

Large-area photo-patterning of initially conductive EGaIn particle-assembled film for soft electronics

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Eutectic-gallium-indium particle (EGaIn*) is considered one of the promising conducting materials for soft electronics due to its enhanced stability compare to bulk EGaIn and constant conductance under strain. However, its practical implementation has thus far been limited due to the challenges of achieving initial electrical conductivity and the incompatibility with well-developed fabrication strategies. Here, we report materials and manufacturing methods that allow large-area multi-layered patterning of 'polystyrene sulfonate (PSS)-attached EGaIn* (EGaIn*:PSS)' thin-film with the conventional cleanroom process. PSS enhances the stability of EGaIn*, which allow uniform thin-film coating and photographic lift-off at a wafer-scale down to 10 μ m features of varying thicknesses. Using dimethyl sulfoxide as the solvent during lift-off induces cohesion between EGaIn*:PSS, resulting in initial electrical conductivity without an additional activation process. Demonstrations of stretchable display, multilayer pressure sensing systems, and soft artificial finger validate the versatility and reliability of this manufacturing strategy for soft electronics.

Keywords: Liquid metal particles; Photo-patterning; Solution process; Multiscale/layer fabrication; Soft electronics

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With the rising demand for next-generation wearable healthcare devices, soft robotics, and conformable implantable devices, soft electronics are increasingly becoming of pivotal importance [1–4]. To enable the aforementioned applications, it is critical that the fabrication method be compatible with the well-established

standard photolithographic process so that high-resolution, large-area, and high-performance devices can be made [5]. The critical component that imparts softness to electronics are the stretchable electrodes and interconnects, and there are two main ways to fabricate them [6]. One method is to fabricate serpentine or wavy structures with standard metallic materials [7,8], and the other is to use stretchable materials such as conductive carbon composites [9,10], metal nanowire composites [11], or conductive polymers [12]. In the former case, high-density integration is difficult due to the extra space required by the geometric design, and the stretchability of electronics is limited due to the relatively small fracture strain of metallic materials. In the latter case, the stretchable materials either lack sufficient conductivity and/or are incompatible with the conventional photolithographic process.

Eutectic-gallium-indium (EGaIn) has recently drawn a great deal of attention as it can potentially overcome the aforementioned limitations, owing to its exceptional deformability and stretchability, and high conductivity comparable to that of metals [13–15]. However, its fluidity (since it is in liquid state), ultrahigh surface tension, immediate formation of oxide layer, and conductance change under strain render it challenging for integration and use as interconnects or direct contact pads [16]. For instance, Brand et al. demonstrated bulk EGaIn patterning with electron beam lithography [17]; however, this technique requires a specific metal adhesion layer for transferring EGaIn, and due to its liquid state, direct use as electrodes and multilayer integration are challenging.

To cope with this matter, EGaIn particles (EGaIn*) have been investigated due to their mechanical/chemical stability compared to liquid-state bulk EGaIn [18–20]. Nevertheless, the applicability of EGaIn* as interconnects or direct contact pads has been limited due to the formation of native oxide on the surface, which acts as an insulator. Mechanical stimuli for removing this insulating layer is required for electrical activation, converting EGaIn* back into bulk EGaIn, thus re-introducing the issues of bulk liquid metal. Also, such additional process introduces uncertainties (shorted or opened circuits), degrading the yield and reproducibility. Recently, initially conductive EGaIn* have been reported; however, they require annealing at high temperature for a long time to achieve electrical conductivity [21,22]. Furthermore, due to the limited controllability of EGaIn*, it has only been utilized for embedding in an elastomer matrix [13,23–25], direct printing [26,27], and screen printing [28,29], which has limited their applicability towards high-resolution patterning.

Herein, we introduce initially conductive large-area multilayered patterning of EGaIn* using standard photolithographic process, thus enabling integration with current industrial manufacturing process. Supplementary Table 1 presents a comparison between the conventional EGaIn-based soft electronics fabrication strategies and our technique. Our EGaIn* was encapsulated with poly(styrene sulfonate) (PSS) (this will be referred to as EGaIn*:PSS from this point forward) (Fig. 1a). The PSS enables compact particle assembly without rupturing during solution shearing, resulting in uniform and densely packed EGaIn*:PSSbased thin-film on top of photoresist (PR)-patterned substrate (Fig. 1b). Using dimethyl sulfoxide (DMSO) as the lift-off solvent, cohesion between EGaIn*:PSS was induced to render the film conductive (Fig. 1c, Refer to Supplementary Fig. 1 for schematic of the lithography process). Here, the particle-assembled morphology was critical for clean lift-off (Fig. 1d(i)). Fig. 1d(ii) and (iii) show conductivity without an activation step and waferscale patterning of EGaIn*:PSS, respectively. The chemical/mechanical stability and initial conductivity of the EGaIn*:PSS enabled repeated lithography processing and layer-to-layer electrical connection, thus allowing multilayered patterning (iv). Furthermore, EGaIn*: PSS can be transferred or directly patterned on a variety of substrates (e.g., PDMS, PET, and gelatin-based biogel [30]) (v), rendering our technique a highly versatile process for the fabrication of various soft electronic devices. Our activation-free large-area patterning of EGaIn* with standard well-developed fabrication process make it industrially feasible and can therefore be applicable to the large-scale manufacturing of soft and stretchable electronics.

