Stretchable Polymerized High Internal Phase Emulsion Separators for High Performance Soft Batteries

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Powering soft embodiments of robots, machines and electronics is a key issue that impacts emerging human friendly forms of technologies. Batteries as energy source enable their untethered operation at high power density but must be rendered elastic to fully comply with (soft) robots and human beings. Current intrinsically stretchable batteries typically show decreased performance when deformed due to design limitations, mainly imposed by the separator material. High quality stretchable separators such as gel electrolytes represent a key component of soft batteries that affects power, internal resistance, and capacity independently of battery chemistry. Here, polymerized high internal phase emulsions (polyHIPEs) are introduced as highly ionically conductive separators in stretchable (rechargeable) batteries. Highly porous (>80%) separators result in electrolyte to polyHIPE conductivity ratios of below 2, while maintaining stretchability of ≈50% strain. The high stretchability, tunable porosity, and fast ion transport enable stretchable batteries with internal resistance below 3 Ω and 16.8 mAh cm⁻² capacity that power on-skin processing and communication electronics. The battery/separator architecture is universally applicable to boost battery performance and represents a step towards autonomous operation of conformable electronic skins for healthcare, robotics, and consumers.

Soft and malleable forms of wearable electronic interfaces, from on-skin medical sensors to electronic skins (e-skins) for virtual reality, lead to new possibilities in healthcare and unprecedented user experiences. However, the collective power consumption of sensors, displays, and data transmission requires amounts of energy that only batteries can deliver wirelessly and continuously. Ideally, wearable applications or even (soft) robots distribute their energy sources over large areas available on the skin or within the body. Intrinsically stretchable batteries effectively make use of available space while perfectly complying to deformations and maintaining comfort for the wearer.

Recent developments exploit high-energy-density materials, such as Zn and Li-ion based batteries, to enhance battery capacity and close the gap to rigid-island designs. Despite the progress in capacity and rechangeability, the principle design of planar intrinsically stretchable batteries has largely remained unchanged. The interplay of electrodes, efficient current collectors, highly ionically conductive separators/gel-electrolytes, and their conformal orientation must collectively be optimized to boost battery performance. Promising solutions for single components, including multilayer current collectors, tough electrodes, and self-healing gel-electrolytes were demonstrated recently. However, many prototypes still use the coplanar orientation of electrodes as proposed for the first soft batteries. This design sacrifices performance due to increased ionic pathways in the gel electrolytes. Introducing a sandwich design of stacked battery components, greatly reduces ionic pathways and therefore the internal resistance of the battery. Soft (sandwich-design) batteries require electron-insulating separators, which have to meet high ion conductivity, extensibility, and chemical inertness. Hydrogels are often used in water-based systems, serving as gel...
electrolyte and separator at the same time. Although hydrogels contain large amounts of electrolyte (>80%), they have an intrinsically lower conductivity due to interactions of ions with the hydrophilic polymer network. They are also limited to water-based electrolytes, impeding the use of ionic liquids. An ideal separator however requires battery-chemistry independent formulations with high processability and scalability to be applicable for various types of stretchable batteries.

Highly porous membranes produced by polymerization of high internal phase emulsions (polyHIPEs) represent a promising materials' choice for energy storage devices. Due to extremely high porosities of 75–95%, polyHIPEs are being explored across many fields including tissue engineering, supported liquid membranes for the extraction of metal ions from solutions, drug delivery systems, and as spring elements in flexible energy harvesting devices. Without addressing stretchability, polyHIPEs were proposed as separators for Li-Ion batteries and supercapacitors. Introducing stretchability to highly porous battery separators remains challenging. First demonstrations utilize non-solvent internal phase separation (NIPS) to create separators with 60% porosity. Despite good stretchability, the ionic conductivity of those NIPS is still lower as the conductivities found in polyacrylamide hydrogels. Highly stretchable polyHIPEs with porosities greater than 75% can provide record high ionic conductivities, yet they are unexploited for the use in stretchable batteries.

