Photo-patternable, large-area solid-state liquid metal film coated via solution shearing for soft electronics

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Article

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Abstract

Liquid metal (LM) is considered one of the most promising conducting materials for soft electronics due to its unique combination of metal-level high conductivity with exceptional deformability and stretchability. However, their practical applicability has thus far been limited due to the challenges of generating chemically and mechanically stable film over a large-area and the need for non-standard fabrication approaches. Here, we report materials and manufacturing methods that enable multiscale patterning (from microns to centimeters) and multilayer integration of ‘solid-state liquid metal (SSLM)’ with the conventional cleanroom process. In this work, solution shearing of a polyelectrolyte-attached LM particle ink is used to generate SSLM films. The stabilized LM particles were observed to form a close-packed thin-film without particle rupture when coated under evaporative regime. This is essential in enabling a subsequent photolithographic lift-off process at wafer-scale to produce high-resolution features (~ 10 µm) of varying thicknesses irrespective of the substrate. Demonstrations of wearable multilayer tactile sensing systems and stretchable skin-interfaced electronics validate the simplicity, versatility, and reliability of this manufacturing strategy, suggesting broad utility in the development of advanced soft electronics.

Introduction

With the rising demand for next-generation wearable healthcare devices, soft robotics, and conformable implantable devices, soft electronics are increasingly becoming of pivotal importance. 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. The critical component that imparts softness to electronics are the stretchable electrodes and interconnects, and there are generally two main ways to fabricate them. One method is to fabricate serpentine or wavy structures with standard metallic materials, and the other is the use of stretchable materials such as carbon nanotube composite, metal nanowire composite, and conductive polymers. In the former case, high-density integration is difficult due to the extra space needed by the geometric design, and the stretchability of electronics is limited due to relatively small fracture strain of metallic materials. In the latter case, the stretchable materials either lack sufficient conductivity and/or are incompatible with conventional photolithographic process.

Gallium-based liquid metal (LM) has recently drawn a great deal of attention as it can potentially overcome the aforementioned limitations, owing to its exceptional deformability and stretchability, and due to its high conductivity comparable to that of metals. However, its liquid state, ultrahigh surface tension, and immediate formation of oxide layer render it very challenging for patterning and integration. For instance, Brand et al. demonstrated LM patterning with electron beam lithography; however, this technique requires a specific metal adhesion layer for transferring LM, and because LM is in a liquid state, direct use as electrodes and multilayer integration are challenging. Kramer-Bottiglio et al.
and Zheng et al. utilized stencil printing to pattern LM;\textsuperscript{16,17} however, the patterning dimension was limited to hundreds of micrometers due to the inherent resolution limit of stencil mask.

Herein, we introduce a solution shearing-based deposition of solid-state LM (SSLM) film using specifically designed LM particle ink for the fabrication of wafer-scale, multi-layered, and high-resolution, lithographically patterned soft electronics. Supplementary Table 1 presents a comparison between conventional LM-based soft electronics fabrication strategies and our technique. Importantly, our ink formulation with the inclusion of polyelectrolyte greatly stabilizes the dispersion of LM particles (Fig. 1a) and induces self-assembly during deposition by forming bridges between particles, which results in uniform and densely packed LM particle-based thin-films, while without particle rupturing, during solution shearing (Fig. 1b). Such features enabled effective patterning of the SSLM film using standard lift-off process (Fig. 1c). LM particle-based inks have previously been reported.\textsuperscript{18} However, they have only been utilized by embedding in an elastomer matrix,\textsuperscript{11,19,20,21} direct printing,\textsuperscript{22,23} and screen printing,\textsuperscript{24,25} mainly due to the lack of precise control of LM particles and rupturing during processing, which have limited their applicability towards high-resolution and high-performance soft electronics. As depicted in Fig. 1d (i) and (ii), the thickness of SSLM film can be precisely controlled, and the film can be patterned at wafer-scale with micron-level resolution. Furthermore, due to the chemical and mechanical stability of the film, multilayer patterning is possible (iii). Finally, SSLM can be transferred or directly patterned on a variety of substrates (\textit{e.g.}, PDMS, PET, and gelatin-based biogel\textsuperscript{26}) (iv), rendering our technique a highly versatile process for the fabrication of various soft electronic devices. Our technique facilitates high-resolution patterning and large-area fabrication. Furthermore, it is simple, cost-effective, tunable, and environmentally friendly, which is therefore industrially feasible and can be applied to large-scale manufacturing of soft and stretchable electronics.

Results

Preparation and characterization of ink with functionalized LM particles.

