ADVANCED ENGINEERING MATERIALS
www.aem-journal.com

Elastomers for Soft Electronics: A Review from the Material's Perspective

*Soumili Chakraborti, Pratip Sankar Banerjee, Debdipta Basu, Sven Wießner, Gert Heinrich, Amit Das, and Shib Shankar Banerjee**

Elastomers fall in a distinctive class of flexible and soft materials driven by their long-term load-bearing capability under wide range of static and dynamic stresses, high compliance and stretchability, low cohesive energy density, and low glass transition temperature. Attributing to suitable functionalization, elastomers are coming to the fore and are of utmost importance for the soft electronics domain translating into soft robotics, electronic skins, sensors, displays, health monitoring, energy-harvesting devices, etc. This review provides a comprehensive analysis of different classes of elastomers and functional fillers, highlighting their potential to meet the specific requirements of these advanced applications. The current state-of-the-art feasible fabrication–integration technologies of elastomers for soft electronics are highlighted. The fundamental mechanisms for boosting functionality with the help of polymer–filler interactions leading to several effects like the tunnelling effect, disconnection mechanisms in overlapped composites, crack propagation in thin films, and many more are included with theoretical and experimental evidences. Additionally, this review emphasizes the dynamic mechanical properties of smart elastomers, which are crucial for the durability and robustness of stretchable electronics in the real application field. This review finally reveals the potential of elastomers to be used in soft electronics over a diverse range of applications.

1. Introduction

The notably customizable mechanical, chemical, thermal, and electrical properties along with a high strength-to-weight ratio and compatibility over various substrates, necessitate the use of soft/flexible smart materials in the domains of soft electronics.[1] The state-of-the-art devices for the applications like biomimetic human tissues,[2] surgical robotic assistant,[2–4] energy-harvesting devices,[5] biohybrid engineering,[2–4] soft sensors, actuators, and logic devices[6–8] are designed and developed by infusing multifunctional attributes into elastomeric/soft substrates as depicted in **Figure 1**. The traditional electronic devices based on rigid materials like metals, and ceramic substrates, lack the necessary compliance for soft substrates.[1,3] Properties such as deformability with continuous strain dissipation,[9] stretchability,[10] and strength retention under cyclic loading,[11] along with biocompatibility,[12] are of fundamental importance in this domain which are not often within a single package of a conventional rigid material.[4] Therefore, the study and development of novel materials bearing most of the aforementioned attributes are of utmost importance to boost the rapid progress of our scientific intellect.

Soft electronics is an integrative research field involving specialized materials capable of conforming to any shape, bendability, and stretchability[1,4,8] along with functional attributes like electrical conductivity, high dielectric constant, magnetic steerability, and thermal properties.[13] In this field, organic (polyethylene terephthalate),[14] polyethylene[15] or inorganic materials are embedded on flexible substrates (polyvinyl acetate,[16] polyethylene naphthenate,[17] and polyimide[18]) to obtain compliant, lightweight, transparent, and portable electronic devices. However, these materials generally exhibit a limited stretchability of below 10% strain at break.[19] Therefore, to attain the required critical flexibility with high strain at break (>300%), elastomers pose as a viable alternative for substrate materials to the modern scientific community.[8,13,19] The elastomeric substrates possess elasticity, low modulus, and high stretchability.[20] Flexibility arises from lower intermolecular forces, lower chain-packing tendency and the subzero glass transition temperature.[21] Along with that, chemical or physical cross-links can be induced within the elastomeric

S. Chakraborti, P. S. Banerjee, S. S. Banerjee
Department of Materials Science and Engineering
Indian Institute of Technology Delhi
Hauz Khas, New Delhi 110016, India
E-mail: ssbanerjee@mse.iitd.ac.in

D. Basu
Indian Rubber Materials Research Institute (IRMRI)
East Centre, Rubber Park, Sankrail, Howrah 711302, India

S. Wießner, G. Heinrich, A. Das
Leibniz-Institut für Polymerforschung Dresden e. V
Hohe Straße 6, D-01069 Dresden, Germany

S. Wießner
TUD Dresden University of Technology
Institut für Werkstoffwissenschaft
Helmholtzstraße 7, D-01069 Dresden, Germany

G. Heinrich
TUD Dresden University of Technology
Institut für Textilmaschinen und Textile Hochleistungswerkstofftechnik
Hohe Straße 6, D-01069 Dresden, Germany

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/adem.202402458.

DOI: 10.1002/adem.202402458

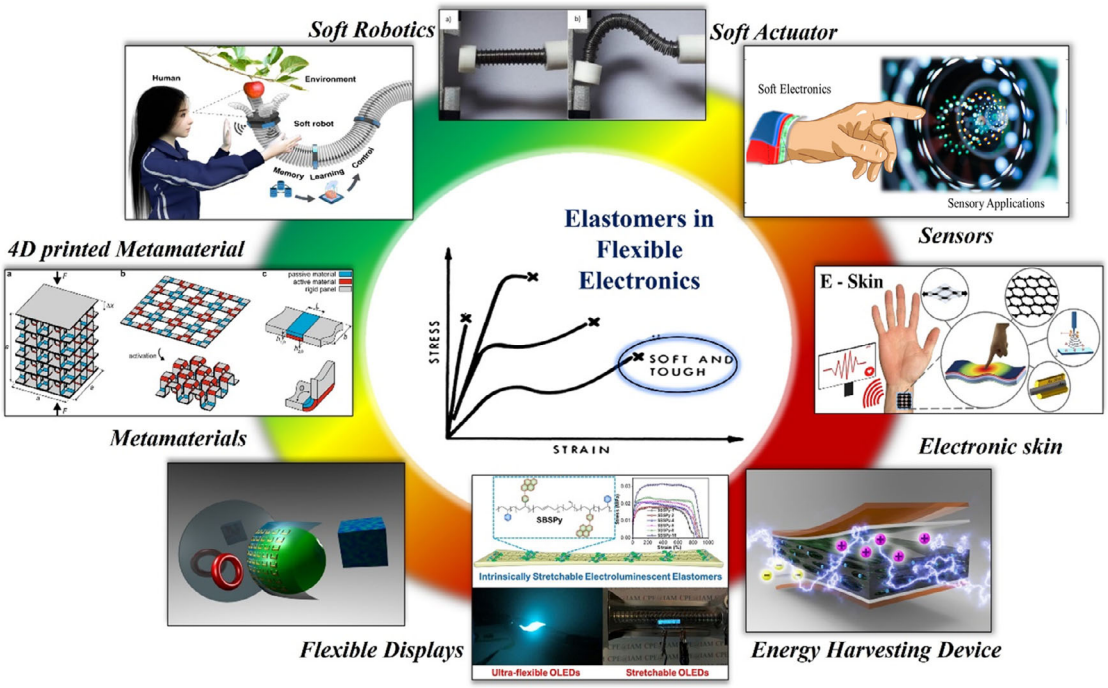


Figure 1. Pioneering and futuristic elastomeric soft electronics in their predominant sectors of application. Elastomers being the soft and tough (Copyright available at free of charge.[259]) materials can be implemented in various futuristic innovations like soft robotics with bimodal sensory surfaces (Reproduced under the terms of the CC-BY license.[260] Copyright 2022, The Authors, Spinger Nature), soft actuators (Adapted with permission.[219] Copyright 2021, Elsevier), and sensors with stimuli-responsive attributes, electronic skin formulations for bio-signaling and health monitoring (Reproduced under the terms of the CC-BY license.[262] Copyright 2021, The Authors, MDPI), energy-harvesting devices like ferroelectret nanogenerators (Adapted with permission.[263] Copyright 2016, Elsevier) and piezoelectret nanogenerators with human-powered responses, flexible displays (Adapted with permission.[241] Copyright 2022, John Wiley and Sons) being the state-of-the-art functional materials in several electronic devices like mobile phones, laptops, light-emitting diodes, etc., new age flexible metamaterials (Reproduced under the terms of the CC-BY license.[243] Copyright 2010, IOP Publishing), 4D-printed self-shape-morphing metamaterials (Reproduced under the terms of the CC-BY license.[244] Copyright 2022, The Authors, Spinger Nature), etc.

chains that improve the strength by eliminating chain slippage.[22] Although the mechanical requirements for flexible electronics substrates appear to be more-or-less addressable using elastomers, there are major limitations for the use of conventional elastomeric materials due to their lack of multifunctionality such as low dielectric strength, insulating behavior, and higher dissipation factor.[23] In addition, commercial rubbers used in the industries like natural rubber (NR), styrene–butadiene rubber (SBR), and polybutadiene rubber (PBR) even deviate from the necessary compliance and softness due to the afore-stated reasons.[7,8,13,23] NR being a natural compound has long molecular chains and high entanglement density giving rise to its nervy nature and high Mooney viscosity. This signifies hindered processibility of gum NR as the entangled polymeric chains take part in strain-induced crystallization thereby incurring stiffness into the chain.[3,6] Other special purpose rubbers like acrylonitrile butadiene rubber (NBR), ethylene–propylene diene monomer (EPDM) rubber, etc., which are tailor-made having comparatively lower Mooney viscosities as compared with NR, although lacking the required electrical and thermal properties.[3,6,19,22] One of the most common methods of inducing multifunctionality is through the incorporation of functional fillers like carbon nanotubes (CNTs),[24] graphene,[25] metallic fillers,[26] ceramic nanoparticles,[27] etc., into the elastomeric matrix.[28] However, higher degrees of functional filler loading render the elastomeric matrix to become rigid which ultimately results in the loss of substrate flexibility.[19] Therefore, achievement of functionality along with adequate elasticity requires necessary modification of the elastomers for applications in niche areas, such as soft and damage-resistant circuitry. Synthesizing graft copolymers composed of polyurethane (PU) and conducting polymers, such as polyaniline, is one way of infusing multifunctionality in the matrix[29,30] though requiring complex experimental conditions and is yet to confirm their long-term reliability. Easier ways of tackling the complication is forming a blend of the conducting polymer and an elastomer.[19,31] For example, in poly(3,4-ethylenedioxythiophene)–PU (PEDOT–PU) blend, a high conductivity of 100 S cm^{-1} was observed with elongation at break greater than 100%.[19,32–34] Mainly electrical conductivity of a blend increases in exchange of the mechanical properties which entails the need for understanding the material design to fulfil the required specifications for the particular applications.[35] Despite different modification strategies employed, incorporation of the rigid fillers into the soft elastomeric matrix leads to incoherent mechanical deformation of the multiple phases causing nonlinearity at dynamic conditions. The rigid fillers remain fixed in space while deformation and the matrix strains under applied stress.

This leads to localized stresses or zones of stress concentration around the high modulus domains. Recent developments and innovations in the fields of flexible and soft electronics pursue the utilization of smart elastomeric materials that respond to stimuli like mechanically active membranes for medical applications like wound dressings. This utilizes PU/poly(vinyl butyral) membranes electrospun together to enable a temperature-driven functional attribute.[36] Similarly, advancements in bio-based elastomers have started with multiple responsiveness due to supramolecular entities for triboelectric nanogenerators (TENGs).[37] However, incorporation and application of the functional domains into an elastomeric matrix require numerous optimizations and the added factor of multiple responsiveness arises at the cost of several internal factors like loading levels due to different polymer–filler interactions. All of this requires systemic monitoring to make it useful with the fulfilment of the basic criteria of soft electronics in dynamic environments.

The review focuses on a comprehensive understanding of the need for elastomeric materials from recent research in the field of soft electronics. Robustness under cyclic dynamic loads is a critical issue, and this is where the requirement for flexible and soft substrates over conventional rigid materials arises. The following fundamental questions are addressed in this review article. 1) What are the particular governing criteria for a material to be appropriate as a soft substrate for soft electronics and why elastomeric matrices are of interest in this regard? 2) How to invoke adequate multifunctionality into elastomeric systems for rendering them suitable for soft electronic devices with mechanisms of electronic sensing and fabrication techniques? 3) What are the specific challenges involved in utilizing different classes of elastomer as a substrate material with additional confrontations in dynamic robustness?

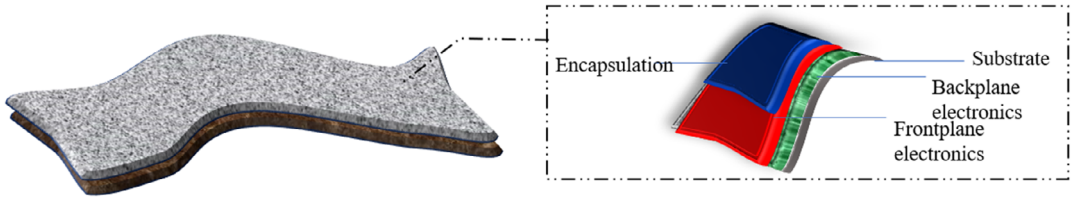
These questions have been addressed thoroughly from elastomeric materials perspective. In addition, the existing strategies for devising soft electronic substrates out of elastomers have been critically analyzed for providing an extrapolative view towards the future development path of this research domain. Plausible areas of applicability based on the advantageous and disadvantageous traits of elastomeric materials are established highlighting the applications of the elastomeric soft electronics. This review is believed to inject a boost to both the academia and the industrial community for developing a sound understanding of the versatility of elastomeric materials in the wake of current and future technological progress in the discipline of flexible electronics.

2. Fundamentals of Soft and Flexible Elastomeric Materials

The broad classification of materials typically concerns the "big three"—metals, ceramics, and polymers. Metals and ceramics are widely known for their excellent functional properties facilitating innumerable real-world applications. However, the fundamental lag concerning these two is the lack of smooth human–machine interface, or in other words, the interaction of devices made out of metals or ceramics with soft, biological tissues is a challenging task. Henceforth, a material bearing a certain degree of softness and flexibility similar to biological tissues so as to ease the human–machine interface is the call of the scientific society and industry alike. Of the "big three," polymers can be in the state of a viscous fluid, an amorphous solid or supercooled liquid (glassy), or in the state that contains both crystalline and amorphous regions, termed as semicrystalline.[38] All of this depends on its molecular properties like chain flexibility, intermolecular force of attraction, alignment of macromolecular chains, and certain external factors like temperature.[20] Elastomers represent the group of amorphous polymers which upon cross-linking can undergo very large, reversible elongations (500–1000%) at low stresses.[39] This type of polymeric materials has high chain flexibility due to low intermolecular force of attraction, chain packability, and glass transition temperature that is lower than the service temperature. The elasticity of rubber originates from the ease of transformation from helical conformation of molecules in the chain backbone in the contorted state to the thermodynamically favored coiled conformation of highest entropy when the stress is withdrawn.[20,39] Some degree of cross-linking facilitates elastic deformation without chain slippage along with some degree of strength.[40] Few elastomers like NR undergo an additional strengthening mechanism by strain-induced crystallization whereby chain orientation in the direction of straining promotes some degree of crystallinity as the chains enable to pack closely.[41] The degree of cross-linking and the consequent mechanical properties of the elastomer covers a wide range depending on distinct end uses. General purpose rubbers (GPRs) have good physical properties with low chemical resistance like NR, SBR, PBR, NBR, and chloroprene rubber (CR).[42] Special purpose rubbers (SPRs) are resistant to degrading action of polar solvents, oils, oxygen, ozone, heat, and frost. For example, EPDM rubber is weathering resistant;[43] isobutylene–isoprene rubber (IIR)[44] is air impermeable; fluoroelastomers (FKMs) have good chemical resistance; and silicone rubber has high thermal stability.[45] Depending on the molecular chain backbone, rubbers are classified as organic and inorganic rubbers. The most commercially used GPRs like NR, SBR, PBR, have an organic (C—C) backbone whereas silicone rubber has an inorganic (Si—O) chain backbone.[46] NR has a chemical name of *cis*-1,4-polyisoprene which has a stereoregular structure enabling strain-induced crystallization generating high gum strength. It is an agricultural product obtained from the tree *Hevea brasiliensis* which causes it to have an uncontrollable molecular weight.[47,48] High molecular weight of the polymer results in high Mooney viscosity from 60 Mooney units (MU) ranging as high up to 100 MU and a nervy nature of the polymeric backbone.[49] Some advantageous properties of NR include high tensile strength (vulcanizate strength reaches up to 17–24 MPa), excellent cut growth resistance arising from its tendency of strain-induced crystallization, high resiliency, and very high green tack.[6,47–51] SBR is the second most commonly used rubber after NR and is a synthetic rubber with monomers including styrene and butadiene.[52] SBR consists of two main grades—emulsion SBR (E-SBR) and solution SBR (S-SBR).[53] E-SBR is a free radically synthesized grade which forms a random/statistical copolymer with greater processibility and broader molecular weight distribution.[54] S-SBR is an anionically synthesized copolymer whose blockiness can be controlled because of the process of synthesis through the control of reactivity ratios. It has better mechanical properties with linear molecular weight distribution.[55] The vinyl content of the SBR chain can be controlled in

this solution-polymerized SBR and hence is used in tire applications for its high abrasion resistance, good wet skid resistance, and lower rolling resistance.[55–57] They are generally insulating in nature. PBR is an unsaturated polymer with high rebound resilience used typically in tire applications, golf balls, etc.[58] It has high cut growth resistance with intrinsic cut growth energy because of the cis counterpart being stereo-regular in nature enabling strain-induced crystallization.[53,59] IIR is a random copolymer of isobutylene (97–98%) and isoprene (2–3%) synthesized by cationic polymerization. The geminal dimethyl groups on the alternating carbons in the chain backbone cause the bond angle to be distorted from usual tetrahedral angle (109.45°) to 123°, this forces the chains to get contorted and elongated, resulting in greater packability and ultimately decreasing the air permeability.[60] They are generally used in inner tubes and tire curing bladders and possesses good vibrational damping characteristics necessary to be used in automotive suspension bumpers.[61,62] PU is a specialty rubber prepared mainly by isocyanates and one another species containing active hydrogen, like alcohols, amines, hydroxyl-terminated esters, ethers, or carbonates.[63] It is synthesized by condensation polymerization without the loss of any by-product.[63,64] It has a wide range of hardness, high load-bearing capacity, flexibility, abrasion and impact resistance, resistance to oil and grease, good insulating properties, wide resiliency range, strong bonding capacities, and wide color varieties. It is also biocompatible in nature with excellent low temperature resistance.[65] However, it has hazardous impact on the environment due to the presence of isocyanate.[66] Silicone elastomers contain an organic moiety (can be changed according to desired properties) in an inorganic chain backbone (Si—O—Si bond).[45] It has good low temperature flexibility due to its low rotational energy barrier of Si—O—Si bond and high temperature stability due to the high binding energy of Si—O bond of 445 kJ mol^{-1} (C—C bond energy is ≈347 kJ mol^{-1}).[45,67] Bigger size of the silicon atom with respect to carbon causes the bond angle to be 125°–160° where sp3 bond angle is 109° 28′. Higher bond strain, higher bond length, and consequently greater flexibility are observed.[45,67,68] It also has good dielectric properties at both high and low frequencies and very low loss tangent values making it suitable for electronic applications.[28,69] However, the tensile strength observed is very low making it unsuitable for various high load bearing applications.[70] Additionally, another class of elastomeric, soft material is the thermoplastic elastomer (TPE). TPEs themselves are a broad category, although thermoplastic PU (TPU) is one of the well-known TPEs used in the industry. TPEs have various advantages compared to other ordinary elastomers because of their reprocessibility, recyclability, and less energy consumptive nature. The thermosets or conventional elastomer vulcanizates require heat/energy forachieving their usual strength and cannot be solvated, which causes melt processing to be highly sensitive. But, in TPEs, the polymer can be solvated due to physical cross-links, allowing it to be melt-processed at lower energy costs. Block copolymers with physical cross-linking are TPEs with the aforementioned properties. Other representatives of polystyrene-based TPEs used in the electronics industry are styrene–butadiene–styrene (SBS) triblock copolymer, styrene–ethylene–butylene–styrene (SEBS) tetramer, styrene–isoprene–styrene (SIS) triblock copolymer, and styrene–isobutylene–styrene (SIBS) triblock copolymer. The generic electronic structure can consist of a substrate, backplane electronics, front plane electronics, and encapsulation, all of which must comply with bending to some degree without losing any functionality to be applicable in the soft electronics industry.[71] A schematic diagram showcasing the general structure of a soft electronics substrate highlighting different components and various suitable elastomers and functional materials is provided in **Figure 2**.