Results

Preparation and characterization of ink with functionalized EGaIn particles.

Configuration of EGaIn*: PSS is depicted in Fig. 2a. To make EGaIn*: PSS, bulk EGaIn and PSS were dispersed in deionized water (DI) containing 5 vol% acetic acid (AA) using tip sonication [13,31]. Contrary to the previously reported ink [32], this ink was reformulated and optimized for lift-off process. Since the zeta potential (ζ) of EGaIn* is +76.8 mV in DI with AA, negatively charged polyelectrolytes such as PSS are easily attached on the EGaIn* via columbic interaction. With the inclusion of PSS in the solution, the zeta potential is a negative value at -32.4 mV, suggesting that the PSS (known as a negativelycharged polyelectrolyte) is surrounding EGaIn*. As presented in the energy dispersive spectrometry (EDS) images in Fig. 2b, the carbon and sulfur peaks from PSS are dispersed on the surface of EGaIn*. As will be discussed below, PSS was chosen as our dispersing polymer since it increases the wettability of ink on substrates for uniform coating, and because it enables cohesion of EGaIn* (to render the film conductive) when reacted with DMSO. AA functioned as a charge neutralizing agent that offsets the repulsive electrostatic force between EGaIn*:PSS (Supplementary Fig. 2), which allowed uniform and compact thin-film formation compare to other solvents (Supplementary Fig. 3). AA was also crucial for enhancing substrate adhesion during and after lift-off (to be discussed below).

For well-defined wafer-scale patterning, it is critical to generate a uniform and compact (i.e., without voids) EGaIn*:PSS film over a large area. We utilized solution shearing as a means to generate the EGaIn*:PSS film due to its large-area scalability and controllable thin-film properties [33,34]. As depicted in Fig. 2c, the solution that included both AA and PSS yielded the best film coverage with low variability between samples (substrate: glass). This can be attributed to the high wettability of the ink on the substrate due to PSS (Supplementary Fig. 4) and the charge neutralizing property of AA. PSS may also be acting as a bridging polymer [32] that enhances ink stability (Supplementary Fig. 5) and aids in the close-packed assembly of EGaIn*. Without PSS, rupturing/reduction of EGaIn* and the formation of gallium oxide were frequently observed (inset SEM image and Supple-



Fabrication process of particle-assembled liquid metal (EGaln*:PSS) film-based soft electronics and demonstration of key features. a, b, c, Schematic illustration of EGaln*:PSS film-based soft electronics fabrication process. **a**, Preparation of functionalized EGaln*:PSS ink. **b**, EGaln*:PSS film coating on photoresist (PR) patterned substrate by solution shearing. **c**, Lift-off-based patterning of EGaln*:PSS film. **d**, Demonstrations of EGaln*:PSS film-based fabrication. (i) Lift-off-based patterning: SEM images of patterned EGaln*:PSS films. Scale bar, 5 μm. (ii) Initial conductivity: photograph of a patterned EGaln*: PSS films integrated with LED. (iii) Multiscale processing: photograph of a patterned high-resolution EGaln*:PSS film on a 4-inch wafer. Scale bar, 2.5 cm. (iv) Multilayer fabrication: optical microscopy image of the stacked structure made with EGaln*:PSS films. Scale bar, 400 μm. (v) Integration with soft substrates: photographs of EGaln*:PSS-based serpentine pattern on PET (left) and gelatin-based biogel (right).

mentary Fig. 6), which rendered it difficult to conduct the lift-off process. Also, both AA and PSS play an important role in imposing stretchability to the film (Supplementary Fig. 7).

Fig. 2d and Supplementary Fig. 8 are particle size distributions as a function of sonication time, respectively. The average particle size decreases with increasing sonication time. The inset of Fig. 2d confirms that with the reduced average particle size, smaller features can be attained. We have determined that to attain a 10 μ m line width, an average particle diameter of ~3.69 μ m is required.

