



Mechanically anisotropic stretchable and transparent composite substrates for distortion-free display

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ABSTRACT

Developing new-form factor devices has led to the creation of foldable and rollable displays. However, next-step stretchable devices with shape changes face a new issue different from conventional displays. When the screen is stretched, screen distortion occurs due to the nature of contraction perpendicular to the stretching direction to preserve the volume. To address this issue, we focused on the composite approach with mechanical anisotropy using continuously aligned fiber fillers. In this study, we fabricated transparent mechanically anisotropic stretchable substrates with near-zero Poisson's ratio by incorporating continuous and aligned transparent ribbon arrays within a transparent, stretchable matrix. The aligned ribbon-reinforced composite substrates have mechanical anisotropy by mainly reinforcing stiffness only in the ribbon-aligned direction. When unidirectional stretchable devices in the direction perpendicular to the alignment are developed, the stiffness of the substrate contracting in the ribbon alignment direction is relatively high compared to the vertical direction, and thus the vertical displacement is diminishing, so substrates with Poisson's ratio close to 0 can be realized. Based on this approach, we realized a light-emitting device (LED) array system with near-zero vertical distortion by attaching LED arrays and printed intrinsically stretchable interconnections on our mechanically anisotropic composite substrate.

1. Introduction

As display devices become essential elements in human lives, these devices have been developed in various directions. Unlike the development during a decade of trends in which display devices develop their screen size, high resolution, and color gamut, recent developments have been directed towards various form factors, leading to the advent of foldable and rollable type devices [1–5]. The next step of these displays is a new-form factor device that works well even in a place with severe curvature or on stretchy skin [6], and even in actively moving situations, like deformable and kinetic displays. On the other hand, these new-type devices with shape changes face a new problem different from the conventional displays. When the screen is stretched, screen distortion occurs on the screen due to contraction in a perpendicular direction to

the stretching direction [7]. The reason screen distortion occurs is that the elastomers have a large Poisson's ratio ($\nu = -\frac{\epsilon_y}{\epsilon_x}$, ϵ_x : strain of stretching direction, ϵ_y : strain of perpendicular direction of stretching) an intrinsic characteristic of mechanical property that seeks shrinkage in the vertical direction to preserve the volume as it stretches. A novel approach to reducing the Poisson's ratio of the stretchable substrate is required to avoid this phenomenon.

Almost all-natural isotropic substances have a positive Poisson's ratio of 0–0.5 to conserve volume in the in-plane direction. To lower the Poisson's ratio, the intrinsic material property, the researchers have attempted various structural engineering of the material [8,9]. The Poisson's ratio of porous structure material is regulated by porosity, porous shape, and intrinsic material Poisson's ratio [10]. Auxetic structures induced by reentrance shape are used for negative Poisson's

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ratio. In particular, since inwardly protruded reentrant structures isotropically expanded and spontaneously formed concavo-convex structures with a negative Poisson's ratio when stretched, it is known as mechanical metamaterials and various application studies in fields such as biomedical, sensors, actuators [9,10], and even stretchable supercapacitors [11] and stretchable lithium-ion batteries [12] are being conducted. However, among these studies using structural mechanics to control the Poisson's ratio, there are few studies related to the distortion of the display devices.

Recently, as stretchable transparent electrodes [13–21] and stretchable light-emitting devices have been developed in a direction that remains stable and does not lose electrical properties even under mechanical deformation [22–24] being developed in academia, concerns about the vertical distortion issue of the stretchable display are starting. The stretchable electroluminescence system with a biaxial micro-wrinkle structure has somewhat relieved vertical distortions as the wrinkle structure is independently unfolded, responding to the tension, but new distortions occur due to their wrinkle structure [22]. Also, a folded auxetic structure was applied to a Kirigami-based flexible circuit board with micro-LED arrays to fabricate a distortion-controlled stretchable display, but the substrate itself was not inherently stretchable, which limited its applicability [7]. Therefore, it is necessary to develop an intrinsically stretchable and transparent substrate that provides the stretchability of a stretchable display and efficiently prevents vertical distortion.

In this study, to fabricate intrinsically stretchable substrates without vertical distortion, we propose a composite approach with mechanical anisotropy using continuously aligned fiber fillers. The continuously aligned fiber-reinforced composites have mechanical anisotropy in which the aligned fibers selectively reinforce the stiffness only in an aligned direction. When transverse tension having a low modulus is applied to the mechanically anisotropic stretchable substrate, it stretches relatively well, but since the vertical (filler alignment direction) modulus of the tension is greatly increased, vertical contraction can significantly reduce. For this reason, the mechanical anisotropy method can be an effective composite-based approach to prevent vertical distortion of stretchable display devices. We fabricated a transparent mechanically anisotropic stretchable substrate with near-zero Poisson's ratio by incorporating continuous and aligned transparent ribbon arrays within a transparent, stretchable matrix. Based on this approach, we realized a light-emitting device (LED) array system with near-zero vertical distortion by attached LED arrays and printed intrinsically stretchable interconnections on our mechanically anisotropic composite substrate. In the future, display trends are shifting

towards moveable displays such as rollable displays. However, in reflecting this trend, rollable displays encounter stretching in one direction accompanied by distortion issues. These distortion issues can be resolved by our fabricated ribbon-aligned composite. The ribbon-aligned composites, having a zero Poisson's ratio in the stretching direction, allow for an accurate and undistorted display.

