Multiscale Pathways to Super tough Polymers: From Bonds to Nanocomposites

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Abstract

Super toughness is defined as a polymer's ability to absorb exceptionally high energy and resist fracture while maintaining structural integrity under extreme stress. Super tough polymers are critical for applications requiring high impact resistance and reliability. Such performance originates from multiscale mechanisms spanning molecular, microscale, and nanoscale levels. At the molecular scale, coordination bonds, hydrogen bonding, and π–π interactions enhance bond strength, network stability, and reversible energy dissipation. Microscale structures, including elastomer domains, phase-separated interfaces, crazing, and core–shell architectures, facilitate controlled crack propagation and energy absorption. Nanoscale reinforcements, such as silicon dioxide (SiO_2) nanofibers, reduced graphene oxide (rGO), and liquid metal (LM) nanoparticles, further improve load transfer, crystallinity, and mechanical robustness. Representative systems, including polyamide 6.6/polyamide 6/maleic anhydride-grafted polyethylene-octene copolymer (PA66/PA6/POE-g-MAH) alloys and freeze-dried and annealed flexible SiO_2 nanofiber/polyvinyl alcohol (FDA-SNF/PVA) hydrogels, show that optimized composition and annealing conditions markedly enhance mechanical performance. Hydrogels with 20 wt% polyvinyl alcohol (PVA) annealed at 120 °C for 90 min (FDA20-120-90) exhibit tensile strength, toughness, and elastic modulus up to 101 times higher than reference systems and support loads over 4600 times their own weight. These findings demonstrate that super toughness is a designable property achievable through hierarchical polymer engineering.

Keywords: Polymer Toughening Mechanisms, Multiscale Mechanical Reinforcement, Dynamic Cross-linked Networks, Nanocomposite, Copolymer Design

1. Introduction

Toughness is a critical property in the design of advanced polymer materials, particularly for applications requiring high structural integrity and resistance to impact or vibration [1,2]. Polymers with high toughness are capable of absorbing significant amounts of energy before fracture, making them suitable for demanding applications such as automotive components, electronics, and structural materials [3,4]. Ensuring resistance to catastrophic failure under external loads is essential for the reliability and safety of polymer-based systems [5].

Toughness is defined as a material's ability to absorb energy and undergo plastic deformation prior to fracture and is commonly represented by the area under the stress–strain curve [1]. In some advanced polymer systems, the concept of super toughness is used to describe exceptionally high impact resistance that far exceeds conventional tough polymers [3]. Although no universal quantitative threshold exists, super toughness generally refers to polymer systems whose toughness or fracture energy is one to two orders of magnitude higher than that of conventional polymers under comparable testing conditions. Such super tough behavior is typically achieved through multi-level energy dissipation mechanisms, including elastomer deformation, interfacial debonding, and crack propagation through multiple crazes [3].

Various strategies have been developed to enhance polymer toughness, including elastomer blending, rigid particle incorporation, and grain refinement [3]. While elastomer blending effectively improves impact resistance, it often leads to reduced tensile strength and modulus [3]. The addition of rigid particles can hinder crack propagation while maintaining strength; however, uniform dispersion and sufficient matrix ductility are difficult to achieve, particularly under low-temperature conditions [3]. Grain refinement at the nanoscale can enhance toughness without compromising stiffness, but this approach typically requires complex and costly processing techniques [3].

Despite these advances, achieving a balance between high toughness, mechanical strength, thermodynamic stability, and dynamic responsiveness within a single polymer system remains a major challenge [5]. Many existing toughening methods involve unavoidable trade-offs or complex processing routes. Consequently, there is a strong need for new material design strategies that enable super tough behavior while preserving tensile strength and modulus, particularly in emerging functional systems such as photohealable or self-healing polymers [5].

"It should be noted that super toughness is a mechanical property, not an intrinsic characteristic of a specific polymer; many polymers can achieve this behavior through appropriate material design strategies, including elastomer blending, rigid particle incorporation, dynamic networks, and multilevel energy dissipation [3,5,8]."

