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Article

Analysis of the Performance of a Gel Actuator Made of Plasticized Polyvinyl Chloride/Carboxylated Cellulose Nanocrystals

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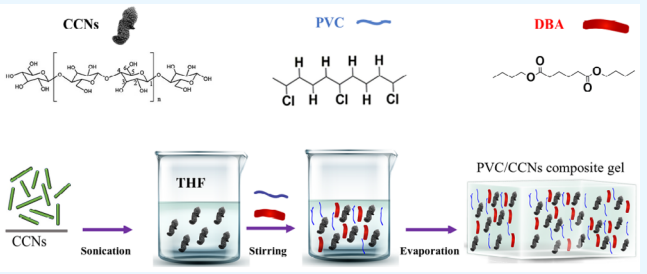
ABSTRACT: Recently, polyvinyl chloride (PVC) gel materials appeared promising for developing actuators, artificial muscles, and sensors. However, their energized response time and recovery limitations restrict their broader applications. Herein, a novel soft composite gel was prepared by mixing functionalized carboxylated cellulose nanocrystals (CCNs) and plasticized PVC. The surface morphology of the plasticized PVC/CCNs composite gel was characterized by scanning electronic microscopy (SEM). The prepared PVC/CCNs gel composites have increased polarity and electrical actuation with a fast response time. Experimental results demonstrated good response characteristics within the actuator model with a multilayer electrode structure when stimulated with a specified DC voltage (1000 V), with deformation of approximately 36.7%. Moreover, this PVC/CCNs gel has excellent tensile elongation, and the elongation at break of the PVC/CCNs gel is greater than the elongation at break of the pure PVC gel under the same thickness conditions. However, these PVC/CCNs composite gels showed excellent properties and development potential and are directed for broad applications in actuators, soft-robotics, and biomedical applications.



INTRODUCTION

Electroactive polymers (EAP), which have received much attention recently, are emerging soft functional materials that respond to specific stimuli and can produce driven deformation results when combined with specific mechanical models.[1,2] Many factors influence material properties (deformation situation, response time, energy consumption, and mechanical properties), including material density, stimulus form, and mechanical structure. EAPs are classified as electric field or ion-driven polymers on the basis of their driving method, stimulus-response mechanism, and transduction mechanism.[1]

Electronically driven EAP materials induce macroscopic mechanical deformation by the action of Coulomb forces under electric fields. Different materials also have exceptional property performance, such as sensing properties, piezoelectric properties, and high transparency. At this stage, EAP materials are widely used in many fields, such as aerospace materials, biomedical devices, and flexible optical components. Electronic EAP requires a high stimulation (high voltage) environment for driving, which is an essential factor hindering its development.[3] In contrast, ionic EAPs are based on an electrochemical reaction combined with a battery structure. This material uses chemical energy as the driving energy, combined with driving components to transform into mechanical deformation. Recently, it has been used primarily in micro soft robots as well as medical robots. Although it has the characteristics of low drive energy consumption and large deformation, the small output force is still one of the problems that needs to be solved.[4,5]

PVC gel is a soft application form of conventional PVC material.[6] The PVC gel studied in this Article is an environmentally friendly electroactive polymer produced by the solution formulation and injection molding methods using PVC resin as the substrate. The simple steps of dissolving in organic solvents, adding plasticizers to plasticize, and then drying to remove the volatile organic components result in a soft colloidal structure with mechanical strength varying with the content of plasticizers.[7,8] However, a more stable fluorinated PVC material has also been used to improve the thermal stability of the material and has been applied to a PVC gel system to enhance the performance. At the same time, the polarity of halogen atoms also increased its actuation effect.[9,10] It was found that elaborating on the relevant deformation principles with Raman tests could provide a clear characterization and interpretation of the response process.[10,11]

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As the external environment changes, smart polymer gels exhibit deformation behavior that distinguishes them from ordinary mechanical structures. For example, some polymer hydrogels change their material shape with changes in solution pH, ambient temperature, external electric field, and other factors, which produce changes in mechanical energy.[12,13] PVC gels have an excellent response to DC voltage. When DC voltage is applied to the gel surface under a reasonable driving structure, the plasticizer particles and polar molecules inside the gel migrate toward the anode under the action of their polarity. The plasticizer particles and polar molecules inside the gel help to migrate toward the anode under their polarity, and thus a creeping deformation at the surface of the anode was observed.[14−16] The low price of raw materials makes it much more economical. Many factors make PVC gels shine in the field of flexible actuators.

