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Piezoelectric cellulose/poly(vinylidene fluoride) glycerogels with synergistically enhanced energy output for wide temperature range

Md. Tariful Islam Mredha^{1,#}, Rumesh Rangana Manimel Wadu^{1,#}, Shuangpeng Li^{2,3,#}, Adith Varma Rama Varma¹, Tanish Gupta¹, Wonoh Lee¹, Chunli Zhang^{2,3,*}, Weiqiu Chen^{2,3,*}, Insu Jeon^{1,*}

¹School of Mechanical Engineering, Chonnam National University, Gwangju 61186, Republic of Korea.

²Key Laboratory of Soft Machines and Smart Devices of Zhejiang Province, Department of Engineering Mechanics, Zhejiang University, Hangzhou 310027, Zhejiang, China.

³Huanjiang Laboratory, Zhuji 311816, Zhejiang, China.

#Authors contributed equally.

*Correspondence to: Dr. Chunli Zhang, Key Laboratory of Soft Machines and Smart Devices of Zhejiang Province, Department of Engineering Mechanics, Zhejiang University, Hangzhou 310027, Zhejiang, China; Huanjiang Laboratory, Zhuji 311816, Zhejiang, China. E-mail: zhangcl01@zju.edu.cn; Dr. Weiqiu Chen, Key Laboratory of Soft Machines and Smart Devices of Zhejiang Province, Department of Engineering Mechanics, Zhejiang University, Hangzhou 310027, Zhejiang, China; Huanjiang Laboratory, Zhuji 311816, Zhejiang, China. E-mail: chenwq@zju.edu.cn; Dr. Insu Jeon, School of Mechanical Engineering, Chonnam National University, 77 Yongbong-ro, Buk-gu, Gwangju 61186, Republic of Korea. E-mail: i_jeon@chonnam.ac.kr

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Abstract

Gel-based piezoelectric materials are stretchable, wearable, and environmentally friendly, unlike their conventional solid counterparts. However, designing environment-tolerant, high-performance piezoelectric gels is challenging. Herein, we develop a piezoresponsive stretchable glycerogel (GG), leveraging the cooperative structure-forming effect of cellulose, poly(vinylidene fluoride) (PVDF) and glycerol (a green extremotolerant solvent). The facile inter- and intramolecular cellulose/PVDF interactions within the hydrogen-bonded network of glycerol generate a highly electroactive crystalline β -phase while retaining mechanical integrity. Therefore, the synergy-driven GG is more piezoresponsive than gels fabricated using individual polymers. Despite having a low polymer density (≈ 16 wt%), the GG exhibits impressive functional attributes such as Young's modulus (≈ 12 MPa), tensile strength (≈ 3 MPa), piezoelectric voltage (92 mV cm^{-2}), and current output (110 nA cm^{-2}). Furthermore, it exhibits long-term stability over a wide temperature range (-20 to 80 °C) owing to its robust structural integrity and thermal



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adaptability. The study findings underscore the viability of preparing high-performance extremotolerant piezoelectric gels for use in next-generation stretchable/wearable piezoelectric sensors and energy devices.

Keywords: Piezoelectric gels, cellulose, poly(vinylidene fluoride), glycerogels, synergistic effect, wide temperature applicability

INTRODUCTION

The use of nonrenewable energy sources has resulted in environmental pollution and exacerbated the energy crisis. Consequently, research has focused on reliable, renewable, and environmentally friendly energy sources as alternatives to mitigate pollution and meet growing energy demands^[1-3]. Mechanical energy is abundant and can be converted into usable energy using piezoelectric nanogenerators to power micro- to nanoscale devices^[4,5]. Commonly used piezoelectric materials, such as lithium niobate [LiNbO₃]^[6], barium titanate [BaTiO₃]^[7], and lead zirconate titanate [Pb(Zr,Ti)O₃]^[8], exhibit excellent piezoelectric energy conversion capabilities. However, existing piezoelectric materials are inherently brittle owing to their highly crystalline structure. They are also toxic, expensive, and environmentally unfriendly^[9,10]. Therefore, researchers are actively attempting to identify new eco-friendly materials that are biocompatible, electroactive, flexible, and lightweight^[11,12]. Soft biocompatible piezoelectric materials are of particular interest in the rapidly expanding fields of flexible electronics, soft robotics, and wearable devices.

The use of gels (a class of semisolid materials featuring swollen polymer networks) is being increasingly explored in soft robotics, biomedicine, sensing/actuation, and flexible electronics. They also show promise for energy generation and energy storage owing to their unique combination of functions, which are unmatched by any existing solid or liquid material. Gels demonstrate various desirable physical properties, such as stretchability, flexibility, electrical conductivity, biocompatibility, biodegradability, and even piezoelectricity^[13-23]. They have recently been used extensively for the fabrication of various stretchable and wearable devices. Proper design strategies can harness the large amount of wasted mechanical energy of gels to generate piezoelectric energy, facilitating the development of sustainable stretchable and wearable devices. Among the many piezoelectric and triboelectric materials that have been developed to harvest this wasted energy^[24-26], gel-based piezo/triboelectric materials are most suitable for wearable devices owing to their inherent stretchability, elasticity, and flexibility. Numerous piezocapacitive or piezoresistive gels and gel-based devices/sensors have recently been developed^[27-29]; however, the development of piezoelectric gels or gel-based devices is particularly challenging because most piezoelectric materials are unsuitable for incorporation into gel systems. Therefore, the development of soft gel-based piezoelectric materials is still in its nascent stages and represents an emerging research area for energy-harvesting, energy-storage, and wearable devices. For example, a supramolecular strategy was implemented to align dipoles to synergistically enhance the piezoelectric response. This was achieved through hydrogen bonding and charge transfer complexation in organogel-based nanogenerators following a complex fabrication procedure^[20]. Moreover, a hydrogel fabricated from graphene-oxide-nanoparticle-doped poly(2-hydroxyethyl methacrylate) was used to generate an electrical current in aqueous media^[21]. Additionally, a polyacrylonitrile/poly(vinylidene fluoride) (PAN/PVDF) hydrogel was reported to exhibit enhanced piezoelectric performance^[22]. Furthermore, the piezoelectric output of a conductive hydrogel was enhanced by introducing β -phase PVDF-trifluorethylene^[23]. However, the piezoelectric-type hydrogels developed to date exhibit poor energy output performances along with weak mechanical and environmental stability.