Despite such promises of being a boon to the materials' perspective, several disadvantages for use in the multifunctional domains for elastomers in general include its limited mechanical stability consequently leading to poor tear resistance and faster fatigue, poor thermal conductivity, high chemical sensitivity leading to degradation, which further depends on the chemical nature of the elastomeric backbone, limited electrical properties which require functional filler incorporation however, leading to



Common Elastomeric Substrates:

PDMS	PU	NBR	CR	Other polar rubbers

Common Functional Electronic Layers:

Conducting Polymers	Nanomaterials	Liquid Metals	Carbon/Metal	Hybrid Materials
Cluster Materials	Nanowires	Organic Inorganic hybrid materials	Biological Functional Materials	Iridium, Platinum interconnects

Common Encapsulating Materials for different Applications:

PDMS	PI	TPU	FKM	SBS block copolymers

Figure 2. Schematic framework and construction of soft electronics with common functional materials for fabrication of soft electronics.[8]

lower electrical and electronic stability, creep and hysteresis also known as the strain softening or Mullin's effect due to cyclic loadings due to short-term stability, limited fabricability without the ability of scalability in industrial perspective, and other environmental concerns. Overcoming these limitations is being aimed by recent researchers for harnessing their use in the multifunctional domains by matching the functionality criteria according to the application ultimately focusing on the structure–property–application dynamics summarized from recent researches shown in **Figure 3**. In regard to the former statement, in applications of electronic skins with biomimetic properties, flexible substrates modulus values range even lower from 10^{-4}–10^{-1} MPa;[72,73] in the liquid-crystalline displays and transmissive bottom-emitting displays, the material used in the substrate should have low birefringence ($\Delta n < 0.002$[74])[75,76] with water vapor transmission rate less than 10^{-6} g m^{-2} day^{-1} and the oxygen permeation rate should be less than 10^{-3}–10^{-5} cm^3 m^{-2} day^{-1}.[71,77–80] Also, the usage of flexible substrates in antennas require minimal dielectric loss with low dielectric constant, low coefficient of thermal expansion (CTE), and high thermal conductivity.[81] In view of the thermal and thermomechanical properties of the material for fabrication of the substrate, the substrate glass transition temperature must be compatible with the maximum fabrication process temperature and any mismatch may cause the electronic interconnect to break during the dynamic thermal cycling associated with the process of fabrication.[71,75–77,82] It has been observed that the product of the temperature difference between substrate and device and the difference of the CTEs has to be below 0.1–0.3%.[71,83] After the complete fabrication and integration with appropriate functionality enhancement according to application, both the GPRs and SPRs can be put into action for soft material substrates in electronics' application as schematically depicted in **Figure 4**a. NR functionalized with polar moieties in the structural backbone through epoxidation forming epoxidized NR allows better interaction with functional fillers like CNTs resulting in an outstanding electrical conductivity of 10^2 S cm^{-1} with enhanced mechanical properties and strain sensitivity beneficial for a strain sensor as seen in Figure 4b.[84] As previously mentioned, the widely studied GPR and SBR have also been reported as a conducting substrate upon

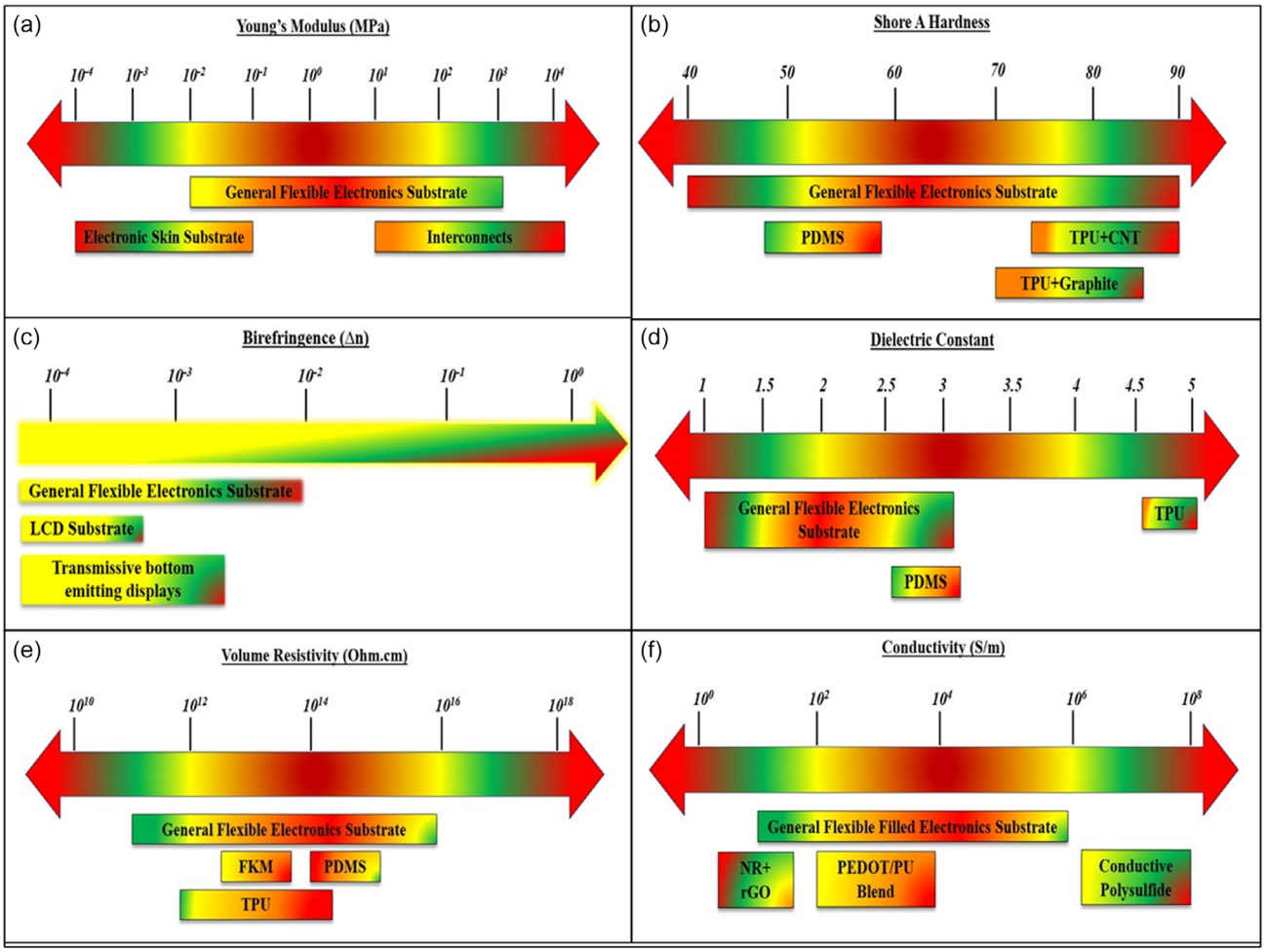


Figure 3. A generalized schematic of the functional properties: a) Young's Modulus (MPa),[72,73] b) hardness (Shore A scale),[3,205,206] c) birefringence,[74–76] d) dielectric constant,[81,264] e) volume resistivity (Ω cm),[153,265,266] and f) conductivity (S m^{-1})[267–271] required in a soft electronics substrate along with few examples.

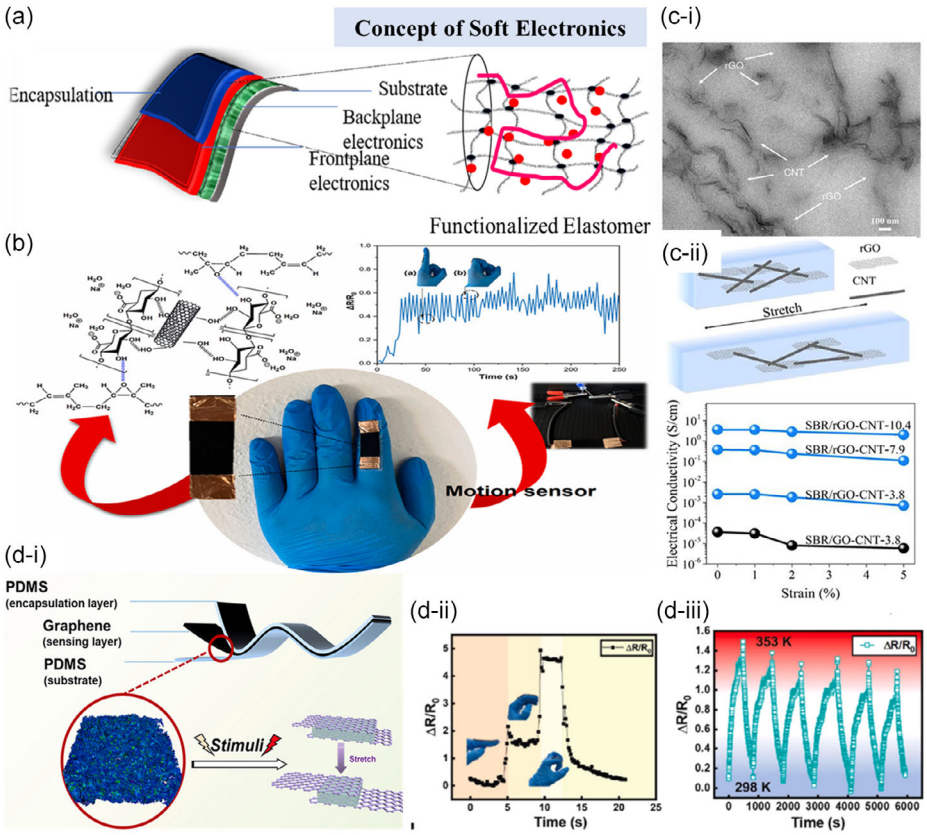


Figure 4. a) Schematic for functionalized elastomer in soft electronics. b) Modified NR–CNT and NR–CNT–alginate composites for motion sensing applications understood by relative resistance measurement upon changing strain (Adapted with permission.[84] Copyright 2024, Elsevier). c-i) SBR composites micrograph for functionalization; (c-ii) SBR composites with sandwiched CNT and reduced graphene oxide (rGO) showing 14 orders of conductivity increase from pristine SBR (Adapted with permission.[85] Copyright 2019, Elsevier). d-i) PDMS composite showing multimodal sensing through large temperature range. (d-ii,iii) Results showing change in relative resistance to show effective multimodal sensing (Adapted with permission.[86] Copyright 2023, Elsevier).

fabrication with CNTs and graphene depicted in Figure 4c(i), c(ii).[85] The most widely used SPR, poly(dimethylsiloxane) (PDMS) has also been functionalized in several cases to meet the final applications of multimodal sensing with graphene moieties with enhanced thermal stability depicted in Figure 4d(i). The changes in relative resistance (RR) shown in Figure 4d(ii),(iii) show its effectiveness in sensing applications.[86] The knowledge of the fundamental sensing mechanisms with respect to required properties has to be critically analyzed for practicality in the industrial domains as discussed in the further sections.

3. Designing of Functional Elastomeric Materials for Soft Electronics

When implementing stretchable and flexible electronics, these stretchable active components are combined with flexible and robust connecting elements that can withstand significant external loads. Even if the mechanical property criterion is fulfilled, limited functionality aspects often can limit the chosen materials to thrive in the soft electronics discipline. Henceforth, fabrication of the elastomeric substrates for soft electronics requires various techniques for enhancement of the multifunctional behavior through the fundamental sensing mechanisms. Functional properties broadly revolve around the electrical, magnetic, thermal, and optical response of the materials and various types of each of these responses can be imbibed by integrating an elastomer with relevant materials bearing the required functionality. However, imbibition of the necessary functionality requires the basic knowledge of the responsive mechanisms causing the effect as schematically represented in **Figure 5**a. The initial understanding of whether the matrix is working as a soft substrate tunes down to the functional filler and polymer interaction to various performance parameters.[87] Responsive behavior in soft substrates during integration of functionality can be due to geometric effects (Figure 5b),[88] piezoresistivity in the material itself,[89] through disconnection in elements,[90] crack propagation (Figure 5d),[91] or tunnelling effect (Figure 5c).[92] These mechanisms for responsive behavior have been mentioned in this study for correlation to the mentioned properties and concisely discussed in the next sections.

Hence, the numerous functional and multifunctional attributes have been induced in an elastomeric material resulting in a soft and flexible material well-suited for this domain, the most highlighted of them are as follows.

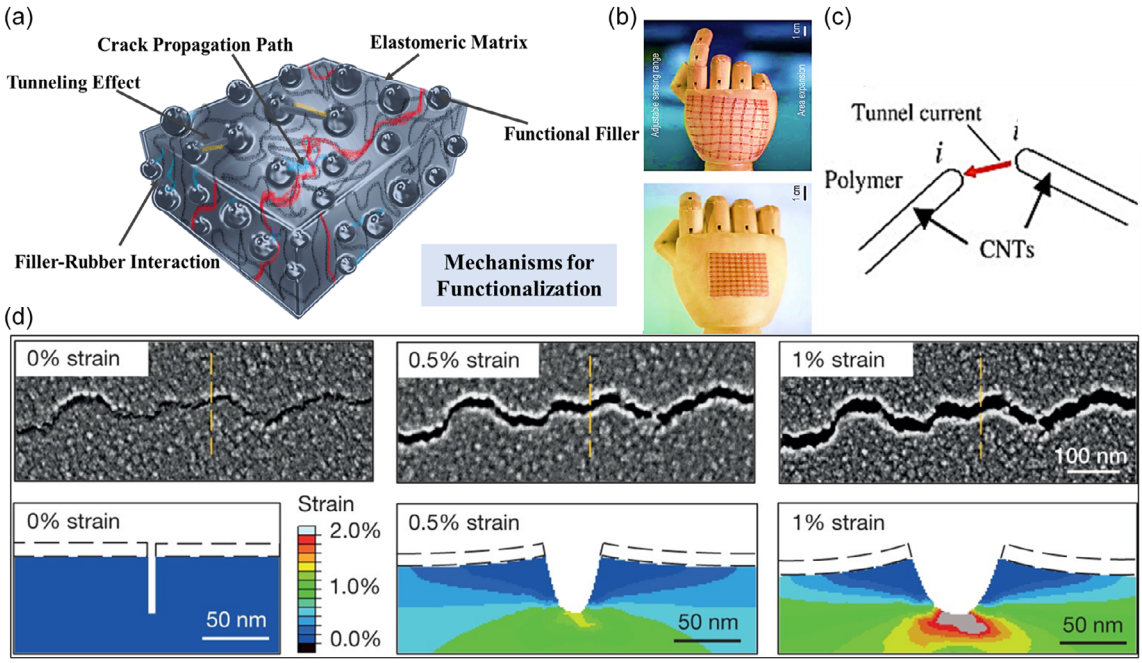


Figure 5. a) Schematic showing all the mechanisms responsible for functional properties in the system through polymer–filler interactions. b) Schematic of geometric effects leading to be the fundamental sensing mechanism (Adapted with permission.[88] Copyright 2022, Elsevier). c) Schematic for understanding quantum tunnelling networks with a tunnel resistance (Adapted with permission.[92] Copyright 2010, Elsevier). d) Schematic of crack propagation dynamics used in fundamental sensing mechanisms (Adapted with permission.[91] Copyright 2014, Springer Nature).

3.1. Piezoresistive Behavior

Piezoresistivity and piezoconductivity both describe the changes in the electrical properties or resistance/conductance in response to varied mechanical deformations used interchangeably. The mechanism of action remains the same for both the behaviors; however, the outcome is physically the opposite in terms of properties. The internal structure of the material in consideration changes with the application of external mechanical stimuli. For example, if conductive fillers are added to the rubbery matrix, under the influence of a mechanical stimulus, the arrangement of them would become disjointed or would orient to a tunnelling pathway, resulting in a decrease or increase in the conductivity, respectively, depending upon the type of functional fillers as depicted by **Figure 6**a. The tunnelling pathway forms after the crossover of electrons through the nonconductive barrier mapping to closely spaced functional materials. The network that forms due to the filler–filler interaction is a dynamic one and the relative position of the functional particles can impact the resistance in the material. The tunnelling resistance can be approximated through Simon's theory for the tunnelling resistance as depicted in the following equation (Equation (1)):[89]

$$R_{\text{tunnel}} = \frac{V}{AJ} \tag{1}$$

where R_{tunnel} is the tunnelling resistance, V is the potential difference, A is the cross-sectional area of the tunnelling junction, and J is the tunnelling current density.[89] This helps in governing the final properties in the functional soft material. These types of materials are used in the domain of soft electronics through pressure and strain sensors for robotic arms and electronic skins.[93] Even without any conducting filler phase, a PDMS-in-PDMS electronic material, due to its internal structure and internal properties like loss tangent and dielectric constant (Figure 6c,d), also exhibits some variation in resistance upon elongation. Yong et al.[94] reported the use of elastomers filled with carbon black (CB) particles as a useful piezoresistive sensor. The author modeled the resistance relaxation with the help of Kelvin–Voigt model which was applicable until the conducting filler had fractured. The hysteresis property indicated that upon retraction, the resistance value did not reach to its initial phase, which was due to the lower tensile strength of the matrix. PDMS was used as a matrix material and it showed lower hysteresis with increasing tensile strength as can be consequently observed from the elasticity of any rubber matrix. This hysteresis property relates down to the dynamic property of the integrated material. Large hysteresis behavior can narrow down to irreversible sensing properties upon dynamic loadings. The modelling of mechanical strain versus the electrical resistance showed significant linearity during extension but nonlinearity at retraction—again corresponding to the lower tensile strength of the matrix due to hysteresis property.[94] This nonlinearity corresponds to the change in morphology from homogeneous to nonhomogeneous and causes further calibration errors. Several other cases have been reported in piezoresistive materials like graphene–elastomer nanocomposites and ionic liquid–based strain sensors[95] for piezoresistive applications in strain and pressure-responsive

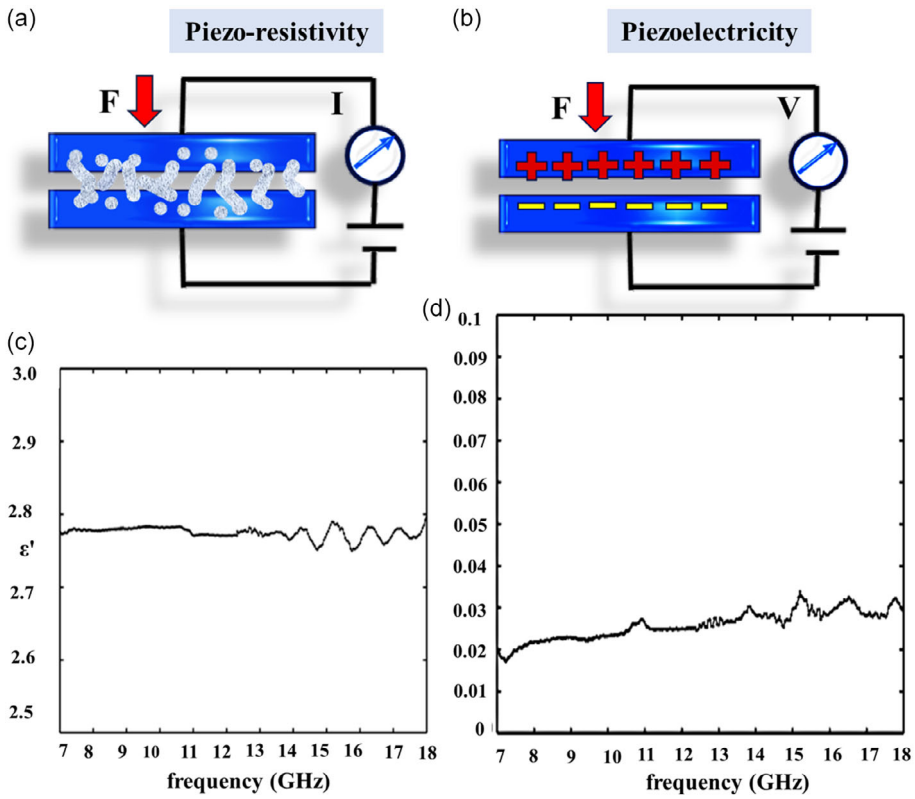


Figure 6. a) Schematic depicting change in current (I) due to piezoresistivity. b) Schematic showing change in voltage (V) due to polarization in piezoelectric materials where F is the force applied. c) Dielectric constant ε' of PDMS measured at RT (Adapted with permission.[272] Copyright 2007, Elsevier). d) Loss tangent tan(δ) of PDMS measured at RT (Adapted with permission.[272] Copyright 2007, Elsevier).

sensors.[96] Park et al.[97] reported a fascinating case on a PDMS—liquid metal (LM) composite forming a 3D nanonetwork allowing giant stretchability up to 225% with 24 100 S cm^{-1} conductivity due to formation of tunnelling networks. The mechanism of the former consequence in this case lies in the local rotations and narrow structural elements in the 3D network. LM in the interstices increases the current carrying capacities and there exists enough cyclic properties for use in interconnects for light-emitting diodes.[97] All of the mechanisms work until a threshold limit of sensitivity due to the change in relative positions of particles causing a change in its tunnelling resistances and accuracy for its applicability and reproducibility in the final performance domain. Natarajan et al.[98] also demonstrated a study on the robust piezoresistive response of NR composites with CNT and CB fillers in dynamic conditions demonstrating stable electromechanical performance with minimal hysteresis, critically emphasizing the role of the uniform distribution and strong interfacial interaction of the fillers in the matrix. Such significant deformation dynamics study that is reproducible should be the era for further research in a more practical sense. Other additional parameters of concern can include the influence of cross-linking density in the elastomer, plasticizers in the chain, and flocculation effects. Cross-linking density in a material would directly affect the resilience in the matrix material and hence the reversibility of the conductive pathway upon stretching. On the contrary, plasticizers would increase the flexibility and interchain distance causing a change in the tunnelling network. Flocculation effects the filler dispersibility, impacting the percolation networks and finally the electrical stability under strain. Hence, under any dynamic condition, the optimization of electrical stability ensures a consistency in signal output reducing the drifts in conductivity and hysteresis, finally enabling a durable performance for the soft electronic materials.