Surface porosity, material polarity, biocompatibility, nontoxicity, electrical conductivity, and other excellent properties have attracted close attention from researchers.[17,18] It was also observed that there is a growing demand for comprehensive research on this material. However, the low solubility of cellulose in weakly polar solvents due to its hydroxyl-rich surface and strong hydrophilic properties has hampered the field of polymeric material compounding, as evidenced by the self-agglomeration and poor solubility of the material in the solvation system.[19,20] This defect is a critical reason for limiting its application scope. However, this problem should be considered: researchers found that CNC can be treated with an oxidized surface (increased functional groups), thus improving its solubility and dispersion in the system.[21]

Furthermore, the thermodynamic compatibility is a crucial factor in polymer science that can significantly affect the characteristics and performance of polymer composite materials. For instance, previous research on a miscible homopolymer−copolymer pair, poly(ethyl methacrylate) (PEMA) and poly(styrene-*co*-butyl acrylate) (SBA), analyzed their miscibility using differential scanning calorimetry.[22] In another study, analog calorimetry was employed to investigate blends of poly(vinyl ester)s and polyacrylates, using hydrogenated monomers as their equivalents. It was found that isomeric esters exhibited only slight heat changes with positive or negative values, depending on the orientation of the COO group.[23]

In recent years, researchers have focused on the plastic modification of PVC materials. Because of the large number of C−Cl− dipole moments inside the PVC molecules, the materials have strong polarity. Inorganic nonmetallic soft materials used in electro-actuated response have been found to have a wide range of application prospects, such as biomimetic engineering, flexible electronics, and soft robots. Because of the soft PVC gel actuation, the problem is that the applied DC voltage for stimulation of PVC gel is still very high (from 200 to 1500 V), and the energy consumption is high for experimental application. To solve this problem, in this study, the highly potential carboxylate nanocellulose-loaded PVC gel is prepared by a simple solution casting method. The prepared gel with nanocellulose was analyzed and tested, and the response time and deformation at the same voltage were tested with laser displacement on the multilayer actuator. The nanocellulose has enhanced the stimulation and mechanical properties, which reduces the driving energy consumption and increases the response time.

MATERIALS AND METHODS

Materials. The materials used for the preparation of PVC/CCNs gel with CAS number and manufacturer's names are listed in Table 1.

Table 1. CCNs Preparation Material Parameters

experimental materials	CAS number	manufacturers
CNC	9004-34-6	Guilin Hongqi Technology Co.
TEMPO	2564-83-2	Changzhou Jia Na Chemical Co.
NaBr	7647-15-6	Beijing Chemical Plant
NaOH	1310-73-2	Beijing Chemical Plant
NaClO	7681-52-9	Beijing Chemical Plant

The equipment used for the preparation of the PVC/CCNs composite gel is given in Table 2.

Table 2. Parameters of Experimental Equipment for CCNs Composite Gel Preparation

test equipment	model	manufacturers
ultrasonic cell crusher	SCIENTZ-IID	Ningbo Xinzhi Biotechnology Co.
tube centrifuge	TDL-5A	Changzhou Yinneng Experimental Instrument Factory
metal conductive copper mesh	40 items	Anping County Hanging Eagle Wire Mesh Products Co.
metallic conductive copper foil	0.1 mm	Shanghai Beacon Industrial Co.

Preparation of Carboxylated Nanocellulose and PVC/CCNs Composite Gel. The preparation of carboxylated nanocellulose by the TEMPO (2,2,6,6-tetramethylpiperidine oxide) method is regarded as the most effective way to oxidize the surface of cellulose material, which is a typical piperidine-type nitroxide radical. The unique characteristics of its functional group make it have distinct selective oxidation properties, mainly reflected in the fact that when NaBr and NaClO act together, the functional group within the TEMPO molecular chain (nitroacyl) is excited, which acts as a condition for the oxidation reaction to occur, turning the hydroxyl group on the surface of CNC into an oxygen-containing carboxyl group.[24]

First, 100 g of CNC raw material solution with a mass fraction of 1% was dissolved with 0.02 g of TEMPO and 0.1 g of NaBr in a trace amount of deionized water and mixed with CNC solution. The stirring parameter of the magnetic stirrer was set to 1000 rad/min, and then NaOH solution with a concentration of 0.1 mol/L was added to adjust the pH value of the suspension at about 10.0.[25] The stirred solution was washed by centrifugation in a centrifuge. Finally, the preparation of CCNs was completed by purifying the prepared material with an appropriate amount of $AgNO_3$ to remove the chloride ions.

