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Characterization of GG/GO hybrid hydrogel for strain sensor application

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Abstract: Hydrogel materials are 3D polymeric materials that have a wide range of applications. Strain-sensor applications, one of the application areas of hydrogels, continue to attract the attention of researchers. In this study, gellan gum-graphene oxide (GG/GO) hybrid hydrogels were synthesized for strain-sensor application. FTIR, XRD, and SEM measurements and strain sensor application analyses of the synthesized hydrogels were performed. It has been observed that the GG/GO hybrid hydrogels obtained as a result of the findings are promising for strain-sensor applications.

Keywords: Strain-sensor; hybrid hydrogel; 3D polymer.

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1 Introduction

The research of flexible and wearable sensors, which fall under the class of smart devices, in the fields of electronic skin for human motion detection and health monitoring has continued exponentially in recent years (Afsarimanesh et al. 2020). Sensors with flexibility, which is one of the main features of new generation wearable electronics, can convert mechanical deformation into repeatable electrical signals due to changes in resistance under external force (Niu et al. 2020). Various attempts have been made, such as metal films, metal-polymer composite structures, metal films on flexible substrates, and nanoscale metal films, to produce wearable sensors that have this feature and can monitor user-related parameters. However, the semiconductor materials used in these devices have the disadvantage that they are brittle and allow only small strains (Liu et al. 2017; Wang et al. 2021; Wang et al. 2019).

Hydrogels are three-dimensional (3D) microstructured polymeric networks with high flexibility and hydration (Kailasa et al. 2022). Conductive hydrogels, which have inherent flexibility and biocompatibility, have received great attention in recent years as soft conductors for wearable strain sensors applications (Rahmani and Shojaei 2021). Studies of conductive hydrogels in high sensitivity, short response time, and multiple recycling applications are available in the literature (Sun et al. 2019).

Conductive hydrogels refer to a class of hydrogels that contain conductive electronic networks composed of conductive nanomaterials or conductive polymers (Zhang et al. 2019). Some issues need attention and continue to be improved in conductive hydrogel applications. E.g., in motion sensor applications, viscoelastic materials generate unwanted noise due to their electrical response to external force, usually due to dynamic changes caused by viscous deformation of conductive networks. Besides, the poor interface between hydrogel matrices and conductive fillers causes an irreversible movement or rearrangement of conductive networks, often irreversibly during deformation. This results in poor long-term reliability and a low signal-to-noise ratio as a result. For this reason, increasing the sensitivity of hydrogels used for motion sensors remains a topic of the current literature (Tang et al. 2020; Xu et al. 2019) .

In this study, gellan gum-graphene oxide (GG/GO) hydrogels with different contents were prepared as conductive hydrogels. In the analysis of strain sensor application, the sensitivities were compared after their responses to wrist joint movements, and the characterization of the hydrogel samples was carried out.

2 Materials and Methods

GG was dissolved in 2.5% (w/v) deionized water under constant stirring for 15 minutes at 85 °C. To prepare GG/$GO_{1.0}$, GG/$GO_{2.0}$ and GG/$GO_{4.0}$ hydrogels, it is prepared by mixing with GO at 85°C for 15 min, when used at a rate of 1.0, 2.0, and 4.0 wt. %, at different rates. The prepared solutions were was poured into molds and then cooled to room temperature to obtain three-dimensional GG/$GO_{1.0}$, GG/$GO_{2.0,}$ and GG/$GO_{4.0}$ hydrogels.

The resistance changes of the strain sensor under strain were analyzed using a multimeter (Keithley 2612A Source Meter). Produced sensors were attached to the wrist joint skin using double-sided tape to detect wrist movements. Copper wires were used to connect the sensor attached to the wrist joint. The change in relative resistance was calculated using the Eq. (1):

$$\Delta R/R_0 = (R - R_0)/R_0 \quad (1)$$

where R_0 and R denote the resistance of the original state and the resistance of real-time, respectively.