2. Results and discussion

The severe vertical contracted deformation of the stretchable display on typical stretchable substrates can be seen in Fig. 1 a and b. To prevent this contracted distortion of the stretchable substrate, a composite structure in which unidirectionally aligned continuous filler bundles are incorporated into the stretchable substrate is introduced, as shown in Fig. 1 c and d. For a continuously and unidirectionally oriented fiber-reinforced composite material, the composite moduli in longitudinal (E_{cl}) and transverse (E_{ct}) directions can theoretically be calculated as follows (Fig. 1(d)) [25],

$$E_{cl} = E_m V_m + E_f V_f \quad (1)$$

$$E_{ct} = \frac{E_m E_f}{E_m V_f + E_f V_m} \quad (2)$$

, where E and V denote the elastic modulus and volume fraction, whereas the subscripts m and f represent matrix and fiber phases. Since the modulus of elasticity in the longitudinal direction of the composite has the average arithmetic value of the fiber and the matrix, and the modulus of the composite in the transverse direction has the harmonic average, the composite can exhibit mechanical anisotropy when the modulus difference between the two materials is considerable. Suppose polyethylene terephthalate (PET) with a modulus of ~ 3 GPa or polypropylene (PP) with a modulus of ~ 0.3 GPa and polyurethane (PU) with a modulus of ~ 3 MPa, which will be described later, is used as a fiber filler and an elastic matrix. In that case, the anisotropic ratio ideally reaches 48.5 (PET) and 5.6 (PP) of times even if only 5 vol% of the aligned fibers are included. The resulting mechanical anisotropy can be used to fabricate displays that are not vertically contracted when stretched in one direction.

Before the fabrication of a mechanically anisotropic and stretchable composite substrate, we executed a stress analysis simulation based on the finite element method to optimize the design of the internal structure of the substrate for mechanical stability against 1D tensile deformation. Composite materials containing fillers such as fibers exhibit superior mechanical properties compared to single materials, but due to the

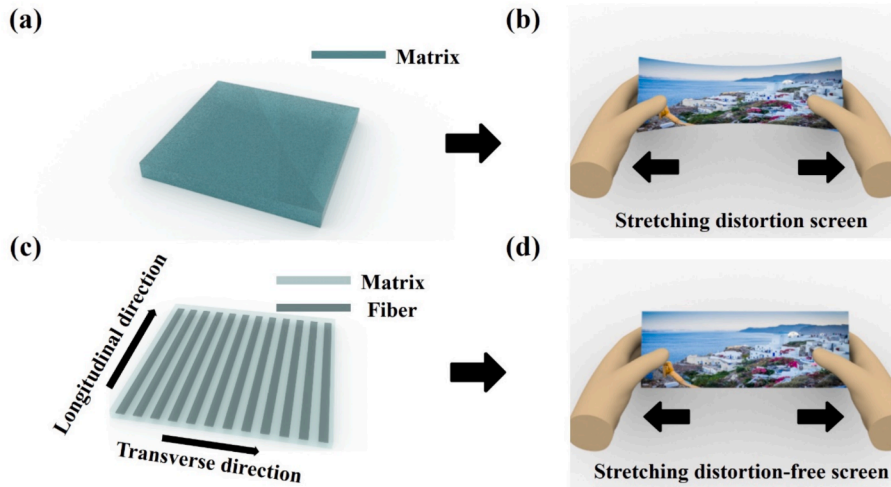


Fig. 1. (a) Bare transparent stretchable substrate (b) Stretching behavior stretchable display on bare stretchable substrate (c) The ribbon-aligned composite substrate to prevent the display distortion. (d) Stretching behavior stretchable display on ribbon-aligned composite substrate.

structural issue, local stress concentration may occur at the interface of different materials when mechanical strain is applied. To enable concentrated stress relief through structural control, a numerical analysis was performed to determine the effect on the spacing between fillers within the composite. In the simulation, the stress distribution of a fiber-aligned composite containing fibers (modulus ~ 3 GPa) of 1 mm diameter interspersed with an elastomer (modulus ~ 3 MPa) of 2 mm thickness under a 50 % strain can be changed by varying the spacing between the fibers. Closely contacted fibers (spacing ~ 0 mm) in the matrix (Fig. 2a) show the maximum stress was 880 MPa and the minimum stress was 0.84 kPa. On the other hand, when the spacing between the fibers was set to 0.5 mm in Fig. 2b, the maximum stress was significantly decreased to 2.5 MPa and the minimum stress was 8.8 kPa. As the spacing increases, the difference between the maximum and minimum stresses decreases, and the stress could be well distributed. In particular, if stress is concentrated at the interface of the composite, delamination and failure can occur at the interface between the matrix and the filler, so it is necessary to secure an appropriate spacing between fibers so that the stress can be well distributed.

In addition, the effect on the strain variation at the thickness direction was investigated depending on the cross-sectional shape of the aligned fillers (Fig. 2c, d). When circular fibers were used, vertical deformation caused undulation along the circular curved surface, which can cause problems in integration with non-deformable planar devices.

For this purpose, we introduced a ribbon filler with a wide rectangular cross-section. In the simulation, when the composites were stretched by 50 %, the vertical strain could be differently applied on the filler and no-filler region, and vertical undulation could be formed along the interface with the fillers. In particular, in the case of cylindrical fillers, the interfaces are curved, so the neutral deformation area is inevitably narrowed. However, when thin, flat ribbon fillers were incorporated, the neutral strain region along the thin ribbon could form much wider than the strain of the cylindrical filler composite, so there is no difficulty in integrating existing non-stretchable thin film devices. In the same way as cylindrical fillers, ribbon fillers are also affected by the spacing between the fillers, with closer spacing leading to increase stress concentration and delamination. To prevent this, a spacing of 0.5 mm should be maintained. In addition, since it is necessary to manufacture the substrate thinner for the same performance, it is more advantageous to use a long ribbon-shaped filler with a rectangular cross-section instead of a filler with a fibrous circular cross-section. Furthermore, rather than a process such as pultrusion that impregnates fibers in the molten matrix, the lamination process after cutting the ribbon from the film has the advantage of producing a faster and more uniform composite film. Based on these results, as shown in Fig. 2(e), stretchable composite films with ribbon-shaped fillers were fabricated with a film thickness of 2 mm, a fiber spacing of 0.5 mm, and a ribbon width of 2.5 mm.