LM particle-embedded ink was prepared as presented in Fig. 2a. Eutectic GaIn-based LM and polystyrene sulfonate (PSS, molecular weight (MW): 70,000 g/mol) were dispersed in deionized water (DI) containing 5 vol\% acetic acid (AA) using ultrasonication. The ultrasonication provides mechanical and thermal energy that induces the formation of LM particles covered with gallium oxide layer.\textsuperscript{11,27} Fig. 2b are photographs and zeta potential values of LM dispersed in DI (left), DI/AA (middle), and DI/AA/PSS (right). The DI by itself does not effectively disperse the LM, as evident by the near transparency of the solution and the sedimentation of LM at the bottom of the container. However, with the addition of AA, the solution became turbid with a gray color, confirming the dispersion of LM in the solution. As previously been reported, this can be attributed to the carboxylic acid group in the AA.\textsuperscript{28,29} Furthermore, the addition of AA increased the zeta potential of LM particles from +42.5 mV to +76.8 mV (Fig. 2b, Supplementary Fig. 1), due to the increased acidity of the solution, and such an increase may enhance the electrostatic coupling of PSS to LM particles. With the inclusion of PSS in the solution, the zeta
potential was a negative value at -5.4 mV, suggesting that the PSS (known as negatively charged polyelectrolyte) is surrounding the LM particles.\textsuperscript{30,31,32} We have concluded that such an interaction also induces the bridging of LM particles via PSS, as schematically depicted in Fig. 2a.\textsuperscript{33} The ink without and with PSS behaved differently upon the addition of HCl. Since HCl dissolves the gallium oxide layer, in the case of ink without PSS, all of the LM particles merged into one large LM droplet in the solution.\textsuperscript{27} For the ink with PSS, such behavior was not observed, where the LM remained dispersed in the solution (Supplementary Fig. 2). This further corroborates our assumption of PSS surrounding the LM particles, thereby enhancing their stability.

\textbf{Fig. 2c} are UV-Vis spectra of three different solutions: pure DI/AA solvent without LM, LM dispersed in DI/AA, and LM dispersed in DI/AA/PSS. The change in the absorption peak at 200~240 nm with the addition of PSS indicates a coupling reaction between LM particles and bridging polymers (Supplementary Fig. 3). To study the rheological behavior of the LM ink (DI/AA/PSS), apparent viscosity was measured as a function of shear rate (Fig. 2d). With increasing PSS concentration, the apparent viscosity decreased. Previous studies indicate that with higher variability in particle size, the viscosity decreases (inset of Fig. 2d, Supplementary Fig. 4).\textsuperscript{34} SEM images confirm that at high PSS concentration, LM particles in some regions aggregated and showed less spherical geometry (Supplementary Fig. 5). This suggests that the aggregated particles behave as large particles, thus increasing the effective particle size variability of the dispersion.

To verify the compatibility of our ink for solution shearing process, wettability on polyimide substrate (hydrophobic surface) was measured by observing contact angles of different inks (Fig. 2e).\textsuperscript{35,36} Firstly, the contact angle of 50 \( \mu l \) droplet was measured; subsequently, 25 \( \mu l \) was withdrawn from the droplet and the contact angle was measured again. The ink without PSS exhibited contact angle of 74.61° and 50.34° before and after ink withdrawal, respectively; whereas, that of the ink with PSS exhibited contact angle of 63.99° and 16.47°, respectively. The droplet with PSS did not decrease in diameter after ink withdrawal (i.e., the droplet was pinned), which was the reason for the larger decrease in contact angle. On the contrary, for the droplet without PSS, the decrease in contact angle was not as large due to the decrease in diameter upon ink withdrawal. This result suggests that PSS acts as a surfactant that decreases the interfacial energy between the ink and the substrate, which enhances the wettability of ink (Supplementary Fig. 6, and Supplementary Movie 1, 2). Such a forced wetting property of PSS-incorporated LM ink enables the formation of uniform liquid layer and thin-film during solution shearing, as described below.

\textbf{Fig. 2f} are coverage ratios (based on multiple samples for each ink) of solution-sheared SSLM films for three different LM inks: DI/PSS, DI/AA, DI/AA/PSS. DI/PSS had the lowest coverage and uniformity, likely due to poor dispersion of LM particles in ink. DI/AA exhibited improved coverage and uniformity; however, the coverage was still incomplete. This can be attributed to the lack of forced wetting ability and self-assembly of LM particles in the absence of bridging polymers. Furthermore, without PSS, rupturing and reduction of LM particles was frequently observed (Supplementary Fig. 7), which rendered it difficult to
conduct the lift-off process, as we will describe below. DI/AA/PSS generated a completely covered and uniform SSLM film (Fig. 2g), which can be ascribed to well-dispersed LM particles and the bridging between them that aids their self-assembly during thin-film formation. We have also confirmed the uniform distribution of components through energy-dispersive X-ray spectroscopy (Supplementary Fig. 8).

Fig. 2h and 2i are representative SEM images and particle size distribution as a function of sonication time, respectively, for the SSLM films generated with two different MW of PSS: 70,000 and 1,000,000 (the two LM solutions contained the same molarities of PSS). In the case of larger MW PSS film, non-spherical LM particles were commonly observed, where the interfacial area between the particles was relatively large. On the other hand, for low MW PSS film, the LM particles were dominantly spherical. Such a difference in film morphology can be attributed to reduced surface energy and stronger inter-particle attraction of the LM particles with the longer polyelectrolyte chains. Moreover, as evident in Fig. 2i, the average LM particle size can be tuned via sonication time (Supplementary Fig. 9). The inset of Fig. 2i confirms that with reduced average LM particle size, smaller features can be attained. We have determined that to attain a 10 μm line width, an average particle size of ~3.69 μm² is required.

