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Processing of Self-Healing Polymers for Soft Robotics

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Soft robots are, due to their softness, inherently safe and adapt well to unstructured environments. However, they are prone to various damage types. Self-healing polymers address this vulnerability. Self-healing soft robots can recover completely from macroscopic damage, extending their lifetime. For developing healable soft robots, various formative and additive manufacturing methods have been exploited to shape self-healing polymers into complex structures. Additionally, several novel manufacturing techniques, noted as (re)assembly binding techniques that are specific to self-healing polymers, have been created. Herein, the wide variety of processing techniques of self-healing polymers for robotics available in the literature is reviewed, and limitations and opportunities discussed thoroughly. Based on defined requirements for soft robots, these techniques are critically compared and validated. A strong focus is drawn to the reversible covalent and (physico)chemical cross-links present in the self-healing polymers that do not only endow healability to the resulting soft robotic components, but are also beneficial in many manufacturing techniques. They solve current obstacles in soft robots, including the formation of robust multi-material parts, recyclability, and stress relaxation. This review bridges two promising research fields, and guides the reader toward selecting a suitable processing method based on a self-healing polymer and the intended soft robotics application.

1. Introduction

Due to their inherent softness and compliance, soft robots are often used for grasping delicate objects,^[1,2] including fruits and vegetables in food packaging,^[3,4] agriculture,^[5] electronics, and even corals.^[6] Their inherent flexibility also provides the required safety in applications where close human-robot interaction is needed,^[7,8] like industrial co-workers,^[9] social robots,^[10] surgical robots,^[11] and wearable robotics^[12] including prosthetics^[13] and exoskeletons.^[14] Typically, soft robots are constructed out of soft materials that have a stiffness similar to the materials found in living organisms (0.1 MPa to 1 GPa).^[15] Although softness provides many advantages, it also introduces a drawback: soft materials are more prone to damage. This is specifically problematic for soft robotic systems which find their application in unstructured, dynamic, and unpredictable environments.^[12,16] The few examples of soft robots in the industry report a limited lifetime^[17] or a clear warning not to use

the robot with sharp objects.^[18] The lifetime of the Versaball that was commercially available is reported as 50 000 grips, but reduces to only 5000 grips when working with abrasive objects.^[17] This limited lifetime of soft robots is a key issue that should be addressed before widespread adoption in the industry can be done economically and ecologically. In addition, most currently developed soft robotic components are manufactured from elastomers, for example, silicones,^[19] that are not fully recyclable. This nonoptimal recyclability, in combination with the limited lifetime, leads to unsustainable future applications.

Recently, the solution has been proposed to make soft robots out of self-healing polymers.^[20–24] These self-healing polymers^[25] have the intrinsic ability to heal microscopic and macroscopic damage and recover their functional performance, either fully autonomously, without the need of any external intervention, or by means of an external stimulus, for example, heat or light. Researchers have proven that this self-healing concept in soft robotics is not only useful in pneumatic,^[26,27] electrostatic,^[28] tendon driven,^[29] or even passive actuators,^[30] but also in their electronics^[31] and sensors.^[32,33] The polymers used to create self-healing soft robotic actuators are all based on the incorporation of reversible (physico)chemical bonds into their polymer network structure.^[34] This renders the

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DOI: 10.1002/adma.202104798

network polymerization reversible upon the application of the adequate stimulus,^[20] usually heat. Consequently, these reversible polymer networks can be reshaped, reprocessed, and recycled,^[35–37] in contrast to many traditional elastomers used in soft robotics.^[19] This review is dedicated to the new opportunities this (re)processability of reversible elastomers adds to the manufacturing of smart (soft) robotic structures with increased complexity and reliability. As illustrated in this paper, the reversibility of polymerization opens new possibilities.

The field of soft robotics^[43,44] and, in particular, the recently established sub-field of self-healing soft robotics^[20–24] are multidisciplinary research fields in which both material science and mechanical engineering are playing an active role. This work intends to bring both research fields closer together by discussing several processing methods for self-healing polymers, as processing can be considered the bridge between them (Figure 1). Most of the work in soft robotics is currently done using a limited number of commercially available silicones, for example, EcoFlex or DragonSkin (Smooth-On, Inc.), while a good choice of material can have a major impact on the mechanical performance of soft robotic systems. On the other hand, material scientists are developing smart, complex materials, including self-healing materials, while many of these innovations remain to be tested in actual applications. More intense collaborations will lead to opportunities to share and discuss the capabilities, limitations, requirements, and drivers among the two fields to stimulate innovation and technology. However, this can only be facilitated when the link between both fields is available, for example, when these materials can be shaped in soft robotic components (Figure 1). This review discusses the processing possibilities for self-healing polymers to manufacture soft robots. Manufacturing soft robotics out of self-healing polymers, not only integrates a healing ability in soft robotic components, but provides new opportunities and perspectives for advanced manufacturing and recycling. Both fields can benefit from the advantages that self-healing elastomers can bring on a manufacturing level.

This review starts with listing the processing requirements for manufacturing techniques for soft robotics. Next, the different classes of self-healing mechanisms and elastomers are introduced. Thereafter, the formative and additive processing methods as well as manufacturing methods specific to intrinsic self-healing polymers, for example, assembly and binding techniques, are introduced and described. For each manufacturing technique, the advantages and disadvantages for soft robotics are discussed and linked to predefined requirements, specifically looking at the processing of self-healing polymers. In addition, this review intends to guide the reader toward selecting a suitable processing method for different situations.

1.1. Processing Requirements for Soft Robots

The ideal technique to manufacture soft robots should in the first place be of low complexity (i), yet allow for a large design freedom (ii) to permit the development of innovative robotic systems with a large embodied intelligence.^[45] In this respect, the technique would allow for a wide range of materials (iii) to be processable or have a low enough entry barrier for new ones. A wide scope of processable materials allows not only to process materials with a wide range of properties, but also to combine multiple materials and different components within a single design. This multi-material (iv) processing could permit the incorporation of interesting components such as sensors,^[46] hydraulic fluids,^[47] heaters^[48] in a single manufacturing technique or even step, without the need for a complex multi-step process or for post-processing. The produced components should have excellent material properties (v), as a result of minimal deterioration of the material properties during manufacturing or even enhanced properties due to processing. In addition, in many applications, an excellent surface finish (vi) is required for proper functioning of the component, as well as for aesthetics. In food-related applications, a bad surface finish

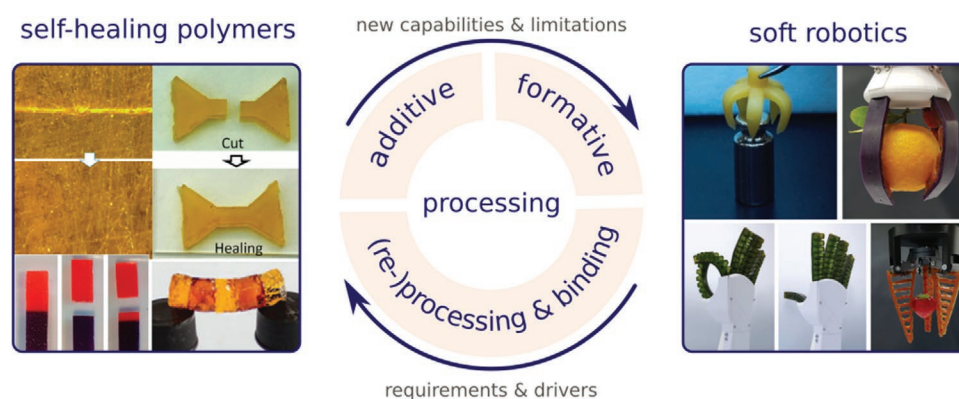


Figure 1. This review intends to form a bridge between two fast-growing fields of research: self-healing polymers, and soft robotics. Processing methods allow to convert these polymers into self-healing soft robotics, and are discussed in this review. Self-healing polymers: Top-left image: Reproduced with permission.^[26] Copyright 2017, The Authors, published by American Association for the Advancement of Science (AAAS). Top-right image: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (<https://creativecommons.org/licenses/by/4.0>).^[38] Copyright 2019, The Authors, published by Springer Nature. Bottom-left image: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (<https://creativecommons.org/licenses/by/4.0>).^[39] Copyright 2019, The Authors, published by MDPI. Bottom-right image: Reproduced with permission.^[40] Copyright 2014, Royal Society of Chemistry. Soft robotics: Top-left image: Reproduced with permission.^[41] Copyright 2019, American Chemical Society. Top-right image: Reproduced with permission.^[29] Copyright 2019, IEEE. Bottom-left image: Reproduced with permission.^[42] Copyright 2020, IEEE. Bottom-right image: Reproduced with permission.^[30] Copyright 2020, Mary Ann Liebert Inc.

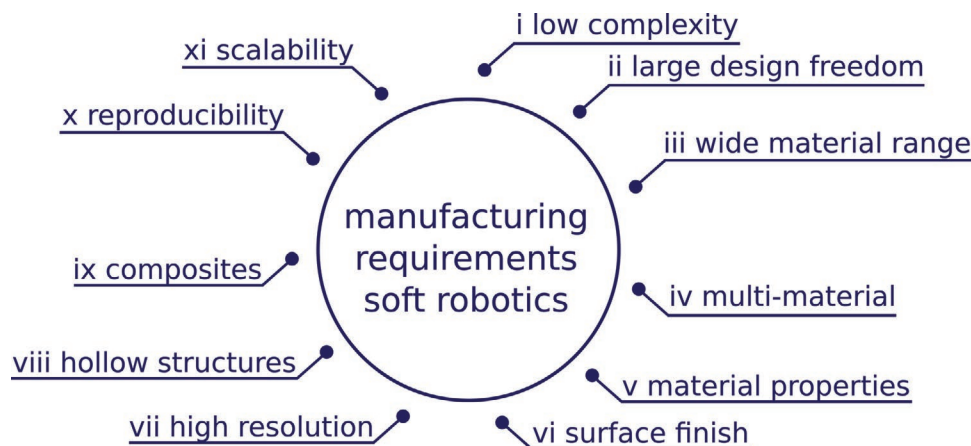


Figure 2. The manufacturing of soft robots poses different requirements on the processing techniques.

can lead to undesired bacteria growth in the ridges on the surface. A high manufacturing resolution (vii) can provide this and allows to produce components with high precision on small scales. With the general trend of making (soft) robotic systems ever lighter and more compact, to increase performance and efficiency, high resolution manufacturing techniques will be increasingly used. Although post-processing steps such as UV curing or a heat treatment increase the time and complexity of manufacturing, it is sometimes useful to enhance the material properties or to enhance surface finish.

There exist many different actuation principles for soft robots.^[49,50] While there exist passive designs,^[30,51] most rely on an external source for the actuation. This ranges from tendon-driven,^[29,52] where the actuation depends on a change in length of one or more tendon wires, to electrical (dielectric,^[28] piezoelectric^[53]) or even magnetic stimulation.^[54] Heat is also a common trigger, controlling different types of actuation mechanisms, including the shape memory effect of metal alloys^[55] or polymers,^[56] and liquid-gas transformations.^[57] But the most common systems are pneumatically actuated by pressurizing their inner structures using an external pressure source^[58,59] or internal explosions.^[60] Consequently, to be able to produce these soft pneumatic robots, the manufacturing technique should allow to create hollow structures (viii).

More and more, soft electronics^[61,62] and specifically soft sensors are playing an active role in the development of soft robotics. These sensors enlarge the potential of soft robots and allow them to estimate their own state (proprioception) and the state of their environment (exteroception). As they are flexible themselves, the integration of these electronics does not compromise the flexibility, however, like the soft parts in which they are embedded, these soft sensors are vulnerable to damage as well. Consequently, sensors embedded in self-healing soft robots, have the additional requirement of being self-healing as well.^[63] These sensors often are mostly manufactured out of conductive self-healing polymers. Electrically conducting self-healing elastomers^[64,65] can be achieved by polymer backbones that are intrinsically conductive, including polypyrrole^[66] and polyaniline,^[67] or it can be achieved by the addition of fillers to the (self-healing) polymer matrix. Typical conductive fillers include carbon black,^[68] carbon nanotubes,^[69] silver nano-wires,^[70]

or liquid metal droplets.^[71] The ideal manufacturing technique allows to directly integrate these soft electronics in the soft robotics body.^[72] Therefore, the manufacturing technique should be able to process polymer composites (ix), for example, polymer matrices compounded with fillers. This is also useful when manufacturing systems with an electric or magnetic actuation source, as these often require the use of composites.

For widespread adoption in the industry, it is important that manual labor during manufacturing is reduced as much as possible. This improves the quality of the manufacturing process by increasing repeatability and reproducibility (x). However, automation is not the only factor to take into account. For example, the recent advancements of additive manufacturing techniques^[73–75] have greatly increased the level of automation during manufacturing, but are not easily scalable. This xi) scalability is essential when soft robots step out of the lab and go toward the market.

Unfortunately, a manufacturing technique that fulfills all requirements (Figure 2) does not exist. Compromises have to be made based on the requirements for both the materials and the robotic design (Figure 2). This paper discusses the limitations and advantages of each technique in more detail, and links them to the traditional types of elastomers and novel reversible polymer networks used. Whereas other review papers provide excellent overviews on manufacturing^[76,77] and additive manufacturing^[74,78,79] of elastomers for soft robotics, this work focuses on the advantages and opportunities that reversible covalent and supramolecular networks introduce in the manufacturing processes. These reversible chemistries used to introduce the healing ability in these elastomeric materials and to improve their reprocessability, are introduced in the following Section 1.2.