We here introduce widely applicable materials choices, general design rules, and fabrication approaches for intrinsically stretchable batteries with unmatched low internal resistance for wearable electronics. Utilizing highly porous stretchable polyHIPEs as separator in a sandwich-design battery leads to unprecedented low electrolyte to separator conductivity ratios and increased performance when mechanically deformed. Our polyHIPEs are readily incorporated in stretchable zinc-carbon batteries as synthesized, without the need for drying or internal phase extraction by a solvent and subsequent soaking in ionic liquids. Despite containing up to 85% electrolyte in their pore structures, our separators are reversibly stretchable to up to 50% strain, which exceeds occurring strains of human skin. Readily scalable fabrication of our soft batteries will enable widespread employment from industry and healthcare to consumer electronics. Seeking imperceptible human-machine interfaces, we demonstrate an untethered, wearable, and soft electronic (audio) controller that wirelessly communicates via Bluetooth with any suitable receiver (audio systems, smartphones, and computers).

The fabrication of polyHIPEs involves a base monomer, forming the continuous yet minority external phase of an emulsion template, which is polymerized in the presence of liquid electrolyte (used as internal phase). The emulsion template is formed by dropwise addition of the internal to the continuous phase under constant stirring and is stabilized using a surfactant. The continuous phase of polyHIPEs can consist of a vast number of different polymers. PolyHIPE research focuses strongly on the use of styrene (St) and divinylbenzene (DVB) as monomers, resulting in friable and brittle open-porous structures. This limitation is resolved by the use of 2-ethylhexylacrylate (EHA) as comonomer. Increasing EHA content results in lower glass transition temperatures, yielding elastomers characterized by low Young's moduli.

We use urethane diacrylate oligomer as crosslinker, which was shown to result in robust and tough polyHIPEs, diluted with high amounts of EHA (>50 wt%) as monomer containing small amounts of surfactant as continuous phase. After dropwise addition of (NH₄Cl/ZnCl₂) electrolyte the resulting viscous emulsion is doctor bladed into thin sheets, which are UV polymerized (Figure S1, Supporting Information). This polymerization leads to a high number of interconnected pores that render an open-porous structure enabling fast ion transport through the membrane (Figure 1b). Quantitatively, a separator’s ability for fast ion transport is described by the MacMullin number,

\[
N_M = \frac{\sigma_0}{\sigma} = m \cdot \frac{\rho}{\rho_0}
\]  

(1)

which is a measure for the ratio of electrolyte conductivity \(\sigma_0\) and separator conductivity \(\sigma\) (electrolyte-filled separator). Using Archie’s exponent for porous structures \(m\), it can be related to the porosity \(p\), being the volume ratio of internal phase to total emulsion volume, using a power law, with \(\rho_0\) and \(\rho\) being the specific resistivity of the free electrolyte and separator respectively.

\[
\sigma = \frac{\rho}{\rho_0} = \frac{p}{\rho_0} \cdot \frac{\sigma_0}{\sigma}
\]  

(2)

For achieving high-rate performance the separator resistance has to be minimized, which for a given electrolyte system corresponds to minimizing the MacMullin number. Values for existing stretchable battery separators are plotted versus separator conductivity in comparison to our results (Figure 1c). With our

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**Figure 1.** Stretchable battery with highly conductive polyHIPE separator. a) A sandwich-design stretchable primary cell with polyHIPE separator. b) Ions travel freely through the open porous structure of the polyHIPE. c) Comparison of separator materials. Most suitable for batteries are separators that combine high ionic conductivity with low MacMullin number.
polyHIPEs we achieve a MacMullin number of less than 2, unmatched by any other stretchable separator technology.\[8–10,40–48\] Introducing potassium hydroxide (KOH) as electrolyte boosts the separator conductivity to record high $0.39 \pm 0.05 \text{ S cm}^{-1}$ while keeping the MacMullin number below 2, corroborating the performance increase achievable with polyHIPE separators.

The unprecedented low MacMullin numbers stem from the high porosity and the interaction-free (with the polymer matrix) transfer of ions. While the porosity is tuned by the phase volume ratio of the emulsion, the size distribution of the spherical pores is highly non-uniform (Figure 2a). Fabricated membranes with 75%, 80%, and 85% porosity have pore-sizes in the range of $\approx 5 \text{–} 30 \mu\text{m}$ (Figure 2b), similar to other reported battery separators.\[10,46,49\] We characterize the conductivity of our separators with impedance spectroscopy utilizing a custom-made measuring cell (Figure S2, Supporting Information). The specific resistance of the electrolyte-filled separator and of the electrolyte itself is determined from the high-frequency plateau of the real part of the impedance (Figure 2c). Therefore, we model the impedance response of the whole measurement cell using an appropriate equivalent circuit, and calibrate the cell with liquids of known conductivity (Figure S3, Supporting Information). Increasing the polyHIPE porosity from 75% to 85% results in a decrease of the specific resistance from

