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Advanced silicone materials for soft actuator applications†

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Soft actuators are at the forefront of the innovation tide in medicine, manufacturing, and aerospace because they are able to mimic the behavior of biological tissue and adapt to complex, unstructured environments. Of all the materials used, silicone-based elastomers have drawn enormous attention since they offer a superb combination of mechanical flexibility, biocompatibility, thermal stability, and long-term durability. In the past few years, there has also been a rapid pace of material evolution, additive manufacturing, and biointegration that has enhanced the performance and applications of silicone-based soft actuators. However, there is no focused and timely review compiling these advances. This review seeks to address that need by critically discussing recent advancements in advanced silicone materials, exploring new fabrication methodologies, and discussing emerging applications that range from wearable devices to implantable robotics. We also present suggestions for directions and the problems which must be addressed in order to further develop the performance and potential of silicone-based soft actuators, justifying the relevance and urgency of this effort.

1. Introduction

Soft actuators represent the next generation of materials that are engineered to mimic the adaptive and dynamic traits of biologically based tissues.[1–19] In comparison to regular rigid actuators, they can perform large, reversible changes in shape, thereby imitating the natural movement of muscles and other soft biological entities.[1,2,4–25] Such a multidirectional ability to change shape upon exposure to different stimuli is very useful in application sectors requiring high compliance and flexibility, *e.g.* those that deal with interaction with humans or fragile environments. Hence, soft actuators find a growing number of applications in areas like soft robotics, wearable systems, biomedical devices, and artificial muscles.[1,6–27]

The design and development of soft actuators are greatly influenced by the responsive mechanical properties in nature.[1–19] A prime example is the simple expansion and contraction of muscles, which generate precise motion with the exertion of controlled force without being damaged. Soft actuators follow the same principles, enabling large strains—usually over 500%—to provide great flexibility and mobility.[1–25,28–36] This inherent dynamism and sensitivity make soft actuators ideal for applications where adaptability, performance, safety, and efficiency are paramount, such as prosthetics, exoskeletons, medical implants, and soft robotics.[14,20,22–35,37–44]

Among the materials that have been explored for use in soft actuators, silicone-based materials have emerged as unparalleled.[45–52] Silicone offers an ideal compromise, having excellent flexibility and high elasticity with requisite durability[53–59] as shown in Fig. 1. Such synergy is necessary to ensure stable performance against dynamic and repetitive movements. With high tensile strength and very long operational durability, silicone-based actuators are durable against continuous deformation and failure, even under harsh environments. Aside from these superior mechanical properties, silicone materials also possess excellent biocompatibility, a feature of utmost importance for biomedical applications, where direct contact with living tissues is made.[57,60–67] This characteristic safety permits silicone-based actuators to be integrated into medical devices, such as implantable

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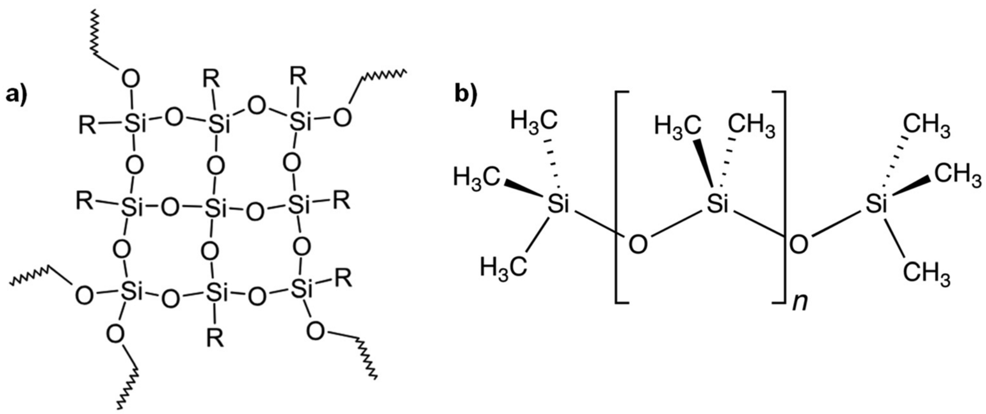

Fig. 1 Chemical structures of silicone materials: (a) silicone resin, (b) siloxane.

sensors, drug delivery systems, and biofeedback devices and systems, where tissue irritation and toxicity are of key concern.[57,60–74]

The second strong advantage of silicones is their ability to retain their mechanical properties under a wide variety of environmental conditions. These materials exhibit excellent thermal stability, and they are flexible and helpful at very low or high temperatures.[53,75–78] Additionally, silicones are unusually chemically stable and resistant to UV light, oxidation, and humidity.[55,76,79] This characteristic robustness in harsh environments expands the application of silicone-based soft actuators into ranges where reliability and durability are vital. These qualities make silicone well suited for continuous unmanned operation under extreme conditions, such as those encountered in space, underwater missions, and extreme industrial processes. They are thus well suited for application in extreme environments, such as those in industrial robotics or aerospace, where actuators need to function optimally with considerable temperature variation and under mechanical stress.

The elasticity and high performance of silicone-based materials qualify them as the most promising candidate for current soft actuator research. Their unusual combination of high elasticity, biocompatibility, mechanical stiffness, and chemical stability make them promising materials for next-generation soft robotics, adaptive devices, and biomedical devices.[57,60–67] As research into silicone-based materials continues to advance, unique applications will undoubtedly rise. These continuing developments are paving the way for the fabrication of smart, flexible, and safe actuators capable of performing complex processes within dynamic and stimulating environments.

To date, several reviews and perspectives have been published that have dealt with the subject of silicone materials or soft actuators in a broad sense.[80] However, it is hard to find a comprehensive and focused review, which not only unifies but also updates the recent developments regarding (i) advances in silicone chemistry, (ii) multifunctional properties, (iii) fabrication strategies, and (iv) their integration in the next-generation soft actuator systems. Here it is aimed at filling such a void by providing a complete material to the reader that starts from the innovations in materials—such as new silicone hybrids and bio-inspired silicones—with the fabrication techniques like 4D printing and laser-based structuring, and the newly surfaced functionalities like self-healing and electroactive responses. Besides, we give comparative insights and the application-specific conversations that existing literature barely touches on. Therefore, our review becomes a timely and instrumental resource to the researchers and developers working on the soft robotics and smart materials domains.

Fig. 2 shows a development roadmap of soft actuators over the past seven decades, highlighting major actuator types, material innovations (particularly silicones and hydrogels), and application domains. The timeline illustrates how soft actuation technology evolved from pneumatic origins to biohybrid and self-healing systems, reflecting the diversification and integration of advanced materials in soft robotics.[6,8]

This review starts with a summary of the essential characteristics and categorization of advanced silicone materials for soft actuators. After that, we explain the innovative manufacturing techniques and material processing strategies that open the way for precise performance control of the actuators. The applications of silicone-based soft actuators in different areas such as medical devices, wearable robotics, and aerospace technologies are discussed in detail too. Finally, we wrap up with a talk about the existing issues and upcoming ideas to help the work of researchers and developers in this fast-moving field to continue.