The mass of the mixed solution was controlled at about 100 g to ensure that the mass fraction of the prepared CCNs solution was still 1%. It has been pointed out that it is reasonable to choose PVC:DBA:THF = 1:4:12 as the mass ratio of the gel material for the multilayer membrane structure soft drive. Therefore, in this study, this mass ratio was used as the initial parameter. The initial amount of PVC should be 1.67 g, and 0.05% by mass of the CCNs solution was added to the mixed system. The nanocellulose fiber solution was introduced into tetrahydrofuran solvent for preliminary

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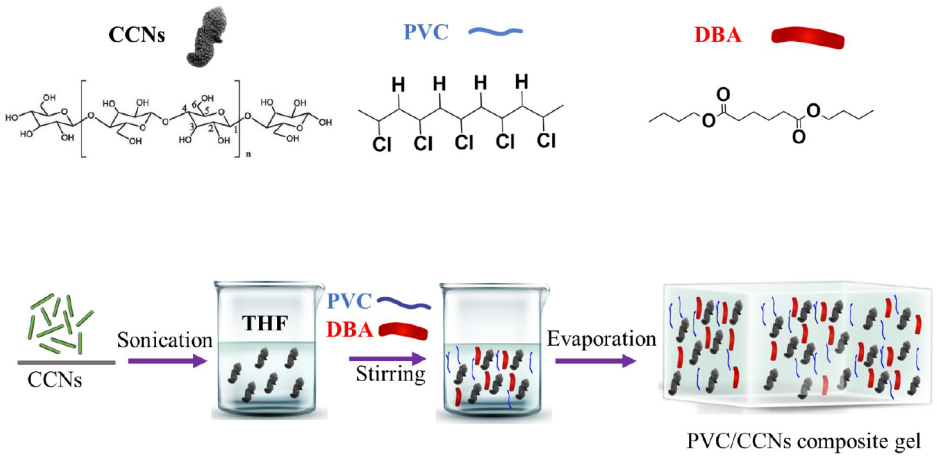

Figure 1. Preparation process of the PVC/CCNs-based gel.

dissolution. The dissolution process was carried out in an ultrasonic cell crusher with 50% output power for 10 min at 900 W power for ultrasonic dispersion. The ultrasonic gap was 5 s on and 5 s off to make the cellulose uniformly dispersed and easier to dissolve. The preparation of the composite gel was combined with the experimental steps in adding PVC resin powder, plasticizer, and CCNs at a stirring time of 12 h, stirring speed of 1000 rad/min, with the stirring temperature set to 60 °C. The prepared gel was dried at room temperature for 7 days under room temperature conditions to remove the volatile components and to prepare the nanocomposite gel required for the experiment.

In the case of PVC, its compatibility with other materials like CCNs and plasticizers, such as DBA, is of particular interest. The interaction parameter between PVC and DBA is anticipated to be positive, indicating favorable compatibility between the two materials. This results in a homogeneous blend with enhanced characteristics such as increased flexibility, softness, and lower processing temperatures.[1,26] The Flory–Huggins interaction parameter has been reported as a useful tool for understanding PVC's thermodynamic compatibility with CCNs and optimizing PVC-based composite formulations and processing. When evaluating the compatibility between PVC and the common plasticizer DBA, the Flory–Huggins interaction parameter can also be used. DBA is known to penetrate the PVC matrix and disrupt polymer–polymer interactions.[1,26] The interaction parameter between PVC and DBA can be influenced by several factors, such as DBA's molecular weight and concentration, temperature, and plasticizer–PVC ratio. Additionally, CCNs possess hydroxyl groups on their surfaces that can interact with the polar groups of PVC through hydrogen bonding, electrostatic attraction, or dipole–dipole interactions.[27,28] Figure 1 provides details on the preparation procedure of the PVC/CCNs composite gel.

Design and Construction of the Actuator Setup. The gel, insulating layer, positive metal mesh (anode), and negative copper foil (cathode) were assembled to construct the actuator setup. The multiple electrodes are coupled and stacked, a four-layer electrode stack structure is set, each electrode is set as a closed circuit, and the overall parallel structure is adopted. Finally, two wires are connected to the high-voltage DC power supply to realize electrode drive control.

In the positive and negative metal electrode selections, the positive electrode is selected from metal mesh with a thickness of 1 mm and a specification of 40 mesh, that is, 40 holes per square inch, which is converted to 6 holes per square centimeter. The most significant degree of deformation was caused by the 40 mesh metal–copper mesh due to the larger voids of the metal mesh, making it easier for the gel to creep deformation toward the anode under applied voltage. Thus, we obtained the higher deformation results of the gel. For the negative electrode material, a metallic copper foil with a thickness of 0.1 mm was chosen to reduce the effect of gravity on the driving impact. For the insulating material, a slide was selected and cut for the setup as the insulating layer material. The electrodes and insulating materials involved in the experiments were cut into square materials of length by width 20 × 20 mm, and the positive and negative materials were designed with electrode extensions to connect the wires.