A formidable challenge in developing high-performance piezoelectric gels is that their structural integrity is naturally disrupted in the highly solvated microenvironment of gels. This can make the material brittle and

dramatically less piezoresponsive. To date, few approaches have been devised for upgrading the piezoelectric output of gels. Another concern is the poor environmental adaptability of conventional gels, which limits their practical applications. Recently developed water-based piezoelectric hydrogels only function in aqueous environments because water is required to maintain the gel properties. Thus, the lifetime of these hydrogels is constrained in nonaqueous (e.g., air) or extreme environments (such as extremely high or low temperatures). This limitation arises from the drying and freezing properties of water^[13,16,30,31]. To overcome these limitations, we have formulated a molecular engineering technique to integrate carefully chosen piezoelectric polymer(s) and an extremotolerant solvent. Subtle chemical interactions among the selected polymer(s) and solvent are leveraged to simultaneously achieve two goals: (i) a structure with a high degree of piezoactivity; and (ii) robust mechanical integrity in the gel state. This ensures the efficient dissipation of the input mechanical signal through the piezoactive structure, maximizing the piezoelectric output.

Based on this strategy, we developed a novel class of extremotolerant piezoelectric glycerogels (GGs) comprising cellulose and PVDF swollen in glycerol, prepared using a facile method [Figure 1A]. PVDF and its copolymers are semicrystalline piezoactive polymer materials that are widely used in the field of energy harvesting because they are biocompatible and exhibit excellent piezoelectric and dielectric properties^[32,33]. Among the different crystalline phases of PVDF (α , β , γ , δ , and ϵ), the polar β -phase has a non-centrosymmetric structure. It exhibits the highest dipolar moment per unit cell (8×10^{-30} C m), indicating a high piezoelectric response^[32,33]. Despite the excellent prospects of PVDF, using it to fabricate wearable and stretchable piezoelectric gels is challenging because of its inherent hydrophobicity, brittleness, and structural mismatch with hydrophilic gel components; therefore, mechanical energy cannot be readily transferred to the crystals in PVDF-based gels. Conversely, cellulose - the most abundant natural biopolymer - is extensively used to develop tough semicrystalline gels owing to its nontoxicity, biocompatibility, low cost, and satisfactory piezoactive properties^[34,35]. The rigid backbone of cellulose and its ability to easily crystallize through hydrogen bonding can reinforce the composite gel structure; the rigid networks also facilitate the mechanical signal transferability. Recently, we demonstrated that glycerol- as a green solvent - can be used to integrate a wide range of polymers and crosslinkers to develop remarkably tough extremotolerant GGs; this was facilitated by facile and stable polymer-solvent interactions over a wide temperature range^[14,36-38]. Considering these aspects, we rationally designed a facile method to integrate both cellulose and PVDF using glycerol as the solvent to develop tough, flexible, and extremotolerant cellulose/PVDF GGs with synergistically amplified piezoelectric properties compared with those of gels fabricated using the individual constituent polymers. The molecular-level integration and formation of inter- and intramolecular hydrogen bonds among cellulose, PVDF and glycerol promoted the crystallization of the electroactive β -phase, and enhanced the gel's toughness and extreme temperature tolerance [Figure 1B]. The rigid network of cellulose in the developed GGs efficiently transferred the applied mechanical energy to orient the dipoles of crystalline PVDF, resulting in impressive piezoelectric properties across a wide temperature range [Figure 1C and D]. Although glycerol has previously been used as a plasticizer for piezoelectric materials^[26], to the best of our knowledge, this is the first time piezoelectric GGs (glycerol content: > 80 wt%) or any gel-based piezoelectric material have been demonstrated to operate at such diverse temperatures.

EXPERIMENTAL

Materials and instrumentation

Glycerol, N,N'-dimethylacetamide (DMAc), lithium chloride (LiCl), and ethanol were purchased from Samchun Pure Chemicals (Republic of Korea). Cellulose filter paper (ADVANTEC) was procured from Toyo Roshi Kaisha (Japan). PVDF (Mw \approx 275,000) and poly(vinyl alcohol) (PVA, Mw: 89,000-98,000) were obtained from Sigma-Aldrich. Dimethyl sulfoxide (DMSO) was acquired from Daejung Chemicals & Metals (Republic of Korea). All chemicals were used as received without further purification. Ultrapure deionized (DI) water was used to prepare all hydrogels.

