Synthesis of Dual Stimuli-Responsive Semi-Interpenetrating Polymer Network (Semi-IPN) Hydrogel from CMC/PNIPAM for Biomedical Applications.

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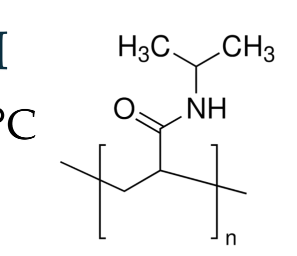
ABSTRACT

This research focuses on the synthesis of a smart hydrogel, a hydrogel system containing polymers that exhibit response to environmental stimuli particularly pH and temperature variations. The hydrogel was developed as a semi-interpenetrating polymer network (Semi-IPN) composed of carboxymethyl cellulose/crosslinked-poly (N-isopropyl acrylamide) (CMC/PNIPAM). Potassium persulfate (KPS) was employed as the initiator, tris[2-(dimethylamino) ethyl] amine (Me_6TREN) as the catalyst, and poly (ethylene glycol) diacrylate (PEGDA) as the crosslinker of PNIPAM moieties, with the molar ratio of crosslinker set as 0.125 % mol. The PEGDA molecular weight was kept constant at 575 Da. The synthesis process was monitored by characterizing the chemical structure using Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FTIR). Additionally, thermo-responsive behavior of solutions with 9% and 10% (w/w) semi-IPN polymer concentrations was analyzed using rheometer to monitor the change of viscoelastic properties of the materials as a function of temperature. The sol-gel phase transition of the polymer solutions was observed at 32 °C by test tube inversion method and the polymer concentration of 10% (w/w) exhibited the highest storage modulus of 121.23 Pascal. The on-going study includes increase the molecular weight of the crosslinker to 700 Da and the viscoelastic properties of the synthesized Semi-IPN will be further analyzed.

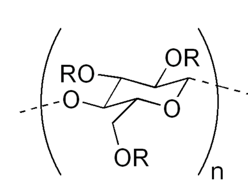
INTRODUCTION

Hydrogels are three-dimensional polymer networks made from natural or synthetic materials which can absorb large amounts of water. They do not dissolve but swell in water and aqueous solutions. They are also called 'smart' networks when these hydrogels exhibit significant physical and chemical changes in response to slight changes in their environment. Semi-Interpenetrating Polymer Networks (semi-IPNs) are built from two or more polymers, one of which is crosslinked, while the other is intercalated into the first one, either with a linear or branched architecture. Semi-IPNs in this study were synthesized from carboxymethyl cellulose (CMC) and poly(N-isopropyl acrylamide) (PNIPAM).

PNIPAM
LCST ~ 32 °C



- Thermo-responsive properties.
- Biocompatibility.



CMC
pKa ~ 4.3-4.7

R = H or CH_2CO_2H

- Hydrophilic polymer.
- pH-responsive properties.