The actuator assembly is placed under the laser displacement meter to measure the deformation. The iron frame table is used for the control height and the position. When voltage is not applied, it is set at the initial position to facilitate recording and measurement. The schematic of the actuator assembly is depicted in Figure 2, and the real assembled actuator is shown in Figure 3.

Testing and Characterization. The samples were cut into different shapes for testing. The gel samples used for mechanical property testing were all set up in a rectangular shape of 10 × 60 × 1 mm and selected for testing by the universal material testing machine.[21] The gel samples used for light transmission tests were all set up in the shape of a rectangle of 5 × 30 × 10 mm and selected for testing by a visible light spectrophotometer. The gel samples used for the actuator placement in the electrode assembly were set up in the shape of a disk of φ 10 mm × 1 mm.

The electrode assembly is cylindrical and consists of a positive metal inner ring with plastic insulating barrier and a negative metal outer ring, all three concentric and having the same height. The gel is laminated to the electrode surface, and a DC voltage of 1000 V is applied. The gel deformation and the response time data are observed and recorded using a laser displacement meter. The equipment used for the testing of the PVC/CCNs composite gel is shown in Table 3.

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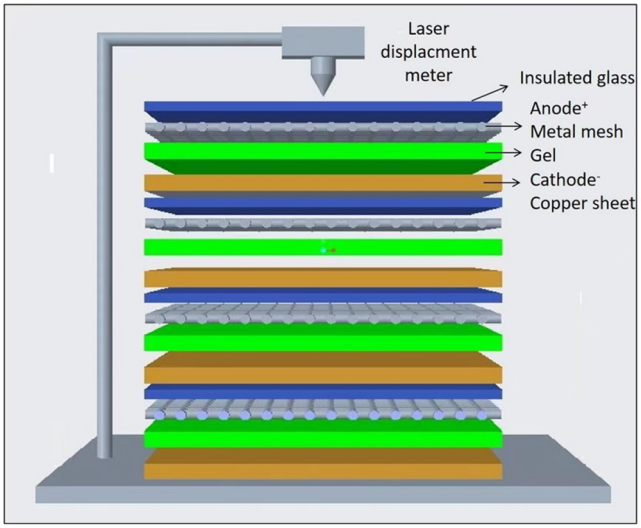

Figure 2. Schematic diagram of the driving model and measurement structure.

Figure 3. Actual driving model construction diagram.

Table 3. Testing Instruments and Parameters

universal material testing machine	9002-86-2	Dongguan Ruiyu Chemical Co.
visible spectrophotometer	F98	Shanghai Prism Technology Co.
scanning electron microscope	Phenom Pro	Fuana Scientific Instruments Co.
vacuum freeze-dryer	LC-12N-50A	Hunan Lichen Instrument Technology Co.
laser displacement collector (mm)	HC-C1030	Shanghai Industrial Control Group Co.

RESULTS AND DISCUSSION

Morphological Analysis of the Pure PVC and PVC Composite Gels. The composite gel was prepared by the simple casting process. Figure 4 shows the white agglomerates that appear on the surface of the gel. According to the literature review, it is due to the incomplete dissolution of the excess nanocellulose and the agglomeration caused by water during the reaction process. The reason for the agglomeration is the insufficient dissolution of nanocellulose. However, the drying process of the gel before the gel is formed makes cellulose. It has been dissolved into shorter fibers, which attract each other due to the introduction of fewer carboxyl groups on the surface. Hence, the internal hydrogen bonding leads to agglomeration in the prepared gel and then to the "snowflake" white agglomerated spots.

Figure 4. PVC/CCNS composite gel.

Further, we characterized the gel sample's part with agglomerated dots using SEM electron microscopy. The gel was frozen before scanning, placed in a cryostat at −20 °C for 24 h, and then dried in a vacuum freeze-dryer for 12 h to remove the effect of volatile components and water.[29] The surface was sprayed with gold, and the scanning results are shown in Figure 3. Figure 5a−c shows the micrographs of the PVC gel, PVC/CNT gel, and PVC/CCNs gel. Overall, the micrographs confirmed a uniform surface morphology of PVC gel with good dispersion of CNTs and CCNs. However, some degree of agglomerations was observed in the PVC/CNTs and CPVC/CCNs gel samples due to low the dosage of CNTs and CCNs in the PVC gel.[28] Moreover, from Figure 5b and c, we can clearly observe a significant change in the internal structure that occurred due to significant physical interactions between CNTs and PVC gel and CCNs and PVC gel. From the results, the dipole−dipole interaction and hydrogen-bond formation among PVC, DBA, and CCNs are observed. It is proposed that

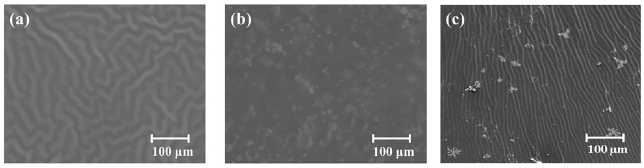

Figure 5. SEM micrographs of (a) pure PVC gel, (b) PVC/CNT, and (c) PVC/CCNs composite gels.