2. Properties of advanced silicone materials

Advanced silicone materials are emerging as highly promising for use in soft actuator devices, where resilience, flexibility, and adaptability are crucial. These specific materials are engiMicromolding: A Powerful Tool for Large-Scale Production of Precise MicrostructuresMicromolding: A Powerful Tool for Large-Scale Production of Precise MicrostructuresMicromolding: A Powerful Tool for Large-Scale Production of Precise Microstructuresneered to have a unique blend of properties that

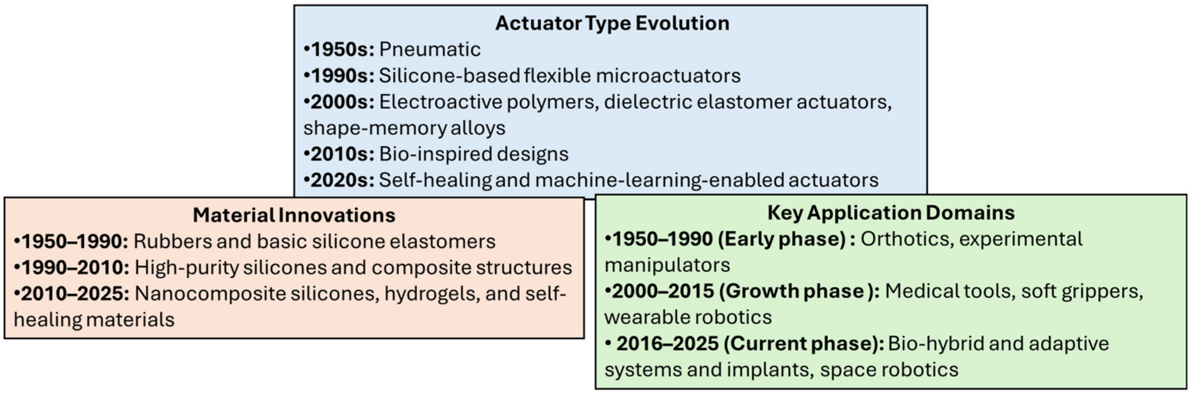

Fig. 2 Development roadmap of soft actuators.

are frequently unachievable with conventional elastomers or polymers.[48,53,59,63,76,81–84] This enhanced performance significantly boosts the capabilities of soft actuators to operate in very specialized applications, ranging from medicine and robotics to numerous other sophisticated fields. The following sections elaborate some of the key properties that characterize these sophisticated silicone materials, and Fig. 3 provides a visual representation of these properties.

2.1. High elasticity

Advanced silicone materials are defined by their high capacity for large elastic deformations without permanent deformation or failure. Most silicone materials possess strain capacities in excess of 500%, a critical property for applications that demand high responsiveness and flexibility.[85–92] Silicone materials get their characteristic elasticity from a unique molecular architecture, which is principally the result of a flexible and highly stable silicon–oxygen (Si–O–Si) backbone. This backbone is significantly different than the carbon–carbon (C–C) chains found in many organic polymers. The Si–O bond is intrinsically longer and holds greater rotational freedom in comparison with a C–C bond. This helps in greater movement and conformational changes along the polymer chain.

For silicone to exhibit rubber-like elasticity, these long polymer chains are naturally cross-linked, creating a 3D network. When exposed to tensile force, the individual coiled silicone chains straighten. However, the presence of these cross-links does not permit the polymer chains to move entirely apart. Once the external force is relaxed, the entropic favoring of the chains to revert to their more disordered coiled conformation drives the material back to its original shape. Cross-link density directly influences the material's stiffness and allows for a tunable range of elastic behavior.

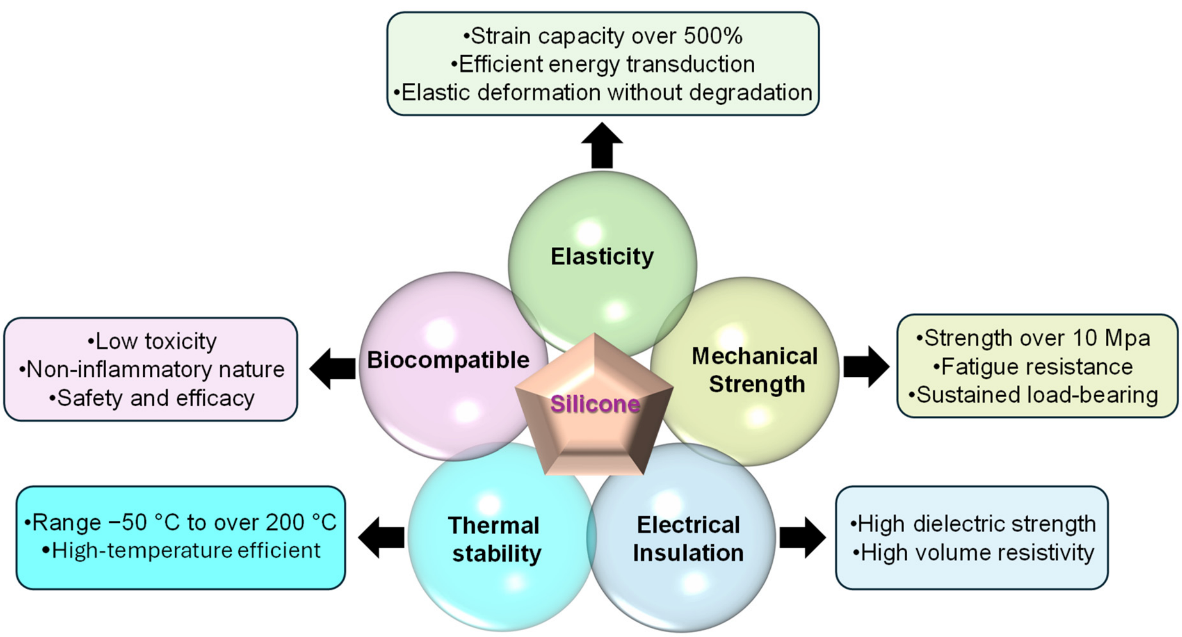

Fig. 3 The image highlights the key properties of advanced silicone materials.

The inherent high elasticity of silicone materials enables efficient energy transduction in soft actuators, thereby enabling complicated, muscle-like movements.[47,59,93–95] This property is particularly beneficial in applications that require reversible soft deformations across a broad spectrum of mechanical states, for instance, in wearable exosuits and soft robotics. This ability to undergo considerable deformation without performance degradation allows these actuators to repeatedly return to their original form, which is necessary for continued movement or actuation in dynamic environments. In soft actuators, this large deformation ability is especially critical for mimicking the function of biological tissues, such as the extensive stretching and contracting of muscles. This property is foundational for developing flexible and efficient systems for repetitive tasks in fields such as manufacturing and medicine. Furthermore, the high strain tolerance of some developed silicone materials reduces the need for complex and costly maintenance or replacement of parts in soft robotic systems.[44,45–48,81,83]

2.2. High strength

Current advances have allowed for the development of highly advanced silicone materials that have high tensile strengths (typically greater than 10 MPa) and yet retain their superior elasticity.[76,85,96–98] Silicone materials exhibit high tensile strengths due to the Si–O backbone's intrinsic stability and resistance to degradation. Intrinsic stability enables the material to maintain its structural integrity well under stress. One of the most important aspects of such mechanical toughness is cross-linking, whereby the extended silicone polymer chains are interconnected to form a highly resistant 3D network. Upon the application of tensile stress, the cross-links dynamically help to prevent total separation of individual chains and thereby prevent the effective stress distribution in the material. Consequently, increased cross-link density considerably increases silicone material's resistance to deformation and cracking under tension. This combination of mechanical strength and high elasticity is important because it permits silicone-based soft actuators to tolerate substantial mechanical stress during operation without the integrity of the material being compromised. For applications in robotics, prosthetics, or wearable devices, soft actuators must be able to withstand loads, manipulate objects, or employ precise mechanical force while simultaneously upholding their ability for elastic deformation. The higher tensile strength of silicone materials makes them particularly well suited for high-stress applications requiring sustained process.[44–48,81–83] For applications such as soft robotics, for instance, actuators often experience repetitive operation with controlled force. The capability of silicones to endure high tensile stress while resisting breakage or fatigue enables continuous functionality, even under constant load-bearing situations. This aspect is essential for applications such as robotic grippers, soft exosuits, and medical devices, which need both robust strength and inherent flexibility from a single material component.