1.2. Self-Healing Elastomers

1.2.1. Classification of Self-Healing Mechanisms

In this work the focus is on self-healing polymers,^[25,80] rather than ceramics,^[81–83] or metals,^[84,85] as they are more generally used in soft robotics because of their mechanical properties. Our previous review^[20] provides insight in the progress of self-healing materials in autonomous robotics and a detailed

evaluation of the suitability of different classes of self-healing polymers for soft robotics, based on five criteria: i) the ability to heal macroscopic damage, ii) the ability to perform multiple healing cycles, iii) the recovery of initial properties after healing, iv) the ability to synthesize high strength elastomers and v) their reprocessability and recyclability. Although more details can be found in previous review,^[20] we introduce the classification of self-healing polymers in this review, to support the following sections on manufacturing of these novel reversible elastomers for soft robotics.

Self-healing polymers can be divided into different categories, either based on their healing mechanism or on the stimulus needed for healing.^[86] Two categories of healing mechanisms can be distinguished. Extrinsic healing mechanisms are achieved by incorporating into reservoirs a healing agent that is not inherent to the material structure. These reservoirs can be micro-^[87] or nanocapsules,^[88] hollow fibers,^[89] or vascular systems^[90] that are embedded in the material matrix. Upon damage, the reservoirs are broken and the healing agent leaches out to close the crack and restore the mechanical properties upon chemical reaction. The number of healing cycles is limited due to depletion of healing agent stored in the finite capsule volume or clogging of the vascular system. Moreover, the crack needs to be able to propagate and break a non-empty reservoir before the crack grows into macroscopic failure, limiting these healing mechanisms to hard, brittle materials.^[91] In general, the flexible elastomeric characteristics required for soft robotics, cannot be achieved in combination with extrinsic self-healing mechanisms. Furthermore, extrinsic self-healing materials require a complex production process that allows the incorporation of reservoirs loaded with healing agent. This makes them less suited for use in soft components.

Intrinsic self-healing polymers rely on physicochemical interactions^[92–95] or reversible covalent bonds^[36,96–98] in their chemical structure that can be broken in a reversible fashion and reformed to restore the material structure and properties, often upon the application of the adequate stimulus.^[34] Whereas the stimulus for extrinsic materials is often the

damage itself, due to consecutive breaking of the capsules or vascular system, intrinsic self-healing materials have been designed to work with a wide range of stimuli. The most common stimuli are heat^[26,99] and UV light.^[100,101] Because heat is easily available, and can be generated in various ways (electrical,^[31] IR light,^[102] inductive^[103]) it is currently the most used stimulus for self-healing soft robots. For some mechanisms, no external trigger is needed, and the healing is said to take place autonomously.^[42,93] If an external stimulus is required for healing, and this stimulus is available in the environment (e.g., mild heat, sunlight), healing can still take place autonomously. Alternatively, if the stimulus is not readily available, the healing process is referred to as non-autonomous.

An overview of intrinsic self-healing mechanisms is shown in **Figure 3**. Physicochemical interactions^[92,94,95] (Figure 3a–c), such as hydrogen bonding,^[92] ionic interactions,^[104] and metal–ligand coordination,^[105] are generally weak bonding interactions (few tens of kJ mol^{-1}) that can be easily broken and quickly reformed in a reversible fashion. Especially, hydrogen bonding interactions have attracted much attention for the synthesis of self-healing polymers due to their ability to quickly reform without the need for an external stimulus. Upon damage, the physicochemical bonds are mechanically broken, as these are the weakest bonds in the polymer. As they are reversible, these bonds are reformed when the fracture surfaces are brought back in contact and given sufficient time.

Similarly, introducing reversible covalent bonds^[36,96–98] into a polymer network structure leads to stronger cross-links that can still be broken and reformed in a reversible fashion, upon the application of an external stimulus, such as heat (for example, the thermoreversible Diels–Alder equilibrium reaction^[106]) or light (e.g., photoreversible anthracene^[107] or coumarin photodimerization^[101,108]). The reversibility of these covalent bonds either follows an associative (Figure 3d) or a dissociative (Figure 3e) mechanism. Dissociative reversible covalent bonds, as in the thermoreversible Diels–Alder equilibrium reaction,^[106,109] diaryl-bibenzofuranone,^[110] phenol-carbamate,^[111] or urea bonds,^[112] are broken upon the activation by

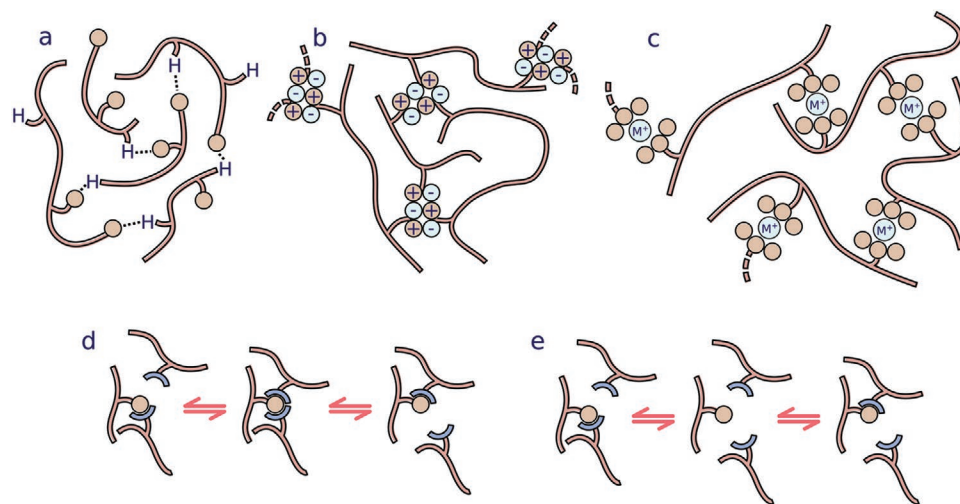


Figure 3. Overview of different intrinsic self-healing mechanisms: a–c) physico-chemical interactions; d,e) reversible covalent interactions. a) Hydrogen bonds. b) Ionomers. c) Metal–ligand. d) Associative bonds. e) Dissociative bonds. a–e) Reproduced with permission.^[20] Copyright 2021, Elsevier

the adequate stimulus. Depending on the intensity and duration of the application of the stimulus, more bonds are broken and the cross-link density of the polymer network decreases. At a certain point, the connectivity of the network structure drops below the critical gel conversion. This results in a transition from predominantly elastic to viscous flow behavior, referred to as the (reversible) gel transition. Reversible covalent polymer networks based on associative reversible covalent bonds, often referred to as vitrimers^[113–115] and employing processes or reversible bonds such as transesterification,^[116,117] disulfide bonds,^[118,119] or thiol-disulfide^[120] exchange reactions, do not show this change in connectivity. The rate at which these reversible associative bonds exchange bonding partners increases with higher stimulus intensity. Compared to physicochemical bonds, these covalent interactions have higher bond strengths (150–550 kJ mol⁻¹), leading to higher mechanical strength and stability of the networks. However, because of this higher bond strength, the healing of polymers with reversible covalent bonds is in general non-autonomous and requires an external stimulus in the form of heat or light. Nevertheless, a judicious choice of the reactive groups, tuning of the reaction kinetics and design of the connectivity in the network architecture led to self-healing polymers based on disulfide bonds^[121,122] or Diels–Alder bonds^[42] in which macroscopic damage can be healed without the need for the thermal stimulus.

The most common approach in intrinsic self-healing polymers is to use the physicochemical or reversible covalent bonds as cross-links, to construct polymer networks.^[97] Networks with a relatively low cross-link density and flexible chain segments will exhibit an elastomeric behavior, required for application in soft robotics. These intrinsic self-healing elastomers have a different thermomechanical behavior in comparison with traditional elastomers, as explained in the following section. Alternatively, intrinsic self-healing polymers can also be designed by incorporating these (physico)chemical bonds in linear polymer chains.^[123,124] This approach does not lead to a network structure, which is required to achieve elastomeric properties.

1.2.2. Classification of Elastomers

Traditionally, elastomers could be divided into two main classes. Thermoplastic elastomers^[125,126] are co-polymers that consist of soft blocks of flexible polymer chains that are physically cross-linked by hard blocks, which are immobile glassy phases (e.g., styrene block copolymers (TPS), like styrene–butadiene–styrene (SBS), styrene-ethylene-butylene-styrene (SEBS), and styrene-isoprene (SIS)) or crystalline phases (e.g., thermoplastic polyurethanes (TPU), thermoplastic polyolefins (TPO), thermoplastic polyamides (TPA), like polyether block amide (PEBA)). Between the respective glass transitions or melting transitions of the soft and hard blocks, the thermoplastic elastomers exhibit rubber-like properties. These thermoplastic elastomers can be reprocessed thermally above the highest transition temperature of the hard blocks, or by dissolution in a known solvent. The physical cross-links, the crystalline or vitrified phases, are larger compared to chemical cross-links, hence the flexibility is limited to the soft block content. As a result, Young's moduli below 1 MPa and large recoverable strains (>300 %), which are required in

some soft robotic applications, cannot be achieved without significant plastic deformation, whereas they can be reached by chemical cross-linking. Chemically cross-linked Thermosetting elastomers (e.g., natural rubber or silicones) chemically react to form an irreversible polymer network structure. Note that the term “thermosetting” is used here to differentiate between the formation of physical and permanent chemical cross-links, regardless of the glass transition temperature or the resulting properties. Although the irreversible covalent cross-links allow to synthesize hyper flexible elastomers (down to 10 kPa), they do not allow reprocessing of the permanent polymer network.

Intrinsic self-healing elastomers can be regarded as transient polymer networks and can also be divided into two main categories. Supramolecular networks^[92,94,127] are made up of macromolecules that are connected by non-covalent, physicochemical interactions. These supramolecular assemblies exhibit rubber-like behavior at ambient temperatures. Their physicochemical bonds can be easily broken and reform quickly, which makes them attractive candidates for self-healing elastomers. However, due to the weakness of the bonding interactions, these supramolecular networks are often susceptible to plastic deformation and creep. By increasing the number of bonding interactions and the polar or ionic strength of these interactions, these supramolecular networks have been made much stronger.^[128] When heating supramolecular elastomers, the dynamic character of the noncovalent bonding interactions is drastically sped up, resulting in liquid-like flow. In general, this transition is more gradual than in thermoplastic elastomers and is often already observed at much lower temperatures.^[129]

Reversible covalent networks,^[36,96,97,130] also referred to as covalent adaptable networks (CANs)^[131] or Dynamers,^[98] are created by using reversible covalent chemistries as nonpermanent chemical cross-links in the polymer network structure. These reversible covalent bonds are much stronger than the physical cross-links in thermoplastic elastomers and the physicochemical interactions in supramolecular networks. Consequently, the network is stronger and has a higher mechanical stability. Nonetheless, the reversible cross-links are weaker than the permanent covalent cross-links in thermosetting elastomers. Upon the application of the adequate stimulus, such as heat^[26,30] or light,^[132] reversible covalent networks based on dissociative cross-links (Figure 3e) can be broken down. The cross-link density of the polymer network decreases by gradually dissociating the reversible cross-links, until finally the material transitions into liquid-like behavior when the critical connectivity to form a polymer network is lost and the polymer exhibits viscous flow. This dramatic change in the viscoelastic properties and lowering of the viscosity with the continued dissociation, offers a lot of opportunities for (re)processing and manufacturing, as discussed in the next sections. The reversible solid to liquid transition is called the degelation transition and occurs at the related gelation temperature (T_{gel}). Multiple approaches exist to determine this temperature, among these, it can be defined using dynamic rheometry measurements as i) T_{gel} as the temperature at which the storage modulus equals the loss modulus (or loss angle equals 45°)^[133] or ii) T_{gel} as the temperature at which the loss angle is frequency independent.^[134–136] Dissociative reversible polymer networks combine the strength of thermosetting (permanently cross-linked) elastomers with the processability

of thermoplastic elastomers. Reversible covalent networks based on associative cross-links (Figure 3d), do not show this sharp dramatic change in viscoelastic properties. The dynamic character of the associative reaction accelerates upon stimulation with higher intensity, resulting in a slight lowering of the viscoelastic properties and increased chain mobility, without a net change in network connectivity. The topological vitrification temperature (T_v)^[116] of these vitrimers is defined as the temperature at which the material has a viscosity of 10^{12} Pas, conventionally selected as the viscosity at which the material transits from solid to a viscoelastic liquid. The associative characteristic of the cross-links results in still highly viscous liquids at elevated temperatures, often requiring high pressures to achieve sufficient flow.^[116] Self-healing elastomers exploiting such reversible cross-linking chemistries, combine the advantages of high flexibility and reprocessability with self-healing capabilities.

Reversible cross-linking chemistries, especially supramolecular assembly have also been extensively used in hydrogels, embedding an intrinsic healing capacity in these hydrophilic, swollen polymer networks.^[137] While out of the scope of this review, hydrogels are increasingly finding their way to soft robotic applications, for example, as grippers in aqueous environments^[138,139] and electronic skin.^[140] In general, hydrogels have lower mechanical strength than required in typical soft robotic applications and are less stable, as their mechanical properties depend highly on the humidity of the environment in which they operate.

Figure 4 gives an overview of multiple self-healing polymers found in literature, and classified per (overarching)

thermoreversible healing mechanism, with high potential for adoption in soft robotics, as discussed in more detail in our previous review paper.^[20] For these materials, the required healing temperature was plotted as function of the ultimate stress and strain, measured in tensile testing until fracture. First, it can be seen that the polymers with the highest ultimate stress, also require the highest healing temperatures. This higher ultimate stress is a consequence of the stronger reversible bonds (bond energy $150\text{--}550\text{ kJ mol}^{-1}$), and the resulting tougher polymer network structures. More energy, and hence a higher temperature, is required to break these bonds in a reversible fashion to establish the necessary chain segmental mobility and reactivity required for successful damage healing.^[20] Supramolecular elastomers such as hydrogen-based polymer networks often have a high ultimate strain, but a low ultimate stress. Due to the limited bond strength (bond energy of several tens of kJ mol^{-1}) and fast (re)formation of these physicochemical interactions, they are often able to heal at room temperature. However, these materials often suffer from non-negligible creep behavior, which can be a disadvantage for soft robotics. This is a clear trade-off, and the material choice depends on the application. For soft robots applications where only low forces are required, low strength autonomous self-healing polymers can be a good option. Alternatively, if the robot should perform a heavier task, for example, lifting heavier objects, higher strength self-healing polymers are preferred, at the cost of needing a higher healing temperature, and as a consequence a stimulus providing system (e.g., an external or internal heater) should be added to the system, increasing its complexity.

