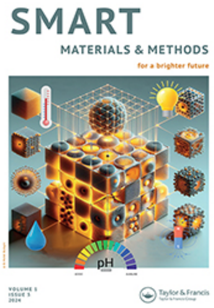

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REVIEW ARTICLE

Vitrimers for sustainable electronics and control of e-waste

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ABSTRACT
Vitrimers offer an innovative approach to recycling polymer waste, crucial in addressing the environmental impact of electronic devices and circular economy. The increasing role of electronics in various industries has heightened concerns regarding material consumption, electronic waste, landfill costs, and energy usage. Vitrimers, as adaptable polymer networks, employ dynamic cross-linkages that activate bond exchange reactions under external stimuli, enabling topology rearrangements. These networks are instrumental in developing sustainable electronics, offering rehealability, shape memory, recyclability, and reprocessing capabilities. This review examines the latest research on vitrimers in sustainable materials for electronic devices, exploring various chemistries in vitrimer design and their impact on device properties. It highlights the potential of vitrimer topology rearrangement and exchange reactions in creating shape memory, self-healing, and recyclability properties. Additionally, the review categorizes vitrimer-based electronic devices, including triboelectric generators, wearable electronics, electro-skins, sensors, supercapacitors, electromagnetic shielding, soft actuators, flexible electronics, printed circuit boards, ionotronics, moisture generators, and thermoelectric devices. The article aims to foster further research on vitrimers in industrial applications, promoting the development of sustainable electronics.

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KEYWORDS Vitrimers; electronic waste; sustainability; circular economy; recyclability; rehealability; shape memory

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1. Introduction

With development of human tools and the gradual change of life and daily needs, the development of technologies related to these changes has become equally important [1–3]. This has caused electronics to become a key concept today so that it can be transformed into technologies based on pharmaceutical, security, information technology, electronic skins, energy, and other fields in the future [4–8]. Apart from the current research in the development of electronics, the future of materials used in new designs of electronic devices is also a crucial issue. Polymers are one of the things that have entered the field of electronics today to advance electronics with their special properties [9,10]. Today, one of the most important problems in the world is to deal with pollution caused by these polymeric materials, which may limit the benefits of using polymers. In this regard, recent research has introduced some types of polymer networks called vitrimers, possessing dynamic and adaptable covalent networks. They can use different chemistry to a structural rearrangement that ultimately leads to the properties such as high flexibility, shape memory, self-healing, reprocessability, and recyclability [11–13]. Therefore, the use and introduction of vitrimers to electronic devices can be the beginning for the development and design of a sustainable and eco-friendly design with unique properties, such as shape memory, reprocessability, rehealability, and recyclability. This review article explores recent advancements in using vitrimers for developing sustainable electronic materials and devices and covers their unique properties such as shape memory, self-healing, and reprocessing. After examining the properties with the chemical structure of vitrimer and the type of exchange reaction applied in the synthesis, it also highlights a discussion of recycling and sustainability. Furthermore, this review classifies the types of vitrimer-based electronic devices in today's industries. It includes several sections such as vitrimer-based triboelectric generators, wearable electronics, electro-skins, sensors, supercapacitors, electromagnetic shielding, soft actuators and soft robotics, flexible electronics, printed circuit boards, ionotronic, moisture generators, and thermoelectric devices. This review article is expected to motivate more research in the use of vitrimers in industrial applications for the development of sustainable electronics.

2. Recycling

2.1. Physical and chemical recycling of polymer waste

Before examining vitrimerization strategy, it is better to introduce the general types of recycling methods in general polymer waste, especially electronic waste. Generally, these techniques are classified into two main groups of physical (mechanical) and chemical recycling.

2.1.1. Physical recycling

One of the best methods in polymer waste recycling is physical or mechanical recycling. Concurring to ASTM D7209–06 and ISO 15,270 benchmarks [14,15], this technique comprises two fundamental categories including primary [16,17] and secondary [18,19] recycling. In other words, primary recycling is known as a process of re-extrusion, re-use, and closed-loop. But secondary recycling represents downgrading. Recent secondary methods based on dissolution/reprecipitation, additive manufacturing (three-dimensional (3D) printing) [20,21], and pure cycle technology [21] are used in physical recycling. In addition to the mentioned items, blending and fabrication of composites [22–26], utilizing additives [27–29], and addition of chain extenders are also included in this classification. Among all the methods mentioned in the classification, injection moulding, extrusion, rotational moulding, and hot pressing are very popular. The traditional method of primary physical recycling includes the steps of shredding, crushing, milling, and additional cleaning [16]. The traditional method of secondary physical recycling is also based on cutting and shortening of polymer chains, which does not harm the chemical structure of the polymer while reducing the molecular weight. Also, to preserve the final properties of the recycled product, using stabilizers and applying conditions such as drying, applying vacuum, and degassing can be mentioned. [18]. Among the advantages of the primary physical recycling method, which is also known as closed-loop recycling, are: reproducing the same polymer product in a completely clean (pre-consumer), environment-friendly method, maintaining the primary properties of the polymer, and versatility with various processing methods such as injection moulding, extrusion, rotational moulding, and heat pressing. However, regarding the disadvantages of primary physical recycling, it is possible to mention things such as having additional steps such as shredding and milling, and cleaning steps [16,17,30]. It pointed out the advantages of secondary physical recycling such as the stability of the chemical structure of the polymer and maintaining the final properties of the product by using adding a group of stabilizers and applying conditions such as drying, vacuum, and degassing. The disadvantages of this include the loss of molecular weight and solvent extraction to distinguish the polymer from other waste materials [20,21]. Additive manufacturing (open-source (OS) 3D printing) is one of the emerging methods in physical recycling. Advantages such as being economical, reducing greenhouse gas emissions, and not using solvents are advantages in the direction of this method becoming more widespread. But the most important disadvantage of this method is the low recycling rate [20,31].

Along with all the existing limitations in all kinds of physical recycling methods, existing pollutions are also important. Generally, pollutants are divided into two categories: volatile organic contaminants (VOCs) [32–34] and solid contaminants. Existing physical recycling processes can cause negative effects at the molecular and morphological level. In fact, this action leads to the oxidation of residues, which causes the creation of carbon-centred (alkyl) and oxygen-centred (alkyl) free radicals leading to the transformation of the structure [35]. Degassing, Filtration and Supercritical fluid techniques are also used to remove contamination [33,34,36].

2.1.2. Chemical recycling

Chemical recycling is another important strategy for polymer waste management. The most important advantage of this method is to regain primary monomers and other primary reactants. This strategy incorporates pyrolysis [37], solvolysis (chemolysis) [38], hydrogenolysis [36], hydrosilylation [39], hydroboration [40], cyclodepolymerization of condensation polymers [41], and cross alkane metathesis (CAM) [42]. The pyrolysis method is available in two types: thermal and catalytic. Using the catalytic method requires less activation energy and the selectivity of the product is higher. Therefore, higher thermal stability, which results in easier separation of gas and liquid products. Another effective technique in breaking the chemical structure of polymers is solvolysis, including the carbonyl bond [43]. The sort of strategy that falls under this category depends totally on the dissolvable that's utilized; Illustrations incorporate glycolysis (ethylene glycol) [44], hydrolysis (water) [45], methanolysis (methanol) [43], aminolysis (amine) [46], and ammonolysis (smelling salts) [47]. In all these methods, catalysts can be used to increase product selectivity and facilitate the recycling process. In another case, condensation polymers can be changed catalytically into macrocyclic oligomers (MCOs) through cyclodepolymerization (CDP), also known as ring-closing depolymerization. In a summary of the disadvantages of the chemical recycling method, some parameters are highlighting such as high energy requirement, expensive equipment, existence of complex chemical reactions, solvent recovery, and catalyst deactivation.

2.2. Physical and chemical of electronic waste

2.2.1. Physical recycling

This method is known as one of the simplest methods and includes the lowest cost in equipment and energy [48]. But due to their adaptability in the direction of progress over time, they have a suitable potential to be used. Also, this method is known as the most widely used method in electronic waste recycling in developed countries such as the European Union and the United States. There are three important techniques in this method, which are: supergravity separation, electrostatic separation, and inverse flotation. In the first case, i.e. Supergravity separation, the effective separation of metals from other materials is based on centrifugal force. Gravitational force with high intensity will cause separation based on difference in melting temperature. The innovation has been effectively connected to recuperate uncommon soil metals, valuable metals, aluminium combinations, and other metals from various mechanical squanders [49]. When the supergravity field is applied, it causes selective melting of metals, which will cause their separation. The molten metal phase is ejected into the environment under centrifugal force while the undissolved solid particles remain [49,50]. After this stage, the melted metals can be collected after the freezing process. One of the most important parameters in increasing separation efficiency is intense mixing and heat transfer under supergravity, which causes homogenization and diffusion. Regarding the effect of gravity coefficient and temperature on separation, the work of Meng et al. can be mentioned [49]. The second technique is Electrostatic separation, which is based on the difference in electrical conductivity of metallic and non-metallic particles in crushed waste and will lead to selective separation. Also, this technique separates metals with high purity while preserving the bromine-containing non-metals intact for further recycling. It should be noted that the cost of the equipment available in this method is moderate and leads to sustainable and comprehensive recycling [49,50]. Effective factors in separation efficiency include particle size distribution, feed rate, electric field strength and roller rotation speed. The third well-known and widely used method is Inverse flotation, which is a froth flotation technique that can be used to recycle metals and plastics from electronic waste. This technique is based on the natural hydrophobicity difference of the components and provides the possibility of cost-effective and relatively simple separation. This technique is used to separate non-metallic components in a mixture of crushed waste due to preferential flotation [51]. For e-waste, reverse melting is preferable to direct melting of metals or plastics because the smaller metal fraction is separated and product loss is avoided. It should be noted that metals comprise less than 30% of electronic waste and this amount reaches more than 30% for plastics. Therefore, using this method and floating plastics, the separation of metals will reach the maximum [51]. Due to the hydrophobic nature of plastics and the hydrophobicity of metals in the waste mixture, when air bubbles enter, the plastics will stick to the bubbles and float, and the metals will remain in suspension. This means that the two phases can be separated without the use of chemical additives. The direct flow of metals becomes ineffective due to low natural hydrophobicity [51].

2.2.2. *Chemical recycling*

Although the physical recycling method requires cheap equipment, it has the ability to separate hazardous substances such as halogens and heavy metals from e-waste [52]. For this reason, chemical recycling has also been introduced and three of the most widely used techniques of this method are: dimethylformamide technique, supercritical fluid technique, and pyrolysis. Dimethylformamide is the investigated technique and is used especially for waste printed circuit boards (WPCBs). An innovation in this method is the use of the organic solvent dimethylformamide (DMF) to dissolve the brominated epoxy resin (BER) that binds the layers of metals and glass fibres in WPCBs [53]. This newly developed method can reduce hazardous emissions while separating metals, glass fibre, and BER. The noteworthy point is that in order to increase the recycling efficiency, the solvent dissolution kinetics should be used to optimize the conditions. Supercritical fluid (SCF) is the second emerging method that is also environmentally friendly. The main advantage of SCF treatment is the rapid, complete, and green release of hazardous substances such as flame retardants, and the release of metals for recycling [54]. This method works based on the use of solvents such as water, CO_2, or alcohol in conditions above the critical point, which include fluids that allow them to dissolve and react entirely with organics in the waste [54]. It is also an addition that for future research and industrialization of this method, designing suitable reactors is essential. However, experimental results confirm that supercritical fluids can recycle e-waste [54]. The third discussed technique is Pyrolysis. In general, this process is known as a thermochemical decomposition process that breaks down organic materials by heating them to high temperatures, typically between 300–1200°C without oxygen. Through the thermal decomposition of non-oxygen organic matter, there are many benefits for waste management, energy production and chemical treatment [55]. Another point is that the lack of presence of oxygen prevents the combustion process and gives the opportunity to break down the residual macromolecular structure into smaller pieces. In order to control the relative amounts of transfer due to hydrolysis, parameters such as temperature, heating rate, pressure, and residence time should be controlled [55].

3. Concept of vitrimers

3.1. Covalent adaptive networks

The basis of the design of this class of materials goes back to the concept of covalent adaptable networks (CANs). These networks are also formed using dynamic covalent bonds (DCBs) and energetically, they are part of the bonds that can be broken by applying special conditions or stimuli, such as heat, pH, and UV light (Figure 1(a)) [56,57]. Among these bonds, we can mention esters [58], urethanes [59], disulphides [60], imines [61], silicon-oxygen [62], and boron-oxygen [63]. Here, CANs are divided into dissociative and associative categories (Figure 1(b)), which is based on the reversibility of dynamic bands. The difference is that in the type of associative CANs, the bonds are broken, and new bonds are formed at the same time where density of transverse connections in the polymer network remains constant. However, in dissociative CANs, the elimination/addition mechanism is advanced, the result of which is the breaking of

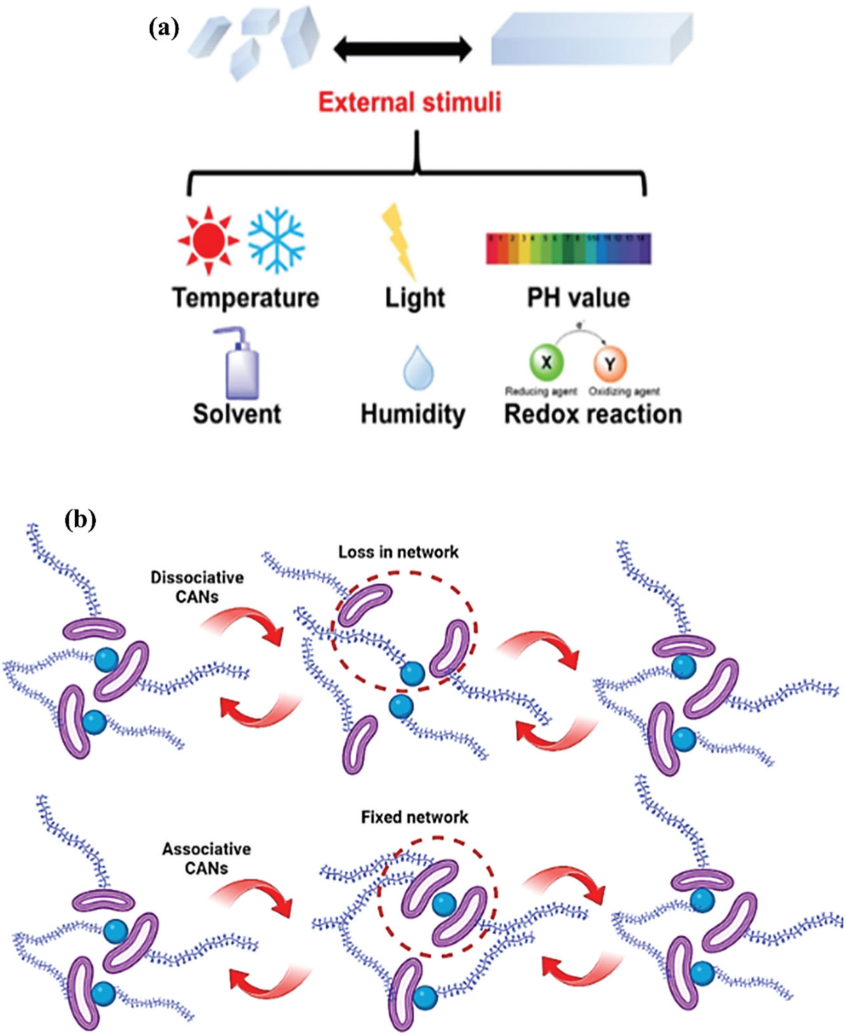

Figure 1. (a) stimuli for CANs (reproduced from reference [56] under open access creative commons license) and references [64] by the permission Elsevier), (b) schematic mechanism of dissociative and associative exchange reaction (reproduced from reference [57] by the permission of Elsevier).

bonds and the failure to re-form all of them, and this means the lack of stability in the density of transverse connections in the polymer network after reprocessing [56,57,64]. Figure 2 shows the types of common chemistries in the synthesis of vitrimeric polymer networks based on catalytic and non-catalytic categories [57,65,66]. It should be mentioned that for a complete study of the types of vitrimer chemistry with details and mechanisms, you can refer to two references [56] and [57] of this article.