To confirm whether this aligned composite structure can provide

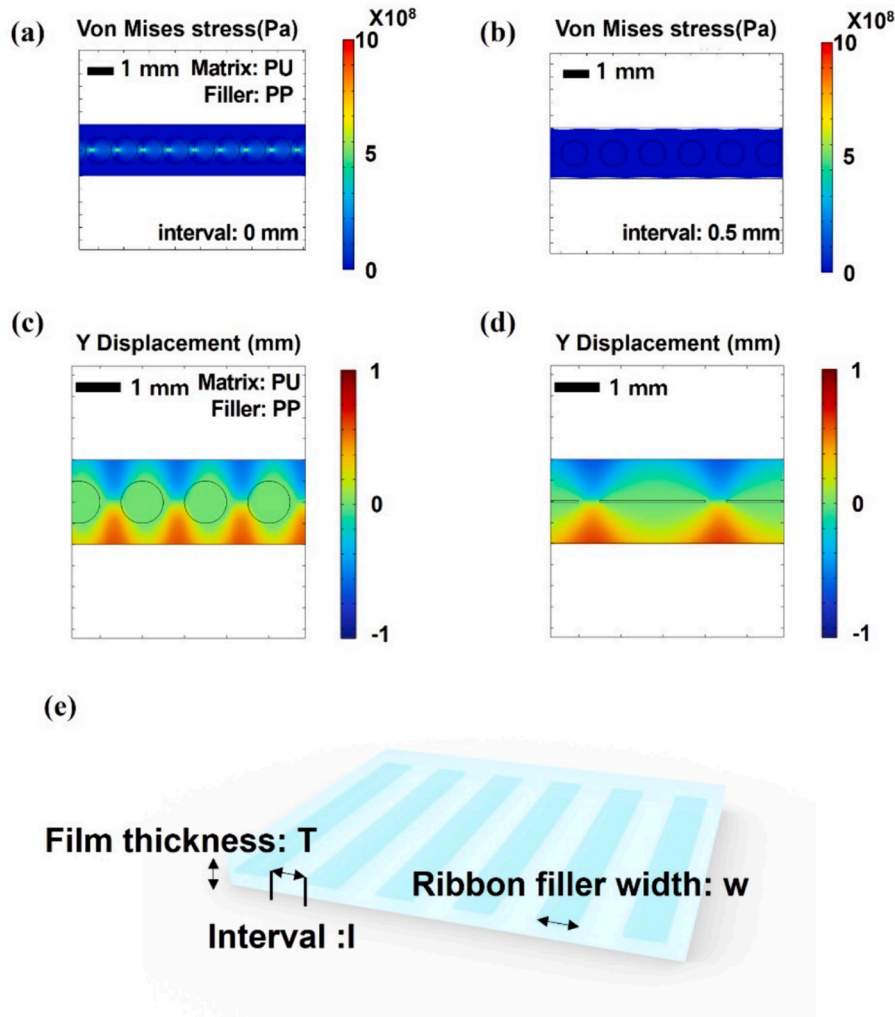


Fig. 2. (a and b) stress analysis simulation under uniaxial 50 % tensile strain stretching /stress simulation in different filler intervals of (a) 0 mm and (b) 0.5 mm. (filler radius: 0.5 mm, matrix $80 \times 80 \times 2$ mm³) (c and d) stress analysis simulation under uniaxial 50 % tensile strain stretching/stress simulation in different filler shape (c) fiber and (d) ribbon. (e) dimension of the ribbon-aligned substrate.

macroscopic mechanical anisotropy, the mechanical properties of various aligned composites in longitudinal and transverse directions were measured through tensile testing using a universal testing machine (UTM), as shown in Fig. 3a. We determined that zero Poisson's ratio could be obtained when the mechanical anisotropy was greater than 10 times. A difference in Young's modulus of more than 10 times can cause a strain rate of 1/10, so Poisson's ratio, which is close to 0.5, can be lowered to less than 0.1. Thus, the samples to be made were prepared with the following materials and conditions. We prepared 5 aligned composites by inserting unidirectionally aligned ribbon arrays with constant width (~ 2.5 mm) and spacing (~ 0.5 mm) but different thicknesses and materials (250 μm and 200 μm of polyethylene terephthalate (PET) ribbons ($E_{\text{PET}} \sim 4.5$ GPa), 40 μm of tri-acetyl cellulose (TAC) ribbons ($E_{\text{TAC}} \sim 4.3$ GPa), 40 μm and 20 μm of PP ribbons ($E_{\text{PP}} \sim 2.5$ GPa), respectively) into the PU matrix ($E_{\text{PU}} \sim 1.16$ MPa, Figure S1 in Supplementary Information) of the thickness of 2 mm. The volume fractions of the 250 μm and 200 μm PET ribbons in the composites are about $\sim 9.4\%$ and $\sim 7.5\%$, and those of the 40 μm TAC ribbon and the 40 μm and 20 μm PP ribbon are $\sim 3\%$, $\sim 3\%$ and $\sim 1.5\%$, respectively. Based on the above equations and the material properties, the mechanical moduli of the ideal anisotropic composite can be calculated as shown in Table 1. When thick and stiff PET ribbon arrays were inserted, high moduli in the orientation direction and very high mechanical anisotropy with a moduli ratio of 330 and 270 between the longitudinal and transverse directions could be expected. Even in the case of a thin yet rigid TAC ribbon array composite, high mechanical anisotropy of more than 100 times can be expected. In the case of the composite impregnated with a thin and relatively soft PP ribbon arrays, the moduli in the orientation direction did not increase significantly compared to the previous composites, but the mechanical anisotropy ratio could still be as high as 63 and 33 times.

