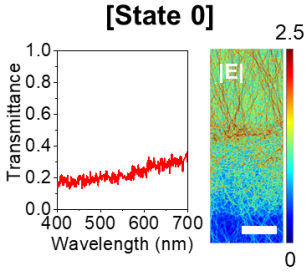
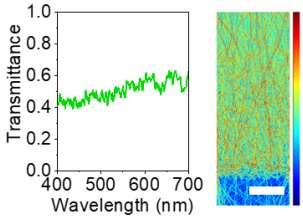
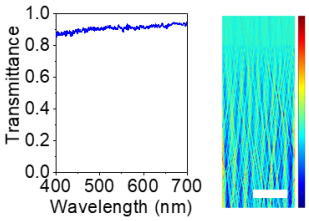
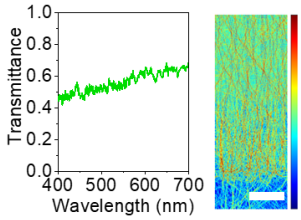
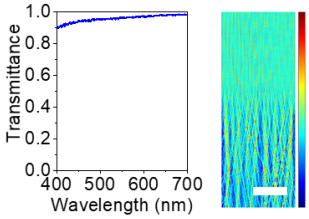


Fig. S7. Transmittance of TTS media of each transparency state. The transmittance was measured after immersing the media in the liquid for 30s.

[0] Opaque		[1] Semi-transparent	[2] Transparent
Without liquid penetration in TTS media	**Polymer solubility in liquid**	**With small amount of liquids penetration**	**With large amount of liquid penetration**
[State 0]  (H)Empty (P)Solid	Insoluble	**[State 1-IS]**  (H)Filled or empty (P)Partially swollen	**[State 2-IS]**  (H)Filled (P)Fully swollen with large liquid absorption
	Soluble	**[State 1-S]**  (H)Filled or empty (P)Partially dissolved	**[State 2-S]**  (H)Filled (P)Fully dissolved

(H) State of the hollow particles and pores
(P) State of the polymer matrix

Swollen film ratio: 75% (1-s1)
Dissolved film ratio: 75% (1-d)
Liquid content in the swollen polymer matrix: 67% (1-s1), 4.7% (1-s2), 67% (2-s)

Fig. S8. Calculated transmittance (left) and electric field distribution (right) of the proposed physical model for each transparency state. The scale bar denotes 15 μm.

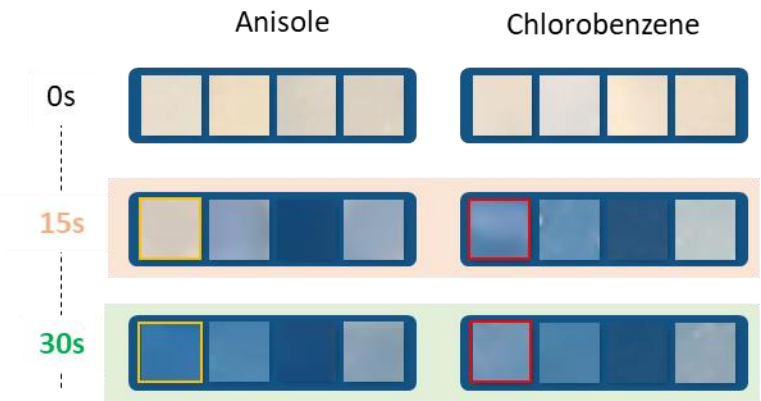


Fig. S9. Transparency patterns of an array in the dried state (0s) and after being immersed in the liquids for 15 and 30 seconds. PMMA, PVK, PS, and PDMAM are used as polymer matrices (from left to right). Photo credit: K. M. Song (KAIST).