Fig. 2j and 2k are the grey-scale color distribution of optical images and SEM images, respectively, of solution-sheared SSLM films generated with different solvents. The narrow distribution in the pixel color in the case of AA is an indication of uniform SSLM film and is visually verified in Fig. 2g and 2h. The other solvents, on the contrary, yielded poor uniformity, as evident by the broad pixel color distribution and the SEM images. Furthermore, rupturing of LM particles was observed in the films made with the other solvents. In the case of DMSO and acetone, since solvents damage the photoresist (PR), lithographical patterns could not be achieved.

Large-area coating of SSLM film through solution shearing.

Fig. 3a is a schematic representation of SSLM thin-film formation using solution shearing. Solution shearing is a technique analogous to blade coating, where the solution is sandwiched between a heated substrate and a moving blade. A meniscus (curved liquid-air interface) naturally forms between the blade and the substrate, and as the blade moves, thin-film is deposited across the substrate via liquid-to-solid transition occurring near the edge of the meniscus, i.e., contact line (substrate-solution-air interface). Solution shearing enables accurate control of fluid dynamics and localizes the solvent evaporation at the meniscus, enabling uniform coating and precise tuning of thin-film properties such as packing density and film thickness. We have observed that during solution shearing, the LM particles continuously migrated towards the meniscus, which was critical for uniform thin-film formation (Supplementary Movie 3). This can be attributed to effects such as capillary, Marangoni, pressure-driven, and boundary-driven flow that drive the transport of solute towards the contact line. Supplementary Table 2 summarizes the benefits of solution shearing in generating SSLM thin-film compared to that of coating techniques.
As a means to observe the SSLM film formation process in real-time, high speed (1000 frames per second) in-situ microscopy was used. Fig. 3b, 3c, and 3d, 3e are side and top view schematic and video images of the meniscus near the contact line in the presence and absence of pre-existing SSLM thin-film acting as seed particles (i.e., seed film), respectively. In the former case, observation of top and side view real-time videos (Supplementary Movie 4, 5) confirm that LM particles swiftly move towards the contact line and self-assemble into a close-packed film to continue the growth of the seed film. This is on the contrary to the latter case, where the majority of LM particles clump-up on the blade at its edge (Supplementary Movie 6). During solution shearing, the solvent passes through underneath the clumps, ultimately resulting in a non-uniform film with a significant amount of voided regions (Supplementary Movie 7). Our results suggest that for SSLM film formation, apart from the aforementioned effects, the inter-particle attraction is also an important factor in effectively transporting LM particles towards the contact line and self-assembling them into a thin-film.

To achieve lift-off-based photo-patterning of SSLM film, it is critical that the film is coated in the evaporative regime (See Supplementary Fig. 10 for further explanation), where film thickness increases with the decrease of shearing speed (Fig. 3f), and the film grows at the contact line simultaneously with the moving blade (< 2 mm/s). Here, self-assembled packing of LM particles was observed (Fig. 3b, 3c), and such particle-packed morphology (i.e., dry film) maintained after thin-film formation, as shown in Fig. 3g.35 At lower shearing speeds, PSS with long-chain yielded higher film thickness, which can be attributed to the stronger inter-particle attraction. At high coating speeds (> 10 mm/s) (Supplementary Movie 8), known as the Landau-Levich regime, the thickness increases with shearing speed (Supplementary Fig. 11).35 In this regime, the film exhibited bulk-like continuous morphology (Fig. 3h), indicating breakage and merging of LM particles during film formation (see Supplementary Fig. 10 for further explanation). As will be discussed below, this ‘wet film’ was not compatible with the lift-off process. In the wet film, indium oxide peaks were detected in the XPS spectrum; whereas, for dry film coated in the evaporative regime, such peaks were not present (Fig. 3i and Supplementary Fig. 12). The indium oxide likely formed due to the oxidation of EGaIn, as the LM particles were being ruptured and annealed.30

Fig. 3j is the surface roughness profile of SSLM film formed via solution shearing, where the film was coated uniformly over a large-area with an RMS roughness of 710.6 nm. On the contrary, film formed via blade coating could not be deposited uniformly on the substrate (see further discussion in Supplementary Fig. 13 and Fig. 14). Fig. 3k is the pixel color distribution of optical images (25 × 25 mm) of spin-coated and solution sheared films. Spin-coating generated relatively non-uniform film, where the LM particle packing density decreased moving radially away from the center of the substrate (inset of Fig. 3k). These results confirm that solution shearing is, by comparison, a highly feasible SSLM film deposition technique. Furthermore, due to the ink’s excellent wetting ability, the SSLM film can be solution sheared on a variety of surfaces, as depicted in Fig. 3l, making it highly versatile for the fabrication of various devices.