2.3. Biocompatibility

Silicone materials are valued for their excellent biocompatibility, a key characteristic that enables their broad use across the health and medical industries.[57,60–68,70,99–101] Their ability to integrate into biological systems without any damaging effects arises from many key characteristics, one of which is their chemical inertness. Distinct from organic polymers, the silicon-oxygen (Si–O–Si) backbone structure of silicone shows substantial stability, safeguarding against any reactions with tissues or biological fluids. This chemical stability cuts the leaching of harmful substances and the degradation or breakdown of the silicones within the form over lengthy periods, thereby meaningfully reducing inflammatory responses, allergic reactions, or systemic toxicity. The surface characteristics of silicone products also account for their biocompatibility. Their low surface tension and hydrophobic nature can be helpful in many different circumstances because they prevent bacterial colonization and reduce the adhesion of some biological biomolecules, such as proteins. Moreover, surface modification techniques such as plasma treatment, chemical grafting of hydrophilic or hydrophobic groups, and polymer coatings can be used to control the wettability of silicone materials, thereby reducing unwanted biological adhesion in clinical applications.[102] Additionally, medical-grade silicone materials are specifically manufactured to be noncytotoxic and nonimmunogenic, shielding the body from the incitement of immune response, such as inflammation or rejection. This is paramount for long-term implantable devices, where the necessity of constant contact with biological systems requires a material that is neutral to the host body.

Due to their low toxicity and noninflammatory properties, silicone materials have long been a foundational material for medical devices, including catheters, implants, wound dressings, and prosthetics.[57,60–68,70,99–101] This excellent biocompatibility ensures that silicone materials can safely interact with human tissue, making them an extremely good prospect for any device that comes into contact with living organisms. This property is particularly critical for soft actuators, especially in developing implantable devices such as neuroprosthetics, next-generation drug delivery systems, or medical rehabilitation soft actuators. Actuators in all these implantable uses must operate within the body without causing unwanted responses such as inflammation and toxicity. Silicones are exceptionally well suited for wearable or implantable devices that require safe and durable operation because their biocompatibility minimizes the effect of prolonged exposure to body fluids.[44,60–68,70,82,93] They are useful in personal healthcare applications, where soft actuators that are worn on the body may provide vital sign monitoring or therapeutic support.

2.4. Thermal stability

Another notable feature of high-performance silicone materials is their exceptional thermal stability. The mechanical properties of the materials are unexpectedly maintained within an extensive temperature range, usually from as low as

−50 °C to greater than 200 °C, with slight noticeable degradation.[75,103–108] The exceptional thermal stability of silicones is rooted in the robust Si–O backbone that establishes their molecular chain. The Si–O bond holds a very high bond energy (approximately 445–460 kJ mol^{-1}) in comparison with the C–C bond (348–356 kJ mol^{-1}). This substantial bond energy means that a considerable input of thermal energy is necessary to slice the silicone backbone, making it highly resistant against thermal degradation and decomposition, even at high temperatures. Additionally, the flexibility of the Si–O–Si chain plays a vital role in the thermal stability of silicone materials across a considerable temperature range. It makes it possible for the chains of the silicone polymer to retain their elasticity and avoid brittleness even at very low temperatures, as evidenced by the very low glass transition temperatures (T_g) (typically below −100 °C), where other polymers become brittle. Conversely, at high temperatures, the elasticity allows space for the material to expand and contrast with heat without suffering excessive internal stress or cracking and thereby retain its structural integrity.

Furthermore, unlike most organic materials, silicone materials possess extreme resistance to oxidation at high temperatures. The Si–O bond is less susceptible to attack by oxygen in comparison with C–C bonds, which are prone to oxidative degradation at high temperatures. This built-in resistance to oxidation extends the working lifespan of silicone materials in high-temperature, oxygen-enriched conditions. In the same manner, silicone materials exhibit extremely high resistance to UV radiation and ozone degradation, which are known to accelerate degradation of other polymers in outdoor or high-energy environments; this quality is another reason for silicon materials' long-term thermal stability across a broad working environment.

This inherent stability makes silicone materials most suitable for applications that involve repetitive or severe temperature fluctuations (*i.e.*, aerospace, automotive, or industrial robotics).[46–48,81,83,109] The thermal stability of silicon materials is exceptionally high, and soft actuators made of silicon materials can perform well even in extreme high-temperature applications. For instance, in aerospace applications, components are regularly exposed to hot and cold temperatures; the reliability and longevity of silicones exposed to these conditions are therefore invaluable. Similarly, industrial soft actuators undergo significant thermal stress during operation. The ability of silicone materials to remain flexible and intact under extreme temperatures prevents failures and material degradation, thereby enhancing the actuating system's operating life. This resistance against thermal aging enables the fabrication of durable, maintenance-free soft actuators, whether working under fluctuating or constant extreme temperatures and harsh conditions. Therefore, silicone materials are ideal candidates for applications that require high reliability and performance, from day-to-day usage to space deployments for autonomous robotic systems.

2.5. Electrical insulation and dielectric behavior

Advanced high-performance silicone materials are highly regarded for their excellent electrical insulating properties,[56,76,77,90,102,110–113] which contribute significantly to their utility in systems that employ electrical actuation, such as electroactive polymers. The unique molecular structure of silicone materials makes them exceptional electrical insulators. They hold a silicon-oxygen (Si–O–Si) backbone chain, generally decorated with organic side groups (*e.g.*, methyl groups) and covalently bonded to the adjacent silicon atoms. In this molecular structure, all the valence electrons are firmly bound in these covalent bonds, either sandwiched between silicon and oxygen or silicon and the organic groups attached to it. Therefore, there are no free or delocalized electrons available to flow readily through the material and conduct electricity, very much unlike the "sea" of mobile electrons found in highly conductive materials. This electron-deficient framework is responsible for the high dielectric strength of silicone materials. This is the most important property for sound insulation, which allows silicone to withstand high voltages without arc flashes and short circuits. The stable Si–O bonds and the stable molecular network both play a role in this high dielectric strength, and it takes a lot of energy to destabilize the electron distribution in the material.

Furthermore, silicone products also possess very high-volume resistivity, which refers to the resistance of a material to electrical current. A high-volume resistivity shows nominal electrical conduction, efficiently preventing leakage currents. Representative volume resistivity for insulating silicone typically falls within the 10^{14} to 10^{16} ohm-cm range, demonstrating its effectiveness as an electrical insulator.[114]

The robust electrical insulation characteristic in silicone is critical for preventing short circuits or interference within integrated systems where few components stay electrically isolated.[49–51,56,63,87,88] In soft actuators with integrated sensors, actuators, and power systems, this characteristic keeps electrical signals insulated, enabling precise and controlled processes in electroactive devices. For instance, the dielectric properties of silicone materials in bioelectronic devices or medical implants are essential for the precision and safety of implanted electrical systems. Furthermore, the combination of the insulating property and inherent flexibility of silicone materials is instrumental in soft robotics, where actuators must interact with complex electrical circuits.[12,32,49–51] This synthesis of mechanical flexibility and electrical insulation greatly expands the design possibilities for soft robotic systems that are smaller, more flexible, and power-conserving.