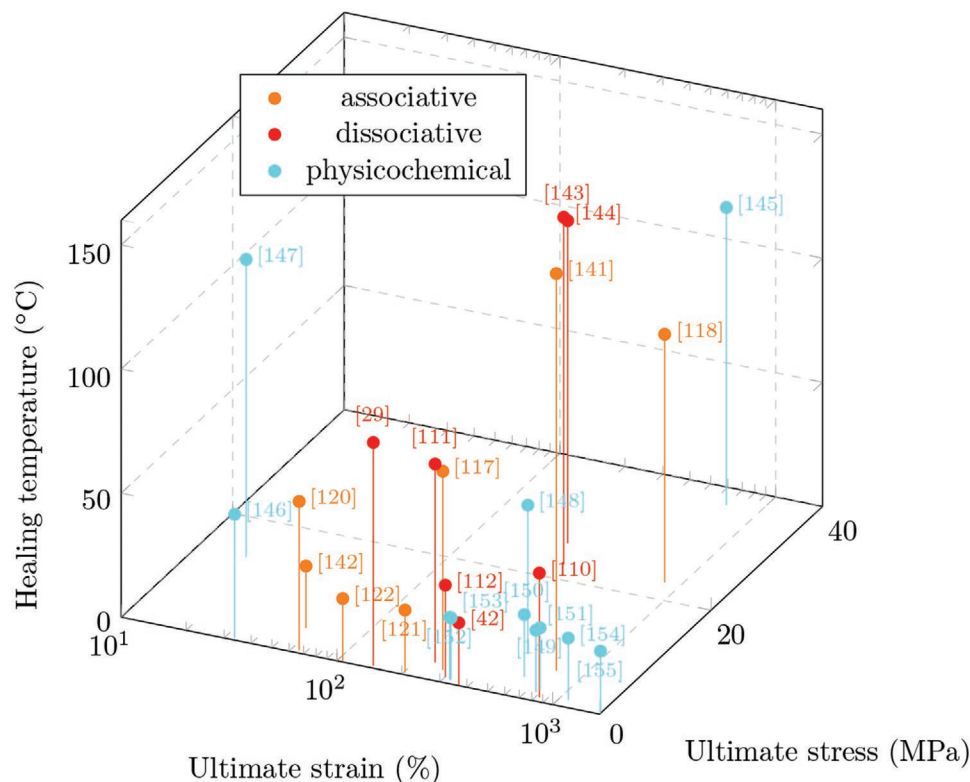


Figure 4. Overview of the properties of different types of self-healing polymers found in literature. The graph is based on data presented in our previous review paper.^[20]

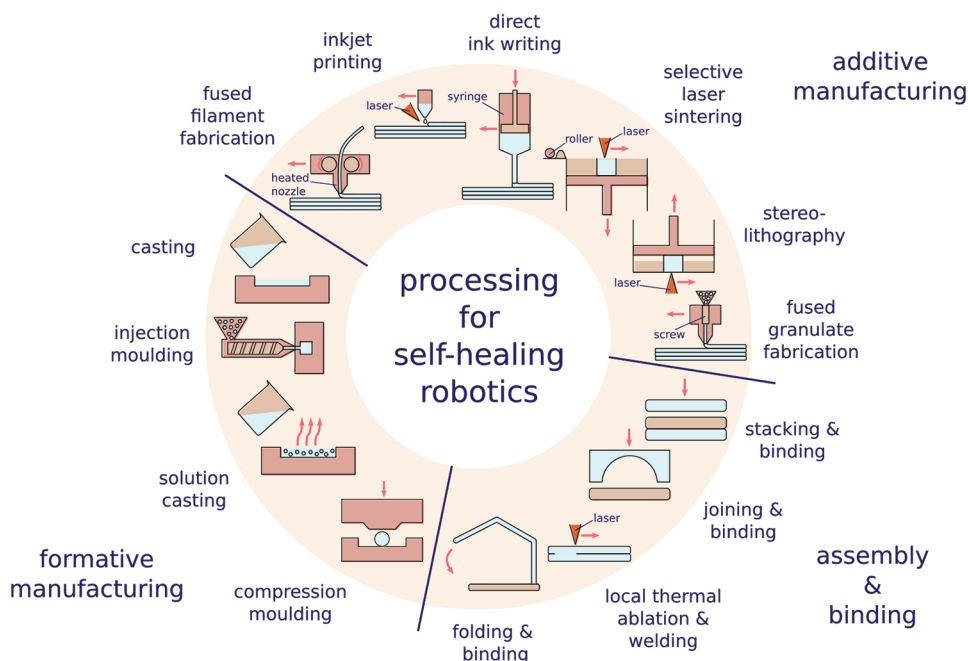


Figure 5. Different techniques are used for the processing of self-healing robotics. In this review, they are split in three categories: formative manufacturing, assembly and binding, and additive manufacturing. For each technique the (dis-)advantages are discussed, together with the opportunities for self-healing soft robots. Images are reproduced with permission.^[141] Copyright 2021, Ellen Roels.

It is important to mention that nonautonomous healing is possible in some thermoplastic elastomers without additional integrated reversible covalent or physicochemical bonds. Upon heating, their physical cross-links (hard phases) can be reversibly mobilized (by melting or heating above the glass transition temperature), while upon cooling, the hard, physical cross-links are reformed by crystallization or vitrification. This thermoreversibility allows healing damage by performing a heat-cool cycle. The main challenge of this self-healing mechanisms is to heat the polymer sufficiently to achieve the mobility needed for healing, while not losing structural stability. As the temperature window of the melting/glass transition is often very narrow, most thermoplastic elastomers lose most of their structural stability upon heating, in contrast to reversible covalent and supra-molecular polymer networks that heal damage in their solid state. For this reason, thermoplastic elastomers are not considered as self-healing elastomers.

2. Manufacturing Techniques for Healable Soft Robots

As it is very challenging and in many cases even impossible to machine elastomers, for example, by milling, turning and drilling, most soft robots are produced by formative manufacturing, mainly through casting, but also via solvent casting, compression and injection molding (IM). Recent developments in additive manufacturing of flexible materials, where the object is printed layer by layer, have led to printing of soft robotics using various techniques, including direct ink writing, fused filament fabrication, fused granular fabrication, selective laser sintering (SLS), and photo-resin printing, for example, stereolithography, and inkjet printing. Although these

manufacturing techniques allow to manufacture complex soft robotics, all of them have restrictions and disadvantages that limit the design freedom and/or performance of the produced components. Based on literature, many can be solved by using reversible elastomers, containing reversible cross-links, in the manufacturing process rather than traditional thermosetting or thermoplastic elastomers. In addition, the reversible polymer networks offer the advantage of intrinsic self-healing properties. This is of high importance in soft robots that are in general highly susceptible to different damage types, including damage by sharp objects, overloads, interfacial debonding, and fatigue. For each formative or additive manufacturing technique a brief introduction will be given that addresses its advantages and limitations. Next, for each manufacturing method (Figure 5), it is elaborated how the use of reversible, self-healing elastomers overcomes certain obstacles and limitations, and opens up new opportunities for higher performance and more robust designs, and in general, to meet the previously listed processing requirements for soft robotics.

3. Formative Manufacturing

Formative manufacturing techniques are extensively used to produce soft robotics.^[17] This is in contrast with traditional stiff robots. These processing techniques include casting, solvent casting, compression molding, and IM.

3.1. Casting

Casting is performed by pouring liquid monomers or (pre) polymers into a mold and letting them solidify through

polymerization or cooling.^[58] Casting-based techniques are the most widely used for manufacturing soft robotics, because they often only require easily available tools, and there exist a lot of commercially available elastomers. Silicones as EcoFlex or DragonSkin (Smooth-On, Inc.), are examples of the most commonly used ones in the field.^[52,145] These thermosetting elastomers are irreversibly cross-linked during polymerization. Most commercially available thermosetting elastomers that are designed for casting, such as the ones mentioned before, consist of one or two reactive components and a catalyst. These components are mixed, preferably degassed under vacuum to remove air bubbles from the mixing process, cast in a mold, and cured (polymerized and/or chemically cross-linked). The curing can take place at room temperature or can be accelerated at higher temperatures in an oven. An advantage of casting is that the internal stresses induced by the manufacturing technique after curing are very low. However, when mixing highly viscous monomers or prepolymers, bubbles can be trapped in the part which can lead to unexpected and unpredictable failure, for example because of leaks in thin-walled pneumatic actuators.^[3,146]

For simple gripper designs, such as the DHAS finger with FinRay effect from Festo,^[18] a single stage casting process is a very efficient way of manufacturing. For more complex designs, such as pneumatic actuators with an internal cavity, this is not possible, and a multi-stage casting process is necessary.^[147] Such a multi-stage casting process is also necessary when combining multiple materials in a single object, which is getting more and more traction in soft robotics^[52,148] as it can improve the actuator's performance.^[52,149,150] The introduced anisotropic deformation response can lead to more complex embodied intelligent behaviors.^[51] In addition, combining less flexible or even stiff materials with flexible membranes in articulated soft robots, allows to increase the force output.^[151] However, multistage casting is time-consuming^[152] and introduces weak (multi-material) interfaces in the actuators, which rely mostly on weak physical interactions. These interfaces are usually broken after a relatively low number of actuation cycles and lead to failure by interfacial debonding and delamination.^[146]

This problem can be addressed by designing soft robotic actuators and their casting procedure, such that these weak interfaces are present in regions that will not undergo large stresses. In addition, scientists try to enhance interfacial strength in multi-stage casting, by casting the mixture of prepolymer on top of a part that was prepared in a previous casting step and that was demolded before being fully cured.^[153] Others propose to use mechanical interlocking at (multi-)material interfaces for improved interface strength and robustness.^[153–155] However, both approaches strongly limit the design freedom for future soft robotics applications. Alternatively, investment casting uses a wax inner mold of the cavities that is melted out after demolding and thus allows for single stage casting of hollow structures.^[156] Such hollow structures can also be made via (multi-)axis rotational casting that exploits the centrifugal effect to deposit a thin layer of elastomer on the walls of the mold.^[157,158] Although the above techniques permit to manufacture robust hollow structures, they cannot be used to create strong multi-material components.

Using self-healing polymers, robust multi-material components for soft robotics can be cast. Some intrinsic self-healing polymers can be produced via the traditional casting techniques when the monomers are liquid and miscible.^[99] In addition, casting of thermoreversible networks can be performed at temperatures above their gel transition temperature T_{gel} , where sufficient dissociative covalent bonds are broken thermally^[29] to ensure a sufficiently low viscosity. In comparison to thermosetting elastomers, self-healing elastomeric parts, produced in different casting steps can be joined together by strong interfaces that rely on strong reversible chemical bonding, often achieved via a heat-cool post-treatment. This was illustrated by Roels et al.,^[29,39] showing that the interface between two reversible covalent network elastomers, with different mechanical properties, is nearly perfect after fusion via a heat-cool cycle. It was shown that the interface was at least as strong as the weakest of the two materials. This merging principle, possible with thermoreversible self-healing elastomers, grants to create robust (multi-material) components using multi-stage casting, making it possible to create complex (hollow) structures for future soft robotic applications. Self-healing composite parts, produced by adding fillers to a self-healing elastomer matrix, can also be joined to parts made out of the pure self-healing elastomer matrix, as demonstrated by Narumi et al.^[159] They combined various cast pieces with and without conductive carbon nanotubes, an electrically conductive filler, to create a robust self-healing sensor. Multi-stage casting with self-healing elastomers could also be performed by casting the prepolymer onto a fully cured part that was produced in a previous casting step. In this case, it is recommended to perform a post-treatment that involves a heat-cool cycle, to ensure a robust part due to the formation of reversible cross-links along the interface. It is clear that the reversible nature of the cross-links in self-healing polymers provides many new opportunities for casting robust (multi-material) soft robots.