Lift-off-based photo-patterning and multilayer electronics demonstration.
Previous lithographic technique utilized the lift-off process of LM in its liquid state, which required pre-patterned gold to form an alloy with the LM.\textsuperscript{15} Furthermore, because LM in its liquid state is mechanically and chemically unstable, multiscale/multilayer fabrication and direct use as metal contacts is infeasible. Since our technique, on the contrary, utilizes LM as a solid-state film, the aforementioned features are achievable, as standard cleanroom processes (\textit{i.e.}, lithographical patterning, spin coating, and reactive ion etching) can be conducted. \textbf{Fig. 4a} is a photograph of SSLM film-based interdigitated electrodes transferred onto a soft elastomer substrate,\textsuperscript{6,40} which was then placed on a finger (Supplementary Fig. 15 and \textbf{Fig. 16}). \textbf{Fig. 4b} are SEM image and depth profile data of patterned SSLM film with 100 μm line width and spacing, demonstrating high uniformity and cleanly lifted off edges. Bath sonication during lift-off (solvent: acetone and isopropanol) was essential in attaining such edges. \textbf{Fig. 4c} are thin (4 μm) and thick (15 μm) SSLM film before and after the lift-off process. Despite the fact that PR thickness was 3 μm, both the thin and the thick SSLM films lifted off cleanly. These results can be attributed to the selective annealing of the SSLM film at the bottom layer (near the substrate-SSLM film interface), which strongly adheres the bottom layer to the substrate surface while the top layers are effectively detached (Supplementary Fig. 10b). Thicknesses of the SSLM film before (dashed lines), and after the lift-off process are presented in \textbf{Fig. 4d}. The solid horizontal line represents PR thickness. The thicker the SSLM film, the greater the decrease in film thickness after lift-off. \textbf{Fig. 4e} is a map of SSLM film thickness as a function of PR thickness, representing four different experimental results. Squares represent conditions where film coating was not possible as the PR was too thick for SSLM film to fully fill-in the gaps between PR. X's represent conditions where the SSLM film could not be patterned as the SSLM film was too thick. The triangles represent conditions where patterning was possible, yet with low yield, where 10-30% of the features were removed. The circles represent conditions where uniform and cleanly patterned features were observed. These results summarize the importance of tuning both the thickness of the SSLM film and the PR in attaining cleanly patterned SSLM film.

In attaining high-density and high-resolution (< 50 μm) features, stronger ultrasonication was required to attained cleanly patterned SSLM film. However, under these conditions (in the case of using 70,000 MW PSS), peeling-off (blue-bordered area) and rupturing (red-bordered area) of the SSLM film occurred, as depicted in \textbf{Fig. 4f}. \textbf{Fig. 4g} is the peel-off ratio (\textit{i.e.}, ratio of peeled off area to total area) for 40 μm and 100 μm resolution serpentine structure, patterned using SSLM film with 70,000 and 1,000,000 MW PSS. The latter case exhibited a lower peel-off ratio, indicating that high MW PSS is necessary for high-density/resolution patterning. This can be attributed to the stronger inter-particle attraction, as mentioned above. \textbf{Fig. 4h} is a wafer-scale pattern of SSLM film with high-resolution (The smallest line width: 20 μm), obtained using 1,000,000 MW PSS. From this point onward, SSLM film with 1,000,000 MW PSS was used.

Lift-off was conducted after coating a wet film of SSLM (left) and with ink without PSS (right) (\textbf{Fig. 4i}). For both films, continuous bulk-like morphology was attained (Fig. 3h, Supplementary Fig. 7). Since this results in highly attractive interaction within the bulk of the film, both of the films could not be patterned
via lift-off process.\textsuperscript{30} This emphasizes the importance of forming particle-packed morphology for lift-off-based patterning.

In order to fabricate highly-integrated, large-scale electronics, SSLM film should be compatible with conventional cleanroom-based fabrication processes such as multilayer deposition, spin-coating, and reactive ion etching (RIE). Fig. 4j are cross-sectional SEM images of multilayer SSLM films with (right) and without (left) polyimide film in between, confirming that vertically insulating or conductive architectures can be fabricated. In the former case, polyimide needed to be spin-coated (4000 rpm) on top of the SSLM film, and the SSLM film remained firmly attached to the substrate during spinning, owing to the strong adhesion of SSLM film to the substrate surface and the mechanical stability of the film itself (Supplementary Fig. 17). For multilayer lithography, the use of RIE with oxygen plasma is inevitable to remove polymer layers. Unfortunately, RIE can also damage the metal layers, which can deteriorate the electrical performance or even destroy the entire device. The SSLM, on the other hand, maintained the same chemical composition and morphology after RIE. Fig. 4k and Supplementary Fig. 18 are XPS spectra of elements in SSLM before and after RIE. There was no change in XPS spectra, further confirming the stability of SSLM film under RIE. To the best of our knowledge, this is the first demonstration of utilizing standard cleanroom processes to pattern and incorporate LM, opening up a wide variety of possible soft electronic applications that require multilayer, high-resolution, and high-density features over a large-area.