3.2. Piezoelectricity

Piezoelectricity can be defined as the coupling between the mechanical and electrical polarization in a material, i.e., in the presence of a mechanical stimulus, a change in the polarizability of the material induces and vice versa as depicted in Figure 6b.[99] Piezoelectric property arises from atomic scale polarization and piezoelectricity depicts a reverse process of mechanical deformation created in a crystal once an electric field is applied.[100] PDMS being a low modulus material has been reported to be used as piezoelectrets and ferroelectrets for mechanical microsensors and actuators.[101] The characteristic low modulus of the polymeric material leads to stress relaxation, a behavior analogous to viscoelastic liquids stated precisely by Maxwell stresses. The general equation for Maxwell stress[102] is provided in Equation (2).

$$\sigma(t) = \sigma_0 \ \exp(-\lambda t) \quad (2)$$

In this equation, $\sigma(t)$ represents the Maxwell stress at a certain time t and σ_0 represents the initial stress at $t = 0$. λ represents the relaxation time of the material given by the ratio of intrinsic properties of the material, elastic modulus (E), and viscosity (η). Piezoelectrets and ferroelectrets are polymeric films with cellular morphology. The positive and negative charges get trapped on the gas–polymer interfaces in the voids forming macroscopic dipoles and hence giving a piezoelectric effect.[103] Kachroudi et al.[101,103] reported a piezoelectric coefficient (d_{33}) of 350 pC N^{-1} for PDMS piezoelectrets using dielectric resonance spectroscopy, which is 10 times more than polyvinylidene fluoride (PVDF) with a d_{33} of 28 pC N^{-1}. Also, Shi et al.[104] reported that the piezoelectric coefficient of a cellular PDMS ferroelectret to be 70 pC N^{-1}, which increased to 170 pC N^{-1} after plasma treatment for 120 s, which may be due to the improved surface charge density in PDMS.[104] A peak piezoelectricity or the maximum magnitude of electrical charge generated per unit of applied mechanical stress, of 220 pC N^{-1} was observed for PDMS ferroelectrets.[105] The elastomer's internal structure distorts resulting in the separation of the positive and negative charges to a large extent, with the generation of high electrical potential. This value of piezoelectricity signifies the potential for development of advanced sensing systems like in actuators, energy conversion, and transducing applications. Energy-harvesting materials with multifunctional technology witnessed high potential of $ZnSnO_3$ nanostructures due to their sustainability, nontoxicity, and high polarizability (≈59 μC cm^{-2}). However, the pure phase of the material is difficult to achieve because of its metastability. Therefore, Rovisco et al.[106] reported a microstructured composite of nanowires of $ZnSnO_3$ synthesized with PDMS to produce nanogenerators, ultimately enhancing the piezoelectric effect enabling the formation of nanogenerators with output voltage of 120 V, current 13 μA, and instantaneous power density of 230 μW cm^{-2}. Performance of these nanogenerators within an active area of less than 1 cm^2 enables them as a power source for multiple LEDs and small electronic devices, befitting a greater potential in soft electronics.[106,107] Similar devices can be fabricated by filling PDMS with piezoelectric barium titanate alone[108] or with multi-walled CNT creating pyramid microstructures.[109,110]

The electrically conductive composites of EPDM are versatile in its applications due to their flexibility, physico-mechanical stability, and superior weathering attributes. The elastomer renders piezoelectric responses suitable for energy-harvesting devices as in flexible robot arms.[111,112] Conductive fillers like metal powders,[113] inorganic oxide particles,[114] CNTs,[115] graphene,[116] and conductive nanomaterials[117] have been used for rendering the required conductivity in the elastomeric matrix. An EPDM rubber–based functional elastomeric material also has been able to monitor biopotential signals because of its favorable electrical properties and piezoelectric responses.[118] Elastic deformations in the internal chambers of the conductive composite allow it to act as a pneumatic actuator useful in human robots.[111] Also, ceramic structures in the CR matrix have been interestingly reported to be useful in hydrophone applications as dielectric and piezoelectric materials.[119] The major challenge in developing a piezoelectric elastomeric substrate is the attainment of permanent polarization within the elastic materials which is a difficulty due to the disordered nature of the elastomers, particularly above their glass transition temperatures. Optimization of the mechanical properties with respect to the conductive and dielectric properties of the elastomers has been a major task for putting the material in applicability.

3.3. Triboelectricity

Triboelectricity refers to a coupling of frictional contact and electrical charge generation. "Tribos" is a Greek word that refers to rubbing two surfaces. This frictional contact creates charge transfer from one domain to the other allowing charge generation at the surface equivalent to static electricity.[120] Individuality in self-powered sensors has been exploited in the recent years through nano- or triboelectric generators or sensors in the health monitoring or electronic skin domain. The triboelectric effect is achieved by stacking polymeric or elastomeric films over each other and generating electrical charges upon mechanical friction through movement. Triboelectricity is the phenomenon that gives rise to self-sustainability to a soft electronic device in the long run. Park et al.[121] reported a fascinating work of porous PDMS with single-walled CNT (SWCNT) film with the characteristic of combined tactile pressure sensing and triboelectricity. To have superior characteristics than human skin with more sensitization, multifunctionality of electronic skins have been investigated. Arief et al.[122] explored the state-of-the-art research on the development, fabrication, and performance of TENGs designed for tactile sensing operations. The use of microstructured FKM and laser-induced graphene electrodes have been studied and fabrication done by the process of transfer printing. The TENG produced reflected a maximum power density of 715 mW m^{-2} and voltage of 148 V. It had a maximum current output of 9.6 μA. The composite also showed robust performance or electrical output even at 200% elongation.[122] Bionic and electronic skins remain the potential areas of application for the particular composite being discussed. Multifunctionality for commercial rubbers has always been an issue but addressal has been reworked on by the research community. One such example is reported by Natarajan et al.[123] where the triboelectric self-powering ability is studied by employing commercial GPRs fabricated through conventional solid-state mixing and vulcanization. A terpolymer of epichlorohydrin, ethylene oxide, and allyl glycidyl ether known as poly(epichlorohydrin-*co*-ethylene oxide-*co*-allyl glycidyl ether) (GECO) rubber shows the best output characteristics/power density as per the analysis done by the former. The highly positive polyethylene oxide renders the best triboelectric property when compared to other GPRs and silica-filled GECO proffers the best triboelectric performance with respect to other functional fillers like the conducting CB and graphene nanoplatelets. Surface modifications enhanced the power generation attribute for the matrix through micro-indentation increasing the surface area density.[123] Enhancement of the surface charge generation with high energy conversion efficiency in a practical way have been a primary challenge in the domain of elastomeric triboelectric substrates for industrial scalability.

3.4. Dielectric Functionality

Capacitance is the ability of a material to store charge and dielectric properties govern the material's ability to store and release electrical energy responding to an electric field. Upon fabrication of silicone rubber composites, the dielectric properties change due to the size and interfacial properties of the filler inclusions. Notably, Ibrahim et al.[124,125] found that the dielectric constant of silica/silicone nanocomposites is smaller than the corresponding unfilled network due to limited ability of the polarizable groups/segments to spontaneously orient with the applied electric field attributing to a discernible change in polarization in the frequency range of 10^3–10^5 Hz. This particularly refers to the dipolar orientation that gets restricted due to the higher interaction area in nanosize inclusions. The decrease in the size of inclusions causes an increase in the interfacial area further increasing the filler polymer interaction leading to additional restrictions on the mobile polar groups. Thus, the addition of excessive functional fillers may always not be a viable solution for enhancing functionality. It is important to note that even at lower frequencies provided with higher time scales of response, the free charges, defects, or grain boundaries at the interface can migrate under an external electric field causing an extrinsic polarization through non-Ohmic contacts. This phenomenon of space charge accumulation can contradictorily increase the dielectric constant in the nanocomposites depending on the frequency at which the study is performed. Klonos et al.[126] reported the interfacial phenomenon of core–shell nanocomposites of PDMS/silica nanoparticles. Observations and experimental studies through broadband dielectric spectroscopy showed that the interfacial relaxation was absent in neat PDMS but became predominant after surface modification and interfacial area increment upon the addition of fillers at frequencies ranging from 5×10^0 to 10^2 Hz. The dynamics of the interface or the semi-bound polymer chains become accelerated due to the non-Ohmic polarizability causing an increase in the dielectric constant compared to the neat samples. Functional properties of NR and its composites like the dielectric permittivity, dissipation factor, and insulating property can be tuned by the addition of fillers like barium titanate ($BaTiO_3$)[127,128] or nickel metallic nanoparticle.[129] In the former case, higher percentages cause deterioration in the flexibility of the compound however increases the dielectric constant of the rubber with better charge-storing capacity. In **Figure 7**a, the NR/$BaTiO_3$ composites dielectric study has been shown with a trend of increasing dielectric constant with increasing filler amount from 2.6 to 3.9 at 1 Hz due completely to interfacial polarization of the matrix in each case. However, with increasing frequency, the dipoles in the matrix are unable to orient themselves causing a decrease in the dielectric constant. Higher filler amounts at low frequency ranges cause lower dielectric losses whereas reach a saturation level without much change with filler amount. Increasing frequency causes a lowering in dielectric losses due to stronger interfacial interaction between filler–polymer networks.[127,128,130–132] In the case of Ni nanoparticles, dielectric permittivity and elastic modulus increased with increasing filler content attributable to the interfacial polarization from the heterogenous samples and there is an evidence of magnetization of the sample as depicted in Figure 7d.[129] Ceramic fillers with high dielectric constant like $BaTiO_3$ have been incorporated into NBR matrix for forming high-performance dielectric elastomer in soft electronics. The dynamic mechanical analyzer (DMA) curves illustrate the storage modulus (E′) and tan δ at different filler loadings in Figure 7c. Postulations from the figure include obstruction in chain mobility by the $BaTiO_3$ filler in the rubber matrix decreasing chain flexibility and reducing mechanical damping attributes also due to the good filler–polymer interactions. The dielectric constant study as shown in Figure 7b depicts the increased dipolar orientation of the composite with increasing filler loadings; however, with increasing frequency, the value drops due to varying electric fields. The frequency region studied in the figure is from 10^5 to 10^7 Hz where the space charge polarization effects cannot be considered.[133] Additionally, the polarity-induced adhesiveness of the elastomer along with hydrophobicity, oxidation resistance, flexibility, and cyclability allows it to be used in conductive adhesives in flexible interconnects.[134] CNT/silver (Ag) networks with NBR matrix has been reported to be used as conductive flexible adhesives also attributing to the stronger affinity of nitrile groups in NBR toward Ag.[134–136] Among the GPRs, comparative analysis depicts that NBR can be more suitable as a soft electronic substrate because of its functional property, high dielectric constant accompanied by low glass transition temperature and orientational polarization eradicating issues of incompatibility with functional fillers as seen in the case of nonpolar GPRs. In the vulcanized form of butyl rubber (IIR), it shows good dielectric properties along with excellent weathering properties. At the microwave region, IIR has a dielectric constant of 3.4 at around 8–12 GHz (X band) and 2.3 at higher frequencies (K band).[137] The dielectric constant decreases with increasing frequency due to limited orientation polarization. Various reports on rigid polymer ceramic composites have been found so far but elastomer ceramic composites in dielectric applications are obscure.[138] Butyl rubber has a relative permittivity around 2.4 and dielectric loss tangent at the order of 10^{-3} at microwave frequencies[139] where the permittivity was reported to be enhanced by 0.42 volume fraction of $SrTiO_3$ (strontium titanate) ceramic filler without compromising on the loss and maintaining flexibility and stretchability in the composite.[140] Around 1000% strain at break was reported signifying no loss of chain flexibility inherently present in the elastomer upon addition of the filler. CTE also decreased upon inclusion of the ceramic filler for the composite to be qualifiable as a key candidate in soft electronics at microwave frequencies.[141,142] Similar reports have been observed for functional fillers like $Sr_2Ce_2Ti_5O_{16}$,[143] $Ba(Zn_{1/3}Ta_{2/3})O_3$,[144] SiO_2,[145] $BaTiO_3$,[146,147] $Ba_{0.7}Sr_{0.3}TiO_3$,[148] and TiO_2[149] composites in the microwave domain. Butyl rubber and alumina are reported to be favorable in electronic packaging materials due to the low CTE of the latter combined with the flexibility of the former.[150] Soft electromagnetic interference (EMI) shielding applications are also looked into through composites of SWCNT and butyl rubber.[151,152] In all these cases, the dielectric relaxation along with electron mobility accounts for the enhancement of relative permittivity. For SWCNT composite, interfacial and electronic polarizations increase the relative permittivity in the composite along with the conductivity due to the characteristic EMI shielding. Functionality induction through carbon particles like CNTs, graphene sheets, CB, and metallic inclusions like Ag particles, nanowires and flakes, and Au nanotubes is in

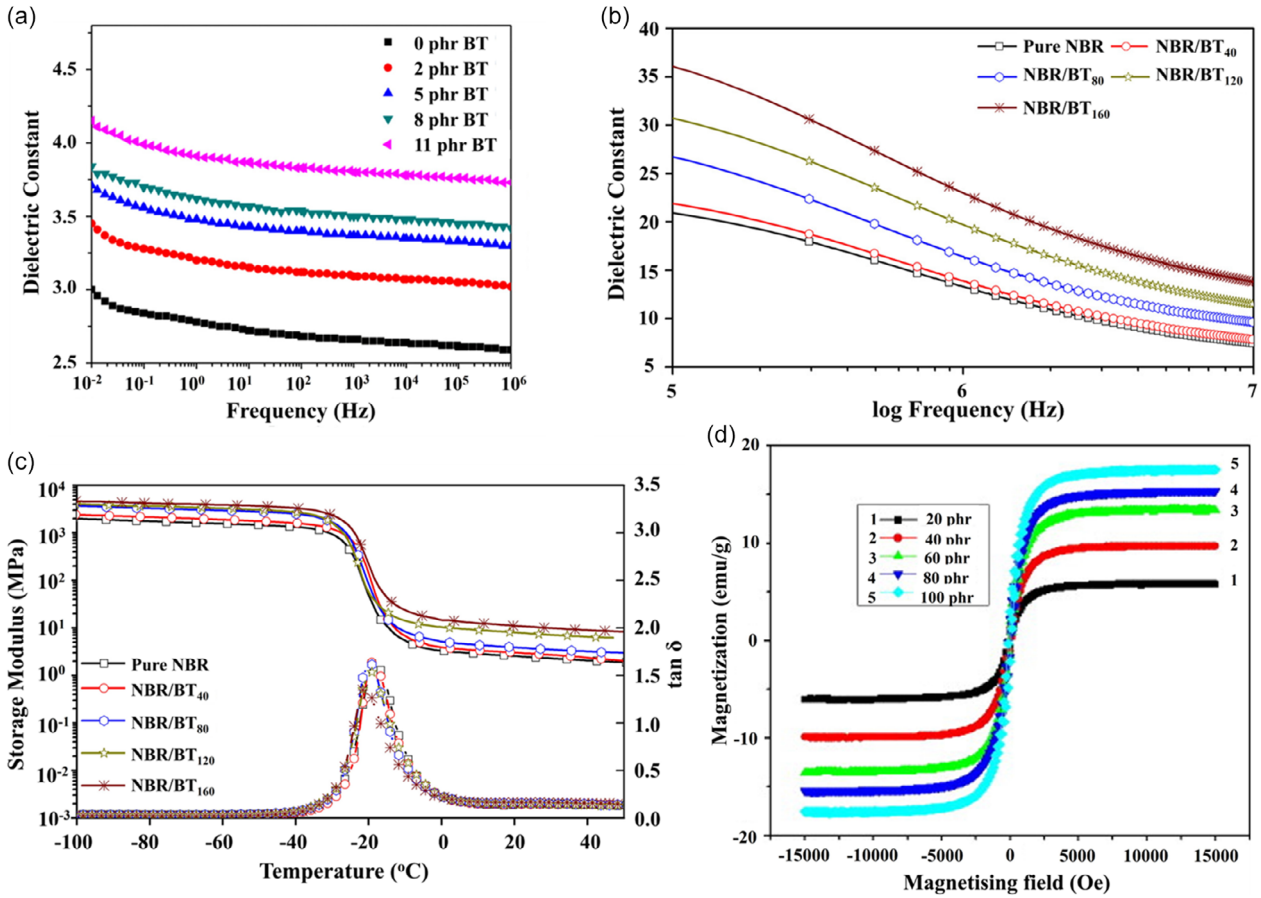


Figure 7. a) Dielectric constant study of natural rubber and barium titanate filler at various parts per hundred rubber (Adapted with permission.[132] Copyright 2021, John Wiley and Sons). b) Dielectric constant for NBR/barium titanate composites and pure NBR. Increasing filler amount increased the dielectric constant due to presence of permanent dipoles in the ceramic filler; however, there was a decrease observed with frequency due to loss of dipole orientations at varying electric fields (Adapted with permission.[133] Copyright 2020, John Wiley and Sons). c) Storage modulus (E′) and tan δ versus temperature for different NBR/barium titanate composites and pure NBR showing a decreasing chain flexibility with increasing filler amount (Adapted with permission.[133] Copyright 2020, John Wiley and Sons). d) Magnetic hysteresis of NR/Ni composites showing ferromagnetic behavior with increasing saturation magnetization with nickel content (Adapted with permission.[129] Copyright 2009, Elsevier).

common research.[153] However, LM incorporation combining high strength-to-weight ratio forming soft and compliant materials without mechanical property degradation in flexible PDMS substrates has been in light recently.[1,3] This has been in use for applications in soft capacitive pressure sensors and innumerable functional circuits. Similarly, another highly functional filler for electrical conductivity induction in PDMS is graphene foam.[154] The proof of high dielectric actuation has also been developed with dynamic property analysis through the studies of Shakun et al.[155] on actuation performance of normal diene rubbers. It focused on the dynamic stability of the matrices with stable resilience and electrical properties. The conclusion came to the fact that dielectric elastomeric actuators should not be bound to silicone and acrylic commercial elastomers but should also be tuned to various combinations by fine-tuning the matrix with the addition of plasticizers and varying acrylonitrile content in the matrices. There was a generation of stronger force or larger strain depending on the application of use.[156] Upon summarizing the recent researches, it can be concluded in a sense that hysteresis becomes a very important design parameter while designing capacitive sensors for practical applications. Hysteresis is a parameter that arrives directly from the viscoelastic nature of the polymers hence cannot be neglected but requires optimization while designing such devices under dynamic load conditions. High linearity with lower response times is more expected from the capacitive sensors than the resistive ones through the analysis of hysteresis behavior.[89]

3.5. Conducting Soft Materials for Compliant Electrode

Electrodes with good mechanical compliance and electrical properties are a key component in any soft electronic device. Gao et al.[157] reported the use of Ag nanowires in to PDMS substrate to fabricate a transparent and intrinsically stretchable electrode. This allowed fabrication over large-area electronic skin and with a low sheet resistance of ≈18 $'\Omega\,m^{-2}$. PDMS is transparent in the UV–visible range following applications in transparent electrodes for flexible touchscreens, organic light emitting diodes, solar

cells, and wearable electronics,[158] all in the domain of soft electronics. This transparency can also be modulated using optical fillers. In addition, the refractive index of PDMS is around 1.4 at wavelengths from 589 to 1554 nm.[153,159] Tuning of refractive indices allows the formation of optical waveguides solely based on PDMS integrated in a microfluidic system.[153] Nanocomposites with high refractive index have been reported for siloxane-derivatized zirconia ZrO_2 nanoparticles in a PDMS matrix (refractive index ≈1.39–1.65 by varying ZrO_2 content) with transparency in the UV–vis >93%.[160,161] Other composites like TiO_2 nanoparticle dispersion in PDMS are done to achieve refractive index up to 1.9.[162,163] Formation of stretchable conductors also allows the fabrication of conducting electrodes as explored by Narongthong et al.[95] Commercially available CB-filled SBR can be used as a stretchable conductor upon fabrication with ionic liquids to prevent modulus mismatch and enhance the stretchability of the matrix. The ionic liquids enable the alignments of the aggregates even better at stressed conditions allowing maximum conducting strain.[95,164,165] The roadmap to sustainable elastomers has been explored with the help of recyclable vegetable oil–based PU elastomeric binder and Ag flakes. This composite makes up a printed sustainable elastomeric conductor with 350% of stretchability, 12 833 S cm^{-1} conductivity, and very low hysteresis (0.333) even after 100% cyclic stretching. This pioneering work has been done by Jian et al.[166] for the development of food maturing sensors in the domain of soft electronics. Thin-film gold deposition has also been a major contributor as flexible electrodes due to its excellent conductivity, chemical stability, and biocompatibility. Thin-film interconnects have been made through various fabrication techniques like electroplating and electroless plating due to the plethora of favorable properties. The electroplating technique involves the use of electrolysis to deposit the gold whereas the electroless plating makes use of the electric current. McClain and co-workers[167] have established neuro-electrodes with thin-film PDMS and gold deposition with minimal modulus mismatch with the neural tissues. The fundamental sensing mechanisms behind the thin films can be through disconnection mechanisms or crack propagation strategies.[89,90] In the conductive thin films formed of functional materials like nanofillers and elastomers, the electrons can form a percolation network to pass through and stretching for stimuli generation can cause breakage in that network increasing the electrical resistance of the substrate. This can be addressed by improving the interfacial bonding between the phases and diminishing the modulus or stiffness mismatch, the latter being the most evident reason of failures in elastomeric substrates and rigid fillers. Cracks also initiate at stress concentration points to release the accumulated stress at nodal points of modulus mismatch and external stimuli can even increase the microcracks to a larger length-scale leading to propagation and increase in electrical resistance.[90] Another important factor is the choice of material as an electrode material for a specific application. For example, the critical design aspect lies in the fact that it is extremely difficult to assign elastomeric electrodes to a flexible substrate where both the components have similar deformation behavior. However, mechanical characteristics are essential as in the case of a dynamic application; a differential deformation of different layers would result in delamination and device failure. In this regard, the fundamental question that can be framed is whether the electrode and the substrate matrices be identical or the functional filler that ought to be utilized be similar. If research evidences of the most suitable conducting fillers, such as CNT, graphene, and CBs are to be employed, loss of matrix compliance is imminent. However, to overcome such difficulty, rather than a filmlike electrode, engineered structures like wavy structures, 2D serpentine springlike structures, or meshes, and coil-like structures which can render even a brittle material stretchable to some extent, can be utilized.[168,169] The engineering complexity would agreeably arise in such a case which is subject to extensive research, but the loss of flexibility due to rigid filler loading might be amicably countered.