3.2. Compression Molding

When heating intrinsic self-healing polymers, containing reversible (physico)chemical cross-links, above their gel transition or topology freezing temperatures, the resulting polymer melt often has a high viscosity. This restricts flow and consequently these polymers can often not be cast under atmospheric conditions. However, by applying pressure during compression molding, the viscous polymer can be shaped into the cavity of a hot mold. Upon cooling, the thermoreversible network is reformed and the polymer solidifies into the desired shape. In material science, this technique is often used to prepare samples for mechanical testing or to illustrate reprocessing of reversible elastomers by heat treatment, for example, for reversible covalent elastomers^[111,122] or supramolecular elastomers.^[160] This technique can also be used to shape a self-healing elastomer during polymerization, on the condition that its monomers are miscible, as illustrated by Zhu et al.^[161] Compression molding is in particular popular for vitrimeric systems, as the associative nature of their reversible cross-links prevents low viscosities at high temperatures.^[162,163] Although less common than casting, compression molding is often used

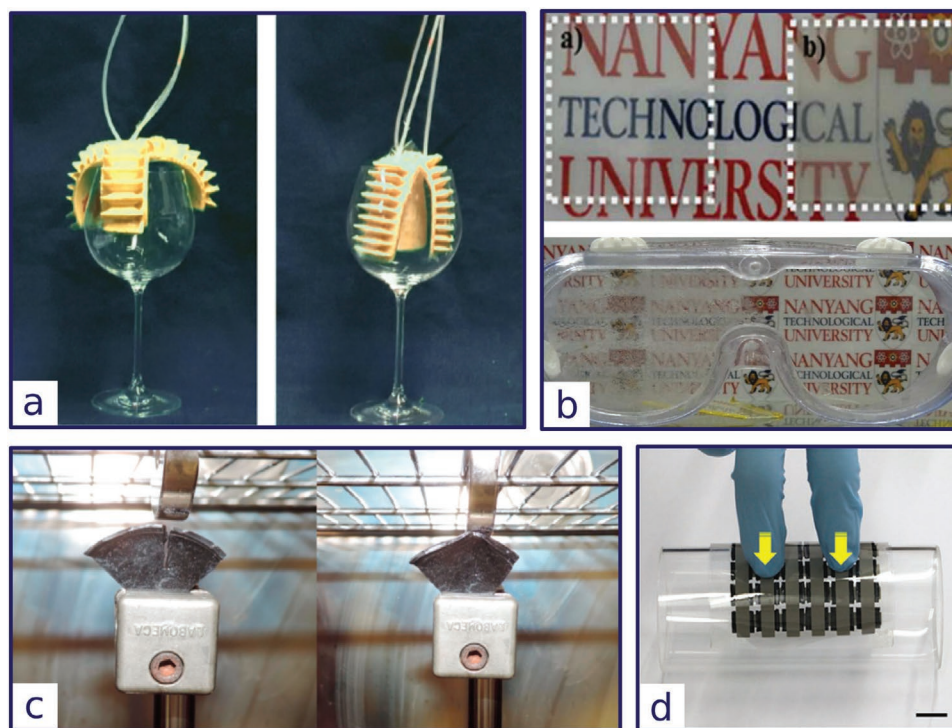


Figure 6. Self-healing soft materials and robots made using formative techniques. a) EcoFlex/Kevlar self-sealing actuator. Reproduced with permission.^[27] Copyright 2013, Wiley-VCH. b) Flexible and transparent heater based on a Diels–Alder network spray coated with silver nanowires. Reproduced with permission.^[42] Copyright 2017, Royal Society of Chemistry. c) Magnetic fillers allow an external driving force to close large damages. The Diels–Alder material is solvent cast, but getting a good dispersion of the particles is challenging. Reproduced with permission.^[43] Copyright 2020, Elsevier. d) Solvent cast self-healing pressure sensor array. Using an intrinsic conductive elastomer avoids the particle dispersion issue. Reproduced with permission.^[44] Copyright 2018, Wiley-VCH.

to manufacture pneumatic actuators both on the millimeter-scale^[164] and cm-scale^[27] (Figure 6a). The damage resilient pneumatic actuator by Shepherd et al. was developed via cold compression molding of silicone while embedding polyaramid (Kevlar) fibers and was one of the first reports on damage resilient soft robotics. It shows a self-sealing effect attributed to the composite of silicone and polyaramid fibers.^[27]

Although compression molding is a very promising technique to develop healable soft robotics from intrinsic self-healing elastomers, it is not yet widely exploited. The technique was used to mold a hard, brittle self-healing thermoset by Terryn et al.^[165] to create a sacrificial mechanical fuse. The fuse was molded in two parts using compression molding of a reversible covalent network based on the thermoreversible Diels–Alder reaction. The two parts of the fuse were joined together upon heating to act as a sacrificial element, breaking at a designed force and protecting more vital parts of a stiff robotic actuator. Miyake et al. took a similar approach by creating a fuse^[166] for a linear actuator using a thermoplastic polymer. While thermoplastic polymers are not generally regarded as self-healing materials themselves, the concept of using the thermoplastic fuse and a controlled thermal trigger does also result in a self-healing system concept.

Moreover, compression molding is a better technique compared to (solvent) casting when working with composites, as shown by Fox et al.^[167] Due to the typical higher viscosity and the limited flow of the material, filler sedimentation is avoided and filler dispersion is influenced less. As for casting, making

hollow structures using compression molding alone is not possible. However, when using intrinsic self-healing elastomers, two parts produced via separate molding steps can be joined together, for example, via a heat–cool cycle.

3.3. Injection Molding

In IM, a viscous polymer melt is injected through a small inlet (gate) into the cavity of a closed mold. For elastomers, two types of IM can be distinguished: reaction IM (also known as liquid IM) for thermosetting elastomers, and thermoplastic IM for thermoplastic elastomers. During reaction IM, one or multiple liquid monomers or prepolymers are injected and cured in the mold. For thermoplastic IM, pellets or polymer granulates are fed into a heated extruder, where they are heated, melted and mixed into a homogeneous polymer melt. This viscous melt is injected into the mold, where it solidifies as it cools down before being demolded. Usually, the polymer is fed and pressurized by a screw. The technique requires the use of more extensive equipment to control the temperature and injection pressure, which makes it not a preferred or widely used processing technique for small scale research prototypes. However, some reports were found on the academic use of this technique for the manufacturing of soft robots. Li et al. used a commercially available SBS block copolymer that is flexible, soft, and optimized for plastic IM, to manufacture strain

limiting pneumatic actuators.^[168] Bell et al. used reaction IM for manufacturing their dome-shaped robots out of commercial silicone.^[169] IM allows for the small-scale features required in their design that are not possible to be manufactured using casting, as the viscosity of the prepolymer was too high. Besides, IM allows to directly integrate flexible sensors, as illustrated by Georgopoulou et al. They incorporated a piezoresistive sensor fiber into a silicone elastomer matrix using reaction IM of a two-component silicone rubber into a mold that held the sensor fiber in place.^[170]

Although not yet exploited for manufacturing healable soft robots, IM is promising for the production of components out of intrinsic self-healing elastomers. This is particularly true for reversible covalent networks based on exchange reactions (vitrimers) that exhibit high viscosities and require high pressures during processing due to the associative nature of the reversible crosslinks.^[116] The dynamic exchange (Figure 3d) of the reversible crosslinks is sped up by increasing the temperature, which allows the material to flow, notwithstanding the network connectivity. However, the change of their viscoelastic properties with temperature is quite slow. As the network structure remains present at high temperatures, the viscosity of the polymer remains high and, consequently, most vitrimeric elastomers can be processed only using high pressure. This makes (reaction) IM and compression molding especially useful for vitrimers. IM is also promising to scale up part manufacturing for dissociative reversible elastomers, both during the first polymerization and for reprocessing. Furthermore, the dynamically reversible nature of the cross-links in self-healing polymers allows to relax the stresses induced by the IM procedure. In thermosetting elastomers, this could lead to warping (undesired deformation) upon demolding or to weak spots that over time lead to failure.

IM is most suited for large scale production, because the cost of the IM equipment and the cost to make a mold is high. As such, this technique will be exploited when healable soft robots will be industrialized and commercialized. For large batch production, this technique is economical, fast, and consistent, producing high quality components with smooth surface finish. Manufacturing objects out of multiple materials is challenging using IM. There exists specialized equipment for coinjection of multiple polymers inside a mold, resulting in parts with a core of one material encapsulated within an outer layer formed by the second polymer. Consequently, the design freedom of multi-material parts using this technique is very limited.^[171] Nevertheless, IM offers a lot of potential to produce self-healing composite parts: it is often used to produce polymer composites, as the injection screw is used to mix and disperse fillers inside the polymer matrix or blend different polymers or grades.^[172]

Similar to compression molding, making hollow structures in one step is not possible with a normal extrusion-IM setup. However, two molded parts of thermoreversible polymer networks can be welded by a heat-cool cycle, exploiting their thermally reversible nature. Injection blow molding (Figure 7a)^[173] can offer a solution, as a specialized IM technique to produce hollow structures, however the extensional rheology and melt strength it requires make its implementation for self-healing elastomers not straightforward. Negshell casting^[174] offers another solution, by using sacrificial cores that are broken after demolding. The broken pieces remain in the finished part, yet

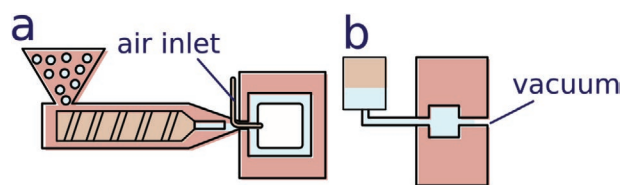


Figure 7. Besides reaction IM (for thermosetting elastomers) and thermoplastic IM (for thermoplastic elastomers), other types of IM exist. a) blow molding for hollow structures. b) Vacuum-assisted resin transfer molding. Images are reproduced with permission.^[141] Copyright 2021, Ellen Roels.

allow movement of a pneumatic actuator. When the viscosity of the monomers is low, vacuum-assisted resin transfer molding can be used (Figure 7b). In this technique a low viscosity mixture is drawn into a closed mold through an inlet by bringing the cavity under vacuum. Park et al.^[99] used vacuum-assisted resin transfer molding to manufacture laminate self-healing composites by putting a closed mold containing carbon fibers under vacuum and injecting a Diels-Alder polymer. Due to the low viscosity of the mixture, static mixers, like the counter flow mixer used by Park et al.^[99] can be used instead of an active screw. Although only applicable to a more limited number of materials, vacuum-assisted resin transfer molding can provide a cheaper solution for producing healable soft robots in academia.

3.4. Solvent Casting

For many self-healing elastomers, the monomers are not miscible, highly viscous or solid particles at ambient temperature. An alternative to melt mixing, including extrusion, is to dissolve them in an appropriate solvent. Subsequently, self-healing parts can be created through solvent casting in a mold and solvent removal. The cast mixture polymerizes in the mold, similar to normal casting methods. Depending on the solvent, this solvent casting is performed at atmospheric conditions or under vacuum, at low or high temperatures. This technique gives the best results when the molded part is thin and has a large surface area to evaporate the solvent. In research, solvent casting is typically used to manufacture sheets out of which test samples can be cut.^[175–177] The same principle is also used for spin coating or drop casting.^[178–180]

Solvent casting is ideal to produce thin structures and robots.^[181] As such, a lot of self-healing sensors, which are typically thin structures, have been manufactured using this technique. In addition, solvent casting allows to disperse fillers (e.g., conductive or magnetic particles) in the monomer solution to yield self-healing composites. Conductive self-healing composites are used to produce self-healing flexible electronics,^[182,183] including soft sensors^[63] and flexible heaters^[142] (Figure 6b), which find applications in soft robotics^[24] and wearable sensing applications.^[184] Magnetic composites allow for an additional driving force to close damage (Figure 6c), or to manufacture magnetic sensors. However, as pointed out by Cerdan Gomez et al., it should be taken into account that for self-healing composites, the dispersion of fillers might not be optimal due to aggregation and sedimentation of the fillers during casting.^[143] Wang et al.^[144] used solvent casting without

fillers with a self-healing network based on dynamic hydrogen bonding and electrostatic interactions that is inherently conductive because of polyaniline in the network, to make self-healing bending and pressure sensors that have potential for the implementation in soft robotic applications (see Figure 6d).

4. (Re-)assembly and Binding

As briefly mentioned in the previous sections, self-healing parts manufactured out of reversible networks, can be bound together via strong (physico)chemical bonds by subjecting the parts to a heat-cool cycle after bringing them in contact. This joining procedure can be performed for parts composed of the same reversible networks but made in a different manufacturing step, or for parts composed of reversible polymers with different mechanical properties but identical reversible cross-links in their networks. For this method, noted as “assembly and binding,” three subclasses were defined depending on the assembly type and the use of global or local heating: stacking/joining and binding,

folding, and covalently binding, and local thermal ablation and welding. The first two are performed in the solid state, (well) below the transition temperatures (T_{gel} or T_V) of the involved reversible polymers, thus ensuring the mechanical stability of the components. This type of manufacturing is specific to intrinsic self-healing elastomers and cannot be performed using thermosetting elastomers, as they are not reprocessible. Nor can these techniques be used for thermoplastic elastomers. In order to bind, these networks need to be heated above their T_m or T_g , where they lose structural stability. “Local thermal ablation and welding” follows the same principle, but in this case parts are joined by raising the temperature locally only. As heating takes place at a specific location, the elastomer can be heated above its transition temperature, without the risk of losing mechanical stability throughout the parts being joined, while effectively fusing them together locally. As such, these local thermal ablation and welding techniques can be applied both on self-healing elastomers and thermoplastic elastomers. As described more in detail in the following sections, these assembly and binding techniques permit to create robust multi-material components