In order to understand the benefits of advanced silicone materials over traditional elastomers in a numerical manner, Table 1 gathers the main mechanical, thermal, and biocompatibility properties that are of interest for soft actuator applications. The advanced silicones display a substantially greater variation of stiffness and strength that can be adjusted, as well as a better elongation at break, thus enabling the actuators to be used under more demanding mechanical conditions. Besides that, the improved thermal stability and the specially designed biocompatibility feature, make their use possible in more severe environments and as implants in the biomedical field. On top of that, these materials are set apart even further

Table 1 Comparative properties of conventional *vs.* advanced silicone materials[54,106,115,116]

Property	Conventional silicone elastomers	Advanced silicone materials	Notes/significance
Young's modulus (MPa)	0.1–1.0	0.01–10.0	Advanced materials cover broader stiffness range
Tensile strength (MPa)	1–5	5–15	Enhanced mechanical robustness in advanced silicones
Elongation at break (%)	200–700	300–1800	Greater stretchability improves actuator flexibility
Thermal stability (°C)	Up to 200	Up to 300	Allows use in harsher environments
Biocompatibility	Generally good	High (with tailored surface chemistries)	Critical for implantable and wearable devices
Self-healing capability	Absent	Present in some formulations	Improves durability and lifespan
Processability	Conventional molding	Additive manufacturing, patterning	Enables complex geometries and miniaturization

by emerging features such as self-healing ability and compatibility with additive manufacturing techniques.

Understanding the fundamental properties of advanced silicone materials lays the groundwork for exploring how these materials have been further engineered and improved in recent years. The following section discusses advancements in silicone-based material formulations that enhance or expand upon these key properties.[117,118]

3. Advancements in silicone-based materials

Recent advances in silicon-based materials have significantly enhanced the prospects of soft actuators such that they can match rising demands in different industries, such as robotics, medicine, and aviation.[57,65,80,82,109–113] Such advancements encompass more than just the manufacture of better versions of the underlying properties of silicone elastomers; they involve the design of new hybrid materials with composite properties that render them more useful, responsive, and functional. The following sections discuss some of the most critical innovations in the area of silicone-based soft actuators. Particular focus is given to new material types, hybrid compositions, and bio-inspired designs.

3.1. Novel silicone elastomers

Novel silicone elastomers represent a significant step forward from traditional silicone rubbers. These elastomers are developed with better or entirely novel properties to meet the demanding needs of advanced uses. Although classical silicones are renowned for their inherent elasticity, biocompatibility, and thermal stability, these new elastomers exceed these fundamental characteristics by incorporating novel functionalities such as self-healing, improved mechanical strength, and responsiveness to external stimuli.[31,46,47,53,59,64,88,93,94,119,121,122] Among other innovations, the addition of reinforcing fillers, such as silica nanoparticles, metal nanoparticles, carbon nanotubes, and graphene, has been crucial in developing silicone elastomers.[98,121–132] Standard fillers enhance the mechanical properties of elastomers in the form of increased tensile strength, better fatigue endurance, and higher material toughness in general. For instance, silica nanoparticles can substantially strengthen the silicone matrix, enabling it to withstand better wear and tear from cyclic deformations; this is essential for actuators operating under cyclic motion or repetitive loading.[128–130] Carbon nanotubes and graphene significantly improve the tensile strength of silicone materials, enabling them to bend under higher actuation forces.[127–132] These developments will enable new scenarios for actuator design for applications in harsh environments, such as prosthetics and industrial robotics, where high actuation forces and mechanical strength are required. The synthesis and development of high-performance silicone elastomers with engineered properties make it possible to construct optimally designed soft actuators for specific tasks. For example, a very elastic silicone elastomer may be the best choice for a prosthetic that is going to need lots of stretching, whereas a soft robot intended for repetitive stress applications would benefit from mechanically robust silicone materials that provide long-term reliability.

3.2. Silicone–polymer hybrids

Silicone–polymer hybrid formation is one of the promising approaches in the development of soft actuators based on the selective addition of silicone and other specific polymers such as polyurethanes, thermoplastic elastomers, or polyolefins.[133–137] This mutual combination allows the application of the desired characteristics of each polymer to generate new hybrid silicone materials. These materials can be engineered to display a much greater range of mechanical properties, ranging from increased flexibility to increased tensile strength and designed sensitivity to environmental stimuli. This ability to achieve desired characteristics is helpful when a single material by itself is not adequate to offer the whole set of balanced properties needed for next-generation soft actuator devices. For example, whereas silicone materials are very flexible and self-biocompatible, they do not necessarily possess the tensile strength, the mechanical strength, or the ability to resist mechanical fatigue required for demanding applications.

Adding polyurethane to silicone increases mechanical strength and the degree of resistance to abrasion in hybrid materials. Moreover, silicone–polymer hybrid materials can be designed to be highly responsive to environmental stimuli. The combination of formable and resilient thermoplastic elastomers with silicone in actuator development can create a novel material with increased temperature, humidity, or external force response. Advanced hybrid materials will facilitate advanced soft robotics applications with superior speed and agility (*e.g.*, interactive wearable robots or sensitive medical devices). The built-in tunability of silicone–polymer hybrids enables the creation of very specialized soft actuators with customized functionalities, extending their applications from medical implants to advanced soft robot arms.

3.3. Bio-inspired silicone-based materials

One of the most promising research areas in soft actuation is the development of bio-inspired silicone materials that mechanically mimic natural tissues such as skin, muscle, or cartilage.[52,57,64,65,68,70,74,138–147] These next-generation silicones are envisioned to confer mechanically engineered attributes to soft actuators while simultaneously enabling them to adapt their behavior regarding their environment when pushed with external forces, as happens with biological tissue.[1,13,21,39,40,42,52,139,146] This kind of adaptability can provide lifelike motion and flexibility. One of the keys focuses on bio-inspired silicones is the search for materials with significantly enhanced flexibility and adaptivity. For instance, silicon can be designed to possess mechanical behavior that is like the flexibility of human muscle or skin.[1,26,30,34,37,57,63,70,93,128] Actuators capable of elongation, curvature, and shape change can be built (like living muscles). This is particularly important in soft robotics, where actuators must interact with the world safely and humanely, for instance, in the manufacturing of exoskeletons or assistive devices.

In addition to flexibility, bio-inspired silicones can also be designed to exhibit a more sophisticated mechanical response to applied load. This can facilitate the creation of soft actuators with sophisticated stiffness profiles akin to the different stiffnesses of biological tissues in location or function (*e.g.*, bone *versus* muscle tissue).[1,13,21,39,40,42,52,139,146] In prosthetic limbs, the same material would yield a more humanlike pressure and deformation response to achieve more natural movement and sensation. Bio-inspired silicones also heal themselves so that the actuator will repair minor damage on its own and have a longer lifetime; this quality is of very high value for wearable technologies and medical devices, where longer lifespans and reduced maintenance are of great importance.[146] In prosthetic limbs, such materials would provide a more humanlike deformation and pressure response, leading to more natural movement and sensation. Bio-inspired silicones also self-heal, meaning that the actuator would heal minor impairment on its own and last longer. This would be mainly beneficial for wearable technologies and medical devices, where durability and reduced maintenance are essential.

As new silicone materials continue to emerge, the development of compatible and innovative fabrication techniques becomes essential for realizing their full potential. The next section explores recent innovations in fabrication methods that enable precise structuring and integration of these advanced materials.

4. Innovations in silicone fabrication methods

Advanced manufacturing processes, advanced fabrication technologies, and advanced techniques are significant for realizing the highest potential of soft actuator materials. Utilizing these techniques makes it possible to create complex geometries, multicomponent composites, and high-accuracy features that are very useful in high-performance soft actuator production. Through these new methods, material properties on the nanoscale and microscale can be accurately designed. This accuracy control enables greater responsiveness, longevity, and overall functionality of the resultant devices. This manufacturing innovation is pivotal in pushing the boundaries in fields such as soft robotics, biomedical devices, and other high-end uses, allowing for the necessary transition from lab prototype to standard, mass-producible systems. The following section discusses the latest fabrication methods in detail.