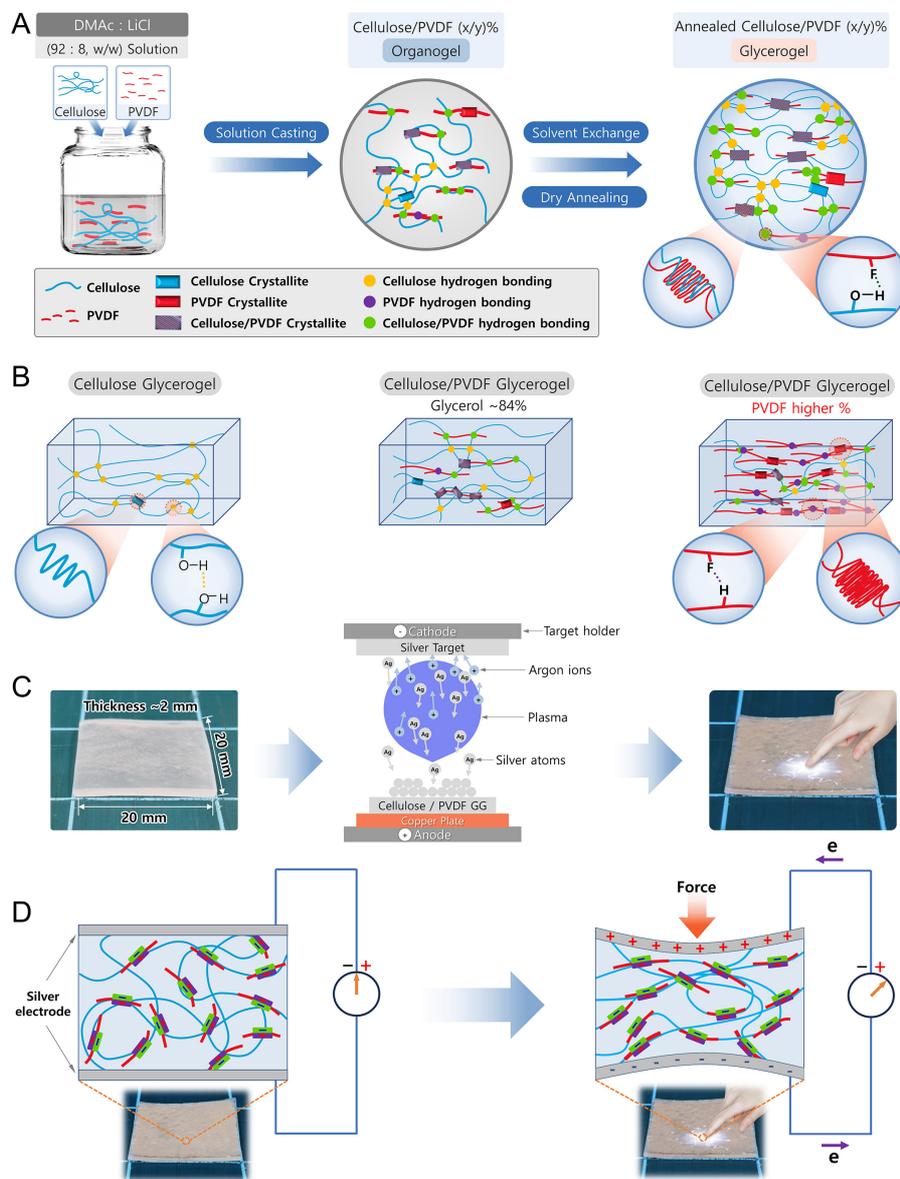


Figure 1. Design and fabrication of cellulose/poly(vinylidene fluoride) (PVDF) glycerogels (GGs) for high-performance piezoelectric applications. (A) Schematic of the process employed to prepare cellulose/PVDF GGs. (B) Magnified schematics of neat cellulose, (1/2)% cellulose/PVDF gel, and (0.5/5)% cellulose/PVDF gel; the circular area indicates the type of bonding and crystalline structures. (C) Illustration of the silver-sputtering process, and photograph of the silver-sputtered (1/2)% cellulose/PVDF GG, which was used to generate piezoelectricity. (D) Illustrative mechanism of force-induced polarization in the (1/2)% cellulose/PVDF GG-based device to generate piezoelectricity for an external circuit. DMAc: N,N'-dimethylacetamide.

All tests were performed under ambient conditions (humidity, 30%-60%; temperature, ~25 °C) unless otherwise specified. The PVDF phases and crystallinity of the gels were investigated using a Fourier-transform infrared (FTIR) spectrometer (PerkinElmer Spectrum IR, USA) and an X-ray diffractometer (PANalytical X'Pert Pro MRD, the Netherlands). In the FTIR measurements, eight scans were accumulated, and four spectral resolutions were examined. X-ray diffraction (XRD) was conducted using a CuK-radiation source ($\lambda = 0.1541874$ nm) operating at 40 kV and 30 mA. The scanning rate was $8.5^\circ \text{ min}^{-1}$, with a step size

of 0.026° . A field-emission scanning electron microscope (FE-SEM; S-4700, Hitachi, Japan) was used to investigate the gel structure. The samples were freeze-dried at -80°C for 1 d and sputtered with platinum before observing the gel surfaces by FE-SEM. The surface potential of GGs was determined through Kelvin-probe force microscopy (KPFM), which was conducted using atomic force microscopy (AFM; NX10, Park Systems, Republic of Korea) at the Energy Convergence Core Facility in Chonnam National University. KPFM was performed on a scanning area of $5\ \mu\text{m} \times 5\ \mu\text{m}$ using a Cr/Au-coated conductive tip with a spring constant of $0.3\ \text{N m}^{-1}$ and a frequency of 65 kHz. A custom-made device equipped with a force sensor (Nidec-Shimpo FGP-2, Japan) was used to tap the prepared gels. The open-circuit voltage density (OCVD), short-circuit current density (SCCD), and piezoelectric charge constant (d_{33}) were measured using an oscilloscope (Tektronix TBS2204B, USA), a low-noise current preamplifier (SR570, USA), and a d_{33} meter (YE2730A, China), respectively. To calculate the dielectric constant, complex impedance spectra were recorded in a frequency range of 100-1 MHz using a multichannel potentiogalvanostat (Metrohm Multi Autolab/M204, the Netherlands). A commercial tensile testing machine (TO-100-1C, TESTONE, Republic of Korea) was used to mechanically characterize the gel samples using a 10 or 100 kgf load cell based on their qualitatively observed mechanical strength.

Fabrication of cellulose/PVDF GGs

Cellulose filter paper with dimensions of $\sim 5\ \text{mm} \times 5\ \text{mm}$ was sequentially washed with water ($\sim 6\ \text{h}$), ethanol ($\sim 6\ \text{h}$), and DMAc ($\sim 12\ \text{h}$) and then vacuum dried at 60°C for $\sim 24\ \text{h}$. The obtained paper was used to prepare cellulose/PVDF hydrogels. Our previous studies revealed that DMAc/LiCl is excellent for dissolving highly crystalline cellulose by breaking its high-density hydrogen bonds; the resulting solutions were used to fabricate various functional gels with excellent mechanical properties^[14,18,39,40]. We found that DMAc/LiCl could also dissolve PVDF at high concentrations because PVDF is also a largely crystalline polymer dominated by hydrogen bonds. Therefore, DMAc/LiCl was chosen to dissolve and integrate both cellulose and PVDF into a high-performance composite gel. A DMAc/LiCl (92:8, w/w) solvent (at 25°C) was used to dissolve $x\ \text{wt}\%$ cellulose filter paper (0.5-1.5 wt%) and $y\ \text{wt}\%$ PVDF pellets (0.5-5 wt%) to prepare $(x/y)\%$ cellulose/PVDF solutions [Supplementary Table 1]. Each of the resulting cellulose/PVDF solutions was poured into clean, flat glass molds ($80\ \text{mm} \times 80\ \text{mm} \times 3\ \text{mm}$) and exposed to controlled atmospheric conditions (temperature: $\sim 25^\circ\text{C}$; humidity: 30%-60%) for at least 3 d to enable the gelation of cellulose/PVDF through hydrogen bond formation. This cellulose/PVDF organogel was transformed into a hydrogel via sequential equilibration in ethanol (1 d) and DI water (1 d). The resulting hydrogel was converted into a GG by sequentially equilibrating the gel in aqueous solutions of 25%, 50%, 75%, and 100% glycerol for 12, 12, 24, and 24 h, respectively. The prolonged step-wise solvent exchange helped preserve the structural integrity and ensured that water was completely replaced with glycerol in the gel network^[14]. Subsequently, the solvent-exchanged GG was dry-annealed in a closed environment at 120°C for 6 h and then slowly cooled to room temperature to obtain $\sim 2\text{-mm}$ -thick cellulose/PVDF GGs, which were used for further experiments [Figure 1A and Supplementary Figure 1]. Note that all the gel precursor solutions were prepared in closed glass vials, and the precursor solution consisting of DMAc, LiCl, cellulose and PVDF was kept inside a closed fume hood during gelation to prevent DMAc from leaking into the environment. After purification, gelation, and the solvent exchange process, ethanol, DMAc and LiCl can be retrieved and separated using standard fractional distillation methods because of their highly distinct boiling points; therefore, no solvent or chemicals will be wasted when these materials are produced at an industrial scale.