4. Fabrication Approaches of Soft Electronics' Substrates

Fabrication of the polymeric substrates for soft electronics requires various techniques for enhancement of the multifunctional behavior. Elastomers can be subjected to solid-state processing, solution, or melt processing depending on the structural properties of the material or latex compounded.[170] The state-of-the-art functional elastomeric material with enhanced dielectric strength, magnetization, and thermal conductivity is one of the most needful materials as has been discussed in the previous sections for the recent day flexible electronics and soft robotics industries. In the domain of soft electronics substrates, numerous processing methods have been utilized, like solution casting for film formation,[95] compression molding[171] for sheet formation with schematic in **Figure 8**e, extrusion for fiber morphology,[172] and ultimately 3D[173] and 4D printing[174] for stereoscopic and stimuli-responsive stereoscopic morphology generation as part of the newer approaches. The fabrication process can be henceforth synopsized by taking an example of a silicone elastomer for the formulation of a soft electronic substrate. Formulation of the customized silicone elastomer requires an initial drafting of the network structure of the polymer with appropriate functional fillers to be used in the processing further. For thin-film application, PDMS is mixed with an appropriate catalyst and filler and then cast for achieving the required thickness. Curing and post-curing of the samples take place and for further fabrication later-mentioned techniques and processes are implemented.[153]

4.1. Solution Casting

Rana et al.[175] investigated solution-processed SIS block copolymer in toluene and nickel ferrite powder in a high shear mixer for uniform dispersion and cast until required thickness for fabrication of a flexible substrate with uniform and superior functional properties. The schematic of the fabrication method has been illustrated in Figure 8a. Figure 8b–d shows the microstructural change of a solution-cast LM-filled block copolymeric elastomer with increasing filler content from neat to 20 vol% filler. The increase in the particle size upon increasing filler amount can be due to fabrication conditions or a situation known as in situ mechanical sintering. The shearing action of the blades in the high-shear mixer causes the particles to break into finer droplets that can cause coalescence for surface energy reduction. Also,

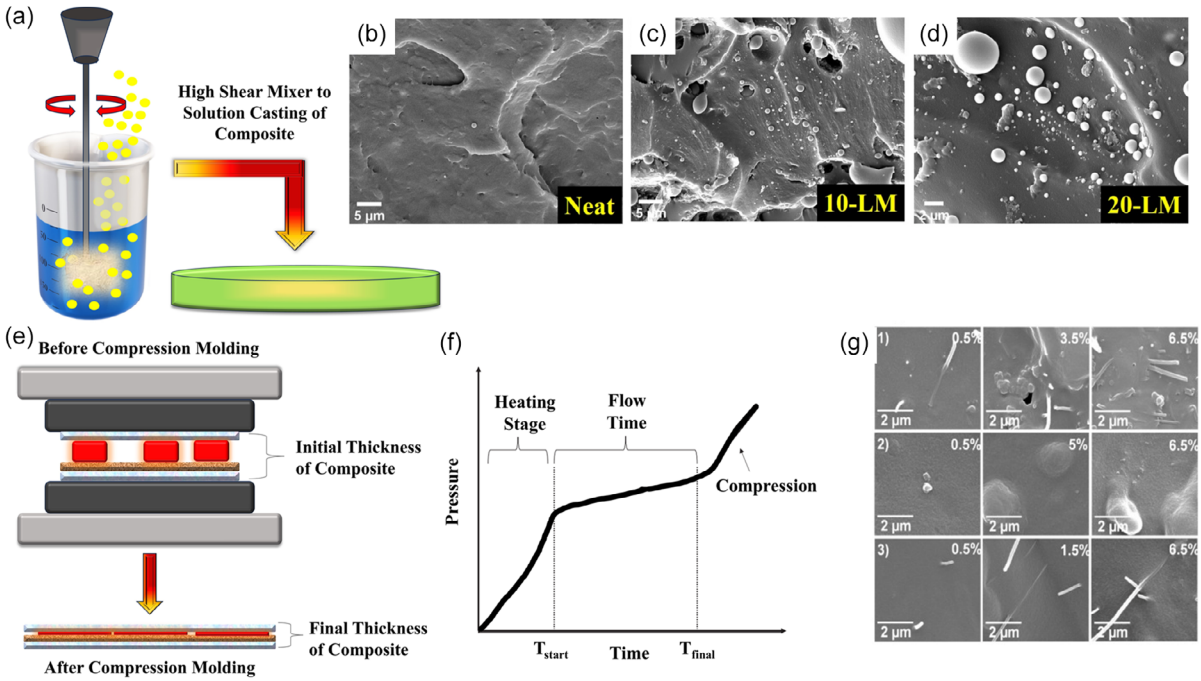


Figure 8. a) Schematic diagram of high shear mixing of composite to solution casting. The microstructure of solution-cast b) neat SEBS-g-MA (SEBS-grafted maleic anhydride) block copolymer; c) 10 vol% LM SEBS-g-MA and d) 20 vol% LM SEBS-g-MA binary composites (Adapted with permission.[87] Copyright 2023, Springer Nature). e) Schematic diagram of the compression molding of a composite.[171] f) Generalized pressure profile in compression molding (Redrawn with permission.[201]). g) SEM (scanning electron microscopy) images of compression molded composites of 1) carbon black—vapor-grown carbon nanofiber SEBS; 2) carbon black—SEBS; and 3) vapor-grown carbon nanofiber SEBS at different filler loadings (Adapted with permission.[202] Copyright 2021, Elsevier).

the localized compressive stresses in the solution-cast sheets cause the coalescence of the particles depicting a larger droplet morphology.[87]

4.2. 3D and 4D Printing

Extrusion being a conventional technique for processing polymeric materials has also been explored in the fabrication of soft electronic substrates. It involves melt processing to form a composite microstructure. This technique has been widely adapted in the state-of-the-art additive manufacturing processes discussed in the further sections and represented in **Figure 9**a. Originated from the conventional extrusion technique for material manufacture, these are the state-of-the-art manufacturing techniques. Substrates with embedded sensors and actuators for robotic applications have been established through shape deposition manufacturing that amalgamated the use of soft and compliant parts.[4] A hybrid process of machining and molding enabled the fabrication through a layer-based deposition process. Corresponding functionalities can thus be integrated into the compliant structures. Sticky Bot is an example of this process in the biomimetic field of soft electronics. Sticky Bot is a gecko-inspired elastomeric robot that can climb up glass-like surfaces also showing dry directional adhesive due to its multi-material limbs.[176] Another such emerging process of fabrication of flexible electronic substrate is smart composite microstructure (SCM) that combines the traditional rigid links to the soft elastomeric or polymeric joints. For manufacturing substrates and structures with integrated compliance and functionality in the mesoscale, this technique is more expedient.[177] Engineering of the flexible body can be done through techniques like nanoimprint (a mold-assisted photolithographic process advancing microscale precision),[178] laser imaging,[179] laser ablation,[180] microinjection molding,[181] embed molding,[182] layup: composite, soft material deposition process, nanocomposite deposition system (functional system manufacturing in layer-by-layer or scaffold pattern), and multiple nozzle deposition system (large-area manufacturing in shorter time).[4] The soft material deposition process is a prompt prototyping system.[183] One of the classes in this process is the fused deposition modeling which is a thermoplastic-based process.[184] The other class in soft material deposition is based on photo-cross-linkable polymer deposition system as in Polyjet.[185] Actuators being one of the important classes of application in the flexible electronics domain can be fabricated by the ensuing methodologies: shape memory alloy embedding process (utilizing shape memory alloy–coiled spring and foil by SCM approach);[186] piezo-composite actuator manufacturing (by the process of SCM to enhance deformations in the actuator).[177] For stretchable electronics in the soft robotics domain, complex anatomy and construction are required in the substrate like the mesh,[187] vertical ripple,[188] planar horseshoe,[189] and spontaneous wrinkling.[190] Taking the instance of the widely used elastomer in flexible electronics that is PDMS, the substrate manufacturing requires a step-by-step approach.

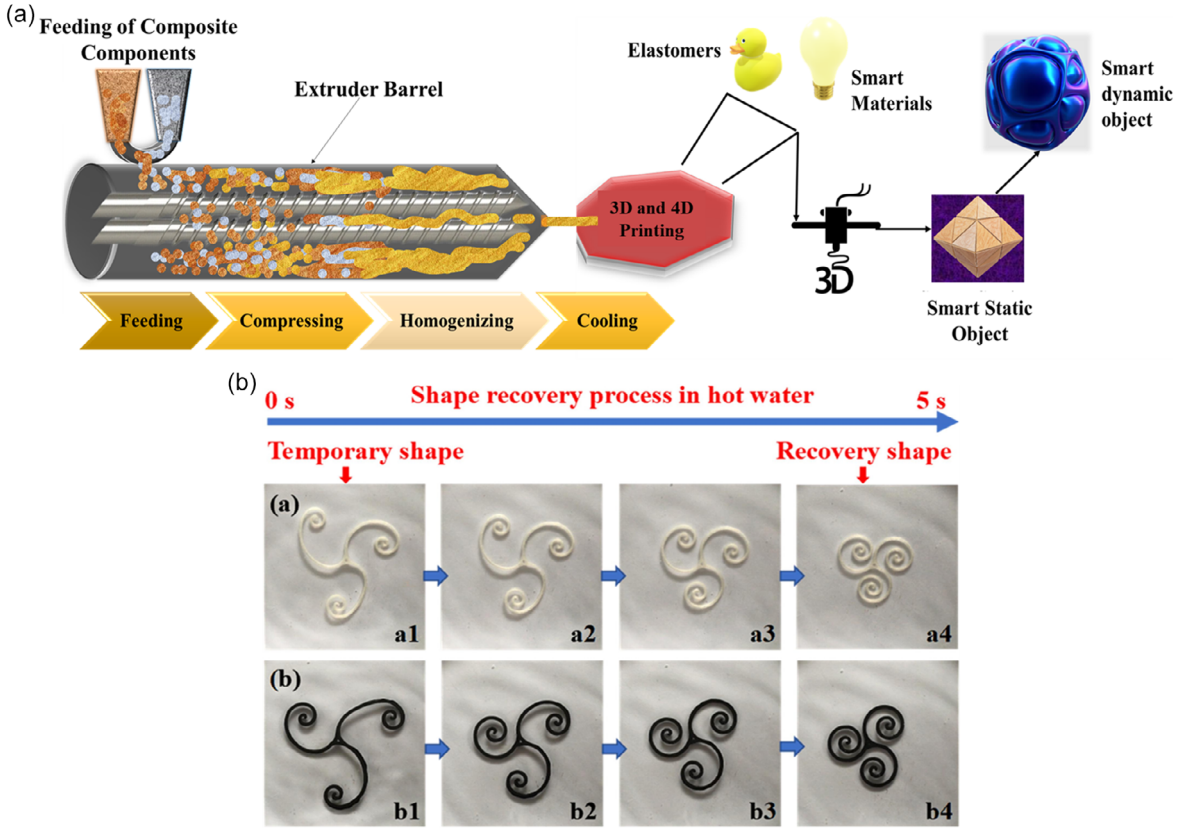


Figure 9. a) Schematic diagram of extrusion process through which the state-of-the-art technologies of 3D and 4D printing emerge. b) Depiction of how smart materials (time as a factor with respect to stimuli [hot water]) emerges in 4D-printing technologies (Adapted with permission.[196] Copyright 2019, Elsevier).

Printing of diversified static parts for soft electronics substrates recently takes credit through additive manufacturing approaches like material extrusion,[191] sheet lamination,[192] material jetting,[193] direct energy deposition,[194] etc. Smart structures with multiple stable conformations have been the pioneering work on fabrication of flexible and soft electronics from 2013 through the process of 4D printing.[195] Time-dependent assembly of numerous sensory and functional parts are done by this process of fabrication depicted in Figure 9b.[196] At times, numerical simulations are performed to quantify and hence testify the performance of the 4D-printed material. The newer printing technologies in additive manufacturing with even smarter soft materials have been in research like the 5D- and 6D-printing technologies.[197]

4.3. Lithography

The combination of producing the best anticipated flexible and soft electronic device requires the combination of large area, compliant substrate, high resolution, and high efficiency with the lowest cost. This proposes a challenge to the manufacturing demands and newer technologies being proposed for more and more scalability and effectivity. Techniques like laser direct writing (LDW), scanning probe lithography (SPL), interference lithography (IL),[198] and directed self-assembly (DSA) have been proposed as methods for fabrication however with some disadvantage in each case. For the LDW, low scan speeds from a single laser and complex control of the focal depths limit its range of applications. For SPL, similar scanning speed and low process efficiency corresponds to less usage of the application. IL and DSA produce large-scale maskless patterning but with high fault rate. Nanoimprint lithography (NIL)[199] has been one of the most recent techniques of fabricating flexible electronic devices with organic polymers, liquid inks or nanomaterials, and composites, anatomically hierarchical, slant, or with multiscale structures. NIL corresponds to a mold–demold process with high resolution and low cost mostly requiring mechanical pressure for filling of molds. The latter is addressed as an issue in the technique however prevented using a differently designed NIL. This process is generally of three types: thermal NIL, UV (ultraviolet) NIL, and electrical NIL. The thermal NIL involves the patterning of thermally molten polymer and UV NIL involves the use of UV curable polymer, both having a drawback of mechanical pressure causing the filling of mold. Electrical NIL however is a modification useful for patterning without mechanical stimuli, because of the use of electric field–induced surface tension force. This process of fabrication helps in the manufacturing of transparent conducting electrodes, optoelectronic devices, flexible microlens, sensors,[200] and bioinspired adhesives.

4.4. Molding

Molding can be done by both injection and compression. Large-scale intricate composites are injection molded; however, the compression molded profiles are more apparent (Figure 8e) in this domain of research. The generalized pressure profile for sheet compression molding is shown in Figure 8f.[201] Cetin et al.[202] investigated the use of compression molding approach for forming hybrid nanocomposites of filled poly[styrene-b-(ethylene-*co*-butylene)-b-styrene] (SEBS). The microstructure of the compression molded soft composite as shown in Figure 8g formed by SEBS block copolymer along with CB and vapor-grown carbon nanofiber (VGCNF). The morphology depicts a synergistic effect of both high shear mixing and compression molding emulating on the highly dispersed polymer–filler matrix. For the CB–SEBS composite (Figure 8g2), spherical particles of CB have been observed to be dispersed in the SEBS matrix with proper conductive networks at the interfaces. For the VGCNF–SEBS composite, a dominant orientation is not found (Figure 8g3) for the filler particles however showing high nanoscale roughness consequently an enhanced polymer–filler interlocking. A significant orientation is yet required for necessary electromechanical properties. In the CB–VGCNF–SEBS composite, CBs have filled the spaces between the nanofibers and matrix which was immediate for electrical conductivity as shown in Figure 8g1.[202]

4.5. Inkjet Printing

Inkjet printing directly forms a deposition of solution processable materials onto a given location of substrates. It has several advantages over other printing techniques—being programmable with variable patterning allowance without extra masks, minimal limitations on optical focus depth consequently obtaining high resolution patterns, and minimal requirements for planar, conductive, smooth printing substrates with high processibility in both organic and inorganic inks. Being a non-contact printing process, it involves least contamination with high-throughput rates at lower temperatures. Commercially being a very cost-effective process in itself, several areas of research become feasible to address the limitations of electrical conductivity, device stretchability, or even mechanical durability. Chemical and physical manipulation of the inks can cause improvement in pattern anatomy and prevent cracking. Nozzle clogging remains a universal problem however can be addressed through further research for a complete full-proof device manufacture.[203] However, as discussed in the preceding section, due to the wide range of the characteristics of the base elastomers, it is extremely difficult to generalize a handful of strategies for designing elastomeric soft electronic substrates. The challenges associated with the processing aspects of each of the individual elastomers, and the routes necessary to establish synergy with either fillers or with other components involved in a device have rendered this as a very niche research domain.

The optimization of the sensing factors in a dynamic environment is crucial apart from fabrication for addressing practical scenarios depicted in the following sections.

5. Mechanical and Dynamic Performance of the Soft and Functional Materials

Impact absorbance in terms of robustness to cyclic dynamic stresses is a critical requirement for the material to be used in the flexible electronics domain. Now, flexibility is a mechanical characteristic to be conveniently classified into three categories: 1) bendable/rollable; 2) permanently shaped; and 3) elastically stretchable (large and reversible deformation).[204] The strain at break should reach limits greater than 300%. Low elastic modulus interconnect layers reduce surface elongation where active components of the circuitry is placed[72] and hence the elastic modulus of the soft substrates should range between 10^{-2} and 10^{3} MPa. The electrode materials used like printed carbon composite, PEDOT:poly(styrenesulfonate) (PEDOT:PSS), iridium, platinum, etc., in the interconnecting layers have modulus values ranging from 1 to 10^{5} MPa. For applications of electronic skins with biomimetic properties, flexible substrates modulus values range even lower from 10^{-4} to 10^{-1} MPa.[72,73] The materials used in flexible electronics generally have a Shore A hardness of 40–90.[3] As reported, PDMS substrates have Shore A range between 48 and 58,[205] although PDMS specimens of yet lower Shore A hardness are possible to such an extent that can mimic the strength of a biological muscle and skin tissue. Furthermore, PDMS electrically conductive adhesive exhibits noticeably retained elasticity under dynamic deformation conditions as evident from a DMA curve in Figure 7f. TPU–CNT composite have a higher Shore A scale up to 90 in 10% carbon filler loading and similarly with graphite has 87 Shore-A hardness at the same filler loading.[206]

The mechanical modulus and the hardness are but one aspect of the structural behavior of a soft electronics substrate. For a static application utilizing a soft substrate, these might suffice, but in case of a dynamic application requiring dimensional and performance stability against long-term cyclic loads factors like fatigue failure, modulus drop, Payne effect (for filled rubbers), and others come into consideration.[207] The most common strategy of characterizing the dynamic performance of a soft, viscoelastic material like elastomers is the DMA, whereby a sinusoidal stress or strain is applied to a specimen as the input signal, typically expressed as a complex strain (ε^*) comprising of both static ($\varepsilon_{\text{static}}$) and dynamic parts ($\varepsilon_{\text{dynamic}}$) given by[208]):

$$\varepsilon^* = \varepsilon_{\text{static}} + \varepsilon_{\text{dynamic}} = \varepsilon_{\text{static}} + \varepsilon_0 e^{i\omega t} \tag{3}$$

Equation (3) corresponds to an input strain signal whereby, the ε_0 is the strain amplitude, ω is the frequency of the input strain and t is the time. The elastomeric material, being viscoelastic, responds in a sinusoidal manner but involves a certain phase lag. The typical dynamic stress (σ_{dynamic}) response is given as:[208]

$$\sigma_{\text{dynamic}} = \sigma_0 e^{i\omega t+\delta} \tag{4}$$

The term δ in Equation (4), is the phase lag and the aforementioned equations can be extrapolated to a term called the complex modulus, G^* expressed as:[209]

$$G^* = G' + iG'' \quad (5)$$

The G' and the G'' terms in Equation (5), are the storage and loss modulus, respectively of which storage implies the portion of the mechanical energy imposed on the specimen is actually used up for useful mechanical work, and loss implies the portion lost by macromolecular occurrences, such as chain orientation, disentanglement, etc. and reflected as viscous heat dissipation. The ratio of these two terms: $G''/G' = tan\ \delta$, or the dissipation factor is an exact quantification of the loss of mechanical energy, and typically for the efficient functioning of a device under dynamic conditions, a low $tan\ \delta$ value is desirable. In addition, the complex modulus can be obtained from the DMA under various parameter sweep conditions such as the strain, frequency, or the temperature sweep. One interesting insight can be drawn from the Payne effect study which is of critical importance in tire rubbers, as critical information regarding filler–rubber and filler–filler interactions can be obtained which empowers the choice of suitable reinforcing filler in tire rubber. Such a comparative plot in tire rubber for CB and silica is shown in **Figure 10**a. The importance of Payne effect and its possible utility can be stated from the fact that it would significantly ensure a judicious choice of a functional filler necessary in an elastomeric matrix, depending upon the area of application, the range of strain amplitude, temperature, and others. However, irrespective of all factors, the general idea should be that the filler–rubber interaction should be maximized so as to ensure a maximum performance synergy and limited incompatibility issues which would otherwise result in cases of premature critical failures. For a more in-depth understanding of the response of the

