

Supplementary Material

A General Simulation-Based Study on Printability of Inks in Direct Ink Writing

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Table S1. Herschel-Bulkley parameters obtained from curve fitting.

Materials		τ_0 (Pa)	$\dot{\gamma}_0$ (s⁻¹)	n	K (Pa-s)
Cement	Unmodified Cement	13.72	3.12	0.58	0.65
	Modified Cement	27.44	1.0134	0.62	13.49
CNC	CNC 6%	2.50	0.16	0.73	4.95
	CNC 8%	10.00	0.004	0.62	200.00
	CNC 20%	687.72	0.004	0.70	10325.30
Epoxy	Epoxy CNT	8.00	1.00	0.43	2.49
	Epoxy CNT modified	500.00	1.02	0.58	276.62
Hydrogel (Alginate-based)	Hydrogel 2	1.5	0.032	0.61	75.32
	Hydrogel 3	25.00	0.030	0.51	976.13
	Hydrogel 4	100.00	0.047	0.40	6190.35
Hydrogel (Ti64-based)	Ti 20%	140.00	1.03	0.60	9.50
	Ti 25%	2350.00	1.10	0.70	64.64
	Ti 30%	3400.00	1.05	0.62	135.17

Table S2. Values of the critical radius for different pre-treated ink compositions and types.

Materials		Critical radius, r_c (mm)
Cement	Unmodified Cement	0.287
	Modified Cement	0.067
CNC	CNC 6%	0.024
	CNC 8%	0
	CNC 20%	0
Epoxy	Epoxy CNT	0.182
	Epoxy CNT modified	0.081
Hydrogel (Alginate-based)	Hydrogel 2	0.004
	Hydrogel 3	0.010
	Hydrogel 4	0.011
Hydrogel (Ti64-based)	Ti 20%	0.265
	Ti 25%	0.304
	Ti 30%	0.305

Table S3. Values of the critical yield shear strength to shear rate ratio (β) for different pre-treated ink compositions and types.

Materials		Rheology Modifier	β, (Pa.s)
Cement	Unmodified Cement	Nano Clay	4.40
	Modified Cement		27.08
CNC	CNC 6%	CNC	9.38
	CNC 8%		2564.10
	CNC 20%		171930.00
Epoxy	Epoxy CNT	Fumed Silica	8.00
	Epoxy CNT modified		490.20
Hydrogel (Alginate-based)	Hydrogel 2	Calcium Salt Cations with cross-linking polymers	46.88
	Hydrogel 3		833.33
	Hydrogel 4		2127.66
Hydrogel (Ti64-based)	Ti 20%	50 vol% Ti-64 powder	135.92
	Ti 25%		2136.36
	Ti 30%		3238.10

Table S4: Values of the printability grading parameter for different ink compositions and types.

Materials		Printability grading parameter, α (Pa.s)
Cement	Unmodified Cement	0.03
	Modified Cement	0.15
CNC	CNC 6%	0.01
	CNC 8%	0.53
	CNC 20%	17.13
Epoxy	Epoxy CNT	0.06
	Epoxy CNT modified	3.19
Hydrogel (Alginate-based)	Hydrogel 2	0.04
	Hydrogel 3	1.38
	Hydrogel 4	9.87
Hydrogel (Ti64-based)	Ti 20%	0.36
	Ti 25%	2.80
	Ti 30%	5.88