One critical aspect of LM-based electronics is the ease in which it can be rendered conductive (i.e., activated).\textsuperscript{18} Our SSLM film becomes activated during the transfer process or during the substrate peel-off process to a resistivity of 8.67x10^{-7} \, \Omega m. The resistivity can be further reduced by applying strain. Fig. 4l is resistance as a function of time under strain at 30%. When strain is applied initially, the resistance suddenly decreases from 1.445 to 1.034 \, \Omega. Thereafter, the resistance continues to decrease under strain but eventually saturate to 1.032 \, \Omega. This value is maintained and only a negligible (0.001 \, \Omega) resistance change was observed under strain release and reapplication. Hence, our SSLM film-based soft electronics can be activated under normal device operation without the need of additional (e.g., mechanical rubbing) activation steps.\textsuperscript{11, 41}

**Characterization and demonstration of soft electronic with SSLM film.**

As mentioned in Fig. 3l, SSLM film can be directly coated on a variety of substrates. Therefore, SSLM film-based electronics can be directly fabricated on soft substrates such as PDMS (see Supplementary Fig. 19 and Methods section for detailed fabrication steps). This direct coating and patterning on soft substrate improve its yield by skipping the cumbersome transfer-printing step. Also, conventional electronic components (\textmu-LED in this case) can be integrated with SSLM-based soft electronics (Fig. 5a). The inset shows that LEDs can be lit using SSLM as electrodes. Interestingly, as presented in Fig. 5b, our SSLM film demonstrated consistent electrical impedance under various strains and AC frequencies. Typically due to skin effect, metal electrodes, including conventional LM, suffer from an increase in impedance with AC frequency.\textsuperscript{42} The absence of such an effect in our film can be attributed to the
presence of ions, as previously been reported. Such consistency in electrical impedance allows stable operation of electrical circuits under various conditions for different applications. I-V curve of an LED connected to an SSLM film-based stretchable interconnector was measured with and without strain to corroborate stable operation (Fig. 5c). Only a negligible change in current was observed under the application of 50% strain.

The variation in resistance with strain can be controlled by tuning the thickness of the film, as depicted in Fig. 5d. In the case of thick film (15 μm), there is only a negligible change in resistance under the application of strain, making it an ideal material for electrodes or interconnects. On the other hand, the resistance of the thin film (4 μm) increases with strain, which therefore can be used as strain sensors, or deformable heaters. The deviation from theoretical gauge factors for both films indicates that the resistivity is decreasing with strain. The dotted line represents theoretical gauge factor (2), assuming resistivity remains constant. For the thicker film, a higher amount of electrical paths are likely to be generating under strain, which largely compensates for the increase in resistance due to geometrical change. Further investigation is needed to elucidate the exact underlying mechanism, and is the subject of our future work.

Utilizing all of the unique capabilities of our SSLM film, (e.g., multilayer, large-area, high-resolution fabrication, film thickness control, and direct patterning on soft substrates), soft artificial finger that can decouple pressure and strain was demonstrated (Fig. 5e). The artificial finger consisted of two SSLM layers electrically connected through a via hole (the multilayer fabrication process is presented in Supplementary Fig. 20). The first layer consisted of thick SSLM film-based interdigitated electrodes. On the interdigitated electrodes, pyramid structured PDMS coated with a conductive polymer (polypyrrole) was laminated, which together functioned as piezoresistive pressure sensor (Supplementary Fig. 21). The second layer consisted of a thin SSLM film, functioning as a strain sensor. Fig. 5f is photograph of the artificial finger placed on a human finger, and Fig. 5g are real-time measurements of pressure and strain. The resistance between terminals 1’ and 3’ (both belonging to the first layer) decreased with pressure, and the resistance between terminal 2” (belonging to the second layer) and terminal 1’ increased with strain (see Supplementary Fig. 22 for description of device architecture and operation). When pressure and strain are independently applied (blue and red regions respectively), only the corresponding signal undergoes change. 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. Furthermore, unlike conventional LM, one key feature of SSLM film is their high stability under repeated contact with another surface. Fig. 5h is resistance variation of the pressure sensor under repeated application of 30 kPa over 10,000 cycles. Inset in Fig. 5h is SEM image of the pyramid surface after cycling test. Despite the repeated contact with the pyramid surface, the SSLM film did not rub off of the substrate. We also demonstrated closed-loop antenna with SSLM film, which can be applied to soft-wireless devices (Supplementary Fig. 23). Such a demonstration was achievable owing to the aforementioned attributes of our SSLM film.
Demonstration of skin-interfaced electronics with SSLM film.

Softness, deformability, biocompatibility, and excellent conductivity of SSLM film make it a compelling option for the construction of bio-integrated electronics.\textsuperscript{3, 14, 25, 49} To demonstrate its potential, we built skin-interfaced wearable electronics for electrophysiological monitoring. More specifically, we fabricated fully-rubbery surface electromyography (sEMG) sensor, integrating SSLM film as skin-interfacing electrodes and as interconnects, as presented in Fig. 6a. Here, by using a mask aligner, SSLM electrode patterns were connected to a conventional wireless system via anisotropic conductive film. The EMG sensor consists of three soft electrodes, which are used for ground, reference, and measurement. The intrinsically flexible, stretchable nature of the sensor allows not only intimate integration with the curvilinear surface of the skin but also dynamic adaptation to skin deformation (Fig. 6b), thereby allowing stable measurement of sEMG signals. Real-time monitoring of sEMG signals on a forearm with repeated wrist bending motions verifies capability of the SSLM skin sensor for high-quality electrophysiological measurements (Fig. 6c). For measuring electrophysiological signals or for delivering electrical stimuli to the skin, direct contact of electrodes with bio-surface is inevitable. In this regard, the stability of the electrode should be examined. The SSLM film does not leave any residue on the skin after intimate contact with the skin for several hours, as seen in Fig. 6d. Furthermore, several studies have shown biocompatibility of GaIn-based LM, suggesting bio-safety of SSLM for use as a skin-interfaced material.\textsuperscript{25, 48} These demonstrations together further confirm the potential applicability of SSLM as bio-integrated electronics, which often requires high-resolution features, metallic electrical properties, while maintaining soft, biocompatible, and stable properties.\textsuperscript{50}

