

# Engineering polymer-solvent interactions for the additive manufacturing of functional polymer gels

**Sheng-Sheng Yu**<sup>1,2,3\*</sup>

<sup>1</sup>*Department of Chemical Engineering, National Cheng Kung University*

<sup>2</sup>*Core Facility Center, National Cheng Kung University*

<sup>3</sup>*Program on Smart and Sustainable Manufacturing, Academy of Innovative Semiconductor and Sustainable Manufacturing, National Cheng Kung University*

*E-mail: ssyu@mail.ncku.edu.tw*

Tough and conductive hydrogels have been intensively studied as wearable electronics to monitor the physiological activities of human bodies. However, it remains a challenge to fabricate robust polymer gels as sensors with complex 3D structures. On the other hand, additive manufacturing, also known as 3D printing, is an emerging technique to fabricate complex and customizable structures. This presentation will provide several examples of multi-functional and 3D printable gels.

First, we demonstrated a simple route to 3D printable ionogels by thermal-induced gelation of cellulose nanocrystals (CNCs) in a deep eutectic solvent (DES). Our strategy significantly reduced the concentration of CNCs needed to prepare printable inks for direct ink writing (DIW). Further photopolymerization of monomers in the composite inks led to ionogels containing multiple dynamic bonding types. The ionogels were then printed as triangular lattice structures to increase the sensitivity of wearable sensors. We also introduce resins based on DES to fabricate a pneumatically driven soft gripper using digital light processing (DLP). By utilizing the intense hydrogen bonding within DES, the resin can be rapidly cured by photopolymerization to form tough ionogels without chemical crosslinkers. The DES ionogels exhibit remarkable toughness and self-healing performance compared to common hydrogels. Furthermore, the ionogels show efficient energy-dissipating behavior and achieve rapid self-recovery. Finally, the DLP-printed soft gripper from the DES-based resin successfully actuates and heals macroscopic damages.

These results demonstrate simple and rapid strategies to fabricate stable and sensitive sensors from cheap and renewable feedstock. 3D printing further expands the design space of functional polymer gels.