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Printability Mapping of HPMC Bioinks for Electrohydrodynamic Jet Printing

Joana Alves Lopes[1,2*], Gabriel Moreira[1], Artur Martins[1], Pedro Miguel Silva[1], Rui M. R. Pinto[1], Michele Michelin[2], Patrícia Sousa[1], Lorenzo M. Pastrana[1], Miguel A. Cerqueira[1]

[1]International Iberian Nanotechnology Laboratory, Avenida Mestre José Veiga s/n, 4715-330 Braga, Portugal;
[2] Centre of Biological Engineering (CEB), University of Minho, 4710-057 Braga, Portugal.
*joana.alveslopes@outlook.com

INTRODUCTION & AIM

In the food technology field, the main consumers' concerns lie in the food safety, label transparency and the environmental impact of the product. While smart packaging innovations enhance monitoring and preservation, gaps remain in sustainable technologies for sensor development. Electrohydrodynamic (EHD) jet printing is a new micro-additive manufacturing technology that uses electric fields to precisely deposit material through a nozzle, achieving high resolutions with high viscosity inks. Seeing as it is a recent technology, bio-based inks have yet to be designed and optimized. Hydroxypropyl methylcellulose (HPMC) stands out as a biodegradable biopolymer with excellent compatibility, paving the way for sustainable smart packaging sensors. The addition of fluorescent biomaterials, such as lignin, can broaden the final application of the jetted bio-structures. Therefore, the objective of this work is to design and optimize bio-based HPMC inks for EHD jet printing to achieve the highest possible resolution.

METHOD

Formulation of bio-inks:

HPMC Concentration %(w/w)		
1	2	3

HPMC powder supplied from Thermo Scientific (viscosity in 2% aquous solution of 40-60 mPa.s at 20°C)

Ethanol/Water Ratio	
0:10	5:5
1:9	6:4
2:8	7:3
3:7	8:2
4:6	9:1

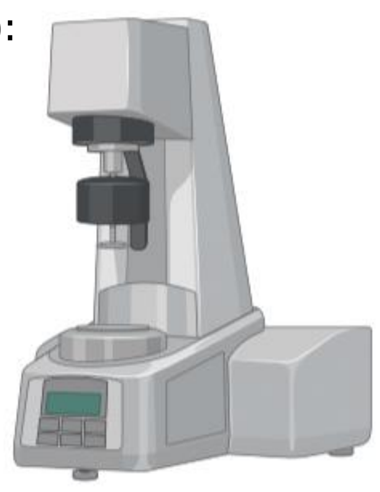
Analysis of bio-ink's properties:

Shear Rate ($\dot{\gamma}$):

$$\dot{\gamma} = \frac{4 \cdot Q}{\pi \cdot r^3}$$

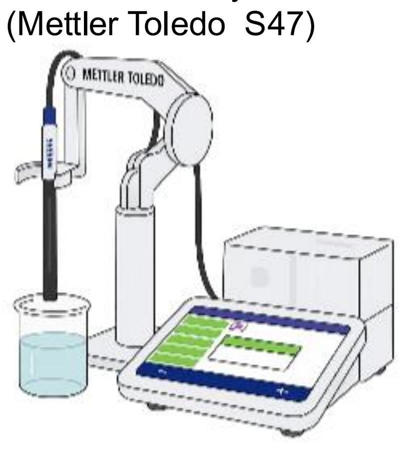
Power Law Approximation:

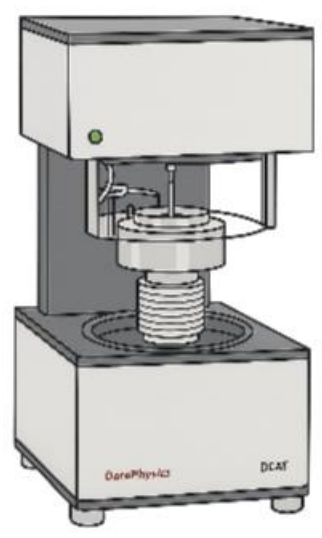
$$\eta = K \cdot \dot{\gamma}^{n-1}$$



Rheology
(Anton Paar, 302e)

Conductivity
(Mettler Toledo S47)



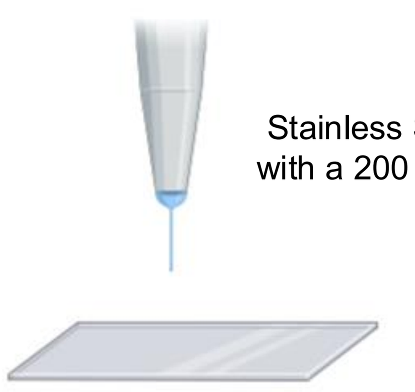


Surface Tension
(Dataphysics, DCAT 8T)

Analysis of printability with fixed parameters:

