## **Supporting Information**

Title: Superhydrophobic Controlled-Released Fertilizer Coated with Bio-Based Polymer with Organosilicone and Nano-Silica Modifications

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<sup>3</sup>Department of Soil and Water Science, Tropical Research and Education Center, IFAS, University of Florida, Homestead, FL 33031, USA. **Analysis of liquefied locust sawdust.** The liquefaction ratio of biopolyols from locust sawdust was 99.12%, and the hydroxy value was 244.56 mg KOH/g (Table S1).

| Treatments     | Liquefaction | Acid value Hydroxy value |            |
|----------------|--------------|--------------------------|------------|
|                | ratio (%)    | (mg KOH/g)               | (mg KOH/g) |
| Locust sawdust | 99.12        | 44.48                    | 244.56     |

 Table S1. Analysis of liquefied indexes

Table S2 Chemical composition of each fertilizer

| Treatments | LWS   | MDI   | OS    | NS   |
|------------|-------|-------|-------|------|
|            | (%)   | (%)   | (%)   | (%)  |
| BPCF       | 33.33 | 66.67 | 0.00  | 0.00 |
| SBPCF-10   | 29.67 | 59.33 | 10.00 | 1.00 |
| SBPCF-20   | 26.33 | 52.67 | 20.00 | 1.00 |
| SBPCF-30   | 23.00 | 46.00 | 30.00 | 1.00 |



Fig. S1 TEM of nano-SiO<sub>2</sub>

**Structure Analysis of Coating Shells by FIIR and <sup>13</sup>C-NMR.** The FTIR spectra of WS, LWS, BPCF and SBPCF revealed chemical shifts during biomass liquefaction and the synthesis process of polyurethane (Figure S2). For example, in WS the existence of -OH, C=C, and C-C bonds corresponded to three characteristic absorption peaks for cellulose at 3416, 1636 and 1121 cm<sup>-1</sup>, respectively (Fig. S2 A-a). In LWS, however, a right shift was observed for -OH at 3256 cm<sup>-1</sup>, C=C-H of the aromatic ring at 1570 cm<sup>-1</sup> and 1475 cm<sup>-1</sup> along with characteristic absorption peak at 1200 cm<sup>-1</sup> representing -C-C-bonds (Fig. S2 A-b). The active group of LWS was more than WS.

The bonds of N-H (3300 cm<sup>-1</sup>), C-H (3100cm<sup>-1</sup>), C=N=O (2256 cm<sup>-1</sup>) C-O-C (1200 cm<sup>-1</sup>) absorption peaks of BPCU shell was observed (Fig. S2 B-a). The results confirmed that polyurethane was created by the reaction between LWS and MDI. For SBPCF, except above bonds, the characteristic absorption peaks at 1103 and 811cm<sup>-1</sup> corresponded to the stretching vibration of -Si-O and C-Si, respectively (Fig. S2 B-b). It indicated that OS was grafted to the polyurethane.

To further confirm the formation of coating materials, <sup>13</sup>C-NMR was employed to detect the change of chemical shift of C atoms in coating materials of BPCF and SBPCF. The peaks at  $\delta$ =40 ppm,  $\delta$ =70.1ppm,  $\delta$ =130-138 ppm and  $\delta$ =155 ppm represented the methylene carbons connected with benzene rings, newly-emerging carbon atoms directly attached to oxygen atoms, aromatic carbons of benzene rings and carbonyl carbons of ester linkages, respectively. They are common for BPCF and SBPCF coatings (Fig. S2 C and D). In addition, the peaks at 61-66 ppm and 118 ppm was observed only in SBCF coatings represented methylene carbon attached to urethane linkage and the aromatic carbons of benzene rings, respectively (Fig. S2 D). These results further indicated the successful fabrication of BPU coating. It can be concluded that BPU coating has been successfully fabricated by the thorough polymerization reaction based on the FTIR and <sup>13</sup>C-NMR results



**Fig. S2** FTIR spectra of WS (A-a), LWS (A-b), BPCF (B-a) and SBPCF (B-b); <sup>13</sup>C-NMR spectra of solid coating materials, BPCF (C) and SBPCF (D).

**Differential scanning calorimetry of five coating materials.** The coatings of BPCU, EMPCUs and EPCU were dried at 40°C for 24h. Then the coating materials were tested with Differential Scanning Calorimeter. Glass transition temperature (Tg) play an important role in polymer applied. The Tg of SBPCFs was higher than that of BPCF (Fig. S3). Hence, the cross-linking degree of SBPCFs was higher than BPCF. As shown in Fig. S4, the void rate of BPCF was higher than SBPCFs. In indicated that SBPCF was more densified than BPCF.



Fig. S3. DSC of the coating materials.



Fig. S4 Void rate of coating materials, different letters above the column indicate statistical significance at the P = 0.05 level within the same measurement date. The testing method was followed with the national standard methods (GB/T 21650.3-2011)



Fig. S5 TGA of coating materials.



Fig. S6 XRD of SBPCF coatings.



Incubation in water for one day, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 7 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 14 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 30 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 60 days, from left to right was 10000x, 20000x and 100000x, respectively

Fig. S7 SEM of BPCF after nutrient release in water



Incubation in water for one day, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 7 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 14 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 30 days, from left to right was 10000x, 20000x and 100000x, respectively



Incubation in water for 60 days, from left to right was 10000x, 20000x and 100000x, respectively

Fig. S8 SEM of SBPCF after nutrient release in water



Fig. S9 Reaction process of LWS (A), Photo of WS and LWS (B), Photo of BPCF

and SBPCF(C).



Fig. S10 AFM roughness analysis of BPCF (A), SBPCF-10(B), SBPCF-20 (C)

and SBPCF-30 (D); Surface energy of four coating materials (E)