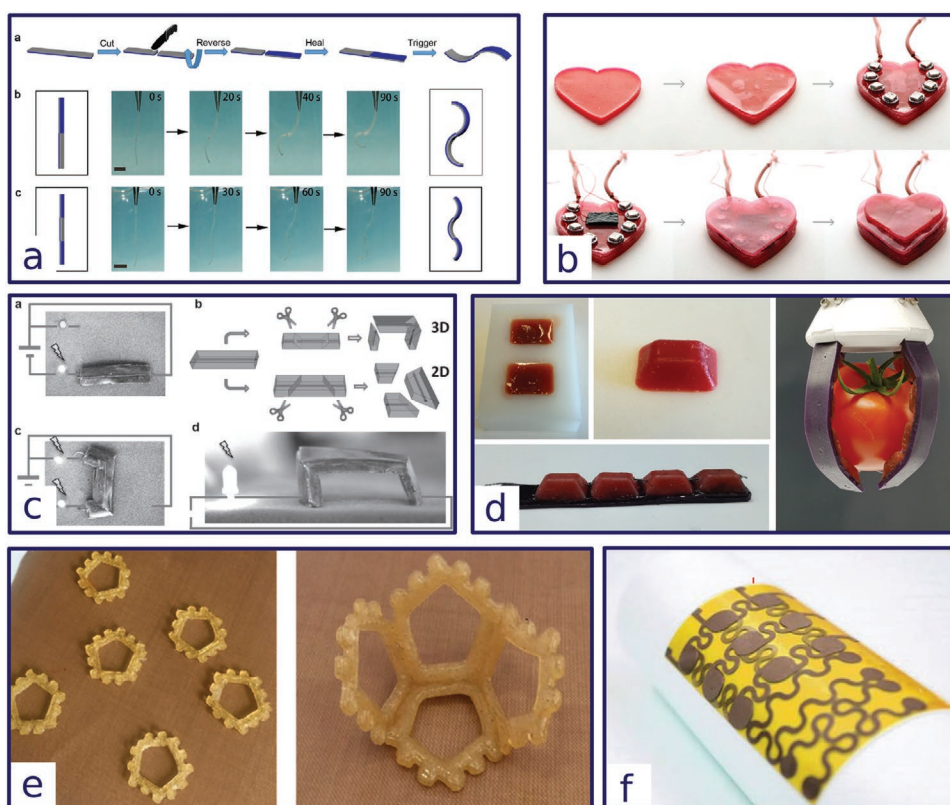


Figure 8. (Re-)assembly and binding is a popular method to manufacture complex self-healing soft robots. a) Reconfigurable actuator that responds to a chemical stimulus (chloroform), manufactured by stacking and joining two layers together. Reproduced with permission.^[185] Copyright 2021, Wiley-VCH. b) Heart with LEDs that can light up, incorporating a conductive self-healing polymer. Healing takes around 6 h at room temperature. Reproduced with permission.^[159] Copyright 2019, ACM Press. c) A reconfigurable conductive circuit based on liquid metal (EGaIn). Reproduced with permission.^[186] Copyright 2013, Wiley-VCH. d) Multi-material gripper out of Diels–Alder polymers. Cast phalanges are joined to a solvent-cast backbone. Reproduced with permission.^[29] Copyright 2019, IEEE. e) 3D structure obtained after joining different objects that are 3D-printed using DIW. Reproduced with permission.^[187] Copyright 2020, American Chemical Society. f) Recyclable robotic skin that incorporates a tactile sensor, flow sensor, humidity sensor, and temperature sensor. Reproduced with permission.^[188] Copyright 2018, The Authors, published by AAAS. Reprinted/adapted from ref. [188]. © The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC) <http://creativecommons.org/licenses/by-nc/4.0/>.

(Figure 8b–d) and hollow structures. In addition, it opens a lot of opportunities for reconfigurable soft robotic systems, able to change their morphology via reassembly and binding.

4.1. Stacking/Joining and Binding

Thanks to the reversible nature of intrinsic self-healing elastomers, the films produced using solvent casting or other processing methods can be further processed using the adequate stimulus, which is in most cases heat, as many intrinsic self-healing elastomers are thermoreversible. By stacking sheets or joining objects (Figure 8e), and performing a heat–cool cycle below the transition temperature (T_{gel} or T_v), the sheets are merged and bound together via (physico)chemical bonds. This way, robust 3D objects can be generated. This allows fabrication of soft robotic actuators, as presented by He et al.,^[189] who developed self-repairable fluid-driven liquid crystal elastomer actuators by stacking multiple layers of disulfide-based self-healing elastomer sheets with complex shapes made via laser cutting. A similar joining technique is shown by Cao et al.,^[190] who developed humidity controlled actuated hygroscopic robots by welding hydrogen-bonding based polymer network parts using a heat–cool cycle. In extension, the stacking method can be used to stack materials with different mechanical properties or fillers, as long as the elastomers or composites contain the same reversible cross-links. For example, by stacking self-healing elastomer and conductive composite sheets, the composite sheet is electrically insulated and healable flexible sensors^[191] or circuits^[186] (Figure 8c) can be generated or even directly integrated in soft robotic bodies.

4.2. Folding and Binding

Aside from stacking, Terryn et al.^[26] developed a technique, referred to as folding and covalently binding. It allows to create airtight and hollow 3D structures out of self-healing Diels–Alder-based elastomer sheets by subsequently folding and exposing the part to a heat–cool cycle (remaining below T_{gel}). This shaping method is illustrated in Figure 9a, and was used to create multiple healable soft robotic parts, including a soft hand,^[39,42] soft gripper,^[26] and artificial muscles.^[26] Deng et al.,^[191] showed that this technique is applicable to vitrimers

as well (Figure 9b). In their 3D flexible structures, they directly integrated healable flexible sensors based on a disulfide-based vitrimeric elastomer matrix filled with silver nanowires. In both stacking and folding and covalently binding techniques, the polymer is heated but remains below its transition temperature (T_{gel} or T_v), illustrating that self-healing polymer structures can be manufactured in solid state. Of course, the (thermo)reversible nature of the self-healing elastomers permits to reshape sheets formed by solvent casting using many other techniques in which the polymers are heated above their transition temperatures (T_{gel} or T_v), as described elsewhere in this review. In the future, it is estimated that solvent casting will continue to be used for the synthesis of elastomeric sheets, produced from nonmiscible reagents. These sheets will subsequently be processed using other manufacturing techniques to produce healable soft robotic structures. In the near future, this principle can be combined with additive folding techniques, like the one described by Yim et al.,^[192] in which 2D slices are threaded by multiple strings, accordion-folded by flexure hinges, and finally stacked into a predefined 3D structure.

4.3. Local Thermal Ablation and Welding

In contrast to heating the entire system, self-healing parts can also be bound using a local thermal treatment, for example, by irradiating it with a laser. The focused light of the laser beam locally heats the material, and depending on the intensity, the material is either ablated,^[193] cut through,^[152,188] or can be welded.^[194] When cutting a part made of intrinsic self-healing polymer, mechanically or thermally (using a laser), and when breaking almost exclusively reversible (physico)chemical bonds, the cut pieces can be rejoined via a heat–cool cycle, as illustrated by Zou et al. in Figure 8f.^[188] Alternatively, the parts could be joined by local heating using a laser. This reversible welding opens up possibilities for reassembly (Section 4.4). As this technique only heats locally, it can not only be used for intrinsic self-healing elastomers, but also to weld thermoplastic elastomers. The major part of the thermoplastic elastomers will remain solid and mechanically stable, as the material is only liquefied locally. Alternatively, thin sheets can be welded together using a localized heat source mounted on a CNC machine, such as a soldering iron^[195] or a heated extruder head^[196] Other

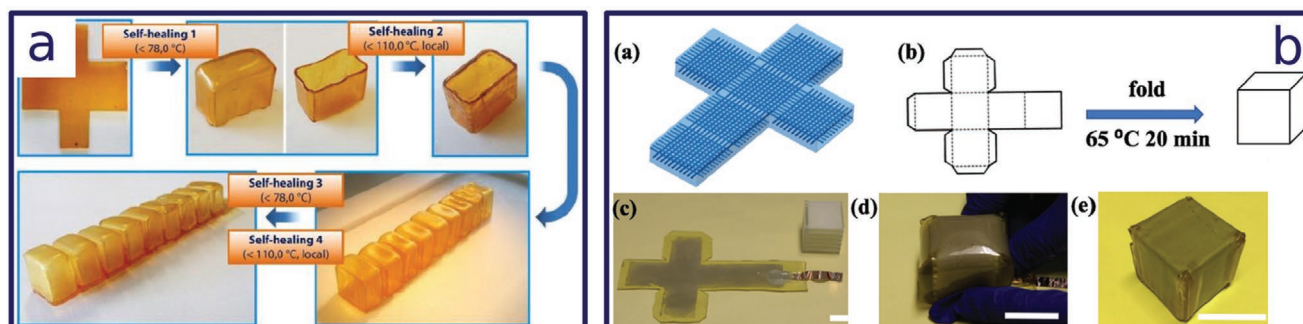


Figure 9. Folding and binding can be used to form 3D objects out of a 2D sheet. a) It can be used to manufacture pneumatic actuators using a Diels–Alder elastomer. Reproduced with permission.^[26] Copyright 2017, The Authors, published by AAAS. b) Or to make a vitrimer based triboelectric nanogenerator that can power wearables. Reproduced with permission.^[191] Copyright 2018, Wiley-VCH.

localized heat sources include impulse sealing^[197,198] and heat stamping.^[199]

These local thermal ablation and welding techniques are fast, precise and repeatable. However, it is in many cases difficult to focus the temperature and, consequently, downscaling these techniques will be challenging. Moreover, only thin sheets can be used as the laser beam is absorbed in the top layers and/or the high heat capacity and low thermal conductivity of the polymer material prevents the heat from penetrating deep into the material. Nonetheless, fusing or welding thin sheets together is a popular method for making thin structure soft robotics, including inflatable pouch actuators, as the technique ensures that they are airtight. These actuators are often soft pneumatic muscles that can be combined into more complex actuators or modules.^[198,200] Another example is the thin-walled, growing soft pneumatic robot by Hawkes et al.^[201] that can be used in constrained or challenging environments. Amiri Moghadam et al.^[194] used a lasercutter to cut and weld TPU films to fabricate a pneumatic robot in a single manufacturing step. Another approach is used by Wehner et al.^[202] and Kim et al.^[203] who used a laser to trim excess material as a post-processing step.

4.4. Reassembly of Modular Systems

Recently there is a rise in popularity of modular^[204] and self-reconfigurable robotic systems.^[205,206] These approaches will facilitate self-repair in robotic systems^[207] by replacing failed modules with new ones completely autonomously. In addition, reversible (physico)chemistries permit to join modules together reversibly via heat-cool cycles. Currently, there exist many coupling mechanisms for modular robotics,^[208] relying on mechanical or magnetic^[209] coupling. So far, modules made from irreversible networks have not been connected via chemical bonding, as the system cannot be disassembled. However, with reversible (physico)chemical bonds these couplings can be made reversibly and modules can be separated from the system by mechanical force, for example, cutting. Lou et al.^[185] used the stacking technique (Section 4.1) to manufacture reconfigurable bilayer actuator modules for soft robotics, composed of two layers of elastomers based on reversible poly(dimethylglyoxime-urethane) and hydrogen bond cross-links that differ in cross-link density (Figure 8a). The bilayer actuator responds to a solvent stimulus (chloroform) due to the difference in the cross-linking densities of the two layers. It can be reconfigured by cutting the reversible network mechanically, bringing it back into contact in a new configuration, and leaving it to rebind. Chemical couplings are interesting as they are compact, compared to magnetic and certainly mechanical couplings. If they rely on reversible covalent interactions, the couplings can be made relatively strong.

5. Additive Manufacturing

Recently, the demands for fast prototyping and high geometrical complexity in soft robotics have led to a fast increase in the use of various additive manufacturing (AM) techniques to print soft robotic structures layer by layer.^[73,78,210,211] Although

self-healing soft robotics is a relatively new subfield, there have been some reports on the use of AM to produce healable soft robots from intrinsic self-healing elastomers (Figure 10), which will be introduced in this section. As was the case for molding techniques, the reversibility of the (physico)chemical bonds in these self-healing networks opens up new possibilities and solves problems that occur when using traditional thermosetting and thermoplastic elastomers.

5.1. Additive Manufacturing via Ink Printing

5.1.1. Direct Ink Writing

Direct ink writing (DIW) is an additive manufacturing technique where liquid monomers or prepolymer, referred to as ink, is extruded through a syringe or nozzle and deposited on the print bed,^[216] where the ink polymerizes and solidifies. The solidification mechanism can vary: it can be the polymerization of a reactive mixture^[217,218] or the solidification of a dissolved polymer through solvent evaporation.^[219] The use of an external stimulus, for example, heat, is in some cases needed to achieve solidification in a short time relative to the printing speed.^[220] Slow solidification, due to slow reaction kinetics, leads to very low printing speeds and mechanical instability of the print. When fast solidification is not available during printing, the object can be post-treated at the cost of design limitations (e.g., no overhang possible).^[212,221] Therefore, the accelerating stimulus is commonly made available in the printing environment. This could be an increased temperature to speed up the curing reaction or evaporation,^[222] or a UV light source.^[223] Nonetheless, DIW is a popular technique used to manufacture soft robots,^[78,139,224] sensors,^[225,226] and heaters.^[71] However, using conventional chemically cross-linked elastomers the network is irreversibly formed upon polymerization on the print bed. These irreversible cross-links prevent the printed parts from being reshaped, reprocessed and recycled.