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the PVC, DBA, and CCNs interactions also lead to better mechanical and thermal properties.

A literature review of the available information on the dissolution agglomeration of nanocellulose revealed information for the dissolution of nanocellulose. Therefore, it is suggested that, in future work, this study will significantly improve the solubility and dispersion of nanocellulose in the gel and further improve the nanocomposite gel's mechanical and electrical stimulation response properties.

It was found that the transparency of the composite gel was less than that of the ordinary gel in the naked eye observation range. In this experiment, the ordinary gel of 20 mm × 20 mm size was intercepted and compared to the composite gel in the visible spectrophotometer. The test results in the visible wavelength range are shown in Figure 6. The reason for the

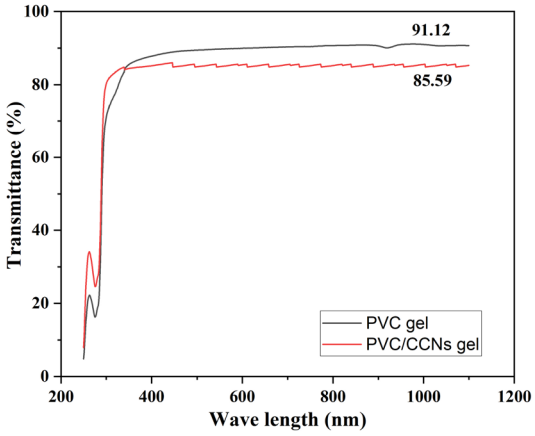

Figure 6. Transparency test of the pure PVC and PVC composite gel samples.

low transparency of the composite gel is that the addition of white nanocellulose leads to the low overall transmittance of the system. The other reason is that the white agglomerates affect the transparency of the gel.

Mechanical Properties of the Pure PVC and PVC Composite Gels. The mechanical properties of the plasticized PVC and PVC/CCNs composite gel were tested with the help of a universal testing machine. The test mainly extracted the elongation data of the material at tensile fracture. The test results are shown in Figure 7, in which the tensile force limit of the PVC/CCNs composite gel is clearly shown as a function of time. The tensile strength of the pure PVC gel and PVC/CCNs composite gel is 26.645 and 38.317 MPa, respectively. The PVC/CCNs composite gel exhibits a higher level of tensile elongation as compared to that of the pure PVC gel. The enhanced tensile properties warrant the CNNs better compatibility with the plasticized PVC and revealed that the used materials for composites gel are mixed properly.[30,31] These remarkable results also showed significant stress−strain behavior, and it is due to the physicochemical interactions among PVC, DBA, and CCNs. For the case of the gel material, the excellent ductility gives the gel more possibilities to use for actuators. For the sample T60 PVC gel, the elongation at the break reached 443%. For the PVC/CCNs composite gel, the maximum elongation was observed at about 697%. It showed better elongation of the same proportion composition than the T60 PVC gel. It was noted that it has considerable

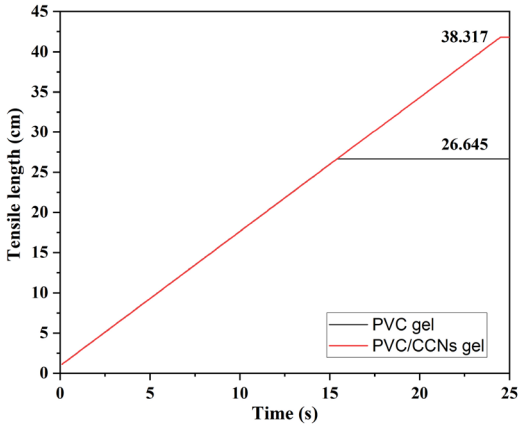

Figure 7. Comparison of tensile elongation of the pure PVC and PVC composite gel samples.