Fig. 2e is a schematic representation of EGaIn*:PSS thin-film formation using solution shearing [35]. As shown in the inset optical microscopy (OM) image, a meniscus (curved liquid–air interface) naturally forms between the blade and the substrate, and as the blade moves, a thin-film is deposited across the sub-

strate via liquid-to-solid transition occurring near the edge of the meniscus, i.e., the contact line (substrate-solution-air interface) [36]. Solution shearing enables the accurate control of fluid dynamics and localizes solvent evaporation at the meniscus, enabling uniform coating and precise tuning of thin-film properties such as packing density and film thickness [35–38]. During solution shearing, the EGaIn*:PSS continuously migrated towards the meniscus, which was critical for uniform thin-film formation (Supplementary Fig. 9, Supplementary Movies 1 and 2) [36]. Other coating techniques, such as doctor blading and spin coating, were not appropriate for the deposition of the EGaIn*:PSS as discussed in Supplementary Fig. 10. Supplementary Table 2 summarizes the benefits of solution shearing in generating the EGaIn*:PSS thin-film compared to that of other coating techniques. Fig. 2f show that the particles are compactly



FIG. 2

Solution shearing of EGaln*:PSS ink and characterization of particle-assembled film. a, Schematic illustration of EGaln*:PSS. **b**, EDS image of EGaln*:PSS film. **c**, The film coverage ratio of each ink. All films are coated on glass substrates with the same solution shearing condition. Inset (left): photograph of the film with EGaln* ink dispersed in DI/PSS. Scale bar, 2 cm. Inset (right): SEM image of the film with EGaln* ink dispersed in DI/AA. Scale bar, 5 µm **d**, Particle size distribution of inks at various tip sonication times. Inset: SEM images of EGaln*:PSS film deposition through solution shearing process. **f**, 3D surface profile data and SEM image of EGaln*:PSS film coated by solution shearing. Scale bar, 30 µm. **g**, Photograph of EGaln*:PSS films coated on various substrates. Scale bar, 3 cm **h**, Film thickness versus shearing speed for inks. Inset is a log-scale fitting graph to calculate the power scale. **i**, Cross-sectional SEM images of the EGaln*:PSS films coated on bare and PR patterned substrate. Scale bar, 50 µm. **k**, Cross-sectional SEM image of the EGaln*:PSS films.

assembled uniformly to form the film. Due to the ink's wetting ability on various surfaces, the EGaIn*:PSS film can be solutionsheared on a variety of surfaces, as depicted in Fig. 2g, making it highly versatile for the fabrication of a variety of devices.

To achieve lift-off-based photo-patterning, the part of the EGaIn*:PSS film coated on top of the PR must be selectively removed. In this regard, a film that is weekly bonded together but is strongly adhered onto the substrate surface is ideal for lift-off. During solution shearing, two different thin-film morphologies were observed: 1) particle-pack morphology where the EGaIn*:PSS were assembled in a close-packed manner, 2) bulk-like continuous morphology, where the EGaIn*:PSS completely merged together to form the film. The former case was observed in the evaporative regime (<2 mm/s), and this was the morphology that yielded cleanly lifted off patterns. This can be attributed to the two properties mentioned above as discussed in Supplementary Fig. 11. The thickness of the particle-assembled film was controllable via shearing speed at the evaporative regime (Fig. 2h and i) [36]. At a given shearing speed, the

particle-assembled film had the same thickness with or without PR, and at different PR thicknesses (Fig. 2j; hence, making it easy to control the film thickness.

Bulk-like continuous film was attained at high coating speeds (>10 mm/s) (Fig. 2k), in the Landau-Levich regime (Supplementary Fig. 12) [36]. This bulk-like film was not stretchable unlike particle-assembled film as presented in Supplementary Fig. 13. For this film, indium oxide peaks were detected in the XPS spectrum; whereas, such peaks were not present in the particle-assembled film (Supplementary Fig. 14). The indium oxide likely formed due to the oxidation of EGaIn as the particles were being ruptured and annealed. This result indicates importance of coating condition to achieve photo-patternable and stretchable EGaIn*:PSS film.

Organic solvent-based electrical activation and photopatterning

One important aspect of patterned EGaIn* film is its initial conductivity. Generally, due to the surrounding native oxide on the



FIG. 3

Organic solvent-based electrical activation and lift-off-based patterning of EGaln*:PSS film. a, Schematic illustration of cohesion of EGaln*:PSS induced by detachment of PSS in DMSO. **b**, SEM and EDS image of EGaln*:PSS film before and after immersion in DMSO. **c**, XPS spectra of EGaln*:PSS film before and after immersion in DMSO. **d**, Conductivity of EGain*:PSS film before and after immersion in DMSO. **e**, Cross-sectional SEM images of the electrically connected multilayer structures (left), and insulated multilayer structure with polyimide (PI) in between (right). Scale bar, 2 µm. **f**, Photograph of a EGaln*:PSS film-based interdigitated electrode on soft substrate. Scale bar, 500 µm **g**, SEM image and depth profile data of a patterned EGaln*:PSS film. **h**, Cross-sectional SEM images before (left) and after (right) patterning with different thicknesses of EGaln*:PSS films. Scale bar, 60 µm. **i**, Photograph of a patterned EGaln*:PSS film on a 4-inch silicon wafer. Scale bar, 6 cm. Inset: zoomed-in optical microscopy image of the patterned EGaln*:PSS film (The smallest line width: 20 µm). Scale bar, 200 µm. **j**, Photographs after the lift-off process of a bulk-like film and a film coated with highly acidic ink or ink without PSS. Lift-off process was impractical in both cases.

