## **Supporting Information**

## Enzyme and Relative Humidity Responsive Antimicrobial Fibers for Active Food

## Packaging

Zeynep Aytac<sup>a#</sup>, Jie Xu<sup>a#</sup>, Suresh Kumar Raman Pillai<sup>b</sup>, Brian D. Eitzer<sup>c</sup>, Tao Xu<sup>a</sup>, Nachiket

Vaze<sup>a</sup>, Kee Woei Ng<sup>a, d, e</sup>, Jason C. White<sup>c</sup>, Mary B. Chan-Park<sup>b</sup>, Yaguang Luo<sup>f</sup>, Philip

Demokritou<sup>a,d\*</sup>

<sup>a</sup> Center for Nanotechnology and Nanotoxicology, Department of Environmental Health, Harvard T. H. Chan School of Public Health, Harvard University, Boston, MA,

02115, USA

 <sup>b</sup> School of Chemical and Biomedical Engineering, Nanyang Technological University, 637457, Singapore
 <sup>c</sup> Department of Analytical Chemistry, The Connecticut Agricultural Experiment Station, New Haven, CT, 06504, USA
 <sup>d</sup> School of Materials Science and Engineering, Nanyang Technological University, 639798, Singapore

<sup>e</sup> Environmental Chemistry and Materials Centre, Nanyang Environment and Water Research Institute, 637141, Singapore

<sup>f</sup>Environmental Microbiology and Food Safety Laboratory, U.S. Department of Agriculture, Agricultural Research Service, Beltsville Agricultural Research Center, 10300 Baltimore Avenue, Beltsville, MD 20705

\* Corresponding author: Philip Demokritou, E-mail: pdemokri@hsph.harvard.edu

<sup>#</sup> These authors contributed equally to this work.

**Table S1.** AI concentrations (%), polymer composition (%), and other operational parameters used for the synthesis of pristine fibers (no AIs), enzyme responsive fibers (no CD-ICs), RH responsive fibers (no free AIs), and multi stimuli responsive fibers.

	Free Als	CD-ICs of Als	Polymer composition (%)	Operational parameters
Pristine fibers	-	-	<ul> <li>41% (w/v) in total,</li> <li>85:15 zein:starch</li> </ul>	<ul><li>Flow rate: 1 mL/h</li><li>Voltage: 35 kV</li><li>Distance: 10 cm</li></ul>
Enzyme responsive fibers	<ul> <li>thyme oil (1%, w/v)</li> <li>citric acid (5%, w/v)</li> <li>nisin (0.2%, w/v)</li> </ul>	-	<ul> <li>47% (w/v) in total,</li> <li>85:15 zein:starch</li> </ul>	<ul> <li>Flow rate: 0.5 mL/h</li> <li>Voltage: 27.5 kV</li> <li>Distance: 10 cm</li> </ul>
RH responsive fibers	-	<ul> <li>thyme oil (1%, w/v)</li> <li>sorbic acid (0.5%, w/v)</li> <li>nisin (0.2%, w/v)</li> </ul>	<ul><li>47% (w/v) in total,</li><li>85:15 zein:starch</li></ul>	<ul><li>Flow rate: 0.5 mL/h</li><li>Voltage: 27.5 kV</li><li>Distance: 10 cm</li></ul>
Multi stimuli responsive fibers	<ul> <li>thyme oil (1%, w/v)</li> <li>citric acid (5%, w/v)</li> <li>nisin (0.2%, w/v)</li> </ul>	<ul> <li>thyme oil (1%, w/v)</li> <li>sorbic acid (0.5%, w/v)</li> <li>nisin (0.2%, w/v)</li> </ul>	<ul><li>41% (w/v) in total,</li><li>85:15 zein:starch</li></ul>	<ul> <li>Flow rate: 0.5 mL/h</li> <li>Voltage: 25 kV</li> <li>Distance: 10 cm</li> </ul>

**Table S2.** Specific surface area (SSA), average pore radius, and total pore volume of pristine fibers, enzyme responsive fibers with only free active ingredients (AIs), relative humidity (RH) responsive fibers with only cyclodextrin-inclusion complexes (CD-ICs) of AIs, and multi stimuli responsive fibers.

	Multipoint BET surface area (m²/g)	Average pore radius (nm)	Total pore volume (cc/g)
Pristine fibers	12.1	2.18	1.32x10 <sup>-2</sup>
Enzyme responsive fibers	19.3	1.64	1.58x10 <sup>-2</sup>
RH responsive fibers	10.4	1.56	0.81x10 <sup>-2</sup>
Multi stimuli responsive fibers	10.9	2.16	1.18x10 <sup>-2</sup>



**Figure S1.** Schematic representation of experiment design for **a.** direct contact assay and **b.** relative humidity (RH) triggered antimicrobial activity test of relative humidity (RH) responsive fibers.



**Figure S2.** Scanning electron microscopy (SEM) images of the degradation of enzyme responsive fibers with only free active ingredients (AIs) in **a.** 0.1 U/mL, **b.** 1 U/mL, and c. 3 U/mL enzyme concentrations at the end of 12 hours.



**Figure S3.** Scanning electron microcopy (SEM) images of relative humidity (RH) responsive fibers with only cyclodextrin-inclusion complexes (CD-ICs) of AIs at **a.** 50% RH and **b.** 95% RH at the end of 4 hours.



Figure S4. Cumulative release (%) of thymol from multi stimuli responsive fibers into food simulant (water) as a function of time.