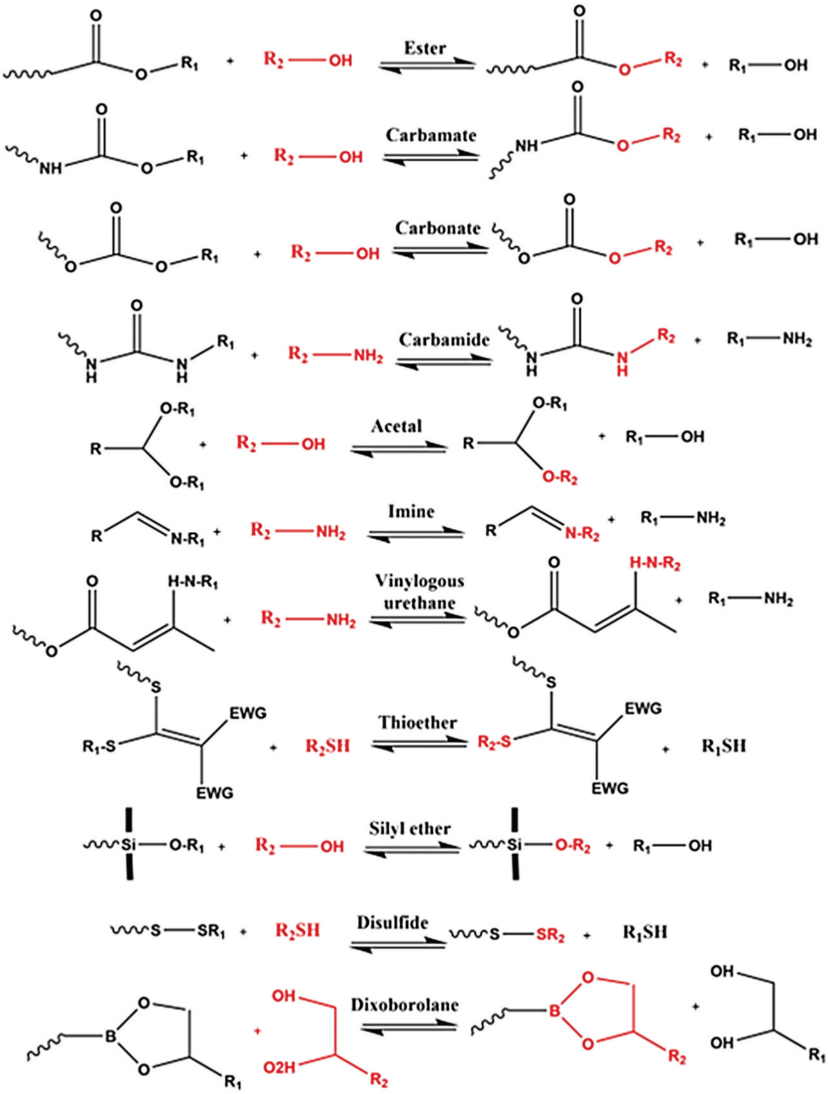

Figure 2. Common DCBs for synthesis of vitrimers (reproduced from reference [57] by the permission of Elsevier and reference [65] under open access creative licence).

3.2. Characterization of vitrimers

3.2.1. Transition temperatures

There are various characterization methods for vitrimers that track and reveal different characteristics. The first one in vitrimers is the transition temperatures. In general, there are two transition temperatures in vitrimers: glass transition temperature (T_g) and topology freezing transition temperature (T_v).

The first transition temperature is the concept of regular and segmental movement of polymer chains, which changes the polymer state from rigid to a mouldable state [56,57,67]. This transition temperature can be measured using dynamic mechanical analysis (DMA) or differential scanning calorimetry (DSC). For the second one, it is worth mentioning that the exchange reactions start around that temperature, and above that, we will have rapid exchange of dynamic bonds. The result of these reactions is the change of the behaviour of the polymer network from viscoelastic solid to a viscoelastic liquid [56,57]. After this transition, the viscosity reaches 10^{12} Pa·s, which suggested as a critical parameter to follow T_v. [64]. Notably, the properties of this vitrimeric network can be overshadowed by the placement of T_v, which can be below or above T_g. In most vitrimers, this temperature is below T_g, so below this temperature, the network is completely rigid and becomes elastic solid as the temperature increases between T_g and T_v, and finally it reaches viscoelastic liquid at T_v. On the other hand, Tv can be potentially below the T_g [12], and in this case, there is no segmental movement below the glass transition temperature, and all the exchange reactions are also frozen, and finally, after passing this temperature, the viscosity decreases. Methods such as dilatometry, stress relaxation, and aggregation-induced-emission (AIE) luminogens can be used for detection of T_v [68].

3.2.2. Chemical characterizations

3.2.2.1. GC-MS. Gas chromatography-mass spectrometry (GC-MS) is a general and versatile tool for samples that contain relatively small molecules. In this technique, there is a heated column that separates the compounds in a sample based on their boiling point difference, and identification is also done by a mass spectrometer. This tool is also very useful for identifying the concentration of components in the sample [69]. This technique is also used in CANs to monitor exchange reactions in small-molecule model compounds linked with the same bonds used in CANs. In this regard, it is possible to link to the desired dynamic band [69]. During these exchange processes, new compounds will be formed that will have different retention times and can be seen through GC-MS [70–73]. Also, by checking the concentration of A-A, B-B and B-A in the sample, the dynamic bond exchange reaction can be checked in more detail, which was done by Snyder et al. in 2018 [74] and Li et al. in 2021 [72]. It is worth mentioning that this technique has also been used to show dynamic balance, which can be attributed to the research of Anales et al. [75]. It is worth mentioning that the coupling of this technique with other existing techniques can also provide a useful tool in investigating the side products of CAN degradation processes, which can be mentioned in the work of Li et al. where coupled TGA-GCMS was used for thiourethanes [76].

3.2.2.2. NMR. Nuclear magnetic resonance (NMR) can also be used to check and track exchanges. The limitation of GC-MS was that compounds with a low boiling point could be detected whereas higher molecular weights can also be studied via NMR. This technique has been used in the study of small molecular models to describe exchange conditions in a variety of cross-linked systems such as polyesters, polyurethanes, and guanidines [75,77,78]. It is worth mentioning that the 1 h NMR technique has been used to investigate the functional groups in the synthesis of CANs and also to optimize the exchange conditions, including the catalyst load and temperature [75]. In addition, this technique has been used to clarify the type of exchange mechanism, such as transesterification kinetics via 1 h NMR for phthalate monoester compounds [79]. In addition, its NMR studies made it possible to classify exchange reactions into two cases, associative or dissociative. The kinetics of imine exchange via transimination and imine metathesis mechanisms for hydrocarbon and ethylene oxide-based polymers are also studied by Schoustra et al. via this method [78]. In addition to thermally activated exchange processes in CANs, photo-activated exchange processes have also been carried out and followed using NMR technique. In this regard, Barsoum et al. showed that the use of photo-switchable internal catalysis can control the bond exchange kinetics between boronic ester and free diol [80].

3.2.2.3. FTIR. Fourier-transform infrared spectroscopy (FTIR) has a more limited application than the other mentioned methods in describing the bond evolution in the covalent dynamic network. However, this technique can be used in the destruction of these networks. In this regard, Gamardella et al. have investigated the degradation mechanisms in thiourethane CAN [81]. Taplan et al. using ATR-FTIR also proved that the chemical composition of CANs does not change during recycling [82,83]. In addition, variable temperature FTIR (VT-FTIR) can be used to describe and track the changes of bonds in a vitrimeric network containing CANs with temperature [84,85]. In this regard, Bongiardina et al. used the VT-FTIR technique to determine the kinetics of bond formation in the thiol-thioester reaction by measuring time to check the dynamic equilibrium [86]. Therefore, it can be concluded that the FTIR technique is useful when the change of functional groups is severe, and this requires that the functional groups are presented with a high concentration during the exchange process. In addition, FTIR characterization is not well-suited to solvent-borne exchange reactions, as the signal from exchange compounds is often drowned out by the solvent signal [69].

3.2.3. Recyclability and reprocessing

Some techniques can be used to check the recyclability process of vitrimers, including rheometry and DMA, stress relaxation, and dilatometry. The first two cases, rheometry and DMA, both can be used to evaluate the viscoelastic properties of the dynamic network [56,57,87], and the difference between these two methods is in the type of measurement, which originates from the difference in the type of applied force. This is because compressive forces in DMA and shear forces in rheometry have priority [88]. In the explanation of the third possible method, stress relaxation, it should be added that vitrimers are subjected to a constant strain and the resulting stress during the test is checked by measuring the relaxation modulus [89–91]. In the last method, dilatometry, the difference in the volume occupied by a thermoset and a vitrimer is compared. The thermosets occupy a smaller volume due to non-dynamic bonds and experience a lower coefficient of thermal expansion [87,92–94]. There are techniques for tracking the reprocessing of vitrimers, which can be mentioned such as compression moulding, welding, and extrusion. Finally, there are also techniques for tracking the reprocessing of vitrimers, which can be mentioned, such as compression moulding, welding, and extrusion [94–96].

3.2.4. Degradation temperature

Degradation temperatures are important because dynamic bonds in CANs are often activated by heating [83,97]. This temperature is defined as a temperature above which an irreversible change is observed in the structure of the polymer network. The application temperature of vitrimers should be lower than this temperature in order to prevent the chemical destruction of the network [98]. The technique used to determine this temperature is thermogravimetric analysis (TGA), which is used by measuring the change in mass as a function of temperature in a controlled atmosphere. Generally, in polymer networks without solvent, the degradation temperature is the temperature at which the polymer network loses 5% of its mass [69].

4. Application of vitrimers in advanced electronics

4.1. Vitrimer-based triboelectric nanogenerator and wearable electronics

The most electronic devices are based on energy storage systems such as batteries and capacitors. For the development of wearable electronics, it is needed to reduce the dimensions of the design and reduce the weight of the parts, which is necessary to have a table and sustainable power supply. Considering this point, it should be said that so far mechanical energy caused by human movement in wearable electronics has been the ideal source of energy. Therefore, the use of traditional electromagnetic generators is no longer compatible, and due to reasons, such as heavy weight, bulky volume, and inflexibility, they are in the role of energy harvesters. Therefore, today, triboelectric nanogenerator (TENG) technology that is based on contact electrification and electrostatic induction has become a promising source that could harvest energy from the surrounding environment, which could be used as wind, waves, vibration, motion, and even friction [99–103]. Firstly, we studied elastomeric vitrimer network containing silver nanowire using disulphide dynamic bands, which was used for the synthesis of mechanical energy harvester and tactile sensing element, *i.e*. designing a vitrimer based triboelectric nanogenerator (VTENG) (Figure 3(a)). Among the properties resulting from this design, we can mention fast structural/functional recovery due to the presence of disulphide bonds and shape adaptivity or configurability, and this self-repair helps increasing the lifespan of the energy harvester, which is a new method in the design of flexible TENG (Figure 3(b)). Also, other properties can be mentioned as changing 2D to 3D structures of this flexible and stretchable VTENG, which can be enlarged in the form of jigsaw puzzles [104]. The next case in the field of self-healable triboelectric nanogenerators (SH-TENGs) has taken advantage of another vitrimerization chemistry. According to Cheng's group's results, one of the mechanisms for designing dynamic bands to achieve vitrimers is hindered urea chemistry employing a bulky urea group, typically *t*-butylamine group, attached to one of the urea nitrogen atoms [105,106]. Among the advantages of using this chemistry, it is degradable and less expensive, and by using facile step-growth polymerization of various polyamines, including multifunctional bulky amines with polyisocyanates, they can be easily synthesized [107,108]. In this case, reprocessable PHU networks have been used, focusing on the synthesis of PHU networks centres on the synthesis of a four-arm star-hindered amine (T-NH) that can be used for step-growth polymerization with a polydimethylsiloxane (PDMS)-based diamine (PDMS-DA) and a polyisocyanate under catalyst-free condition (Figure 4). The examination results of this vitrimeric network have shown that the network is flexible and has excellent mechanical properties, such as tensile strength as high as 1.7 MPa at break. It is also worth mentioning that this network has shown fast and cyclic self-healing properties and is reprocessable under mild conditions, enabling the recovery of triboelectric performances after the complete healing of the damaged surfaces. In addition, surface polarization increases the dielectric constant with the highest triboelectric output performance (169.9 V/cm^2), which can be used in the design of mechanical energy-harvesting devices with stretchability and high recoverability properties (Figure 4) [109].

In the continuation of the investigations related to triboelectric nanogenerator (TENG) as a soft electrical power source, it has been emphasized on properties such as self-healable, stretchable, and transparent at the same time with dynamic band chemistry of imine. In this regard, a thin film with buckled Ag nanowires/poly(3,4-ethylenedioxythiophene) composite electrode is sandwiched with self-healable PDMS elastomers [110]. By activating the imine dynamic mechanism,

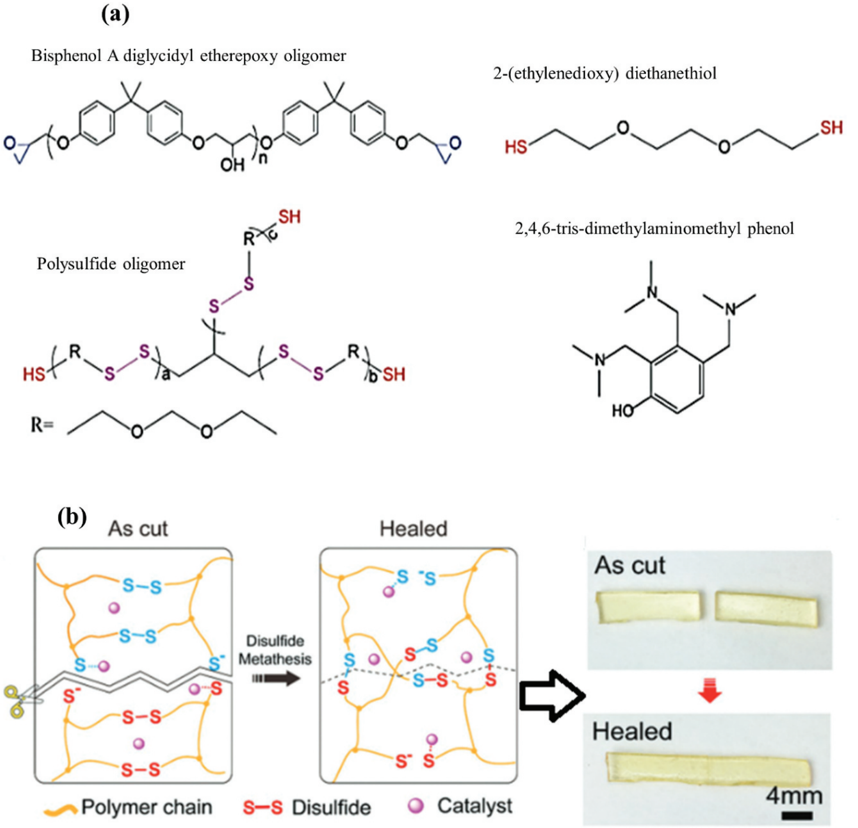

Figure 3. (a) chemical structure of the materials used in vitrimeric polymer network synthesis, (b) self-healing results via disulfide metathesis catalyzed by an organic base in vitrimer network (reproduced from reference [104] by the permission of John Wiley and sons).

the repair efficiency of 94% has been reported, and by adjusting the buckling wavelength of the electrode, two properties of stretchability and transparency of the soft TENG can also be controlled. It is also reported that TENG with 50% stretchability and 73% transmittance can recover the electricity generation even after a cut, which means 100% recovery efficiency [110].