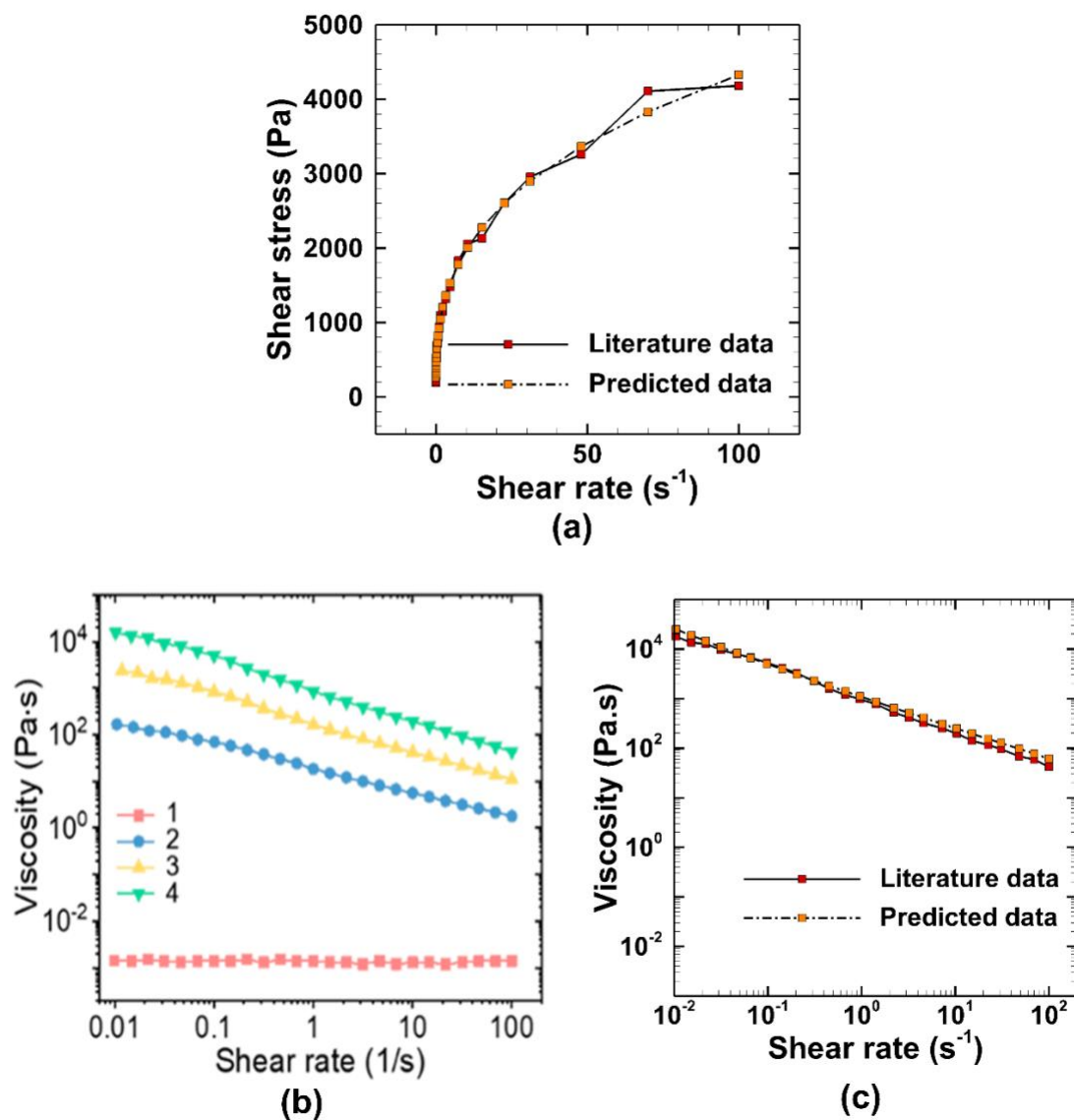


Fig. S1. Illustration of prediction of Herschel-Bulkley parameters via the curve fitting method of Hydrogel 4. (a) curve fitting of shear stress data; (b) the characteristic curve of Hydrogel inks obtained from literature (Ref32); (c) curve fitting of viscosity with the literature data of Hydrogel 4 (Ref32).

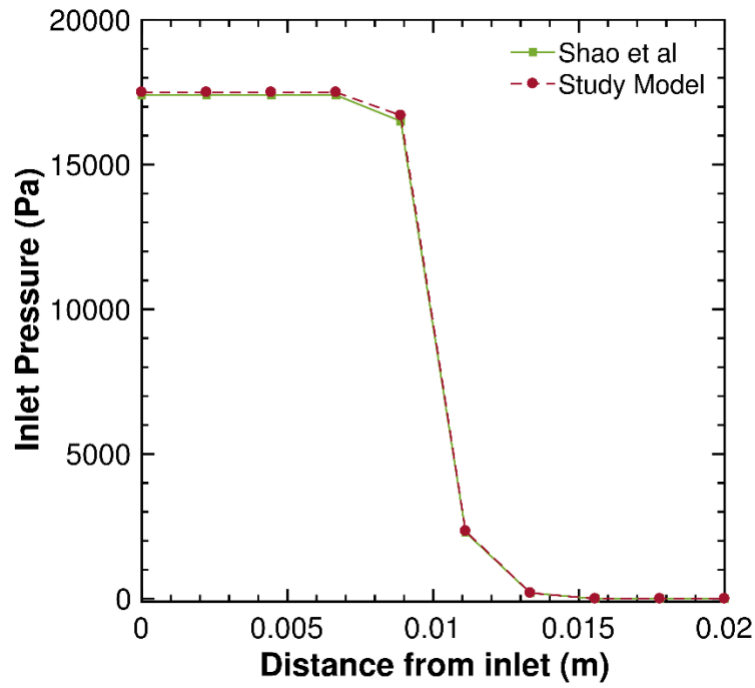


Fig. S2. Comparison of the pressure distribution inside the nozzle for the current numerical simulation and the results obtained by Shao *et al.* (Ref30).

