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Characteristics of Sodium Polyacrylate/Nano-Sized Carbon Hydrogel for Biomedical Patch

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Conductive hydrogels were prepared for biomedical patch in order to improve the electrical conductivity. Sodium polyacrylate and nano-sized carbon were mixed and fabricated by aqueous solution gelation process in various contents of nano-sized carbon with 0.1, 0.5, 1.0 and 2.0 wt%. Sodium polyacrylate/nano-sized carbon conductive hydrogels were investigated by molecular structure, surface morphology and electrical conductivity. The conductivity of the hydrogel/nano-sized carbon conductive hydrogel proved to be 10% higher than conductive hydrogel without nano-sized carbon. However, it was founded that conductive hydrogels with nano-sized carbon content from 0.5 up to 2.0 wt% were remarkably decreased. This may be due to the non-uniform distribution of nano-sized carbon, resulting from agglomerates of nano-sized carbon. The developed hydrogel is intended for use in the medical and cosmetic fields that is applicable to supply micro-current from device to human body.

Keywords: Sodium Polyacrylate, Nano-Sized Carbon, Conductive Hydrogel, Electrical Conductivity, Bio-Patch.

1. INTRODUCTION

Conductive hydrogels represent a distinctive class of materials that synergize the electrically advantageous features of hydrogels.[1,2] The conductive hydrogels have enormous potential to provide a more conductive environment for cell integration.[3] Thus, the applicability of the conductive hydrogels is across a range of biomedical applications such as biosensor, biological patch, and drug delivery system.[2,4]

The common approach to improve the electrical conductivity of hydrogel is either adding conductive particles to gel matrices or synthesizing directly from conjugated polymers.[5] According to the latest research paper, it is normally accepted that the former approach seems to be more appropriate than the latter that, because the latter approach is limited by the inherent rigidity of the conjugated macromolecular chains.[5]

Meanwhile, the use of polymer with carbon materials such as graphite,[6,7] carbon fiber,[8,9] carbon black,[10,11] and carbon nanotube[12] could be a way to develop a superior conductive hydrogel, since that satisfies not only high conductivity but also excellent biocompatibility.[3] However, despite its excellent performance, the complicated manufacturing process, which include free radical polymerization and electro-depositing, remain a limitation in expanding the scope of use. Moreover, although a number of previous studies have attempted to provide information on the conductive hydrogels by considering carbon materials, the effects of nano-sized carbon particles have not been fully elucidated.[13,14]

Therefore, the present study performed a comparative analysis to evaluate the effects of different contents of nano-sized carbon particles in sodium polyacrylate while adopting a facile manufacturing process, which is adding the particles to gel matrices. Moreover, an application of the developed conductive hydrogel in the biomedical fields would be introduced to further extend our knowledge.

2. EXPERIMENTAL DETAILS

2.1. Materials

Nano-sized carbon in powder form (TOKABLACK, #500, −25 nm in diameter) was provided by Tokai carbon Co., Ltd. Sodium polyacrylate, EDTA, Glycerin, Polysorbate 20, Aluminum Stearate, Sodium Chloride and Tartaric

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acid purchased from Sigma-Aldrich. All other reagents were analytical grade and used as received.

2.2. Fabrication of Conductive Hydrogels

Conductive hydrogels composed of sodium polyacrylate/Nano-sized carbon were fabricated by dissolving in a beaker of 500 ml. Polysorbate of 0.1–0.1 g, phenoxyehanol of 0.2–0.5 g and EDTA of 0.1–0.4 g were put together into the beaker and stirred at 300 rpm for 30 min. Glycerin of 25 g, methyl paraben of 0.15 g, sodium chloride of 0.15–0.3 g and tartaric acid of 0.25 g were dissolved in distill water. The resultant solution was then heated at 80 °C for 1 h. On completion of the reaction, the color of specimens was converted into a dark black hydrogel. Sodium polyacrylate/nano-sized carbon hydrogel was prepared.

2.3. Characterizations

The molecular structure of conductive hydrogels was analyzed using a fourier transform infrared vacuum spectroscopy (FT-IR, VERTEX 80V) in KBr at room temperature in the region 4,000–500 cm^{-1}. The surface morphology and chemical composition of the conductive hydrogels was observed using a field emission scanning electron microscopy (FE-SEM, Hitachi, S-4200) equipped with an energy-dispersive X-ray spectroscopy (EDXS). X-ray diffraction (XRD, X'Pert-RRO MRD, Phillips) were recorded with Cu Kα radiation in a 2θ a scanning range from 10 to 80°. The electrical conductivities of conductive hydrogels were measured by inserting an electrical conductivity sourcemeter (Keithley2425) in a cylinder containing each of specimens of 5 g at room temperature. Platinum probe with tip diameter 2.0 μm were used for two probe electrical conductivity analysis. The conductive hydrogels were calculated using following relation:[15]

$$\rho = \left(\frac{V}{I}\right) 2\pi s \tag{1}$$

where ρ the resistivity of the specimens, V is the applied voltage, I is the measured current through the specimens, and S is the distance between electrodes.