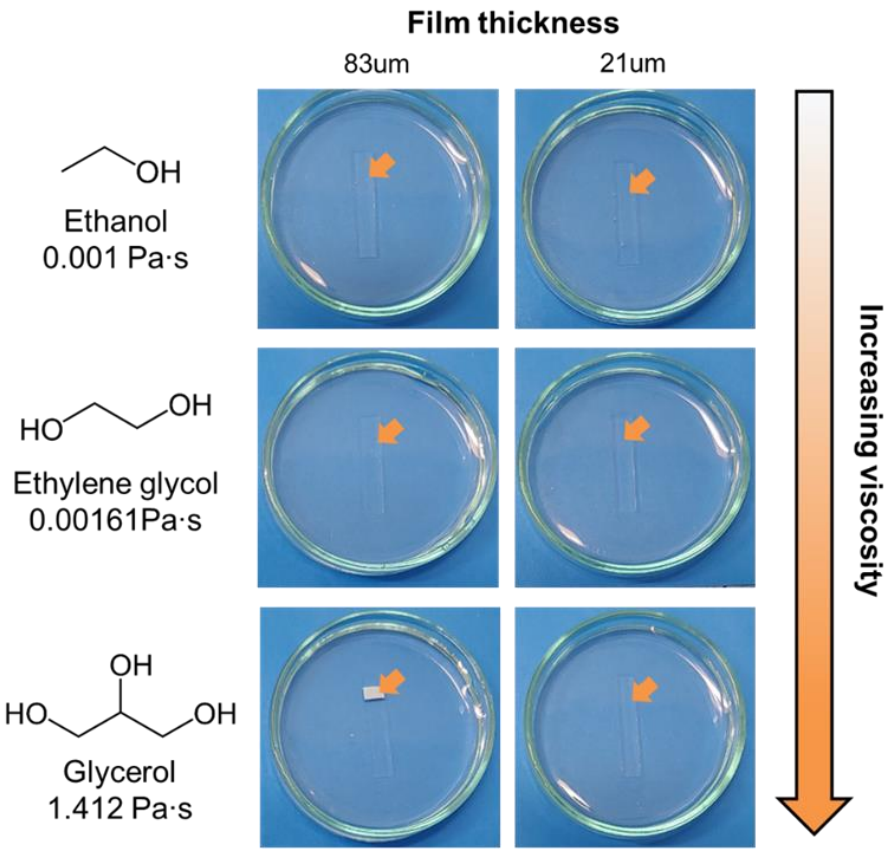


Fig. S10. Photographs of the sensing media immersed in liquid for 30s. The polymer is based on PMMA. Photo credit: K. M. Song (KAIST).

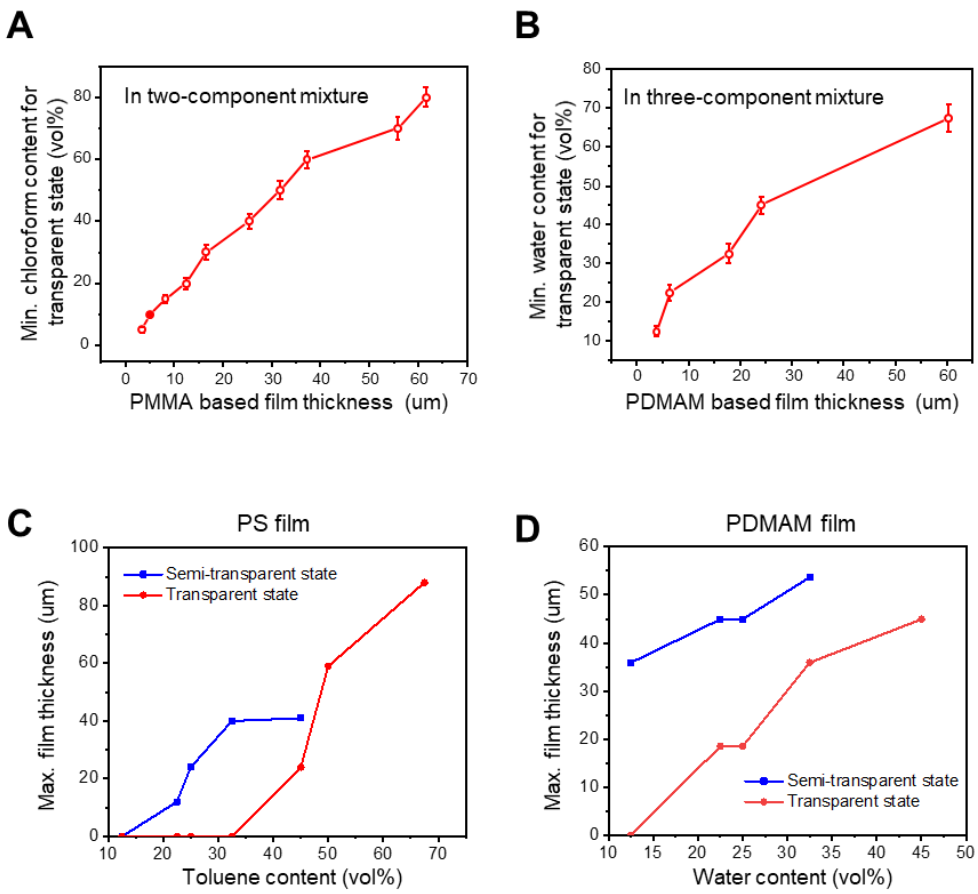


Fig. S11. Identification of liquid mixture composition via film thickness control. Minimum target liquid concentration for the transparent state in (A) binary and (B) ternary liquid mixture as a function of the film thickness. Error bars indicate the standard deviation of at least three measurements with <20% variation. (C-D) Maximum thickness of (C) PS and (D) PDMAM based TTS media as a function of the target liquid concentration.

Supplementary Tables

Table. S1 Material lists of the polymer solution for Fig. 3 (A), Fig. S3, and Fig. S4.

Volumetric ratio of polymer is identical as 10%. The percentage of particles in samples (2) and (4) is the maximum amount just before aggregating.

	Sample	Types of particle	Volumetric percentage of particles (%)	PMMA MW (kg/mole)	Solvent
(1)	Bare film	-	-	15	Chloroform
(2)	Only solid particle	SiO_2 solid particles	5	15	Chloroform
(3)	Solid particle with interphase voids	SiO_2 solid particles	2	1000	Chloroform
(4)	Only hollow particles	SiO_2 hollow particles	5	15	Chloroform
(5)	Hollow particle with interphase void	SiO_2 hollow particles	2	1000	Chloroform

Table. S2 Material lists of the polymer solution for Fig. 2 (C). Volumetric ratio of each component is identical.

Polymer	Molecular weight (kg/mol)	Solvent	Particle
Poly(methylmetacrylate)	1000	chloroform	Hollow silica particle
Poly(N-vinylcarbazole)	1100	Toluene	Hollow silica particle
Poly(N,N-dimethyl acrylamide)	900	Methanol	Hollow silica particle
Poly(styrene)	1200	Toluene	Hollow silica particle