The use of repairable and recyclable triboelectric nanogenerators based on an epoxy-derived vitrimer (EV) is also studied. Here, flexible and elastomer-like EV films can act as the tribo-material, which are prepared by solvent-free, and quasi-thermoforming approach. To design this vitrimeric network, trans-esterification-type bond-exchange reactions have been used, and the excellent mechanical properties and recyclability of the materials are among its most important characteristics. Also, the results showed that by taking the

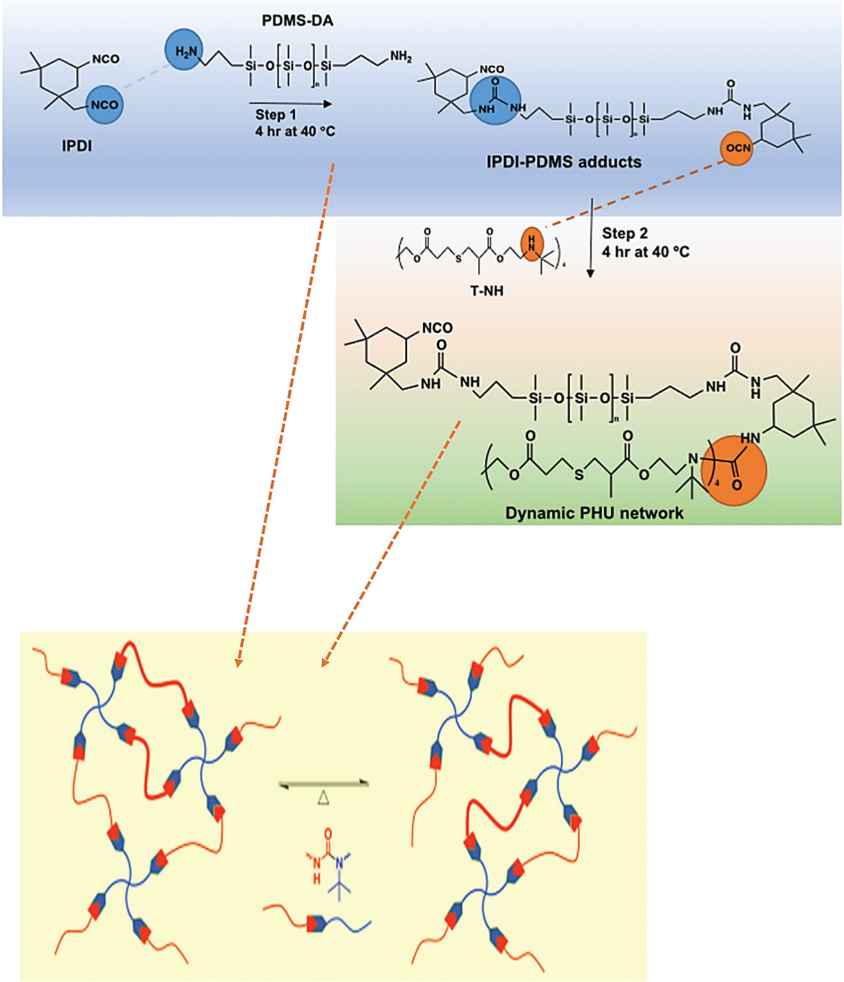

Figure 4. Overview of reversible exchange reaction of dynamic-hindered urea bonds in PHU network during heating and two step reaction paths for PHU vitrimeric network (reproduced from reference [109] by the permission of ACS).

triboelectric signal as a probe for network evolution, there is reparability and recyclability, and 98% repair is reported [111]. Imine-vitrimer elastomer (IVE) has also been used for the development of flexible and compressible TENG with circular life cycles and outstanding electrical output. This vitrimeric network contains PDMS segments, which are synthesized by cross-linking 3-methoxy-4-(oxiran-2-ylmethoxy) benzaldehyde (MB) with PDMS-NH_2, the TENG designed with this vitrimeric style is known as IVE-TENG. Due to imine bonds, compressibility along with high electrical output has also been observed, so that output voltage density and short-circuit current density of

IVE-TENG are measured to be 506.4 kV m^{-2} and 21,620 μA m^{-2}, respectively [112]. Also, stretchable and self-healable vitrimer triboelectric nanogenerator (VITENG) with the property of high stretchability and good healability is investigated, which is based on elastomeric PDMS-based vitrimer (VPDMS) which compensates for the mechanical defects of TENG. This vitrimeric network is synthesized from diacrylate poly(dimethylsiloxane) (AA-PDMS) *via* a fast thiol-Michael reaction, which includes 2,3-dihydroxypropyl methacrylate (DHPMA) and increases the flexibility and transesterification reaction efficiency. This factor has caused 100% repair efficiency and also showed good performance with an output voltage of 135 V under a load of 10 N [113]. Ionic cross-linked transcarbamoylation vitrimer network has also been used, which has the properties of triple shape memory and thermomechanical performances. For the synthesis of this vitrimeric network of dual-cross-linked poly(acrylonitrile-*co*-butyl acrylate-*co*-hydroxyethyl methacrylate-*co*-zinc methacrylate) (Zn-PABHM), a facile and one-pot free radical polymerization is used. Based on Figure 5(a), it is clear that a divinyl carbamoyl derivative (HEMAH) was synthesized from the reaction between 1,6-diisocyanatohexane and hydroxyethyl methacrylate, which acted as a crosslink for dynamic transcarbamoylation anchor point. Then, dual cross-linked poly (acrylonitrile-*co*-butyl acrylate-*co*-hydroxyethyl methacrylate-*co*-zinc methacrylate) (Zn-PABHM) is synthesized (Figure 5(b)). The results showed that the presence of ionic cross-linking helps stabilizing the permanent shape and two temporary shapes of the obtained vitrimer ionomer [114]. The use of ionic liquid to dimethylglyoxime-urethane (DOU)-based CANs is reported to adjust both reversible bond dynamics and polymer chain mobility using chemical coupling and physical lubrication. This synthesis was based on the introduction of functional small-molecule additive, an ionic liquid (1-ethyl-3-methylimidazolium bis(trifluoromethyl-sulfonyl) imide, [EMI][TFSI]) to DOU group, in which the presence of liquid ion can catalyse dynamic band exchange [50]. Also, the ionic liquid has the ability to break the hydrogen bond between the chains and cause an increase in mobility, which results in a decrease in the activation energy of dynamic reactions from 110 to 85 kJ mol^{-1}. In addition, the presence of the ionic liquid causes conductive properties and the concept introduces ionic CAN (iCAN) which can be a powerful tool in the construction of wearable electronics responsive to multiple stimulations including temperature, strain, and humidity [115].

4.2. Vitrimer-based electro-skin

TENGs have been highly regarded in the application of electronic skin due to their flexibility, structural diversity, and self-powered properties [116–118]. Double-crosslinked self- healing PDMS (DCS-PDMS) was considered to achieve elasticity and special mechanical properties. In this network, imine chemistry

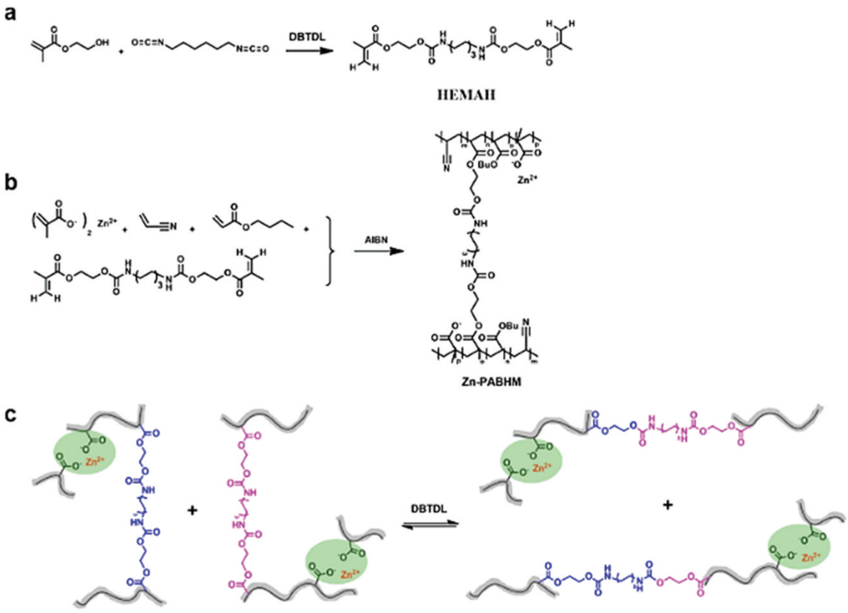

Figure 5. (a) synthesis of carbamoyl-containing divinyl crosslinker (HEMAH), (b) synthesis of Zn-pabhm vitrimer, (c) schematic of exchanging mechanism via transcarbamoylation reaction in the Zn-pabhm vitrimer (reproduced from reference [114] by the permission of ACS).

was also used for dynamics, which ultimately self-healed the network due to the formation of H-waves between MXene and lightly cross-linked PDMS. According to Figure 6, DCS-PDMS is synthesized using polycondensation reaction of aminopropyl terminated PDMS (NPDMS) with isophorone diisocyanate (IPDI) and terephthalaldehyde (TPAL). It is worth mentioning that the presence of isocyanate group of IPDI and the amino group on NPDMS can form urea and cause hydrogen bonding; so, IPDI can cross-link the aldehyde group on TPAL and the amino group on NPDMS. Also, the reports related to the self-healing of the samples are also shown in Figure 6 [119].

4.3. Vitrimer-based sensors

The use of elastomer-based devices is a key practice in the design of stretchable electronics, which today includes strain sensing (Figure 7(a)) [120–122]. One of the requirements for creating intelligent interaction between electronics and users is to introduce human-readable visual readouts such as colour diversity. In this regard, the use of structural colours derived from the interaction of light with nanostructures based on photonic crystals (PCs) is used [123,124]. One of the most important innovations was created when mixing

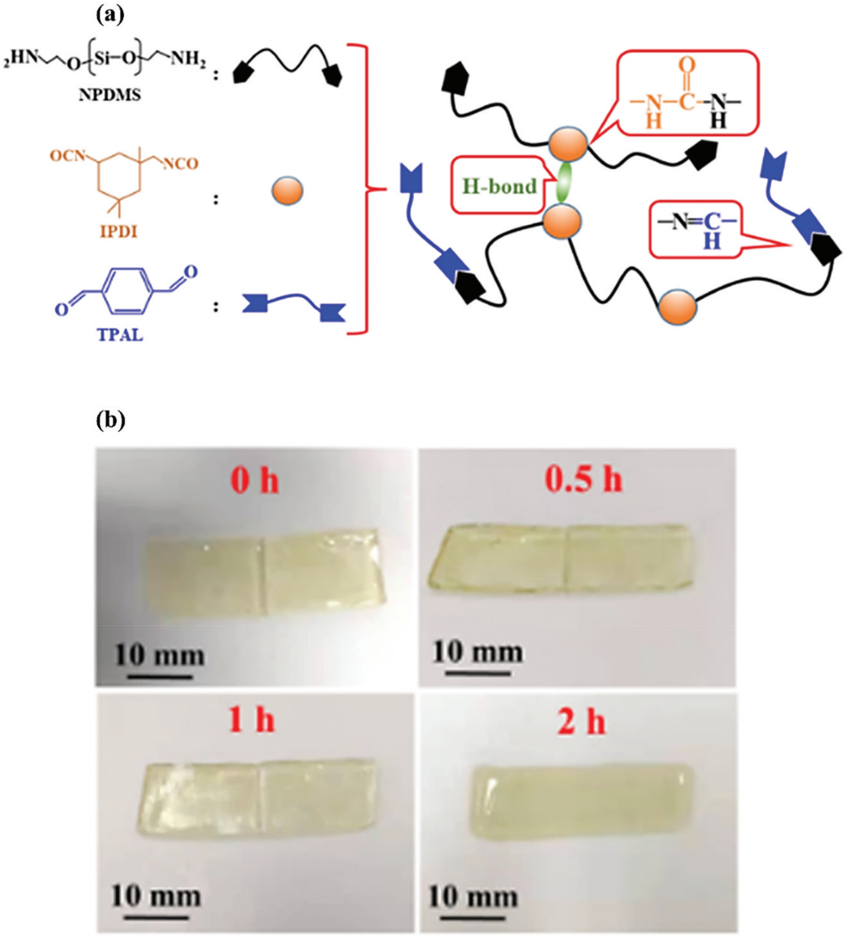

Figure 6. (a) synthesis of DCS-PDMS, (b) self-healing behavior (reproduced from reference [119] by the permission of Elsevier).

colloidal PCs with elastic polymer matrices leading to colour change during stretching [125,126]. One of the studies conducted in this regard is photonic vitrimer-based electronics (PVBEs) through manipulating synchronous structure variations of PCs and carbonized cotton textile (CCT) induced by vitrimer deformation. Therefore, poly(urethane-urea) vitrimer elastomers were synthesized using the polycondensation method between polytetramethylene ether glycol (PTMG), poly(1,4-butanediol) bis(4-aminobenzoate) (PBDAB), isophorone diisocyanate (IPDI) and glycerine (GLY). The results showed that in addition to excellent mechanical properties and high flexibility, 93% efficiency of self-healing has also been achieved (Figure 7(b)) [127]. Also, the introduction of PCs and CCT into vitrimer caused

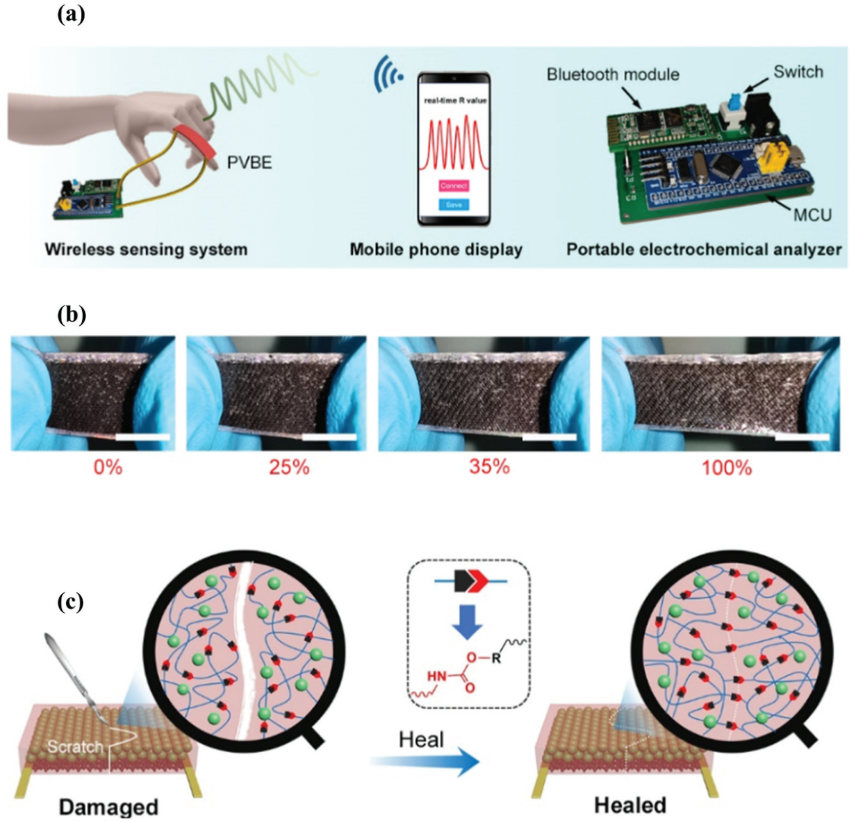

Figure 7. (a) PVBE incorporated with a controller unit for wireless sensing system, (b) PVBE upon various strain content, (c) PVBE self-healing mechanism via exchange reaction (reproduces from reference [127] by the permission of Elsevier).

