ABSTRACT

Hydrogels have existed for more than 60 years and fascinate researchers for their wide applications in material and biomedical fields of research. It is a three-dimensional crosslinked network of polymeric chains comprised of hydrophilic functional groups which allows the swelling of the hydrogel to a great extent. Though hydrogels swell in water, they maintain their structure due to the chemical and physical cross-linking of individual polymeric chains. Due to the significant water content in the hydrogel, they possess a degree of flexibility very similar to that of natural tissue. Hydrogels are found to have a vast array of applications in various fields such as in biomedical areas, wastewater remediation, actuators, hygiene products, etc. The tunable property of the hydrogel allows researchers to tune hydrogel with various properties such as self-healing, adhesive, photocatalytic, and electroactive properties. In addition, the ability of the hydrogel to respond to various stimuli such as pH, temperature, light, electric field, etc. makes it a versatile material in different areas and termed as 'smart hydrogel.' These smart hydrogels, when embedded with specific properties such as adhesive or photocatalytic are highly advantageous.

In recent years, the applications of hydrogel to wet or underwater surfaces have gained significant attention as it is one of the most challenging properties of an adhesive. Therefore, the fabrication of a hydrogel-based adhesive is one of the key hurdles in achieving tough adhesion between hydrogels and wet surfaces. The present thesis deals with the synthesis, characterization, and plausible application as wet adhesives and pH-responsive drug release have been investigated. In addition, this thesis also deals with the development of photocatalytic hydrogel by photopolymerization under visible light and its applications for the removal of organic dyes through the synergistic effect of adsorption and photopolymerization has also been explored. The dye removal at the varied composition of photocatalysts, different pH along with the recyclability of the photocatalytic material has also been investigated.

Moreover, this thesis also highlights the importance of electric-field responsive hydrogels in various fields. Incorporating conductive properties into a hydrogel allows it to respond to electric-field stimuli by changing the swelling behavior and correspondingly shows bending actuation. The bending actuation of the hydrogel has been determined by changing different parameters. The mechanical strength of the synthesized hydrogels has also been investigated by incorporating a reinforcing agent

into the hydrogel matrix. The whole thesis is compiled into 6 chapters and each chapter is summarized below.

Chapter 1: A general introduction to hydrogel, its properties, and applications in various fields

This chapter describes the motivation behind the present research work. It deals with the general introduction of hydrogel material, and its classification followed by synthetic procedures. The classification based on stimuli-responsive properties is well defined in this chapter. The chapter emphasizes the properties of hydrogel and highlights their importance as adhesives, photocatalysts, and electroactive materials. It also briefly outlines the applications of adhesive, photocatalytic, and electroactive hydrogel in various fields. The last part of the chapter describes the objectives and plan of the current investigation.

Chapter 2: Development of mussel mimetic gelatin-based adhesive hydrogel for wet surfaces with self-healing and reversible properties

In this chapter, gelatin-based adhesive hydrogel has been developed through a facile synthetic method. Gelatin, being a naturally derived biomacromolecule shows good biocompatibility and biodegradability and hence becomes a potential biomaterial in synthesizing adhesive hydrogel. However, to achieve significant adhesive strength under wet conditions and good mechanical properties, gelatin is functionalized with dopamine and acrylic acid. Here, inspired by nature, we have developed a gelatin-based adhesive hydrogel for wet surfaces by incorporating dopamine into a gelatin-poly(acrylic acid) (gel-PAAc) chain. The synthesized hydrogel demonstrates good mechanical strength, high stretchability, reversibility, self-healing, and dynamic adhesive behavior along with long-term reusability. The adhesive strength of the synthesized hydrogel to the tissue surface was 6.5 KPa when applied under submerged conditions. Moreover, the swelling behavior of the hydrogel reveals that hydrogel has limited swelling ability thereby retaining adhesive properties under a fully swollen state. Hemolysis results reveal the biocompatible nature of hydrogel. Thus, this hydrogel emerged as a promising bioadhesive for applications in various fields, mostly in biomedical devices.