For comparison with predicted mechanical properties, experimental stress-strain curves of 5 ribbon-aligned composites in the longitudinal direction and in the transverse-direction were obtained in Fig. 3b-f. All five stress-strain curves show a large difference in the slope between the longitudinal and transverse directions. As thinner and softer fillers are inserted, the slope in the longitudinal direction decreases, while the slope in the transverse direction has similar values for all five samples.

Table 1

Measured parallel and vertical directions of elastic modulus of aligned ribbon array-incorporated composites with different thicknesses and materials of ribbons at 3 % tensile strain using a UTM machine.

Materials	PET		TAC	PP	
Thickness	250 μm	200 μm	40 μm	40 μm	20 μm
Calculated longitudinal modulus (E_{cl})	424 MPa	339 MPa	130 MPa	76 MPa	39 MPa
Calculated transverse modulus (E_{ct})	1.28 MPa	1.25 MPa	1.20 MPa	1.20 MPa	1.18 MPa
Measured longitudinal modulus (E_{cl})	58.7 MPa (± 7.2)	49.6 MPa (± 5.3)	25.5 MPa (± 2.4)	12.9 MPa (± 3.9)	6.5 MPa (± 1.6)
Measured transverse modulus (E_{ct})	1.3 MPa (± 0.3)	1.6 MPa (± 0.4)	2.0 MPa (± 0.2)	2.4 MPa (± 0.4)	2.1 MPa (± 0.1)

Based on slopes up to 3 % of the curves (yield strains of the non-stretchable ribbons), the longitudinal and transverse moduli were obtained and are shown in Table 1. We measured the Young's modulus at 3 % strain because this is the strain at which the yield strength of the internal fillers occurs. The overall longitudinal moduli were smaller than expected but tended to increase for thicker and stiffer ribbons, indicating that the orientation of the ribbon array may not be perfect. Additionally, the vertical modulus did not differ between samples, as expected, but was slightly increased for thinner and softer ribbons, which is believed to be due to the partial separation of the ribbons from the matrix in the tensioning transverse direction in the case of composites with large modulus differences. Based on these results, the aligned ribbon array composites were confirmed to have high mechanical anisotropies and wide anisotropy ratios of 45, 31, 13, 5.4, and 3.1 times, respectively, in experimental tensile tests by controlling the ribbon thicknesses and materials. To examine the long-term stability of the substrate, a mechanical cyclic tensile strength of the composite substrate was executed in Figure S2 in Supporting Information.

This ribbon-aligned composite with mechanical anisotropy also

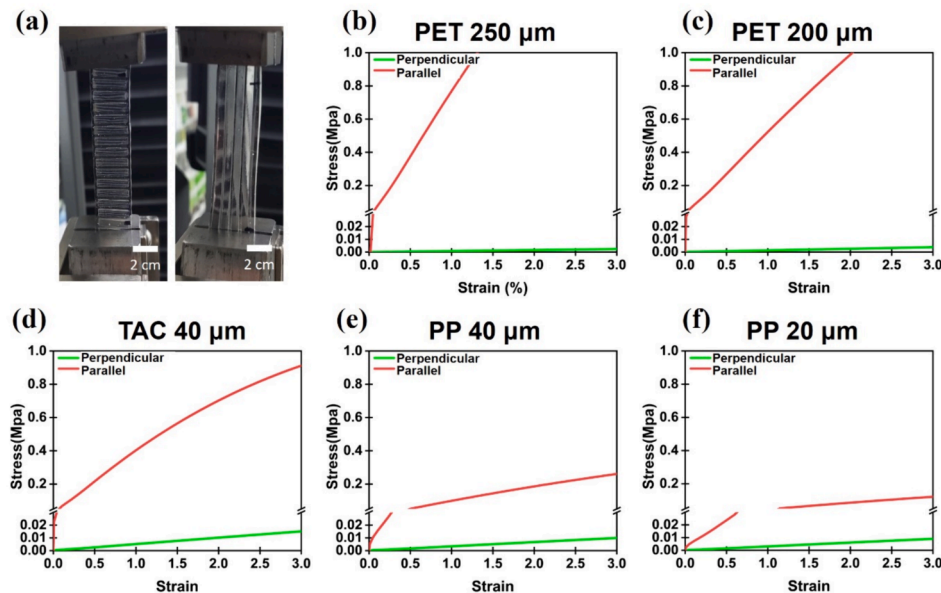


Fig. 3. (a) Photographic images of the UTM tensile test of aligned ribbon (aluminum coated for visibility of ribbon filler) incorporated PU composites in direction parallel and perpendicular to the aligned ribbon direction. (b-f) Stress-strain curves of PU/ribbon composite with (b) 250 μm -thick PET, (c) 200 μm -thick PET, (d) 40 μm -thick TAC, (e) 40 μm -thick PP, and (f) 20 μm -thick PP ribbons embedded 2 mm-thick PU matrix. The graphs above include an omission of part of the stress on the Y-axis. This omission was made to show the difference in the stress-strain curves of the composites based on their alignment. By doing so, we were able to comprehensively display the stress-strain curve of the parallel direction, which has higher stress, in comparison to the perpendicular direction.

possesses transparency. Its transparency is maintained by utilizing highly transparent materials such as PU, PET, PP, and TAC film, and combining them while matching their refractive indices. (Refractive index: PU 1.48822, PET 1.570, PP 1.480, TAC 1.49) Fig. 4a and b demonstrate that the composite, fabricated in this method, remains transparent to the naked eye. When transparency was measured, all composites achieved over 90 % transparency across the entire visible light spectrum. Furthermore, it was observed that the transparency at a wavelength of 550 nm, which serves as an indicator of transparency, also approached 95 %. These experimental results indicate that a substrate has been developed that both preserve transparency and maintain mechanical properties by matching the refractive indices. However, as shown in Fig. 4b, there is a visible disadvantage in that under tensioned states, the deformation between the ribbon and the non-ribbon area is different, resulting in undulation in the thickness direction on the surface.