Figure 2. Electro-mechanical characterization of polyHIPE separators. a) SEM image of the crosssection of a polyHIPE sample with 85% porosity. b) First-order pores show numerous interconnecting pores. c) Real and imaginary part of impedance as measured for a polyHIPE with porosity of 75% as well as the corresponding regression with an equivalent-circuit model. d) Specific resistance decreases with increasing porosity. e) Tensile stress–strain curves of polyHIPEs of varying porosity. Variation (gray) is small around a third-order fit (colored). f) Maximum strain increases with porosity while the Young’s modulus decreases.
14.1 ± 1.1 to 11.1 ± 1.7 Ω cm, following a power law (Equation 2). MacMullin numbers are as low as 1.65 ± 0.29, with the specific resistance of the electrolyte being 6.78 ± 0.56 Ω cm (Equation 1).

Uniaxial tensile tests of polyHIPEs demonstrate increasing maximum strain versus porosity, reaching 49 ± 4% at 85% porosity (Figure 2e). These strain levels exceed those naturally occurring in human skin and make them perfectly suitable for wearable applications. The foam-like stress-strain behavior shows low Young’s moduli in the range of a few hundred kPa. PolyHIPEs get softer with increasing porosity, reflecting the volumetric replacement of solid polymer with internal phase (Figure 2f). Repeatedly stretched polyHIPE samples exhibited no visible fractures or irreversible deformations of pores after more than 300 stretch-release cycles as evidenced by SEM analysis (Figure S4, Supporting Information).

We test the performance of the polyHIPE separators (85% porosity) in stretchable primary cells serving as a model system (Figure 3a). The polyHIPE separates paste-based electrodes, which work in combination with ultrathin wrinkled metal-foils as current collectors. Those foils feature high conductivity and stretchability at the same time, are fabricated in a scalable approach, and are unsusceptible to damage upon repeated deformation.[50] Zn powder bound in the electrolyte solution with xanthan gum forms the anode, a mixture of carbon powder, manganese dioxide, and electrolyte solution the cathode. The polyHIPE separator is bonded to the elastomeric casing via a cyanoacrylate dispersion to prevent delamination (Figure S5 and S6, Supporting Information). We investigate the characteristic V–I curve of our intrinsically stretchable primary cell in both pristine and stretched state, showing linearity over the whole range for all states (Figure 3b). While the open-circuit voltage decreases with strain, the short-circuit current increases and reaches a plateau at ~20% strain (Figure 3c). Overall, the internal resistance is reduced from 3.4 Ω in the relaxed state to as low as 2.1 Ω when fully stretched (Figure 3d). An initial rise is observed during clamping of the battery into the extensor. Our batteries prove their resilience even under dynamically varying conditions, when discharged with 10 mA under repeated deformation between 0% and 20% strain (Figure 3e). The stretchable thin-film current-collectors prove to be intact even after 600 repeated stretch-release cycles to 20% strain (Figure S7, Supporting Information). The wrinkled metal-foils exhibit no significant fatigue under deformation of an assembled battery, also reflected by the absence of any noticeable changes in electrode resistance. Constant–current discharging (10 mA) of our batteries under a constant deformation (20% strain) leads to a specific capacity of 16.8 mAh cm⁻² (Figure 3f). The universal battery architecture readily allows fabrication of secondary cells. Replacing ZnCl₂/NH₄Cl electrolyte with 3.6 m KOH renders a rechargeable alkaline battery with characteristic performance. As typical for this system,[51] the normalized capacity drops to 50% after ~50 charge-discharge cycles with 2 mA current (Figure 3g,h). This however corresponds to a fivefold life-time increase compared to similar approaches towards stretchable, rechargeable alkaline batteries.[52] Higher energy densities and more charge cycles are possible with lithium ion based chemistries. PolyHIPE separators are compatible with state-of-the-art Li-based electrolytes containing Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), rendering them applicable for chemistries that utilize LiMn₂O₄-cathode and V₂O₅-anode compositions,[10] or NMC/graphite-cells using graphite anodes and Li[Ni₀.₅Mn₀.₅Co₀.₁]O₂-cathodes.[53] In combination with 11 m LiTFSI we find a conductivity of 9.8 ± 1.2 mS cm⁻¹, resulting in a MacMullin number as low as 3.0 ± 0.7 (Figure S8, Supporting Information).