surface of the EGaIn^{*}, it is initially non-conductive, thus requiring activation in the form of mechanical stimuli (i.e., rubbing) [13,39]. However, this is not a preferred methodology, especially for high-resolution complex patterns, as it stochastically introduces defects in the circuit. In our work, no activation is required: the film becomes conductive during lift-off process by using DMSO as the PR stripper. Unlike that of previous work [21,22], no high temperature processing is required, thus enabling direct patterning on soft elastomeric substrates. We hypothesize that when EGaIn^{*}:PSS is immersed in DMSO, electrostatic interaction between the EGaIn^{*} and PSS is reduced due to the high polarity of DMSO [32], which in turn induces the cohesion (i.e., partial merging) between EGaIn* (Fig. 3a). The other lift-off solvents (acetone and toluene) did not generate high electrical conductivity in the film (Supplementary Fig. 15). Fig. 3b presents EDS mapping of carbon (i.e., an element in PSS) before and after immersion in DMSO. Carbon attached on the particles before immersion was detached and aggregated after immersion in DMSO. This mechanism is similar to the interaction between PEDOT:PSS and DMSO; doping with polar solvents (i.e., DMSO, ionic liquid) induces the phase separation between the insulating PSS chain and PEDOT, which in turn causes the tight cohesion and interconnection of conductive PEDOT [40,41]. Fig. 3c indicate the presence of Ga peak in XPS after

immersion in DMSO [32]. This result can be attributed to the partial extrusion of bulk EGaIn during the cohesion between particles. After immersion in DMSO, EGaIn*:PSS shows the metal level conductivity with 2.2×10^6 S/m, as presented in Fig. 3d. Such post activation-less fabrication process overcomes the key limitation of EGaIn*-based electronics, which has impeded the wide-spread application of EGaIn* towards industrially feasible soft electronics [13,39].

One distinguishing feature of our photo-patterning technique, contrary to previous report that pattern bulk EGaIn, is the mechanical and chemical stability of the EGaIn*:PSS film that renders it compatible with conventional cleanroom-based fabrication processes such as spin coating, and reactive ion etching (RIE). This enables multi-layered patterning as seen in Fig. 3e and further discussed in Supplementary Figs. 16 and 17. Furthermore, transfer printing onto a soft-substrate is also possible [7,42]. Fig. 3f is a photograph of EGaIn*: PSS film-based interdigitated electrodes (IDT) patterned on a soft elastomer substrate, which was then placed on a finger (see Supplementary Fig. 18 for fabrication steps and Supplementary Fig. 19 for the electrical characteristic of IDT). The mechanical stability enables use as direct electrodes without the need of an encapsulating layer. Fig. 3g are SEM image and depth profile data of a patterned EGaIn*: PSS film with 30 µm line width and spacing, demonstrating high uniformity and cleanly lifted-off edges. Here, to attain the high-density and high-resolution features, MW of PSS plays crucial role as discussed in Supplementary Fig. 20. Fig. 3h are thin (4 µm) and thick (15 µm) EGaIn*:PSS films before and after the lift-off process. Despite the fact that PR thickness was 3 µm, both the thin and the thick EGaIn*:PSS films lifted off cleanly (patterning feasibility depending on PR and film thickness is presented in Supplementary Fig. 21). These results can be attributed to the selective annealing of the EGaIn*:PSS film at the bottom layer (Supplementary Fig. 11). As shown in Fig. 3i, reliable wafer-scale patterning of EGaIn*:PSS film with highresolution (The smallest line width: 20 µm) was achieved with standard cleanroom-based fabrication.

The left panel of Fig. 3j depicts a bulk-like film after lift-off, while the right panel illustrates a film that has been coated using a highly acidic solvent or lacks PSS after lift-off. Both films show that clean patterning is not achievable, which as alluded above, is likely due to the strong internal bonding of the film that prevents partial removal of the film [43]. Previously, we have developed printable liquid metal ink with acidic solvents [32]. However, this composition of ink was not compatible with lift-off. For this work, the ink formula was optimized for photopatterning by reducing the acidity of solvent. To the best of our knowledge, this is the first demonstration of utilizing standard cleanroom processes for wafer-scale patterning of initially conductive EGaIn*, opening up a wide variety of possible soft electronic applications that require multilayer, high-resolution, and high-density features over a large area.