Fabrication of PVA/PVDF GGs

First, a PVA/PVDF precursor solution was prepared by dissolving PVA/PVDF at different weight percentages (10/5, 10/10, 5/10 wt%) in DMSO with continuous stirring at 120°C for 1 d. The solution was poured between rectangular glass molds to a thickness of $\sim 3\ \text{mm}$. Then, gelation was performed by freeze-thawing, in which the molded solution was frozen at -50°C (12 h) and then thawed at room temperature

(12 h). This process was repeated for three cycles for complete gelation. The resulting gel was removed from the mold and equilibrated in water for 3 d to obtain a water-equilibrated hydrogel, which was then converted to a GG by sequentially equilibrating it in aqueous solutions of 25, 50, 75, and 100% glycerol for 12, 12, 24, and 24 h, respectively. Next, the GG was dry-annealed at 120 C for 6 h in a closed environment and then slowly cooled to room temperature to obtain the final PVA/PVDF GG (thickness \approx 2 mm), which was used for further characterization.

Device fabrication for piezoelectric response analysis

For the piezoelectric response experiments, Ag was sputtered onto the surface of the developed GG (20 mm \times 20 mm \times \sim 2 mm) using a commercial direct current (DC) ion-sputtering machine (magnetron sputtering system, ALPHAPLUS, Republic of Korea). For sputtering, the GGs were placed on the anode component of the machine. The chamber was maintained at a vacuum pressure of 1.5×10^{-4} Pa in an inert environment (argon gas, flowrate: 20 cm³ min⁻¹) for 5 min with a DC of 2 A. The sputtered samples were used for piezoelectric property assessment [Figure 1C].

RESULTS AND DISCUSSION

Physicochemical characterization of cellulose/PVDF GGs

Cellulose/PVDF GGs were fabricated with different compositions. The piezoelectric performances of the gels were anticipated to depend on both their piezoelectric crystal structures and mechanical integrity^[21,41,42]. Although gels were successfully obtained for each selected composition, increasing the amount of PVDF resulted in physically weak gels.

The chemical structure of the cellulose/PVDF GG matrix [Supplementary Figure 1] was analyzed by FTIR spectroscopy [Figure 2A]. The bands at 763, 795, and 975 cm⁻¹ for pristine PVDF indicated the presence of the α -phase, whereas those at 841, 1,275, 1,403, and 1,431 cm⁻¹ were attributed to the β -phase [Figure 2B(i)]^[41]. In contrast, the spectra of the cellulose/PVDF GGs showed bands characteristic of PVDF at 841, 1,275, 1,403, and 1,431 cm⁻¹ [Figure 2B(ii)], revealing the presence of only the β -phase PVDF in the hybrid gel^[41]. This result suggests that the incorporation of cellulose and glycerol facilitated the formation of a highly piezoelectric β -phase. It also eliminated the non-electroactive α -phase of PVDF in the hybrid gel. The facile hydrogen bonding ability of rigid-cellulose polymers and hydroxyl-rich glycerol molecules could facilitate the lateral arrangement of PVDF. This arrangement created non-centrosymmetric (β -phase) crystal structures. The intensity of the characteristic β -phase bands increased with the addition of large amounts of PVDF to the gel matrix without the appearance of the α -phase band; this suggests that the cooperative phenomenon of β -phase crystallization is effective even at higher PVDF concentrations at which cellulose concentrations are comparatively insignificant. This result also verifies that in addition to cellulose, the glycerol in the gel network also plays an active role in β -phase formation. However, a GG based only on PVDF could not be fabricated in this study; this was because the rigid-cellulose polymer was important as a mechanical support for stabilizing the PVDF-based GG.

In the XRD profile of pristine PVDF [Figure 2C], the characteristic crystalline peaks appeared at 2θ values of 18.3° and 19.9°, which have been ascribed to the α -phase peaks^[41,43], whereas that of cellulose appeared at 20.4°. However, the incorporation of PVDF into the cellulose matrix decreased the full width at half maximum (FWHM) of the characteristic cellulose peak and shifted it slightly to $2\theta \approx 20.6^\circ$ (β -phase; Figure 2D)^[22,41]. These changes indicate a higher degree of crystal alignment in the cellulose/PVDF gel compared with that in the neat cellulose gel and validate its cellulose-mediated cooperative β -phase crystallization, which is particularly important for ensuring the mechanical integrity of the β -phase in the gel network. Furthermore, the surface structure of the gel was comparatively investigated by FE-SEM

