**Electronic Supplementary Information**

# A Methodology to Produce Eco-Friendly Superhydrophobic Coatings Produced from All-Water-Processed Plant-Based Filler Materials

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# I. DPOD Phase Inversion

When initially testing the wettability of DPOD, it was found that the hydrophobicity of DPOD-only films on glass slides were dependent on both thermal curing time and temperature. It was found that droplets dispensed onto a DPOD film that were not sufficiently heat treated penetrated the polymer film (see ***Figure S1a***). Under appropriate heating conditions, droplets dispensed on a DPOD film showed no penetration into the surface (see ***Figure S1b***).


**Figure S1:** Behavior of a water droplet deposited onto a pure DPOD film for various levels of heat treatment for 5 minutes. **(a)** 100°C, and **(b)** 200°C.

In the as-received DPOD solution (***Figure S2a-i***), the hydrophilic Primacor™ (shown in orange) acts as dispersing agent for the more hydrophobic Affinity™ (in purple). After spraying the DPOD solution onto a glass slide and allowing the film to cure at room temperature, Primacor™-covered spherules of Affinity™ form on the surface (***Figure S2a-ii***). Mild heat treatment (30 minutes at 100°C) is sufficient to melt these spherules (***Figure S2a-iii***). Although the DPOD film is now flat, the surface is not homogeneous and is rich in the Primacor™ phase. Upon further heat treatment (***Figure S2a-iv***), the two phases separate until there is a homogeneous Affinity™-rich phase at the coating surface (***Figure S2a-v***). This completes the inversion process, and the hydrophilic component is now adjacent to the solid substrate, while the hydrophobic component is entirely at the surface. The phase separation process was confirmed through scanning electron microscopy (SEM) images taken at different stages of this process. In ***Figure S2b-i***, after curing a DPOD film at room temperature, spherules of Affinity™ covered in Primacor™ are seen on the sample surface. After heat-treating the DPOD film for 30 minutes at 100°C, the phase separation process begins and the resulting surface comprises both Primacor™ and Affinity™. This is first demonstrated in ***Figure S2b-ii***, where dark regions (a representative one is circled with a white dotted-line) correspond to Affinity™, and lighter regions correspond to Primacor™. Further heat treatment (5 minutes at 165°C; ***Figure S2b-iii***) brings the Affinity™ phase to the outer surface, until this surface is comprised only of Affinity™. The two phases in DPOD films treated for 5 minutes at 200°C were found to have completely separated, as the SEM image in ***Figure S2b-iv*** shows a homogeneous surface consisting of the hydrophobic component.