Intrinsic self-healing polymers can again provide a solution to this non-ecological situation. DIW is already used to produce healable soft robotic components. Yuan et al.^[212] mixed a reactive trifunctional furan compound with bismaleimide and used this mixture to print Diels–Alder polymer structures. Damage in the centimeter scale was healed with healing efficiencies of 80 % (Figure 10b). For printing, the liquid mixture is heated above the transition temperature of the produced polymer to prevent undesired solidification in the piston and/or syringe, in this case above its T_{gel} , at 120°C. The produced components are healable, but also reprocessable. DIW with thermoreversible elastomers allows also to start from solid elastomeric material, which is degelled upon heating in the piston. This principle is presented by Yang et al.,^[227] by heating solid Diels–Alder network particles above their T_{gel} , after which the viscous material was pushed through the nozzle and deposited on a heated print bed. The print bed is often heated, remaining below T_{gel} , as higher temperatures lead to faster kinetics of the Diels–Alder reaction, resulting in faster solidification of the printed layers. Recently, DIW has become a popular option for self-healing thermosets,^[212] elastomers,^[187,228] and hydrogels.^[33,229,230]

A disadvantage DIW shares with fused filament fabrication (FFF, Section 5.2.1), is its rough surface finish due to the layer

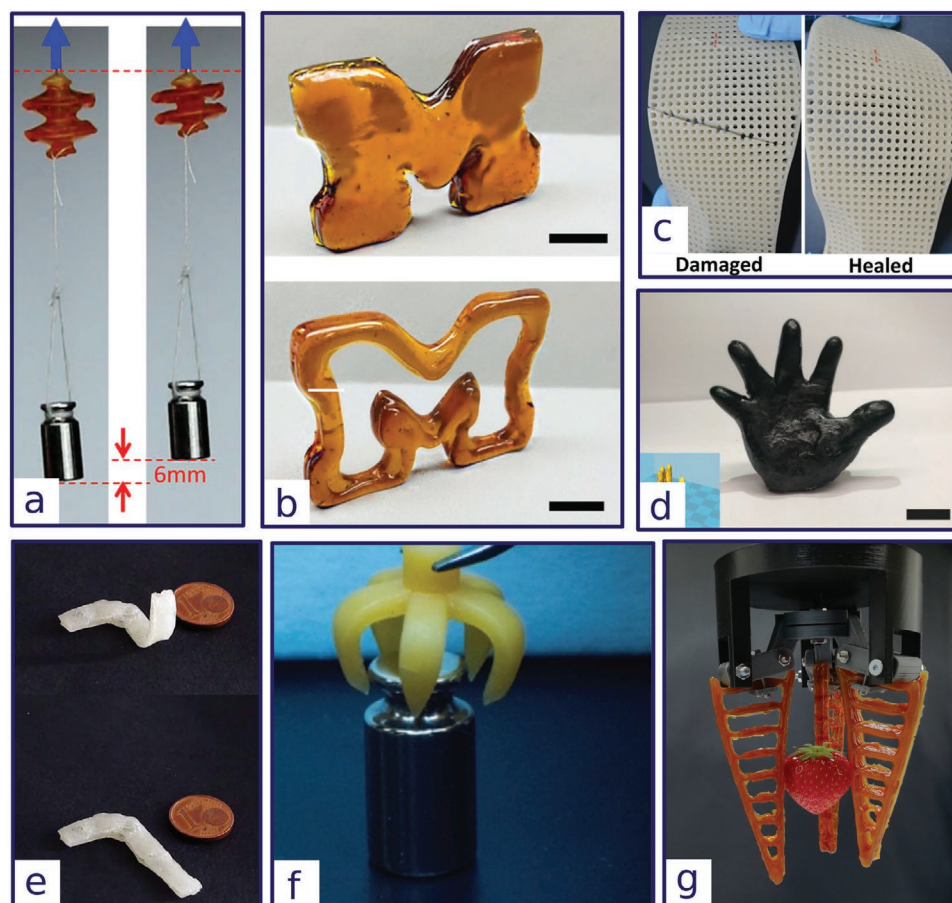


Figure 10. Self-healing polymers can be processed using many types of additive manufacturing techniques. a) Soft pneumatic muscle, based on PDMS, printed using SLA/DLP. Adapted under the terms of the CC-BY Creative Commons Attribution 4.0 International license (<https://creativecommons.org/licenses/by/4.0>).^[38] Copyright 2019, The Authors, published by Springer Nature. b) Diels–Alder thermoset printed using DIW. The objects are postcured overnight. Reproduced with permission.^[212] Copyright 2020, Royal Society of Chemistry. c) A self-healing orthotic insole printed using SLS. Reproduced with permission.^[213] Copyright 2021, Elsevier. d) A thermoreversible polyurethane printed using DIW in the shape of a hand. It can be actuated by locally shining IR light on an individual finger. Reproduced with permission.^[214] Copyright 2019, Royal Society of Chemistry. e) A shape memory effect can be introduced in self-healing objects printed using DLP. Reproduced with permission.^[215] Copyright 2019, The Authors, published by IOP Science. f) Gripper actuated using a shape memory effect, printed using DLP. Reproduced with permission.^[41] Copyright 2019, American Chemical Society. g) FinRay gripper printed using FFF out of a Diels–Alder elastomer. Reproduced with permission.^[30] Copyright 2020, Mary Ann Liebert, Inc.

by layer depositing. However, as shown in **Figure 11d**,^[30,227,228] the prints produced with reversible elastomers can have a good surface finish, as well as isotropic mechanical properties in the print. This results from high interfacial (physico)chemical bonding between the printed layers, slower solidification, and higher mobility (lower viscosity) to seal the gaps between layers. It should be taken into account that there exists a trade-off in print resolution at the sides of the print and surface finish, which is inversely affected by the solidification speed. If required, the interlayer strength in printed self-healing parts can be increased via post-curing at elevated temperature below the gel transition temperature, where mechanical stability is guaranteed. Liu et al.^[231] printed a double hydrogel network in which one network is formed during printing, while the other one is formed during a post-processing step to improve mechanical properties of the print.

Whereas molding techniques need multiple stages to obtain multi-material parts, DIW allows to print one object using

several materials, each printed from a different nozzle,^[232,233] or by switching multiple materials through a single nozzle.^[234] This is, for example, shown by Gul et al.,^[232] who used several printing heads to print a spider-like soft robot using nonreversible urethane- and epoxy-based photopolymers combined with a shape memory alloy wire. It also allows to print composite inks with particles as graphene or NdFeB to make them electrically conductive or magnetic, respectively, which leads to new possibilities in printing sensors or actuators.^[235,236] This multi-material printing has also been shown for self-healing polymers.^[214,228] It allows researchers to create actuation (rolling^[237] and shape-memory behavior,^[214,228,238] see **Figure 10d**) and sensing (gas detection,^[239] motion,^[33,66,231] strain gauge^[240,241]) concepts that can be useful in soft robotics. In the near future, it is expected entire robotic systems will be printed in a single, multi-material printing process.

Another advantage of DIW is that there is very little pre-processing needed for the material compared to other additive

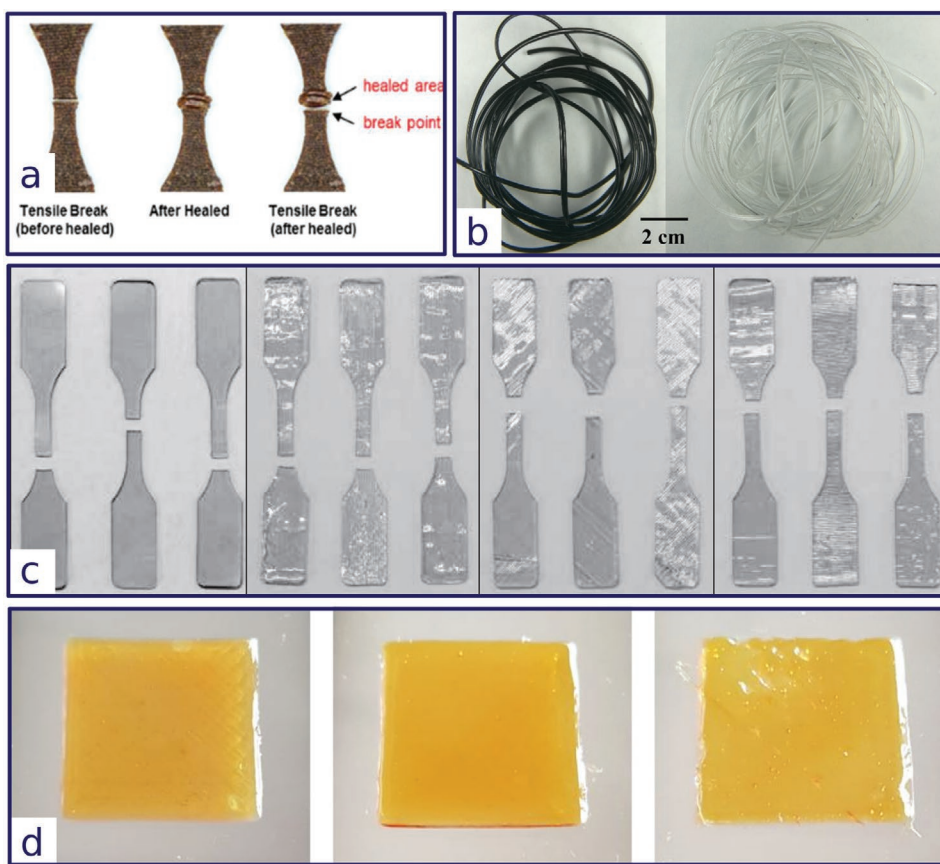


Figure 11. Additive manufacturing of self-healing polymers has several advantages, especially for extrusion-based techniques. a) Blending a Diels–Alder based mending agent with PLA, decreases the anisotropy in printed parts. Reproduced with permission.^[246] Copyright 2017, Royal Society of Chemistry. b) For FFF, filament needs to be produced, which can be a challenge. Calderon-Villajos et al. produced smooth filament of a commercially available ionomer. Reproduced with permission.^[247] Copyright 2019, Elsevier. c) By printing thermoreversible polymers, very low anisotropy can be achieved, thanks to the strong covalent bonds formed over the interface. These dogbones are printed in different directions and show anisotropy as low as 5%. Reproduced with permission.^[227] Copyright 2017, Wiley-VCH. d) Samples of Diels–Alder polymer printed at different temperatures. The layer marks and surface roughness that is typically observed during extrusion-based printing is reduced when using thermoreversible polymers. Reproduced with permission.^[30] Copyright 2020, Mary Ann Liebert, Inc.

manufacturing techniques, such as FFF (Section 5.2.1), which requires high quality filament, and selective laser sintering (SLS, Section 5.2.3), which uses powder with narrow size distribution. Pellets or pieces of reversible polymer with random shapes can be placed in a piston, liquefied by heating above T_{gel} or T_v , and used in the DIW process. In addition, DIW allows to print flexible elastomers that are very challenging to print with FFF, as flexible filament has the undesired tendency to buckle in the extruder head of FFF printers. This eventually leads to failure of the print or low quality prints.^[242] However, the viscosity of the ink is required to be within a window of about $0.1\text{--}3 \times 10^3$ Pas for optimal printability,^[216,221] as too high viscosity prevents flow out of the nozzle. The viscosity increase when leaving the nozzle and the subsequent solidification have to be relatively fast to achieve mechanical stability of the printed layers. For intrinsic self-healing materials that rely on reversible reactions, this means that the kinetics of the reversible reaction should be sufficiently fast. Yang et al.^[227] showed that thin, high structures could be printed using Diels–Alder based self-healing polymers. However, for printing overhangs the solidification was not fast enough. Zhou et al.^[228] showed that the reversible nature of the cross-links in these self-healing elastomers permits joining

printed parts via a heat–cool cycle to form overhang structures. If adequate solidification speeds cannot be reached, even with an elevated print bed temperature that increases kinetics, embedded direct ink writing can be a solution.

5.1.2. Embedded Direct Ink Writing

In embedded direct ink writing (EDIW), a needle prints the polymer part inside of a medium, which can be a liquid^[243] or a gel.^[203,244] A low viscosity or a long solidification time is no longer an issue, as the ink is supported by the medium. This was shown by Sparrman et al.^[245] who printed a two-component silicone with a standard curing time of 6 h inside a self-healing gel. This illustrates that self-healing gels find also applications in the support material for this EDIW printing technique, as gaps and tears in the medium, generated by the needle that moves through the medium, should be healed relatively fast. Although not yet used to develop healable soft robotic parts, EDIW is very promising for many intrinsic self-healing polymers, in particular the ones based on reversible dissociative covalent cross-links, as they have slow reaction kinetics, leading to slow solidification. As printing

can be performed at lower viscosities of the ink, finer needles or nozzles can be used, and the resolution can be increased.

5.2. Additive Manufacturing via Reprocessing

5.2.1. Fused Filament Fabrication

FFF, also known under the trademarked name “fused deposition modeling”, is a printing technique in which filament is fed to an extruder and pushed through a hot nozzle to deposit strands of material on the print bed, similar to DIW (Section 5.1.1). Compared to DIW, a higher extent of preprocessing is required: it needs smooth filament with a constant diameter (typically 1.75 or 2.85 mm). This filament is produced via an extrusion process. During the printing of conventional thermoplastics, the polymer is typically heated above its glass transition (for amorphous thermoplastics like ABS) or melting temperature (for semicrystalline thermoplastics like PLA). Upon deposition on the print bed, the polymer solidifies by vitrification or crystallization. FFF is well-known in soft robotics for manufacturing of molds, used for casting silicones for soft robotic components. Commercial thermoplastic elastomers, including TPU^[248,249] and SBS, have been used abundantly to print soft robots.^[73] However, only a limited number of elastomeric filaments is commercially available and the order of mechanical moduli is limited to the 1 MPa–1 GPa range. Nevertheless, many soft robotics applications require materials with lower moduli of 100 kPa–1 MPa.^[19,250] There are two reasons for the lower limit in flexibility. First, printing with highly flexible filament is challenging, as buckling and compression of the filament limit the pressure that can be applied to push the polymer through the nozzle. However, in new direct extruders designed specifically for flexible filament, these undesired effects are reduced. The second reason, is that the flexibility of thermoplastic elastomers is limited, with moduli not lower than 1 MPa, because the physical cross-links are formed by an immobile phase, a crystalline phase (SBS), or a phase with a high glass transition (TPU), formed by phase separation in the block co-polymer. In reversible elastomers cross-linked by (physico)chemical interactions or reversible covalent bonds, lower moduli can be achieved. Furthermore, these intrinsic self-healing elastomers can be printed using FFF, by exploiting the reversible nature of the (physico)chemical cross-links, in contrast to conventional permanently cross-linked elastomers. By heating a filament made of an extruded thermoreversible network above its transition temperature, the filament de-gels and the viscous polymer is pushed through the nozzle. This principle is referred to as reactive FFF printing, as its liquefaction and its solidification are based on a (physico)chemical reaction, instead of a physical process. If the hardware of the printer allows it, elastomeric soft robotic components with higher flexibility can be printed from self-healing elastomers, expanding the use of FFF in this field.