Figure 3. Stretchable battery with low internal resistance. a) Stretchability of zinc–carbon battery with polyHIPE separator. b) Voltage–current characteristics of the battery mounted in an extensor, 20% stretched, and as prepared. c) Open-circuit voltage and short-circuit current versus strain. d) Internal resistance decreases during stretching of the battery. e) Constant-current discharge with 10 mA under repeated cyclic strains of 20%. f) Constant-current discharge with 10 mA under 20% constant strain. g) Normalized capacity of a rechargeable alkaline cell over 125 charge–discharge cycles. h) Voltage and current of charge–discharge cycles at half-life of the battery.
Intrinsically stretchable batteries deploy their full potential when used as power source for on-skin electronics. Their soft embodiment makes use of large areas that are available on the skin and enable distributed energy supply as it is found in nature. We demonstrate the applicability of our batteries by integrating them in a soft and stretchable electronic skin (e-skin), which is used as an untethered communication device for audio control (Figure 4a,b). A Bluetooth low energy (BLE) microchip reads input signals from five touch sensors, processes the data, and sends control commands to a paired Bluetooth receiver, for example a multimedia system like a smartphone (Figures S9 and S10, Supporting Information). Two soft batteries are connected in series to power the logic chips with 2 V voltage. The constant voltage supply is achieved by introducing a voltage regulating circuit beforehand. All interconnects are fabricated in a meander shape and rendered stretchable to accomplish a completely conformal electronic skin (Figure 4c). Songs that are played on a smartphone are paused, volume controlled, or switched using the respective buttons on the e-skin (Figure 4d). With an average power consumption of 16 mW (8 mA at 2 V), the microchip can be operated for ≈2 h with our battery. Longer operation

Figure 4. Untethered stretchable audio controller. a) Schematic of the controller architecture and function. Touch input triggers wireless transmission of a control command to a receiving audio device. b) Soft electronic skin fixed on the human arm. c) Deformation of the audio controller during operation. d) Operation of the soft audio controller and respective changes on a smartphone.
times are readily achieved by connecting more batteries in parallel. In summary, we report a new type of stretchable highly porous separator for soft batteries, with unprecedented low MacMullin number of 1.65. We fabricate the separator from polymerized high internal phase emulsions (polyHIPEs) with a porosity of 85% to achieve beyond skin-like stretchability of 50% strain. Utilizing this separator in intrinsically stretchable batteries lowers their internal resistance to less than 3 Ω and even further under deformation. We use these batteries to power an unattended audio controller that is worn on the body to demonstrate autonomous operation of fully stretchable electronics. The controller includes Bluetooth data transmission, processing, and five free-programmable input sensors to enable wireless communication with any receiver device. Stretchable polyHIPEs emerge as a material class for highly ionically conductive separators for soft batteries.

Experimental section

Materials: All chemicals were used as received without further purification. PolyHIPEs consisting of 2-ethylhexylacrylate (EHA, Sigma Aldrich, Germany), urethane diacrylate (UDA, Ebecryl 8402, Allnex, Germany), and polymeric surfactant (Hypermer B246, Croda International Plc, UK) were UV-photo-polymerized using the photoinitiator phenylbis(2,4,6-trimethylbenzoyl)phosphine oxide (irgacure 819, Sigma-Aldrich) photoinitiator. Ammonium chloride (NH₄Cl, Sigma Aldrich), and zinc chloride (ZnCl₂, YVR, USA) were used as electrolyte solution. Zinc powder (Grillo-Werke AG, Germany), xanthan powder (Sigma Aldrich), manganese(IV) oxide (MnO₂, Sigma Aldrich), polyacrylic acid (PAA, Sigma Aldrich), and acetylene carbon black (ABC R GmbH & CoKG, Germany) were used for the battery electrodes. Ecoflex 00–30 (Smooth-On, USA), acrylic elastomer adhesive (VHB 467MP, 3M, USA), and 1200-OS Primer (Dow Corning, USA) were used for the battery casing and the electronic skin. 2-ethyl-cyanoacrylate (RS, UK) and 2,2,4-trimethylpentane were used (C₈H₁₈, Sigma Aldrich) as an adhesive for the battery. A 1:4 mixture of 2-ethylhexylacrylate (EHA, Sigma Aldrich) solution was used for fabricating polyHIPEs holding a high-conductivity internal phase used for conductivity measurements and secondary cells. A 11 m bis(trifluoromethane)sulfonimide lithium salt (LiTFSI, Sigma Aldrich) solution was used for fabricating polyHIPEs applicable for Li-ion batteries.