3. RESULTS AND DISCUSSION

3.1. Influence of Cross-Linker Concentration in the Conductive Hydrogel

Figure 1 shows the FT-IR spectrum of the conductive hydrogels under different weight percent of nano-sized carbon particles to analyze the molecular structure according to the nano-sized carbon particles ((a) without nano-sized carbon, (b) 0.1 wt%, (c) 0.5 wt%, (d) 1.0 wt% and (e) 2.0 wt% nano-sized carbon, respectively). From the result, the peak values arose at certain points of wavelength at 3,449, 2,958, 1,564, and 1,046 cm^{-1} regardless of the ratio of nano-sized carbon. The values of 3,449 and 2,958 cm^{-1} were assigned the O-H stretching vibration in carboxylic groups, and the value of 1,564 cm^{-1}

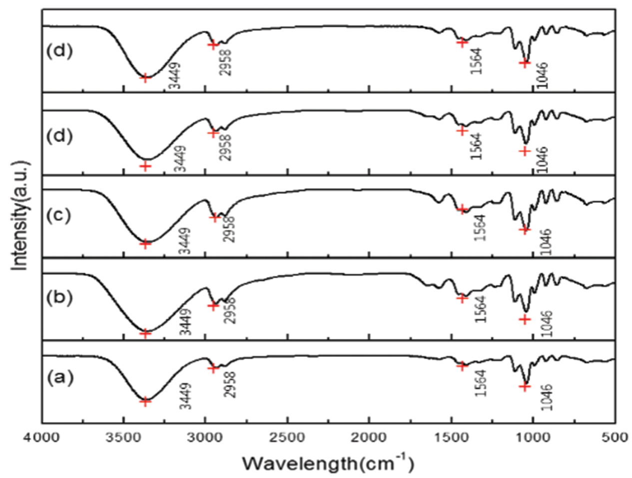

Figure 1. FT-IR spectrum of the conductive hydrogels under different weight percent of nano-sized carbon; (a) without nano-sized carbon, (b) 0.1 wt%, (c) 0.5 wt%, (d) 1.0 wt% and (e) 2.0 wt% nano-sized carbon, respectively.

was attributed to the –COO bending vibration. Compared with our expectation, the results imply that electrical conductivity in conductive hydrogels is not determined by the molecular structure and the structure of the polymer network.

Based on the result, we can estimate that the conductive hydrogels combined with sodium polyacrylate and nano-sized carbon may be formed by absorption onto a polymer network by physical or chemical combination. In here, the entire system is in the form of a nano-sized carbon particle structure adsorbed on the polymer network, and the conduction channels are formed in such a way that the carbon particles are in contact with each other.

3.2. Morphology of Nano-Sized Carbon Amount in Conductive Conductive Hydrogel

Figure 2 shows the the morphological differences according to different ratio of nano-sized carbon weight percent ((a) without nano-sized carbon, (b) 0.1 wt%, (c) 0.5 wt%, (d) 1.0 wt% and (e) 2.0 wt% nano-sized carbon, respectively). In Figure 2(a), which is the case of without nano-sized carbon, the extent of roughness on the surface seems to be weak and uniform. However, as the amount of the nano-sized carbon increases, the surface roughness was also increased proportionally. This phenomenon may be due to the enhanced aggregation of carbon particles.

In addition, EDXS analysis was performed to analyze the chemical composition according to the proportion of nano-sized carbon particles. The Table I showed that the main components consist of such elements as carbon, Oxygen, Sodium Nitrogen. It was confirmed that carbon content is considerably increased by adding nano-sized carbon.

As a result, the increased amount of the nano-sized carbon particles is expected to enhance the possibility