To investigate the effect of mechanical anisotropy on the, we obtained the Poisson's ratios of the ribbon-aligned composites with various mechanical anisotropy ratios by measuring the vertical strain at various strains up to 50 % in direction normal to the aligned ribbons, as shown in Fig. 5a and b. While the pristine PU substrate had a high Poisson's ratio of 0.35 or more in all tested strains up to 50 %, the ribbon-aligned composite substrates all have significantly lower Poisson's ratios. In particular, in the cases of 250 and 200 μm -thick PET ribbon substrates with very high anisotropy ratios of 45 and 31, it was confirmed to have a Poisson's ratio of less than 0.03 over a wide strain range, and in the case of a 40 μm -thick TAC ribbon substrate with a high anisotropy ratio of 13, it was confirmed that it consistently had a Poisson's ratio of less than 0.05. Even in the case of PP ribbon composites with low anisotropy ratios, 40 μm -thick ribbon substrate with a ratio of 5.4 times had a Poisson's ratio as low as 0.07 in most strain areas, while 20 μm -thick ribbon substrate shows a relatively high Poisson's ratio of 0.17 on average due to the low anisotropy ratio of 3.1. Based on these results, it was confirmed that as the anisotropy increases, the Poisson's ratio decreases closer to 0, and that there is no significant difference above a certain anisotropy of approximately 10 or more. In addition, since the

Poisson's ratio can be different between the part on the ribbon and the part without the ribbon in the thickness direction, each part of the 40 μm -thick PP ribbon substrate was measured separately (Fig. 5c). The Poisson's ratios of both the ribbon and non-ribbon areas were sufficiently lower than that of the original PU matrix, and the ratio difference was about 0.02, which was not a large difference, confirming that the Poisson's ratio was lowered overall in the substrate.

To assess the x-axis (vertical) local strain on the substrate in real-time and the actual behavior of the substrate along with local deformations, we investigate the local strains by performing simulations and digital image correlation (DIC) measurements until the substrate is stretched up to 50 % (Fig. 5d-g). In the vertical strain simulation, while pristine PU substrates can experience significant vertical strain in most areas except the clamped edges, aligned composites show local vertical strain close to zero over the entire area, indicating that the substrate remains distortion-free at most points. In actual strain measurements obtained through DIC equipment to confirm these simulation results, pristine PU substrate shows significant vertical strains in most areas with slightly weak vertical deformation near the clamp. On the other hand, our aligned ribbon composite in the DIC experiment also shows a vertical strain value close to 0 in most areas except the edge. However, because the influence of the aligned ribbon filler is weakened at the end of the ribbon filler in the composite, we observed that the vertical strain was slightly higher than 0 near the edges. Through this measurement, we confirmed that our aligned composite substrate can demonstrate distortion-free properties with a vertical strain close to 0 even under up to 50 % of tensile strains, which is the same as the theoretical expectation.

To demonstrate that this distortion-free substrate can be used as an ideal substrate for practical stretchable displays, a stretchable display was fabricated by attaching 16 mini-LEDs in a 4×4 arrangement 5 mm apart from each other on pristine PU substrate and the 40 μm PP ribbon-aligned composite substrate and connecting each LED with stretchable interconnection made from EGaIn liquid metal, as schematically shown in Fig. 6a [26]. The mini-LED arrays fabricated on the two stretchable substrates were turned on and stretched to 50 % to check whether

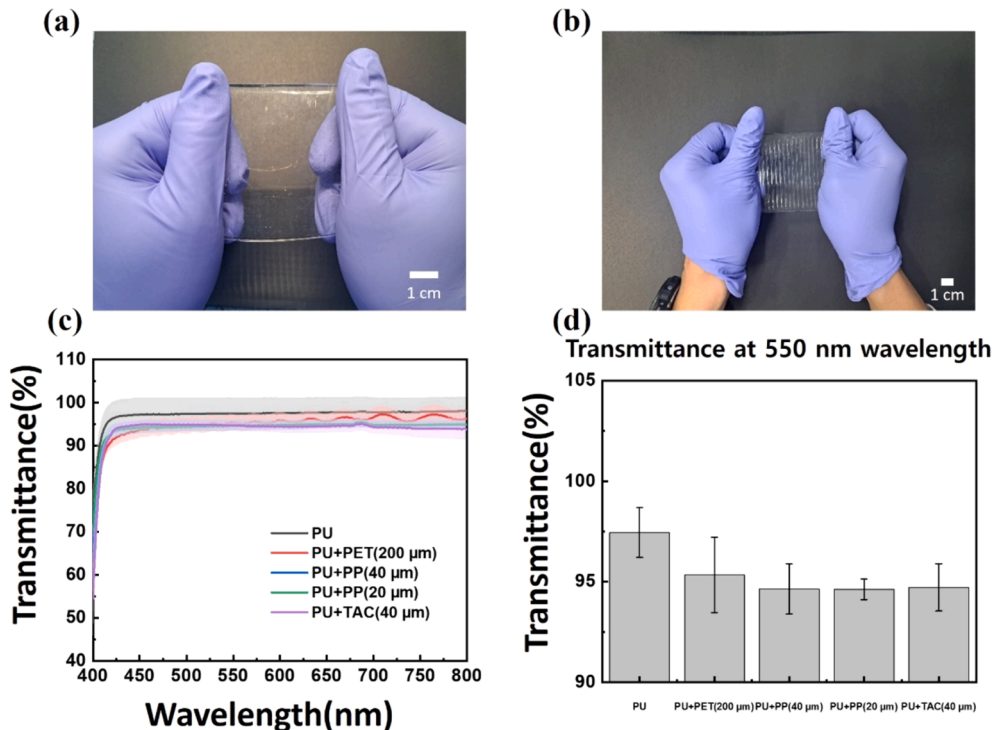


Fig. 4. (a) Before stretching the transparent ribbon-aligned stretchable PU + PP 40 μm composite substrate (b) After stretching the transparent ribbon-aligned stretchable PU + PP 40 μm composite substrate (c) at UV-visible wavelength scale (d) at 550 nm wavelength.