4.1. 4D printing

Four-dimensional (4D) printing significantly advances conventional 3D printing by introducing a time factor such that created objects can alter their shape, properties, or function autonomously with time in response to specific environmental stimuli.[35,148–165] Unlike passive 3D-printed structures, 4D-printed structures are fabricated to experience dynamic transformation. This novel technology exploits the high-precision layer-by-layer manufacturing of smart materials, which elastically react to environmental stimuli such as temperature, humidity, light, pH, or electric fields. Four-dimensional printing design hence blends both the initial 3D shape and engineered material response to enable the production of self-assembling, self-shaping, or functionally adaptive structures without subsequent external input following fabrication (Fig. 4a). This additive manufacturing paradigm enables the development of soft actuators with the capacity to adjust their shape or their properties dynamically, providing unprecedented flexibility. Using intelligent materials, it is feasible to design actuators that can self-assemble, possess dynamic motion, or evolve to intricate shapes.

Silicones are well suited for 4D printing applications, particularly for the development of soft actuators and adaptive systems.[155,157–166] The intrinsic flexibility and elasticity of silicones, along with their broad operating temperature range and their biocompatibility, offer a strong foundation for developing dynamic structures that can be transformed in real time. Embedding desired functionalities in silicone material directly using the additive manufacturing process is possible through

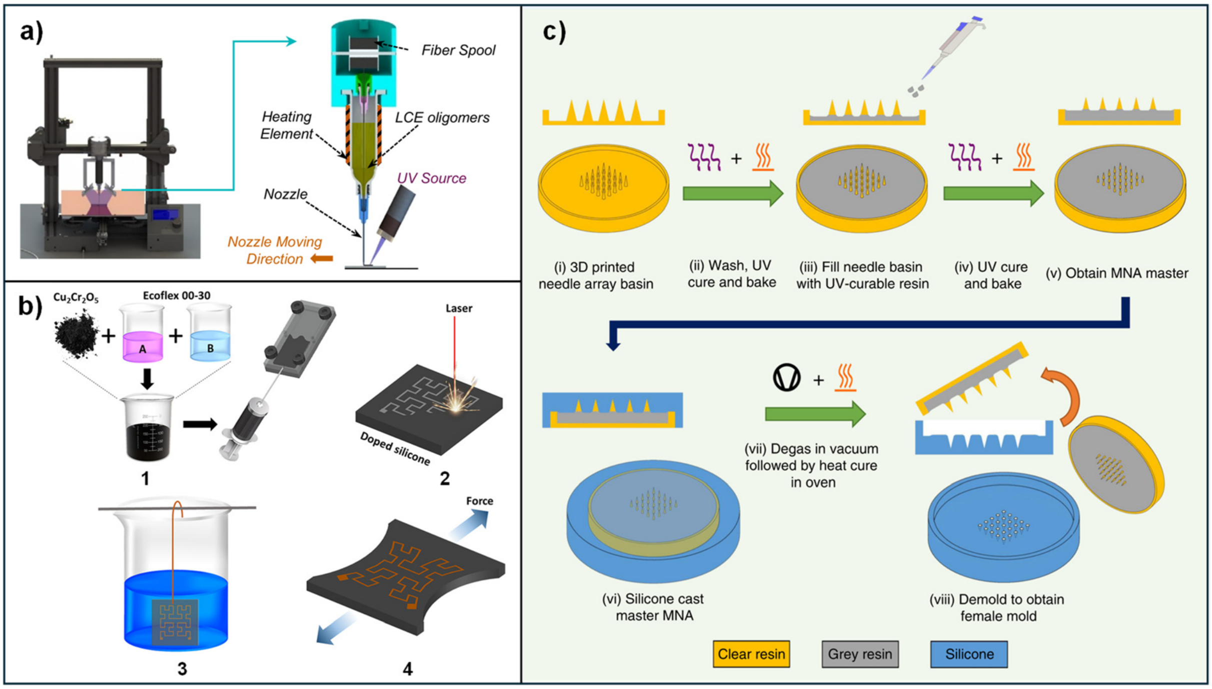

Fig. 4 (a) Schematic diagram of the 4D printing setup with a printer head, reproduced from Jiang *et al.*[165] under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License. (b) Schematic diagram of laser direct structuring on silicone process: (1) mixing and casting; (2) laser structuring; (3) electroless copper plating; (4) resulting circuit; reprinted with permission from Yoo *et al.*,[167] Copyright 2022 American Chemical Society. (c) Microneedle arrays master mold fabrication method, reproduced from Krieger *et al.*[179] under a Creative Commons Attribution 4.0 International License.

4D printing.[157–166] For example, thermoresponsive silicone materials are printed with high precision to develop shape-changing actuators (*e.g.*, expand or contract) automatically in response to temperature changes. This decreases the necessity for external control systems, enabling self-sustaining and energy-efficient behavior. This property is particularly applicable in adaptive robotics, where actuators need to respond to environmental or user inputs without any sophisticated external control systems.

Similarly, 4D printed electroactive silicone-based actuators that deform appropriately when an electric field is applied will open new doors of opportunity for high-end soft robotics and dynamically reconfigurable components.[157–166] Multimaterial 4D printing techniques will facilitate the strategic combining of silicone material with other responsive polymers. Patterning the different recipes of silicone or other polymer materials precisely will enable a sophisticated and localized response to be engineered through 4D printing. The differential expansion leads to folding or bending of the 4D printed silicone structures into a preprogrammed complex shape upon heating. This places 4D printing of silicone materials as a revolutionary technology for uses involving real-time adjustment, self-assembly, and more human–device interaction. The creation of 4D printing technology for actuators in soft robotics offers an exciting potential for highly enhanced functionality, autonomy, and less maintenance, where the possibilities of self-assembly and self-healing are provided in real time. These higher-order characteristics put 4D printing on the threshold of widespread application in medical devices, smart fashion, and soft robots, mainly in actuators that need to adapt securely and discreetly in real-world settings while interacting with the environment.[157–166]

4.2. Laser-based fabrication

Laser processing techniques, including laser sintering, laser ablation, and laser-induced forward transfer, are exact techniques for microfabrication and nanofabrication of silicone-based soft actuators.[167–174] These methods are particularly beneficial for the fabrication of actuators with intricate geometries or microstructures that might be challenging to produce using conventional molding or casting techniques. Laser fabrication makes it possible to fine-tune the shape of silicone structures with very high resolution to create soft actuators of complex, multilayer geometries (Fig. 4b).[167–174] Such geometries are especially pertinent for applications such as soft robotic grippers, advanced medical implants, and sensor or fluid channel integrated devices. The techniques also allow for complex surface patterning, which enhances the actuator's interaction with foreign objects, thereby increasing its grip or flexibility. Above all, laser-based techniques allow for rapid production of high-quality, low-cost prototypes. This is essential for speeding up the design of new soft actuator devices. Furthermore, the techniques are highly scalable, making them particularly well suited for the mass production of small and complex actuator components.

4.3. Micro molding

Micro molding is a highly specialized production method tailored to produce tiny high-precision components.[175–180] Micro molding creates complex shapes with extremely close tolerance, frequently on the order of micrometers. The process is essentially a miniaturized version of conventional injection or compression molding for commodities that are lighter than 1 g and that exhibit submillimeter features (Fig. 4c). The basic micro molding process is to inject or pressurize molten or fluid material into a closely machined mold cavity, which then solidifies creating the desired micropart.[175–182] Micro molding machines are equipped with high-precision injection units and advanced control systems to maintain infinitesimal shot sizes as well as consistent material flow into the infinitesimally small mold cavities.