2. Fundamentals and Mechanisms of Toughness in Polymer Systems

Toughness in polymer systems is a critical mechanical property that determines a material's ability to resist fracture under external loading. It reflects the combined capability of polymers to withstand deformation, absorb mechanical energy, and dissipate it through multiple, often synergistic, mechanisms. These responses are strongly governed by polymer microstructure, molecular interactions, and network architecture.

2.1 Mechanical Response of Polymers Under External Loading

Polymers exhibit a wide range of mechanical responses under tensile or impact loading, which directly influence their toughness and applicability in structural materials. Stress–strain behavior provides fundamental insight into these responses. For example, high-performance aramid fibers such as Kevlar® exhibit a high elastic modulus but relatively brittle behavior, characterized by sudden failure at low strains of approximately 2%–3% [6]. In contrast, super tough polymer systems can simultaneously achieve high tensile strength and extremely large elongation at break, with reported stretchability exceeding 1900% [6].

Polymer fracture is typically governed by complex and distributed damage mechanisms rather than localized failure. In aramid fibers, damage initiates with transverse crack formation at strains above 2%, followed by crack propagation in Mode I (opening mode), revealing underlying microfibrillar structures and large crack opening displacements of up to 3 µm [6,7]. At later stages, finer longitudinal cracks develop and propagate in Mode II (shear mode), interconnecting transverse cracks into complex fracture networks [7]. This widespread damage significantly enhances fracture resistance and overall toughness [6,7].

The hierarchical microstructure of polymers plays a crucial role in these processes. Features such as pleated-sheet structures, microfibrils, and crystalline domains strongly influence stress redistribution, crystallite reorientation, and strain-hardening behavior during deformation [3,6]. In twisted fiber systems, crack propagation often follows the twist angle, further highlighting the importance of fibrillar architecture in polymer toughening [6].

2.2 Energy Absorption and Dissipation Mechanisms in Polymers

Efficient energy absorption and dissipation are fundamental to achieving high toughness in polymer materials. These mechanisms vary depending on polymer chemistry, morphology, and network design.

Non-covalent interactions, including hydrogen bonds, coordination bonds, and π–π interactions, play a key role in dissipating mechanical energy, particularly in elastomers and dynamic polymer networks [6]. For instance, Cu(II)-coordinated benzoquinone dioxime-carbamate units can enhance hydrogen bond strength and optimize π–π stacking interactions, resulting in improved toughness and thermodynamic stability [6]. The reversible dissociation of large numbers of hydrogen bonds during cyclic loading has been identified as a major energy dissipation pathway in such systems [6].

Dynamic polymer networks further contribute to toughness by enabling reversible bond exchange and segmental rearrangement under stress. Dynamic cross-linking units, such as those formed via Cu(II) coordination, allow polymers to recover their original structure after deformation while lowering the energy barrier for network reconfiguration [5]. This dynamic behavior significantly enhances energy dissipation efficiency and mechanical resilience [5].

In multiphase polymer systems and alloys, elastomer deformation and interfacial debonding represent dominant energy dissipation mechanisms. Elastomer domains act as stress concentrators, deforming and debonding from the matrix under load, which absorbs large amounts of energy and initiates-controlled crack formation [5]. The associated formation of silver streaks and shear bands effectively suppresses craze propagation and prevents catastrophic fracture [5].

Additional energy dissipation pathways are observed in systems containing crystalline domains and entangled macromolecular chains. In hydrogels, crystalline regions function as physical cross-links that require significant energy to break, thereby redistributing stress and hindering crack growth [5]. Simultaneously, the untangling and stretching of polymer chains during deformation dissipates substantial energy, leading to enhanced tensile strength and toughness [5].

Sliding cross-link strategies provide another advanced route to super toughness. In systems such as poly rotaxanes, ring molecules are able to slide along polymer chains, enabling reversible molecular slippage under external force. This mechanism efficiently releases stress, enhances stretchability, and significantly improves toughness without compromising mechanical strength [3,6].