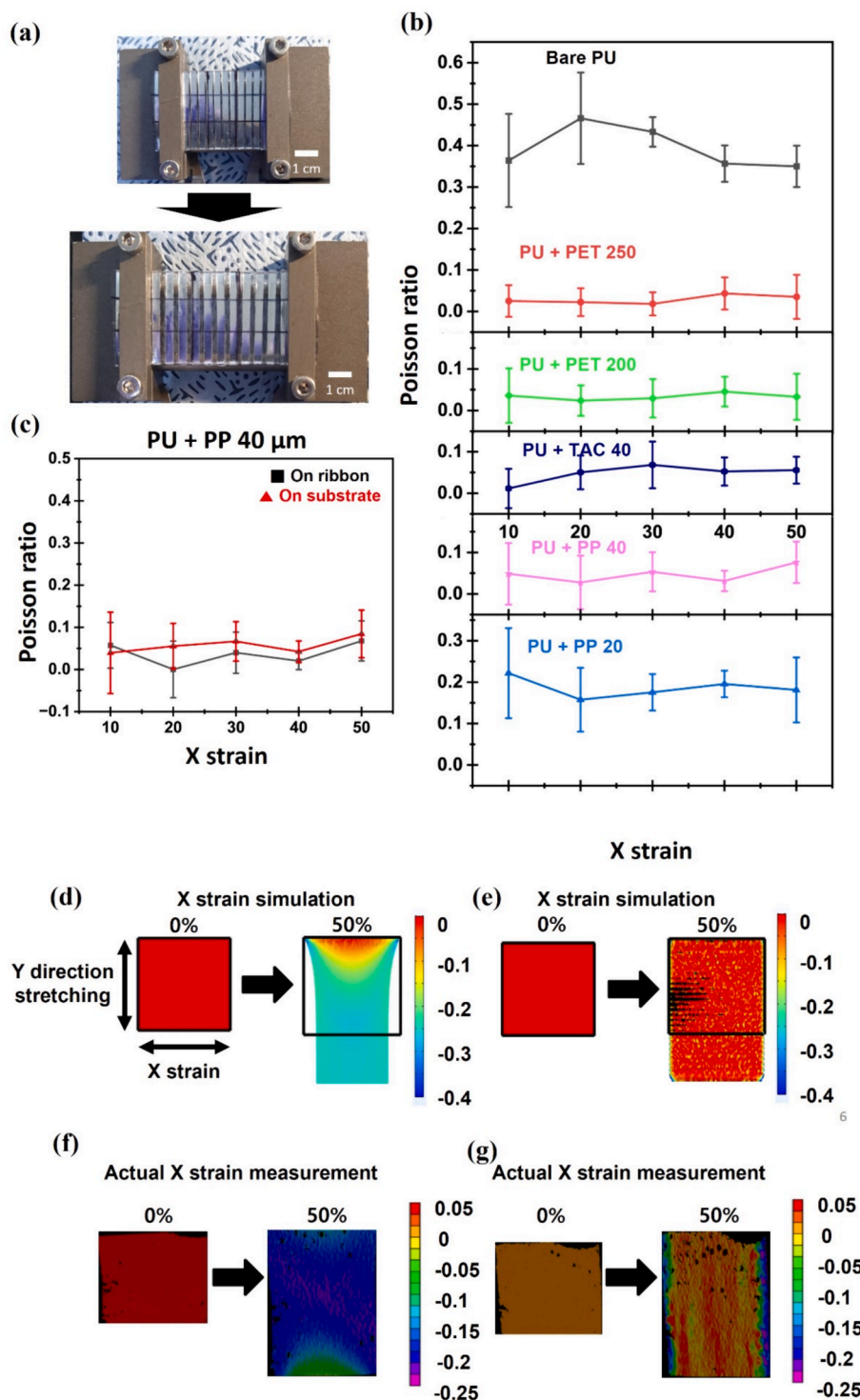


Fig. 5. (a) Photographic image of tensile testing of vertically aligned ribbon arrays in PU matrix to measure Poisson's ratios. (b) Average strain-Poisson's ratio at from 10 % to 50 % strains of pristine PU, 250 μm -thick PET, 200 μm -thick PET, 40 μm -thick TAC, 40 μm -thick PP, and 20 μm -thick PP ribbon incorporated PU composites. (c) Poisson's ratios of 40 μm -thick PP ribbon composite at the ribbon area and non-ribbon area of the substrate. (d and e) x-direction (vertical) strain simulations of (d) pristine PU and (e) 40 μm -thick PP ribbon composite at 0 % and 50 % of y-direction tensile strain. (f and g) x-direction (vertical) strains from digital image correlation (DIC) measurement of (f) pristine PU substrate and (g) 40 μm -thick PP ribbon arrays in PU matrix under 0 % and 50 % uniaxial tensile strain.

successfully prevented vertical distortion in the real display. Fig. 6b shows that the square-shaped LED array on the pristine PU substrate was distorted by vertical contraction at the center of the square under 50 % of tensile strain due to the high Poisson's ratio. Also, the distances between LEDs in 4×4 arrays were reduced average of 27.1 % at 50 % strain. The stretchable LED device was quite unstable from frequent

contact failure with the deformed substrate. However, the aligned ribbon composite substrate showed almost no distortion of the square LED array, only the length in the stretching direction increased monotonically, and the vertical distance between LEDs in the square array decreased to about 2.8 % which was difficult to discern with the eye, as shown in Fig. 6c. In addition, by attaching the LEDs on the ribbon area