Demonstration of large-scale and high-resolution soft electronics with EGaIn*:PSS film

Scalability, capability to integrate with electronic components, and stretchability of the EGaIn*:PSS film can enable the construction of large-scale soft electronics. As mentioned in

Fig. 2g, EGaIn*:PSS film-based electronics can be fabricated on various soft substrates (e.g., PDMS) with different dimensions (see Supplementary Fig. 18 and Methods section for detailed fabrication steps). Also, conventional electronic components (e.g., LEDs) can be easily integrated with EGaIn*:PSS-based soft electronics (Fig. 4a), as with traditional printed circuit boards (PCBs). Another important feature of the EGaIn*:PSS approach for the design of soft electronics is that its positive piezo-conductivity allow the negligible change in resistance under the application of strain unlike bulk EGaIn as shown in Fig. 4b [44]. Resistance variation under strain over 50% is presented in Supplementary Fig. 22. I-V curves of an LED connected to EGaIn*:PSS filmbased stretchable interconnects were measured with and without strain to corroborate stable operation (Fig. 4c). A negligible change in current was observed under the application of 50 % strain. Such consistency in electrical characteristics allows the stable operation of electrical circuits under dynamic deformation, opening opportunities for flexible and stretchable electronics. Using these attributes, for the demonstration of large-scale soft electronics, we fabricated a stretchable LED display built on a 6-inch wafer (Fig. 4d). Unlike conventional metals, which often show limited principal strains of under 1%, the sufficiently large stretchability of EGaIn*:PSS allows stable operation under the application of strain. These features may enable new opportunities for large-scale wearable electronics as demonstrated in Supplementary Fig. 23. Furthermore, EGaIn*:PSS shows highly consistent and durable operation as a stretchable conductor under repeated loading and unloading of strain (Supplementary Fig. 24) and long-term operation capability (Supplementary Fig. 25).

Utilizing all the unique attributes of the EGaIn*:PSS film, (i.e., high-resolution and large-area patterning capability on soft substrates, chemical and mechanical stability, and multilayer processing capability), we also demonstrated an all-soft highresolution pressure sensing array (SPSA). Fig. 4e shows a photograph of the SPSA conformally integrated on the curved surface of a robotic hand to provide high-resolution pressure sensing (see Supplementary Fig. 26 for the deformability of SPSA). To mimic the pressure sensing capability of the human skin, a highspatial resolution (~5 mm or smaller) is required. To acquire such small sensing pixels with high density while minimizing crosstalk, we designed an SPSA consisting of two EGaIn*:PSS layers electrically connected through via holes through an insulating layer (Fig. 4f, see Supplementary Fig. 27 for the fabrication process). The bottom EGaIn*: PSS layer has a microelectrode array (MEA) and its interconnect (size of MEA is 500 μm imes 500 μm and width of interconnect is 250 µm), and the top layer contains pressure sensing electrodes directly contacted with pyramidpatterned PDMS layer coated with a conductive polymer (polypyrrole) (Supplementary Fig. 28). As pressure is applied, the contact resistance between the pyramids and the electrode decreases. Overall, 8×8 pressure sensing pixels are integrated into the SPSA. The size of each pixel is 3 mm \times 3 mm and the overall size of the array is about 3 cm \times 3 cm. The SPSA with closely-packed 64 sensors allows the detection of the contour of a lightweight object (a commercial pen drive) through mapping the pressure distribution (Fig. 4g). One notable characteristic of EGaIn*:PSS films is their high stability when repeatedly coming into contact



FIG. 4

Electrical characterization of particle-assembled liquid metal (EGaIn*:PSS) film and various soft electronics demonstrations. a, Photograph of soft electronics with EGaIn*:PSS film-based interconnect connected to a μ-LED. **b**, Relative resistance change as a function of applied strain to EGaIn*:PSS film and bulk EGaIn. **c**, I-V curve of LED connected to a stretchable EGaIn*:PSS film-based interconnector with (red) and without (black) 50% strain. **d**, Photograph of a large-area (6-inch) stretchable LED display with EGaIn*:PSS film-based interconnects. **e**, Photograph of the EGaIn*:PSS-based SPSA integrated on a robotic hand. **f**, Illustration of a multilayer EGaIn*:PSS film-based all-soft high-resolution pressure sensing array (SPSA). Pyramid-patterned PDMS coated with polypyrrole was laminated on the electrode for pressure sensing. **g**, Pressure mapping of a pen drive placed on the EGaIn*:PSS-based SPSA. **h**, Loading and unloading test with a repeated application of 50 kPa for over 10,000 cycles. Insets show a zoomed-in view of the graph (left) and an SEM image of a pyramid-structured pressure sensor after the cyclic test (right).

with another surface, a feature that conventional EGaIn does not possess. This mechanical stability is evidenced by Fig. 4h, which shows consistent resistance variation of the pressure sensor when subjected to repeated contact with an applied pressure of 50 kPa over 10,000 cycles. Additionally, due to its mechanical robustness and biocompatibility, EGaIn*:PSS can serve as a biointerfaced electrode for surface electromyogram (sEMG) sensors, as presented in Supplementary Fig. 29. This application highlights the versatility of EGaIn*:PSS in soft electronics.