Poly(EHA-co-UDA)/HIPE Synthesis: An electrolyte solution was prepared by mixing 60 wt% DI-water, 5 wt% NH₄Cl, and 35 wt% ZnCl₂ representing the internal phase of the polymer. The external phase consisting of 54.1 wt% EHA, 41.09 wt% UDA, 4.79 wt% Hypermer B246, and 0.02 wt% irgacure 819 was stirred continuously using a magnetic stirrer at 350 rpm, while being purged with nitrogen. The internal phase was added dropwise to the external phase until 75 vol%, 80 vol%, and 85 vol% were reached, respectively. The emulsion was coated on a glass plate and cured under UV-light (365 nm, LED-Spot 100 with LED Powerdrive 40, Dr. Hönle AG, Germany) resulting in a layer of 0.9 mm thickness.

High conductivity PolyHIPE separators were prepared as described above with additional exchange of the ZnCl₂/NH₄Cl electrolyte with 3.6 m KOH solution. The electrolyte was exchanged by soaking the separators in deionized water for 12 h four times, and subsequently soaking the separators in 11 m LiTFSI solution for 12 h each.

Scanning Electron Microscopy: Samples were gently dried with tissue and stored at ambient conditions for a week. Additionally, the samples were prepared by thermally evaporating 10 nm (0.1-0.5 nm s⁻¹, 1 x 10⁻⁶ mbar) of Au (Oegussa, 99.99%) to improve surface conductivity. Measurement was performed using a Zeiss 1540 XB CrossBeam scanning electron microscope with acceleration voltage of 3 keV.

Impedance Spectroscopy: A custom-made measurement cell was used to determine the impedance of electrolyte-filled polyHIPE membranes. The impedance was measured between 1 mHz and 10 MHz using an impedance analyzer (Novocontrol Alpha A Analyzer) connected to the cell. If not stated otherwise, all measurements were conducted at 25 °C applying 1 mV AC Voltage. The bulk resistance was determined from the high frequency plateau of the real part of the impedance. The conductivity and specific resistance were calculated by using the bulk resistance and the membrane geometry.

Mechanical Characterization: Tensile tests were conducted with a dual column universal testing machine (2005, Zwick Roell) equipped with a 100 N load cell at a constant speed of 50 mm min⁻¹. Test samples containing electrolyte were laser cut (Trotec Speedy 300, CO₂-laser) into a standardized dumbbell-shape geometry (EN ISO527-2:1996(SA)). Fatigue tests were performed by stretching polyHIPEs 300 times to 20% strain at a speed of 0.2 m s⁻¹. SEM images were recorded of unstrained and strained samples.

Battery Fabrication: The anode paste was prepared by mixing of 62 wt% zinc powder, 34 wt% electrolyte solution, and 4 wt% xanthan powder. The cathode paste was prepared by mixing 12 wt% manganese(IV) oxide and 12 wt% acetylene carbon black in 76 wt% electrolyte solution. For the current collectors, a chromium-copper bilyayer (3 nm Cr, 0.2 nm s⁻¹ 300 nm Cu, 3 nm s⁻¹) was used as anode and a chromium–gold bilyayer (3 nm Cr, 0.2 nm s⁻¹ 200 nm Au, 3 nm s⁻¹) used as cathode were thermally evaporated on 1.4 μm thick PET foils. Ecoflex 00–30 was degassed and cast between two sheets of acrylic elastomer adhesive that were treated with 1200-OS primer. The stack was cured for 3 h at 70 °C resulting in elastomer sheets of 0.5 mm thickness. Parts that enclose the battery were laser cut (Trotec Speedy 300, CO₂-laser). Current collector stripes were applied to the prestretched (200% strain) closing lids and relaxed afterwards, forming wrinkles. A 1:4 mixture of cyanoacrylate/trimethylpentane dispersion was spin-coated (10 rps for 5 s) on the elastomer casing in contact with the separator, resulting in instant tough bonding of the two materials. The stretchable battery was assembled layer by layer.

Secondary Cells: Fabrication steps of secondary cells are as described above, utilizing 3.6 m KOH as electrolyte. PolyHIPE separators of 85% porosity filled with KOH electrolyte and slightly modified anode and cathode paste were used. The anode was prepared by mixing of 47 wt% zinc powder, 47 wt% electrolyte solution (3.6 m KOH), and 6 wt% PAA. The cathode paste was prepared by mixing 11.6 wt% manganese(IV) oxide, 5.7 wt% carbon black, 5.7 wt% PAA, and 77 wt% electrolyte solution.