2.3 Advanced Mechanical Responses and Functional Toughness

Beyond conventional energy dissipation, advanced polymer systems may exhibit additional mechanical responses that further enhance durability and functionality. Self-healing polymers, for example, utilize photothermal effects or reversible bond dynamics to repair damage after fracture. Photohealable elastomers coordinate thermodynamic stability, kinetic activity, and external field responsiveness, enabling efficient light-driven healing processes [3]. Materials such as PIB5Cu, with optimized π–π interactions and Cu (II)-catalyzed carbamate dynamics, demonstrate high near-infrared photothermal healing efficiency [6].

Thermal stability and viscoelastic behavior also play important roles in long-term mechanical performance. Strong intermolecular interactions contribute to thermodynamic stability, while sufficient molecular mobility ensures kinetic activity and viscoelastic adaptability [3]. Polymers incorporating high-energy locking units, such as Cu-BQDU, exhibit exceptional creep resistance and stability under prolonged mechanical stress [4]. Additionally, robust cross-linked polymer networks often display excellent solvent resistance, maintaining structural integrity even after extended solvent exposure [4].

2.4 Link to Super toughening Mechanisms

Collectively, these fundamental mechanical responses and energy dissipation pathways form the basis for various super toughening strategies in polymers. By tailoring polymer microstructure, intermolecular interactions, and network dynamics, it is possible to design materials that effectively suppress crack propagation and achieve exceptional toughness. These principles provide a foundation for the classification of super toughening mechanisms, including elastomer toughening, rigid particle toughening, grain refinement, interfacial engineering, and multilevel energy dissipation approaches, which are discussed in the following section.

3. Structural Design Strategies for Super Toughened Polymers

One of the most effective approaches for enhancing polymer toughness involves polymer blending and copolymer design, which enables stress redistribution, energy dissipation, and controlled crack propagation through tailored microstructures and intermolecular interactions.

Elastomer toughening is a widely used strategy in which elastomers are incorporated into semi-crystalline polymer matrices. In these systems, elastomer particles act as stress concentration sites that deform and debond from the surrounding matrix under external loading. This process induces the formation of numerous silver streaks and shear bands, which hinder crack propagation and suppress catastrophic fracture, thereby significantly enhancing toughness. However, this approach is often accompanied by a noticeable reduction in elastic modulus. The toughening efficiency is strongly dependent on elastomer particle size, and an optimal range—typically around 0.2–0.3 µm—is required to maximize toughness enhancement within the polymer matrix.

Polymer blending with controlled phase separation provides another effective route to toughness improvement. Mixing polymers such as PA66 and PA6 creates multiple phase interfaces within the material. These interfaces serve as preferential sites for crack initiation and propagation, leading to substantial energy consumption during crack growth and, consequently, enhanced impact resistance. For example, PA66/PA6/POE-g-MAH ternary alloys exhibit higher crack propagation energy than corresponding binary systems due to the synergistic interaction between POE-g-MAH and the PA66/PA6 matrix.

Copolymers can also be deliberately designed to introduce strong and specific intermolecular interactions. A notable example is the incorporation of Cu(II)-coordinated benzoquinone dioxime-carbamate (Cu-BQDU) units, where coordination bonds effectively polarize hydrogen bonds and increase their strength to 5.6 kcal/mol—the highest reported value for carbamate–carbamate segments. This coordination further extends molecular conjugation length and optimizes π–π interactions, resulting in exceptional thermodynamic stability and a record-high toughness of 236.0 MJ/m^3.

3.1 Double and Interpenetrating Polymer Networks

Advanced network architectures, including double and interpenetrating polymer networks, offer powerful strategies for achieving high toughness without sacrificing stiffness or strength.

Core–shell structured systems involve the formation of rubbery or crystalline phases encapsulated within a shell material. These architectures enhance interfacial friction and reduce interparticle distance, which promotes efficient energy dissipation. For instance, the introduction of a ductile β-polypropylene interface in core–shell structured polypropylene random copolymers reduces crack-tip stress concentration, leading to a several-fold increase in fracture energy compared to systems with weak or rigid interfaces. By optimizing the content of ductile interfaces, impact toughness can be improved from approximately 10.8 kJ/m^2 to 18.2 kJ/m^2.