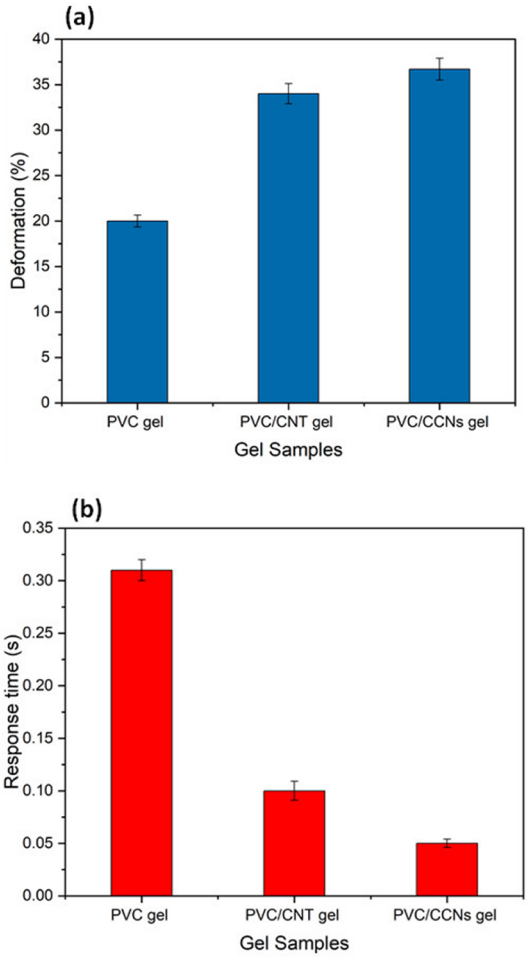

Figure 8. Comparison of results: (a) deformation of the pure PVC gel, PVC/CNT, and PVC/CCNs composite gel, and (b) response time of the pure PVC gel, PVC/CNT, and PVC/CCNs composite gels.

improvement. The above gel elongation calculated by a formula is as follows:

$$\delta = \Delta L/L \times 100\% \quad (1)$$

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where δ is the ductility of the material, ΔL is the total deformation of the material, and L is the original length of the material.

These results revealed that the CCNs are merely mixed with plasticized PVC gel. The PVC/CCNs composite gel has good physicochemical interactions specifically due to the molecular forces, hydrogen bonding, and dipole−dipole interactions among the PVC, DBA, and CCNs materials.

Electro-responsiveness of the Pure PVC and PVC Composite Gels. In this study, the deformation and response time data were recorded on a self-fabricated actuator setup of pure plasticized PVC, PVC/CNTS, and PVC/CCNs composite gels under applied voltage. The deformation of gel samples is recorded as about 34% and 36.7%, and the response time of the four-layer actuator is 0.32, 0.1, and 0.05 s after the application of the 1000 V electric force as given in Figure 8a and b. The high deformation and fast response time of PVC/CCNs composite gel are recorded as compared to pure PVC and PVC/CNTs gels. The deformation occurred due to the Maxwell stress effect and was attributed to the fact that the plasticizer DBA molecules were charged under applied voltage. Therefore, the plasticizer DBA polarized and resulted in the PVC chains opening and DBA being easily passed from the PVC gel network.[32] Whenever voltage was applied, the negative charges of the cathode passed from the gels moved toward the anode and caused bending deformation. This high deformation and fast response signified that there were strong interactions among PVC, DBA, and CCNs. Such a high displacement of PVC/CCNs is satisfactory for the actuator and artificial muscles.[9] It was observed that the results of this study are higher than those reported in the literature to date, as shown in Table S1.

CONCLUSION AND FUTURE DIRECTION

In this study, carboxylated nanocellulose was introduced into the PVC gel. It was first introduced in the plasticized PVC gel. The nanocellulose crystals were treated and converted into carboxylated nanocellulose with abundant carboxyl elements by the TEMPO oxidation method. The carboxyl part of such nanocellulose would combine with the carbon−oxygen double bond of the plasticizer in DBA. It was also combined with the hydrogen atoms on the molecular chain of PVC. The gels' tensile properties were also evaluated. The PVC/CCNs composite gel has excellent elongation at break, with a maximum elongation of 697%, which is the highest elongation of the same proportion composition gel known so far, and the elongation at break of the T60 PVC gel is 443% in the previous study.

Furthermore, the SEM results showed that the agglomerated nanocellulose and the agglomeration phenomenon also affected the transparency of the PVC/CCNs composite gel. It was discussed in previously reported work. A light transmission test was conducted on the composite gel, and the results showed that the light transmission of the PVC/CCNs composite gel was only 85%, which was lower than the light transmission of the PVC gel. To address this phenomenon, this Article suggests using a better solvent to dissolve the cellulose material and strictly controlling the effect of water vapor on the material during the experiment. The actuator performance achieved is 36.7%, demonstrating that this composite gel has an excellent electrical response. The material selection demonstrated that nanocellulose material performance is superior and could be used in actuators and soft robotics.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.3c01172.

Table S1. Plasticized PVC and PVC composite gel based multilayered actuators characteristics properties (PDF)

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Notes

The authors declare no competing financial interest.