simultaneous colour variation and electromechanical performance upon stretching. However, the self-healing mechanism in this vitrimeric network was the exchange of dynamic urethane bonds. In such a way that the interface between the two parts is separated, new covalent bonds are formed by bond exchange reactions (Figure 7(c)) [127,128]. In addition, high-performance composite strain sensors based on vanillin vitrimeric network have also been discussed, and silver nanowires (AgNW) have also been used due to their excellent conductivity and mechanical flexibility. It is also worth mentioning that one-pot reactions involving the thiol-epoxy and aldehyde-amine were carried out to synthesize vanillin-based vitrimers. The results showed that good adhesion and stable properties between the lower and upper layers were achieved due to the proper interaction between the film and the filler, and high UV absorption ability was observed due to the formation of Schiff base network. It also confirms the results of wide linear response range (0 to 100%) and a high gauge factor (15.8) [129]. The synthesis of self-healable photonic vitrimer using photonic crystals based on elastomeric matrix is also reported. At first, elastomeric vitrimers were synthesized by polycondensation of hydroxyl-terminated polybutadiene (HTPB), polytetrahydrofuran (PTMG), glycerol (GLY), and isocyanate (IPDI). Here, in addition to the attractive property of self-healing, optical functions, specifically bright structural colour and colour-changing ability under strain, *i.e.*, mechanochromism, are also visible. Other reported properties of this elastomeric vitrimeric network include high toughness, large strength, excellent optical creep-resistance, and excellent durability. Due to the band gap of photonic crystals and the presence of dynamic covalent network, self-repairing capability of structural colour and visualized mechano-chromism during stretching is also available, which causes the use of this vitrimeric network in interactive sensor for visual monitoring human motion without an external power supply [130]. The use of MXene acrylate vitrimer network (MAVIN) in the application of electromechanical sensor development is also reported. This vitrimeric network has a low glass transition temperature, which is due to the use of flexible siloxane backbone (PDMS) in the process of vitrimer synthesis [131]. Also, the high concentration of hydroxyl groups, particularly in the proximity of ester bonds (β-hydroxyl ester), can accelerate the transesterification rate [132]. It is noteworthy that the activation of this exchange reaction requires heat, which may cause the oxidation of MXene in the presence of solvent [133], which can be prevented by a thiol-Michael addition reaction. Also, due to strong microwave absorption by $Ti_3C_2T_x$, damaged MAVIN sensor can be repaired by microwave radiation with a high healing efficiency of 92.4% within 1 minute and voltage stability of 3 volts [130].

4.4. Vitrimer-based supercapacitors and electromagnetic shielding

Smart devices based on paper-based composite are also a group of materials that have recently received much attention in electronics, especially cover electronics and flexible energy storage and supercapacitors [134,135]. In this application, one of the researches deals with the synthesis of flexible original paper with pencil-drawing (OPD) @ polyaniline (PN)-vitrimer (V) hybrid using a combination of painting, electropolymerization, and dip-coating methods. Therefore, paper-based materials are classified into three layers, one layer of conductive graphite is on the (near) surface of both sides of the paper covered with pencil, which makes the paper (near) surface conductive and creates conditions for electropolymerization of high capacity PN nanofibers. The next layer is vitrimer (V), which is incorporated into the OPD@PN hybrid on the (near) surface of both sides of paper, and OPD@PN-V layers are used as supercapacitor electrodes and an insulating separating layer between the two electrodes [136]. It should be mentioned that a double ring precursor was prepared for the vitrimer synthesis at first. For this, diphenyl carbonate was used to heat at 140°C and after adding bis(trimethylolpropane), the reaction was carried out at the same temperature for 48 hours. After that, ether is added to the obtained bicyclic precursor and it was stirred for two hours to remove diphenyl carbonate. Bicyclic precursor and tris (2-aminoethyl) amine dissolved in a proper amount of DMSO were used for the synthesis of vitrimer (Figure 8(a)). The results show that in addition to the special properties obtained by the supercapacitor such as easy moulding, self-healing, and shape memory, ultrahigh gravimetric and areal specific energy (power) density of 56 Wh kg^{-1} (78 kW kg^{-1}) and 785 μWh cm^{-2} (286 mW cm^{-2}) has been obtained in terms of cyclic stability [136]. It is also worth mentioning that one of the inevitable growing pollutions in the world is electromagnetic pollution. In addition, due to the growth of the use of flexible wearable electronics, the use of high-efficiency electromagnetic shielding (EMI) is necessary to maintain human health [137,138]. In this regard, the research study confirms the synthesis of a vitrimeric composite

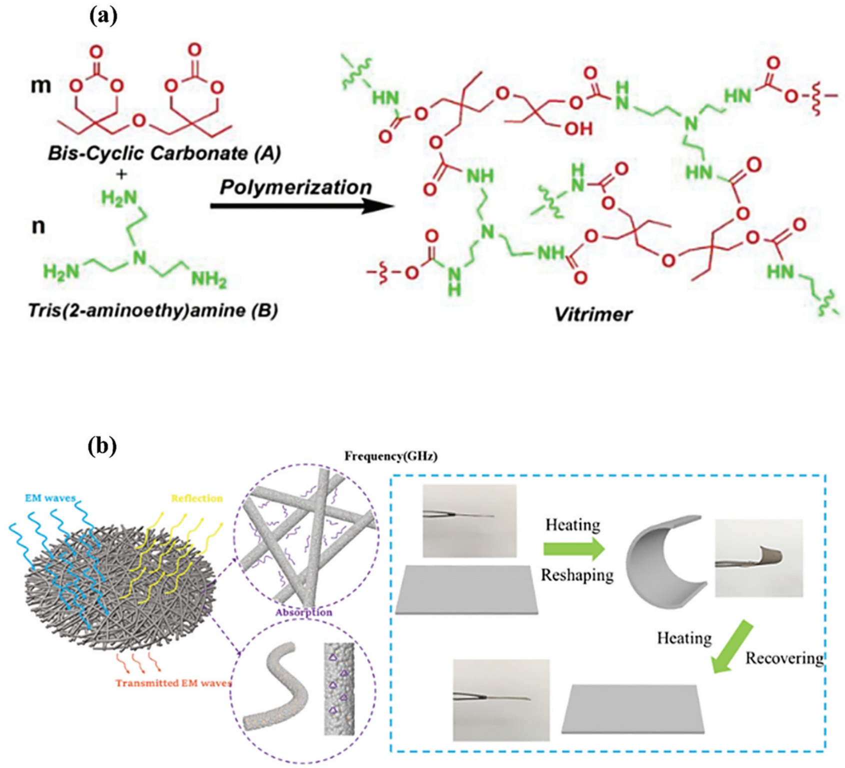

Figure 8. (a) synthesis of vitrimer (reproduced from reference [136] by the permission of Elsevier), (b) EMI shielding mechanism of CPPA composite and CPPA-V composite shape memory behavior (reproduced from reference [139] by the permission of ACS).

that is prepared from growing metallic silver nanoparticles inside the cellulose paper (CP) modified with polydopamine (PDA) and is called CP@PDA@Ag (CPPA) composite and has properties such as induced electromagnetic shielding, sensing, Joule heating, and antimicrobial. Also, due to its high flexibility, this composite has the ability to bend and twist, and due to the synergy of silver nanoparticles and porous conductive fibre networks, it has high conductivity, *i.e.* 2090 S/m and remarkable EMI shielding performance reaching 51.3 dB in the thickness of 0.17 mm. To achieve the shape memory properties of this composite, a vitrimer has been used by polymerizing a bicyclic precursor with tri (2-aminoethyl) amine and it is called CPPA-V, which is shown in Figure 8(b) [139].

4.5. Vitrimer-based soft actuators and soft robotics

Sensors and transducers provide the interaction between robots and the environment [140,141]. Among the disadvantages in their design, we can mention rigidity, lack of self-healing, and biodegradability [142–144]. In other words, electronics are designed without considering the issue of longevity and stability. Today, in this field, the focus is on introducing the concepts of environmentally friendly, degradability, and recycling in order to avoid scarcity, toxicity, short lifetime, and recycling complexity, and this is because electronic waste is the fastest growing type of waste. Therefore, in this regard, one of the reports on biobased and biodegradable vitrimer as a binder for a conductive ink comprising graphene nanoplatelets (GnPs) and carbon nanofibers (CnFs) has been used [145]. It is worth mentioning that this ink is mentioned as green ink because soybean oil as a precursor was used in the synthesis of vitrimer along with ethanol solvent. These inks can be applied to different substrates such as natural rubber, paper, and glass. They have also been used in human-imitating soft robotic skin due to its self-healing properties with a glass transition temperature of 10°C, suitable mechanical properties, and low resistance conductivity (10^{-4} Ω m). In addition to the above, achieving characteristics such as temperature sensitivity, degradability, recycling of the substrate and the coating itself in ethanol solvent for this synthetic vitrimer is also worth mentioning (Figure 9) [145]. It should be mentioned that soft actuators have the ability to change their shape, volume, and dimensions due to the application of an external stimulus such as light [146,147], heat [148–150], electricity [151,152], moisture [153,154], magnetism [155], and solvent [156], which can lead them to be nominated in the development of smart and sustainable electronics [157–159]. Therefore, in this application, it is important to apply the special properties obtained in other investigated cases. In this regard, multifunctional, recyclable, thermosetting, and vitrimer-based soft actuators are reported that respond to four

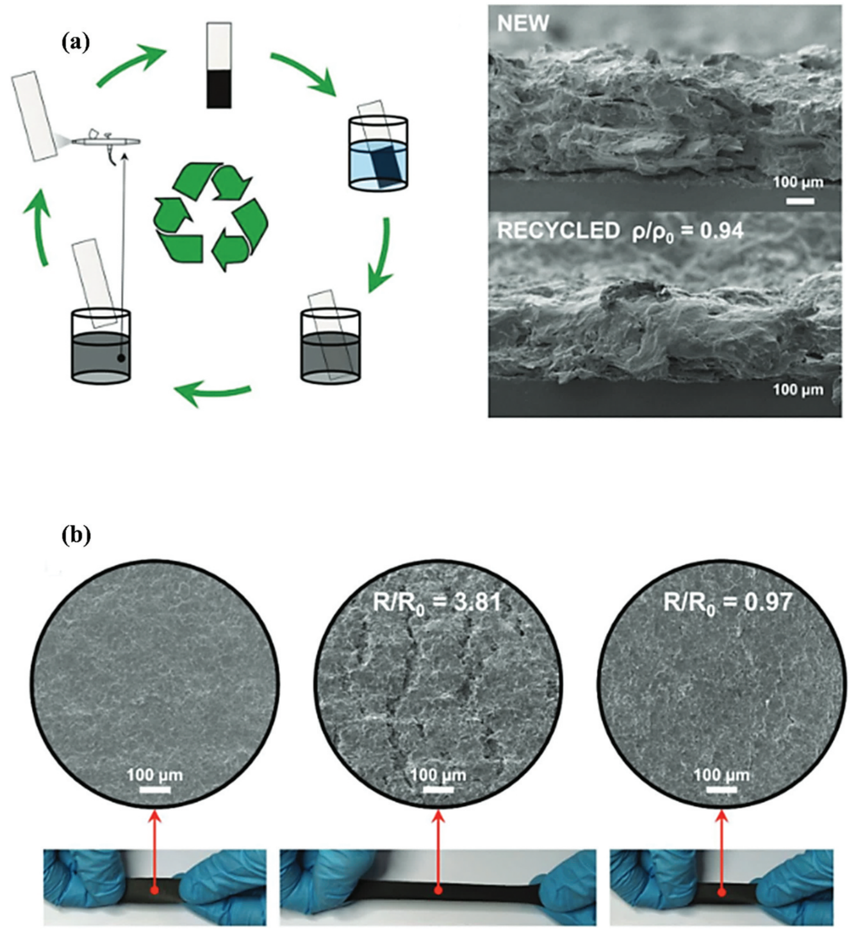

Figure 9. (a) vitrimer conductive coating (VCC) recycling process with SEM images of the neat and recycled VCC20 coating on glass via ratio between the resistivity obtained after recycling neat sample, (b) vitrimer conductive coating strain and strain relaxation behavior for 24 h (reproduced from reference [145] under open access creative license).

stimuli. For the synthesis of this vitrimer network, hot-pressing carbonized silk fabric (CSF) into the surface of vitrimers was used and it was called CSF-vitrimer. The vitrimer of the epoxy is synthesized from the mixture of diglycidyl ether of bisphenol A and dodecanedioic acid, heating at 180°C, and the presence of triazabicyclodecene [160].

4.6. Recyclable vitrimers for flexible electronics

Today, the use and development of flexible electronics has become a serious competitor for electronics based on Si wafer or glass plate [161,162]. Flexible electronics can revolutionize the future of the electronics industry due to features such as lightweight, robust, stackable, foldable, and the ability to be twilled by printing method [163,164]. For sustainable electronic design, it is very common to use two polymers, polyethylene terephthalate (PET) and polyethylene naphthalate (PEN), which are also recyclable and rehealable. However, due to the linear structure of these polymers, they have poor dimensional stability at high temperature and inappropriate resistance to solvent [165]. Generally, to strengthen these properties, metallic nano-wires /nanoparticles or metal liquids are used, which can cause metal oxidation, toxicity, leakage, and limited stretchability and bendability. Nowadays, the use of malleable thermosets has been suggested as a sustainable alternative way due to the presence of features such as excellent mechanical properties, high resistance to solvent, reprocessability, and recyclability [166,167]. In this classification, one of the attractive polymers is cross-linked polyimines, in which bond exchange reactions through reversible imine chemistry can be activated [168,169]. This network can be reprocessed using the depolymerization reaction and completely recycled, which also have restorative properties. Therefore, in this regard, one of the studies deals with the synthesis of flexible conductive composite based on malleable polyimine thermosets and multiwalled carbon nanotubes (MWCNTs). The results of this network showed that the production method was very cheap and simple, and at the same time, they showed good conductivity, bendability, and stretchability, and also rehealability, reprocessability, and recyclability (Figure 10) [170]. Another result is that this network can be easily changed into different shapes without compromising the conductivity. The reason for using MWCNTs as a conductive filler is its unique tubular structure, extremely high aspect ratio, and excellent stiffness and strength [170].

4.7. Recyclable vitrimer-based printed circuit boards

At the end of the life of electronic components, printed circuit boards (PCBs) constitute a significant part of electronic waste that is dangerous for the environment. Due to the fact that most of them are composed of heat-hard epoxies and are irreversible, their recycling faces challenges [171]. Therefore, in order to solve this challenge of recyclability, steps have been taken that are mentioned in one of the studies using sustainable PCB solution using transesterification vitrimers (vPCBs). In the recycling cycle, it is worth mentioning that at first fibre-reinforced vitrimer (GFRV) composites are prepared. In the second stage, by using the ability of exchange bands in the vitrimeric network, physically damaged vPCBs have been repaired for more than four cycles while maintaining the mechanical and electrical properties. The third stage also belongs to the overall recycling of vPCBs. In this stage, some solvents cause swelling of vitrimer, which will cause disintegration of

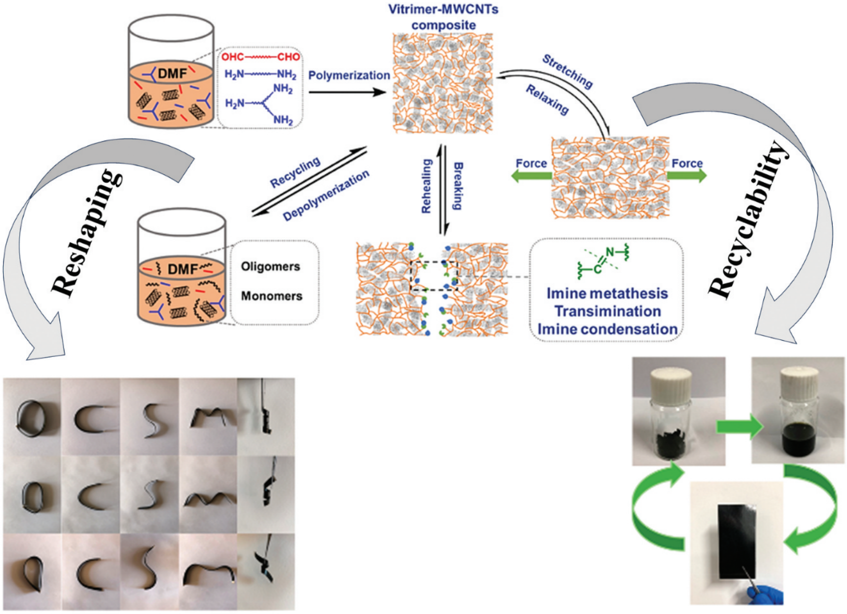

Figure 10. Schematic for synthesis of flexible vitrimer-mwcnt composite with reshaping, recyclability and rehealability mechanism and results (reproduced from reference [170] by the permission of ACS).

transesterification vitrimer composites with solid inclusions and metal attachments (Figure 11) [172]. The results report 98% recovery of the vitrimer polymer and 100% recovery of glass fibres, and even a 91% recovery of the solvent at the end of the recycling process. Also, the results of the comparison between vPCB and PCBs in life-cycle assessment (LCA) have shown 47.9% improvement in global warming potential (GWP), 65.5% in ozone depletion potential, and 80.9% in human cancer [172].