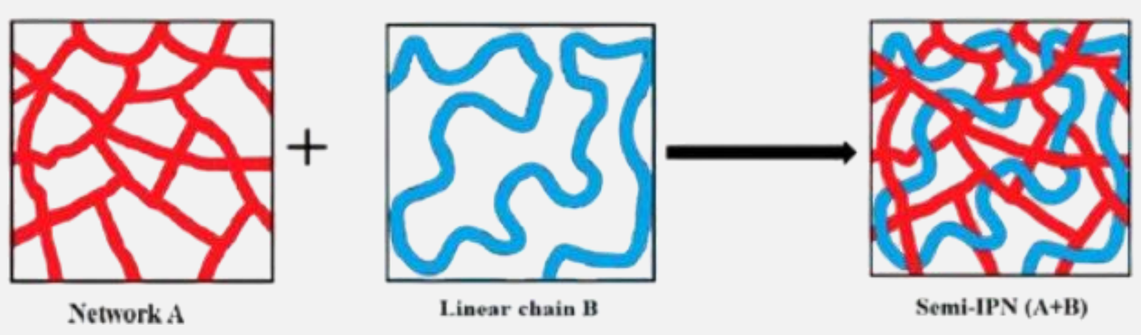


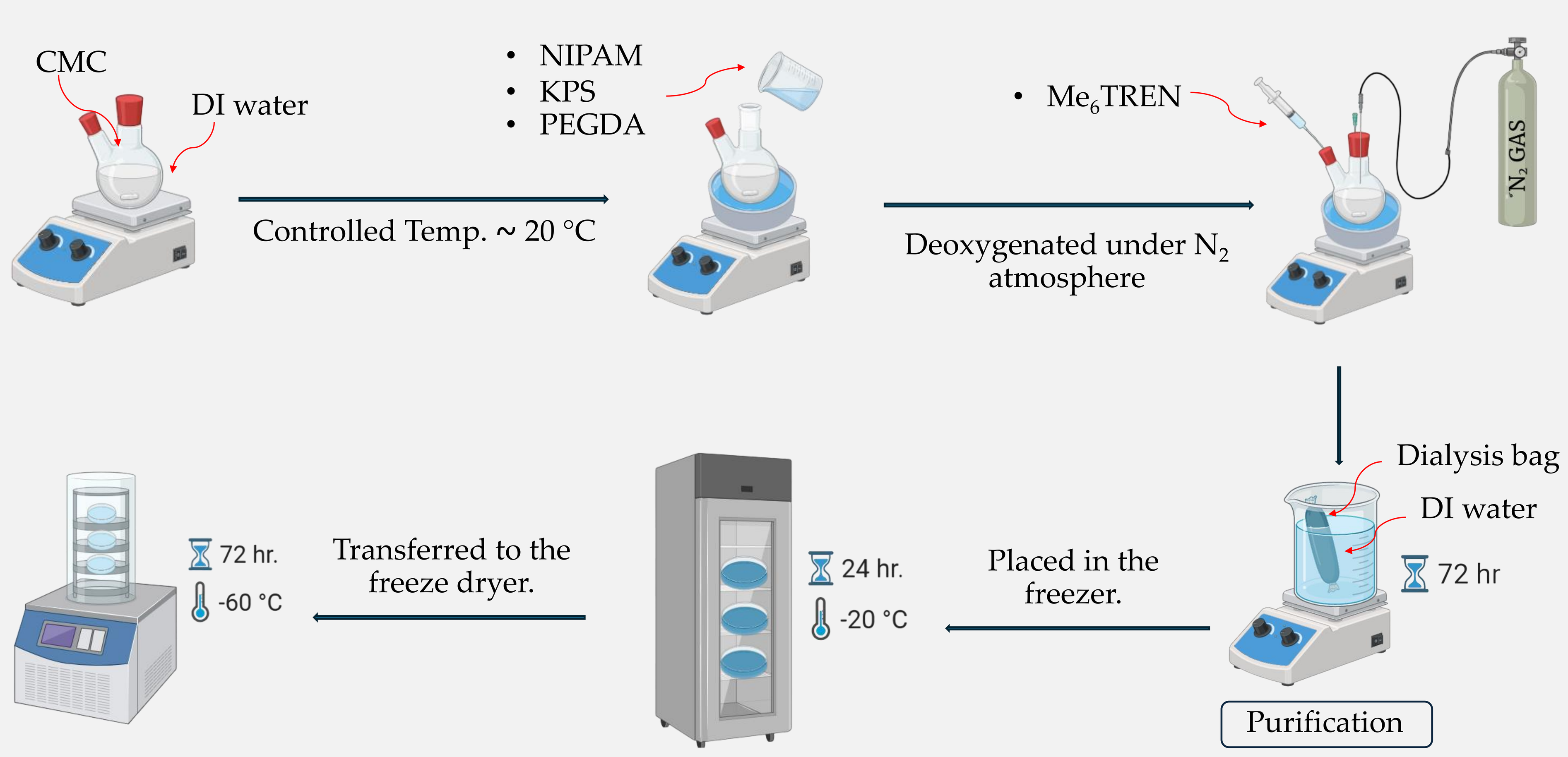
Figure 1. Schematic formation of Semi-Interpenetrating Polymer Network (Semi-IPN) structures.