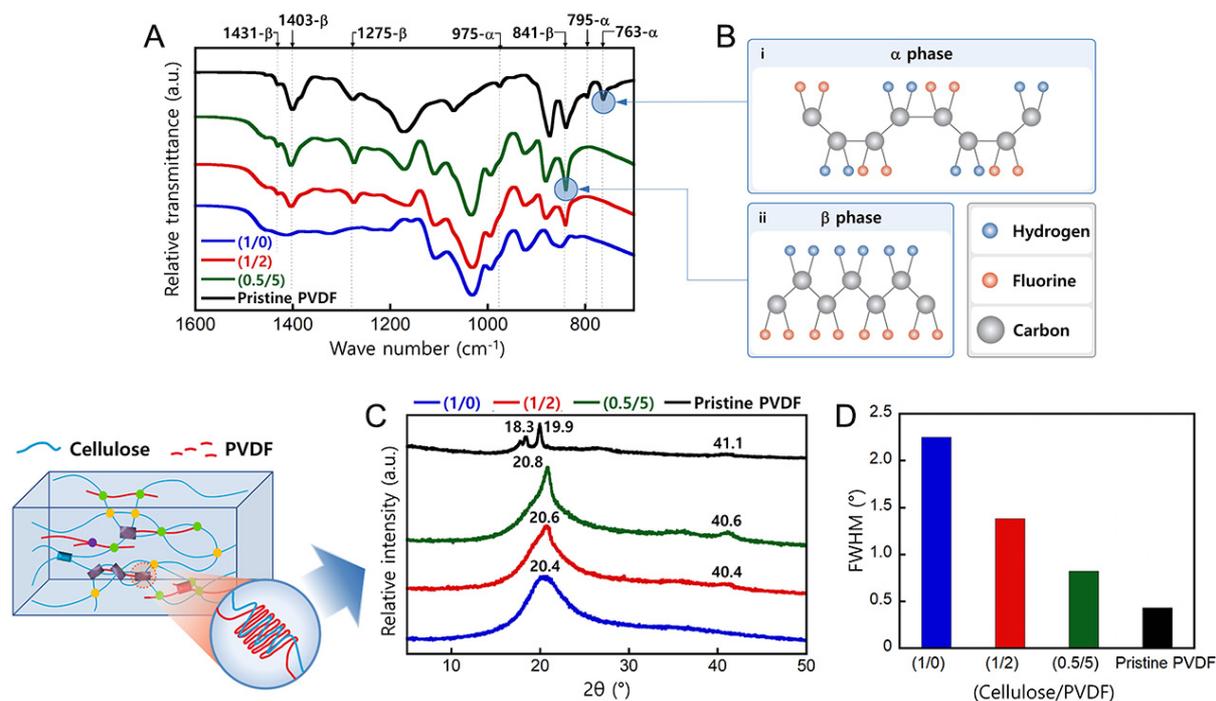


Figure 2. Structural characterization of the cellulose/PVDF GGs. (A) FTIR spectra of the cellulose/PVDF GGs. (B) Crystalline phases of PVDF: (i) α -phase and (ii) β -phase. (C) XRD patterns of cellulose/PVDF GGs and (D) corresponding full width at half maximum (FWHM) values.

[Supplementary Figure 2]. Owing to the high glycerol content in the GGs, investigating the surface morphology of the freeze-dried GG by FE-SEM was difficult^[14]. Therefore, the corresponding freeze-dried hydrogels were used. The FE-SEM images of the neat cellulose gel indicate the presence of irregular polymer aggregate structures, whereas those of the (0.5/5)% cellulose/PVDF gel show globular and micro-scale phase-separated structures. Additionally, the polymer structures appearing in the FE-SEM images of the (1/2)% cellulose/PVDF gel are homogeneous with less aggregated and less globular structures. These results suggest that a critical balance is required between the two polymers, which have contrasting chemical properties, to integrate them at the molecular level, form homogeneous crystals, and manipulate their preeminent functions. KPFM was used to understand the surface potential of the GGs. The contact potential difference between the tip and sample was measured to reveal the surface potential. The (1/2)% cellulose/PVDF GG exhibited an enhanced surface potential of 86.8 ± 0.1 mV, whereas pure cellulose GG showed a near-zero surface potential of 7.3 ± 0.1 mV [Supplementary Figure 3]. This enhancement observed with (1/2)% cellulose/PVDF suggests that the synergistic effect is a result of the molecular dipole orientation of PVDF in GG.

Piezoelectric response of cellulose/PVDF GGs

The piezoelectric responses of the cellulose/PVDF GGs were evaluated by their output OCVD and SCCD measurements [Figure 3A]. The pure cellulose gel exhibited a low OCVD of 22.78 ± 3.07 mV cm⁻² and SCCD of 25.02 ± 3.06 nA cm⁻², potentially because of the inferior piezoelectric properties of cellulose^[44,45]. Adding PVDF to the composition of (1/2)% cellulose/PVDF significantly enhanced the voltage and current outputs. Maximum values reached 86.06 ± 6.00 mV cm⁻² and 104.93 ± 6.66 nA cm⁻², respectively, owing to the synergistic contribution of the piezoelectric β -phase crystals and the mechanical strength of the hybrid gel network. However, further increasing the PVDF content significantly decreased the output. This was because excess PVDF weakened the gel by creating phase-separated structures, which possibly curtailed its