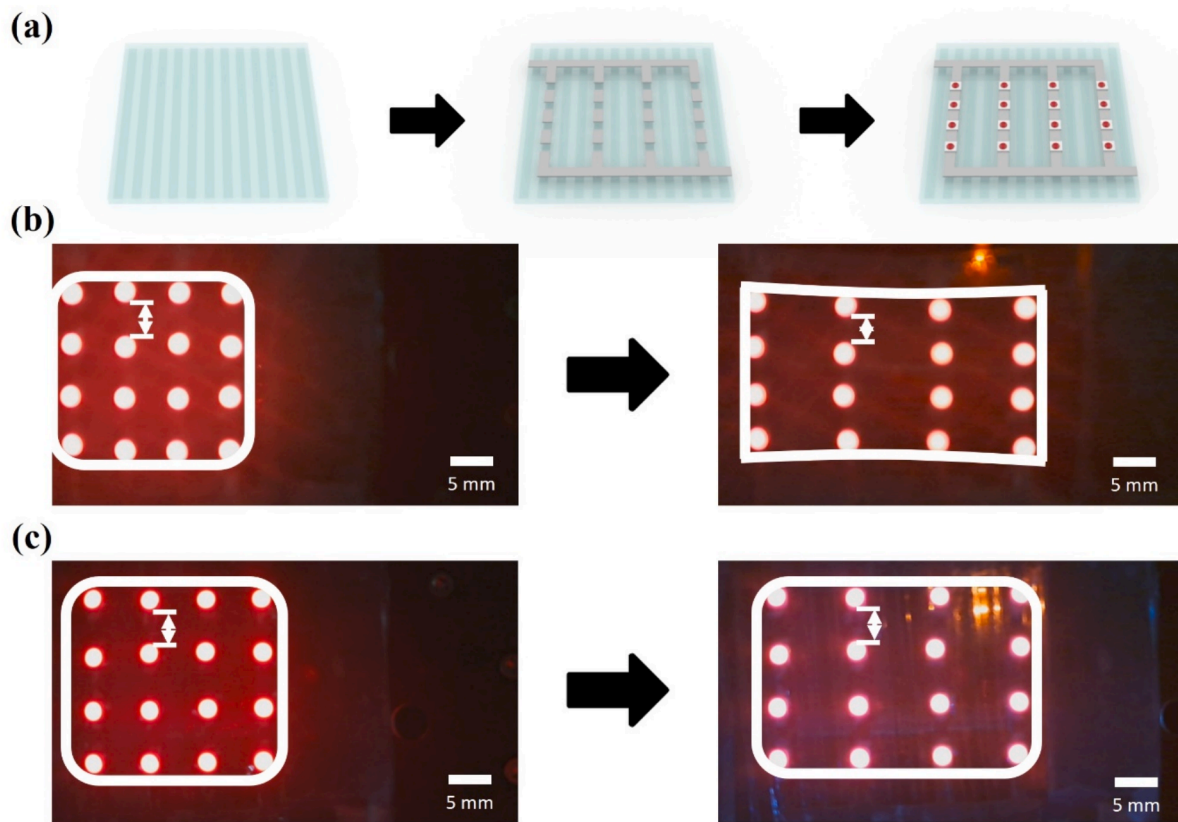


Fig. 6. (a) Fabricating process the mini-LED 4×4 array on 8 cm substrate. (b) 50 % stretching behavior LED 4×4 array on pristine PU bare substrate and (c) 50 % stretching behavior LED 4×4 array on PP 40 μm ribbon embedded composite substrate.

with a relatively small strain in the tensile direction, stable attachment of the LED was maintained even under tension of more than 50 %, confirming that it works well as a stretchable display device.

3. Conclusion

In the field of stretchable devices, where much research has been actively conducted recently, consideration of mechanical properties and strain analysis related to the deformation of the device, which had previously been overlooked, has become very important. The mechanical properties of the deformable substrates on which these devices are mounted have also become important. Conventional stretchable materials exhibit high Poisson's ratios to minimize volume changes, resulting in varying rates of vertical deformation with position and thus distortion in display applications. We have suggested a mechanically anisotropic, ribbon-aligned composite for a distortion-free stretchable substrate that exhibits high stiffness in the direction parallel to the ribbon and low stiffness perpendicular to the ribbon direction. By changing the modulus and thickness of the aligned ribbon fillers, the mechanical anisotropy of the composite could be tuned without compromising transparency. This mechanical anisotropy can reduce vertical contraction because when stretched in one direction, the stiffness in only the shrinkage direction is strengthened. Through this approach, we were able to implement a stretchable transparent substrate that shows little vertical deformation when stretched, and by attaching a square-arranged LED array on the substrate. We demonstrated a vertical zero Poisson's ratio stretchable display under uniaxial strain. Based on this approach, we think that this aligned composite substrate can be practically used as a deformable substrate for rollable displays with a very small radius of curvature that requires deformation in one direction or stretchable displays and since it is a process using a roll-to-roll process or a pultrusion process, substrates can be mass-produced without the need to build new existing

infrastructure. Furthermore, in that we have demonstrated that control of vertical deformation through mechanical anisotropy is possible in stretchable device technology, we believe that we have presented a new approach to the realization of form factor-free configuration of stretchable or deformable devices.

4. Material and method

Material. Transparent thermo-curable polyurethane (PU) elastomer (ClearFlex™ 30) was purchased from Smooth-On, Inc. Polyethylene terephthalate (PET) 250 μm and 200 μm thicknesses films or ribbons were obtained from UB Light Co, Ltd. Tri-acetyl cellulose (TAC) 40 μm thickness films were obtained from Hyosung Vina Chemicals Co., Ltd. (Seoul, Korea). 40 μm and 20 μm thick polypropylene films were obtained from YoulChon Chemical Co., Ltd. (Seoul, Korea). Acetone (99.5 %) was purchased from Daejung Chemical & Metal Co., Ltd (Seoul, Korea). Eutectic Gallium Indium alloy (EGaIn, Gallium 75.5 %, In 24.5 %, 99.99 %) was purchased from Sigma-Aldrich.