4.8. Vitrimer-based ionotronic devices

Ionotropic is a concept that is effective for integration with electronics. In addition to electrons, ionotronic devices also rely on ion-based charge effects. These devices have created a new field for creating electrically switchable memories [173]. Ionogels are a group of materials that consist of ionic liquid enclosed in a polymer network that forms the mobile ions of the ionic headlight and the presence of the polymer network causes softness, elasticity, and stability. These materials can be a blend of thermoplastic and ionic liquid (IL), or confinement of IL in a cross-linked polymer network [174] and they have abilities such as healing, welding, reshaping, and recycling which makes them a hope for sustainable soft electronics. In this regard, researchers vitrimer ionogel based on

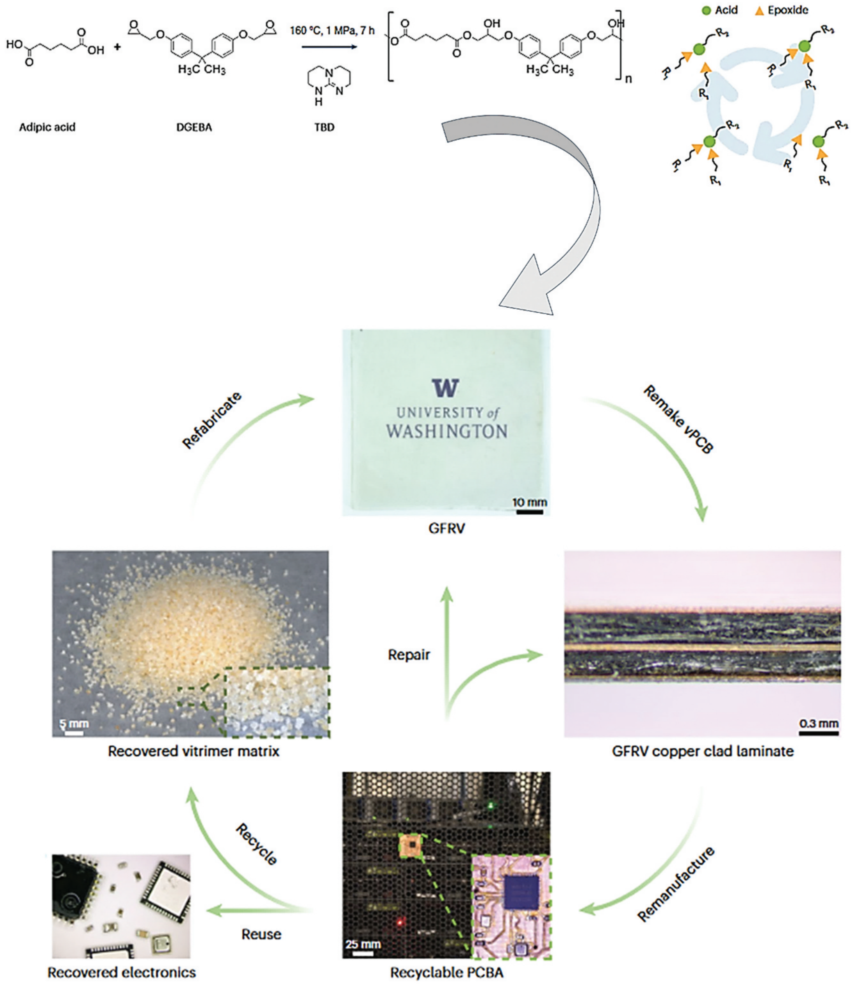

Figure 11. Synthesis of glass fiber-reinforced vitrimer composite with bond exchange via enables healing and recyclability when heated beside fully recyclable PCB process (reproduced from reference [172] by the permission of springer nature).

transesterification was synthesized *via* the epoxy ring-opening reaction with carboxylic acids of functionalized poly (ethylene glycol) (PEG) and triglycidyl ether crosslinkers in the presence of the ionic liquid 1-ethyl-3-methylimidazolium bis (trifluoromethylsulfonyl)imide (EMIm TFSI) (Figure 12) [175]. The results showed that due to the plasticizing effect and the introduction of mobile ions, in the presence of 50 *wt*. % EMIm TFSI, 300% strain, low elastic modulus (60 kPa) and 2.6 × 10 4 S cm^{-1} conductivity have been obtained. Also, the presence of EMIm TFSI increases the chain mobility, which facilitates the transesterification reaction of exchangeable β-hydroxy ester linkages, which results in increased

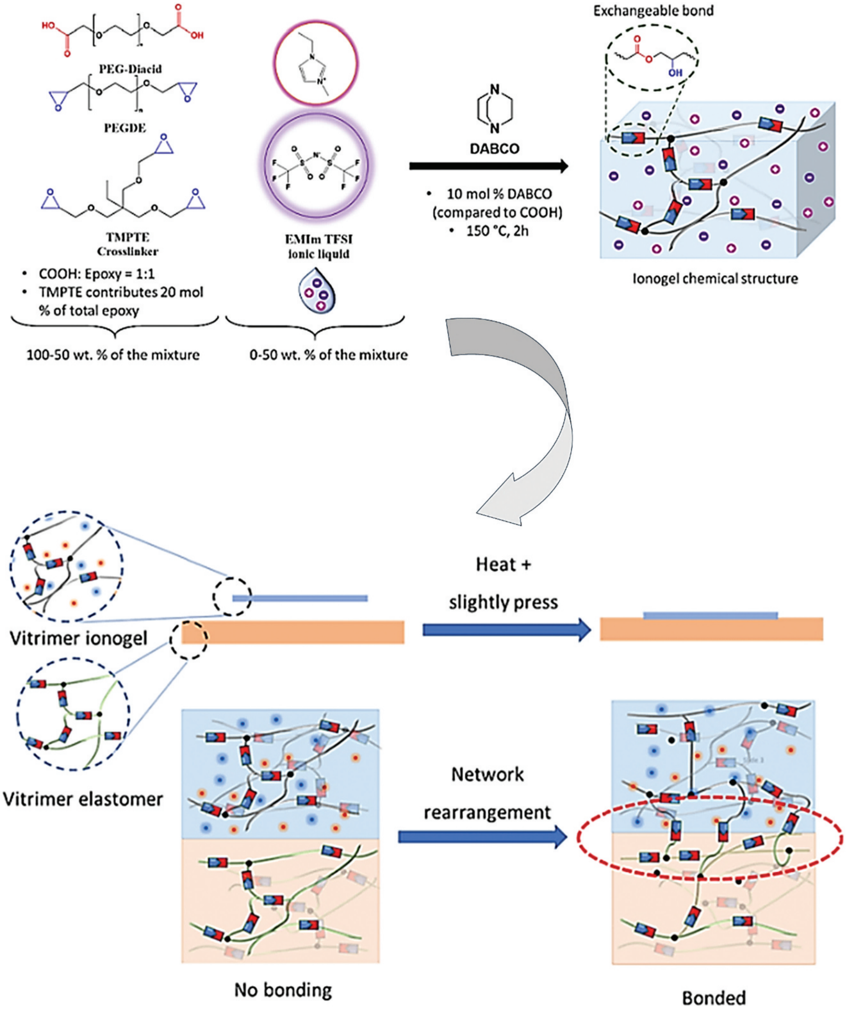

Figure 12. Synthesis route of ionogel-based vitrimer with surface bonding mechanism (reproduced from reference [175] by the permission of Elsevier).

efficiency. This vitrimeric network was also used as a strain sensor, soft electrode, and ionic cable [175].

4.9. Vitrimer-based moist-electric generators and thermoelectric devices

Another interesting recent application is related to unique microscopic interactions of water with functional materials in order to develop moist-electric generators (MEGs) and extract green energy directly from water [176–178] because water, as a recyclable resource, absorbs 35% of the incident solar energy [179]. Finally, problems such as non-recyclability, lack of self-healing, weak mechanical resistance and other issues caused vitrimers to enter the field in this application as well. Therefore, in this direction, moist-electric generators (MEGs) based on a polyhydroxyurethane (PHU) vitrimer have been developed. This vitrimer was synthesized based on amines and cyclic carbonates in an environmentally friendly way (Figure 13), which is dynamic covalent bond-based networks with plenty of dangling hydroxyl groups. The results showed that unlike traditional polymer MEG, vitrimer moist-electric generators (VMEGs) have shown high and stable voltage outputs at high temperature and high humidity without structural collapse [180]. Also, after doping with Ti_3C_2, VMEG finds the ability to simultaneously convert water and solar energy into electrical energy and provide continuous power for electronic devices, such as timers, under a natural environment. The property of self-healing and transparency is also noteworthy for these VMEGs [180]. In recent years, thermometric devices have been another concept that has been used to generate energy and develop sustainable electronics. In this application, like other applications, the resulting waste has become a challenge that can limit the scope of application of these devices [181,182]. Therefore, in this direction, an epoxy vitrimer matrix containing dynamic silyl ether bonds has been used to develop stable thermoelectric generators (TEGs) with the ability to recycle and reprocess. The results have shown that the use of dynamic silyl ether linkage was employed as the encapsulation device inducing excellent mechanical properties, conformability to surfaces, and more importantly, reprocessability. Another feature of this system is low thermal penetration, which supports suitable thermal insulation. Compared to common TEG, vitrimer encapsulated TEG system has shown higher power generation up to 70 mW for temperature gradients below 30 K that is enough as a power source for a small LED. Also, this system has the ability to be separated for reuse, as well as the ability to recycle that the remanufactured device retains its power generation performance [183].

5. Remarkable conclusions

Existing traditional methods for recycling polymer waste as an important concern of the world have some merits. Expenditure, polymer molecular weight loss, solvent recovery, existence of energy-consuming processes such as crushing, cleaning, pressing and heating are the most famous of them. These reasons caused attention to be focused on vitrimer strategy as a new method to maintain the structure and final properties of polymer.

The synthesis and development of the new materials with special properties is imperative in electronics developments today. This however leads to a substantial rise of electronic waste, which can be a serious warning for

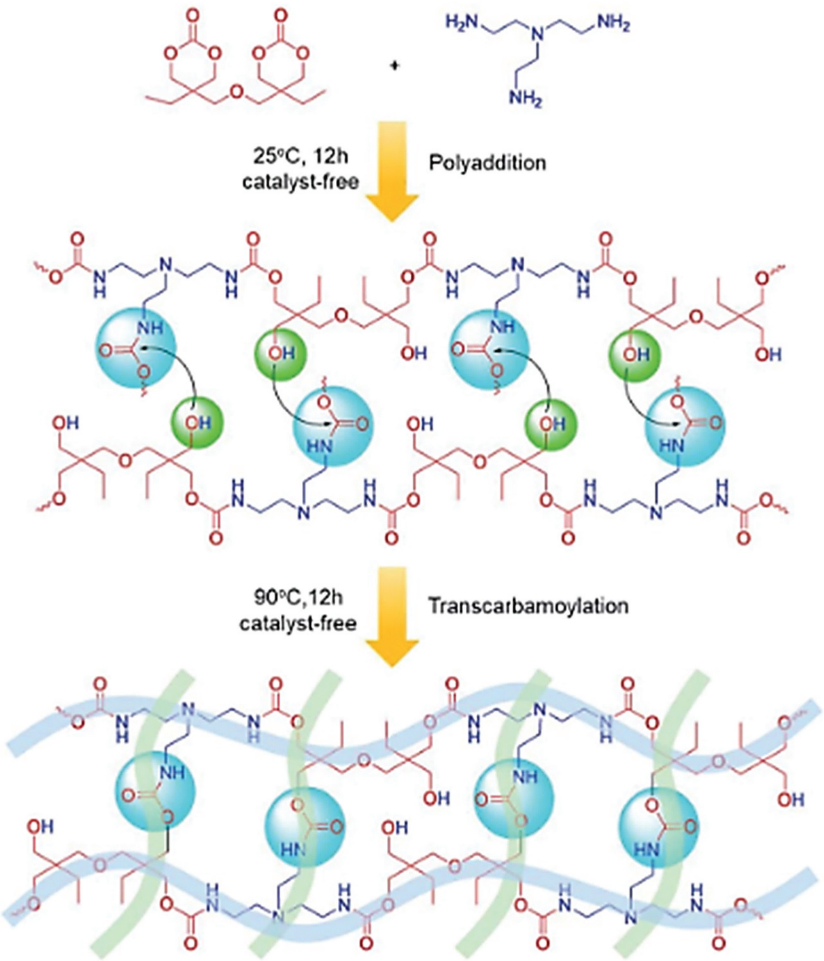

Figure 13. Synthesis of the PHU vitrimer from BCC (bis (6-membered cyclic carbonate)) and TREN (tris(2-aminoethyl) amine) with exchange reaction mechanism for healing and recycling (reproduced from reference [180] by the permission of royal society of chemistry).

environmentalists. To simultaneously solve the problem of electronic pollution control and turn them into valuable materials that preserve desired properties, opinions have been drawn from a new class of polymers that use covalent dynamic bonds in their chemical structure, which ultimately leads to the synthesis of polymer networks. These adaptable polymer networks are named vitrimers, which benefit from two very important transition temperatures, namely T_g and T_V. Thanks to the presence of these two transition temperatures, along with the dynamic nature of chemical bonds, the rearrangement of the topology is achieved by the application of an external stimulus. This is a prelude to the induction of special properties such as self-healing, shape memory, reprocessability, and recyclability.

Several dynamic band chemistries are of interest in the field of electronic synthesis materials research, which include disulphide, hindered urea, imine, transesterification, transcarbamylase dimethyl glyoxime urethane, and ionic chemistry-based covalent adaptable networks. This review article aimed to explore recent research on vitrimers for developing sustainable electronic materials and devices. After a full introduction of the applied chemistry in the synthesis of vitrimeric networks, the obtained properties were reported and their correlation with the chemical nature of the network was mentioned. Our goal in this article was to achieve electronic devices with shape memory, rehealability, and recyclability properties simultaneously to increase the lifespan and efficiency. The main results/opinions based on this review can be detailed below:

- The use of vitrimer chemistry has been one of the most effective ways to achieve the shape memory property. From our viewpoint, this can be the basis for the design of shape programmable electronic devices.
- In the investigated cases, the use of vitrimer chemistry has also resulted in achieving the reparability property. This property can further develop the future of self-healing electronics, which is critical in increasing performance efficiency and uptime.
- The use of vitrimer chemistry provides the potential to achieve simultaneous properties including rehealability, shape memory, reprocessability and recycling. It is believed that such synchronization will be one of the most important mechanisms in increasing the lifespan of devices and maintaining their quality and performance.
- All the synthesized samples are able to maintain their mechanical properties well after recycling. This result is proof of the usefulness for combination vitrimer networks with electronic applications.
- It is possible to advance designing and properties towards the use of other dynamic band chemistries such as bronic-ester, silyl-ether, acetal, carbonate, carbamide, thioether, and dixoborolane for a multifunctional design.

