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# A Multifunctional Injectable Hydrogel with Dynamic Covalent Adhesion and Bioelectronic Activity for Repair and Regeneration

Nisha Arunachalam<sup>1</sup>, Jitendra P Mata<sup>2</sup>, Jitraporn Vongsvivut<sup>3</sup>, Anita Quigley<sup>4</sup>, Naba Kumar Dutta<sup>1\*</sup>, and Namita Roy Choudhury<sup>1\*</sup>

<sup>1</sup> *Chemical and Environmental Engineering, School of Engineering, RMIT University, Melbourne, VIC 3000, Australia; [s4040569@student.rmit.edu.au](mailto:s4040569@student.rmit.edu.au) (N.A.)*

<sup>2</sup> *Australian Center for Neutron Scattering, ANSTO, Lucas Heights, NSW 2234, Australia; [jtm@ansto.gov.au](mailto:jtm@ansto.gov.au)*

<sup>3</sup> *Infrared Microspectroscopy Beamline, Australian Synchrotron, 800 Blackburn Road, Clayton, VIC 3168, Australia; [jitrapov@ansto.gov.au](mailto:jitrapov@ansto.gov.au) (J.V.)*

<sup>4</sup> *Biomedical Engineering, School of Engineering, RMIT University, Melbourne, VIC 3000, Australia;; [anita.quigley@rmit.edu.au](mailto:anita.quigley@rmit.edu.au)*

\*Corresponding author E-mail:

[naba.dutta@rmit.edu.au](mailto:naba.dutta@rmit.edu.au) (N.K.D.); [namita.choudhury@rmit.edu.au](mailto:namita.choudhury@rmit.edu.au) (N.R.C.)

## ABSTRACT

Achieving robust adhesion to wet and dynamic tissues while enabling active modulation of cell-material interactions remains a critical challenge in the design of next-generation bioadhesives. In particular, conductive hydrogels have emerged as promising platforms for interfacing with biological tissues, as electrical cues are known to influence cell adhesion, spreading, and signaling. However, most existing conductive systems lack strong interfacial adhesion under physiological conditions or fail to integrate through dynamic bonding mechanisms required for tissue compliance. Herein, we report a conductive, injectable bioadhesive hydrogel based on oxidized pectin (OP) and gelatin methacrylate (GelMA), designed to achieve robust wet tissue adhesion while enabling electrical modulation of cellular responses. Oxidized pectin introduces aldehyde functionalities that form reversible Schiff-base linkages with amine groups in GelMA, generating a dynamic, self-healing network capable of adapting to mechanically active environments. To further enhance interfacial adhesion, tannic acid is incorporated as a multifunctional polyphenolic adhesive motif, providing synergistic interactions through hydrogen bonding and interfacial complexation with tissue surfaces.

To impart electrical functionality, 2D materials are integrated within the hydrogel matrix, forming a conductive percolation network while preserving injectability and mechanical integrity. The resulting system exhibits shear-thinning behavior, enabling minimally invasive delivery, followed by rapid stabilization through secondary photo-crosslinking of GelMA. This dual crosslinking strategy allows independent tuning of adhesion strength and bulk mechanical properties. The internal network structure was probed using Small-Angle Neutron Scattering (SANS) and Synchrotron Macro ATR-FTIR Microspectroscopy, revealing nanoscale heterogeneity and insights into polymer domain organization and crosslink density, which correlate with the observed mechanical resilience and adhesive performance. The hydrogel exhibits controlled swelling and porosity, supporting nutrient transport while maintaining cohesive integrity.

Importantly, the incorporation of 2D material enables the hydrogel to function as a bioelectronic adhesive interface. In vitro studies using soft tissue cell lines demonstrate high cytocompatibility and enhanced cell adhesion, with preliminary investigations indicating that electrical stimulation can further

modulate cell spreading and activity. These findings suggest that the conductive network not only supports adhesion but also provides a platform for electrically guided regulation of cell-material interactions. Overall, this work presents a multifunctional conductive bioadhesive hydrogel that integrates dynamic covalent bonding, polyphenol-mediated adhesion, and electrical functionality. The system offers a promising strategy for developing next-generation adhesive biomaterials capable of both mechanically stabilizing tissues and actively influencing cellular behavior, with potential applications in wound healing, soft tissue repair, and bioelectronic interfaces.

#### **KEY WORDS**

*Bioadhesive, Wet adhesion, conductivity, Dynamic Covalent bonding, Injectable hydrogel, Self-healing, Cell-material interaction, Soft tissue regeneration*

#### **BIOGRAPHY**

Nisha Arunachalam is a researcher at RMIT University, working in the field of biomaterials and bioadhesives. Her research focuses on the development of protein-polymer systems, particularly modular proteins and modified polysaccharides, for advanced biomedical applications. She has experience in designing and characterizing hydrogel-based materials, with an emphasis on understanding their structure-property relationships. Nisha is also skilled in cell-based studies, enabling evaluation of material-cell interactions for translational outcomes. Her work aims to develop innovative, functional bioadhesives for healthcare applications, bridging material science and biology.

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