OBJECTIVES

I. To synthesize the CMC/PNIPAM based semi-IPN hydrogels.
II. To characterize the chemical structure of semi-IPN hydrogels.
III. To study the viscoelastic properties of the synthesized semi-IPN hydrogels.

METHODOLOGY

- **Synthesis of CMC/PNIPAM Semi-Interpenetrating Polymer Network (Semi-IPN) hydrogel.**



RESULTS AND DISCUSSIONS

- **Structural characterization:** FTIR spectra of CMC, PNIPAM, Semi-IPN hydrogel (575) and Semi-IPN hydrogel (700)

Figure 2. Physical appearance of hydrogel after purification and hydrogel after freeze dry.

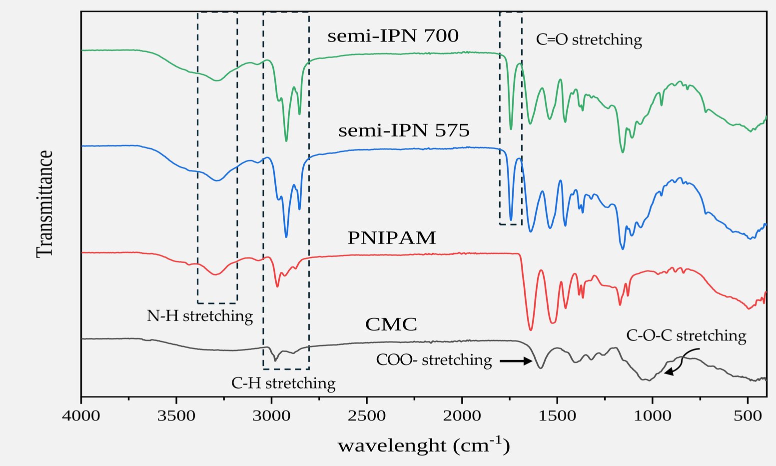


Figure 3. FTIR spectra of CMC, PNIPAM, Semi-IPN hydrogel (575) and Semi-IPN hydrogel (700)

- **Rheology:** Amplitude sweep test and Frequency sweep test of 9% and 10% (w/w) CMC/PNIPAM hydrogels both with 0.125% crosslinker.