Fabricate pristine PU substrate. The prepolymer and cross-link agents of PU (1:1 w/w) were mixed by a paste mixer for 4 min and poured into a mold that made the PU substrate 2 mm thick. The PU in the mold was vacuumed for 1 h at room temperature. After that, the PU was cured for 2 h at 80 $^{\circ}\text{C}$.

Fabricate PU ribbon-aligned composite. The sheet was cut into ribbon shape patterns (width: 2.5 mm interval: 0.5 mm) by a cutting machine (Cameo 3, Silhouette America). The cut sheet had O_2 plasma treatment (100 W, 100 sccm for 30 s, VITA 8, FemtoScience Inc.) to improve adhesion between the ribbon and PU. After plasma treatment, the ribbon arrays were fixed below the mold and the PU mixture (1:1 w/w) was poured into the mold, making the PU substrate 1 mm thick. The PU in the mold was placed in a vacuum chamber for 1 h at room temperature. After that, the PU was cured for 2 h at 80 $^{\circ}\text{C}$. After curing PU 1

mm substrate and ribbon arrays, we reversely located the cured PU and ribbon arrays in the 2 mm mold. Then, the PU mixture (1:1 w/w) was poured for 2 mm thickness and cured at 80 °C for 2 hr. (Figure S3 in Supporting Information).

FEM simulation. FEM analysis was used to simulate the mechanical behavior by COMSOL Multiphysics 6.0. The design of the substrate sample is $4 \times 4 \text{ mm}^2$ and thickness is 2 mm. The fibers and ribbons were designed the same way as the fabricated sample. Mechanical deformation and induced Von Mises stress, principal strain, and displacement as a function of imposed displacement or stretching were mapped using the “Solid Mechanics” module. Elastic modulus and Poisson’s ratio of PP and PU were taken to be 2.4 GPa and 2.0 MPa, 0.24 and 0.49, respectively. Also, we used a model that uses two parameters among the Mooney Rivlin model to elastomer, which is one of the hyper-elastic models (C_{10} : 145090 Pa, C_{11} : 20850 Pa).

Mechanical stretching test. The tensile test samples were prepared by cutting rectangle shapes $2 \times 8 \text{ cm}^2$ respectively and five identical samples per composite with different ribbon thicknesses were fabricated. To do the tensile test, we used the universal testing machine (UTM, 5567, Instron). The stretching rate is 12 cm min^{-1} . The Poisson’s ratio samples were prepared by cutting rectangle shape $3 \times 6 \text{ cm}^2$. And the sample was stretched by stretching equipment (lab-made) that measured the Poisson’s ratio. The stretched samples were taken at 10 %, 20 %, 30 %, 40 %, and 50 % respectively to measure the Poisson’s ratio by using an image analysis program (Image J). In Poisson’s ratio measurement experiment, at least five samples were prepared for each composite and measured the Poisson’s ratio. The Digital Image Correlation (DIC) samples were prepared by cutting rectangle shape $3 \times 6 \text{ cm}^2$. The corresponding x strain and y strain were obtained by using DIC software and VIC-2D (from correlated SOLUTIONS) [27].

Transmittance measurement. To do the transmittance measurement, the transmittance spectra of wavelengths were collected using an ultraviolet–visible–near-infrared spectrometer (V-600, Jasco).

Fabricate 4×4 array LED display. To fabricate the square arranged 4×4 LEDs (width: 2.8 mm, length: 3.5 mm, height: 1.9 mm, ASMM-CR03-AS402, Broadcom) array for stretchable display, the stretchable PU + EGaIn composite electrode was fabricated. First, to synthesize LM microparticles (LMPs), 30 mL of acetone was placed in a glass vial, and 1.35 g of LM was injected into the vial. A probe sonicator was then applied with a 63 μm amplitude (44 %) for 20 min in a water bath at room temperature to generate LM microdroplets. LM droplets were isolated by centrifugation using a centrifugal machine for 30 min at 3000 rpm. The solvent was then carefully discarded. The PU (1:1 w/w) was added to the vial and mixed by a paste mixer. Once composite ink was mixed, it was patterned onto the substrates by a screen-printing process. The mask was made by a cutting machine. After patterned, the patterned polymer composite was cured for 2 h at 80 °C. After the drying process, the film was fixed on the bath with scotch tape, and the bath was filled with water. An acoustic field was applied to the printed composite using a probe sonicator with a 43 μm amplitude (30 %) for 30 s. [26] The LEDs were placed on the pattern and fixed by silver paste (CANS ELCOAT, P-100). The fabricated sample was stretched until 50 % strain at a rate of 1.8 mm min^{-1} by stretching machine (lab-made equipment) (Figure S4. in Supporting Information).

CRedit authorship contribution statement

Hyeongsu Oh: Writing – original draft, Methodology, Investigation. **Jung Hur:** Investigation, Conceptualization. **Soojin Jeong:** Methodology, Investigation. **Seung-hwan Heo:** Investigation. **Dong-chun Lee:** Supervision, Funding acquisition. **Yongtaek Hong:** Supervision, Project administration. **Seungjun Chung:** Supervision, Project administration, Funding acquisition. **Jonghui Lee:** Supervision, Project administration, Funding acquisition. **Jeong Gon Son:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jeong Gon Son reports financial support was provided by Korea Institute of Science and Technology. Jeong Gon Son reports financial support was provided by National Research Foundation of Korea. Jeong Gon Son reports a relationship with Korea Institute of Science and Technology that includes: employment. Jeong Gon Son has patent #KR 10-2299529 licensed to Jeong Gon Son. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.compositesa.2024.108338>.

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