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REFERENCES

(1) Ali, Imdad; Li, Xudong; Chen, Xiaoqing; Jiao, Zhiwei; Pervaiz, Mohammad; Yang, Weimin; Li, Haoyi; Sain, Mohini A review of electro-stimulated gels and their applications: Present state and future perspectives. *Mater. Sci. Eng., C* **2019**, *103*, 109852.

(2) Chen, Yujie; Chen, Chi; Rehman, Hafeez Ur; Zheng, Xu; Li, Hua; Liu, Hezhou; Hedenqvist, Mikael S. Shape-memory polymeric artificial muscles: Mechanisms, applications and challenges. *Molecules* **2020**, *25*, 1−27.

(3) Shin, Eun-Jae; Park, Won-Hyeong; Kim, Sang-Youn Fabrication of a High-Performance Bending Actuator Made with a PVC Gel. *Appl. Sci.* **2018**, *8*, 1284.

(4) Zhang, Wei; Wang, Ruixing; Sun, ZhengMing; Zhu, Xiangwei; Zhao, Qiang; Zhang, Tengfei; Cholewinski, Aleksander; Yang, Fut-Kuo; Zhao, Boxin Rattapol Pinnaratip et al "Catechol-functionalized hydrogels: biomimetic design, adhesion mechanism, and biomedical applications. *Chem. Soc. Rev.* **2020**, *49*, 433−464.

(5) Xue, Bin; Meng, Qin; Wang, Tiankuo; Wu, Junhua; Luo, Dongjun; Jiang, Qing; Li, Ying; Cao, Yi; Wang, Wei Electrically Controllable Actuators Based on Supramolecular Peptide Hydrogels. *Adv. Funct Mater.* **2016**, *26*, 9053−9062.

(6) Hwang, Taeseon; Frank, Zachary; Neubauer, Justin; Kim, Kwang Jin High-performance polyvinyl chloride gel artificial muscle actuator with graphene oxide and plasticizer. *Sci. Rep* **2019**, *9*, 2−10.

(7) Ali, Imdad; Ali, Ahsan; Ahmed, Ali; Ramzan, Muhammad; Hussain, Khalid; Li, Xudong; Jin, Zhan; Augusto, Otavio; Dias, Titton; Yang, Weimin; Li, Haoyi; Zhang, Liyan; et al. Highly electro-responsive composite gel based on functionally tuned graphene filled polyvinyl chloride. *Polym. Adv. Technol.* **2021**, *32*, 1−10.

(8) Tim Helps; Taghavi, Majid; Rossiter, Jonathan Thermoplastic electroactive gels for 3D-printable artificial muscles. *Smart Mater. Struct* **2019**, *28* (8), 1−11.

(9) Ali, Imdad; Latif, Abdul; Hussain, Khalid; Shehzad, Farooq Khurum; Ahmed, Ali; Faisal, Rehman; Li, Xudong; Titton Dias, Otavio Augusto; Yang, Weimin; Li, Haoyi Ionic liquids enhanced performance of PVC gels actuator. *J. Appl. Polym. Sci.* **2021**, *138*, 1−7.

(10) Mijangos, C.; Calafel, I.; Santamaría, A. Poly(vinyl chloride), a historical polymer still evolving. *Polymer (Guildf).* **2023**, *266*, 125610.

(11) Yamano, M.; Ogawa, N.; Hashimoto, M.; Takasaki, M.; Hirai, T. A contraction type soft actuator using poly vinyl chloride gel. *IEEE Int. Conf. Robot. Biomimetics* **2008**, 745−750.

(12) Asaka, K.; Hashimoto, M. Electrical properties and electro-mechanical modeling of plasticized PVC gel actuators. *Sensors Actuators, B Chem.* **2018**, *273*, 1246−1256.

(13) Carpi, F.; Frediani, G.; Turco, S.; De Rossi, D. Bioinspired tunable lens with muscle-like electroactive elastomers. *Adv. Funct Mater.* **2011**, *21*, 4152−4158.

(14) Shian, S.; Diebold, R. M.; Clarke, D. R. Tunable lenses using transparent dielectric elastomer actuators. *Opt Express* **2013**, *21*, 8669−8676.

(15) Rizzello, G. A Review of Cooperative Actuator and Sensor Systems Based on Dielectric Elastomer Transducers. *Actuators* **2023**, *12*, 46.

(16) He, Qingsong; Pan, Hui; Zhao, Zefang; Zhang, Hao; Yin, Guoxiao; Wu, Yuwei; Cai, Lei; Yu, Min; Duan, Jinjun; Shen, Qi; et al. Polyvinyl chloride gel based electro-active switchable bio-inspired adhesive. *Compos Part A Appl. Sci. Manuf* **2022**, *163*, 107180.