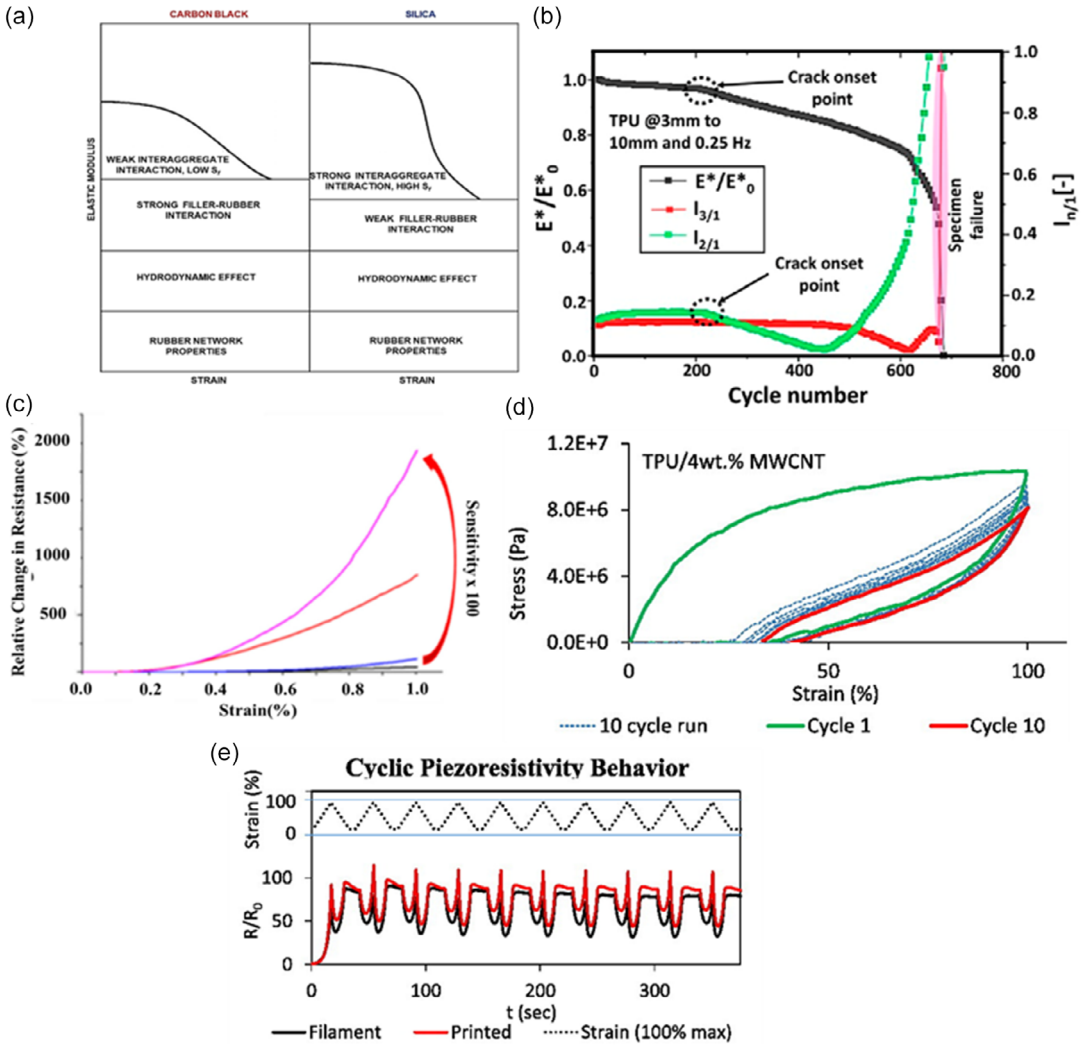


Figure 10. a) Comparative plot showing Payne effect in tire rubber for carbon black and silica (Redrawn with permission.[207]); b) FT rheological analysis of a TPU specimen showing the point of crack origination based on the fingerprint harmonics obtained from a Taylor expansion of the specimen's response to cyclic stress (Adapted with permission.[208] Copyright 2024, Sage Publications); c) representation how increasing strain increases sensitivity toward change in relative resistance; d) hysteresis analysis of a dynamic strain sensor with elastomeric matrix (Adapted with permission.[273] Copyright 2017, Elsevier); and e) cyclic piezoresistivity sensor for dynamic durability/endurance testing for the elastomeric soft matrix for applicability in practical scenarios (Adapted with permission.[90] Copyright 2020, Elsevier).

material toward such cyclic stresses and the fatigue failure, the works of Wilhelm et al.[210] Lacroix et al.[211] and Hirschberg et al.[212] on Fourier transform (FT) rheology of elastomeric materials, are of utmost significance. In an FT-rheology experimentation, the complex modulus (G^*) of a specimen under complex strain of Equation (3) is expanded by a Taylor expansion as[208]

$$G^*(t) = G_1\varepsilon^*(t) + G_2[\varepsilon^*(t)]^2 + G_3[\varepsilon^*(t)]^3 + G_4[\varepsilon^*(t)]^4 \ldots \quad (6)$$

$$\sigma^*(t) = I_0 + I_1 e^{[i(\omega t+\delta_1)]} + I_2 e^{[i(\omega t+\delta_2)]} + I_3 e^{[i(\omega t+\delta_3)]} + I_4 e^{[i(\omega t+\delta_4)]} \ldots \quad (7)$$

Equation (6) indicates the Taylor expansion of the complex strain and the alternate version in terms of harmonics is provided in Equation (7). The harmonics I_1, I_2, and I_3 are the fingerprint of a material and the first harmonic is indicative of the linear response while the second and third harmonics deals with the nonlinear material response arising from the crack formation and propagation in the specimen under deformation. Particularly, the second harmonics articulates any anisotropy due to the appearance of macroscopic cracks given by a sharp drop in its value and the decreasing third harmonics correspond to the formation of a crack, its gradual propagation, followed by a sudden rise due to critical failure. Such a similar observation is showcased by Awasthi et al.[208] in case of TPU and its blends. And the important facet of this method is the accurate information which can be drawn about the specimen service life preventing a catastrophic mishap or damage of any device (Figure 10b). This kind of mechanical understanding is not only important for choosing the optimal soft matrix for a suitable application but also can be coupled with devices requiring a continuous stress input, such as piezoelectric or piezoconductive devices, actuators, and transducers or in any case where mechanical energy is coupled with any other functional property. Additionally, mechanical properties at the soldering positions or the joints require the study of dynamism and cyclic behavior for tactile sensors forming soft electronic devices. Various polymers used in the soft electronics have been studied and modeled according to their viscoelastic responses.[213] The TPU matrix generally shows a hyper-viscoelastic mechanical behavior and high tensile rubbers tend to have the elastic behavior. Modeling of hyper-elastic nature of the elastomers at finite deformations requires an appropriate strain energy function to replicate the experimental data. The Neo–Hookean, Mooney Rivlin, and Ogden models have been used to obtain the accurate hyper-elastic strain energy function for nonlinear elastic responses, however having drawbacks in fitting data accurately without the proper representation of material instability under the different deformation modes. The resistance values do not matter much in the domain of soft sensors at the different modes of deformation. Here, the change in resistance is a much more crucial factor to map and model the deformation behavior represented by different metrices like RR and gauge factor (GF). This helps in quantifying the change in resistance and sensitivity in the stretchable sensors as per Equation (8) and (9).

$$\mathrm{RR} = \Delta R/R_o \quad (8)$$

$$\mathrm{GF} = \mathrm{RR}/\in \quad (9)$$

ΔR is the change in the intrinsic resistance, R_o is the initial resistance of the sensor, and ε is the logarithmic strain of the sensor represented in Figure 10c. Kouchakzadeh et al.[214] investigated and simulated the behavior of a TPU sensor with 4 channels and obtained a GF greater than 2 showing high sensitivity to applied strain through mapping and modeling of strain energy function, GF, and RR. A GF greater than 2 indicates a high strain sensitivity and effectivity.[214] Another crucial drawback of the aforementioned hyper-elasticity models is the estimation of the material to be an incompressible material having a Poisson's ratio of 0.5,[215] which might be suitable for a pristine elastomer but may deviate significantly in the case of a rubber blend or a functional filler-loaded elastomeric composite.[216] In such cases, it would be safer to take volumetric deformation of the material under consideration in the strain energy expression. Other performance factors when considering the dynamic behavior includes hysteresis, dynamic durability, and linearity.[89] Hysteresis property is the most important factor when used for skin-mountable or wearable applications depicted in Figure 10d.[89] The response and recovery time behaves in response to the hysteresis property of the matrix. The long-term durability or endurance of the material can also be speculated from the dynamic durability design factor corresponding to how many load cycles the matrix can sustain as in Figure 10e.[89] Linearity is a directly proportional relationship between the applied stimuli and electrical signal obtained. Complicated regression analysis is required for the linearity analysis in the matrices and viscoelastic nature of the polymer does not allow the same.[202]

6. Applications

The preceding sections attests to the certitude of stretchable and flexible electronics surpassing the currently used rigid and bulky forms of materials through their robustness, continuous strain dissipation, and versatility. These rubbery materials are used for wide range of applications including bioelectronics consisting of electronic skins[217] and flexible wound dressing–integrated electronics,[218] sensors,[6] actuators,[219] self-powered devices (energy-harvesting devices),[220] electronic displays,[221] soft robotics,[13] new age metamaterials,[222] etc., depicted pictorially as a scale of increment from the basic matrix materials and functional fillers to ultimate applications in **Figure 11**. These applications have also been tabulated in **Table 1** with regards to the recent developments in the associated academic community also highlighting the challenges associated with the fabrication of such devices in the compliant form. Elaborative discussions on each of the aspects have been covered in this section.

6.1. Soft Robotics

Human–machine interaction, being one of the refinements in the technologies of flexible electronics and soft robotics, enhanced by wireless communication mitigates any disordered

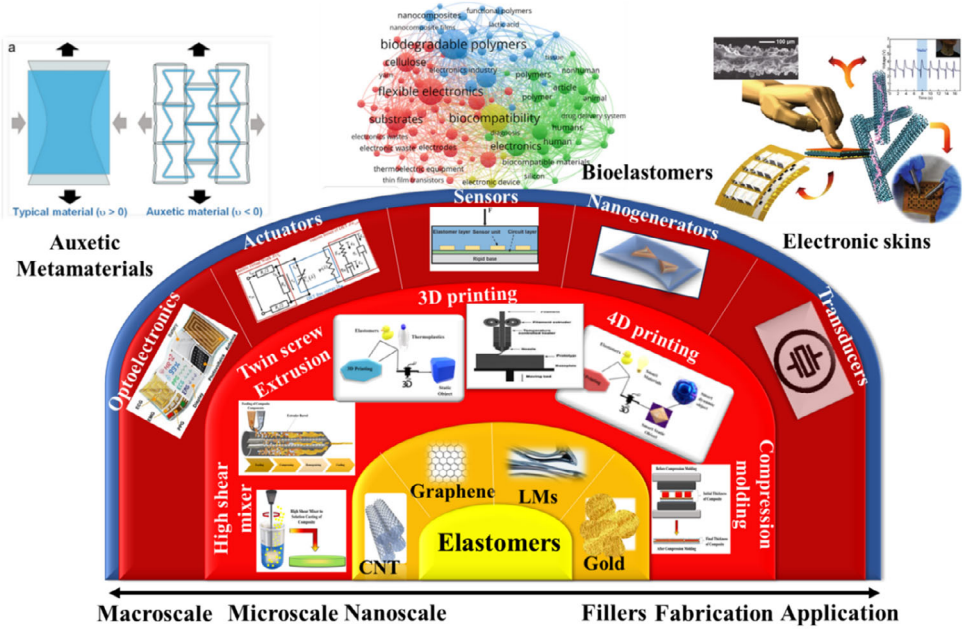


Figure 11. Schematic overview of the functionalized elastomers with fillers like CNT (Adapted with permission.[274] Copyright 2022, Springer Nature), graphene (Reproduced under the terms of the CC-BY license.[275] Copyright 2018, The Authors, Spinger Nature), LMs (Adapted with permission.[276] Copyright 2022, Springer Nature), gold, etc., along with their fabricating techniques like high shear mixing for solution casting, twin screw extrusion, 3D and 4D printing (Adapted with permission.[277] Copyright 2021, Springer Nature), and compression molding. The primary applications of flexible and stretchable elastomers include sensors (Adapted with permission.[278] Copyright 2022, Springer Nature), actuators (Reproduced under the terms of the CC-BY license.[279] Copyright 2022, The Authors, Spinger Nature), optoelectronics (Adapted with permission.[280] Copyright 2021, Springer Nature), transducers[281,282] and nanogenerators, and the multiparametric approaches lead to the new age applications of electronic skins (Reproduced under the terms of the CC-BY license.[283] Copyright 2019, The Authors, Spinger Nature), bio-elastomers for sustainable electronics (Adapted with permission.[284] Copyright 2024, Springer Nature) and auxetic metamaterials (Adapted with permission.[285] Copyright 2023, Springer Nature).

restrictions in the advancement of the same. The introduction of tunability of the electrical response upon application of stimuli in the sphere of flexible electronics has been an entailing attribute to broaden practicality. Soft matter engineering and soft robotics have been extensively reviewed by Majidi et al.[13] to emphasize on the fact that, elastomeric grippers schematically represented in **Figure 12**b(i), have high leverage on the tunability of stiffness and compliance to establish a conformal and adaptive contact with objects finally allowing humanlike sensing due to actuation capabilities.[13] Xu and co-workers[223] reported the development of a viscoelastic sensor with magnetic on- and off-switch through LM-filled magnetorheological PU matrix. The LM and the magnetic carbonyl iron particles used in the study would form a chain-like structure upon application of magnetic field via magnetic domain alignment facilitating a conductive path for generating electrical responses depicted in Figure 12b(ii). Also being an insulator at room temperature (RT), and upon application of magnetic field, temperature, and force, feedback signals would be conveniently generated for a multifunctional multi-sensing characteristic performance, or for magneto-responsive fast charging devices, thermoregulatory devices, and many more. However, there remains a limitation due to the assumption of separated LM droplets upon application of magnetic field in varied directions so further studies need to be done. Another stumbling block in the field of switchable and tunable electrical responses for multiple sensing ability lies in the fact of signal cross talk between the interfering parameters. This cross talk creates an interfering signal due to undesired capacitive, inductive, or conductive coupling. Strategies and researches analyze electrical cross talks for improving and manifesting decoupling mechanisms under multiple stimuli.[9,224]

6.2. Electronic Skin

Enlightening on the revolutionary need for bioelectronic materials for stimulations, monitoring, and tactile sensing for the holistic understanding of disease conditions, electronic skins through elastomeric functional materials have proved to be a pioneer in mimicking the myriad attributes of human skin schematically represented in Figure 12a(i). The human skin can be largely strained without any permanent deformation enabling mobility to the human body. Along with that, self-healing attribute of the human skin allows it to have a longer lifetime. Intrinsically stretchable materials like elastomers hence proffer the advantage of stretchability, high sensitivity for large-scale deformation, and low-cost processibility. Commercial elastomers used in this segment of application as stretchable insulators are PDMS,[225] PU,[226] and SEBS[227] with low dielectric constant ($k < 3$) consequently operating in high voltages.[228] As conductors with stretchability, elastomeric matrices have been used with functionalized fillers like LMs[229] or metal flakes.[230] LMs ensure no compromise to stretchability and electron mobility of the system as they are metals

Table 1. Summary of all key applications with their effective strategies of fabrication and challenges.

Targeted application	Key properties	Material selection criteria	Functionalization strategies	Different preparation methods	Challenges	References
Soft actuators	Electroresponsiveness with mechanical stability	Optimum Young's Modulus and Elongation at break with minimal hysteresis and quick response time, e.g., shape memory PU (SMPU)	Polymer fluids, shape memory elastomers, liquid metals, composites of ionic polymer and metals, liquid crystalline elastomers	Solution casting, in situ polymerization, 3D and 4D printing, machine learning (ML)-assisted additive manufacturing	Uniform stress distribution and mechanical stability with quick electroresponsiveness with equivalent recovery time in each cycle	[286–288]
Wearable electronics	Electrical conductivity, high flexibility, thermal stability, self-healing capability	Stable and durable at repeated stretch cycles with biocompatibility, e.g., elastomeric ionogels	Ionic elastomer composites, thermoresponsive elastomers, self-healing (e.g., disulfide-based) elastomers, nanocomposites, surface modifications	Thermal, free radical, ring opening polymerization, click chemistry, 3D printing, solution casting, electrospinning	Durability under different stretch cycles with uniform distribution of functional fillers	[261,289,290]
Sensors	High sensitivity and low noise with stimuli-responsive behavior, biocompatibility for biological sensors	Sustained equivalent outputs over operating ranges, e.g., styrene isoprene propylene styrene photo-cross-link-able systems	Stimuli-responsive elastomers, functional cross-links, nanoparticle integration, and surface modification	Electrospinning, printing, casting, UV cross-linking	Equivalent sensitivity at all environmental conditions; less modulus mismatch issues	[291–293]
Energy-harvesting devices	Stretchability with high dielectric or piezoelectric properties	High dielectric constant, low dielectric loss, high mechanical stability, e.g., electro-active elastomers	Incorporation of high dielectric constant fillers with self-healing linkages into a compliant electrode	Solution casting, molding, coaxial wet spinning, electrospinning, 3D and 4D printing	Achieving high energy conversion efficiency with durability at repeated cycles	[294–296]
Soft robotics	Compliance with human–machine interaction ability, communicability, piezoresponsive ability	Uniform stress distribution, functionality equivalent to biological organisms	Liquid metal–embedded elastomers, polymeric gels, electroactivity incorporation	Additive manufacturing, molding techniques	Multifunctionality incorporation as of biological organisms	[297,298]
Optical electronics	Optical transparency, high signal to noise ratio, electrical or optical stimuli-responsiveness	Highly flexible and transparent substrate with energy collecting efficiency, e.g., functionalized PDMS	Copolymerization and functionalization—physical and chemical with optical fillers with required refractive index	Solution casting and additive manufacturing	Optical clarity and stimuli-responsive activity with fast response time and energy efficiency	[299,300]

and metallic alloys like gallium,[231] eutectic gallium–indium,[232] gallium–indium–tin alloy,[233] and others having melting temperatures near about RT. The incorporation of LM mitigates the modulus gap between the matrix and filler phases as commonly experienced in the case of rigid filler-integrated composites. Furthermore, in addition to other advantageous attributes, the LM-based systems favor the mimicking of the human skin as the resulting materials bear an incredibly low reaction time of ≈60 ms and even lower.[234–236] Active layers of elastomeric components with semiconducting polymers like polythiophenes have allowed increased strain at break with efficient charge transport.[237] Recent developments in the novel electronic skin, apart from the sensing ability through its individuality and artificial intelligence, have been in greater importance due to its everlasting permanence. The self-healing attribute through healing agents or dynamic reversible bonds in elastomers. Supramolecular zwitterionic-network-assisted self-healing ionic elastomers serve the purpose of adaptable multifunctional electronic skin for biomedical applications.[238] Kim et al.[239] reported the recent day pursuit of multi-stimuli-sensitive prosthetic skin equipped with temperature sensing ability for utilization in pioneering flexible electronics.[239]

6.3. Smart and Flexible Displays

Human–machine interactions have been emerging through the most generally used smart and flexible displays enabling a technological boom with industries coming into action very rapidly with the ever-developing notions in this field schematically represented in Figure 12c(i),(ii).[240] Highly stretchable organic light-emitting diodes with retained electroluminescent property have been a challenge to develop; however, feasible options have been proposed by the utilization of intrinsically stretchable electroluminescent elastomers like SBS block copolymer and pyrene units.[241] Multilayered dielectric elastomeric actuators have been proposed for wearable information transferring flexible haptic devices going in hand with the vastly developing enriched mobile computing industry.[242] Zhao et al.[224] studied the multifunctional lanthanide metallopolymer showing significant photo-responsiveness through the use of cross-linked functionalized PU with lanthanide coordinate and hydrogen bonds. The material also showed 100% self-healing efficiency within 24 h through the study of its mechanical response. The functionalized moieties allowed dual-emitting performances with multiple luminescence control switches. The cyclic photostability,

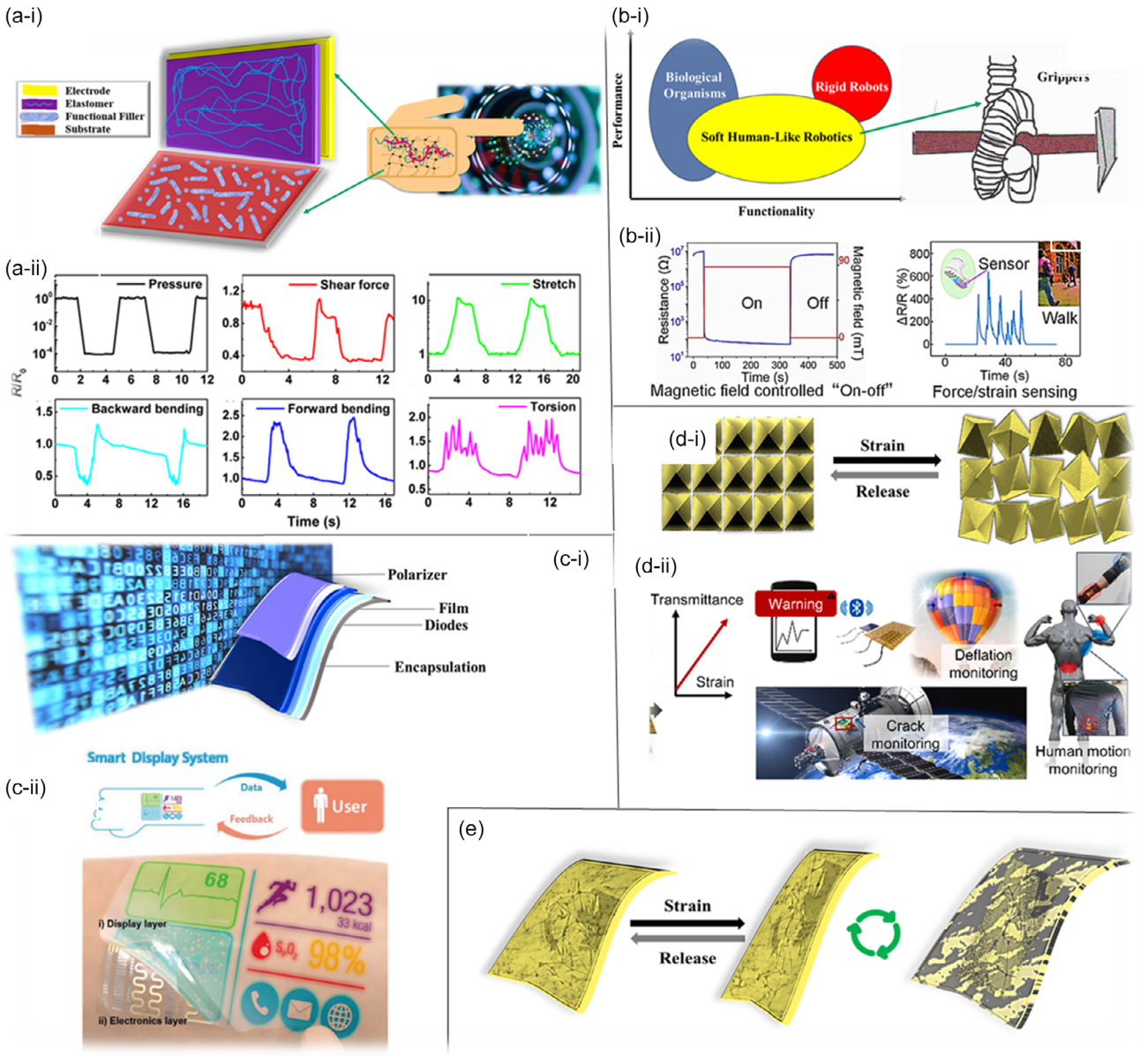


Figure 12. a-i) Schematic diagram of electronic skin; a-ii) pressure and force sensing ability of e-skin (Adapted with permission.[236] Copyright 2014, American Chemical Society); b-i) definition of soft robotics; (b-ii) sensor-based human–machine interaction with magnetic on–off switch (Adapted with permission.[223] Copyright 2021, Elsevier); c-i) schematic diagram of soft and wearable displays; (c-ii) application of soft and wearable displays in daily life (Adapted with permission.[240] Copyright 2018, Elsevier); d-i) schematic to understand metamaterial design; (d-ii) applications where metamaterial allowing higher transmission on strain is useful (Adapted with permission.[248] Copyright 2021, Elsevier); and e) schematic of biodegradable flexible electronics.

when studied with its robustness proven in a scalable upper limit, can be integrated into futuristic adaptive stretchable optoelectronic displays. Exploring through the world of adaptive displays, pioneering work has been done in establishing a smart elastomeric material as a dynamic encryption device. Feng et al.[9] studied such a bilayer material displaying time-resolved information by varying the rheology of the layers through distinct optical stages. The change in the viscoelasticity caused a particular change in the elastic modulus upon programming UV due to different wrinkling wavelengths through interaction networks, finally aiming for advanced dynamic encryption in real-world security applications.[9] Although, th precise control over the optical transitions across wide ranges still remains a challenge, yet useful in adaptive dynamic smart displays.

