

Supplementary Materials

Additive Manufacturing of Shape-Changing Oral Tablets via Powder-Based Extrusion 3D Printing of Natural Cellulose and Polyvinyl Alcohol

Kasidit Dokhom ¹, Pensak Jantrawut ^{1,2}, Pattaraporn Panraksa ¹, Suruk Udomsom ^{3,4}, Wirongrong Tongdeesoontorn ^{5,6}, Baramee Chanabodeechalermrung ¹, Pornchai Rachtanapun ^{2,7}, and Tanpong Chaiwarit ^{1,*}

¹ Department of Pharmaceutical Sciences, Faculty of Pharmacy, Chiang Mai University, Chiang Mai 50200, Thailand; kasidit_dokh@cmu.ac.th (K.D.); pensak.j@cmu.ac.th (P.J.); pattaraporn.pan@cmu.ac.th (P.P.); baramee.c@cmu.ac.th (B.C.); tanpong.ch@cmu.ac.th (T.C.)

² Center of Excellence in Agro Bio-Circular-Green Industry (Agro BCG), Agro-Industry, Chiang Mai University, Chiang Mai 50100, Thailand

³ Department of Electrical Engineering, Faculty of Engineering, Chiang Mai University, Chiang Mai, 50200 Thailand; suruk.u@cmu.ac.th (S.U.)

⁴ Office of Research Administration, Chiang Mai University, Chiang Mai 50200, Thailand

⁵ School of Agro-Industry, Mae Fah Luang University, 333 Moo 1 Tasud, Chiang Rai 57100, Thailand; wirongrong.ton@mfu.ac.th (W.T.)

⁶ Research Center of Innovative Food Packaging and Biomaterials Unit, Mae Fah Luang University, 333 Moo 1 Tasud, Chiang Rai 57100, Thailand

⁷ Faculty of Agro-Industry, Chiang Mai University, Mae-Hea, Mueang, Chiang Mai 50100, Thailand; pornchai.r@cmu.ac.th (P.R.)