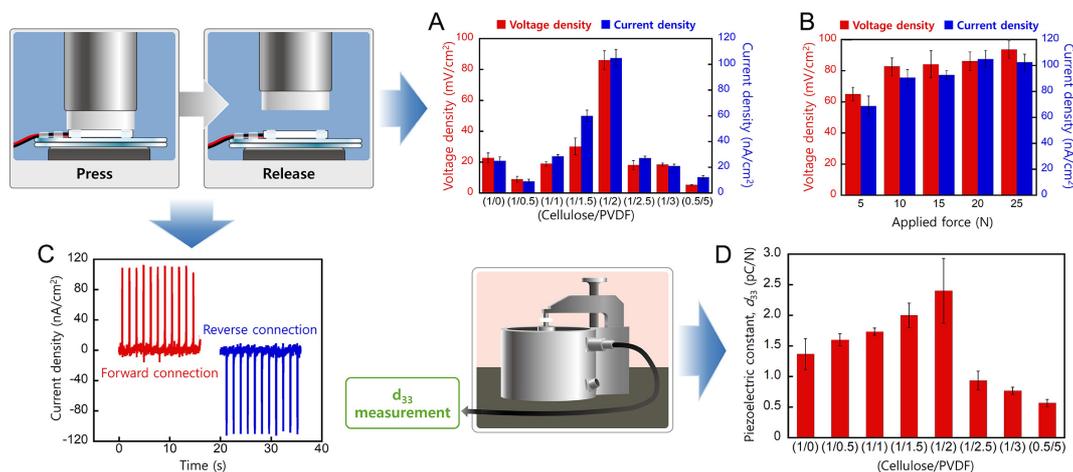


Figure 3. Piezoelectric response properties of the cellulose/PVDF GGs. (A) Piezoelectric output OCVD (mV cm^{-2}) and SCCD (nA cm^{-2}) of ≈ 2 -mm-thick cellulose/PVDF GGs (surface area: $2 \text{ cm} \times 2 \text{ cm}$) measured under a cyclic load of 20 N at a frequency of $\approx 0.7 \text{ Hz}$. (B) Voltage and current outputs of the (1/2)% cellulose/PVDF gel under different magnitudes of cyclic loads ranging from 5 to 25 N. (C) Representative current output of the (1/2)% cellulose/PVDF gel under a cyclic load of 20 N in the forward and reverse connection modes of an external circuit. (D) Variation in the piezoelectric charge constant d_{33} with the concentration ratio of cellulose/PVDF GGs. Data in (A, B and D) are presented as average values with mean absolute deviation from the measurements of three samples.

mechanical signal transduction ability. Furthermore, GGs with different cellulose concentrations were tested to identify the optimal piezoresponsive gel [Supplementary Figure 4]. For a given cellulose concentration, increasing the PVDF content in the gel produced consistent piezoelectric response behavior patterns. The (1/2)% cellulose/PVDF gel delivers the maximum output values, as discussed previously.

Subsequently, the output response of the optimized gel was investigated under loads ranging from 5 to 25 N. The piezoelectric output voltage and current densities increased linearly with the applied force [Figure 3B, Supplementary Figure 5]. The increases occurred at rates of $1.2125 \text{ mV cm}^{-2} \text{ N}^{-1}$ and $1.6335 \text{ nA cm}^{-2} \text{ N}^{-1}$, respectively. The higher the load magnitude, the larger the distortion of the piezoelectric crystals, which can generate larger energy outputs. These results also validate the ability of the hybrid gel to efficiently transform mechanical energy into piezoelectric energy. The output signal of the (1/2)% cellulose/PVDF gel was measured in the reverse connection mode using an external circuit. The trends in current [Figure 3C] and voltage outputs [Supplementary Figure 6] were similar to those in the forward connection, albeit in the opposite direction. This verified that the generated signal was solely based on piezoelectricity^[22].

To further verify the piezoelectric properties of the cellulose/PVDF GGs, their d_{33} values were measured [Figure 3D]. Owing to the inferior piezoelectric properties of cellulose, the neat 1% cellulose gel exhibited a d_{33} value of $1.37 \pm 0.25 \text{ pC N}^{-1}$. The variation in the d_{33} values of the gels was consistent with the piezoelectric output behavior of the cellulose/PVDF gels. The (1/2)% gel delivered the highest output ($2.4 \pm 0.53 \text{ pC N}^{-1}$). A further increase in the PVDF content of the gel matrix drastically reduced the d_{33} value owing to its weak phase-separated structure. Furthermore, the dielectric properties of the gels were examined over a frequency range of $100\text{-}10^5 \text{ Hz}$ at room temperature. Among the gels with 1% cellulose, the optimized piezoresponsive (1/2)% cellulose/PVDF gel exhibited the highest dielectric constant at any given frequency. The gel with less cellulose and the highest PVDF content [(0.5/5)% gel] showed the highest dielectric constant among all the tested gels [Supplementary Figure 7]. The neat cellulose gel exhibited the lowest dielectric loss at the selected frequencies, and adding PVDF slightly increased this loss [Supplementary Figure 7]. Although the (0.5/5)% gel exhibited the most impressive dielectric properties, it had a low d_{33} value and low energy outputs, whereas the optimized (1/2)% gel exhibited a high d_{33} value and high energy outputs despite its

moderate dielectric properties. This was attributed to the efficient mechanical-signal-transferring ability of the optimized gel, which was unlikely in the case of the gel with the highest PVDF content [(0.5/5)%] owing to its phase-separated structure.

Extremotolerance and mechanical properties of cellulose/PVDF GGs

The (1/2)% cellulose/PVDF gel demonstrated excellent stability under long-term exposure to air (~7 d) at temperatures of -20 to 80 °C. In particular, its weight remained nearly unchanged at 25 °C, increased by ~2.8 wt% at -20 °C, and decreased by ~15.4 wt% at 80 °C [Figure 4A and B]. This small increase and decrease in gel weight were presumably related to the absorption of atmospheric moisture and the loss of glycerol from the GG, respectively.

Subsequently, the tensile performance of the cellulose/PVDF gels was evaluated [Figure 4C, Supplementary Figure 8]. Among the tested samples, the neat 1% cellulose gel exhibited the highest Young's modulus (12.25 ± 1.34 MPa), which decreased with the addition of PVDF. Young's modulus of the optimum piezoresponsive (1/2)% gel (10.42 ± 1.47 MPa) was close to that of the neat cellulose gel. However, Young's modulus of the gel with the highest PVDF content [(0.5/5)%; 1.83 ± 0.09 MPa] was approximately 5.7 times lower than that of the (1/2)% gel. Moreover, the tensile strength of the gels remained higher than the neat cellulose gel with the addition of up to 2% PVDF to the gel network; however, further addition of PVDF significantly reduced the gel strength. The (1/0.5)% cellulose/PVDF gel exhibited the highest tensile strength among the tested samples (3.36 ± 0.39 MPa; ~1 MPa higher than that of the neat 1% cellulose gel). The optimal piezoresponsive (1/2)% gel exhibited a fracture stress of 2.86 ± 0.09 MPa, whereas that with the highest PVDF content [(0.5/5)% gel] exhibited the lowest fracture stress (6 ± 0.7 kPa; approximately 477 times lower than that of the optimum gel). Adding PVDF to a cellulose matrix reduces the stretchability of piezoelectric gels because PVDF is inherently brittle and tends to form crystals^[46]. Consistent with this finding, the stretchability of the optimum (1/2)% gel ($50.49\% \pm 2.2\%$) was 1.6 times lower than that of the 1% cellulose gel ($82\% \pm 3.37\%$). Among all the tested gels, the specimen with the highest PVDF concentration [(0.5/5)%] exhibited the lowest stretchability ($5.69\% \pm 0.68\%$; approximately 8.9 times lower than that of the optimum (1/2)% gel). Furthermore, the (1/0.5)% gel exhibited the highest work of extension value (1.33 ± 0.15 MJ m⁻³; 0.62 MJ m⁻³ higher than that of the (1/2)% gel). The (0.5/5)% gel exhibited the lowest work of extension value (0.0003 ± 0.00008 MJ m⁻³; more than 2,350 times lower than that of the optimum piezoresponsive gel). The compression test data [Figure 4D, Supplementary Figure 9] indicated that the compressive strength of the gel increased up to the (1/2)% cellulose/PVDF composition at low strains (< 50%). A further increase in the PVDF concentration led to a reduction in the compressive strength owing to the formation of a weaker gel.