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. Herein, liquid metal is the most appropriate material for interconnects and electrodes, and their applicability relies on their compatibility with well-established cleanroom based photolithographic patterning, as large-area, multilayer, multiscale, and high-resolution features are needed for integration with conventional electronic systems. These features have not yet been realized due to the difficulty of making mechanically and chemically stable LM film uniformly over a wafer-scale that can also be lithographically patterned. Our new ink formulation stabilizes the LM particles and enables the formation of highly uniform and stable solid-state film using solution shearing. We also confirmed that the particle-packed morphology of SSLM film achieved by coating under the evaporative regime allows photo-patterning with a conventional lift-off process. The film can be coated and patterned on a variety of substrates, and its thickness control enables tuning of conductance. These attributes were utilized to fabricate SSLM multilayer interconnects, tactile sensors, and skin-interfacing electrodes for sEMG measurements. We anticipate that our technique will provide the pathway for significant advancement of 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 acquired from Sigma-Aldrich unless otherwise mentioned. To prepare and characterize the LM ink, eutectic gallium indium alloy (EGaIn, Rich-Metals, China), poly(styrene sulfonate) (PSS, with an average molecular weight of 70,000 and 1,000,000), acetic acid (99%), ethanol (SAMCHUN, 99.5%), acetone (SAMCHUN, 99.5%), isopropyl alcohol (IPA, SAMCHUN, 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,2H-perfluorooctyl)silane (PFOCTS, 97%), poly(methyl methacrylate) (PMMA), anisole (99%), polyamic acid solution (PI), PDMS (Sylgard 184, Dow Corning), NR-9 3000 and 8000 py (Lift-off PR, Futurrex). For fabrication of pressure sensor, pyrrole monomer, polypyrrole solution, and Iron(III) p-toluenesulfonate hexahydrate were used.

Preparation of functionalized LM particles-embedded ink for solution shearing. Bulk LM (1.4 g), and 0.07 g (7×10^-8 mol) of PSS were dissolved in diluted acetic acid (5 vol% in DI water, 2 ml). This compound solution was tip sonicated (VC 505, Sonics & Materials) for 30 minutes unless stated otherwise.

SSLM film coating via solution shearing. The LM ink was coated on the various substrate (glass, PET film, Si wafer, PI film, PR film, PDMS, Au) by using a customized shearing machine. Before solution shearing, samples 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 × 75 mm/ 50 mm × 75 mm) were utilized 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 the films coated. 100 μl of LM ink was injected between the coating blade and substrate right before solution shearing. After the shearing process, SSLM 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 vertically standing), and for the wet film coating, the speed of moving substrate increased to > 10 mm/s.

SSLM film patterning with lift-off process. For patterning of the SSLM film, NR9-3000py, and 8000py (lift-off PR) were spin-coated and selectively exposed to UV light using a mask aligner (MJB4). 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 MIF 300 developer for 50 sec. Finally, the substrate was exposed to oxygen plasma treatment for surface activation. Then, SSLM film was coated on the PR patterned substrate via solution shearing as described above. SSLM film coated sample was immersed in acetone/IPA (4:1) and sonicated in bath sonication for lift-off.

Fabrication of soft electronics through transfer printing of patterned SSLM film. Schematic illustration in Supplementary Fig. 15 depicts the overall patterning and transfer printing process of SSLM film. For this process, PMMA working as a sacrificial layer, PI working as a temporary substrate, and PR layers for
patterning are coated on the substrate (glass, silicon wafer) sequentially. PMMA solution (4 wt% dissolved in anisole) was spin-coated (3000 rpm, 30 sec) on oxygen plasma-treated substrate. Subsequently, PMMA was then annealed at 180 °C for 3 min, and the edges were removed with acetone. PI solution was spin-coated onto the PMMA-coated glass at 4000 rpm for 60 sec and cured at 250 °C for 60 min. Thereafter, PR patterning, SSLM film coating, and lift-off-based patterning were conducted, as described above. Once SSLM film was patterned, PMMA sacrificial layer was removed by putting the sample in the acetone overnight. Then, PI film with patterned SSLM film was delaminated from the original substrate and transferred to a soft substrate by stamping or using conventional thermal releasing tape. After transferring, oxygen plasma etching was conducted (100 sccm, 200 W, 1 hour) to remove PI film.