Flow rate (Q) (28.28 μL·h-1)

Working distance (1 mm)



Stainless Steel nozzle with a 200 μm diameter

Glass substrate with 100 nm layer of tungsten-titanium

CONCLUSION

- HPMC shows great potential as a bio-ink;
- The ideal properties of the inks were viscosity: 21-105 mPa·s; surface tension: 29-39 mN·m^{-1} and conductivity: 18-54 μS·cm^{-1};
- Resolutions lower than 100 μm were achieved (viscosity: 78.22 mPa·s; surface tension: 35.05 mN·m^{-1} and conductivity: 43.03 μS·cm^{-1});
- Fluorescent microstructures were obtained with 0.02% (w/v) of lignin incorporated into the HPMC-based bio-inks;
- Multiple applications can be seen in the food and biomedical fields.

RESULTS & DISCUSSION

Bio-inks' properties

Figure 1. Different HPMC bio-inks concentrations (1, 2 and 3% (w/w) in 80% (v/v) ethanol (left) and 90% (v/v) ethanol (right).

- HPMC is insoluble in ethanol concentrations above 90% (v/v)
- Viscosity starts to decrease above 50% (v/v) of ethanol
- Surface Tension does not vary with the HPMC content but is influenced by the ethanol concentration

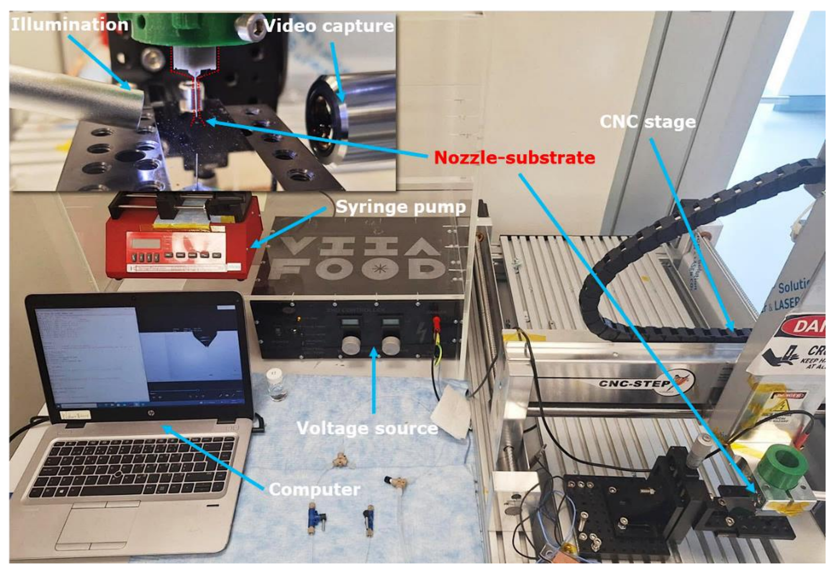


Figure 2. Home-made EHD jet printer.