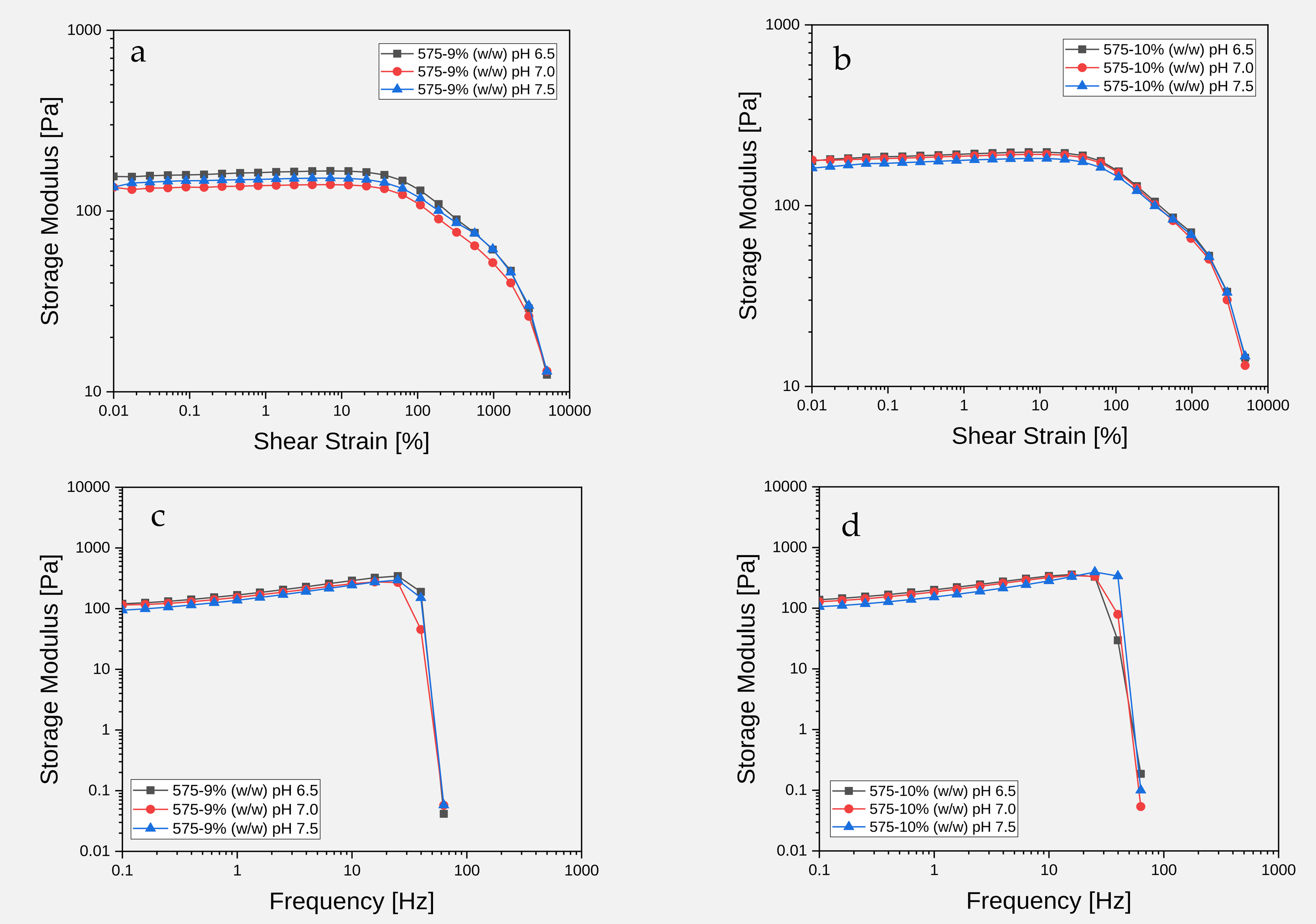


Figure 4. Rheology measurement ; **(a)** Amplitude sweep test of 9% (w/w) CMC/PNIPAM, **(b)** Amplitude sweep test of 10% (w/w) CMC/PNIPAM, **(c)** Frequency sweep test of 9% (w/w) CMC/PNIPAM and **(d)** Frequency sweep test of 10% (w/w) CMC/PNIPAM.

- **Thermo-responsive gelation :** CMC/PNIPAM hydrogels of 9% and 10% (w/w), 0.125% crosslinker.