Regarding the industrialization of this strategy, it can be said that, it originates from the concepts of chemical and mechanical recycling and is a suitable method to maintain the final properties of the recycled product. Until today, no articles have been reported about the industrialization of vitrimerization on a real and large scale. It is probably due to challenges such as the cost of the equipment and the special pilot required to perform vitrimerization such as expensive reactive extrusion and so on.

Abbreviations

[EMI][TFSI]	(1-ethyl-3-methylimidazolium bis(trifluoromethyl-sulphonyl) imide
2D	Two-dimensional
3D	Three-dimensional
AA-PDMS	Diacrylate poly(dimethylsiloxane)
AgNW	Silver nanowires
AIE	Aggregation-induced-emission
CANs	Covalent adaptable networks
CCT	Carbonized cotton textile
CnFS	Carbon nanofibers
CP	Cellulose paper
CSF	Carbonized silk fabric
DCBs	Dynamic covalent bonds
DCS-PDMS	Double-crosslinked self- healing PDMS
DHPMA	2,3-dihydroxypropyl methacrylate
DMA	Dynamic mechanical analysis
DOU	Dimethylglyoxime-urethane
DSC	Differential scanning calorimetry
EMI	Electromagnetic shielding
EMIm TFSI	1-ethyl-3-methylimidazolium bis (trifluoromethylsulfonyl)imide
EV	Epoxy-derived vitrimer
GFRV	Fiber-reinforced vitrimer
GLY	Glycerin
GnPs	Graphene nanoplatelets
GWP	Global warming potential
HEMAH	Divinyl carbamoyl derivative
HTPB	Hydroxyl-terminated polybutadiene
iCAN	ionic covalent adaptable network
IL	Ionic liquid
IPDI	Isophorone diisocyanate
IVE	Imine-vitrimer elastomer (IVE)
LCA	Life-cycle assessment
MAVIN	MXene acrylate vitrimer network
MB	3-methoxy-4-(oxiran-2-ylmethoxy) benzaldehyde
MGEs	Moist-electric generators
MWCNTs	Multiwalled carbon nanotubes
NPDMS	Aminopropyl terminated PDMS
OPD	Original paper with pencil-drawing
PBDAB	Poly(1,4-butanediol) bis(4-aminobenzoate)
PCBs	Printed circuit boards
PCs	Photonic crystals
PDA	Polydopamine
PDMS	Polydimethylsiloxane
PDMS-DA	Polydimethylsiloxane-based diamine
PEG	Poly(ethylene glycol)
PEN	Polyethylene naphthalate
PET	Polyethylene terephthalate
pH	Potential of hydrogen or power of hydrogen
PHU	Polyhydroxyurethane

PN	Polyaniline
PTMG	(Poly(tetramethylene ether)glycol)
PVBEs	Photonic vitrimer-based electronics
SH-TENGs	Self-healable triboelectric nanogenerators
TEGs	Thermoelectric generators
TENG	Triboelectric nanogenerator
T_g	Glass transition temperature
T-NH	Four-arm star-hindered amine
TPAL	Terephthalaldehyde
T_v	Topology freezing transition temperature
Ultra Violet	UV
V	Vitrimer
VITENG	Vitrimer triboelectric nanogenerator
VMEGs	Vitrimer moist-electric generators
vPCBs	Vitrimer-based PCBs
VPDMS	Polydimethylsiloxane-based vitrimer
VTENG	Vitrimer based triboelectric nanogenerator
Zn-PABHM	Poly(acrylonitrile-*co*-butyl acrylate-*co*-hydroxyethyl methacrylate-*co*-zinc methacrylate)

Disclosure statement

No potential conflict of interest was reported by the author(s).

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Data availability statement

No data was used for the research described in the article.

CRediT author statement

Amirhossein Enayati-Gerdroodbar: Conceptualization, Validation, Writing – Original Draft. **Amirreza Khayati**: Writing – Original Draft. **Ali Zolfagharian**: Formal Analysis, Writing – Review & Editing. **Mehdi Salami-Kalajahi**: Conceptualization, Resources, Writing – Review & Editing, Supervision. **Mahdi Bodaghi**: Conceptualization, Validation, Resources, Formal Analysis, Visualization, Supervision, Project Administration, Writing – Review & Editing.

References

[1] Bouhamed A, Binyu Q, Böhm B et al. A hybrid piezoelectric composite flexible film based on PVDF-HFP for boosting power generation. Compos Sci Technol. 2021;208:108769. doi: 10.1016/j.compscitech.2021.108769

[2] Cao X, Xiong Y, Sun J et al. Piezoelectric nanogenerators derived self-powered sensors for multifunctional applications and artificial intelligence. Adv Funct Mater. 2021;31(33):2102983. doi: 10.1002/adfm.202102983

[3] Koo D, Jung S, Seo J, et al. Flexible organic solar cells over 15% efficiency with polyimide-integrated graphene electrodes. Joule. 2020;4(5):1021–1034. doi: 10.1016/j.joule.2020.02.012

[4] Xu J, Wang S, Nathan Wang G-J, et al. Highly stretchable polymer semiconductor films through the nanoconfinement effect. Science. 2017;355(6320):59–64. doi: 10.1126/science.aah4496

[5] Gao W, Emaminejad S, Yin Yin Nyein H, et al. Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis. Nature. 2016;529(7587):509–514. doi: 10.1038/nature16521

[6] Chen L-F, Huang Z-H, Liang H-W et al. Flexible all-solid-state high-power supercapacitor fabricated with nitrogen-doped carbon nanofiber electrode material derived from bacterial cellulose. Energyamp Environ Sci. 2013;6(11):3331–3338. doi: 10.1039/C3EE42366B

[7] Enayati-Gerdroodbar A, Eliseeva SN, Salami-Kalajahi M. A review on the effect of nanoparticles/matrix interactions on the battery performance of composite polymer electrolytes. J Energy Storage. 2023;68:107836. doi: 10.1016/j.est.2023.107836

[8] Zardehi-Tabriz A, Ghayebzadeh Y, Enayati Gerdroodbar A et al. Polyampholyte Polymers-Based sensors: a review on stimuli and applications. Macromol Mater Eng. 2023;308(12):2300179. doi: 10.1002/mame.202300179

[9] Mackanic DG, Chang T-H, Huang Z et al. Stretchable electrochemical energy storage devices. Chem Soc Rev. 2020;49(13):4466–4495. doi: 10.1039/D0CS00035C

[10] Yang Y, Zhang H, Liu Y et al. Silicon-based hybrid energy cell for self-powered electrodegradation and personal electronics. ACS Nano. 2013;7(3):2808–2813. doi: 10.1021/nn400361p

[11] Caffy F, Nicolaÿ R. Transformation of polyethylene into a vitrimer by nitroxide radical coupling of a bis-dioxaborolane. Polym Chem. 2019;10(23):3107–3115. doi: 10.1039/C9PY00253G

[12] Zhang H, Cui J, Hu G et al. Recycling strategies for vitrimers. Int J Smart Nano Mater. 2022;13(3):367–390. doi: 10.1080/19475411.2022.2087785

[13] Krishnakumar B, Prasanna Sanka RVS, Binder WH et al. Vitrimers: associative dynamic covalent adaptive networks in thermoset polymers. Chem Eng J. 2020;385:123820. doi: 10.1016/j.cej.2019.123820

[14] ASTM D7209-06. Standard guide for waste reduction, resource recovery, and Use of recycled polymeric materials and products (withdrawn 2015). West Conshohocken (PA): ASTM International; 2006. Available from: www.astm.org

[15] ISO 15270:2008. Plastics — guidelines for the recovery and recycling of plastics waste. Int Organ Standardization (ISO). 2008. Available from: https://www.iso.org

[16] Maris J, Bourdon S, Brossard J-M et al. Mechanical recycling: compatibilization of mixed thermoplastic wastes. Polym Degrad Stab. 2018;147:245–266. doi: 10.1016/j.polymdegradstab.2017.11.001

[17] de Camargo RV, Saron C. Mechanical–chemical recycling of low-density polyethylene waste with polypropylene. J Polym Environ. 2020;28(3):794–802. doi: 10.1007/s10924-019-01642-5

[18] Ignatyev IA, Thielemans W, Beke BV. Recycling of polymers: a review. ChemSusChem 2014. 2014;7(6):1579–1593. doi: 10.1002/cssc.201300898

[19] Zhao Y-B, Lv X-D, Ni H-G. Solvent-based separation and recycling of waste plastics: a review. Chemosphere. 2018;209:707–720. doi: 10.1016/j.chemosphere.2018.06.095

[20] Sanchez FAC, Boudaoud H, Hoppe S et al. Polymer recycling in an open-source additive manufacturing context: mechanical issues, add. Manufact. 2017;17:87–105. doi: 10.1016/j.addma.2017.05.013

[21] Byard DJ, Woern AL, Oakley RB et al. Green fab lab applications of large-area waste polymer-based additive manufacturing. Add Manufact. 2019;27:515–525. doi: 10.1016/j.addma.2019.03.006

[22] Sharifzadeh E, Salami-Kalajahi M, Hosseini MS et al. Synthesis of silica janus nanoparticles by buoyancy effect-induced desymmetrization process and their placement at the PS/PMMA interface. Colloid Polym Sci. 2017;295(1):25–36. doi: 10.1007/s00396-016-3977-5

[23] Lejeail M, Fischer HR. Investigations on the replacement of bismaleimide by the bio-based bisitaconimide for recyclable thermoset composites based on thermo-reversible Diels-Alder cross-links. Eur Polym J. 2020;131:109699. doi: 10.1016/j.eurpolymj.2020.109699

[24] Araujo LMG, Morales AR. Compatibilization of recycled polypropylene and recycled poly (ethylene terephthalate) blends with sebs-g-ma. Polímeros. 2018;28(1):84–91. doi: 10.1590/0104-1428.03016

[25] Gere D, Czigany T. Future trends of plastic bottle recycling: compatibilization of PET and PLA. Polym Test. 2020;81:106160. doi: 10.1016/j.polymertesting.2019.106160

[26] Li H, Sui X, Xie X-M. High-strength and super-tough PA6/PS/PP/SEBS quaternary blends compatibilized by using a highly effective multi-phase compatibilizer: toward efficient recycling of waste plastics. Polymer. 2017;231:240–246. doi: 10.1016/j.polymer.2017.07.024

[27] Moqadam S, Salami-Kalajahi M, Mahdavian M. Synthesis and characterization of sunflower oil-based polysulfide Polymer/Cloisite 30B Nanocomposites, Iran. J Chem Chem Eng. 2018;37:185–192.

[28] Han W, Zhang M, Kong Y et al. Pentaerythritol stearate ester-based tin (II) metal alkoxides: a tri-functional organotin as poly (vinyl chloride) thermal stabilizers. Polym Degrad Stab. 2020;175:109129. doi: 10.1016/j.polymdegradstab.2020.109129

[29] Tavares AA, Silva DFA, Lima PS et al. Chain extension of virgin and recycled polyethylene terephthalate. Polym Test. 2016;50:26–32. doi: 10.1016/j.polymertesting.2015.11.020

[30] Evens T, Bex G-J, Yigit M et al. The influence of mechanical recycling on properties in injection molding of fiber-reinforced polypropylene. Int Polym Proc. 2019;34(4):398–407. doi: 10.3139/217.3770

[31] Vollmer I, Jenks MJF, Roelands MCP et al. Beyond mechanical recycling: giving new life to plastic waste, Angew. Chem. 2020;59(36):15402–15423. doi: 10.1002/anie.201915651

[32] Liu SS, Hu JY, Qin D et al. Analysis of the volatile organic compounds (VOCs) during the regeneration of post-consumed poly (ethylene terephthalate) using HS-GC-MS method. Mater Sci Forum. 2019;944:1208–1214. doi: 10.4028/www.scientific.net/MSF.944.1208

[33] Cabanes A, Valdés FJ, Fullana A. A review on VOCs from recycled plastics. Sustain Mater Technol. 2020;25:e00179. doi: 10.1016/j.susmat.2020.e00179

[34] Cristancho DE, Guzman JD, Taylor C et al. Supercritical extraction of volatile organic components from polyethylene pellets. J Supercrit Fluids. 2012;69:124–130. doi: 10.1016/j.supflu.2012.05.004

[35] Amtai A, Elkoun S, Robert M et al. Mechanical recycling of thermoplastics: a review of key issues. Waste. 2023;1(4):860–883. doi: 10.3390/waste1040050

[36] Oya SI, Kanno D, Watanabe H et al. Catalytic production of branched small alkanes from biohydrocarbons. ChemSuschem. 2015;8(15):2472–2475. doi: 10.1002/cssc.201500375

[37] Miandad R, Rehan M, Nizami AS et al. Ismail, he energy and value-added products from pyrolysis of waste plastics. In: Karthikeyan O, K Heimann S Muthu, editors. Recycling of solid waste for biofuels and bio-chemicals. Environmental footprints and Eco-design of products and processes. Singapore: Springer; 2016. p 333–355. doi: 10.1007/978-981-10-0150-5_12

[38] Soni RK, Dutt K, Jain A et al. A novel route of synthesis, characterization of terephthalic dihydrazide from polyethylene terephthalate waste and its application in PVC compounding as plasticizer. J Appl Polym Sci. 2009;113(2):1090–1096. doi: 10.1002/app.29842

[39] Feghali E, Cantat T. Room temperature organocatalyzed reductive depolymerization of waste polyethers, polyesters, and polycarbonates. ChemSuschem. 2015;8(6):980–984. doi: 10.1002/cssc.201500054

[40] Cao X, Wang W, Lu K, et al. Magnesium-catalyzed hydroboration of organic carbonates, carbon dioxide and esters. Dalton Trans. 2020;49(9):2776–2780. doi: 10.1039/D0DT00465K

[41] Hodge P. Cyclodepolymerization as a method for the synthesis of macrocyclic oligomers, react. Funct Polym. 2014;80:21–32. doi: 10.1016/j.reactfunctpolym.2013.12.008

[42] Hodge P, Philip. Recycling of condensation polymers via ring–chain equilibria. Polym Adv Technol. 2015;26(7):797–803. doi: 10.1002/pat.3469

[43] Liu S, Wang Z, Li L et al. Butanol alcoholysis reaction of polyethylene terephthalate using acidic ionic liquid as catalyst. J Appl Polym Sci. 2013;130(3):1840–1844. doi: 10.1002/app.39246

[44] Purohit J, Chawada G, Choubisa B et al. Polyester polyol derived from waste poly (ethylene terephthalate) for coating application on mild steel. Chem Sci J. 2012;CSJ-76:1–7. Available from: http://astonjournals.com/csj.html

[45] Abdelaal MY, Sobahi TR, Makki MSI. Chemical degradation of poly(ethylene terephthalate). Int J Polym Mater. 2018;57(1):73–80. doi: 10.1080/00914030701329080

[46] Sebestyén Z, Rajnai EB, Bozi J et al. Thermo-catalytic pyrolysis of biomass and plastic mixtures using HZSM-5. Appl Energy. 2017;207:207 114–122. doi: 10.1016/j.apenergy.2017.06.032

[47] Mittal A, Soni RK, Dutt K et al. Scanning electron microscopic study of hazardous waste flakes of polyethylene terephthalate (PET) by aminolysis and ammonolysis. J Hazard Mater. 2010;178(1–3):390–396. doi: 10.1016/j.jhazmat.2010.01.092

[48] Abdelbasir SM, El-Sheltawy CT, Abdo DM. Green processes for electronic waste recycling: a review. J Sustain Metall. 2018;4:295–311. doi: 10.1007/s40831-018-0175-3