Hydrogel samples were analyzed using FTIR (Spectrum Two Perkin-Elmer Co.) in the wavelength range of 4000 cm^{-1}-400 cm^{-1} by scanning at 4 cm^{-1} resolution. Morphological studies were observed using scanning electron microscopy (SEM, JEOL, JMS 6060). The hydrogels, which were previously lyophilized and cut into suitable pieces, were fixed on conductive carbon bands and then covered with a thin layer of gold to ensure conductivity (Polaron CS7620). For the phase characterization of the synthesized hydrogels, analysis was carried out between 5° and 90° using Cu Ka radiation (λ= 0.15406 nm) operating at 40 kV, 30 mA, and X-ray diffractometry (XRD, D/Max 2200 LV).

3 Results and Discussion

After mounting a hydrogel sensor on a volunteer's wrist, the 90° flexing wrist stretched the gel and produced reproducible $\Delta R/R_0$ of 30%, 37%, and 34% for hydrogels GG/$GO_{1.0}$, GG/$GO_{2.0,}$ and GG/$GO_{4.0}$, respectively (Fig. 1). The obtained results demonstrate the great potentials of conductive hydrogels fabricated for flexible wearable devices. The sensitivity of the hydrogel sensors increased up to GG/$GO_{2.0}$ and decreased at GG/$GO_{4.0}$. The slight decrease in $\Delta R/R_0$ when the concentration of the additive material reaches 4% by weight can be attributed to the high density of the additive in the GG polymeric networks, making them difficult to replace when stretched. Therefore, GG/$GO_{2.0}$ as the optimum selected sample was used for subsequent characterizations and named GG/GO.