Fabrication of soft electronics through direct patterning of SSLM film. Schematic illustration in Supplementary Fig. 19 depicts the overall direct fabrication process of soft electronics with SSLM film. Before coating PDMS film on 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 PI tape prior to 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, 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 cured at 70 °C to form a PDMS film. Subsequently, PI solution was spin-coated 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. Afterwards, oxygen plasma etching was conducted (50 sccm, 200 W, 30 min) to remove PI film in the regions absent of PR. Subsequently, SSLM was coated and patterned directly on the PDMS film by following the abovementioned methods. Remaining PI was removed by oxygen plasma etching (100 sccm, 200 W, 1 hour). For the multilayer structure, we repeated the same sequence from the PI coating on the SSLM patterned PDMS. Finally, the sample was cut along the hydrophobic/hydrophilic border, and the PDMS was lifted off of the glass substrate.

Fabrication of pressure sensor. Schematic illustration in Supplementary Fig. 21 depicts the overall fabrication process of the conductive polymer-coated pyramid pressure sensor. For the structuring of the pyramid pattern, the silicon substrate is chemically etched as previously reported and subsequently coated with PFOCTS to facilitate the delamination of PDMS film. 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. PDMS was cured at 80 °C for 30 min and was peeled off from the mold. Then, the surface of pyramid patterned PDMS film was treated with oxygen plasma to activate the 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%) with the presence of Iron(III) p-toluenesulfonate hexahydrate catalyst (1.6 wt%) for 12 hours.

Characterization. Chemical, rheological, and morphological characterization. Zeta potential values of LM inks were measured by DLS (Zetasizer nano zs, Malvern). Each ink was characterized by 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 LM inks with different proportions of PSS was measured by MCR 302 rheometer (Anton Paar) at a shear rate of $10^2 \sim 10^2$ s$^{-1}$ at room temperature. To determine the wettability of each ink, the contact angle was measured through a contact angle analyzer (SEO Phoenix). Measurements were conducted two times for each sample: when 50 $\mu$l of the sample was dropped and when 25 $\mu$l of that was withdrawn.

To observe the morphology of the coated film, SEM images were taken by S4800 (Hitachi). Surface morphology and roughness of the films were analyzed by a 3D surface profiler (TOF-SIMS5, ION-TOF GmbH).

**Electrical Characterization.** To measure the resistance, impedance, and the capacitance LCR meter (4284A, HP) was used. The samples were cut to the same size (1 mm $\times$ 25 mm) unless stated otherwise. I-V curve under application of strain was 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 stand with a motor (Mark-10), and a customized manual stain machine. The reflection coefficient of the SSLM-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 RF cable.

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

**Experiments on human subjects** All experiments 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.

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**Declarations**

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Author contributions

G.-H.L., J.-W.J. and S.P. conceived the concept and designed experiments. G.-H.L. conducted all the experiments and performed the electrical characterization. H.K. conducted ink preparation and film coating and performed ink and thin-film characterization. J.L. assisted soft electronics fabrications and sEMG measurement. H.K. and J.K. provided comments regarding data analysis. C.Y and S.P. were involved in the soft electronics demonstration. Z.B. provided comments regarding experiment configuration and manuscript preparation. J.-W.J. and S.P. were responsible for managing all aspects of this project. G.-H.L. and H.K. wrote the draft and Z.B., J.-W.J. and S.P. revised the manuscript. All authors discussed the results and the manuscript.

Additional information

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Figures
Figure 1

Fabrication process of solid-state liquid metal (SSLM) film-based soft electronics and demonstration of key features. a,b,c, Schematic illustration of SSLM film-based soft electronics fabrication process. (a) Preparation of functionalized LM particles-embedded ink. (b) SSLM film coating with solution shearing. (c) Lift-off-based patterning of SSLM film. d, Demonstrations of SSLM film-based fabrication. (i) Thickness control: SEM images of thick (top) and thin (bottom) patterned SSLM film. Scale bar, 5 μm. (ii) Multiscale processing: photograph of patterned high-resolution SSLM film on 4-inch wafer. Scale bar, 2.5 cm. (iii) Multilayer fabrication: optical microscopy (OM) image of the stacked structure made with SSLM film. Scale bar, 400 μm. (iv) Integration with soft substrates: photographs of SSLM-based serpentine pattern on PET (left) and gelatin-based biogel (right).
Figure 2

Characterization of functionalized LM ink for solution shearing. a, Schematic illustration of LM particle formation by tip sonication with the addition of a bridging polyelectrolyte (PSS) acetic acid (AA). b, Photographs and zeta potential values of inks with different components. c, UV-Vis absorbance spectra of inks with different combinations. d, Apparent viscosities according to a shear rate of LM inks with different PSS concentrations. Inset is a conceptual depiction of viscosity as a function of particle size variability in the suspension. e, Contact angles of LM ink droplet without (left) and with (right) PSS before (blue) and after partial ink withdrawal (red). The contact angle of bare LM is presented as a black dotted line. Bottom photographs are droplet images with contact angles after withdrawal. f, 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 LM ink dispersed in DI/PSS. Scale bar, 2 cm. Inset (right): SEM image of the film with LM ink dispersed in DI/AA. g, Photograph of SSLM film coated on glass with LM ink dispersed in DI/AA/PSS. Inset: SEM image of SSLM film. Scale bar, 30 μm. h, SEM images of SSLM film with different molecular weights (MW) of PSS. Scale bar, 25 μm. i, Particle size distribution of inks with the two different PSS MW, at various tip sonication times. Inset: SEM images of LM particles (PSS MW 70,000) and OM images of line patterned SSLM film with different sonication times. j, Optical image color distribution of each film. Uniform SSLM film shows a sharp peak in color distribution. Solvents were diluted (5 vol%) solutions in DI water. k, SEM images of coated films with different solvents. All films are coated on a glass substrate with the same condition. Scale bar, 5 μm.
Figure 3