Demonstration of artificial finger with EGaIn*:PSS film

A soft artificial finger that can decouple pressure and strain was demonstrated by leveraging the unique capabilities of our EGaIn*:PSS film, which includes multilayer construction, large area coverage, high-resolution fabrication, precise control over film thickness, and direct patterning on soft substrates (Fig. 5a). The artificial finger consisted of two EGaIn*:PSS layers electrically connected through a via hole (the multilayer fabrication process is presented in Supplementary Fig. 30). The first layer consisted



FIG. 5

Characterization of EGaIn*:PSS-based artificial finger. a, Illustration of the artificial finger. **b**, Photograph of the EGaIn*:PSS-based artificial finger. **c**, Resistance variation between terminal 1' and 3' according to pressure. **d**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain. **e**, Resistance variation between terminal 1' and 2" according to strain.

of thick EGaIn*:PSS film-based interdigitated electrodes. On the interdigitated electrodes, pyramid-structured PDMS coated with a conductive polymer (polypyrrole) was laminated, which together functioned as a piezoresistive pressure sensor. The second layer consisted of a thin EGaIn*:PSS coated on nano Pt film, functioning as a strain sensor. Fig. 5b is a photograph of the artificial finger placed on a human finger.

To monitor pressure and strain with the artificial finger, three terminals were connected to a monitoring system. Terminal 1' and terminal 3' were located in the first layer (' represents the first layer) and were made up of thick EGaIn*:PSS films. Terminal 2" was located in the second layer (" represents the second layer) made up of a thin EGaIn*:PSS film. Pressure can be monitored by measuring the resistance between terminals 1' and 3' as presented in Fig. 5c. The resistance between 1' and 3' was determined by the contact resistance of the pyrrole-coated pressure sensor. To monitor the strain, the thin EGaIn*: PSS film was electrically connected to one of the thick electrodes in layer 1 using a 'via hole.' Since the resistance of a thin-film varies with strain, strain can be determined by measuring the resistance between terminals 1' and 2" as shown in Fig. 5d. Here, the resistance of a thick EGaIn*:PSS film is stable under the application of strain since the resistance of a thick EGaIn*:PSS film is negligibly low compared to that of the conductive polymer coated on the pressure sensor. Therefore, pressure can be independently monitored even when mechanical strain is applied to the artificial finger (Fig. 5e).

Fig. 5f are real-time measurements of pressure and strain. The resistance between terminals 1' and 3' (both belonging to the first layer) decreased with pressure, while the resistance between terminal 2'' (belonging to the second layer) and terminal 1' increased with strain. When pressure and strain are independently applied (blue and red regions respectively), only the corre-

sponding signal changes. Moreover, when both stimuli are applied simultaneously (yellow region), the corresponding signals change concurrently without cross-interference, verifying that pressure and strain can be effectively decoupled.

Conclusion

Soft electronics are expected to play a critical role in the forthcoming electronic applications, where devices will make intimate and conformal contact with the soft tissues of the body. EGaIn* is an appropriate material for interconnects and electrodes for soft electronics, and their applicability relies on their compatibility with well-established cleanroom-based photolithographic patterning, as large-area, multilayer, and high-resolution features are needed for integration with conventional electronic systems. Our technology enables the formation of a highly uniform and stable EGaIn* film over a large-area via solution shearing. The particle-assembled morphology of the film achieved in the evaporative regime enables photo-patterning with a conventional lift-off process on a variety of substrates, and its stability allows a cleanroom-based multi-layered fabrication process. Furthermore, this film does not need a separate mechanical activation step to impart conductivity. These attributes were utilized to fabricate interconnects, stretchable display, and multilayer tactile sensors. We anticipate that our technique will provide the pathway for significant advancements in soft electronics, thereby bringing forth new opportunities in soft robotics, and wearable and implantable devices in the near future.

Method

Materials

All chemicals were used without further purification and were acquired from Sigma-Aldrich unless otherwise mentioned. To

prepare and characterize the EGaIn*:PSS ink, eutectic gallium indium alloy (EGaIn, Rich-Metals), poly(styrene sulfonate) (PSS, with an average molecular weight of 1,000,000), acetic acid (99%), ethanol (99.5%), acetone (99.5%), isopropyl alcohol (IPA, 99.5%), dimethyl sulfoxide (DMSO, 99.9%), hydrochloric acid (HCl, 37%), and Span 80 were used. For lithographical patterning and device fabrication, we used trichloro(1H,1H,2H,2Hperfluorooctyl)silane (PFOCTS, 97%), poly(methyl methacrylate) (PMMA), anisole (99%), polyamic acid solution (PI), PDMS (Sylgard 184, Dow Corning), NR-9 3000py and 8000py (Lift-off PR, Futurrex), and AZ 300 MIF (developer, MERCK). For the fabrication of pressure sensor, pyrrole monomer, polypyrrole solution, and Iron(III) p-toluenesulfonate hexahydrate were used.