6.4. Metamaterials

Metamaterials are the newer class of materials having supernatural properties hence unfound in the naturally occurring substances. These metamaterials can exhibit myriad attributes for

necessary functional applications in the revolutionary field of flexible electronics. Flexible metamaterial electronics is a groundbreaking burgeoning research dimension to reach the inaccessible and unconventional.[195,222,243,244] Metamaterials possess attributes like tunable Poisson's ratio inducing biaxial stretchability for auxetic metamaterial formation schematically represented in Figure 12d(i).[245] Auxetic materials possess a negative Poisson's ratio, i.e., a surprising property of increasing lateral dimension under longitudinal stretch. CNT in auxetic PDMS has been reported as an auxetic metamaterial being applicable in adaptive imagers, soft crawlers, etc. It is a brand-new research dimension that enables devising and revising of electromagnetic and elastic waves with programmable morphability.[246] Flexible reconfigurable electronics that are instrumental in implantable applications are possible through higher adaptability and compliance.[247] Photonic sensors devise electromagnetic waves for multifunctional flexible smart skin which has been in high demand since decades.[248] It allows invisibility cloaking for stealth aircraft technology.[249] Harne et al.[250] articulated a new genre of magnetorheological elastomer having tunable static and dynamic properties as in the elastic metamaterials with mechanisms of internal collapse. Research in this domain is yet at the tip of an iceberg and has a lot to be discovered due to challenges faced due to inefficient knowledge of fabrication techniques for the new day intricate metamaterials. The currently used methods introduce 3D/4D printing. Kim et al.[251] proposed a wireless auxetic three-layered *meta*-material with PDMS as the elastomeric material. The conductive layers into the elastomeric material were introduced through 3D printing of microfluidic channels of LM. The 4D printing opens a new realm of 3D printing with smart materials for customized research mainly in the domains of soft electronics. With the advent of these new fabrication techniques for intricate smart materials, a lot of spectral attributes can be invoked in the matrices of flexible materials with toughness and compliance, like elastomers. The 4D printing, being time-responsive, has propelled into the dynamics of soft electronics enabling shape modeling along with the unlocking of smart features. Stimuli-responsive shape memory polymers or shape-morphable-layered multifunctional multiparametric substrates can be fabricated using the aforementioned technique. Encoding of multiple parameters in 4D printing allows to program these shape-morphing behaviors in liquid-crystal elastomers. Intricate metamaterials still remain a challenge and multiple simulations have to be performed to appropriately predict the human–machine interaction. These advantages in the automation of digital technologies contradict the global sustainability goal. Thus, expanding the range of intelligent applications, biodegradability has been introduced in the soft robotic domain for the direct manufacturing and scalability through 3D- and 4D-printing technologies.

6.5. Biodegradable Soft Robotics—A Circular Economy

The circular economy proposed and suggested by various technological environmentalists is to build biodegradable soft robotics fabricated by sustainable additive manufacturing methods schematically represented in Figure 12e. Agriculture,[252] biomedicine,[252–254] sustainable electronics,[254] and microlocomotive robots[255] have been a few of the sectors where the usefulness of these 3D/4D-printed biodegradable soft actuators and sensors come into play. The state-of-the-art material used for the fabrication of soft sustainable electronics is a polyethylene glycol–acrylic acid–based elastomer for pneumatic gripping.[256] Poly(lactide-*co*-caprolactone) has also been a super elastic biodegradable elastomer to be readily fabricated for substrates in deformable electronics by Jiang et al.[257] However, being in the state of infancy, this field has little commercial fabrication so far and requires significant development to rise from conceptualism to reality. This field requires additional research to fill the void and prevent detrimental environmental impacts. A particularly distinct challenge in the area of biomedical soft electronics substrate and tissue engineering is its biocompatibility at the interface. Modulus mismatch, phase separation due to the absence of any chemical interaction at the interface of the biomedical electronic substrate with the living matter hinder their application. This issue is somewhat prevented using elastomer-hydrogel systems[258] or termed as bio-elastomers like PUs and thermoset elastomers like polyesters forming an interpenetrating network with the hydrogel system. In-depth research in the advances of these interactions of bio-inspired elastomer hydrogel systems can produce novel classes of materials even on a commercial level with high bio-functionality and easy processibility.

7. Summary and Future Outlook

The current review describes the use of elastomeric materials in the field of soft electronics and critically analyses the relevant criteria for the suitability of an elastomer material as a flexible substrate through th fundamental sensing mechanisms. The compliant matrix with dynamic reversibility would allow it to solve the problems faced by conventional rigid electronic substrates. However, the disadvantages associated with the use of different classes of elastomers, as a substrate material, due to modulus mismatch, lack of functionality, phase separation, nonpolarity, and difficulty in dynamic reproducibility have been discussed in detail. The introduction of adequate multifunctional properties into an elastomeric material (e.g., through functional fillers) was discussed stating suitable instances. The existing strategies for devising and fabricating flexible electronic substrates out of elastomers have been categorically analyzed for providing an extrapolative view toward the future development. Plausible and credible areas of applicability based on the traits of elastomeric materials are established highlighting the applications of the elastomeric soft electronics in electronic skins, sensors, energy-harvesting devices, health monitoring, electronic displays, soft robotics, and metamaterials. The most fascinating advantage of the elastomers is owed to the natural integration ability of the same to the soft biological systems like organs and tissues in a seamless manner.

The construction of soft and stretchable electronics from elastomers is a newer approach and is in the recent-day researches requiring a stringent technical pathway due to certain attributes. The applications that have been discussed and conceived with high functionality in the rubbery electronics are yet a challenge. The mobility of the carriers as well as the conductivity in the elastomeric electronics are significantly low with large device size and lower integration density. Collectively, there remains a low performance to achieve the most important functions of

electronics like switching and amplifying. Emphasizing on these challenges and developments reveals a lot of scope to refill the lacuna in the domain of soft electronics for the future of soft matter engineering. Despite the fact that soft electronics are growingly overcoming the drawbacks of its conventional rigid coordinate, commercialization and market adoption have been limited. To speed up and urge on its development, distinguishing the bottlenecks that hinder the progression of the same is crucial through propositions of potential solutions. For example, the designing and fabrication for the high-performance soft electronics in accordance to its structure–property relationship with detailed theoretical and practical evidences are highly needed. In addition to the material's perspective, the assembly of the device components with packaging technologies, in a scalable manner, is appreciably required for its broadness in applications. Hence, greater cognizance on the ingenuity of elastomeric materials has to be infused to the associated academic and industrial community for the future technological progress in the application of soft electronics.

Acknowledgements

S.C. and P.S.B. contributed equally to this work. G.H. and S.W. acknowledge the DFG (Deutsche Forschungsgemeinschaft) research training group "Interactive Fiber Rubber Composites" Project 380321452/GRK2430. P.S.B acknowledges the Prime Minister's Research Fellowship (PMRF) scheme.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Soumili Chakraborti: conceptualization (lead); data curation (lead); formal analysis (lead); investigation (lead); visualization (lead); writing—original draft (lead). **Pratip Sankar Banerjee**: conceptualization (equal); formal analysis (supporting); investigation (supporting); validation (supporting); visualization (supporting); writing—original draft (supporting); writing—review & editing (lead). **Debdipta Basu**: conceptualization (supporting); validation (supporting); visualization (supporting). **Sven Wießner**: conceptualization (supporting); validation (supporting); visualization (supporting). **Gert Heinrich**: conceptualization (supporting); formal analysis (supporting); investigation (supporting); supervision (supporting); validation (equal); visualization (supporting); writing—review & editing (supporting). **Amit Das**: conceptualization (supporting); supervision (supporting); validation (supporting); visualization (supporting); writing—review & editing (supporting). **Shib Shankar Banerjee**: conceptualization (supporting); supervision (lead); validation (equal); visualization (supporting); writing—review & editing (supporting).

Keywords

dynamic performances, elastomers, fabrication–integration technologies, multifunctional properties, soft electronics

Received: October 22, 2024
Revised: February 28, 2025
Published online:

[1] S. S. Banerjee, S. Mandal, I. Arief, R. K. Layek, A. K. Ghosh, K. Yang, J. Kumar, P. Formanek, A. Fery, G. Heinrich, A. Das, *ACS Appl. Mater. Interfaces* **2021**, *13*, 15610.
[2] M. D. Bartlett, E. J. Markvicka, C. Majidi, *Adv. Funct. Mater.* **2016**, *26*, 8496.
[3] P. S. Banerjee, D. K. Rana, S. S. Banerjee, *Adv. Colloid Interface Sci.* **2022**, *308*, 102752.
[4] K.-J. Cho, J.-S. Koh, S. Kim, W.-S. Chu, Y. Hong, S.-H. Ahn, *Int. J. Precis. Eng. Manuf.* **2009**, *10*, 171.
[5] Q. Shi, T. He, C. Lee, *Nano Energy* **2019**, *57*, 851.
[6] S. S. Banerjee, I. Arief, R. Berthold, M. Wiese, M. Bartholdt, D. Ganguli, S. Mitra, S. Mandal, J. Wallaschek, A. Raatz, G. Heinrich, A. Das, *Appl. Mater. Today* **2021**, *25*, 101219.
[7] M. D. Bartlett, A. Fassler, N. Kazem, E. J. Markvicka, P. Mandal, C. Majidi, *Adv. Mater.* **2016**, *28*, 3726.
[8] Z. Zhou, H. Zhang, J. Liu, W. Huang, *Giant* **2021**, *6*, 100051.
[9] D. Feng, Q. Guo, Z. Huang, B. Zhou, L. Gong, S. Lu, Y. Yang, D. Yu, Z. Zheng, X. Chen, *Adv. Mater.* **2024**, *36*, 2314201.
[10] K. Lv, G. Tian, Y. Yan, H. Zhou, Q. Fan, L. Liang, N. Liu, D. Wang, Z. Song, F. Xu, G. Zhao, *Chem. Eng. J.* **2024**, *500*, 157534.
[11] R. Fang, B. Yao, T. Chen, X. Xu, D. Xue, W. Hong, H. Wang, Q. Wang, S. Zhang, *Adv. Funct. Mater.* **2024**, *34*, 2310225.
[12] J. H. Lee, K. Cho, J. K. Kim, *Adv. Mater.* **2024**, *36*, 2310505.
[13] C. Majidi, *Adv. Mater. Technol.* **2019**, *4*, 1800477.
[14] Z. Kang, Y. Zhang, M. Zhou, *Chem. Eng. J.* **2019**, *368*, 223.
[15] G. C. Righini, J. Krzak, A. Lukowiak, G. Macrelli, S. Varas, M. Ferrari, *Opt. Mater.* **2021**, *115*, 111011.
[16] C. Wang, Y. Jiang, Q. Ji, Y. Xing, X. Ma, *J. Clean. Prod.* **2024**, *435*, 140503.
[17] W. A. MacDonald, *Large Area Flex. Electron.* **2015**, 291.
[18] H. Ling, S. Liu, Z. Zheng, F. Yan, *Small Methods* **2018**, *2*, 1800070.
[19] J.-S. Noh, *Polymers* **2016**, *8*, 123.
[20] B. M. Mandal, *Fundamentals of Polymerization*, World Scientific, Singapore **2013**.
[21] D. W. Haines, W. D. Wilson, *J. Mech. Phys. Solids* **1979**, *27*, 345.
[22] N. Bosnjak, M. N. Silberstein, *Science* **2021**, *374*, 150.
[23] N. Kazem, T. Hellebrekers, C. Majidi, *Adv. Mater.* **2017**, *29*, 1605985.
[24] J. Tang, *Flexible, Wearable, and Stretchable Electronics*, CRC Press, Boca Raton, FL **2020**, pp. 137–156.
[25] R. You, Y. Liu, Y. Hao, D. Han, Y. Zhang, Z. You, *Adv. Mater.* **2020**, *32*, 1901981.
[26] S. Naghdi, K. Y. Rhee, D. Hui, S. J. Park, *Coatings* **2018**, *8*, 278.
[27] R. A. Golda, A. Marikani, E. J. Alex, *J. Electron. Mater.* **2021**, *50*, 3652.
[28] L. Chen, L. Lu, D. Wu, G. Chen, *Polym. Compos.* **2007**, *28*, 493.
[29] E. Carone, L. D'ilario, A. Martinelli, *J. Appl. Polym. Sci.* **2002**, *83*, 857.
[30] G. Abbati, E. Carone, L. D'ilario, A. Martinelli, *J. Appl. Polym. Sci.* **2003**, *89*, 2516.
[31] U. Lang, N. Naujoks, J. Dual, *Synth. Met.* **2009**, *159*, 473.
[32] T. Hansen, K. West, O. Hassager, N. Larsen, *Adv. Funct. Mater.* **2007**, *17*, 3069.
[33] D. S. Vicentini, G. M. O. Barra, J. R. Bertolino, A. T. N. Pires, *Eur. Polym. J.* **2007**, *43*, 4565.
[34] H. Yoshikawa, T. Hino, N. Kuramoto, *Synth. Met.* **2006**, *156*, 1187.
[35] K. K. Sadasivuni, D. Ponnamma, S. Thomas, Y. Grohens, *Prog. Polym. Sci.* **2014**, *39*, 749.
[36] Y. Dong, X. Zhang, Y. Chen, J. Yu, X. Li, B. Ding, *Adv. Funct. Mater.* **2024**, *34*, 2402252.
[37] H. Wang, Y. Yin, Z. Su, C. Chen, L. Zhang, C. Wang, W. Yang, Y. Huang, P. Xu, P. Ma, T. Liu, P. Ma, *Adv. Funct. Mater.* **2024**, *34*, 2311649.
[38] R. O. Ebewele, *Polymer Science and Technology*, CRC Press, Boca Raton, FL **2000**.