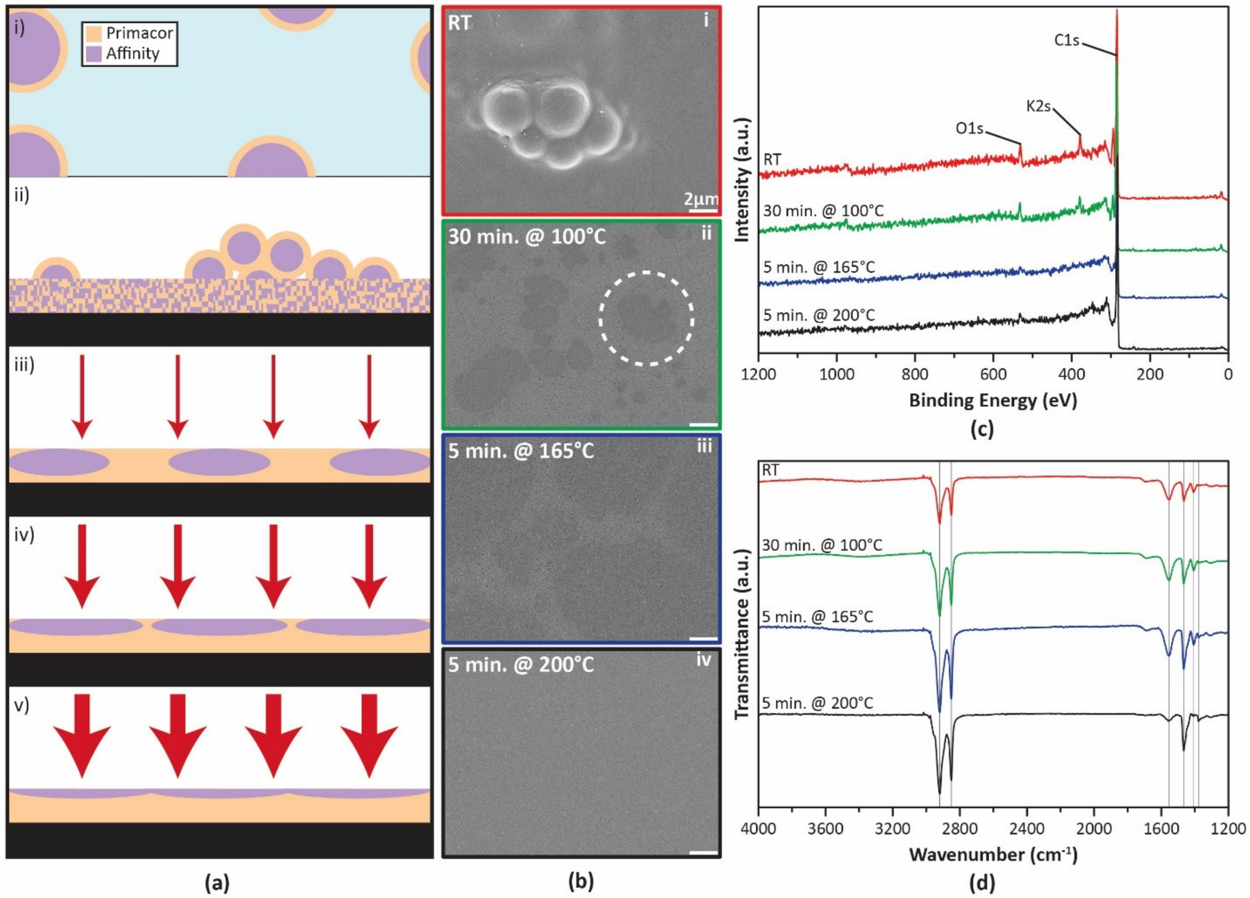


**Figure S2: (a)** Schematic demonstrating the phase separation process of DPOD: (i) DPOD in aqueous solution, (ii) DPOD film cured at room temperature, (iii) DPOD film after mild heat treatment (30 minutes at 100°C), (iv) DPOD film after intermediate heat treatment (5 minutes at 165°C), and (v) DPOD film after intense heat treatment (5 minutes at 200°C). **(b)** SEM images of the DPOD components phase-separation process, **(c)** XPS spectra, and **(d)** FTIR spectra of a DPOD film during the phase separation process in which the film was subjected to the same heat treatment as in (a-ii through a-v). The scale bar in each SEM image denotes 2μm.

X-ray photoelectron spectroscopy (XPS) and Fourier-Transform Infrared Spectroscopy (FTIR) were used to distinguish the various stages of the phase separation process. During the synthesis of DPOD, it is required that Primacor™ be partially neutralized with potassium hydroxide (KOH). Knowing this information, XPS survey spectra were collected for different stages of the phase separation process (shown in ***Figure S2c***). For a DPOD film cured at room temperature, the XPS spectrum (shown in red) shows a K2s peak, indicating Primacor™ at the surface (only Primacor™ contains potassium). At the various stages of heat treatment (*e.g.* transition from red to black XPS spectra), the K2s peak disappears, indicating that Affinity™ has moved to the surface. In addition, FTIR analysis for a DPOD film undergoing phase separation (***Figure S2d***), also supports the sequence depicted in ***Figure S2a***. Initially, for a film cured at room temperature, the FTIR spectrum (***Figure S2d*** – red curve) shows primary polyethylene-like peaks (at 2920, 2851, 1466 $cm^{-1}$) and peaks representative of poly(acrylic acid) in the fingerprint region (at 1550, 1408 $cm^{-1}$). After heat treatment, in the FTIR spectrum for a fully phase-separated DPOD film (***Figure S2d*** – black curve), the poly(acrylic acid) peaks have disappeared, indicating that the phases of the DPOD film have fully separated.