Fused filament fabrication of thermoplastic elastomers has another disadvantage: as the material solidifies through a physical transition (glass transition or crystallization), the interlayer adhesion in printed parts relies entirely on the molecular (inter) diffusion at the interface between the print bed and the just deposited filament, and on the formation of physical cross-links through entanglement, crystallization, or vitrification at the

interface, rather than on the formation of chemical bonds. The resulting weak interlayer strength often leads to undesired anisotropy and poor mechanical properties in the direction perpendicular to the print layers.^[251,252] Hence, it is challenging to print pneumatic actuators airtight.^[248] Self-healing polymer networks can be of great interest for FFF, as they can solve the problem of weak interlayer adhesion. The presence of reversible bonds in the elastomer allows to create (physico)chemical bonds between the layers, increasing the mechanical strength in the direction perpendicular to the printed layers. This leads to a reduction of the anisotropy (Figure 11c).^[227] This was shown to be the case in the Diels–Alder based self-healing parts printed by Roels et al.,^[30] in which anisotropy is reduced to only 13%, measured by comparing the fracture stress of samples strained along the print direction to samples strained perpendicular to the print layers. Similarly, Appuhamillage et al.,^[246] greatly improved isotropy in printed PLA samples, by blending them with a Diels–Alder network (Figure 11a). Similar to DIW, FFF printing with intrinsic self-healing polymers can increase the surface smoothness of the prints. Roels et al.^[30] printed a self-healing gripper via FFF out of Diels–Alder elastomer, of which the fingers can heal from being cut completely in half (Figure 10g). In addition, the printed surfaces are smooth, an important feature for food grippers, as this facilitates cleaning and reduces the potential of dirt and bacteria to stick in the ridges on the surface.

Multi-nozzle FFF allows for multi-material printing,^[253] which is an increasing demand in soft robotics as multi-material designs can improve the system’s performance. By printing conductive and nonconductive polymers in a single print, sensors can be integrated directly.^[254,255] Multi-material FFF using multiple self-healing materials that have different mechanical properties, yet identical reversible chemistry, will have the advantage that the multi-material interfaces in the print will be chemically bonded together during the printing process or by post-curing. These strong interfacial bonds are not only important for robust multi-material soft actuators, but also in soft sensors, where strong adhesion between the flexible conductive sensor and its matrix is required.^[256]

When working with non-commercial materials, as is currently the case for self-healing elastomers, filament has to be extruded prior to printing. For high quality extrusion, relatively expensive equipment is required, so DIW (Section 5.1.1) is often preferred as an alternative as there are no preprocessing requirements for this technique.^[227] Calderon-Villajos et al.^[247] have described the filament extrusion and printing process of the self-healing Surlyn ionomer (DuPont, Inc.) both with and without carbon nanotubes for reinforcement (Figure 11b). These ionomers are quite stiff (Young’s modulus ≈300 MPa). More flexible (Young’s modulus ≈8 MPa) materials have been printed by Roels et al.^[30] who did report problems with obtaining smooth filament. Nevertheless, it was possible to print a soft robotic gripper using a slightly adapted printer. Fused granulate fabrication (FGF), is another alternative to avoid the need for extensive preprocessing.

5.2.2. FGF

FGF or granulate printing is a variation on fused filament fabrication. Instead of extruding the filaments with a screw extruder

first, the screw extruder is incorporated in the printing head. Pellets or granulates are printed directly,^[257] which circumvents the need of extruding smooth filament first, making FGF a more economical option. The added weight of the (small) extruder mounted on the print head reduces the maximum printing speed compared to FFF. Khondoker et al.^[258] solved this issue by fixing the extruder on the printer frame and leading the material through a heated tube toward the nozzle. Because no filament is used, this technique lends itself well to print very soft materials (shore hardness <60 A, which appears to be the lower limit for current commercially available FFF materials^[259,260]). They have successfully printed a pneumatic finger in styrene–ethylene–butylene–styrene thermoplastic elastomer with a shore hardness of ≈47 A.^[258] However, using softer (physico)chemical cross-linked networks, the flexibility can be reduced further, making this technique highly suitable for highly flexible soft robotic applications. Because this technique is similar to FFF, the material properties of the printed parts are similar.^[261]

5.2.3. SLS

SLS is the overall third most popular additive manufacturing technique, after FFF and stereolithography (SLA). The polymer is prepared as a powder, and a roller deposits a thin layer of polymer powder that is heated up to just below its melting point. Next, a laser locally heats the powder to fuse (sinter) it together. After sintering the desired regions, the roller can deposit a new layer of powder. The most used polymer for SLS is polyamide, which is relatively stiff, but can nevertheless be used for flexible robotics using flexure hinges, as described by Roppenecker et al. and Krieger et al.^[262,263] While they are not very common, flexible TPU's are commercially available for use in SLS down to a shore hardness of about 45 A, bringing it on par with FFF regarding the ability to print soft materials using thermoplastic elastomers. Scharff et al.^[264] used a TPU with shore hardness of 92 A to manufacture a soft pneumatic hand. This illustrates that hollow structures can be printed using this SLS technique, due to the powder that supports the object during printing. However, a large enough opening has to be foreseen to get the excess powder out of the cavity. The surface of the finished part is rough and SLS generates a lot of waste material. Because the powder bed is heated, the powder quality degrades and can generally not be reused. In addition, care needs to be taken when cleaning a part as the fine powder is an inhalation hazard. However, SLS printers have a high precision and a resolution of typically around 100 μm, which is significantly better than FFF or DIW.

A disadvantage of SLS is that extensive preprocessing is needed, as powders with narrow size distribution are required.^[265] Powder production can be performed using (cryo-)milling and sieving. Because extensive preprocessing of the powder is required, only few examples are found of SLS being applied to self-healing polymers. Sun et al.^[213,266] sintered a dynamic TPU, containing a reaction mechanism of the halogenated bisphenol carbamate that allows for both healing and reprocessing through SLS (Figure 10c). Due to the sintering, the inter-layer adhesion is stronger in comparison with FFF and there is less anisotropy in the finished parts.^[267] Nonetheless, Sun et al.^[213,266] show that using their self-healing TPU, even higher

isotropic mechanical properties are achieved compared to a commercially available TPU. In general, SLS printed parts have mechanical properties that are inferior to those of molded parts due to porosity. Reversible chemistries in self-healing elastomers can provide a solution for this in the future, as these cavities can be sealed through post processing. Although multi-material SLS is very challenging, new powder depositing principles (for metals) are currently being developed that allow multi-material printing,^[268] but design freedom is and will be very limited.

5.3. Additive Manufacturing via Photo-Resin Printing

Photo-resin printing (or shorter: resin printing) is an overarching term for all additive manufacturing techniques where liquid prepolymer (resin) is polymerized layer by layer via photopolymerization. It includes Digital Light Processing (DLP), (laser) SLA, multi-photon lithography and inkjet printing. The low viscosity resin is usually polymerized by a UV laser or LED on a layer by layer basis. While inkjet printing uses a nozzle to spray resin droplets in specified locations (similar to fused filament fabrication, see Section 5.2.1) that are instantly polymerized, DLP and SLA use a reservoir filled with monomer that is polymerized selectively. These techniques also show the best resolution of the additive manufacturing techniques discussed, and are therefore suitable for models with fine details.

5.3.1. Digital Light Processing and Stereolithography

DLP differs from SLA by the fact that the whole layer is irradiated simultaneously (using a combination of an LED and an LCD), whereas the light source (laser) scans the layer in SLA. When a layer is finished, a plunger moves the object from the source and a new layer is polymerized. In multi-photon lithography, which is specifically for sub-millimeter-scale printing, there is no moving plunger. When a layer is finished, the focal point of the laser is shifted to solidify the next layer. Photo-resin printing is limited to polymers that can photopolymerize under irradiation at a certain wavelength. Most elastomer resins are based on acrylates and urethane acrylates with flexible chains, but also other chemistries were investigated.^[269] For SLA and DLP, after printing, usually a post-processing step is performed, which consist of washing away all non-polymerized resin and post-curing under UV light. Thanks to the post-curing step, resin-based printing shows the lowest anisotropy (1–2 %) in finished parts of all common additive manufacturing techniques.^[251] Objects manufactured using resin printing have a good surface finish and the technique is generally much faster compared to other additive manufacturing techniques, such as DIW or FFF.

Due to the patented nature of inkjet printers, these printers can currently not be used with custom materials and thus all resin printing for self-healing materials described is performed using reservoir-based printing techniques. Intrinsic self-healing elastomeric parts can be produced from resin that contains photoreactive components required for photo-resin printing, as well as reversible (physico)chemistries needed to introduce healability.^[215] Compared to FFF and DIW, there is less need to optimize the rheological properties of the material.^[270] Liu et al.^[271]

combined the thiol-ene photoreaction with reversible (physico) chemical hydrogen bonds to print self-healing elastomeric parts in an SLA printer, while Yu et al.^[38] combined it with reversible disulfide bonds (Figure 10a). Li et al.^[272] polymerized the network using DLP by a hydroxyethyl acrylate photoreaction and incorporated disulfide reversible bonds for healing. Sanders et al.^[270] used an SLA printer to print an extrinsically self-healing polymer through the free radical photopolymerization of a photocurable resin modified with polymer and solvent-filled microcapsules. Not only a healing ability, but also a shape memory effect can be introduced in DLP printed elastomeric parts (Figure 10e,f).^[41,215] Suriano et al.^[215] used the cross-linking photopolymerization of a methacrylated polycaprolactone (PCL) with a monomer containing 2-ureido-4[1H]-pyrimidinone functionalities for forming hydrogen-bonded complexes. Vitrimers have been printed by Rossegger et al. using thiol-acrylate chemistry, exploiting the acrylate ester bonds for transesterification.^[273,274]

For photo-resin printing, the reaction rate of the photopolymerization has to be relatively fast, within the order of seconds, and directly impacts the printing time.^[271] In addition, the viscosity of the resin directly impacts the printing time. When the plunger moves to prepare for the next layer, enough time should be given for the liquid resin to even out. This time depends on the resin viscosity and should be sufficiently low. Various upper limits are found in literature: while Bartolo et al.^[275] put 5 Pa·s as upper limit, Zhang et al.^[41] find 42 Pa·s a reasonable upper limit, as a simulation shows this corresponds to a waiting time of 1 min per layer for it to even out. Heating the resin can reduce the viscosity, and thus the total printing time, significantly. Post-curing may also be a vital step of photo-resin printing of (self-healing) elastomers. An interpenetrating polymer network (IPN)^[190,276] was formed by printing the first network or so-called 'green body', while the second network is formed during a post-treatment (UV-curing process,^[277] heating^[271]). As IPNs are a promising route in developing self-healing elastomers that have the potential to combine excellent fast healing at moderate temperatures with high mechanical strength and stability, this approach has a lot of potential for a new generation of self-healing materials for soft robotics.^[20]

When printing hollow structures, the internal cavities of an object will be filled with liquid unreacted resin. For soft pneumatic actuators, similar as in SLS (Section 5.2.3), a hole in the design allows to let the resin flow out to create air chambers that can be actuated by compressed air.^[278–281] However, the cavities can also be closed, and uncured resin can be encapsulated and used as actuation fluid. Even upon post-curing via light irradiation, the encapsulated resin will not cure, as it is not subjected to the light, which typically has a limited penetration depth in the polymer network shell. Aside from acting as actuation fluid, the embedded resin can also be used to incorporate an extrinsic healing mechanism in the system.^[282] This was demonstrated by Wallin et al.,^[47] who manufactured a hydraulic actuator via SLA, in which uncured photo-resin is used as actuation fluid for a bidirectional bending actuator. It permits to heal damages caused by sharp objects in the actuator walls via photocuring. Upon damage, photo-resin escapes through this puncture and is exposed to ambient sunlight, under which it instantaneously photopolymerizes and solidifies, sealing and

healing the damage. Zhang et al.^[283] on the other hand, printed hollow structures using DLP, in which functional liquids are injected as post-processing step. Liquid metals are injected to create integrated soft sensors, while UV curable resin with a modulus that differs from the DLP printed matrix, is used to limit strains locally to enhance the performance of the pneumatic actuators. The latter resin solidifies via post UV-curing.

Due to the need of a liquid reservoir, it is challenging to make multi-material parts. Nonetheless, there are some resin reservoir printers that support multi-material printing by dipping the object in a second reservoir^[284] or material puddles,^[285] but the availability is low and the price high.^[38] Magnetic-field-assisted projection SLA can also be used to vary the amount of magnetic filler within a printed object, which allows to create gradients of mechanical and magnetic properties.^[286,287] This allows to program complex motions in a soft robot in a single manufacturing step. The approach also guards from issues that arise when using resin printing for filled materials, in which the filler has to remain well dispersed within the resin during printing to obtain a homogeneous part.^[275,288] Post-injection of UV resin, as presented by Zhang et al.^[283] and discussed before, is also a (limited) possibility to manufacture multi-material parts. However, it is clear that for multi-material SLA and DLP, the design freedom is and will be limited.

5.3.2. Inkjet Printing

In inkjet printing, the photo-resin is printed in lines, similar to DIW (Section 5.1.1) on a print bed on which it photopolymerizes via a (UV) laser or LED. In contrast to SLA and DLP, this technique allows to print multi-material objects by having multiple nozzles.^[152,289–291] This was demonstrated by Hughes et al.,^[51] who printed a piano-playing soft robotic hand with passive intelligence in a single inkjet printing procedure with three distinct materials, of which one was used as soluble support. By depositing a ratio of two different photo-resins on a single location, prior to UV exposure, gradients of mechanical properties can be printed in soft robotics as well, as demonstrated by Bartlett et al.^[292] in their combustion-powered soft robot. MacCurdy et al.^[293] used multi-nozzle inkjet printing to print multi-material soft robotic components, in which one material is a noncuring liquid. This embedded liquid acts as the actuation fluid in their design. An additional advantage of inkjet printing is that in general no post-processing is needed. Although the technique is promising, to the authors knowledge, no healable soft robots have been manufactured using inkjet printing. However, there have been reports on inkjet printing with self-healing elastomers, as presented by Rahman et al.,^[294] who printed using a thiol-ene photoreaction and incorporated healing by disulfide cross-links.