Battery Characterization: The characteristics of the batteries were recorded using a Keithley 2611A Source Measuring Unit (SMU) connected to the battery current collectors. For discharge measurements and characteristic curves in stretched states, the battery was mounted within a uniaxial extensor and stretched to the desired strains. For discharge measurements under cyclic stretching, a custom made tensiometer was used in parallel to the SMU. Secondary cells were continuously charged/discharged with a constant current of 2 mA to a cut-off voltage of 0.9 V for discharging, and 1.65 V for charging.

Electronic Skin: Signal processing and communication were performed by the Bluetooth-module CYBLE-222014 fabricated by Cypress Semiconductor Corp. This module was connected to a Programmer/Debugger (KitProg, Cypress) and flushed with a Bootloader program, which enabled the user to update the module over-the-air (OTA) via Bluetooth. The application of the module was programmed in PSoC Creator 4.2 and updated with the program CySmart 1.3 using the USB-dongle CY5677 CySmart BLE 4.2. A low-dropout-regulator (AP7313-205AG-7, Diodes Incorporated) and a ferrite bead (WE-CBF 1206, Würth Elektronik) were connected to the Bluetooth-module beforehand, to ensure a stable voltage supply of 2 V. An additional switch (CVS-018,
Copal Electronics) was included to cut-off the power supply of the battery. Two free-programmable LEDs were included in the electronic circuit to show the state of the Bluetooth-module. All interconnects are made of meander-shaped copper wires (>70 µm diameter wires were manually bent) and soldered to the components with a standard non-RoHs solder (Sn60Pb39Cu1, RS Components). The five touch input buttons were designed as open electrodes and operated with SmartSense, which was an included feature in the Bluetooth-module.

The whole circuit was placed onto a 0.5 mm thick acrylic elastomer (VHB, 3M) sheet and the open electrodes are bent orthogonal to the sheet. The circuit was embedded in a layer of silicone elastomer (cured at 40 °C for 10 h), to achieve a reversibly stretchable electronic skin.

To achieve the required voltage levels an array of two single cells (fabricated as described above) were connected in series. The 3M-VHB elastomer lid and base of the battery were pre-stretched ≈50% using an accordion extender, to render biaxially stretchable electrodes. The electrodes were guided to the bottom side of the battery, to enable direct contact with the open connections of the electronic skin, when mounted on the equally sticking VHB base sheet.

The hand shown in Figure 4 and Movie S1, Supporting Information is that of D.D., who has given her permission to publish this media.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Author Contributions
F.H., S.B., M.K. conceived the research project; W.P. developed the polyHIPE materials with input from A.B.; D.D., F.H., R.S. designed the experiments; D.D. prepared the materials; D.D., F.H. conducted electro-mechanical characterization; S.D. recorded the SEM images; D.D., R.S. conducted the dielectric modeling; R.P., D.D. designed and fabricated the electronic skin; D.D., F.H. analyzed the data; F.H., D.D., and M.K. wrote the manuscript; all authors contributed to editing the manuscript. F.H., M.K. supervised the research.