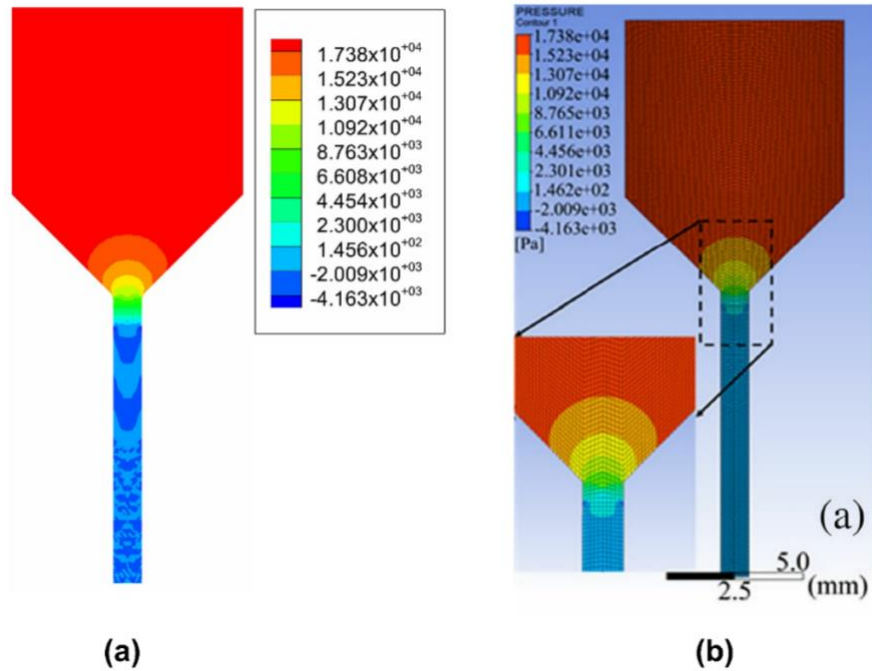


Fig. S3. Comparison of the pressure contour obtained by: (a) current numerical simulation; (b) Shao *et al.* (Ref30).

Formulation of ink materials

This study has used ink materials from existing literature. The material preparation procedures found in the literatures are briefly explained in this section. Detailed information can be found in the literatures.

Cement inks

The preparation of cement inks are as follows (Ref15)-

Materials: Class G Portland cement, bentonite nano-clay, Hydroxyethyl-cellulose, Polycarboxylate ether, and 2-Ethyl-1-hexanol.

Preparation of inks: The initial step involved combining a suitable quantity of nano-clay and hydroxyethyl-cellulose with Class G Portland cement. Subsequently, a solution was created by combining polycarboxylate ether and 2-ethyl-1-hexanol with water utilizing a Vortex Mixer. The components were combined in a small container and blended using a Planetary Centrifugal Mixer.

Cellulose inks

The preparation of CNC inks are as follows (Ref31)-

Materials: HEMA (98%), methacrylic anhydride (94%), pyridine, difunctional PUA oligomers (PUA-BR571 and PUA-BR3741AJ), Irgacure 819, DMF ($\geq 99.5\%$), Hydrophobic pyrogenic silica, eucalyptus pulp, and sulfuric acid.

Preparation of inks: CNC suspensions/gels were made by diluting deionized water with quantities varying from 8 to 20 wt.%. Subsequently, the HEMA monomer was introduced and stirred for a few minutes. To obtain CNC gel, water was evaporated using a rotavapor at a temperature of 60°C and a pressure of 30 mbar. Ultimately, the gel is supplemented with PUA and Irgacure, and then thoroughly blended using a planetary mixer.

Epoxy-CNT inks

The preparation of CNC inks are as follows (Ref33)-

Materials: Multi-walled CNTs (Nanocyl NC7000), Epon 826, Jeffamine D230, and Fumed Silica (8.5 wt.%).

Preparation of inks: Initially, the Epon 826 and Jeffamine D230 (epoxy resin) were subjected to a temperature of 100°C in order to decrease its viscosity. Subsequently, CNTs were introduced into the heated resin at concentrations of 0.1%, 0.3%, and 1 wt.%. Subsequently, the mixture was homogenized utilizing a planetary centrifugal mixer. The Epoxy-CNT mixture was additionally heated to 130°C in order to decrease viscosity for the purpose of sonication. Subsequently, the heated mixture was moved to a hot plate that was set at a temperature of 100°C. Following sonication, a concentration of 8.5 wt.% fumed silica was introduced into the Epoxy-CNT mixture to formulate modified Epoxy-CNT. Finally, the mixture was stirred using the planetary mixer.

Hydrogel (alginate-based) inks

The preparation of alginate-based Hydrogel inks are as follows (Ref32)-

Materials: Acrylamide (AAM), N, N'-Methylenebis(acrylamide) (MBAA), Ammonium persulfate (APS), Sodium alginate and Calcium chloride.

Preparation of inks: Initially, AAM, MBAA, and APS were dissolved in distilled water, often in amounts of 10 mL. Subsequently, varying quantities of sodium alginate and calcium chloride were used to produce four distinct hydrogel inks. A spatula was employed to thoroughly blend the ingredients, as the alginate necessitated intense mixing.

Hydrogel (Ti powder-based) inks

The preparation of alginate-based Hydrogel inks are as follows (Ref20)-

Materials: Pluronic F127 (poly(ethylene oxide)-poly(propylene oxide)-poly(ethylene

oxide), Ti64 powder (Ti-6Al-4V), and Distilled water.

Preparation of inks: At first, the binder solutions were prepared by dissolving Pluronic F127 in distilled water at concentrations of 20, 25, and 30 wt.%. The solutions were held at a temperature of 4°C to ensure that they remained in their liquid condition below the temperature at which they transition into a gel. Then the binder solution was filled with Ti64 powder at a volume fraction of 50%. Finally, the mixtures were stirred utilizing a centrifugal mixer.