5.3.3. Multi-Photon Lithography

A third additive manufacturing technique based on photo-resin polymerization, is multi-photon lithography or direct laser writing. This technique has many similarities with SLA/DLP, but is specifically designed for sub-millimeter-scale printing, and

provides an excellent resolution: down to 50 nm.^[295] In contrast with SLA and DLP, there is no moving plunger. When a layer is finished, the focal point of the laser is shifted to solidify the next layer of resin. The high resolution and low working volume, make multi-photon lithography an excellent choice for printing micro-actuators^[296] or their molds.^[297,298] Typical materials used for multi-photon lithography are epoxies and acrylates, but more recently, different hydrogels were successfully printed into actuators.^[299,300] Also a reversible Diels–Alder network was used to support the object during the printing process while the laser irreversibly cross-linked the network selectively using radical reactions with suspended thiol molecules.^[301] This led to improved design freedom as the support material is solid, and overhangs can thus be printed more easily. So far, there are no reports of self-healing actuators manufactured using this technique, but as more and more stimuli-responsive materials (light, pH, heat,...) can be processed using this technique,^[296] it is a promising path yet to be explored for intrinsic self-healing materials.

6. Selection of Manufacturing Techniques for Healable Soft Robots

All processing methods described in this review paper are useful for soft robotics and have already shown their potential, but they also have their limitations. Many of these limitations are resolved when selecting intrinsic self-healing elastomers. The final decision on the choice of material and manufacturing technique depends heavily on the needs imposed by the application. The most important requirements that can occur for soft robotics were introduced in Section 1.1. **Table 1** indicates for each processing method and type of elastomer whether the different requirements are satisfied (green bullet), not satisfied (red square) or only partially satisfied (orange triangle), facilitating a fair comparison between the different techniques. For example, it becomes clear that formative techniques have generally a lower tolerance for complexity as compared to the additive techniques, however they tend to result in a superior surface finish. Extrusion-based additive manufacturing result in a lower resolution compared to other additive techniques, but they have the advantage of being compatible with a larger range of materials, and they are often better suited for printing multi-material parts. It also becomes clear that for many manufacturing techniques, especially additive manufacturing, scalability remains a major challenge.

Table 1 also shows that some requirements that are not or only partially met when using thermosetting or thermoplastic elastomers, can be satisfied when using self-healing elastomers. Parts made of self-healing elastomers can be made out of a single material and assembled into multi-material parts using any of the techniques given in Section 4. This gives the user more choice in processing method, as the requirement for being able to make (strong) multi-material parts is now automatically satisfied. Using assembly and binding, also allows to create hollow structures using techniques that otherwise do not support this. Moreover, the use of self-healing elastomers in extrusion-based additive manufacturing permits improving the surface finish.

From the table, one could conclude that casting self-healing polymers is the ideal technique, as it satisfies all requirements.

However, besides this table, and depending on each individual application and material, there can be additional constraints that need to be taken in account when deciding on a processing technique to go from a material to a soft robot. Consequently, Table 1 should be used as a guideline to select possible techniques and rule out others. An important constraint not indicated in the table, is imposed by the viscosity of the monomer (mixture) or the polymer in liquid state (above its transition temperature). Various manufacturing techniques require different viscosities. Typical viscosity ranges for some of the techniques are given in **Figure 12**. It becomes clear that a material with a high viscosity will, for example, not be the ideal candidate for casting, instead FFF or FGF could be better option. Alternatively, by adding adequate fillers or pre-heating the material, the viscosity could be improved to fit the desired processing technique. Although the solidification time is an important parameter, highly influencing the manufacturing time, it is impossible to specify acceptable solidification times for each technique as these will highly depend on the acceptable cost of manufacturing of the component.

Another constraint is imposed by the solidification speed of the polymer network. If the material solidifies slowly, a formative process is preferred, as there is only one solidification step. In additive processes, each layer typically has to solidify before the next can be deposited on top. Whereas it is not impossible to use these techniques, it would take a long time before the part is finished. If the material solidifies very quickly, the handling time (also referred to as “pot life”) might be too short for a formative technique as casting, and an additive technique can be used instead with mixing of the reagents just before printing or using a triggered solidification.

7. Conclusion and Perspective

Both the fields of self-healing materials and soft robotics have progressed significantly during the past decades and will continue to do so in the coming years at an increasing pace. In recent years, these fields combined to create self-healing soft robots. The processing and manufacturing methods to create complex, self-healing structural components were thoroughly reviewed in this paper, as this bridge between these two fascinating research fields will spur advances and enhance future interdisciplinary research.

The reversible covalent bonds and physicochemical interactions used to create intrinsic self-healing polymers do not only endow these polymeric systems and structures with the ability to repair damage. They also offer enhanced (re)processability, as opposed to irreversibly cross-linked thermosetting elastomers like silicones, and improved (hyper)flexible behavior and mechanical stability, compared to thermoplastic elastomers like polyurethanes. The self-healing ability increases the lifetime of the soft robot, while their reprocessability further reduces their ecological impact. The authors believe that this ecological incentive will lead to an increased use of these smart materials in robotics.

Many techniques for manufacturing soft robots have been developed. Nonetheless, all of these techniques have drawbacks and limitations when used with traditional types of materials. The use of intrinsic self-healing materials helps to overcome

Table 1. Processing methods and their advantages and disadvantages for irreversible and self-healing reversible polymers. An overview is given that details which of the requirements for soft robotics are satisfied for each of the methods and elastomer types.

| Formative | Elastomer | i) Low complexity | ii) Large design freedom | iii) Materials range | iv) Multi-material | v) Material properties | vi) Surface finish | vii) High resolution | viii) Hollow structures | ix) Composites | x) Reproducibility | xi) Scalability | Comments | Ref. |
|-------------------------|---------------|----------------------|-----------------------------|-------------------------|-----------------------|---------------------------|-----------------------|-------------------------|----------------------------|-------------------|-----------------------|--------------------|---|----------------------|
| | | | | | | | | | | | | | | |
| Casting | Thermoset | ● | ● | ● | ▲* | ● | ● | ● | ▲† | ● | ● | ● | *multi-stage casting only | [52, 58, 157] |
| | Self-healing | ● | ● | ● | ●* | ● | ● | ●* | ● | ● | ● | ● | †Multi-stage casting, rotational casting *using assembly and binding | [29, 143, 159] |
| Compression moulding | Thermoplastic | ● | ▲ | ▲ | ■ | ● | ● | ■ | ■ | ● | ● | ● | *using assembly and binding | [166] |
| | Self-healing | ● | ▲ | ▲ | ●* | ● | ● | ●* | ● | ● | ● | ● | *using assembly and binding | [111, 160, 163, 167] |
| Injection moulding (IM) | Thermoset | ■ | ● | ● | ▲* | ● | ● | ▲ | ▲ | ● | ● | ● | *co-injection only | [169, 170, 174] |
| | Thermoplastic | ■ | ● | ● | ▲* | ● | ● | ▲ | ▲ | ● | ● | ● | *co-injection only | [168] |
| | Self-healing | ■ | ● | ● | ●* | ● | ● | ●* | ● | ● | ● | ● | *using assembly and binding | [99] |
| Solvent casting | Thermoset | ● | ■ | ■ | ▲* | ● | ● | ■ | ■ | ▲ | ▲ | ■ | *multi-stage casting only | [181] |
| | Self-healing | ● | ■ | ■ | ●* | ● | ● | ■ | ■ | ▲ | ▲ | ■ | *using assembly and binding | [63, 142, 144] |
| Additive | | | | | | | | | | | | | | |
| DIW | Thermoset | ● | ● | ● | ● | ▲ | ■ | ▲* | ▲ | ● | ▲ | ■ | *highly depends on the solidification speed | [217, 218, 225] |
| | Self-healing | ● | ● | ● | ● | ▲ | ▲ | ●* | ● | ● | ▲ | ■ | *using assembly and binding | [187, 212, 214] |
| Embedded DIW | Thermoset | ▲ | ▲ | ● | ● | ▲ | ■ | ●* | ● | ● | ▲ | ■ | *if the medium is removed after the print | [245] |
| | Self-healing | ▲ | ▲ | ● | ● | ▲ | ▲ | ●* | ● | ● | ▲ | ■ | *using assembly and binding | |
| FFF | Thermoplastic | ▲ | ● | ▲ | ● | ▲ | ■ | ● | ● | ● | ▲ | ■ | | [73, 248, 249, 254] |
| | Self-healing | ▲ | ● | ● | ● | ▲ | ▲ | ● | ● | ● | ▲ | ■ | | [30, 247] |
| FGF | Thermoplastic | ▲ | ● | ● | ▲* | ▲ | ■ | ● | ● | ● | ▲ | ■ | *not yet done, but possible | [257] |
| | Self-healing | ▲ | ● | ● | ●* | ▲ | ▲ | ● | ● | ● | ▲ | ■ | *using assembly and binding | |
| SLS | Thermoplastic | ■ | ● | ▲ | ■ | ▲ | ▲ | ▲ | ▲ | ● | ● | ▲ | | [262–264] |
| | Self-healing | ■ | ● | ● | ●* | ▲ | ▲ | ●* | ● | ● | ● | ▲ | *using assembly and binding | [213, 266] |
| SLA/DLP | Thermoset | ▲ | ● | ■ | ■ | ● | ● | ● | ● | ■ | ● | ▲ | | [47, 278, 281] |
| | Self-healing | ▲ | ● | ■ | ●* | ● | ● | ● | ● | ■ | ● | ▲ | * using assembly and binding | [38, 41, 215] |
| Inkjet printing | Thermoset | ■ | ● | ■ | ● | ● | ● | ● | ● | ■ | ● | ▲ | *due to the patents on this technology | [51, 152, 293] |
| | Self-healing | ■ | ● | ■ | ● | ● | ● | ● | ● | ■ | ● | ▲ | | [294] |

● requirement satisfied ▲ partially satisfied ■ requirement not satisfied

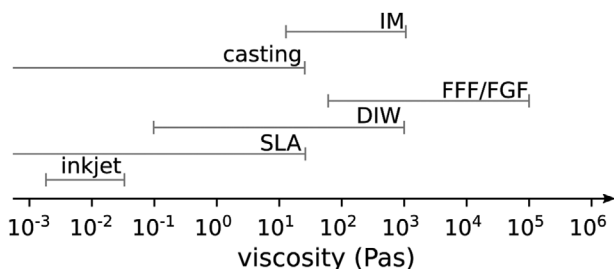


Figure 12. Typical viscosity ranges for different additive and formative processing methods. Values taken from refs. ^[41,216] [221,302–304].

some of these issues and opens up new opportunities that were not available for thermosetting and thermoplastic elastomers. As such, these reversible covalent and supramolecular polymer networks blur the lines between reactive and thermoplastic processing.

Casting of conventional thermosetting elastomers is extensively used to produce soft robotics components and systems in academia. However, it introduces a lot of issues when fabricating complex and multi-material parts, including the introduction of weak links at (multi-)material interfaces. The same is true for compression molding and IM, two formative manufacturing techniques for thermoplastic elastomers that hold a lot of potential for producing soft robots on an industrial scale in the future. Intrinsic self-healing elastomers, made up of networks cross-linked by reversible covalent or physicochemical bonds allow the formation of much stronger connections across the interface of prefabricated parts by way of their reversible character, activated using an adequate trigger.

It is expected that in the future many intrinsic self-healing polymers will still be synthesized via solvent casting, due to the lack of miscibility of the reagents involved. However, the produced sheets can be reprocessed via other (formative) techniques, thanks to the reversible nature of the cross-links. Besides, 3D structures can be made from 2D sheets via different manufacturing techniques, including stacking, welding, and folding and covalently binding. During this welding procedure, the polymer parts remain solid and preserve their mechanical stability. With the general trend in soft robotics of using multi-material designs for increased performance or/and embedding sensors, the benefits of using intrinsic self-healing elastomers will be increasingly exploited.

Intrinsic self-healing polymers can be (re)processed using formative manufacturing techniques, such as compression and IM, and produced parts can be joined and merged by activating the reversibility of the cross-linking bonds using the adequate trigger. Structures that would not be possible to print due to limited overhangs, can now be produced. Intrinsic self-healing materials have found their way to many types of additive manufacturing techniques, including direct ink writing, fused filament fabrication, fused granulate printing and inkjet printing, often by exploiting their thermally reversible reactivity. Many of these techniques allow to create multi-material parts in a single printing process, which opens a lot of opportunities for higher performance soft robotic designs and for embedded soft electronics. The use of self-healing polymers in these multi-material printing techniques ensures strong interfaces between the

different materials printed. The reversible nature of the cross-links in these materials also highly enhances the interlayer/granular bonding that is limited using traditional thermoplastic elastomers. This results in a higher mechanical strength and robustness, often superior to thermoplastic elastomer alternatives. Incorporating a thermally or photochemically reversible chemistry in photo-resins embeds a healing ability in printed parts using light-based manufacturing techniques. In addition, the produced components can be reprocessed or recycled using formative or additive manufacturing techniques by activating the reversible nature of the cross-links using the adequate trigger.

The reviewed advantages these reversible covalent and supramolecular elastomers offer for the formative and additive manufacturing of complex, self-healing structural components, will continue to spur their use in different types of structural applications, especially in soft robotics and flexible electronics.^[305–317]

Acknowledgements

This literature review was performed in relation to and funded by the EU FET Open RIA Project SHERO (828818), the EU Marie Curie ITN project SMART (860108) and the FWO SBO project AMSeR (G028218N). In addition, the authors gratefully acknowledge the FWO (Fonds Wetenschappelijk Onderzoek) for the personal grants of Roels (1S84120N), Terryn (1100416N), and Brancart (12W4719N).

Conflict of Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: A.W.B. is employed by SupraPolix BV and hold shares in SupraPolix BV.

Keywords

additive manufacturing, processing, self-healing, soft robotics

Received: June 23, 2021

Revised: September 27, 2021

Published online: November 6, 2021

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