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[1] H. U. Chung, B. H. Kim, J. Y. Lee, J. Lee, Z. Xie, E. M. Ibler, K. H. Lee, A. Banks, J. Y. Jeong, J. Kim, C. Ogle, D. Grande, Y. Yu, H. Jang, P. Assem, D. Ryu, J. W. Kwak, M. Namkoong, J. Bin Park, Y. Lee, D. H. Kim, A. Ryu, J. Jeong, K. You, B. Ji, Z. Liu, Q. Huo, X. Feng, Y. Deng, Y. Xu, K. I. Jang, J. Kim, Y. Zhang, R. Chaffari, C. M. Rand, M. Schau, A. Hammars, D. E. Weese-Mayer, Y. Huang, S. M. Lee, C. H. Lee, N. R. Shanbhag, A. S. Paller, S. Xu, J. A. Rogers, Science 2019, 363, eaau0780.
[2] J. Ge, X. Wang, M. Drack, O. Volkov, M. Liang, G. S. Cañón Bermúdez, R. Illing, C. Wang, S. Zhou, J. Fassbender, M. Kaltenbrunner, D. Makarov, Nat. Commun. 2019, 10, 4405.
[3] F. Hartmann, M. Drack, M. Kaltenbrunner, Sci. Rob. 2018, 3, eaat9091.
[4] R. Kumar, J. Shin, L. Yin, J. M. You, Y. S. Meng, J. Wang, Adv. Energy Mater. 2017, 7, 1602096.
[5] H. Li, Y. Ding, H. Ha, Y. Shi, L. Peng, X. Zhang, C. J. Ellison, G. Yu, Adv. Mater. 2017, 29, 1700898.
[6] Y. Zhang, W. Bai, J. Ren, W. Weng, H. Lin, Z. Zhang, H. Peng, J. Mater. Chem. A 2014, 2, 11054.
[7] W. Weng, Q. Sun, Y. Zhang, S. He, Q. Wu, J. Deng, X. Fang, G. Guan, J. Ren, H. Peng, Adv. Mater. 2015, 27, 1363.
[8] L. Ma, S. Chen, D. Wang, Q. Yang, F. Mo, G. Liang, N. Li, H. Zhang, J. A. Zapien, C. Zhi, Adv. Energy Mater. 2019, 9, 1803046.
[9] J. Liu, M. Hu, J. Wang, N. Nie, Y. Wang, Y. Wang, J. Zhang, Y. Huang, Nano Energy 2019, 58, 338.
[10] X. Chen, H. Huang, L. Pan, T. Liu, M. Niederberger, Adv. Mater. 2019, 31, 1904648.
[11] L. Yin, J. K. Seo, J. Kurniawan, R. Kumar, J. Lv, L. Xie, S. Liu, S. Xu, Y. S. Meng, J. Wang, Small 2018, 14, 1800393.
[12] M. Gu, W.-J. Song, J. Hong, S. Y. Kim, T. J. Shin, N. A. Kotov, S. Park, B.-S. Kim, Sci. Adv. 2019, 5, eaav1879.
[13] N. Boaretto, J. Almenara, A. Mikhalchan, R. Marcilla, J. J. Vilatela, ACS Appl. Energy Mater. 2019, 2, 5883.
[14] A. J. D’Angelo, M. J. Panzer, Chem. Mater. 2019, 31, 2913.
[15] C. Yan, X. Wang, M. Cui, J. Wang, W. Kang, C. Y. Foo, P. S. Lee, Adv. Energy Mater. 2014, 4, 1301396.
[16] H. W. Zhu, J. Ge, Y. C. Peng, H. Y. Zhao, L. A. Shi, S. H. Yu, Nano Res. 2018, 11, 1554.
[17] W. J. Song, J. Park, D. H. Kim, S. Bae, M. J. Kwak, M. Shin, S. Kim, S. Choi, J. H. Jang, T. J. Shin, S. Y. Kim, K. Seo, S. Park, Adv. Energy Mater. 2018, 8, 1702478.
[18] M. Kaltenbrunner, G. Kettlgruber, C. Siket, R. Schwödiauer, S. Bauer, Adv. Mater. 2010, 22, 2065.
[19] D. Wirthl, R. Pichler, M. Drack, G. Kettlgruber, R. Moser, R. Gerstmayr, F. Hartmann, E. Bradt, R. Kaltseis, C. M. Siket, S. E. Schausberger, S. Hild, S. Bauer, M. Kaltenbrunner, Sci. Adv. 2017, 3, e1700053.
[20] S. S. Sekhon, Bull. Mater. Sci. 2003, 26, 321.
[21] R. T. Woodward, D. W. H. Fam, D. B. Anthony, J. Hong, T. O. McDonald, C. Petit, M. S. P. Shaffer, A. Bismarck, Carbon 2016, 101, 233.
[22] R. Owen, C. Sherborne, T. Paterson, N. H. Green, C. G. Reilly, F. Claeyssens, J. Mech. Behav. Biomed. Mater. 2016, 54, 159.