[39] G. Odian, *Principles of Polymerization*, John Wiley & Sons, Hoboken, NJ **2004**.
[40] M. A. Akiba, A. Hashim, *Prog. Polym. Sci.* **1997**, *22*, 475.
[41] I. J. Rao, K. R. Rajagopal, *Int. J. Solids Struct.* **2001**, *38*, 1149.
[42] R. A. Shanks, I. Kong, *Advances in Elastomers I: Blends and Interpenetrating Networks*, Springer, Berlin **2013**, pp. 11–45.
[43] A. S. Deuri, A. K. Bhowmick, *J. Appl. Polym. Sci.* **1987**, *34*, 2205.
[44] Y. Liang, W. Cao, Z. Li, Y. Wang, Y. Wu, L. Zhang, *Polym. Test.* **2008**, *27*, 270.
[45] S. C. Shit, P. Shah, *Natl. Acad. Sci. Lett.* **2013**, *36*, 355.
[46] M. Morton, *Rubber Technology*, Springer Science & Business Media, Berlin **2013**.
[47] S. Kohjiya, *Natural Rubber*, Smithers Rapra, Shawbury, Shrewsbury, Shropshire, UK **2015**.
[48] S. Kohjiya, Y. Ikeda, *Chemistry, Manufacture and Applications of Natural Rubber*, Woodhead Publishing, Darya Ganj, New Delhi, India **2021**.
[49] D. Ponnamma, K. K. Sadasivuni, C. Wan, S. Thomas, M. Al-Ali AlMa'adeed, *Flexible and Stretchable Electronic Composites*, Springer, Berlin **2016**, pp. 1–35.
[50] W. Kaewsakul, K. Sahakaro, W. K. Dierkes, J. W. M. Noordermeer, *Rubber Chem. Technol.* **2013**, *86*, 313.
[51] Y. Hirata, H. Kondo, Y. Ozawa, *Chemistry, Manufacture and Applications of Natural Rubber*, Elsevier, Amsterdam **2014**, pp. 325–352.
[52] K. H. Rahiman, G. Unnikrishnan, A. Sujith, C. K. Radhakrishnan, *Mater. Lett.* **2005**, *59*, 633.
[53] P. T. Hao, H. Ismail, A. S. Hashim, *Polym. Test.* **2001**, *20*, 539.
[54] A. Zhang, Y. Zhang, Y. Zhang, *Polym. Test.* **2020**, *89*, 106600.
[55] A. F. Halasa, J. Prentis, B. Hsu, C. Jasiunas, *Polymer* **2005**, *46*, 4166.
[56] T. Yuasa, T. Tominaga, T. Sone, *Nippon Gomu Kyokaishi* **2013**, *86*, 249.
[57] P. Sae-Oui, K. Suchiva, U. Thepsuwan, W. Intiya, P. Yodjun, C. Sirisinha, *Rubber Chem. Technol.* **2016**, *89*, 240.
[58] A. Yoshioka, K. Komuro, A. Ueda, H. Watanabe, S. Akita, T. Masuda, A. Nakajima, *Pure Appl. Chem.* **1986**, *58*, 1697.
[59] R. J. Dhanorkar, S. Mohanty, V. K. Gupta, *Ind. Eng. Chem. Res.* **2021**, *60*, 4517.
[60] P. J. Flory, *Rubber Chem. Technol.* **1946**, *19*, 552.
[61] R. N. Webb, T. D. Shaffer, A. H. Tsou, Butyl rubber. Encyclopedia of Polymer Science and Technology, **2002**.
[62] E. Kresge, H. C. Wang, Butyl rubber. Kirk-Othmer Encyclopedia of Chemical Technology, **2000**.
[63] Z. S. Petrović, J. Ferguson, *Prog. Polym. Sci.* **1991**, *16*, 695.
[64] C. Hepburn, *Polyurethane Elastomers*, Springer Science & Business Media, Berlin **2012**.
[65] C. Prisacariu, *Polyurethane Elastomers: From Morphology to Mechanical Aspects*, Springer Science & Business Media, Berlin **2011**.
[66] M. Szycher, *J. Biomater. Appl.* **1988**, *3*, 297.
[67] K. E. Polmanteer, *Rubber Chem. Technol.* **1988**, *61*, 470.
[68] K. E. Polmanteer, *Rubber Chem. Technol.* **1981**, *54*, 1051.
[69] S.-H. Kim, E. A. Cherney, R. Hackam, *IEEE Trans. Electr. Insul.* **1992**, *27*, 610.
[70] P. A. L. S. Martins, R. M. Natal Jorge, A. J. M. Ferreira, *Strain* **2006**, *42*, 135.
[71] W. S. Wong, A. Salleo, *Flexible Electronics: Materials and Applications*, Vol. *11*, Springer Science & Business Media, Berlin **2009**.
[72] N. Adly, S. Weidlich, S. Seyock, F. Brings, A. Yakushenko, A. Offenhäusser, B. Wolfrum, *npj Flex. Electron.* **2018**, *2*, 15.
[73] S. Park, H. Park, S. Seong, Y. Chung, *Sci. Rep.* **2020**, *10*, 7660.
[74] Y. Jeon, J. Choi, D. Seo, S. H. Jung, J. Lim, *Polym. Chem.* **2023**, *14*, 1117.
[75] Q. Zhang, *Front. Phys.* **2021**, *16*, 1.
[76] S. M. Venugopal, D. R. Allee, M. Quevedo-Lopez, B. Gnade, E. Forsythe, D. Morton, in *2010 IEEE Inter. Reliability Physics Symp.*, IEEE, Piscataway, NJ **2010**.
[77] I.-C. Cheng, S. Wagner, *Flexible Electronics: Materials and Applications*, Springer, Berlin **2009**, pp. 1–28.
[78] T. Chen, D. Wuu, C. Wu, C. Chiang, Y. Chen, R. Horng, *Plasma Process. Polym.* **2007**, *4*, 180.
[79] K. L. Jarvis, P. J. Evans, N. A. Cooling, B. Vaughan, J. Habsuda, W. J. Belcher, C. Bilen, G. Griffiths, P. C. Dastoor, G. Triani, *Surfaces Interfaces* **2017**, *9*, 182.
[80] S. J. Kang, C. Liu, Y. Y. Noh, *Large Area and Flexible Electronics*, Wiley, Hoboken, NJ **2015**, pp. 275–290.
[81] M. Wagih, Y. Wei, S. Beeby, *IEEE Antennas Wirel. Propag. Lett.* **2019**, *18*, 49.
[82] A. Russo, B. Y. Ahn, J. J. Adams, E. B. Duoss, J. T. Bernhard, J. A. Lewis, *Adv. Mater.* **2011**, *23*, 3426.
[83] W. Zhou, Y. Cheng, K. Chen, G. Xie, T. Wang, G. Zhang, *Adv. Funct. Mater.* **2020**, *30*, 1903829.
[84] A. Krainoi, K. Boonkerd, *Mater. Today Sustain.* **2024**, *26*, 100698.
[85] Z. Liu, Z. Qian, J. Song, Y. Zhang, *Carbon* **2019**, *149*, 181.
[86] Z. Chen, Z. Yang, T. Yu, Z. Wei, C. Ji, B. Zhao, T. Yu, W. Yang, Y. Li, *Compos. Sci. Technol.* **2023**, *232*, 109881.
[87] P. S. Banerjee, S. S. Banerjee, *J. Mater. Sci.* **2023**, *58*, 14009.
[88] Y. Wang, C. Xu, X. Yu, H. Zhang, M. Han, *Mater. Today Phys.* **2022**, *23*, 100647.
[89] M. Amjadi, K. Kyung, I. Park, M. Sitti, *Adv. Funct. Mater.* **2016**, *26*, 1678.
[90] L. Duan, D. R. D'hooge, L. Cardon, *Prog. Mater. Sci.* **2020**, *114*, 100617.
[91] D. Kang, P. V. Pikhitsa, Y. W. Choi, C. Lee, S. S. Shin, L. Piao, B. Park, K.-Y. Suh, T. Kim, M. Choi, *Nature* **2014**, *516*, 222.
[92] N. Hu, Y. Karube, M. Arai, T. Watanabe, C. Yan, Y. Li, Y. Liu, H. Fukunaga, *Carbon* **2010**, *48*, 680.
[93] F. S. Irani, A. H. Shafaghi, M. C. Tasdelen, T. Delipinar, C. E. Kaya, G. G. Yapici, M. K. Yapici, *Micromachines* **2022**, *13*, 119.
[94] S. Yong, K. Aw, *Sens. Imaging* **2022**, *23*, 22.
[95] J. Narongthong, A. Das, H. H. Le, S. Wießner, C. Sirisinha, *Composites, Part A* **2018**, *113*, 330.
[96] D. Niu, W. Jiang, G. Ye, K. Wang, L. Yin, Y. Shi, B. Chen, F. Luo, H. Liu, *Mater. Res. Bull.* **2018**, *102*, 92.
[97] J. Park, S. Wang, M. Li, C. Ahn, J. K. Hyun, D. S. Kim, D. K. Kim, J. A. Rogers, Y. Huang, S. Jeon, *Nat. Commun.* **2012**, *3*, 916.
[98] T. S. Natarajan, S. B. Eshwaran, K. W. Stöckelhuber, S. Wießner, P. Pötschke, G. Heinrich, A. Das, *ACS Appl. Mater. Interfaces* **2017**, *9*, 4860.
[99] B. C. Sekhar, B. Dhanalakshmi, B. Srinivasa Rao, S. Ramesh, K. Venkata Prasad, P. S. V. Subba Rao, B. Parvatheeswara Rao, *Multifunctional Ferroelectric Materials*, IntechOpen, London, UK **2021**, pp. 71–88.
[100] J. Tichý, J. Erhart, E. Kittinger, J. Přívratská, *Fundamentals of Piezoelectric Sensorics: Mechanical, Dielectric, and Thermodynamical Properties of Piezoelectric Materials*, Springer, Berlin **2010**, pp. 1–14.
[101] A. Kachroudi, S. Basrour, L. Rufer, F. Jomni, *J. Phys.: Conf. Ser.* **2015**, *660*, 012040.
[102] R. J. Crawford, P. J. Martin, *Plastics Engineering*, Butterworth-Heinemann, Oxford, UK **2020**.
[103] A. Kachroudi, S. Basrour, L. Rufer, A. Sylvestre, F. Jomni, *Smart Mater. Struct.* **2015**, *24*, 125013.
[104] J. Shi, D. Zhu, S. Beeby, *J. Phys.: Conf. Ser.* **2014**, *557*, 012104.
[105] J. Shi, Z. Luo, Z. Dibin, S. Beeby, *Smart Mater. Struct.* **2019**, *28*, 075010.

[106] A. Rovisco, A. Dos Santos, T. Cramer, J. Martins, R. Branquinho, H. Águas, B. Fraboni, E. Fortunato, R. Martins, R. Igreja, P. Barquinha, *ACS Appl. Mater. Interfaces* **2020**, *12*, 18421.
[107] K. K. Sadasivuni, D. Ponnamma, J.-J. Cabibihan, M. Al-Ali AlMa'adeed, *Flexible and Stretchable Electronic Composites*, Springer, Berlin **2016**, pp. 199–228.
[108] O. Okhay, A. Tkach, *Nanomaterials* **2023**, *13*, 988.
[109] Y. Qiu, Y. Tian, S. Sun, J. Hu, Y. Wang, Z. Zhang, A. Liu, H. Cheng, W. Gao, W. Zhang, H. Chai, H. Wu, *Nano Energy* **2020**, *78*, 105337.
[110] J. Pan, S. Liu, Y. Yang, J. Lu, *Nanomaterials* **2018**, *8*, 413.
[111] N. Gariya, P. Kumar, B. Prasad, *J. Intell. Robot. Syst.* **2023**, *109*, 19.
[112] C. Liu, Y. Zhuang, A. Nasrollahi, L. Lu, M. F. Haider, F.-K. Chang, *Sensors* **2020**, *20*, 2830.
[113] Y. He, X. Zhang, J. Guan, *J. Energy Eng.* **2016**, *142*, 04016011.
[114] T. Zaharescu, *Mater. Chem. Phys.* **2019**, *234*, 102.
[115] J. E. Jam, M. Ahangari, *Polym.-Plast. Technol. Eng.* **2012**, *51*, 1474.
[116] L. Valentini, S. Bittolo Bon, M. A. Lopez-Manchado, R. Verdejo, L. Pappalardo, A. Bolognini, A. Alvino, S. Borsini, A. Berardo, N. M. Pugno, *Compos. Sci. Technol.* **2016**, *128*, 123.
[117] N. Jha, G. Sarkhel, S. Mahapatra, *Mater. Today: Proc.* **2021**, *45*, 3850.
[118] Y.-H. Chen, M. De Beeck, L. Vanderheyden, E. Carrette, V. Mihajlović, K. Vanstreels, B. Grundlehner, S. Gadeyne, P. Boon, C. Van Hoof, *Sensors* **2014**, *14*, 23758.
[119] H. Banno, *Ferroelectrics* **1983**, *50*, 3.
[120] S. Niu, Z. L. Wang, *Nano Energy* **2015**, *14*, 161.
[121] J. Park, D. Kim, Y. T. Kim, *Nanotechnology* **2021**, *32*, 385403.
[122] I. Arief, P. Zimmermann, S. Hait, H. Park, A. K. Ghosh, A. Janke, S. Chattopadhyay, J. Nagel, G. Heinrich, S. Wießner, A. Das, *Mater. Horiz.* **2022**, *9*, 1468.
[123] T. S. Natarajan, S. Finger, J. Lacayo-Pineda, E. S. Bhagavatheswaran, S. S. Banerjee, G. Heinrich, A. Das, *ACS Appl. Electron. Mater.* **2020**, *2*, 4054.
[124] I. A. M. Ibrahim, A. A. F. Zikry, M. A. Sharaf, J. E. Mark, K. Jacob, I. M. Jasiuk, R. Tannenbaumn, *IOP Conf. Ser.: Mater. Sci. Eng.* **2012**, *40*, 012011.
[125] P. Klonos, I. Y. Sulym, M. V. Borysenko, V. M. Gun'ko, S. Kripotou, A. Kyritsis, P. Pissis, *Polymer* **2015**, *58*, 9.
[126] P. A. Klonos, O. V. Goncharuk, E. M. Pakhlov, D. Sternik, A. Deryło-Marczewska, A. Kyritsis, V. M. Gun'Ko, P. Pissis, *Macromolecules* **2019**, *52*, 2863.
[127] N. González, M. À. Custal, G. N. Tomara, G. C. Psarras, J.-R. Riba, E. Armelin, *Eur. Polym. J.* **2017**, *97*, 57.
[128] S. Salaeh, N. Muensit, P. Bomlai, C. Nakason, *J. Mater. Sci.* **2011**, *46*, 1723.
[129] E. Muhammad Abdul Jamal, P. A. Joy, P. Kurian, M. R. Anantharaman, *Mater. Sci. Eng. B* **2009**, *156*, 24.
[130] A. Rattanapan, *Creat. Sci.* **2018**, *10*, 45.
[131] A. S. Sethulekshmi, A. Saritha, K. Joseph, *Int. J. Biol. Macromol.* **2022**, *194*, 819.
[132] A. Kumar, D. Ahmad, K. Patra, M. Hossain, *J. Appl. Polym. Sci.* **2021**, *138*, 50991.
[133] P. Luangchuang, N. Chueangchayaphan, M. A. Sulaiman, W. Chueangchayaphan, *Polym. Int.* **2021**, *70*, 154.
[134] W. Du, H. Zou, M. Tian, L. Zhang, W. Wang, *Polym. Adv. Technol.* **2012**, *23*, 1029.
[135] S. Kwon, R. Ma, U. Kim, H. R. Choi, S. Baik, *Carbon* **2014**, *68*, 118.
[136] C. M. Ajmal, M. M. Menamparambath, H. R. Choi, S. Baik, *Nanotechnology* **2016**, *27*, 225603.
[137] J. Charles, S. Muthusamy, *J. Appl. Sci. Eng. Methodol.* **2016**, *2*, 206.
[138] M. Sebastian, L. Namitha, *Microwave Materials and Applications 2V Set*, John Wiley & Sons, Ltd, Hoboken, NJ **2017**, pp. 537–574.
[139] M. T. Sebastian, *Dielectric Materials for Wireless Communication*, Elsevier, Amsterdam **2010**.
[140] D. Thomas, C. Janardhanan, M. T. Sebastian, *Int. J. Appl. Ceram. Technol.* **2011**, *8*, 1099.
[141] M. Shiva, E. Esmaeli, M. Zivdar, Effects of Ceramic Fillers on the Mechanical and Thermal Transport Properties of Butyl Rubber in the Presence of Carbon Black **2021**.
[142] M. T. Sebastian, H. Jantunen, *Int. J. Appl. Ceram. Technol.* **2010**, *7*, 415.
[143] C. Janardhanan, D. Thomas, G. Subodh, S. Harshan, J. Philip, M. T. Sebastian, *J. Appl. Polym. Sci.* **2012**, *124*, 3426.
[144] J. Chameswary, M. Sebastian, *J. Mater. Sci.: Mater. Electron.* **2013**, *24*, 4351.
[145] A. A. M. Ward, B. Stoll, W. Von Soden, S. Herminghaus, A. A. Mansour, *Macromol. Mater. Eng.* **2003**, *288*, 971.
[146] J. Chameswary, M. Sebastian, *J. Mater. Sci.: Mater. Electron.* **2015**, *26*, 4629.
[147] M. Ruan, D. Yang, W. Guo, S. Huang, Y. Wu, H. Wang, H. Wang, L. Zhang, *RSC Adv.* **2017**, *7*, 37148.
[148] J. Chameswary, M. T. Sebastian, *Ceram. Int.* **2013**, *39*, 2795.
[149] J. Chameswary, M. T. Sebastian, *Ceram. Int.* **2014**, *40*, 7439.
[150] J. Chameswary, L. K. Namitha, M. Brahmakumar, M. T. Sebastian, *Int. J. Appl. Ceram. Technol.* **2014**, *11*, 919.
[151] N. Joseph, C. Janardhanan, M. T. Sebastian, *Compos. Sci. Technol.* **2014**, *101*, 139.
[152] X. Guo, G. Liu, *Polymers* **2023**, *15*, 2101.
[153] M. P. Wolf, G. B. Salieb-Beugelaar, P. Hunziker, *Prog. Polym. Sci.* **2018**, *83*, 97.
[154] Z. Chen, C. Xu, C. Ma, W. Ren, H. Cheng, *Adv. Mater.* **2013**, *25*, 1296.
[155] A. Shakun, Soft elastomeric material with improved dielectric permittivity, **2014**.
[156] M. Poikelispää, A. Shakun, A. Das, J. Vuorinen, *Polym. Adv. Technol.* **2017**, *28*, 130.
[157] Z. Gao, C. Yiu, Y. Liu, D. Li, L. Mei, Z. Zeng, X. Yu, *J. Mater. Chem. C* **2020**, *8*, 15105.
[158] Y. Xu, J. Liu, *Small* **2016**, *12*, 1400.
[159] I. Martinček, I. Turek, N. Tarjányi, *Opt. Mater. Express* **2014**, *4*, 1997.
[160] H. Cho, D. Lee, S. Hong, H. Kim, K. Jo, C. Kim, I. Yoon, *Nanomaterials* **2022**, *12*, 2328.
[161] S. Lee, H.-J. Shin, S.-M. Yoon, D. K. Yi, J.-Y. Choi, U. Paik, *J. Mater. Chem.* **2008**, *18*, 1751.
[162] A. Hosseinzadeh, C. T. Middlebrook, M. E. Mullins, in *Organic Photonic Materials and Devices XVII*, SPIE, Bellingham, Washington **2015**.
[163] A. Dalod, O. Grendal, A. Blichfeld, V. Furtula, J. Pérez, L. Henriksen, T. Grande, M.-A. Einarsrud, *Nanomaterials* **2017**, *7*, 460.
[164] J. Narongthong, H. H. Le, A. Das, C. Sirisinha, S. Wießner, *Compos. Sci. Technol.* **2019**, *174*, 202.
[165] K. Subramaniam, A. Das, K. W. Stöckelhuber, G. Heinrich, *Rubber Chem. Technol.* **2013**, *86*, 367.
[166] J. Lv, G. Thangavel, Y. Xin, D. Gao, W. C. Poh, S. Chen, P. S. Lee, *Nat. Commun.* **2023**, *14*, 7132.
[167] M. A. Mcclain, I. P. Clements, R. H. Shafer, R. V. Bellamkonda, M. C. Laplaca, M. G. Allen, *Biomed. Microdevices* **2011**, *13*, 361.
[168] T. Cheng, Y. Zhang, W. Lai, W. Huang, *Adv. Mater.* **2015**, *27*, 3349.
[169] S. Huang, Y. Liu, Y. Zhao, Z. Ren, C. F. Guo, *Adv. Funct. Mater.* **2019**, *29*, 1805924.
[170] B. Dong, L. Zhang, Y. Wu, *J. Mater. Sci.* **2016**, *51*, 10561.
[171] S. Naderizadeh, G. Santagiuliana, W. Tu, D. Marsh, E. Bilotti, J. J. C. Busfield, *IEEE Sens. J.* **2023**, *23*, 18013.
[172] Y. Gao, M. Xu, G. Yu, J. Tan, F. Xuan, *Sens. Actuators, A* **2019**, *299*, 111625.
[173] K. K. Meena, I. Arief, A. K. Ghosh, H. Liebscher, S. Hait, J. Nagel, G. Heinrich, A. Fery, A. Das, *Nano Energy* **2023**, *115*, 108707.

