SUPPORTING INFORMATION

Novel LDPE/Halloysite Nanotubes Composites with Sustained Carvacrol Release for Broad-Spectrum Antimicrobial Activity

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Figure S1 depicts representative Fourier-transform infrared spectroscopy (FTIR) spectra of pure carvacrol, neat LDPE and LDPE/(HNT/carvacrol hybrid) films. It is clearly seen that the high-temperature melt compounding process does not affect the chemical structure of carvacrol, as the FTIR spectrum of the LDPE/(HNT/carvacrol hybrid) film combines all distinctive spectral elements from both LDPE and pure carvacrol.

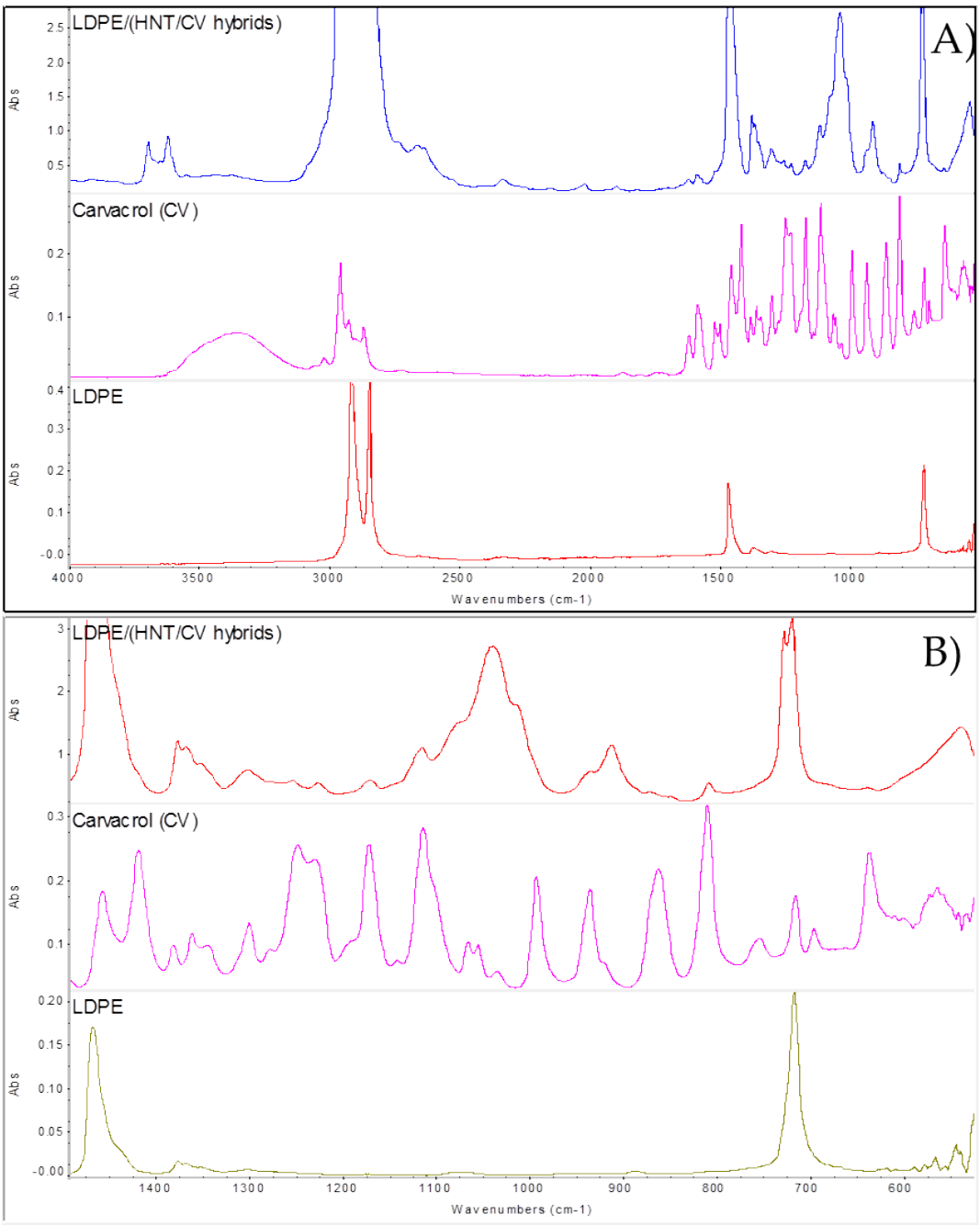

Figure S1: (A) FTIR spectra of pure carvacrol, neat LDPE and LDPE/(HNT/carvacrol hybrid) films. (B) View of the same spectra, zoomed at the wavelength range of 1500-500 cm^{-1}.

Figure S2 presents the results of the different LDPE-based films on *L. innocua*-inoculated white soft cheese. We adapted the FDA Bacteriological Analytical Manual (BAM) for Detection and Enumeration of *L. monocytogenes* [52] and accordingly inoculated the cheese with *L. innocua* to contain 4 log CFU/mL bacteria. The cheese is then incubated (at 26°C for 22 h) with the different films (in direct contact and indirect-headspace contact), followed by Listerial count. Figure S2 shows that the LDPE/(HNTs/carvacrol hybrid) films retard the growth of *L. innocua*; generating a profound and reproducible decrease of 1.5 log CFU/mL (Fig. S2b) and 1 log CFU/mL (Fig. S2c) in *L. innocua* population for indirect and direct contact assays, respectively.

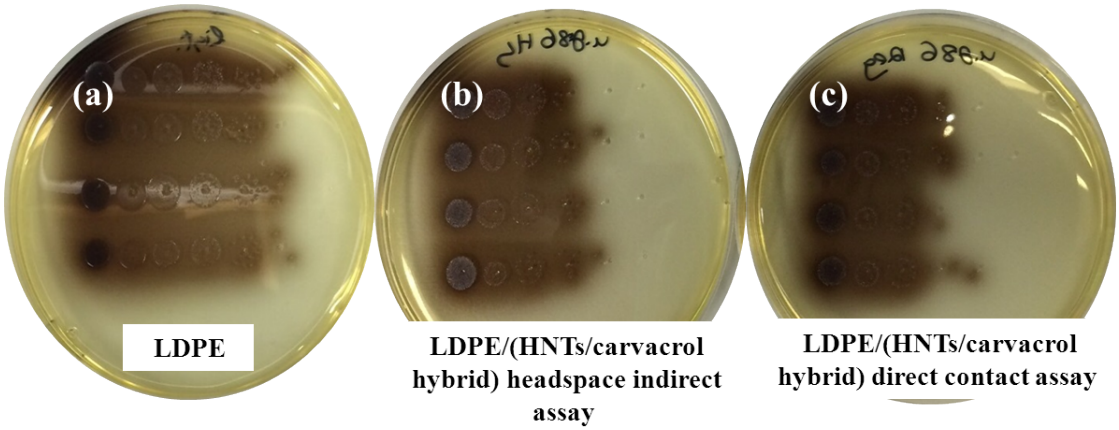

Figure S2: Inoculated soft cheese experiment, assessing the effect of LDPE-based films on *L. innocua* growth. Images present the appearance of *L. innocua* colonies on a modified oxford plate after incubation at 30°C for 48 h. (a) neat LDPE film; (b) LDPE/(HNTs/carvacrol hybrid) film, measured by an indirect-headspace assay; (c) LDPE/(HNTs/carvacrol hybrid) film, measured by a direct contact assay. *L. innocua* appears as black colonies with a black halo.