Preparation of functionalized EGaIn*:PSS-embedded ink for solution shearing

Bulk EGaIn (1.4 g), and 0.14 g of PSS (MW: 1,000,000) were dissolved in diluted acetic acid (5 vol% in DI water, 2 ml). This compound solution was tip sonicated (VC 505, Sonics & Materials, 3 mm microtip) at 500 W and 20 kHz for 30 min unless stated otherwise.

EGaIn*:PSS film coating via solution shearing

The EGaIn*: PSS ink was coated on various substrates (glass, PET film, Si wafer, PI film, PR film, PDMS, Au) by using a customized shearing machine. Before solution shearing, substrates were treated with oxygen plasma (CUTE, Femto Science) at 100 W for 1 min to clean and activate the surface. The substrates were heated to 70 °C during film coating. Here, two types of glass slides (25 mm \times 75 mm/ 50 mm \times 75 mm) were used as the coating blade depending on the width of the film needed. We fixed the angle (5°) and the gap between blade and substrate (200 µm) for all samples. 100 µl of EGaIn*:PSS ink was injected between the coating blade and substrate right before solution shearing. After the shearing process, EGaIn*:PSS film-coated substrates were placed on a 70 °C hot plate to completely evaporate away any remaining solvent. For blade coating, the shearing angle was modified to 90° (blade standing vertically), and for bulk-like film coating, the speed of the moving substrate increased to >10 mm/s.

DMSO-based patterning and electrical activation

For the patterning of the EGaIn*:PSS film, NR9-3000py, and 8000py (lift-off PR) were spin-coated and selectively exposed to UV light using a mask aligner (MJB4). Here, we note that other PR easily removed by DMSO can be used such as AZ series (AZ Electronic Materials). Spin-coating and UV exposure conditions were tuned according to the thickness of the PR. The exposed PR was baked at 110 °C (150 sec) and developed in a developer (AZ 300 MIF developer, MERCK) for 50 sec. Subsequently, the substrate was exposed to oxygen plasma treatment for surface activation. Then, EGaIn*:PSS film was coated on the PR-patterned substrate via solution shearing as described above. Finally, the sample coated with EGaIn*:PSS film was immersed in DMSO and sonicated by bath sonication for lift-off. After sonication, the patterned sample was placed on the hot plate at 180 °C to remove DMSO.

Fabrication of soft electronics through the direct patterning of a EGaIn*:PSS film

Schematic illustration in Supplementary Fig. 18 depicts the overall direct patterning process of soft electronics with a EGaIn*:PSS film. Before coating the PDMS film on the rigid glass substrate, the glass substrate was modified as a hydrophobic surface with PFOCTS by chemical vapor deposition to facilitate the delamination of the film. Here, all four edges of the glass were sealed with a PI tape prior to the hydrophobic coating process to maintain hydrophilicity, so that delamination can be prevented during solution-based processing (e.g., development, lift-off). After chemical vapor deposition, the PDMS solution (base and curing agent were mixed in a weight ratio of 10:1) was poured on a glass slide and spin-coated (1000 rpm, 30 sec), and was cured at 70 °C to form a PDMS film. Subsequently, the PI solution was spincoated on the PDMS-coated glass at 4000 rpm for 60 sec and cured at 250 °C for 60 min. PR was patterned with the aforementioned methods. Afterward, oxygen plasma etching was conducted (50 sccm, 200 W, 30 min) to remove the PI film in the regions absent of PR. Then, EGaIn*: PSS was coated and patterned directly on the PDMS film by following the abovementioned methods. The remaining PI was removed by oxygen plasma etching (100 sccm, 200 W, 1 h). For the multilayer structure, we repeated the steps from PI coating onwards. Finally, the sample was cut along the hydrophobic/hydrophilic border, and the PDMS was lifted off the glass substrate.

Fabrication of SPSA

The illustration in Supplementary Fig. 25 depicts the overall fabrication process of SPSA. Firstly, the MEA and interconnects were directly patterned on a spin-coated PDMS (800 rpm, 30 sec) by using the direct patterning method. After patterning the bottom layer, a PDMS solution was poured on a patterned EGaIn*:PSS layer and was spin-coated (2500 rpm, 30 sec) for electrical insulation. For the electrical connection between the bottom layer and the top electrodes, via holes were created in the MEA layer by laser engraving (MD-U1000C: UV laser cutter, Keyence). Then, direct contact electrodes were patterned on the top layer by aligning with the bottom MEA. These top direct contact electrodes and bottom electrodes were electrically connected through the via holes. Finally, a pyramid-structured PDMS pressure sensor was laminated on the direct electrode to map the pressure.