Solution shearing of LM ink and characterization of SSLM film. a, Schematic illustration of SSLM film deposition through solution shearing process. b, c, Schematic illustration, and OM images of SSLM film formation during solution shearing with pre-coated seed particles. Side view (b) and top view (c). d, e, Schematic illustration, and OM images of SSLM film formation during solution shearing without seed particles. Side view (d) and top view (e). f, Film thickness vs. shearing speed for LM inks with different PSS MW. Inset is a log scale fitting graph to calculate the power scale. g, Cross-sectional SEM images of dry SSLM film with different shearing speeds. Scale bar, 10 μm (top), 5 μm (bottom). h, Cross-sectional SEM image of the wet film. Scale bar, 50 μm. Inset: photograph of the wet film after solution shearing at high speed (> 10 mm/s). i, XPS spectrum of the wet film and the dry film on In 3d. j, 3D surface profile data of SSLM film coated with solution shearing. k, Optical image color distribution for solution sheared and spin-coated film. Inset: SEM image of the spin-coated film. Scale bar, 30 μm. l, Photograph of SSLM film coated on various substrates. Scale bar, 3 cm.
Figure 4

Lift-off-based photo-patterning of SSLM film and its characterization. a, Photograph of SSLM film-based interdigitated electrode on soft substrate. b, SEM image and depth profile data of patterned SSLM film. c, Cross-sectional SEM images before (left) and after (right) patterning with different thicknesses of SSLM film. Scale bar, 60 μm. d, Depth profile data of SSLM film after lift-off process. Dashed lines indicate SSLM film before lift-off and solid orange line indicates PR thickness. e, Patterning feasibility based on PR and SSLM thickness. Only the conditions represented by circles yielded fully patterned features. f, SEM image of SSLM film, which contains ruptured (red-bordered) and peeled off (blue-bordered) regions. Scale bar, 10 μm. g, Peel-off ratio of patterned SSLM film with different resolutions. Insets are SEM images of serpentine structure with 40 μm resolution for different PSS MW. h, Photograph of patterned SSLM film on a 4-inch Si wafer. Scale bar, 6cm. Inset: zoomed-in OM image of the patterned SSLM film (The smallest line width: 20 μm). Scale bar, 200 μm. i, Photographs after lift-off process of wet film and film coated with LM ink that does not contain PSS. Without maintaining its particle-packed morphology, lift-off-based patterning was impractical. j, Cross-sectional SEM images of the electrically connected multilayer structures (left), and insulated multilayer structure with polyimide in between (right). Scale bar, 2 μm. k, XPS spectrum of SSLM film before (black) and after (red) reactive ion etching (RIE) on In 3d and Ga 2p. There is no significant difference in XPS peaks before and after RIE. l, The resistance change of SSLM film versus time under strain and release.
Figure 5

Electrical characterization of SSLM film and various soft electronic demonstrations. a, Photograph of large-scale soft electronics with SSLM film-based interconnect connected to a μ-LED. Inset is the μ-LED being turned on. Scale bar, 3 cm. b, Impedance variation of SSLM film and bare LM film as a function of strain and frequency. c, I-V curve of the LED connected to a stretchable SSLM film-based interconnector with (red) and without (black) 50% strain. d, Relative resistance change versus strain of a thick (~15 μm) and a thin (~4 μm) SSLM film. Inset presents the initial resistances of thick and thin SSLM films. e, Illustration of multilayer SSLM film-based artificial finger that can measure and decouple pressure and strain. Pyramid-patterned PDMS coated with polypyrrole was laminated on the interdigitated electrode for pressure sensing. Inset is a cross-sectional SEM image of a stacked structure. f, Photograph of SSLM-based artificial finger placed on a glove. g, Real-time monitoring of pressure and strain with SSLM-based artificial finger. Pressure and strain are successfully decoupled. h, Resistance change of the pressure sensor upon repeated application of 30 kPa of pressure for 10,000 cycles. Inset is an SEM image of a pyramid structured pressure sensor after the cycling test.

Figure 6
Demonstration of a fully-rubbery SSLM device for applications in skin-interfaced wearable electronics. 

a, Illustration of SSLM film-based skin electronics for measuring surface electromyogram (sEMG). Insets are photograph of the sensor (left) and OM image of SSLM film connected to Au pads using an aligner. 
b, Photograph of SSLM film-based skin electronics attached on the subject’s wrist. 
c, Real-time monitoring of sEMG upon wrist bending. 
d, Photograph of the wrist after peeling off the SSLM-based skin electronics. No residue remained after peel-off.

Supplementary Files

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