Micro molding is a sophisticated technique that enables highly reproducible and precise manufacture of silicone microstructures.[183–189] Micro molding is conducted by injecting liquid silicone elastomers into precisely crafted molds with intricate internal patterns (*e.g.*, microfluidic channels, *in situ* sensors, or multilayer actuator geometries).[190,191] The most significant advantage of this process is its capacity to develop actuators with precise functions for application in small or restricted areas; these actuators are highly valuable for micro robotic and advanced medical procedures.[190] Micromolded actuators, for instance, are essential in minimally invasive surgery, where diminutive flexible structures must be applied to deal with soft tissues or perform very delicate interventions within the body.[185,186,189,190] Furthermore, micromolding allows for the creation of large numbers of silicone-based soft actuators. This scalability is important to consider for the mass commercialization of healthcare devices, sensors, and robots. The precision and scalability of micromolding enable the development of sophisticated actuators without the compromise of functionality or price feasibility.

Silicones are specifically well suited for micromolding because of their unique set of attributes, which make them highly useful to produce minute components in applications such as medical devices, microelectronics, and soft robotics.[22,57,64,183–191] The intrinsic elasticity, flexibility, biocompatibility, and chemical inertness of silicones are particularly advantageous in the manufacture of microparts that require conformability, interaction with biological systems, or protection from harsh environments.[57,60–68,70,85–92,96–113] Liquid silicone rubber is frequently employed in micromolded silicones due to its low viscosity, which renders it fluid enough to penetrate tiny and complex mold cavities and fill and reproduce fine microfeatures. Typically, injecting the liquid silicone rubber into a preheated mold cavity, where it vulcanizes (cures) to form the end component solid, is involved. The property of silicone to cure quickly and produce significant byproducts additionally improves the efficiency and precision of the micromolding method. Micromolding allows the manufacture of silicone devices and parts with very fine details, such as microfluidic channels, miniature seals, small diaphragms, and complex connectors, all with excellent tolerances. For example, in medical technology, micromolded silicone components are critical for catheters, drug delivery systems, and implantable sensors, where precision and biocompatibility at a microscopic level are essential. In microrobotics, this technique facilitates the fabrication of compliant actuators and grippers with intricate internal structures that allow highly dexterous activities. With fine-tuned material preparation, injection conditions, and curing cycles in micromolding, high repeatability and sound quality, which are of prime importance, are assured when these minute but functionally important silicone parts are produced in high volume.

Fabrication advancements not only enable complex geometries but also facilitate the integration of novel functionalities into silicone-based actuators. The following section highlights such functionalities, including self-healing, thermoresponsiveness, and electroactivity.

5. New functionalities of advanced silicone materials

New functionalities of silicone-based soft actuators are at the core of the advancement of functionalities enabled by these devices. Outside simple expansion and contraction, novel functionalities such as self-healing, self-sensing, and multimodal actuation (for example, combining electrical and pneumatic responses) allow unique adaptability and intelligence. These advanced characteristics permit soft actuators to function autonomously in dynamic, unpredictable environments; mimic complex biological actions with greater fidelity; and deliver integrated feedback. These developments are crucial for developing intelligent soft robots. These advanced prostheses communicate seamlessly with the human body, and advanced medical equipment can accomplish complex procedures with minimal operator feedback, meaning that there are many essential situations for the use of advanced silicone material. The following sections discuss the state-of-the-art functionalities of silicone-based materials in detail.

5.1. Self-healing properties

The self-healing properties of new silicone materials mean that they have a superior potential to independently heal damage, such as cracks, scratches, or punctures.[192–197] The ability to recover structural integrity and function without any external human involvement significantly enhances the lifetime and reliability of silicone products, particularly in extreme or inaccessible environments. This is achieved by the inclusion of specific chemical mechanisms within the silicone polymer network that allow for the breaking and, upon damage, re-forming of molecular bonds. Self-healing in silicones is typically characterized as being extrinsic or intrinsic.[192,193,196,198] Extrinsic self-healing is when a healing agent, typically encapsulated in microcapsules or contained within a vascular network, is incorporated into the material.[198] When damaged, these capsules are ruptured, or the vascular system delivers the healing agent to the cracked zone. The agent then reacts, usually in the presence of a catalyst, to bond and fill the cracks. Although beneficial, this method usually offers a finite number of healing cycles because the healing agent can be exhausted. Intrinsic self-healing, however, is a more advanced approach that relies on the polymer possessing an inherent ability to heal without utilizing external healing agents.[198] This is typically accomplished by adding dynamic or reversible bonds to the molecular structure of the silicone. When the substance is broken, these bonds are broken, and upon proximity or the use of a weak stimulus (*i.e.*, heat, light, or pressure), they can re-form, effectively reattaching the substance.

The development of self-healing silicone materials has facilitated groundbreaking functions for soft actuators.[20,21,39,42,52,146] These new silicones possess the unique ability to self-repair minor cracks, tears, or damage in the course of routine operations. This natural self-healing capacity significantly enhances the lifespan and durability of soft actuators, particularly under operating conditions of heavy wear or extreme environments, where classical materials would typically degrade or fail over time. Self-healing is very beneficial for repetitive motion and abrasive environment applications of soft actuators (*e.g.*, industrial robots, wearables, and soft exosuits).[20,21,39,42,44,52,82,127,146] Self-healing silicone provides the assurance that even after actuators have undergone minor damage, they will still perform at full capacity, making costly replacements or incessant repairs unnecessary. Alternatives like these not only make equipment stronger and more durable; they also translate into better sustainability and economic viability during the equipment's working life. In addition to enhanced performance in harsh environments, self-healing silicones are also finding critical applications in the medical field, where device integrity directly equates to patient safety. Implantable devices or robotic medical devices, for example, can be modeled with self-healing materials as a surface coating to minimize malfunction or degradation within the body during long-term use.

5.2. Thermoresponsive properties

Thermosensitive silicones constitute a technologically advanced class of smart materials that extensively and frequently change their physical and chemical characteristics (*e.g.*, shape, rigidity, or solubility) upon temperature variations.[76,90,103,105,110,199–204] This dynamic behavior makes thermosensitive silicones extremely valuable for applications that require adaptive responses to temperature stimulations, particularly thermal actuators, soft robotics, and adaptive thermal management systems.[81,120,164,199–204] The underlying mechanism of thermoresponsiveness in silicone materials is a delicate balance of intermolecular forces and chain conformation. Whereas the native polysiloxane backbone ensures flexibility and stability over a broad range of temperatures, the thermoresponsive nature is specially engineered. It is achieved *via* the incorporation of specific functional groups or *via* the construction of hybrid systems that demonstrate a particular transition of phase or change of hydrogen bonding and/or hydrophobic interactions at a specific temperature.[76,90,103,105,108,199–204] For instance, some thermoresponsive polymers are characterized by a lower critical solution temperature, meaning that the polymers become insoluble or undergo a volume phase transition (*e.g.*, collapse or shrink) at a temperature greater than a critical temperature. However, upper critical solution temperature polymers become soluble when above a critical temperature. When temperature exceeds such a clearly defined limit, the balance between interactions within the polymer chains or between the chains and the environment is disturbed. Changes in conformation ensue, such as the collapse or dilatation of polymer segments or in hydrational state. Macroscopically, such molecular changes are expressed as quantifiable shape, rigidity, or even wetting modifications on the surface, enabling the material to function as a temperature-stimulated, intelligent component without regard to mechanical or electrical inputs from the surroundings. In soft robotics, thermoresponsive silicones can be engineered as actuators that can dynamically change their shape upon a temperature change and thereby enable autonomous actuation without external control.[81,120,164,199–204]

For example, silicone actuators can be engineered into systems that either expand or contract based on the ambient temperature and thus can be efficient and self-sustaining in their movement. These thermoresponsive actuators are useful in fields ranging from self-deploying structures to adaptive robotics, where devices must respond to environmental conditions in real time. Thermoresponsive silicones are also highly significant in thermal management systems. These materials can be utilized to create responsive components that regulate temperature in delicate applications such as electronics, wearable electronics, or aerospace.[12,32,49–51,199–204] Materials can be designed to stretch or contract in response to temperature changes, making them excellent for managing heat flow or retention in machines under tight conditions.