The excellent piezoelectric performance of the cellulose/PVDF gel can be explained by the coupling behavior between the mechanical and electrical properties governed by the following constitutive equations^[21,42]:

$$S = c_E \cdot T + e \cdot E, \quad (1)$$

$$D = e^T \cdot T + \epsilon_s \cdot E, \quad (2)$$

where S is the strain tensor, T is the stress tensor, D is the electric displacement vector, E is the electric field vector, c_E denotes the elastic constant (at constant electric field) matrix, e denotes the piezoelectric constant matrix, and ϵ_s denotes the dielectric constant (at constant strain) matrix. The superscript T appearing in Eq. (2) indicates the transposed matrix. The mechanical properties of our gels were consistent with their

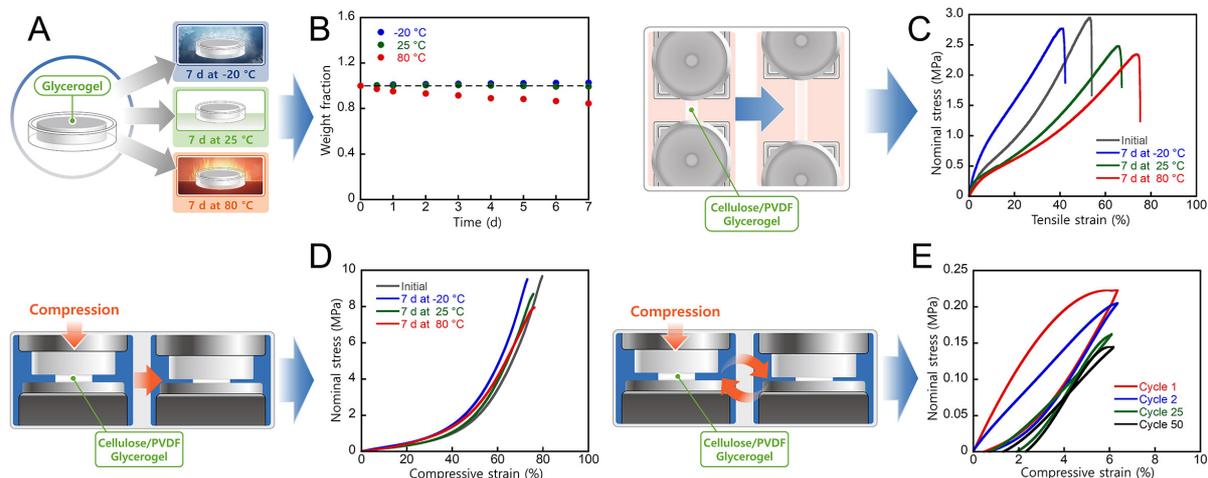


Figure 4. Extremotolerance of cellulose/PVDF GGs. (A) Evaluation of the effect of low (-20 °C), ambient (25 °C), and high (80 °C) temperatures on the cellulose/PVDF GGs. (B) Variation in the weight fractions of the (1/2)% cellulose/PVDF gel during long-term (7 d) exposure to air at -20, 25, and 80 °C and corresponding (C) tensile stress-strain and (D) compressive stress-strain curves. (E) Cyclic compression of the (1/2)% cellulose/PVDF gel at 25 °C with a strain rate of -830 min^{-1} (0.277 mm s^{-1}).

structures and piezoelectric performances. The rigid biopolymer cellulose facilitated the formation of highly electroactive β -phase PVDF crystals while preserving the mechanical integrity of the hybrid gels up to (1/2)% cellulose/PVDF composition; therefore, it exhibited the highest and most synergistically enhanced piezoelectric performance. Further increasing the PVDF content resulted in a phase-separated structure because of the compositional mismatch due to the excessive contribution of hydrophobic PVDF, which in turn significantly weakened its mechanical properties and caused poor piezoelectric performances. These results were also consistent with the solvent content of gels [Supplementary Figure 10]. With the addition of PVDF up to (1/2)% cellulose/PVDF composition, the solvent content decreased to the lowest value, suggesting a better-crystallized structure formation in this concentration. Further rising PVDF increased the solvent content owing to the weakening of structure formation. The piezoelectric, mechanical, and extremotolerant properties of the proposed GGs were compared with those of the existing piezoelectric gels, as summarized in Supplementary Table 2, and the proposed gels remarkably outperformed the existing gels despite their low polymer density ($\approx 16 \text{ wt}\%$).

Cyclic loading-unloading curves of the (1/2)% gel were acquired for over 50 cycles [Figure 4E]. The presence of significant hysteresis loops qualitatively indicated that the cellulose/PVDF gels were viscoelastic materials and toughened by the efficient energy dissipation of hydrogen bonding [Figure 4E and Supplementary Figure 11]. The energy dissipation of our gel also suggests that the mechanical energy input can be efficiently dissipated to the polymer network^[13-16], which would generate a higher piezoelectric output; this phenomenon is usually unlikely to occur in weak gels or hard solid materials. The presence of the double loops suggests that the relaxation rate of the material was lower than the unloading rate, which corresponds to strain-rate-dependent hardening^[21].