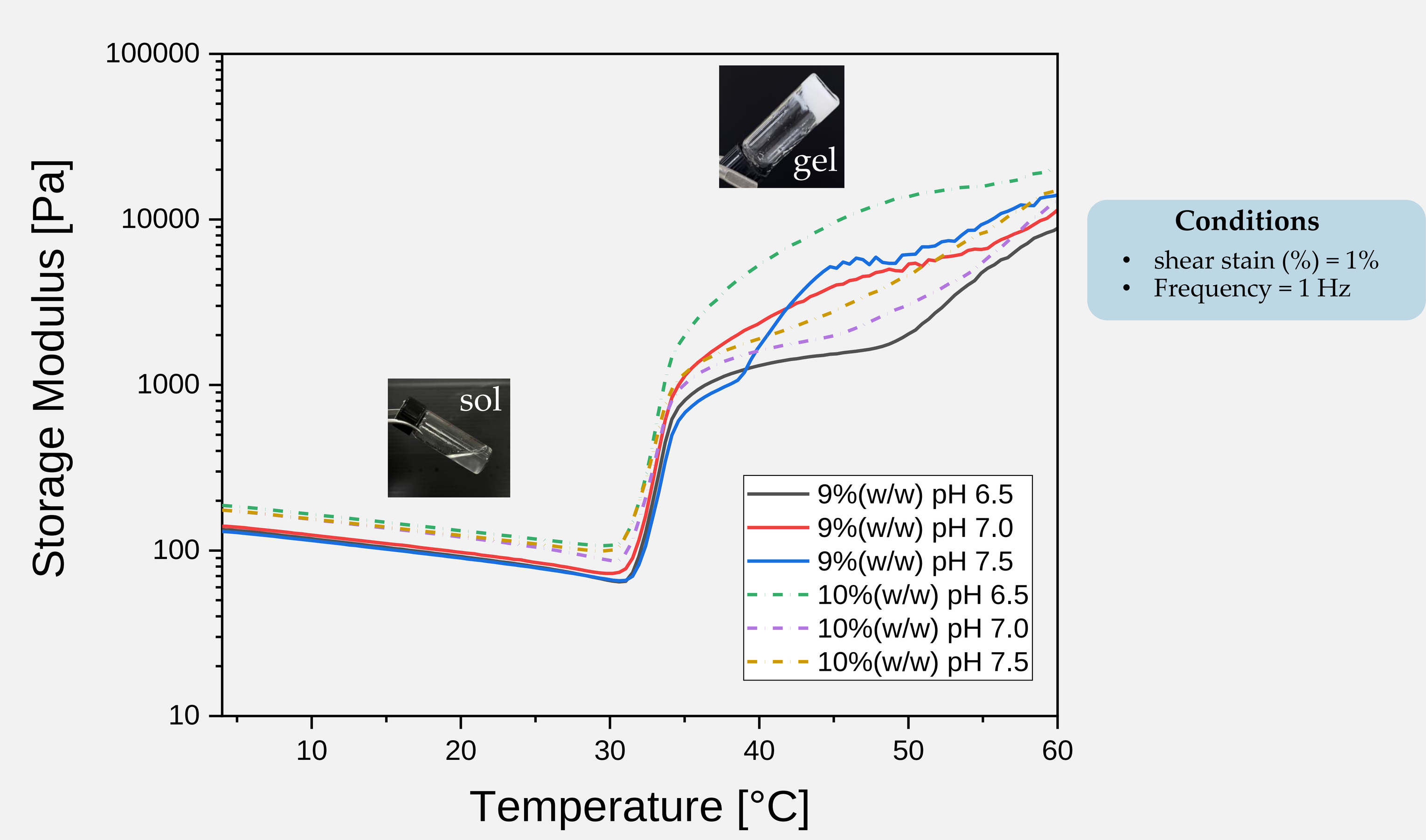


Figure 5. Thermo-responsive gelation of 9% and 10% (w/w) CMC/PNIPAM hydrogels.

ACKNOWLEDGEMENT

The author would like to thanks to Assoc. Prof. Dr. Kiattikhun Manokruang (Advisor), Dr. Wanwanut Chueasupcharoean, Mr. Nuttawut Khammata, Ms. Ain Us Saba, Department of Chemistry, Faculty of Science, Chiang Mai University and Bioplastic Production Laboratory For Medical Applications, Faculty of Science, Chiang Mai University.

CONCLUSIONS

- CMC/PNIPAM semi-IPN hydrogel was successfully synthesized using 0.125% mol of PEGDA as a crosslinker.
- When increasing polymer concentrations, polymer strength was increased. However, the phase transition was decreased with temperature.

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