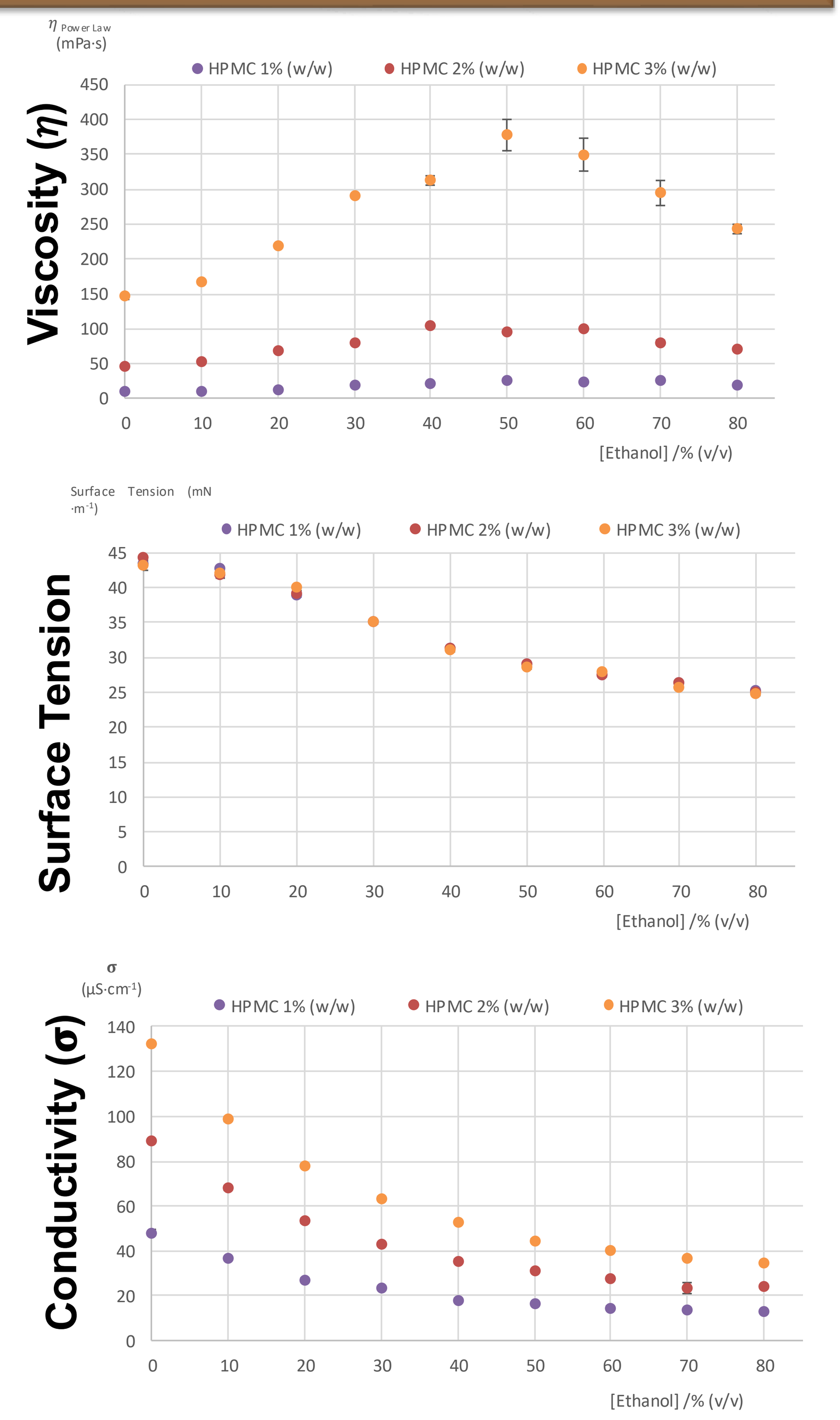


Figure 3. Properties (Viscosity (mPa·s), Surface Tension (mN ·m^{-1}) and Conductivity (μS·cm^{-1}) of HPMC-based bio-inks of different concentrations (1, 2 and 3% (w/w)) in different ethanol concentrations (0-80% (v/v)).

Printable Zone

- Above 50% (v/v) ethanol, evaporation leads to clogging of the nozzle
- At 0% (v/v) of ethanol, the surface tension force exceeds the eletric field force, making jetting impossible

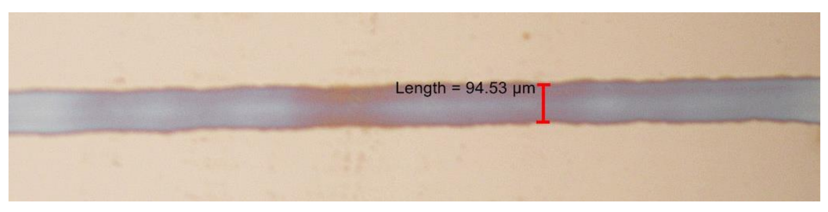


Figure 4. Best resolution (94.53 μm) achieved with 2% (w/w) HPMC in 30% (v/v) ethanol ink.

Printable solutions in fixed parameters:
- 1% (w/w) HPMC in 40 and 50% (v/v) ethanol
- 2% (w/w) HPMC in 20 to 50% (v/v) ethanol

Fluorescence of jetted micro-structures

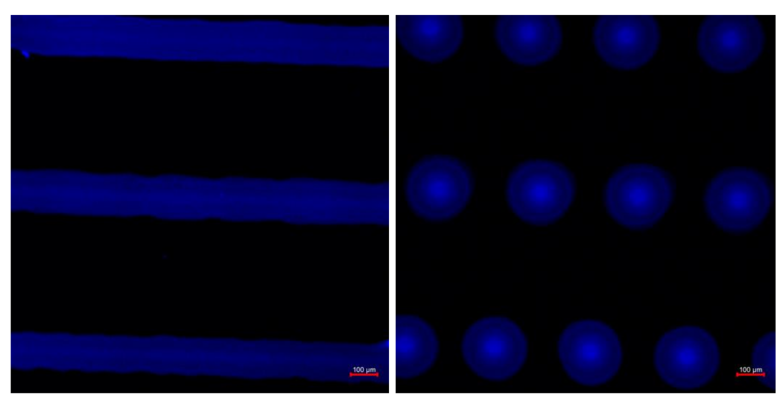


Figure 5. Confocal microscopic images of printed 2% (w/w) HPMC in 50% (v/v) ethanol with 0.02% (w/v) of Brewer's Spent lignin at a 414 nm emission wavelength and 344 nm excitation wavelength.