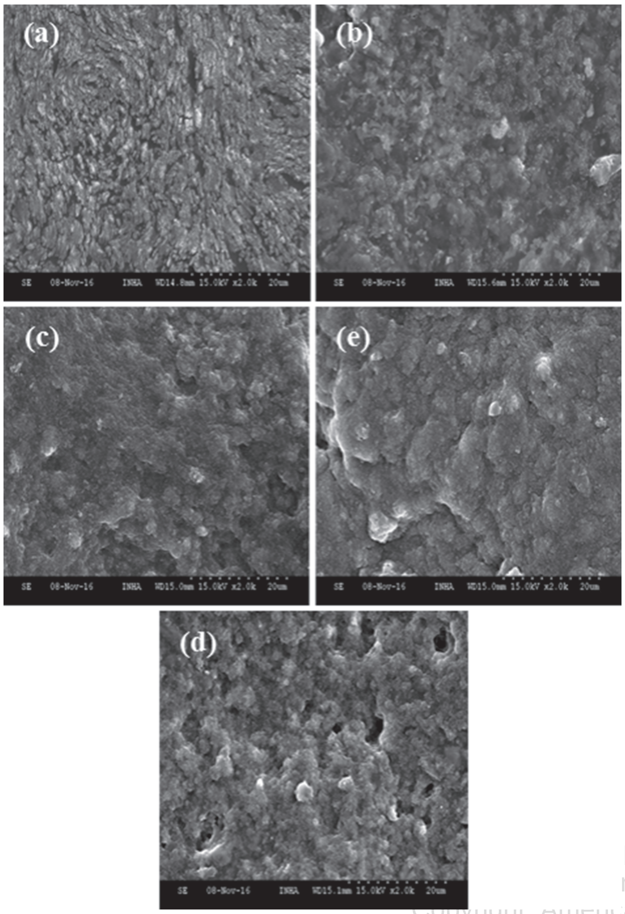

Figure 2. Microscopic images to analyze the morphological differences under different weight percent of nano-sized carbon; (a) without nano-sized carbon, (b) 0.1 wt%, (c) 0.5 wt%, (d) 1.0 wt% and (e) 2.0 wt% nano-sized carbon, respectively.

of agglomeration of nano-sized carbon in the conductive hydrogel, which may affect the surface roughness and the electrical conductivity as well.

3.3. Influence of Nano-Sized Carbon Amount on the Electrical Conductivity of Conductive Hydrogel

Figure 3 shows the electrical conductivity according to different contents of nano-sized carbon in the conductive hydrogels. The electrical conductivity in the case of 0.1 wt% was increased to 5.23 mS/cm compared with that of nano-sized carbon (4.76 mS/cm). Interestingly, the

Table I. The results of EDXS analysis for nano-sized carbon amount in conductive hydrogel.

Specimen (nano-sized carbon)	Element			
	C (wt.%)	O (wt.%)	Na (wt.%)	N (wt.%)
–	26.32	68.84	4.16	0.68
0.1 wt.%	35.52	61.22	2.93	1.84
0.5 wt.%	45.32	41.32	12.52	1.03
1.0 wt.%	45.32	29.30	13.49	1.28
2.0 wt.%	65.38	26.71	7.12	0.79

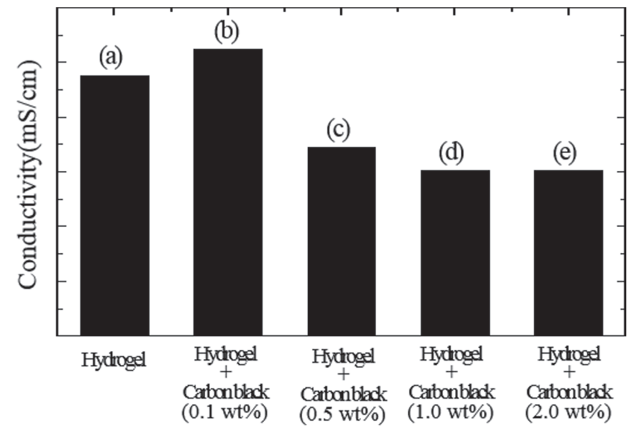

Figure 3. The conductivity of the electrical conductive hydrogel with various nano-sized carbon weight percent; (a) without nano-sized carbon, (b) 0.1 wt%, (c) 0.5 wt%, (d)1.0 wt% and (e) 2.0 wt% nano-sized carbon.

electrical conductivity was decreased when the specific content of nano-sized carbon (the case of 0.1 wt%) was exceeded, and the values of hydrogel with nano-sized carbon are observed to be lower than that of hydrogel without nano-sized carbon.

From the results, we can estimate that the addition of nano-sized carbon may induce the enhancement of electrical conductivity; however, this phenomenon is expected to occur only in the specific extent of contents. From now, we should develop and improve the homogenous distribution process of nano-sized carbon in hydrogel matrix for enhanced electrical conductivity of hydrogel.

3.4. Application of the Conductive Hydrogel with Micro-Current Stimulating Device

A significant application of high conductive hydrogel is micro-electrical current therapy (MET). Since MET uses extremely low-level electrical currents to treat various health challenges such as inflammation, muscle pain, and skin repair, the electrical conductivity of contact terminal (generally used the patch type of hydrogel) from device to human body is most important.

We performed clinical test to evaluate the effectiveness on the skin care by adopting the initially developed hydrogel patches. From the test, we can concluded that MET is effective on the enhancement of skin qualities.[16] This positive effect may be due to a kind of cellular (skin) exercise stimulated by external micro-current. In here, the most important concept is that the conductive hydrogel should be designed to have the ability of exact and high transmission. Our current research would be widely used to improve the accuracy of that kind of applications in the future.

4. CONCLUSION

We fabricated the Sodium polyacrylate/nano-sized carbon for the electrical conductivity hydrogel via the aqueous gelation process. For the chemical bonding in the five different specimens, FT-IR analysis was performed from 500 cm^{-1} to 4000 cm^{-1} even though the structural properties were somewhat same. Based on both SEM and EDXS, as the nano-sized carbon constituents into the specimens increased, the surface roughness is gradually increased due to the agglomeration of each particle. Under 0.1 wt% carbon condition, the electrical conductivity of nano-sized carbon indicated the highest value (5.23 mS/cm) compared with the other samples. This concept using controllable elemental composition may be open up the new possibility of biomedical patch with beauty treatment although further detailed studies were required in the near future.

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