[49] Meng L, Gao J, Zhong Y et al. Supergravity separation for recovering Pb and Sn from electronic waste. Sep Purif Technol. 2018;191:375–383. doi: 10.1016/j.seppur.2017.09.041

[50] Wang K. Advances in E-Waste recycling: physical and chemical treatment methods. Highlights Sci Eng Technol. 2023;73:378–383. doi: 10.54097/hset.v73i.13106

[51] Wang Z, Li H, Chen H et al. Recovery of residual metals from− 0.5 mm non-metallic fractions in WPCBs using electrostatic separation enhanced by air classification. J Mater Cycles Waste Manag. 2022;24(3):980–991. doi: 10.1007/s10163-022-01373-1

[52] Flores-Campos R, Estrada-Ruiz RH, Velarde-Sánchez EJ. Study of the physico-chemical effects on the separation of the non-metallic fraction from printed circuit boards by inverse flotation. Waste Manag. 2017;69:400–406. doi: 10.1016/j.wasman.2017.08.049

[53] Verma HR, Singh KK, Raj Mankhand T. Dissolution and separation of brominated epoxy resin of waste printed circuit boards by using di-methyl formamide. J Cleaner Production. 2016;139:586–596. doi: 10.1016/j.jclepro.2016.08.084

[54] Li K, Xu Z. A review of current progress of supercritical fluid technologies for e-waste treatment. J Cleaner Production. 2019;227:794–809. doi: 10.1016/j.jclepro.2019.04.104

[55] Andooz A, Eqbalpour M, Kowsari E et al. A comprehensive review on pyrolysis of E-waste and its sustainability. J Cleaner Production. 2022;333:130191. doi: 10.1016/j.jclepro.2021.130191

[56] Gerdroodbar AE, Alihemmati H, Bodaghi M et al. Vitrimer chemistry for 4D printing formulation. Eur Polym J. 2023;197:112343. doi: 10.1016/j.eurpolymj.2023.112343

[57] Gerdroodbar AE, Karimkhani V, Dashtimoghadam E et al. Vitrimerization as a bridge of chemical and mechanical recycling. J Environ Chem Eng. 2024:112897. doi: 10.1016/j.jece.2024.112897

[58] Akbari-Meinagh M, Safavi-Mirmahalleh S-A, Roghani-Mamaqani H et al. Preparation of stimuli-responsive hydrogel vitrimers as recyclable dye adsorbent for water purification. J Environ Chem Eng. 2023;11(3):109799. doi: 10.1016/j.jece.2023.109799

[59] Beran R, Zarybnicka L, Machova D. Recycling of rigid polyurethane foam: Micro-milled powder used as active filler in polyurethane adhesives. J Appl Polym Sci. 2020;137(37):49095. doi: 10.1002/app.49095

[60] Li X, Yu R, He Y et al. Self-healing polyurethane elastomers based on a disulfide bond by digital light processing 3D printing. ACS Macro Lett. 2019;8(11):1511–1516. doi: 10.1021/acsmacrolett.9b00766

[61] Feng Z, Yu B, Hu J et al. Multifunctional vitrimer-like polydimethylsiloxane (PDMS): recyclable, self-healable, and water-driven malleable covalent networks based on dynamic imine bond. Ind Eng Chem Res. 2019;58(3):1212–1221. doi: 10.1021/acs.iecr.8b05309

[62] Debsharma T, Amfilochiou V, Alicja Wróblewska A et al. Fast dynamic siloxane exchange mechanism for reshapable vitrimer composites. J Am Chem Soc. 2022;144(27):12280–12289. doi: 10.1021/jacs.2c03518

[63] Zhang X, Wang S, Jiang Z et al. Boronic ester based vitrimers with enhanced stability via internal boron–nitrogen coordination. J Am Chem Soc. 2020;142(52):21852–21860. doi: 10.1021/jacs.0c10244

[64] Zheng J, Mao Png Z, Hoe Ng S et al. Vitrimers: current research trends and their emerging applications. Mater Today. 2021;51:586–625. doi: 10.1016/j.mattod.2021.07.003

[65] Tran HT, Sultana Nisha S, Radjef R et al. Recyclable and biobased vitrimers for carbon fibre-reinforced composites—A review. Polymers. 2024;16(8):1025. doi: 10.3390/polym16081025

[66] Hu Z, Hu F, Deng L, et al. Reprocessible triketoenamine-based vitrimers with closed-loop recyclability. Angew Chem. 2023;135(34):e202306039. doi: 10.1002/ange.202306039

[67] Sharifzadeh E, Salami-Kalajahi M, Salami Hosseini M et al. Synthesis of silica janus nanoparticles by buoyancy effect-induced desymmetrization process and their placement at the PS/PMMA interface. Colloid Polym Sci. 2017;295:25–36. doi: 10.1007/s00396-016-3977-5

[68] Yang Z, Wang Q, Wang T. Dual-triggered and thermally reconfigurable shape memory graphene-vitrimer composites. ACS Appl Mater Interface. 2016;8(33):21691–21699. doi: 10.1021/acsami.6b07403

[69] Lagron AB, Zaatari BME, Hamachi LS. Characterization techniques to assess recyclability in dynamic polymer networks. Front Mater. 2022;9:915296. doi: 10.3389/fmats.2022.915296

[70] Kim H, Cha I, Yoon Y et al. Facile mechanochemical synthesis of malleable biomass-derived network polyurethanes and their shape-memory applications. ACS Sustain Chem Eng. 2021;9(20):6952–6961. doi: 10.1021/acssuschemeng.1c00390

[71] Li Q, Ma S, Li P et al. Biosourced acetal and Diels–Alder adduct concurrent polyurethane covalent adaptable network. Macromolecules. 2021;54(4):1742–1753. doi: 10.1021/acs.macromol.0c02699

[72] Li Q, Ma S, Li P et al. Fast reprocessing of acetal covalent adaptable networks with high performance enabled by neighboring group participation. Macromolecules. 2021;54(18):8423–8434. doi: 10.1021/acs.macromol.1c01046

[73] Hernández A, Houck HA, Elizalde F et al. Internal catalysis on the opposite side of the fence in non-isocyanate polyurethane covalent adaptable networks. Eur Polym J. 2022;168:111100. doi: 10.1016/j.eurpolymj.2022.111100

[74] Samanta S, Kim S, Saito T et al. Polymers with dynamic bonds: adaptive functional materials for a sustainable future. J Phys Chem B. 2021;125(33):9389–9401. doi: 10.1021/acs.jpcb.1c03511

[75] Melchor B, Alberto J, Larsen MB. Thermal guanidine metathesis for covalent adaptable networks. ACS Macro Lett. 2020;9(7):937–943. doi: 10.1021/acsmacrolett.0c00352

[76] Li L, Chen X, Torkelson JM. Reprocessable polymer networks via thiourethane dynamic chemistry: recovery of cross-link density after recycling and proof-of-principle solvolysis leading to monomer recovery. Macromolecules. 2019;52(21):8207–8216. doi: 10.1021/acs.macromol.9b01359

[77] Alraddadi MA, Chiaradia V, Stubbs CJ et al. Renewable and recyclable covalent adaptable networks based on bio-derived lipoic acid. Polym Chem. 2021;12(40):5796–5802. doi: 10.1039/D1PY00754H

[78] Schoustra SK, Groeneveld T, Smulders MMJ. The effect of polarity on the molecular exchange dynamics in imine-based covalent adaptable networks. Polym Chem. 2021;12(11):1635–1642. doi: 10.1039/D0PY01555E

[79] Delahaye M, Winne JM, Du Prez FE. Internal catalysis in covalent adaptable networks: phthalate monoester transesterification as a versatile dynamic cross-linking chemistry. J Am Chem Soc. 2019;141(38):15277–15287. doi: 10.1021/jacs.9b07269

[80] Barsoum DN, Kirinda VC, Kang B et al. Remote-controlled exchange rates by photoswitchable internal catalysis of dynamic covalent bonds. J Am Chem Soc. 2022;144(23):10168–10173. doi: 10.1021/jacs.2c04658

[81] Gamardella F, Muñoz S, Flor SDL et al. Recyclable organocatalyzed poly (thiourethane) covalent adaptable networks. Polymers. 2020;12(12):2913. doi: 10.3390/polym12122913

[82] He X, Shi X, Chung C et al. A sustainable manufacturing method of thermoset composites based on covalent adaptable network polymers, composites part B. Engineering. 2021;221:109004. doi: 10.1016/j.compositesb.2021.109004

[83] Taplan C, Guerre M, Du Prez FE. Covalent adaptable networks using β-amino esters as thermally reversible building blocks. J Am Chem Soc. 2021;143 (24):9140–9150. doi: 10.1021/jacs.1c03316

[84] Scheutz GM, Lessard JJ, Sims MB et al. Adaptable crosslinks in polymeric materials: resolving the intersection of thermoplastics and thermosets. J Am Chem Soc. 2019;141(41):16181–16196. doi: 10.1021/jacs.9b07922

[85] Shi X, Soule D, Mao Y et al. A multiscale chemomechanics theory for the solvent–assisted recycling of covalent adaptable network polymers. J Mech Phys Solids. 2020;138:103918. doi: 10.1016/j.jmps.2020.103918

[86] Bongiardina NJ, Long KF, Podgórski M et al. Substituted thiols in dynamic thiol–thioester reactions. Macromolecules. 2021;54(18):8341–8351. doi: 10.1021/acs.macromol.1c00649

[87] Lagron AB, Bassil ME-Z, Leslie SH. Characterization techniques to assess recyclability in dynamic polymer networks. Front Mater. 2022;9:915296. doi: 10.3389/fmats.2022.915296

[88] Meyvis TK, Stubbe BG, Van Steenbergen MJ et al. A comparison between the use of dynamic mechanical analysis and oscillatory shear rheometry for the characterisation of hydrogels. Int J Pharm. 2002;244(1–2):163–168. doi: 10.1016/S0378-5173(02)00328-9

[89] Chakma P, Morley CN, Sparks JL et al. Exploring how vitrimer-like properties can be achieved from dissociative exchange in anilinium salts. Macromolecules. 2020;53(4):1233–1244. doi: 10.1021/acs.macromol.0c00120

[90] Parada GA, Zhao X. Ideal reversible polymer networks. Soft Matter. 2018;14 (25):5186–5196. doi: 10.1039/C8SM00646F

[91] Lingqiao L, Chen X, Jin K et al. Vitrimers designed both to strongly suppress creep and to recover original cross-link density after reprocessing: quantitative theory and experiments. Macromolecules. 2018;51(15):5537–5546. doi: 10.1021/acs.macromol.8b00922.4

[92] Kaiser S, Novak P, Giebler M et al. The crucial role of external force in the estimation of the topology freezing transition temperature of vitrimers by elongational creep measurements. Polymer. 2020;204:122804. doi: 10.1016/j.polymer.2020.122804

[93] Hubbard AM, Ren Y, Konkolewicz D et al. Vitrimer transition temperature identification: coupling various thermomechanical methodologies. ACS Appl Polym Mater. 2021;3(4):1756–1766. doi: 10.1021/acsapm.0c01290

[94] Zeng Y, Yang W, Liu S et al. Dynamic semi IPNs with duple dynamic linkers: self-healing, reprocessing, welding, and shape memory behaviors. Polymers. 2021;13(11):1679. doi: 10.3390/polym13111679

[95] Xu H, Hanzon DW, Yu K. Cyclic welding behavior of covalent adaptable network polymers. J Polym Sci Part B: Polym Phys. 2018;56(5):402–413. doi: 10.1002/polb.2455

[96] Swartz JL, Sheppard DT, Haugstad G et al. Blending polyurethane thermosets using dynamic urethane exchange. Macromolecules. 2021;54(23):11126–11133. doi: 10.1021/acs.macromol.1c01910

[97] Danielsen SPO, Beech HK, Wang S, et al. Molecular characterization of polymer networks. Chem Rev. 2021;121(8):5042–5092. doi: 10.1021/acs.chemrev.0c01304

[98] Lijsebetten FV, Spiesschaert Y, Winne JM et al. Reprocessing of covalent adaptable polyamide networks through internal catalysis and ring-size effects. J Am Chem Soc. 2021;143(38):15834–15844. doi: 10.1021/jacs.1c07360

[99] Yang Y, Zhu G, Zhang H et al. Triboelectric nanogenerator for harvesting wind energy and as self-powered wind vector sensor system. ACS Nano. 2013;7(10):9461–9468. doi: 10.1021/nn4043157

[100] Wang ZL, Jiang T, Xu L. Toward the blue energy dream by triboelectric nanogenerator networks. Nano Energy. 2017;39:9–23. doi: 10.1016/j.nanoen.2017.06.035

[101] Chen J, Zhu G, Yang W et al. Harmonic-resonator-based triboelectric nanogenerator as a sustainable power source and a self-powered active vibration sensor. Adv Mater. 2013;25(42):6094–6099. doi: 10.1002/adma.201302397

[102] Wang J, Li S, Yi F et al. Sustainably powering wearable electronics solely by biomechanical energy. Nat Commun. 2016;7(1):12744. doi: 10.1038/ncomms12744

[103] Niu S, Wang S, Liu Y et al. A theoretical study of grating structured triboelectric nanogenerators. Energyamp Environ Sci. 2014;7(7):2339–2349. doi: 10.1039/C4EE00498A

[104] Deng J, Kuang X, Liu R, et al. Vitrimer elastomer-based jigsaw puzzle-like healable triboelectric nanogenerator for self-powered wearable electronics. Adv Mater. 2018;30(14):1705918. doi: 10.1002/adma.201705918

[105] Ying H, Zhang Y, Cheng J. Dynamic urea bond for the design of reversible and self-healing polymers. Nat Commun. 2014;5(1):3218. doi: 10.1038/ncomms4218

[106] Ying H, Cheng J. Hydrolyzable polyureas bearing hindered urea bonds. J Am Chem Soc. 2014;136(49):16974–16977. doi: 10.1021/ja5093437

[107] Wang Y, Pan Y, Zheng Z et al. Reconfigurable and reprocessable thermoset shape memory polymer with synergetic triple dynamic covalent bonds. Macromol Rapid Commun. 2018;39(10):1800128. doi: 10.1002/marc.201800128

[108] Zhang Y, Ying H, Hart KR, et al. Malleable and recyclable poly (urea-urethane) thermosets bearing hindered urea bonds. Adv Mater. 2016;28(35):7646–7651. doi: 10.1002/adma.201601242

[109] Patel T, Kim MP, Park J et al. Self-healable reprocessable triboelectric nanogenerators fabricated with vitrimeric poly (hindered urea) networks. ACS Nano. 2020;14(9):11442–11451. doi: 10.1021/acsnano.0c03819

[110] Sun J, Pu X, Liu M et al. Self-healable, stretchable, transparent triboelectric nanogenerators as soft power sources. ACS Nano. 2018;12(6):6147–6155. doi: 10.1021/acsnano.8b02479

[111] Zhang W, Wang J, Zhang W et al. Scalable micropatterned epoxy vitrimer films by thermo-triggered bond exchange for repairable and recyclable triboelectric nanogenerators. Appl Mater Today. 2023;32:101789. doi: 10.1016/j.apmt.2023.101789

[112] Feng H, Wang N, Li K, et al. Imine-vitrimer elastomer enabled triboelectric nanogenerators with remarkable electrical output and circular life cycles. Chem Eng J. 2024;480:148214. doi: 10.1016/j.cej.2023.148214

[113] Ye C, Li W, Voet VS et al. Flexible vitrimers for Self-healable triboelectric nanogenerators. Adv Mater Technol. 2023;8(8):2201670. doi: 10.1002/admt.202201670

[114] Niu X, Wang M, Xia Y et al. Self-healing, thermadapt triple-shape memory ionomer vitrimer for shape memory triboelectric nanogenerator. ACS Appl Mater Interface. 2022;14(44):50101–50111. doi: 10.1021/acsami.2c13294