EHD jet printing parameters' evaluation

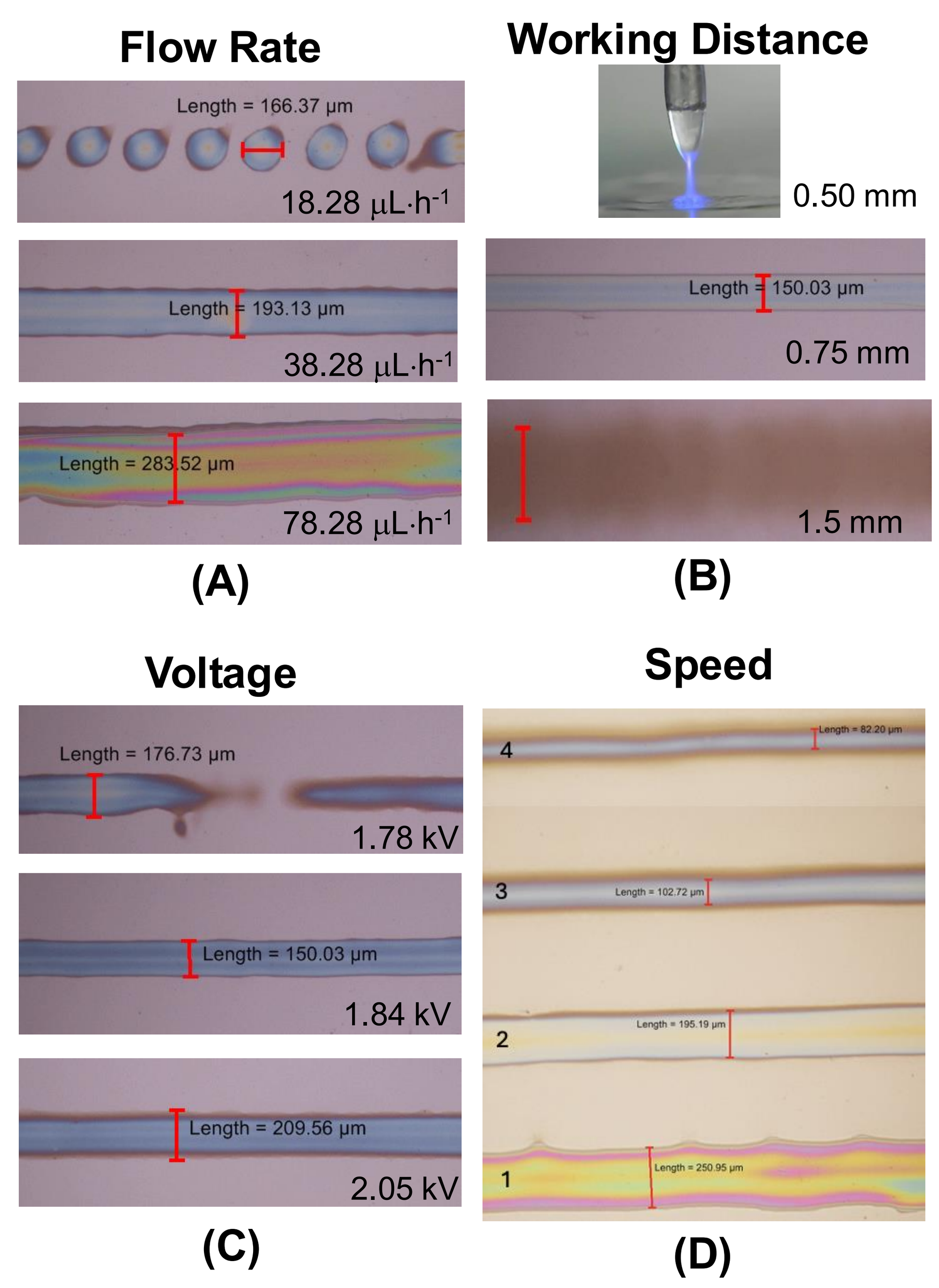


Figure 6. Bio-ink with 2% (w/w) HPMC in 30% (w/w) ethanol, printed **(A)** at a voltage of 2.05 kV speed of 4 mm·s^{-1} and a working distance of 1 mm, varying flow rate; **(B)** printed at a voltage of 2.05 kV speed of 4 mm·s^{-1} and a flow rate of 28.28 μL·h^{-1}, varying working distance; **(C)** at a speed of 4 mm·s^{-1}, a flow rate of 28.28 μL·h^{-1} and a working distance of 1 mm, varying voltage; **(D)** at a voltage of 1.96 kV, a flow rate of 28.28 μL·h^{-1} and a working distance of 1 mm, varying speed (3 to 6 mm·s^{-1}).

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Universidade do Minho
Escola de Engenharia
CENTRE OF BIOLOGICAL ENGINEERING
INL INTERNATIONAL IBERIAN NANOTECHNOLOGY LABORATORY