* Correspondence: tanpong.ch@cmu.ac.th; Tel.: +66882-610-254

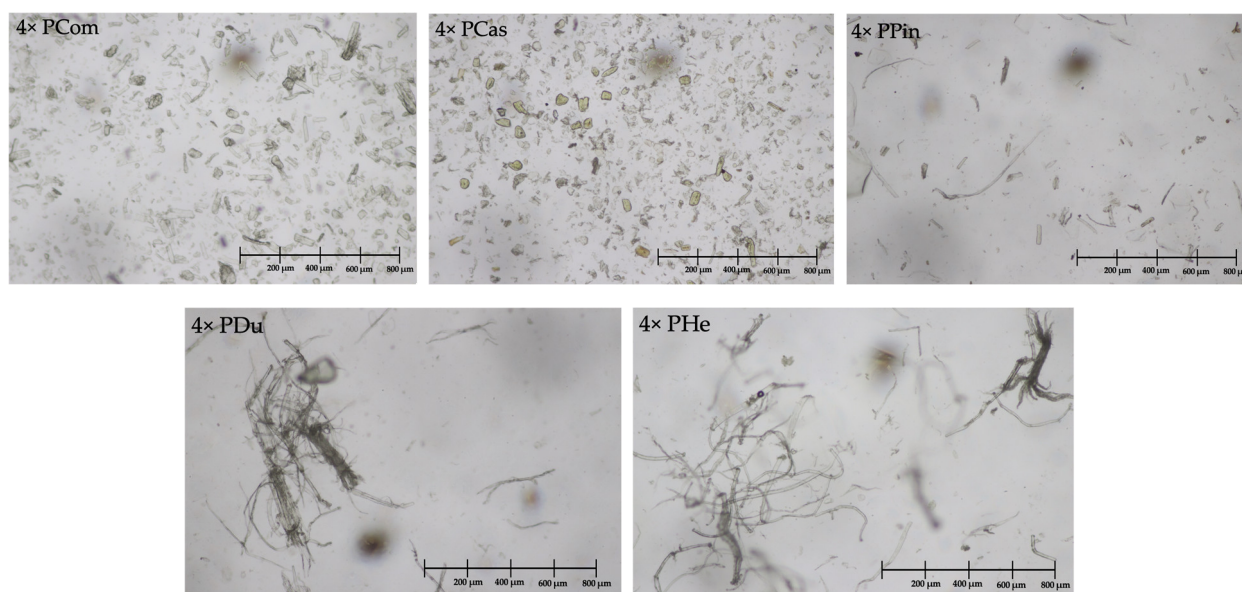


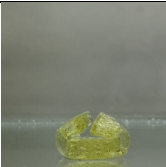
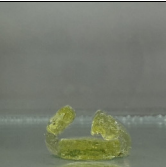
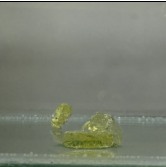
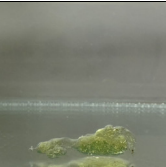
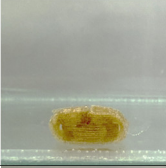
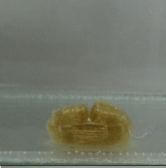
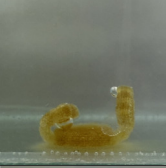
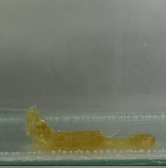
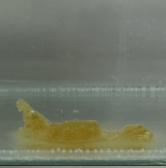
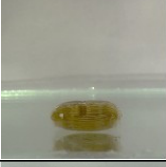
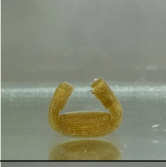
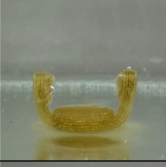
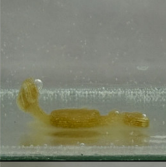
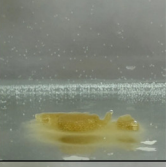
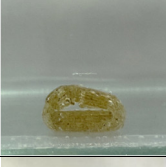
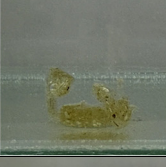
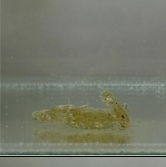
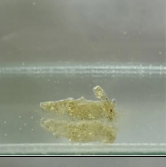
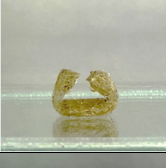
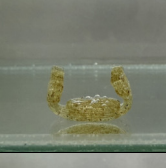
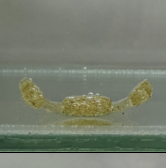
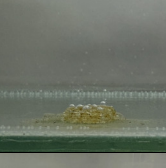
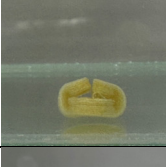
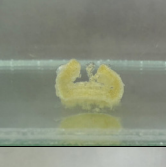
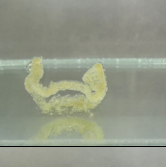
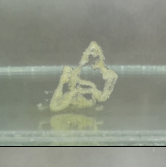
Figure S1. Powder appearance of PCom2.5, PCas2.5, PPin2.5, PDu2.5 and PHe2.5 formulations on 4× magnification under a microscope (Eclipse E200, Nikon Corporation, Tokyo, Japan)

Table S1. Equipment and experimental parameters of Printlets characterization

Section	Equipment	Condition
Morphology	Scanning electron microscopy (SEM; Tescan Clara™, Tescan Essence Software, Brno, Czech Republic)	Morphological characteristic was observed by SEM, operated at 15 kV under high vacuum. Samples were fixed on aluminum stubs using double-sided carbon tape, sputter-coated with gold of 5 nm thickness, and imaged at 50× magnification.
XRD analysis	X-ray diffractometer (Empyrean Series 3; Malvern Panalytical, Malvern, UK)	Crystallinity of samples before and after printing was evaluated by X-ray diffraction (XRD) using an X-ray diffractometer (Empyrean Series 3; Malvern Panalytical, Malvern, UK). Diffraction patterns were collected over 10–60° (2θ) at a scan speed of 10°/min with a step size of 0.01°, operating at 40 kV.
Thermal properties	Differential scanning calorimetry (DSC; Mettler Toledo Stare System, DSC N1 Module, Greifensee, Switzerland)	Samples (2 mg) were sealed in 40-μL platinum pans and analyzed by differential scanning calorimetry (DSC; Mettler Toledo Stare System, DSC N1 Module, Greifensee, Switzerland) over a temperature range of 25–350 °C at a heating rate of 2 °C/min under a nitrogen gas environment (50 mL/min).
Mechanical properties	TA Plus (Stable Micro Systems, Surrey, UK)	The mechanical properties of the printlets were evaluated using a TX. TA Plus (Stable Micro Systems, Surrey, UK) equipped with a 5-kg load cell with a sensitivity of 0.001 N, operating in compression mode. Samples were compressed using a flat, plane-faced probe with a diameter of 2 mm at the rate of 2 mm/s.
Drug loading content	HPLC (Agilent 1260 Infinity; Agilent Technologies, Waldbronn, Germany)	Drug-loaded printlet was dissolved in 100 mL of deionized water (DI), filtered through a 0.45 μm membrane filter (Zhejiang, China), and diluted with DI water, respectively. The drug concentration was determined by HPLC (Agilent 1260 Infinity; Agilent Technologies, Waldbronn, Germany). The HPLC was performed with 5 μL of injection volume. An isocratic mobile phase composed of 20 mM KH ₂ PO ₄ (pH 2.5) and HPLC-grade methanol (95:5, v/v) was delivered at a flow rate of 1 mL/min for 10 minutes through an Ascentis® C18 column (5 μm, 25 × 4.6 mm; Merck KGaA, Darmstadt, Germany) maintained at 40 °C. Detection of the eluent was carried out at 230 nm. Calibration curve of levodopa with high linear relationship (R ² = 0.999) was prepared in ranging from 25 to 200 μg/mL in methanol.

Drug releasing properties	HPLC (Agilent 1260 Infinity; Agilent Technologies, Waldbronn, Germany)	Each sample was placed in a 150 mL conical flask containing 100 mL of deionized water and stirred with a magnetic stirrer at 50 rpm at 37 ± 2 °C. Samples were collected at 1, 5, 10, 15, 45, 240, and 1440 min, filtered through 0.45 µm membranes (Zhejiang, China), and diluted with KH_2PO_4 , respectively. Drug concentrations were determined by HPLC (Agilent 1260 Infinity) using the same chromatographic conditions as described in Section 2.8 (drug loading). Each test was conducted in triplicate
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Table S2. Shaping changing of PPVA, PCom5.0, PCas5.0, PPin2.5, PPin5.0, PPVA, PCom5.0-L, PCas5.0-L, and PPin5.0-L

	Time (min)				
	0	5	10	15	20
PPVA					
PCom5.0					
PCas5.0					
PPin2.5					
PPin5.0					
PPVA-L					
PCom5.0-L	