

Rapid microfluidic hydrogel imprint based on CNT-induced thermoresponsive effect

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Summary:

The study concerns the rapid development of precise, diverse-shaped hydrogel microfluidic structures utilizing CNT-based thermoresponsive effect. Biocompatible self-developed polymer compositions were prepared for the additive manufacturing of hydrogel matrices. Concurrently, 3D-printed structures covered with CNTs were fabricated in the form of heaters to imprint microfluidic channels into the hydrogel matrices. The experiments resulted in the precise formation of channels within the hydrogels, which is essential for further research in microfluidics and biomedical studies.

Keywords: microfluidics, hydrogel matrix, additive manufacturing, carbon nanotubes, imprinting

Background, Motivation, and Objective

Hydrogels made from natural polymers, such as sodium alginate or methylcellulose, are interesting solutions for diverse biomedical research, as they have unique properties. Hydrogel matrices equipped with microfluidic structures can create a specific environment for cellular research at the microscale, imitating the natural cell environment, i.e., ECM. On the other hand, CNTs are increasingly gaining popularity in microfluidic applications, mainly thanks to their high thermal and electric conductivity, chemical resistance, and biocompatibility. Moreover, the incorporation of CNTs into hydrogels can improve their mechanical strength, which expands their application fields significantly [1]. In this particular research, for the first time, CNTs-coated structures were used to obtain spatial microfluidic structures in hydrogel matrices, thanks to the thermoresponsive effect and utilizing the latest technological achievements of a 3D printing technique.

Description of the New Method or System

Our research was mainly focused on the fabrication of 1 W resistive heaters that serve as punches that can melt and form microchannels in a hydrogel matrix at high temperatures (approximately 140 °C). The stamps were 3D printed utilizing the DLP technique (Asiga printer, DentaStudy resin). The conductive elements of

the heater were made of SWCNT (OCSiA's Tuball Batt with 0.4% SWCNT and 0.8% poly(vinylpyrrolidone) (PVP) in H₂O), and the leads were made of silver-coated wire.

Hydrogel matrices fabrication proceeds as follows. Appropriate amounts of natural polymer powders, such as sodium alginate and methylcellulose (different concentrations), were weighed as shown in Tab. 1. A 0.9 % saline solution (NaCl) was then added as a solvent. The mixture was stirred at room temperature for 20 h on a magnetic stirrer. This allowed the obtainment of a homogeneous polymer solution - ink. The extrusion process was performed to obtain the designed matrix (nozzle type: 22G, process temperature: 25 °C, printing speed: 5 mm/s). The structure was then stabilized using a calcium chloride solution (CaCl₂, 1 M) as a crosslinking agent (t = 20 minutes).

Tab. 1: Different hydrogel compositions used for imprinting procedures

Hydrogel symbol	Hydrogels ingredients
1	3% Sodium alginate + 2% Methylcellulose + NaCl
2	5% Sodium alginate + 3% Methylcellulose + NaCl
3	3% Sodium alginate + 2% Methylcellulose + 0.1% CNT + NaCl

Results

The following 300 Ω resistance heaters were used to provide different patterns in the hydrogel structures, as shown in Fig. 1. The procedure for fabricating an imprint takes about 5 seconds, regardless of the sample. The conductive layer made of SWCNT is applied manually over the 3D-printed pattern. Each heater consists of 30 layers of SWCNTs, which corresponds to a total thickness of SWCNT layers of about 15 μm . Example results are shown in Figs. 2, 3, and 4. The results obtained for all the samples allow us to confirm that it is possible to obtain complex shapes in the hydrogel structures.

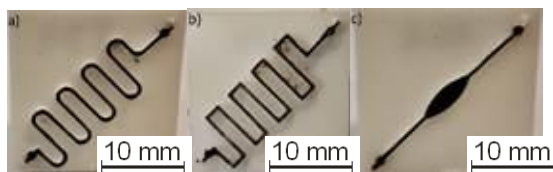


Fig. 1. Heaters with: a) meander rounded microchannel, b) meander right-angled microchannel, c) straight microchannel with widening. The width of the path of the heating element is 0.5 mm, the length of the heaters is 30 mm, and the depth of the pattern is 2 mm. The widening of the pattern from Fig. 1c) is 3 mm at the widest point.