5.3. Electroactive properties

Electroactive properties of advanced silicone materials refer to the capability of these materials to alter their shape, size, or other mechanical properties upon exposure to an electric field.[56,76,77,90,103,110–113,205–209] These materials have long been categorized as electroactive polymers. They are now the subject of much interest due to their potential as artificial muscles with high achievable strains, high response speeds, and high energy densities.[30,37,77,90,93,103,110–113,128] The electroactive behavior of silicones is primarily a result of their use as dielectric elastomer actuators (DEAs). A DEA usually consists of a soft, insulating silicone elastomer film sandwiched between two compliant electrodes.[210–212] Upon the application of an electric field across the two electrodes, the primary reason for deformation is the Maxwell stress (*i.e.*, the electrostatic attractive force between the opposite charges of the electrodes). The Maxwell stress successfully compresses the insulating silicone layer in thickness. Because silicone is near incompressible, this volume compression results in a corresponding increase in the planar area of the material. Although electrostriction, a deformation related to the square of the applied electric field, does play a role, the Maxwell stress effect prevails. This mechanism enables high reversibility and low-voltage-controllable shape and size transformations, thereby enabling electroactive silicones to find application as artificial muscles in a variety of advanced technologies.

This characteristic makes electroactive silicones a universal source of actuation in soft robotics, where actuators can alter their shape with accuracy or execute defined movements based on electrical stimuli.[7,8,32,57,60–67,81,164] The uses of such actuators are diverse in conditions that require precise motions (*e.g.*, prosthetic limbs, wearable robots, and adaptive grippers).[31,45,138] Unlike traditional actuators, electroactive silicones have certain advantages of their own; their inherent softness and pliability enable them to produce detailed organic motion that is challenging to achieve with hard materials. Beyond the realm of soft robotics, electroactive silicones also hold great promise in active tactile display systems that provide users with real-like touch. By using applied electric fields of different magnitudes, these materials can produce a broad range of vibration, contact, or deformation and serve to simulate haptic feedback effectively. This capability is creating new fields of usage in virtual reality, telemedicine, and assistive devices for visually impaired or hearing-impaired individuals.

With enhanced properties and new functionalities, advanced silicone materials are now being deployed in a wide range of real-world applications. The next section discusses emerging uses in areas such as wearable robotics, implantable devices, and soft grippers.

6. Emerging applications of advanced silicone materials

High-performance silicone materials are some of the most critical innovation enablers across most industries. They can be used in highly compliant and adaptable robots for safe human interaction, precise manipulation in manufacturing, or exploration in soft robotics. In biomedical devices, their biocompatibility as well as mechanically tunable nature play key roles in advanced prosthetics, implantable sensors, drug delivery systems, and wearable monitors that naturally innervate the human body. Their hardness, thermal stability, and low weight in the space environment are all essential in seals, gaskets, and protective coatings for aircraft and spacecraft. More recently, they have been used to make shape-morphing devices, self-healing devices, and dynamically adaptive materials in flexible electronics, smart textiles, and 4D printing. The following sections discuss the latest applications of silicone-based materials in detail.

6.1. Soft exosuits

Soft exosuits or wearable robotic systems (so-called *exoskins*) represent a new approach to human augmentation and support.[213–221] Unlike conventional rigid exoskeletons founded upon stiff, external frameworks, soft exosuits are garment-like devices constructed from flexible, lightweight material, with silicone being a fundamental material. These suits are designed carefully to function in parallel with the human body's musculoskeletal system, adding assistive forces and torques to specific joints or groups of muscles through textile-based load paths that are integrated within the suits (Fig. 5a and b).[215–221] The inherent properties of silicone-based actuators (*i.e.*, their high flexibility, strength, and versatility) make them highly suitable for generating human-like movement while ensuring user comfort, safety, and wearability.[57,60–68,70,83–92,96–113,213–221] Unlike traditional, rigid exoskeletons, soft exosuits are typically compliant and lightweight, and they enable users to move about unimpeded by the high encumbrance of large, rigid structures.

Silicone soft exosuits augment human motion through the selective application of silicone-based actuators, which are most expressed in the form of inflatable pneumatic bladders or advanced DEAs.[212–224] The actuators are integrated into the stretchable fabric of the suit. Upon actuation—whether *via* pressurized air or fluid for pneumatic systems or through an electric field for DEAs—the silicone components undergo controlled expansion or contraction. This deformation generates assistive torques and forces that are transmitted by the textile architecture to the joints and limbs of the wearer. A sophisticated control system, based on the quantity of sensors, clocks the actuator's output about the intended movement and gait phase of the user. This allows the exosuit to render precise, natural support, enhancing mobility, reducing metabolic energy consumption, and preventing discomfort due to the compliance and elasticity that are natural in silicone.

Silicone soft exosuits bring significant advantages to human augmentation, primarily due to the intrinsic nature of silicone materials.[212–224] They are constructed as a soft, lightweight framework that offers additional comfort, wearability, and more natural collaboration with the human body than with rigid exoskeletons, leading to less metabolic cost and damage risk. The biocompatibility created for silicone makes such suits safe for long-term contact with the skin, making them very appropriate for medical and assistive purposes. Moreover, the ability of silicone actuators to work silently, unadulterated by external mechanical systems, contributes significantly to making such exosuits comfortable and convenient to wear for everyday activities.

These advantages are realized in different meaningful applications. In rehabilitation, soft exosuits deliver targeted support for gait training and limb drive, assisting those recovering from situations such as stroke, spinal cord wounds, or other issues that weaken mobility. In industrial and occupational applications, these exosuits can meaningfully decrease fatigue and mitigate musculoskeletal damage for workforces carrying out strenuous or tedious everyday jobs. Soft exosuits also harness the significant potential of aging mobility assistance, promoting independence and reducing the risk of falls. Military and emergency personnel are even