(17) Bekatorou, A.; Plioni, I.; Sparou, K.; Maroutsiou, R.; Tsafrakidou, P.; Petsi, T.; Kordouli, E. Bacterial cellulose production using the corinthian currant finishing side-stream and cheese whey: Process optimization and textural characterization. *Foods* **2019**, *8*, 193.

(18) Wu, G.; Gu, Y.; Hou, X.; Li, R.; Ke, H.; Xiao, X. Hybrid nanocomposites of cellulose/carbon-nanotubes/polyurethane with rapidly water sensitive shape memory effect and strain sensing performance. *Polymers (Basel)* **2019**, *11*, 1586.

(19) Koutinas, A. A.; Sypsas, V.; Kandylis, P. Nanotubular cellulose for bioprocess technology development. *PLoS One* **2012**, *7*, 34350.

(20) He, X.; Guo, F.; Ge, T.; Tang, K.; Shi, S.; Geng, M. Preparation and properties of carboxylated nanocellulose by two methods. *Sci. Technol.* **2018**, *2*, 19−24.

(21) Rong, Q.; Lei, W.; Chen, L.; Yin, Y.; Zhou, J.; Liu, M. Anti-freezing, conductive self-healing organohydrogels with stable strain-sensitivity at subzero temperatures. *Angew. Chemie Int. Ed* **2017**, *56*, 14159−14163.

(22) RANA, D.; BAG, K.; BHATTACHARYYA BMM, S. N. Miscibility of poly styrene-co-butyl acrylate with poly ethyl methacrylate.pdf. *J. Polym. Sci., Part B: Polym. Phys.* **2000**, *38*, 369.

(23) Rana, D.; Mandal, B. M.; Bhattacharyya, S. N. Analogue calorimetric studies of blends of poly(vinyl ester)s and polyacrylates. *Macromolecules* **1996**, *29*, 1579−1583.

(24) Isogai, A.; Saito, T.; Fukuzumi, H. TEMPO-Oxidized Cellulose Nanofibers. *Nanoscale* **2011**, *3* (1), 71−85.

(25) Liu, S.; Low, Z. X.; Xie, Z.; Wang, H. TEMPO-Oxidized Cellulose Nanofibers: A Renewable Nanomaterial for Environmental and Energy Applications. *Adv. Mater. Technol.* **2021**, *6* (7), 1−23.

(26) Hassan, F.; Akbar, F.; Rashid, F.; Roomi, S.; Ghafoor, A.; Suleman, M. Assessment of Biodegradability of PVC Containing Cellulose by White Rot Fungus. *Malays. J. Microbiol* **2014**, *10* (2), 119−125.

(27) Sheltami, R. M.; Kargarzadeh, H.; Abdullah, I. Effects of Silane Surface Treatment of Cellulose Nanocrystals on the Tensile Properties of Cellulose-Polyvinyl Chloride Nanocomposite. *Sains Malaysiana* **2015**, *44* (6), 801−810.

(28) Ali, I.; Yang, W.; Li, X.; Ali, A.; Jiao, Z.; Xie, P.; Dias, O. A. T.; Pervaiz, M.; Li, H.; Sain, M. Highly Electro-Responsive Plasticized PVC/FMWCNTs Soft Composites: A Novel Flex Actuator with Functional Characteristics. *Eur. Polym. J.* **2020**, *126*, 109556.

(29) Rana, D.; Kim, H. L.; Kwag, H.; Choe, S. Hybrid Blends of Similar Ethylene 1-Octene Copolymers. *Polymer* **2000**, *41*, 7067−7082.

(30) Rana, D.; Lee, C. H.; Cho, K.; Lee, B. H.; Choe, S.; Al, R. E. T. Thermal and Mechanical Properties for Binary Blends of Metallocene Polyethylene with Conventional Polyolefins. *J. Appl. Polym. Sci.* **1998**, *69*, 2441−2450.

(31) Ali, I.; Weimin, Y.; Xudong, L.; Ali, A.; Zhiwei, J.; Xie, P.; Augusto Titton Dias, O.; Pervaiz, M.; Haoyi, L.; Sain, M. Highly Electro-Responsive Plasticized PVC/FMWCNTs Soft Composites: A Novel Flex Actuator with Functional Characteristics. *Eur. Polym. J.* **2020**, *126*, 109556.

(32) Asaka, K.; Hashimoto, M. Electrical Properties and Electro-mechanical Modeling of Plasticized PVC Gel Actuators. *Sensors Actuators, B Chem.* **2018**, *273*, 1246−1256.

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