Fabrication of pressure sensor

The schematic illustration in Supplementary Fig. 26a depicts the overall fabrication process of the conductive polymer-coated pyramid pressure sensor. For the structuring of the pyramid pattern, the silicon substrate was chemically etched as previously reported [45] and subsequently coated with PFOCTS to facilitate the delamination of the PDMS film. A PDMS solution (base and curing agent were mixed in a weight ratio of 10:1) was then poured on the mold and spin-coated at 800 rpm for 30 sec. The PDMS was cured at 80 °C for 30 min and was peeled off from the mold. Then, the surface of the pyramid-patterned PDMS film was treated with oxygen plasma to activate its surface. Pyrrole monomer was coated on the PDMS through the chemical vapor deposition method (80 °C, 30 min). Finally, polypyrrole was

deposited on the PDMS surface through immersion in pyrrole solution (0.4 wt%) in the presence of Iron(III) p-toluenesulfonate hexahydrate catalyst (1.6 wt%) for 12 h [46].

Monitoring of sEMG

Real-time monitoring of sEMG was conducted with commercial wireless sEMG measurement equipment (BioRadio, Great Lakes NeuroTechnologies). An EGaIn*:PSS-based sEMG sensor was electrically connected with a measurement apparatus via a commercial anisotropic conductive film (line width and pitch: $250 \mu m$, 3 M) and customized PCB.n.

Characterization

Chemical, rheological, and morphological characterization. Zeta potential values of EGaIn*:PSS inks were measured by DLS (Zeta-sizer nano zs, Malvern). Each ink was characterized by a UV/VIS Spectrophotometer (Lambda 1050, Perkin Elmer) in the wavelength range of 200 nm to 500 nm. To determine the chemical composition of the coated film under different coating conditions, X-Ray Photoelectron Spectroscopy (XPS, K-alpha, Thermo VG Scientific) was conducted.

The apparent viscosity of EGaIn*:PSS inks with different proportions of PSS was measured by MCR 302 rheometer (Anton Paar) at a shear rate of 10^{-2} – 10^2 s⁻¹ at room temperature. To determine the wettability of each ink, the contact angles were measured through a contact angle analyzer (SEO Phoenix). A PI film without oxygen plasma treatment was used as the substrate. Measurements were conducted twice for each sample: when 50 µl of the sample was dropped and when 25 µl were withdrawn from the same droplet.

To observe the morphology of the coated film, SEM images were taken by S4800 (Hitachi). The surface morphology and roughness of the films were analyzed by a 3D surface profiler (TOF-SIMS5, ION-TOF GmbH). To evaluate the efficiency of film coating, the coverage ratio was defined as the area coated with the EGaIn*:PSS film divided by the total area. The coverage ratio was measured by mapping the 8-bit optical image of each film with the Image J program.

Electrical Characterization. To measure the resistance, impedance, and capacitance, an LCR meter (4284A, HP) was used. The samples were cut into the same size (1 mm \times 25 mm) unless stated otherwise. I-V curves under the application of strain were determined with a source meter.

Characterization of the artificial finger was conducted with the LCR meter, force gauge (the maximum force is 50 N, Mark-10), a motor stand (Mark-10), and a customized manual strain machine. The reflection coefficient of the EGaIn*:PSS-based closed-loop antenna was measured with the commercial magnetic field reader (HZ-15 RSH400-1, Rohde & Schwarz) connected with a vector network analyzer (ZND, Rohde & Schwarz) through an RF cable.

Experiments on human subjects

All experiments including demonstrations of electronic skin and sEMG sensor on human skins were performed under approval from the Institutional Review Board at Korea Advanced Institute of Science and Technology (protocol number: KH2021-039) and received informed consent from the volunteer subjects.

CRediT authorship contribution statement

Gun-Hee Lee: Conceptualization, Methodology, Visualization, Investigation, Writing – original draft, Writing – review & editing. Hyeonji Kim: Visualization, Investigation, Data curation. Juhyun Lee: Methodology, Investigation. Jae-Young Bae: Methodology, Investigation. Congqi Yang: Investigation, Formal analysis. Hanul Kim: Software. Heemin Kang: Resources. Siyoung Q. Choi: Resources. Seongjun Park: Resources, Methodology. Seung-Kyun Kang: Res4ources, Methodology. Jiheong Kang: Resources, Methodology. Zhenan Bao: Resources, Methodology, Writing – review & editing. Jae-Woong Jeong: Conceptualization, Supervision, Writing – review & editing, Funding acquisition. Steve Park: Conceptualization, Supervision, Writing – review & editing, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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