The mechanical performance of the (1/2)% gel at different temperatures was also evaluated after 7 d of exposure to air. The work of extension values of the gels at all the considered temperatures was similar ($\sim 0.8 \text{ MJ m}^{-3}$), and Young's modulus decreased as the temperature increased, with values of 15.35 ± 1.58 , 9.28 ± 0.95 , and $9.16 \pm 2.18 \text{ MPa}$ at -20, 25, and 80 °C, respectively [Figure 4C and Supplementary Figure 12]. The compressive strains of the gels at all temperatures ranged from 60% to 80% [Figure 4D], and the compression curve behavior was similar at all temperatures except at 80 °C. In

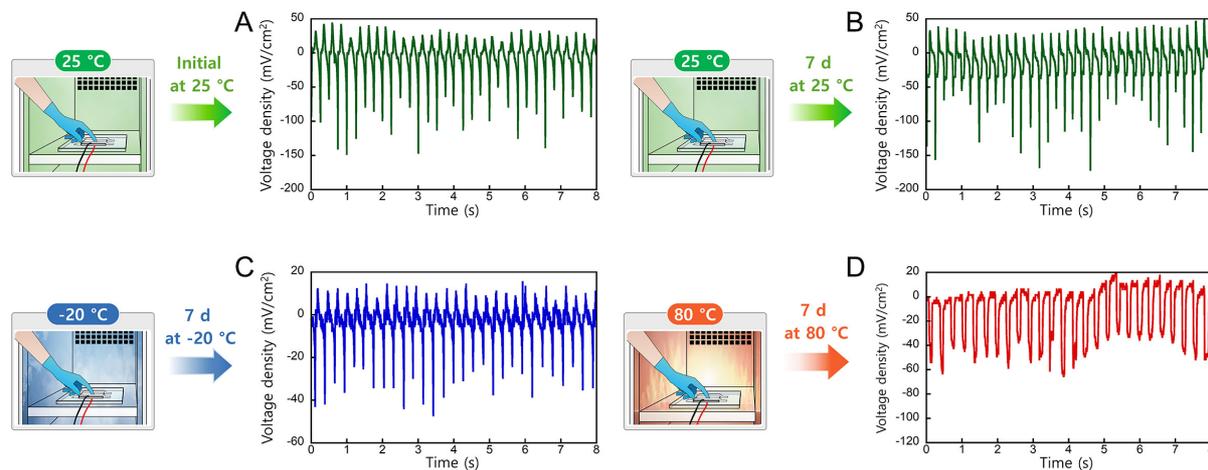


Figure 5. Piezoelectric voltage output in extreme conditions. The piezoelectric voltage output response of the (1/2)% cellulose/PVDF GG subjected to finger tapping: (A) Initial response and response after incubation for 7 d at (B) 25 °C, (C) -20 °C, and (D) 80 °C.

particular, the prolonged exposure (7 d) of the samples to high temperatures (80 °C) weakened the bond strength of the gel matrix, resulting in softer mechanical properties.

Finally, when subjected to finger tapping, the gel's piezoelectric response after 7 d of exposure to air at 25 °C was consistent with the initial values (OCVD $\approx 138.2 \pm 27.6$ mV cm⁻² and SCCD $\approx 146.8 \pm 8$ nA cm⁻²). However, after storage at -20 (80) °C, the OCVD and SCCD values decreased to 50.1 ± 6.2 (63.2 ± 7) mV cm⁻² and 69.1 ± 9 (69.3 ± 8.3) nA cm⁻², respectively [Figure 5A-D, Supplementary Figure 13, and 14; Supplementary Movies 1-3]. Evidently, the variation in the piezoelectric output at extreme temperatures was strongly correlated with changes in the mechanical properties of the gel.

Effects of polymer structure

We anticipated that the mechanical signal transferability of piezoelectric crystals in gel state would be extremely important to achieving a good piezoelectric response. The rigid structure and cooperative crystallization ability of cellulose both help create a robust network channel; this efficiently transfers the input mechanical force and improves the piezoelectric output. To validate our hypothesis we compared our results with another type of PVDF-based hybrid GG, which was fabricated using a relatively flexible polymer, PVA, instead of cellulose. The polymer density and mechanical strength of the best-performing (10/10)% PVA/PVDF GG were similar to that of (1/2)% cellulose/PVDF GG; however, the former GG was significantly less piezoresponsive (4.6 times lower OCVD value) [Supplementary Figure 15]. Both cellulose and PVA chains are rich in hydroxyl groups and easily crystallize through dense hydrogen bond formation^[36,37,39,40]. However, the backbone of cellulose is more rigid than that of PVA^[40]. In the case of the cellulose/PVDF GG, the input mechanical signal can be efficiently transferred through its rigid polymer network; thus, it exhibits much higher piezoelectricity than its PVA-based counterpart.

CONCLUSIONS

We developed a novel piezoresponsive gel by incorporating cellulose and PVDF, which are biocompatible and inexpensive polymers, into glycerol, a green extremotolerant solvent. Although cellulose exhibits relatively poor piezoelectric performance, it serves as a mediator to create highly piezoactive β -phase PVDF crystals and provides good mechanical integrity for an efficient mechanical energy transduction process.

This phenomenon is particularly effective for a relatively rigid polymer, which is more sensitive to a mechanical signal. This effect is aided by the presence of glycerol in the hybrid gel network. Therefore, despite differences in the structure and characteristics, the hybrid gels exhibited excellent piezoelectricity, mechanical properties, and stability over a wide temperature range (-20 to 80 °C). The rationale used in this study could provide insights for developing various high-performance piezoelectric gels through a proper combination of mechanical and piezoactive materials for diverse applications.

DECLARATIONS

Authors' contributions

Contributed significantly to data curation, formal analysis, investigation, methodology, validation, visualization, and writing of the original draft: Mredha, M. T. I.

Contributed significantly to data curation, formal analysis, investigation, methodology, resources, validation, visualization, and writing of the original draft: Wadu, R. R. M.; Varma, A. V. R.; Gupta, T.

Contributed significantly to data curation, formal analysis, investigation, methodology, resources, and writing of the original draft: Li, S.

Provided resources and validation, contributed to conceptualization, investigation, and methodology, and participated in writing the original draft and reviewing and editing the manuscript: Lee, W.

Contributed to investigation, provided resources and supervision, ensured validation, and participated in conceptualization and writing of the original draft and review and editing of the manuscript: Zhang, C.

Performed formal analysis and investigation, provided supervision and validation, and contributed to conceptualization, writing the original draft, and review and editing: Chen, W.

Contributed significantly to conceptualization, formal analysis, funding acquisition, investigation, project administration, supervision, validation, and writing the original draft, as well as reviewing and editing the manuscript: Jeon, I.

Availability of data and materials

The data supporting the findings of this study are available within this article and its [Supplementary Material](#).

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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