Figure S3 displays images of sliced wheat bread (preservative-free) following storage for 11 days at 25°C with the different LDPE-based films. Note that the experiments is carried out without inoculation, by aseptically placing a slice of bread in a sterile Petri dish and films with an area of ~25 cm^2 are attached to the center of the dish lid, assuring no contact between the studied film and the bread. The plates are tightly sealed and incubated for 11 days at 25°C. The test is carried out in triplicates and the fungal growth is monitored and recorded. Figure S3a-c shows that different fungi populations develop onto the bread slices during storage. However, no fungus is detected when the bread is stored with the LDPE/(HNTs/carvacrol hybrid) film.

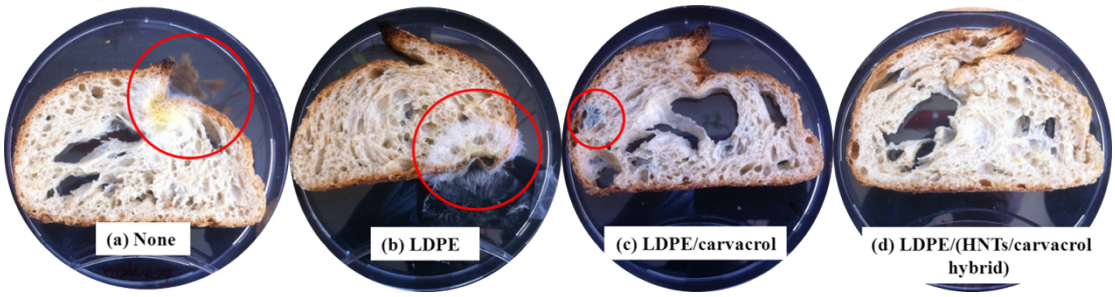

Figure S3: Storage experiment of natural sliced bread (preservative-free). Indirect headspace assay examining the effect of different films on the development of fungi following 11 days of incubation at 25°C in the dark: (a) none (b) neat LDPE film (c) LDPE/carvacrol film (d) LDPE/(HNTs/carvacrol hybrid) film. Fungal colonies are circled in red for clarity.

Figure S4 displays images of the fungi-inoculated bread after storage for 11 days at 25ºC. For bread stored with no film (Fig. S4a) and with neat LDPE film (Fig. S4b), substantial and homogenous fungal growth is observed to occur on the slices. In contrast, incubation of the inoculated-bread with LDPE/(HNTs/carvacrol hybrid) films resulted in total eradication of the fungus (Fig. S4c).

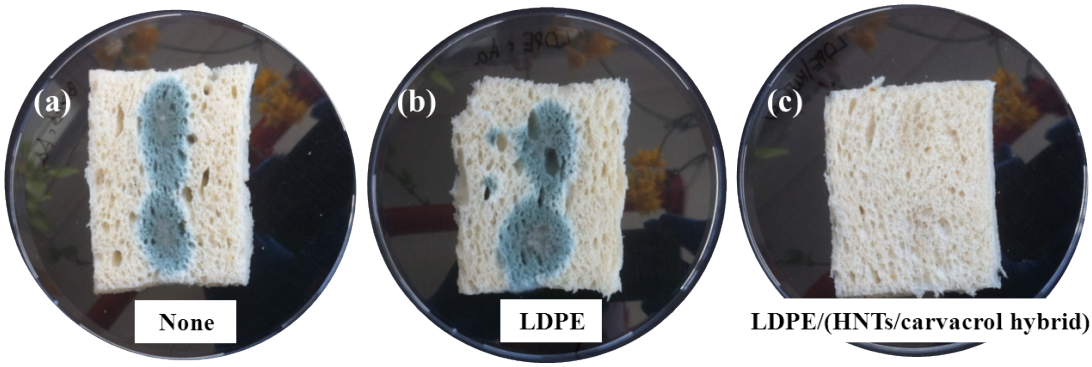

Figure S4: Fungi-inoculated (in two points) sliced bread storage experiment. Indirect headspace assay examining the effect of different films on the development of fungi following 11 days of incubation at 25°C in the dark: (a) none; (b) neat LDPE film; (c) LDPE/(HNTs/carvacrol hybrid) film.