In the case of samples defined as “1” hydrogel symbol, some inconvenience was the uneven surface of the sample, and the bubbles present inside, which deformed the imprinted pattern (Fig. 2).

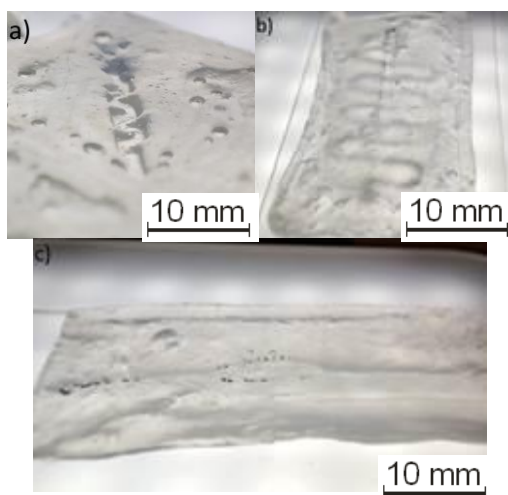


Fig. 2. Hydrogel patterns at a glance (“1” hydrogel symbol): a) meander heater with rounded edges, b) pattern laid on a glass substrate forming a microfluidic channel, c) straight microchannel with widening.

On the other hand, samples titled as “2” hydrogel symbol allow the best results in terms of pattern reproduction quality (Fig. 3). Thanks to the use of a hydrogel substrate, it was possible to create an even channel along its entire length. The only disadvantage of these samples is that making a deeper channel requires a longer annealing time, which causes boiling and consequent burning of the hydrogel.

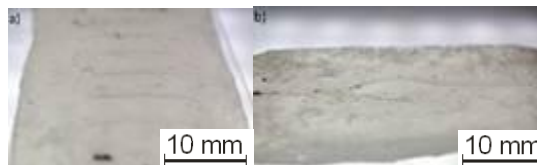


Fig. 3. Hydrogel patterns at a glance: a) meander heater with rounded edges, b) heater with widening.

In the case of the “3” hydrogel symbol, the patterns reproduce best in terms of depth but at the expense of hydrogel dissolution. It is worth mentioning that these samples are characterized by a very uniform structure and surface, which allows obtaining patterns of satisfactory quality, probably due to the addition of CNTs (Fig. 4).

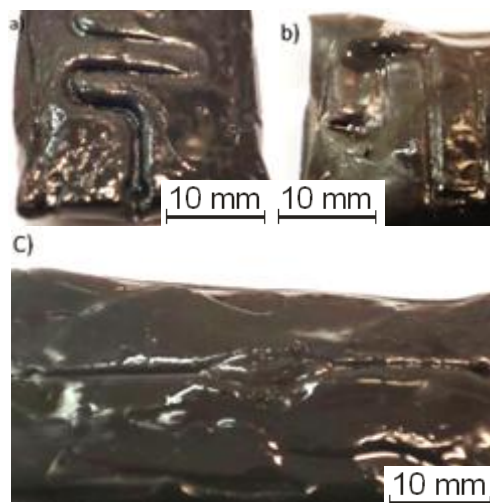


Fig. 4. Hydrogel patterns at a glance: a) meander heater with rounded edges, b) meander heater with right-angled edges, c) heater with widening.

In summary, a simple and easy-to-follow method of stamping patterns with SWCNT-layer resistive heaters makes it possible to swiftly produce microfluidic channels of versatile and complex shapes in hydrogel materials. This innovative idea could be the basis for further development of precise hydrogel structures on a microscale.

References

- [1] Cieślak A. *et al.*, *Microchimica Acta*, vol. 191, no. 10, p. 608, 2024.