[115] Sun L, Huang H, Guan Q et al. Cooperative chemical coupling and physical lubrication effects construct highly dynamic ionic covalent adaptable network for high-performance wearable electronics. CCS Chem. 2023;5(5):1096–1107. doi: 10.31635/ccschem.022.202202037

[116] Lai Y, Wu H, Lin H et al. Entirely, intrinsically, and autonomously self-healable, highly transparent, and superstretchable triboelectric nanogenerator for personal power sources and self-powered electronic skins. Adv Funct Mater. 2019;29(40):1904626. doi: 10.1002/adfm.201904626

[117] Cai Y-W, Zhang X-N, Wang G-G et al. A flexible ultra-sensitive triboelectric tactile sensor of wrinkled PDMS/MXene composite films for E-skin. Nano Energy. 2021;81:105663. doi: 10.1016/j.nanoen.2020.105663

[118] Ganesh RS, Yoon H, Kim S. Recent trends of biocompatible triboelectric nanogenerators toward self-powered e-skin. EcoMat. 2020;2(4):e12065. doi: 10.1002/eom2.12065

[119] Cai Y-W, Wang G-G, Mei Y-C et al. Self-healable, super-stretchable and shape-adaptive triboelectric nanogenerator based on double cross-linked PDMS for electronic skins. Nano Energy. 2022;102:107683. doi: 10.1016/j.nanoen.2022.107683

[120] Chen J, Yu Q, Cui X et al. An overview of stretchable strain sensors from conductive polymer nanocomposites. J Mater Chem C. 2019;7(38):11710–11730. doi: 10.1039/C9TC03655E

[121] Pang H, Xu L, Yan D-X et al. Conductive polymer composites with segregated structures. Prog Polym Sci. 2014;39(11):1908–1933. doi: 10.1016/j.progpolymsci.2014.07.007

[122] Roh E, Hwang B-U, Kim D et al. Stretchable, transparent, ultrasensitive, and patchable strain sensor for human–machine interfaces comprising a nanohybrid of carbon nanotubes and conductive elastomers. ACS Nano. 2015;9(6):6252–6261. doi: 10.1021/acsnano.5b01613

[123] Shang L, Zhang W, Xu K et al. Bio-inspired intelligent structural color materials. Mater Horiz. 2019;6(5):945–958. doi: 10.1039/C9MH00101H

[124] Wu P, Wang J, Jiang L. Bio-inspired photonic crystal patterns. Mater Horiz. 2020;7(2):338–365. doi: 10.1039/C9MH01389J

[125] Lee GH, Min Choi T, Kim B et al. Chameleon-inspired mechanochromic photonic films composed of non-close-packed colloidal arrays. ACS Nano. 2017;11(11):11350–11357. doi: 10.1021/acsnano.7b05885

[126] Zhao K, Cao X, Alsaid Y et al. Interactively mechanochromic electronic textile sensor with rapid and durable electrical/optical response for visualized stretchable electronics. Chem Eng J. 2021;426:130870. doi: 10.1016/j.cej.2021.130870

[127] Zhao K, Wang Y, Guo J et al. Photonic vitrimer-based electronics with self-healing and ultrastable visual-digital outputs for wireless strain sensing. Chem Eng J. 2022;450:138285. doi: 10.1016/j.cej.2022.138285

[128] Shi C, Zou Z, Lei Z et al. Heterogeneous integration of rigid, soft, and liquid materials for self-healable, recyclable, and reconfigurable wearable electronics. Sci Adv. 2020;6(45):eabd0202. doi: 10.1126/sciadv.abd0202

[129] Zhang D, Cheng Z, Gao S et al. Rational structural design of vanillin based vitrimers in one-pot to facilely fabricate highly stretchable and sensitive composite strain sensor. Mater Chem Phys. 2022;279:125730. doi: 10.1016/j.matchemphys.2022.125730

[130] Niu W, Cao X, Wang Y et al. Photonic vitrimer elastomer with self-healing, high toughness, mechanochromism, and excellent durability based on dynamic covalent bond. Adv Funct Mater. 2021;31(13):2009017. doi: 10.1002/adfm.202009017

[131] Ye C, Yan F, Lan X et al. Novel MXene sensors based on fast healing vitrimers. Appl Mater Today. 2022;29:101683. doi: 10.1016/j.apmt.2022.101683

[132] Guerre M, Taplan C, Winne JM et al. Vitrimers: directing chemical reactivity to control material properties. Chem Sci. 2020;11(19):4855–4870. doi: 10.1039/d0sc01069c

[133] Zhang CJ, Pinilla S, McEvoy N, et al. Oxidation stability of colloidal two-dimensional titanium carbides (MXenes. Chem Mater. 2017;29(11):4848–4856. doi: 10.1021/acs.chemmater.7b00745

[134] Shi K, Huang X, Sun B et al. Cellulose/BaTiO3 aerogel paper based flexible piezoelectric nanogenerators and the electric coupling with triboelectricity. Nano Energy. 2019;57:450–458. doi: 10.1016/j.nanoen.2018.12.076

[135] Kim J-H, Lee Y-H, Cho S-J et al. Nanomat Li–S batteries based on all-fibrous cathode/separator assemblies and reinforced Li metal anodes: towards ultra-high energy density and flexibility. Energyamp Environ Sci. 2019;12(1):177–186. doi: 10.1039/C8EE01879K

[136] Xiong C, Li M, Zhao W et al. A smart paper@ polyaniline nanofibers incorporated vitrimer bifunctional device with reshaping, shape-memory and self-healing properties applied in high-performance supercapacitors and sensors. Chem Eng J. 2020;396:125318. doi: 10.1016/j.cej.2020.125318

[137] Xiong C, Wang T, Zhang Y et al. Recent progress on green electromagnetic shielding materials based on macro wood and micro cellulose components from natural agricultural and forestry resources. Nano Res. 2022;15(8):7506–7532. doi: 10.1007/s12274-022-4512-2

[138] Zhang H-Y, Li J-Y, Pan Y et al. Flexible carbon fiber-based composites for electromagnetic interference shielding. Rare Met. 2022;41(11):3612–3629. doi: 10.1007/s12598-022-02057-3

[139] Xiong C, Wang T, Zhang Y et al. Multifunctional conductive material based on intelligent porous paper used in conjunction with a vitrimer for electromagnetic shielding, sensing, joule heating, and antibacterial properties. ACS Appl Mater Interface. 2023;15(28):33763–33773. doi: 10.1021/acsami.3c06926

[140] Irimia-Vladu M. "Green" electronics: biodegradable and biocompatible materials and devices for sustainable future. Chem Soc Rev. 2014;43(2):588–610. doi: 10.1039/C3CS60235D

[141] Feig VR, Tran H, Bao Z. Biodegradable polymeric materials in degradable electronic devices. ACS Cent Sci. 2018;4(3):337–348. doi: 10.1021/acscentsci.7b00595

[142] Li W, Liu Q, Zhang Y et al. Biodegradable materials and green processing for green electronics. Adv Mater. 2020;32(33):2001591. doi: 10.1002/adma.202001591

[143] Cantarella G, Madagalam M, Merino I, et al. Laser-Induced, Green and biocompatible Paper-Based devices for circular electronics. Adv Funct Mater. 2023;33(17):2210422. doi: 10.1002/adfm.202210422

[144] Wu X, Steiner P, Raine T et al. Hybrid graphene/carbon nanofiber wax emulsion for paper-based electronics and thermal management. Adv Electron Mater. 2020;6(7):2000232. doi: 10.1002/aelm.202000232

[145] Spallanzani G, Najafi M, Zahid M et al. Self-Healing, recyclable, biodegradable, electrically conductive vitrimer coating for soft robotics. Adv Sustain Syst. 2023;7(12):2300220. doi: 10.1002/adsu.202300220

[146] Gelebart AH, Jan Mulder D, Varga M et al. Making waves in a photoactive polymer film. Nature. 2017;546(7660):632–636. doi: 10.1038/nature22987

[147] Yang Y, Pei Z, Li Z et al. Making and remaking dynamic 3D structures by shining light on flat liquid crystalline vitrimer films without a mold. J Am Chem Soc. 2016;138(7):2118–2121. doi: 10.1021/jacs.5b12531

[148] Pei Z, Yang Y, Chen Q et al. Mouldable liquid-crystalline elastomer actuators with exchangeable covalent bonds. Nat Mater. 2014;13(1):36–41. doi: 10.1038/nmat3812

[149] Yang Y, Terentjev EM, Zhang Y et al. Reprocessable thermoset soft actuators. Angew Chem Int Ed. 2019;58(48):17474–17479. doi: 10.1002/anie.201911612

[150] Wu Y, Yang Y, Qian X et al. Liquid-crystalline soft actuators with switchable thermal reprogrammability. Angew Chem. 2020;132(12):4808–4814. doi: 10.1002/anie.201915694

[151] Wang H, Yang Y, Zhang M et al. Electricity-triggered self-healing of conductive and thermostable vitrimer enabled by paving aligned carbon nanotubes. ACS Appl Mater Interface. 2020;12(12):14315–14322. doi: 10.1021/acsami.9b21949

[152] Taccola S, Greco F, Sinibaldi E et al. Toward a new generation of electrically controllable hygromorphic soft actuators. Adv Mater. 2015;27(10):1668. doi: 10.1002/adma.201404772

[153] Piotrowska R, Hesketh T, Wang H, et al. Mechanistic insights of evaporation-induced actuation in supramolecular crystals. Nat Mater. 2021;20(3):403–409. doi: 10.1038/s41563-020-0799-0

[154] Arazoe H, Miyajima D, Akaike K et al. An autonomous actuator driven by fluctuations in ambient humidity. Nat Mater. 2016;15(10):1084–1089. doi: 10.1038/nmat4693

[155] Song H, Lee H, Lee J et al. Reprogrammable ferromagnetic domains for reconfigurable soft magnetic actuators. Nano Lett. 2020;20(7):5185–5192. doi: 10.1021/acs.nanolett.0c01418

[156] Zhao Q, Dunlop JW, Qiu X et al. An instant multi-responsive porous polymer actuator driven by solvent molecule sorption. Nat Commun. 2014;5(1):4293. doi: 10.1038/ncomms5293

[157] Cianchetti M, Laschi C, Menciassi A et al. Biomedical applications of soft robotics. Nat Rev Mater. 2018;3(6):143–153. doi: 10.1038/s41578-018-0022-y

[158] Rus D, Tolley MT. Design, fabrication and control of soft robots. Nature. 2015;521(7553):467–475. doi: 10.1038/nature14543

[159] Sun L, Yu Y, Chen Z et al. Biohybrid robotics with living cell actuation. Chem Soc Rev. 2020;49(12):4043–4069. doi: 10.1039/D0CS00120A

[160] Yang Y, Wang H, Zhang S et al. Vitrimer-based soft actuators with multiple responsiveness and self-healing ability triggered by multiple stimuli. Matter. 2021;4(10):3354–3365. doi: 10.1016/j.matt.2021.08.009

[161] Kim D-H, Lu N, Ma R, et al. Epidermal electronics. Science. 2011;333(6044):838–843. doi: 10.1126/science.1206157

[162] Kaltenbrunner M, Sekitani T, Reeder J, et al. An ultra-lightweight design for imperceptible plastic electronics. Nature. 2013;499(7459):458–463. doi: 10.1038/nature12314

[163] LeMieux MC, Bao Z. Stretching our imagination. Nat Nanotechnol. 2008;3(10):585–586. doi: 10.1038/nnano.2008.296

[164] Hammock ML, Chortos A, Tee BC et al. 25th anniversary article: the evolution of electronic skin (e-skin): a brief history, design considerations, and recent progress. Adv Mater. 2013;25(42):5997–6038. doi: 10.1002/adma.201302240

[165] Xu S, Zhang Y, Jia L, et al. Soft microfluidic assemblies of sensors, circuits, and radios for the skin. Science. 2014;344(6179):70–74. doi: 10.1126/science.1250169

[166] Kloxin CJ, Scott TF, Adzima BJ et al. Covalent adaptable networks (CANs): a unique paradigm in cross-linked polymers. Macromolecules. 2010;43(6):2643–2653. doi: 10.1021/ma902596s

[167] Kloxin CJ, Bowman CN. Covalent adaptable networks: smart, reconfigurable and responsive network systems. Chem Soc Rev. 2013;42(17):7161–7173. doi: 10.1039/C3CS60046G

[168] Taynton P, Yu K, Shoemaker RK et al. Heat-or water-driven malleability in a highly recyclable covalent network polymer. Adv Mater. 2014;26(23):3938–3942. doi: 10.1002/adma.201400317

[169] Whiteley JM, Taynton P, Zhang W et al. Ultra-thin solid-state Li-ion electrolyte membrane facilitated by a self-healing polymer matrix. Adv Mater. 2015;27(43):6922–6927. doi: 10.1002/adma.201502636

[170] Zhang J, Lei Z, Luo S et al. Malleable and recyclable conductive mwcnt-vitrimer composite for flexible electronics. ACS Appl Nano Mater. 2020;3(5):4845–4850. doi: 10.1021/acsanm.0c00902

[171] Ogunseitan OA, Schoenung JM, Lincoln J et al. Biobased materials for sustainable printed circuit boards. Nat Rev Mater. 2022;7(10):749–750. doi: 10.1038/s41578-022-00485-2

[172] Zhang Z, Biswal AK, Nandi A et al. Recyclable vitrimer-based printed circuit boards for sustainable electronics. Nat Sustainability. 2024:1–12. doi: 10.1038/s41893-024-01333-7

[173] Sultana S, Ahmed K, Krisma Jiwanti P et al. Ionic liquid-based gels for applications in electrochemical energy storage and conversion devices: a review of recent progress and future prospects. Gels. 2021;8(1):2. doi: 10.3390/gels8010002

[174] Fan X, Liu S, Zhenhua Jia JJK, et al. Ionogels: recent advances in design, material properties and emerging biomedical applications. Chem Soc Rev. 2023;52(7):2497–2527. doi: 10.1039/D2CS00652A

[175] Bui K, Nguyen GT, Vancaeyzeele C et al. Stretchable, healable, and weldable vitrimer ionogel for ionotronic applications. Chem Eng J. 2023;474:145533. doi: 10.1016/j.cej.2023.145533

[176] Xue G, Xu Y, Ding T, et al. Water-evaporation-induced electricity with nanostructured carbon materials. Nat Nanotechnol. 2017;12(4):317–321. doi: 10.1038/nnano.2016.300

[177] van der Heyden, Frank HJ, Jan Bonthuis D et al. Power generation by pressure-driven transport of ions in nanofluidic channels. Nano Lett. 2007;7(4):1022–1025. doi: 10.1021/nl070194h

[178] Shen D, Duley WW, Peng P et al. Moisture-enabled electricity generation: from physics and materials to self-powered applications. Adv Mater. 2020;32(52):2003722. doi: 10.1002/adma.202003722

[179] Stephens GL, Li J, Wild M et al. An update on Earth's energy balance in light of the latest global observations. Nat Geosci. 2012;5(10):691–696. doi: 10.1038/ngeo1580

[180] Feng Z, Zhao W, Yang Z et al. Energy harvesting by vitrimer-based moist-electric generators. J Mater Chem A. 2022;10(21):11524–11534. doi: 10.1039/D1TA11029B

[181] Zhu B, Wang H, Ru Leow W et al. Silk fibroin for flexible electronic devices. Adv Mater. 2016;28(22):4250–4265. doi: 10.1002/adma.201504276

[182] Liu Y, He K, Chen G et al. Nature-inspired structural materials for flexible electronic devices. Chem Rev. 2017;117(20):12893–12941. doi: 10.1021/acs.chemrev.7b00291

[183] Zheng J, Faye Duran Solco S, Jie Ee Wong C, et al. Integrating recyclable polymers into thermoelectric devices for green electronics. J Mater Chem A. 2022;10(37):19787–19796. doi: 10.1039/D2TA00386D