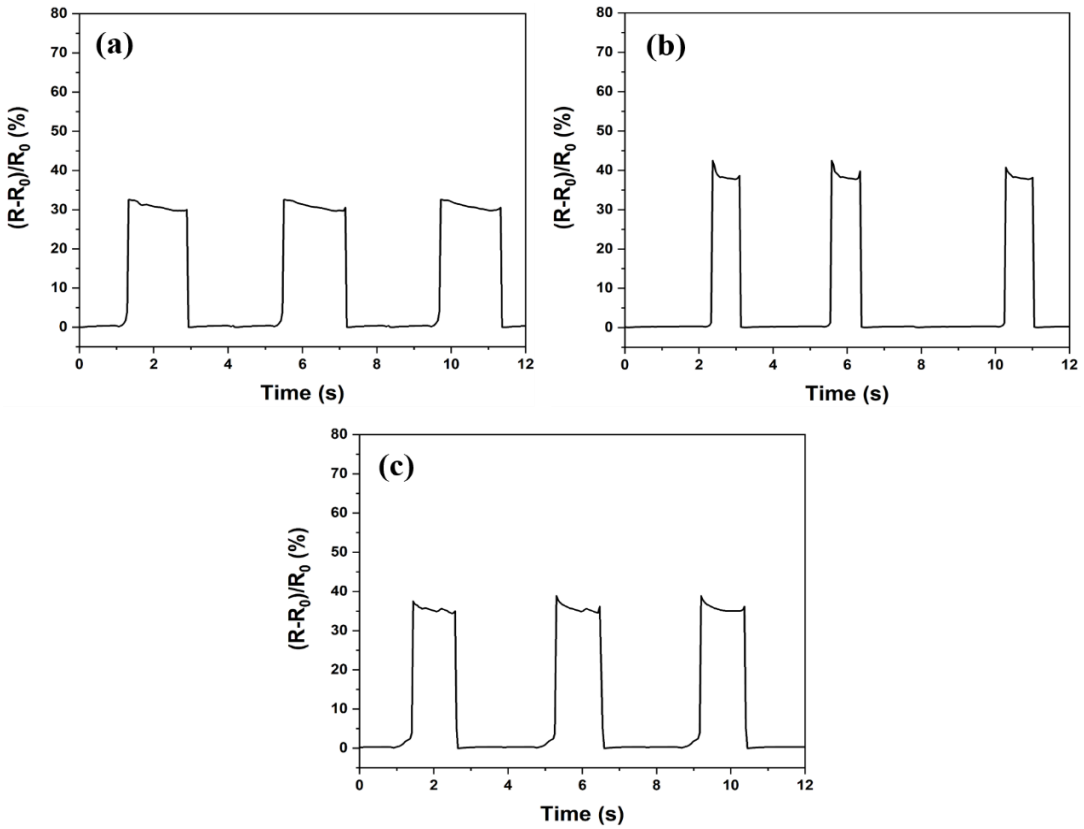

Fig. 1 The real-time detection of human motions when the (a) GG/$GO_{1.0}$, (b) GG/$GO_{2.0}$, and (c) GG/$GO_{4.0}$ hydrogel strain sensors directly adhered to the wrist joint.

Section morphologies of the lyophilized hydrogels were examined using SEM, as shown in Fig. 2. GG-glycerol hydrogel tends to form a homogeneous structure with smaller pores and tighter networks. No obvious agglomerated structure was observed in the hydrogel sample, in which an interconnected porous network was observed.

The obtained hydrogels were also analyzed by FTIR and XRD, as shown in Fig. 3. For the FTIR spectrum of GG (Fig. 3a), the broad band at 3500−3000 cm^{-1} was contributed to the stretching vibration of OH^- groups, the characteristic peak at 2925 cm^{-1} was attributed to the stretching vibration of C–H_2 groups. The characteristic peaks at 1611 and 1415 cm^{-1} were ascribed to the asymmetric and symmetric stretching vibration of carboxyl groups that existed in the salt form, and 1032 cm^{-1} was attributed to the CO^- stretching (Lu et al. 2019; Wang et al. 2021). When looking at the FTIR spectrum of GG/GO, it is observed that there is no significant difference. Fig. 3b displays the X-ray diffraction pattern of GG and GG/GO hydrogels. GG showed a broad peak at approximately 2θ = 20°, indicating their amorphous nature with lower crystallinity. A similar amorphous structure is seen when looking at the FTIR analysis of the GG/GO hydrogel sample.