# II. Contact-Angle Measurements

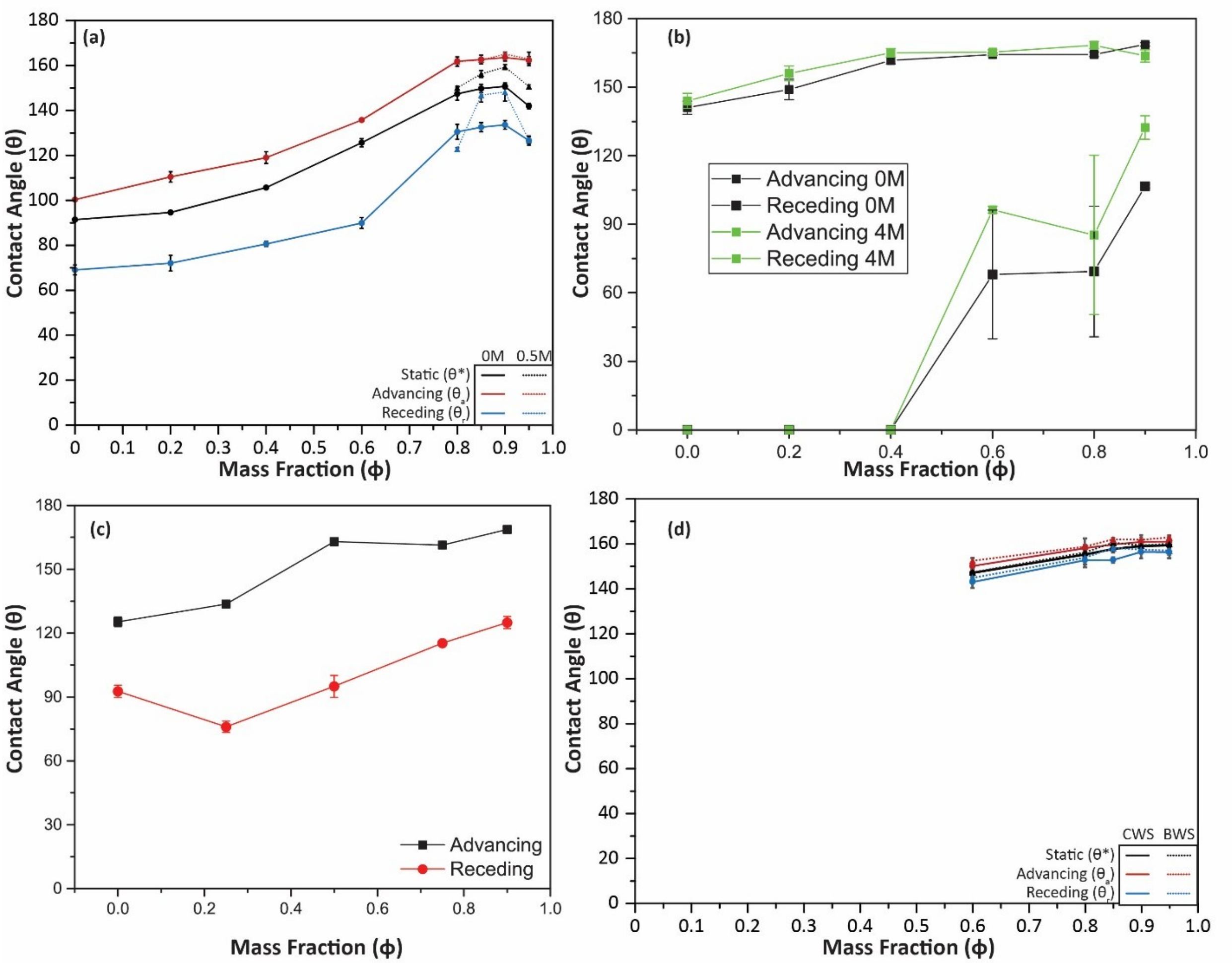


**Figure S3:** Water contact angles as a function of filler mass fraction ($\varphi$) in total solids. **(a)** MCC:DPOD, **(b)** MCC:AKD, **(c)** MCC:DPOD:AKD, and **(d)** Lycopodium with the natural wax suspensions

## III. SEM Cross-section of L:CWS Coatings

Cross section SEM images were taken of the L:CWS coating with mass fraction (φ) of 0.85 on different substrates, specifically glass and polyethylene terephthalate (PET). These images were obtained by spraying the L:CWS coating onto a given substrate and visualizing the edge of the sample (***Figure S4***).

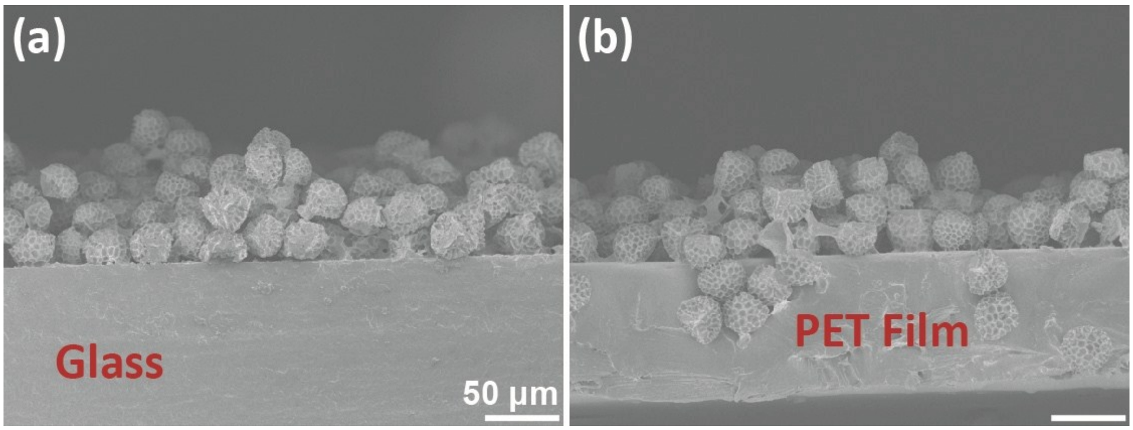


**Figure S4:** Cross section scanning electron microscopy images of the L:CWS coating (φ = 0.85) on **(a)** glass, and **(b)** polyethylene terephthalate (PET) substrates.