[174] B. Subeshan, Y. Baddam, E. Asmatulu, *Prog. Addit. Manuf.* **2021**, *6*, 495.
[175] D. K. Rana, P. S. Banerjee, S. S. Banerjee, *J. Mater. Sci.* **2023**, *58*, 6375.
[176] S. Kim, M. Spenko, S. Trujillo, B. Heyneman, V. Mattoli, M. R. Cutkosky, in *Proc. 2007 IEEE Inter. Conf. on Robotics and Automation*, IEEE, Piscataway, NJ **2007**.
[177] R. J. Wood, S. Avadhanula, R. Sahai, E. Steltz, R. S. Fearing, *J. Mech. Des.* **2008**, *130*, 052304.
[178] S. Y. Chou, P. R. Krauss, W. Zhang, L. Guo, L. Zhuang, *J. Vac. Sci. Technol. B: Microelectron. Nanometer. Struct. Process. Meas. Phenom.* **1997**, *15*, 2897.
[179] G. B. Blanchet, Y.-L. Loo, J. A. Rogers, F. Gao, C. R. Fincher, *Appl. Phys. Lett.* **2003**, *82*, 463.
[180] S. Bo-Sung, C. Won-Suk, W. Kyung-Hyun, *Int. J. Precis. Eng. Manuf.* **2006**, *7*, 56.
[181] G. Fu, N. H. Loh, S. B. Tor, Y. Murakoshi, R. Maeda, *Mater. Des.* **2004**, *25*, 729.
[182] A. M. Dollar, R. D. Howe, *IEEE/ASME Trans. Mechatron.* **2006**, *11*, 154.
[183] K. Elkins, H. Nordby, C. Janak, R. W. Gray IV, J. Helge Bohn, D. G. Baird, in *1997 Inter. Solid Freeform Fabrication Symp.*, The University of Texas, Austin **1997**.
[184] T. N. A. T. Rahim, A. M. Abdullah, H. Md Akil, *Polym. Rev.* **2019**, *59*, 589.
[185] Y. L. Tee, C. Peng, P. Pille, M. Leary, P. Tran, *JOM* **2020**, *72*, 1105.
[186] M. Leester-Schädel, B. Hoxhold, C. Lesche, S. Demming, S. Büttgenbach, *Microsyst. Technol.* **2008**, *14*, 697.
[187] Q. Zou, Z. Ma, S. Li, Z. Lei, Q. Su, *Sens. Actuators, A* **2020**, *308*, 112012.
[188] S. P. Lacour, J. Jones, S. Wagner, S. Teng Li, S. Zhigang Suo, *Proc. IEEE* **2005**, *93*, 1459.
[189] D. Gray, J. Tien, C. Chen, *Adv. Mater.* **2004**, *16*, 393.
[190] S. P. Lacour, S. Wagner, Z. Huang, Z. Suo, *Appl. Phys. Lett.* **2003**, *82*, 2404.
[191] A. Oleff, B. Küster, M. Stonis, L. Overmeyer, *Prog. Addit. Manuf.* **2021**, *6*, 705.
[192] P. M. Bhatt, A. M. Kabir, M. Peralta, H. A. Bruck, S. K. Gupta, *Addit. Manuf.* **2019**, *27*, 278.
[193] Y. L. Yap, C. Wang, S. L. Sing, V. Dikshit, W. Y. Yeong, J. Wei, *Precis. Eng.* **2017**, *50*, 275.
[194] D. Svetlizky, M. Das, B. Zheng, A. L. Vyatskikh, S. Bose, A. Bandyopadhyay, J. M. Schoenung, E. J. Lavernia, N. Eliaz, *Mater. Today* **2021**, *49*, 271.
[195] D. Yang, H. Mei, L. Yao, W. Yang, Y. Yao, L. Cheng, L. Zhang, K. G. Dassios, *J. Mater. Chem. C* **2021**, *9*, 12010.
[196] F. Zhang, L. Wang, Z. Zheng, Y. Liu, J. Leng, *Composites, Part A: Appl. Sci. Manuf.* **2019**, *125*, 105571.
[197] S. K. Georgantzinos, G. I. Giannopoulos, P. A. Bakalis, *J. Compos. Sci.* **2021**, *5*, 119.
[198] A. K. Ghosh, S. Sarkar, T. Tsuda, S. Chae, A. Knapp, M. Nitschke, A. Das, S. Wießner, T. A. F. König, A. Fery, *Adv. Funct. Mater.* **2023**, *33*, 2210172.
[199] J. Shao, X. Chen, X. Li, H. Tian, C. Wang, B. Lu, *Sci. China Technol. Sci.* **2019**, *62*, 175.
[200] T. Higashiki, T. Nakasugi, I. Yoneda, *J. Micro/Nanolithogr. MEMS MOEMS* **2011**, *10*, 043008-043008-7.
[201] J. S. Limkaichong, *Flexible Electronics Encapsulation in Thin Polyester Films*, University of Massachusetts Lowell, Lowell, MA **2020**.
[202] M. S. Cetin, H. A. K. Toprakci, *Composites, Part B* **2021**, *224*, 109199.
[203] K. Yan, J. Li, L. Pan, Y. Shi, *APL Mater.* **2020**, *8*, 120705.
[204] J. Hu, in *2010 28th VLSI Test Symp. (VTS)*, IEEE, Piscataway, NJ **2010**.
[205] M. Kim, B.-U. Moon, C. H. Hidrovo, *J. Micromech. Microeng.* **2013**, *23*, 095024.
[206] N. P. Kim, *Polymers* **2020**, *12*, 1224.
[207] Z. Xu, Y. Song, Q. Zheng, *Polymer* **2019**, *185*, 121953.
[208] P. Awasthi, P. S. Banerjee, V. Hirschberg, S. S. Banerjee, *Prog. Rubber Plast. Recycl. Technol.* **2024**, https://doi.org/10.1177/14777606241270513.
[209] S. Aiswarya, P. Awasthi, P. S. Banerjee, S. S. Banerjee, *Dynamic Mechanical and Creep-Recovery Behavior of Polymer-Based Composites*, Elsevier, Amsterdam **2024**, pp. 113–142.
[210] M. H. Wilhelm, E. Filippidi, J. H. Waite, M. T. Valentine, *Soft Matter* **2017**, *13*, 7381.
[211] F. Lacroix, S. Méo, G. Berton, F. Chalon, A. Tougui, N. Ranganathan, *Constitutive Models for Rubber IV*, Routledge, London **2017**, pp. 77–82.
[212] V. Hirschberg, L. Faust, M. Wilhelm, D. Rodrigue, *Macromol. Mater. Eng.* **2021**, *306*, 2100165.
[213] T. Košir, J. Slavič, *Polymers* **2023**, *15*, 158.
[214] S. Kouchakzadeh, K. Narooei, *Sens. Actuators, A* **2021**, *332*, 113214.
[215] A. Kossa, M. T. Valentine, R. M. McMeeking, *Meccanica* **2023**, *58*, 217.
[216] P. Zhang, M. Morris, D. Doshi, *Rubber Chem. Technol.* **2016**, *89*, 79.
[217] A. P. Gerratt, H. O. Michaud, S. P. Lacour, *Adv. Funct. Mater.* **2015**, *25*, 2287.
[218] M. Li, J. Chen, M. Shi, H. Zhang, P. X. Ma, B. Guo, *Chem. Eng. J.* **2019**, *375*, 121999.
[219] L. Fracczak, M. Nowak, K. Koter, *Sens. Actuators, A* **2021**, *321*, 112578.
[220] M. Fan, S. Li, L. Wu, L. Li, M. Qu, J. Nie, R. Zhang, P. Tang, Y. Bin, *Chem. Eng. J.* **2022**, *433*, 133499.
[221] V. Vishwakarma, C. Waghela, A. Jain, *Microelectron. Eng.* **2015**, *142*, 36.
[222] Y. Jiang, Q. Wang, *Sci. Rep.* **2016**, *6*, 34147.
[223] J. Xu, H. Pang, X. Gong, L. Pei, S. Xuan, *Iscience* **2021**, *24*, 102549.
[224] D. Zhao, L. Guo, Q. Li, C. Yue, B. Han, K. Liu, H. Li, *Adv. Mater.* **2024**, *36*, 2405164.
[225] S. El-Molla, A. Albrecht, E. Cagatay, P. Mittendorfer, G. Cheng, P. Lugli, J. F. Salmerón, A. Rivadeneyra, *J. Sens.* **2016**, *2016*, https://doi.org/10.1155/2016/1736169.
[226] W. B. Ying, Z. Yu, D. H. Kim, K. J. Lee, H. Hu, Y. Liu, Z. Kong, K. Wang, J. Shang, R. Zhang, J. Zhu, R.-W. Li, *ACS Appl. Mater. Interfaces* **2020**, *12*, 11072.
[227] Y. Xu, B. Sun, Y. Ling, Q. Fei, Z. Chen, X. Li, P. Guo, N. Jeon, S. Goswami, Y. Liao, S. Ding, Q. Yu, J. Lin, G. Huang, Z. Yan, *Proc. Natl. Acad. Sci.* **2020**, *117*, 205.
[228] X. Wang, L. Dong, H. Zhang, R. Yu, C. Pan, Z. L. Wang, *Adv. Sci.* **2015**, *2*, 1500169.
[229] S. Xiang, D. Liu, C. Jiang, W. Zhou, D. Ling, W. Zheng, X. Sun, X. Li, Y. Mao, C. Shan, *Adv. Funct. Mater.* **2021**, *31*, 2100940.
[230] J. C. Yang, J. Mun, S. Y. Kwon, S. Park, Z. Bao, S. Park, *Adv. Mater.* **2019**, *31*, 1904765.
[231] Y. Lin, J. Genzer, M. D. Dickey, *Adv. Sci.* **2020**, *7*, 2000192.
[232] Y. Guan, Y. Liu, Q. Li, Y. Shi, H. Li, J. Guo, G. Zhang, C. Liu, W. Li, G. Liu, Z. Liu, *Compos. Struct.* **2022**, *291*, 115653.
[233] J. Zhang, M. Liu, G. Pearce, Y. Yu, Z. Sha, Y. Zhou, A. C. Y. Yuen, C. Tao, C. Boyer, F. Huang, M. Islam, C. H. Wang, *Compos. Sci. Technol.* **2021**, *201*, 108497.
[234] Y. Wu, Y. Zhou, W. Asghar, Y. Liu, F. Li, D. Sun, C. Hu, Z. Wu, J. Shang, Z. Yu, R.-W. Li, H. Yang, *Adv. Intell. Syst.* **2021**, *3*, 2000235.
[235] Y. Gao, H. Ota, E. W. Schaler, K. Chen, A. Zhao, W. Gao, H. M. Fahad, Y. Leng, A. Zheng, F. Xiong, C. Zhang, L. Tai, P. Zhao, R. S. Fearing, A. Javey, *Adv. Mater.* **2017**, *29*, 1701985.
[236] J. Park, Y. Lee, J. Hong, Y. Lee, M. Ha, Y. Jung, H. Lim, S. Y. Kim, H. Ko, *ACS Nano* **2014**, *8*, 12020.

[237] H. J. Son, S. Jeong, I. Jeong, H.-J. Kim, M. Park, *ACS Appl. Nano Mater.* **2022**, *5*, 13027.
[238] D. Pei, S. Yu, X. Zhang, Y. Chen, M. Li, C. Li, *Chem. Eng. J.* **2022**, *445*, 136741.
[239] Y. Lee, J. Park, A. Choe, S. Cho, J. Kim, H. Ko, *Adv. Funct. Mater.* **2020**, *30*, 1904523.
[240] J. H. Koo, D. C. Kim, H. J. Shim, T. Kim, D. Kim, *Adv. Funct. Mater.* **2018**, *28*, 1801834.
[241] X. Li, L. Yao, W. Song, F. Liu, Q. Wang, J. Chen, Q. Xue, W. Lai, *Angew. Chem.* **2023**, *135*, e202213749.
[242] D.-Y. Lee, S. H. Jeong, A. J. Cohen, D. M. Vogt, M. Kollosche, G. Lansberry, Y. Mengüç, A. Israr, D. R. Clarke, R. J. Wood, *Soft Robot.* **2022**, *9*, 1186.
[243] A. D. Falco, M. Ploschner, T. F. Krauss, *New J. Phys.* **2010**, *12*, 113006.
[244] T. Van Manen, V. M. Dehabadi, M. C. Saldívar, M. J. Mirzaali, A. A. Zadpoor, *Commun. Mater.* **2022**, *3*, 43.
[245] X. Li, W. Peng, W. Wu, J. Xiong, Y. Lu, *Int. J. Extreme Manuf.* **2023**, *5*, 042003.
[246] F. La Malfa, S. Puce, F. Rizzi, M. De Vittorio, *Nanomaterials* **2020**, *10*, 2365.
[247] J. Zhu, M. Dexheimer, H. Cheng, *npj Flex. Electron.* **2017**, *1*, 8.
[248] J. Gu, J. Ahn, J. Jung, S. Cho, J. Choi, Y. Jeong, J. Park, S. Hwang, I. Cho, J. Ko, J.-H. Ha, Z.-J. Zhao, S. Jeon, S. Ryu, J.-H. Jeong, I. Park, *Nano Energy* **2021**, *89*, 106447.
[249] G. Beziuk, A. Krajewski, T. C. Baum, K. J. Nicholson, K. Ghorbani, *IEEE J. Microwaves* **2024**, *4*, 13.
[250] R. L. Harne, Z. Deng, M. J. Dapino, *J. Intell. Mater. Syst. Struct.* **2018**, *29*, 265.
[251] C.-H. Kim, H.-Y. Kim, J.-H. Kim, J. Kim, *Funct. Compos. Struct.* **2023**, *5*, 015006.
[252] E. Navas, R. Fernández, D. Sepúlveda, M. Armada, P. Gonzalez-De-Santos, *Sensors* **2021**, *21*, 2689.
[253] T. Ashuri, A. Armani, R. Jalilzadeh Hamidi, T. Reasnor, S. Ahmadi, K. Iqbal, *Biomed. Eng. Lett.* **2020**, *10*, 369.
[254] F. Hartmann, M. Baumgartner, M. Kaltenbrunner, *Adv. Mater.* **2021**, *33*, 2004413.
[255] J.-M. Breguet, S. Johansson, W. Driesen, U. Simu, in *Proc. of the 10th Inter. Conf. on New Actuators (Actuator 2006)*, Bremen, Germany, January **2006**.
[256] C. Yoon, *Nano Converg.* **2019**, *6*, 20.
[257] J. H. Lim, W. B. Han, T.-M. Jang, G.-J. Ko, J.-W. Shin, S. Han, H. Kang, C.-H. Eom, S. J. Choi, K. Rajaram, A. J. Bandodkar, W.-H. Yeo, S.-W. Hwang, *Biosens. Bioelectron.* **2024**, *254*, 116222.
[258] A. K. Bastola, N. Rodriguez, M. Behl, P. Soffiatti, N. P. Rowe, A. Lendlein, *Mater. Des.* **2021**, *202*, 109515.
[259] F. D. S. Larotonda, Biodegradable films and coatings obtained from carrageenan from Mastocarpus stellatus and starch from Quercus suber, **2007**.
[260] W. Liu, Y. Duo, J. Liu, F. Yuan, L. Li, L. Li, G. Wang, B. Chen, S. Wang, H. Yang, Y. Liu, Y. Mo, Y. Wang, B. Fang, F. Sun, X. Ding, C. Zhang, L. Wen, *Nat. Commun.* **2022**, *13*, 5030.
[261] T. Delipinar, A. Shafique, M. S. Gohar, M. K. Yapici, *ACS Omega* **2021**, *6*, 8744.
[262] H. Oh, C. Lee, N. Kim, T. An, G. Kim, *Polymers* **2021**, *13*, 2478.
[263] W. Li, D. Torres, T. Wang, C. Wang, N. Sepúlveda, *Nano Energy* **2016**, *30*, 649.
[264] S. G. Kirtania, A. W. Elger, M. R. Hasan, A. Wisniewska, K. Sekhar, T. Karacolak, P. K. Sekhar, *Micromachines* **2020**, *11*, 847.
[265] A. Abdihamzehkolaei, M. T. Ahad, Z. Siddique, *Polymers* **2021**, *13*, 773.
[266] T. Gurunathan, C. R. K. Rao, R. Narayan, K. V. S. N. Raju, *J. Mater. Sci.* **2013**, *48*, 67.
[267] L. V. Kayser, D. J. Lipomi, *Adv. Mater.* **2019**, *31*, 1806133.
[268] J. Li, B. Song, D. An, Y. Ren, J. Hah, X. Wang, R. Zhang, K. Moon, C. Wong, *J. Appl. Polym. Sci.* **2021**, *138*, 50121.
[269] B. Song, J. Li, F. Wu, S. Patel, J. Hah, X. Wang, K. Moon, C.-P. Wong, *Soft Matter* **2018**, *14*, 9036.
[270] B. Song, F. Wu, K.-S. Moon, C. P. Wong, in *2019 IEEE 69th Electronic Components and Technology Conf. (ECTC)*, IEEE, Piscataway, NJ **2019**.
[271] A. Capezza, R. L. Andersson, V. Ström, Q. Wu, B. Sacchi, S. Farris, M. S. Hedenqvist, R. T. Olsson, *ACS Omega* **2019**, *4*, 3458.
[272] C. Johansson, M. Robertsson, *J. Electron. Mater.* **2007**, *36*, 1206.
[273] J. F. Christ, N. Aliheidari, A. Ameli, P. Pötschke, *Mater. Des.* **2017**, *131*, 394.
[274] M. M. R. Nayini, Z. Ranjbar, *Handbook of Carbon Nanotubes*, Springer, Berlin **2022**, pp. 341–392.
[275] S. Priyadarsini, S. Mohanty, S. Mukherjee, S. Basu, M. Mishra, *J. Nanostruct. Chem.* **2018**, *8*, 123.
[276] H. Wang, R. Li, Y. Cao, S. Chen, B. Yuan, X. Zhu, J. Cheng, M. Duan, J. Liu, *Adv. Fiber Mater.* **2022**, *4*, 987.
[277] A. M. Appusamy, E. Prakash, S. Madheswaran, A. Rajamanickam, V. K. Selvakumar, P. Chandrasekar, in *Advances in Materials Research: Select Proceedings of ICAMR 2019*, Springer, Berlin **2021**.
[278] Q. Diao, W. Chen, Y. Wang, Q. Jiang, Z. Li, in *Inter. Conf. on Intelligent Robotics and Applications*, Springer, Berlin **2022**.
[279] A. E. Masoud, J. Maas, *Acta Mech.* **2023**, *234*, 239.
[280] H. Lee, Z. Jiang, T. Yokota, K. Fukuda, S. Park, T. Someya, *Mater. Sci. Eng. R: Rep.* **2021**, *146*, 100631.
[281] M. Waki, S. Chiba, *Soft Actuators: Materials, Modeling, Applications, and Future Perspectives*, Springer, Berlin **2019**, pp. 645–660.
[282] S. A. Chiba, M. Waki, Y. Tanaka, N. Tsurumi, K. Okamoto, K. Nagase, M. Honma, H. Yokota, K. Odagiri, H. Sato, T. Saiki, J. Kaneko, *Adv. Sci. Technol.* **2017**, *97*, 61.
[283] W. Zhan, L. Sun, X. Han, *Nano-Micro Lett.* **2019**, *11*, 1.
[284] S. Himalyan, V. Gupta, *Qlt. Qty.* **2024**, *58*, 3559.
[285] S. Pyo, K. Park, *Int. J. Precis. Eng. Manuf.-Green Technol.* **2024**, *11*, 291.
[286] P. Prathumrat, M. Nikzad, E. Hajizadeh, R. Arablouei, I. Sbarski, *Polym. Adv. Technol.* **2022**, *33*, 1782.
[287] C. Song, Y. Zhang, J. Bao, Z. Wang, L. Zhang, J. Sun, R. Lan, Z. Yu, S. Zhu, H. Yang, *Adv. Funct. Mater.* **2023**, *33*, 2213771.
[288] S. Ma, P. Xue, C. Valenzuela, X. Zhang, Y. Chen, Y. Liu, L. Yang, X. Xu, L. Wang, *Adv. Funct. Mater.* **2024**, *34*, 2309899.
[289] E. J. Barron, E. T. Williams, B. T. Wilcox, D. H. Ho, M. D. Bartlett, *J. Polym. Sci.* **2024**, *62*, 3818.
[290] G. Ge, Y. Zhang, X. Xiao, Y. Gong, C. Liu, C. Lyu, W. L. Ong, G. W. Ho, Z. Yang, W. Huang, *Adv. Funct. Mater.* **2024**, *34*, 2310963.
[291] Y. Yuan, B. Liu, M. R. Adibeig, Q. Xue, C. Qin, Q. Sun, Y. Jin, M. Wang, C. Yang, *Adv. Mater.* **2024**, *36*, 2310429.
[292] H. Park, M. Kim, H. Park, J. H. Oh, *Adv. Funct. Mater.* **2024**, *34*, 2312034.
[293] H. Dai, C. Zhang, C. Pan, H. Hu, K. Ji, H. Sun, C. Lyu, D. Tang, T. Li, J. Fu, P. Zhao, *Adv. Mater.* **2024**, *36*, 2310145.
[294] J. Nirmala Suresh, H. Liebscher, H. Komber, M. Tahir, G. Gerlach, S. Wießner, *ACS Omega* **2024**, *9*, 4754.
[295] Y. Liu, S. Yue, Z. Tian, Z. Zhu, Y. Li, X. Chen, Z. L. Wang, Z. Yu, D. Yang, *Adv. Mater.* **2024**, *36*, 2309893.
[296] C. Zhi, S. Shi, H. Wu, Y. Si, S. Zhang, L. Lei, J. Hu, *Adv. Mater.* **2024**, *36*, 2401264.
[297] S. Chen, S. Fan, H. Chan, Z. Qiao, J. Qi, Z. Wu, J. C. Yeo, C. T. Lim, *Adv. Funct. Mater.* **2024**, *34*, 2309989.
[298] D. R. Yao, I. Kim, S. Yin, W. Gao, *Adv. Mater.* **2024**, *36*, 2308829.
[299] Y. Liu, J. Ma, Y. Yang, C. Valenzuela, X. Zhang, L. Wang, W. Feng, *Smart Mol.* **2024**, *2*, e20230025.
[300] Y. Wan, X. Li, H. Yuan, D. Liu, W. Lai, *Adv. Funct. Mater.* **2024**, *34*, 2316550.

Soumili Chakraborti is a Master of Technology student at the Indian Institute of Technology (IIT) Delhi, Department of Materials Science and Engineering. She received her B.Tech. in polymer science and technology in 2023 from the University College of Science, Technology and Agriculture, University of Calcutta, India. Her research interests include multifunctional polymeric materials for high end engineering applications.

Pratip Sankar Banerjee is a research scholar at the Indian Institute of Technology (IIT) Delhi, Department of Materials Science and Engineering. He received his M.Tech. in materials science and engineering in 2021 from the Indian Institute of Technology (IIT) Kanpur and B.Tech. in polymer science and technology in 2019 from the University College of Science, Technology and Agriculture, University of Calcutta, respectively, India. His research interests include multifunctional materials and elastomeric composites for soft matter engineering.

Debdipta Basu is a senior assistant director and center head for Indian Rubber Materials Research Institute (East), an autonomous institute under Government of India. He carried out his research work at IPF Dresden and received his Ph.D. degree with *Summa cum laude* (distinction) from TUD Dresden University of Technology, Germany, in the area of block copolymer like phase behaviour in ionic elastomer in 2015. He was bestowed with prestigious DKG award in Merseburg, Germany, for "Outstanding Ph.D. thesis in Rubber Technology" in 2016.

Sven Wießner is currently working as Head of Department "Elastomers" at Leibniz Institute of Polymer Research Dresden (IPF Dresden) and is professor of elastomeric materials at the Institute of Materials Science of the Faculty of Mechanical Science and Engineering at the TUD Dresden University of Technology. He graduated in chemical engineering and earned his Ph.D. in polymer engineering from the Chemnitz University of Technology, Germany in 2009. His research interests cover functional elastomers and TPEs, rubber processing, plastics processing, and conventional and reactive polymer mixing processes. He has about 150 publications in international peer-reviewed journals and several contributions in conferences.

Gert Heinrich received Ph.D. and habilitation degree from the University of Technology at Merseburg, Germany, in 1978 and 1985, respectively. From 1990 to 2002, he was a senior research scientist and head of Materials Research at Continental AG, Hannover, Germany. He has been a professor at the Faculty of Mechanical Engineering, TU Dresden, Germany, since 2003, and served as the director of the Institute of Polymer Materials at the Leibniz Institute for Polymer Research Dresden until 2017. He has supervised numerous doctoral candidates and authored over 800 scientific articles, patents, and book chapters. His h-index (Google Scholar) is 84. Gert Heinrich has been honored with various accolades, including the H. F. Mark Medal (Australia, 2024) and the Charles Goodyear Medal (USA, 2025).

Amit Das holds the distinguished position of a senior scientist at the Leibniz Institute of Polymer Research Dresden e.V. (IPF), Germany. Additionally, he serves as an adjunct professor at Tampere University in Finland. He earned his Ph.D. from Jadavpur University in 2004. As the head of the Research Group "Smart and Functional Elastomers," he has made significant contributions to the field of polymer research. He has been honored with various accolades, including the Innovation Prize from IPF, the Spark Thomas Award from the ACS Rubber Division, and the title of Fellow in the Finland Distinguished Professor Program.

Shib Shankar Banerjee is an assistant professor at Indian Institute of Technology (IIT) Delhi, Department of Materials Science and Engineering. Before joining IIT Delhi, he was a research scientist at IPF Dresden, Germany, and postdoctoral research associate at the University of Massachusetts Lowell, USA. He received his Ph.D. in elastomer science and technology from IIT Patna in 2016. He has more than 70 publications in international peer-reviewed journals and about 30 contributions in conferences. His research is focused on elastomers and additive manufacturing of functional elastomeric/polymeric materials.