[23] R. S. Moglia, J. L. Holm, N. A. Sears, C. J. Wilson, D. M. Harrison, E. Cosgriff-Hernandez, Biomacromolecules 2011, 12, 3621.
[24] Q. Jiang, A. Menner, A. Bismarck, React. Funct. Polym. 2017, 114, 104.
[25] M. Corti, E. Cailieri, S. Perteghella, A. Ferrara, R. Tamma, C. Milanese, D. Mandracchia, G. Brusotti, M. L. Torre, D. Ribatti, F. Auricchio, G. Massolini, G. Tripodo, Mater. Sci. Eng., C 2019, 105, 110060.
[26] Q. Jiang, H. Barkan, A. Menner, A. Bismarck, Polymer. 2017, 126, 419.
[27] S. Kovačić, H. Kren, P. Krajnc, S. Koller, C. Slugovc, Macromol. Rapid Commun. 2013, 34, 581.
[28] N. Shirshova, P. Johansson, M. J. Marczewski, E. Kot, D. Ensling, A. Bismarck, J. H. G. Steinke, J. Mater. Chem. A 2013, 1, 9612.
[29] A. D. Roberts, X. Li, H. Zhang, Chem. Soc. Rev. 2014, 43, 4341.
[30] U. M. Patil, R. V. Ghorpade, M. S. Nam, A. C. Nalawade, S. Lee, H. Han, S. C. Jun, Sci. Rep. 2016, 6, 35490.
[31] M. Shin, W. J. Song, H. Bin Son, S. Yoo, S. Kim, G. Song, N. S. Choi, S. Park, Adv. Energy Mater. 2018, 8, 1801025.
[32] A. Menner, K. Haibach, R. Powell, A. Bismarck, Polymer. 2006, 47, 7628.
[33] N. Zhang, Y. Zhou, Y. Zhang, W. Jiang, T. Wang, J. Fu, Chem. Eng. J. 2018, 354, 245.
[34] Y. Luo, A. N. Wang, X. Gao, Colloid Polym. Sci. 2015, 293, 1767.
[35] Q. Wang, H. Ma, J. Chen, Z. Du, J. Mi, J. Environ. Chem. Eng. 2017, 5, 2807.
[36] N. R. Cameron, D. C. Sherrington, J. Mater. Chem. 1997, 7, 2209.
[37] H. Tai, A. Sergienko, M. S. Silverstein, Polym. Eng. Sci. 2001, 41, 1540.
[38] A. Y. Sergienko, H. Tai, M. Narkis, M. S. Silverstein, J. Appl. Polym. Sci. 2002, 84, 2018.
[39] Q. Jiang, A. Menner, A. Bismarck, Polymer. 2016, 97, 598.
[40] Y. Zhou, C. Wan, Y. Yang, H. Yang, S. Wang, Z. Dai, K. Ji, H. Jiang, X. Chen, Y. Long, Adv. Funct. Mater. 2019, 29, 1806220.
[41] Y. Q. Yang, Z. Chang, M. X. Li, X. W. Wang, Y. P. Wu, Solid State Ionics 2015, 269, 1.
[42] J. Guan, Y. Li, J. Li, Ind. Eng. Chem. Res. 2017, 56, 12456.
[43] M. Park, H. Woo, J. Heo, J. Kim, R. Thangavel, Y. Lee, D. Kim, ChemSusChem 2019, 12, 4645.
[44] S. Choudhury, T. Saha, K. Naskar, M. Stamm, G. Heinrich, A. Das, Polymer 2017, 112, 447.
[45] T. M. Di Palma, F. Migliardi, M. F. Gaele, P. Corbo, Ionics 2019, 25, 4209.
[46] H. Khani, S. Kalami, J. B. Goodenough, Sustainable Energy Fuels 2020, 4, 177.
[47] P. Hu, Y. Duan, D. Hu, B. Qin, J. Zhang, Q. Wang, Z. Liu, G. Cui, L. Chen, ACS Appl. Mater. Interfaces 2015, 7, 4720.
[48] D. Zhou, Y. B. He, R. Liu, M. Liu, H. Du, B. Li, Q. Cai, Q. H. Yang, F. Kang, Adv. Energy Mater. 2015, 5, 1500353.
[49] D. Djian, F. Alloin, S. Martinet, H. Lignier, J. Pow. Sour. 2008, 2, 575.
[50] M. Drack, I. Graz, T. Sekitani, T. Someya, M. Kaltenbrunner, S. Bauer, Adv. Mater. 2015, 27, 34.
[51] D. Linden, Linden’s Handbook of Batteries (Ed: T. B. Reddy), Vol. 4, Mcgraw-hill, New York 2011.
[52] G. Kettlgruber, M. Kaltenbrunner, C. M. Siket, R. Moser, I. M. Graz, R. Schwödiauer, S. Bauer, J. Mater. Chem. A 2013, 1, 5505.
[53] J. Kalhoff, D. Bresser, M. Bolloli, F. Alloin, J.-Y. Sanchez, S. Passerini, ChemSusChem 2014, 7, 2939.