Chapter 3: Mussel-inspired adhesive hydrogel patch for wet surfaces with self-healing and pH-dependent drug delivery for potential transdermal applications

This chapter describes the development of an adhesive hydrogel which is applied as an adhesive for underwater surfaces and as a drug delivery patch. Adhesive hydrogels have attracted significant attention in many areas, including biomedical fields where they have been used as sealants, therapeutic agents for wound healing, or drug delivery systems. However, most hydrogels lack adequate mechanical strength and have poor adhesion to moist or wet surfaces. Here, we have developed a simple route to synthesize dopa-g-Gel/AAc/AAm hydrogel as an adhesive biomaterial with multifunctional properties. The developed hydrogel shows a tensile strength of 5.38 MPa and the high flexibility of the hydrogel is also determined by its repeatable stretching behavior. The hydrogel shows repeatable adhesion to different hydrophilic and hydrophobic surfaces including tissue surfaces under dry and submerged conditions. Notably, the hydrogel also demonstrates stimuli-free self-healing properties. In addition, the pH-responsive swelling behavior of the hydrogel allows the controlled release of the drug molecule. It was found that, within 8 hours, the hydrogel showed a drug release of 60% at pH 5.8, and at pH 7.4, it showed a release of 96%. Again, hemolysis results reveal that hydrogel is biocompatible. Therefore, the above findings emphasize its potential application as a bioadhesive in numerous biomedical areas.

Chapter 4: Robust and highly compressible polyacrylamide co-polymer hydrogel developed through g-C_3N_4 initiated photopolymerisation and its photocatalytic activity towards dye removal

This chapter demonstrates the development of a photocatalytic hydrogel and explores the synergistic effect of adsorption and photodegradation of the hydrogel for removing organic dyes. Photocatalytic hydrogels have drawn significant interest in many research fields including energy conversion and wastewater treatment. However, many synthesized hydrogels show mechanical brittleness which limits their practical application. Here, we have developed a simple, cost-effective, and eco-friendly synthesis method for the photopolymerization of g-C_3N_4/polyacrylamide co-polymer hydrogel under visible light irradiation. The developed hydrogel exhibits high compressibility up to a strain of 230% with a mechanical strength of 0.55 MPa. We have addressed the dual functions of g-C_3N_4 in hydrogel. First, as a photoinitiator and reinforcing agent in the polymerization of a photocatalytic hydrogel. Second, as a photocatalyst embedded in the hydrogel for the degradation of organic dye pollutants. The three-dimensional network of the hydrogel allows rapid permeation of the cationic dye molecules into the hydrogel

matrix thereby accelerating the photodegradation process. The applicability of the synthesized photocatalytic hydrogel towards the selective degradation of different cationic dyes over anionic dyes has been determined. In addition, the synthesized hydrogel demonstrates recyclability for up to 5 cycles, without losing the photocatalytic property. In addition, the hydrogel shows adsorption maximum of 72 mg/g towards CV dye. Thus, the simple and easy preparation of the g-C_3N_4-based hydrogel and its excellent photocatalytic and adsorption properties have proved to be a potential photocatalytic material for green and sustainable water remediation techniques.

Chapter 5: Design of Starch-Based Electro-Responsive Smart Hydrogel with Rapid Bending Actuation Under Low-Electric Field

This chapter combines conductive polyaniline (PANI) with a polyelectrolyte-based hydrogel to synthesize an electric-field responsive hydrogel with improved electrical properties. Remote actuation, easy modulation, and biocompatibility of electric-field responsive hydrogels gained them widespread attention. However, the application of such hydrogel is often limited due to low mechanical strength and the requirement of high voltage for its operation. Here, we have developed g-C_3N_4 incorporated (starch/AMPS/AAc-PANI) hydrogel. The hydrogel shows a maximum tensile strength of 1.02 MPa and a maximum compressive stress of 14.79 KP with repeatable compressibility up to 20 cycles. The improved mechanical strength of the synthesized hydrogel can be attributed to the presence of g-C_3N_4 within the hydrogel and can be tuned by varying its composition. The maximum bending actuation by the hydrogel reached 110° under a low electric field of 10V. The hydrogel also exhibits repeatable actuation on a cyclically applying electric field. Moreover, the rate of bending actuation depends on the thickness and electric field strength and hence can be easily modulated by varying its content. Therefore, the developed hydrogel with rapid and controllable bending actuation has the potential for various actuators and soft robotics applications.

Chapter 6: Conclusion and Future

The last and final chapter of the thesis comprises the chapter-wise concluding remarks and future scope of the research work. The primary goal of the studied research work is to explore the applications of hydrogels in different fields. Our studies reveal that the developed hydrogels exhibit improved properties in the respective fields with better

mechanical strength highlighting their potential applications in environmental remediation, stimuli-driven actuators, and other biomedical applications.