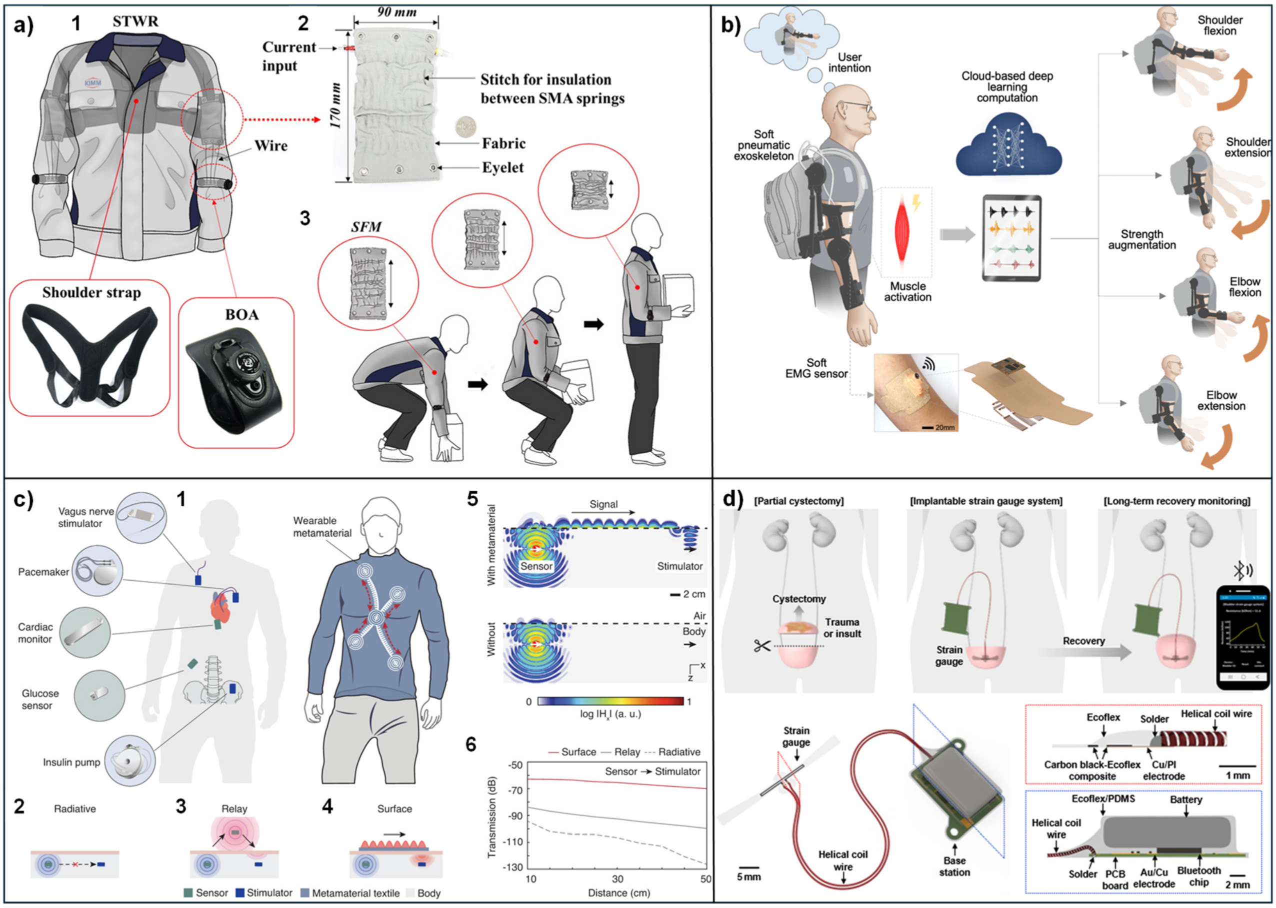

Fig. 5 Examples of soft exosuits. (a) Soft wearable robot for assisting the muscular strength of the arms for lifting heavy objects, reproduced from Park *et al.*[220] under a Creative Commons Attribution 4.0 International License. (b) Overview of smart upper-limb exoskeleton with soft sensors and soft actuators, reproduced from Lee *et al.*[221] under a Creative Commons Attribution 4.0 International License. (c) Implantable devices with wearable metamaterials and wireless connectivity, reproduced from Tian *et al.*[225] under a Creative Commons Attribution 4.0 International License. (d) Wireless implantable system for quantitative monitoring of bladder function, reproduced from Kim *et al.*[226] under a Creative Commons Attribution-NonCommercial-NoDerivatives License 4.0.

investigating them to enhance endurance and load-carrying capability in harsh environments.

6.2. Implantable devices

Silicone-based soft actuators are drawing significant interest for implantable biomedical devices due to their high biocompatibility, environmental sensitivity, and mechanical flexibility.[57,60–68,70,85–92,96–113,225] All these features are crucial for next-generation drug delivery systems, biosensors, and microsurgical robots. Silicone actuators can be implanted inside the human body and engage in interactions with the body's tissues without causing inflammatory and rejection responses.[44,60–68,70,82,93] Implantable drug delivery systems, for example, can utilize silicone-based actuators to accurately release drugs upon sensing specific biological signals, including pH or temperature. These smart systems enable more targeted and site-specific drug delivery, which can make therapies for diseases such as diabetes, cancer, or neurological disorders more efficient (Fig. 5c and d). Silicone-based actuators have also been used in biomedical sensors to detect specific biomolecules or intrabody environmental conditions and thereby allow for real-time monitoring and diagnostics. Due to the ability of silicone to retain its mechanical properties in biological systems, the material is also well suited for microrobotic surgery, which demands precision in gentle intervention. Actuators in such robots can offer flexible, minimally invasive interventions, leading to reduced recovery times as well as improved patient outcomes (Fig. 5d). Of note is the inherent biocompatibility of silicone; this biocompatibility is an absolute advantage for implantable devices because they will neither be rejected by the immune system nor cause long-term complications.[44,60–68,70,82,93] This quality is also conducive to the stability and durability of devices to be left within the body for prolonged periods.

6.3. Soft robotic grippers

Using silicone-based grippers as soft robot grippers is the future solution for accurate manipulation tasks when dealing with fragile, oddly shaped, or soft objects. Unlike rigid robot grippers, silicone-based grippers can conform to the precise shape and surface of an object so that it can be grasped firmly but gently.[31,45,83,138,162] This adaptability significantly reduces the chances of breaking down fragile materials. The compliance and flexibility provided by silicone actuators make these grippers applicable to a wide range of applications. These applications range from the manipulation of sensitive biological samples in research settings to the handling of food items for packaging processes. These grippers are also highly relevant in some surgical procedures, such as in minimally invasive surgery and surgical robotics, where sensitive and precise manipulation of tissues and organs is essential. One of the significant advantages of silicone-based soft grippers is their ability to handle objects with pertinent variations in geometry and material properties. The intrinsic deformability and flexibility of the actuators offer great versatility, and fields of application range from automated manufacturing to assembly lines to service robotics. Additionally, silicone-actuated soft robotic grippers facilitate better safety in human–robot interaction, where their compliance feature mitigates the possibility of causing injury or discomfort upon handling. These applications demonstrate the growing impact of advanced silicone materials across multiple domains. The future perspective and concluding sections summarize key findings and discusses future research directions in the field.

7. Future perspectives and research directions

Even with major breakthroughs in the development of soft actuators, in particular, those made of silicone materials, many potential research paths are still available. A main focus for the future could be the creation of multifunctional silicones that not only have self-healing properties but also feature improved mechanical strength and biocompatibility, thus soft robots and implantable devices that last longer and are more reliable. Combining machine learning and real-time sensing in soft actuation systems is yet another challenging horizon that open adaptive, autonomous behavior which can be a dynamic response to complicated surroundings.

Moreover, the concept of biohybrid systems, wherein living cells or tissues are merged with synthetic materials, to produce actuators with extraordinary functionality and sensitivity is a viable option in the future. On the manufacturing side, the large-scale, highly precise fabrication methods like additive manufacturing and micro-patterning will determine how far the complexity and miniaturization of soft actuators can be taken. The issue of environmental sustainability through the creation of recyclable or biodegradable silicones is going to be as important as the field is moving towards a wider practical deployment. As a whole, these research themes will be instrumental in shaping the following generation of soft robotic systems that are intelligent, durable, and flexible.

8. Conclusion

Advanced silicones are singly transforming the domain of soft actuators *via* a clear and considerable set of qualities, such as high elasticity, biocompatibility, and excellent durability. These qualities and compatibility with a wide range of sophisticated fabrication techniques put silicones at the forefront of breakthroughs in robotics, medicine, and aviation. Their vast potential is spread across a list of future applications. The ever-growing newer materials (*e.g.*, silicone–polymer composites and bio-inspired silicones) and the advanced fabrication technology in the form of 4D printing and laser processing are going to further augment the functionality and multiapplication prospects of soft actuators. Increased and continued research in this fast-growing area is therefore bound to power revolutionary advancement in the performance and multifunctionality of silicone-based soft actuators in the coming years.

Author contributions

KM: writing – review & editing, writing – original draft, visualization, conceptualization. GK: writing – review & editing, writing – original draft, visualization, conceptualization. RA: writing – review & editing.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

This article is a review and does not contain any original experimental data. All data referenced and discussed in this manuscript are available from publicly accessible sources, including peer-reviewed publications and scientific databases, which are properly cited within the text. No new datasets were generated or analyzed during the current study. No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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