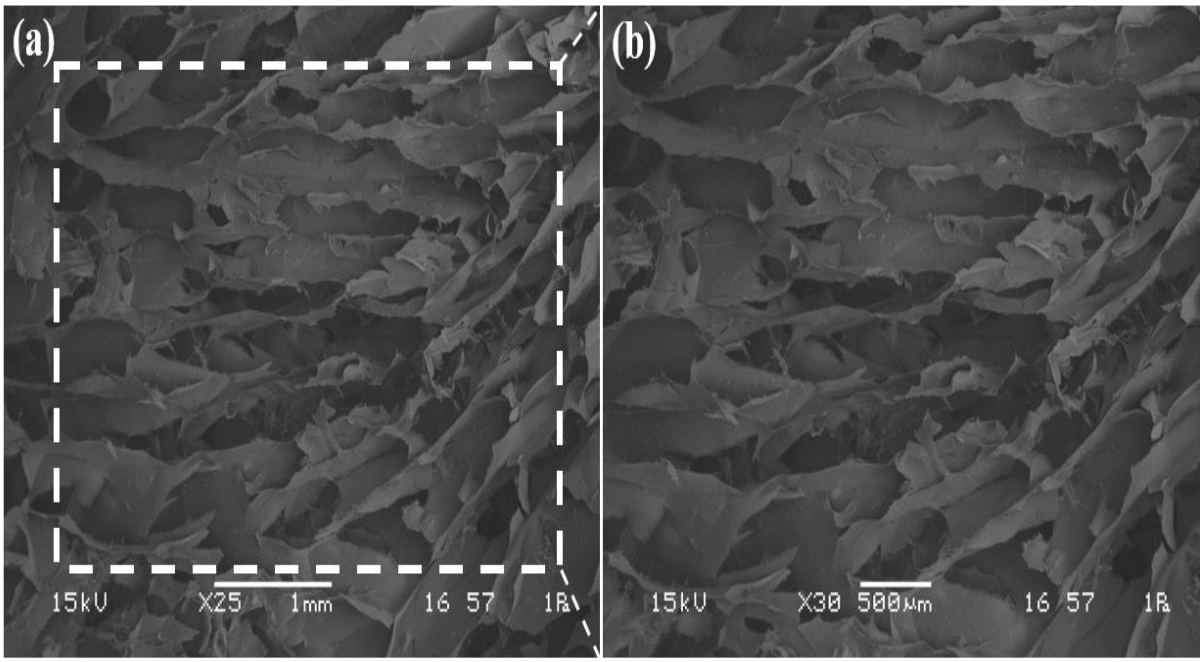

Fig. 2 (a) Cross-section SEM image, and (b) magnified cross-section SEM image of the lyophilized GG/GO hydrogel.

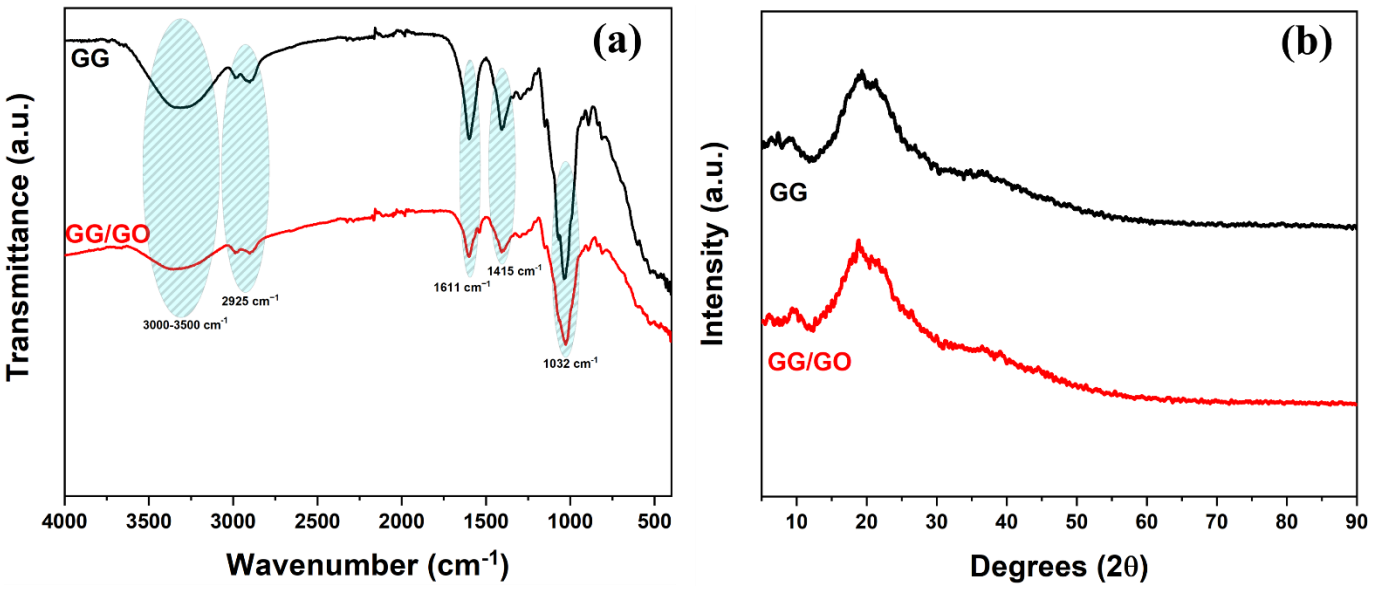

Fig. 3 (a) FTIR spectra, and (b) XRD pattern of the GG and GG/GO hydrogels.

4 Conclusion

GG based hydrogels with different GO content were prepared and measured as a wrist joint movement sensor. As a result of the study, it was concluded that the sensitivity of the motion sensor feature of the GG-based hydrogel was adjusted by changing the additive ratio in its content. In addition, the promise of the prepared hydrogels as a wearable motion sensor was observed in this study. Since it is expected that hydrogel strain sensors will take place with increasing interest in future studies, the susceptibility of the obtained hydrogel to movements and vibrations such as heartbeat, speech can be examined and adhesion tests can be performed.

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