Dynamic cross-linking networks utilize reversible bonding interactions—such as coordination bonds, polarized hydrogen bonds, π–π interactions, and oxime–carbamate bonds—to construct ultra-strong yet adaptable polymer networks. The Lewis acidity of Cu(II) ions can catalyze the dynamic exchange of oxime–carbamate bonds, enhancing self-healing efficiency and photothermal performance. These reversible interactions facilitate rapid segmental rearrangement, enabling the material to recover its original shape and mechanical properties after deformation.

Interpenetrating polymer networks (IPNs), particularly in hydrogel systems, achieve a balance between high stiffness and toughness by optimizing inter-polymer interactions and promoting polymer crystallization. The resulting dense network structure contains numerous entanglement points that require substantial energy to disentangle during stretching, while crystalline domains act as robust physical cross-links that resist fracture and enhance toughness.

3.2 Nanocomposites and Hybrid Systems

Nanocomposite and hybrid polymer systems further expand the toolbox for achieving superior mechanical performance through multiscale reinforcement mechanisms.

The incorporation of nanofillers, such as flexible SiO_2 nanofibers (SNF), into polymer matrices like polyvinyl alcohol (PVA) can dramatically enhance mechanical properties. Strong intermolecular interactions between SNF and PVA enable effective stress transfer and damage dispersion, while crystalline domains serve as physical cross-links that impede crack propagation and dissipate energy. As a result, such nanocomposites can achieve high tensile strength (7.84 ± 0.10 MPa) and toughness (9.9 ± 0.4 $MJ\ m^{-3}$).

In geopolymer systems, the addition of appropriate amounts of graphene oxide (GO) to fly ash–slag matrices significantly improves toughness and mechanical strength. Under alkaline conditions, GO undergoes reduction and forms a strong electron cloud at its edges, altering its binding mode with calcium ions. This process creates a calcium-rich environment that promotes the nucleation and growth of C-(A)-S-H gel phases. Consequently, microscopic pores are filled, crack sizes are reduced, and the overall compactness and structural stability of the geopolymer are enhanced.

Biomimetic layered nanocomposites represent another advanced hybrid strategy inspired by natural materials. In these systems, chain-sliding cross-linking is introduced at interfaces between inorganic nanosheets, such as sulfonated graphene, and polymer layers like polyurethane. Ring molecules sliding along linear polymer chains release accumulated stress and provide sufficient interlayer spacing for energy dissipation. This unique architecture enables the development of ultratough, highly stretchable, and self-healing materials with high tensile strength (22.33 MPa) and exceptional toughness (219.08 MJ m^{-3}).

Overall, these diverse material design strategies share a common objective: to effectively manage stress, enhance energy dissipation, and control crack behavior across multiple length scales, thereby achieving superior mechanical performance in advanced polymeric materials.

4. Experimental and Results

The tensile and impact strengths of PA66/PA6/POE-g-MAH alloys were measured and are presented in Figure 1. As shown in Figure 1(a), the tensile strength of PA66/PA6/POE-g-MAH initially decreased and then increased as the amount of PA66 increased. The relationship between the tensile strengths of PA66/POE-g-MAH and PA6/POE-g-MAH did not exhibit a linear variation. This suggests that there is another significant factor contributing to the enhancement of tensile strength in PA66/PA6/POE-g-MAH alloys, aside from the superior tensile strength of PA66 and the inferior tensile strength of PA6. Specifically, when the ratio of PA66 to PA6 changed from 2:8 to 6:4, the tensile strength of the PA66/PA6/POE-g-MAH alloys remained lower than that of the PA6/POE-g-MAH alloy. The tensile strength of the PA66/PA6/POE-g-MAH alloy at a PA66 to PA6 ratio of 8:2 was found to be greater than that of the PA6/POE-g-MAH, yet lower than that of the PA66/POE-g-MAH. This difference is likely closely related to the quantity of interfaces present between PA66 and PA6. When the PA66 to PA6 ratio ranged from 2:8 to 6:4, numerous interfaces resulted from the distinct separation of the PA66 and PA6 phases. These interfaces were more susceptible to damage during the tensile testing, resulting in lower tensile strength. In samples with a PA66 to PA6 ratio of 8:2, as the amount of PA6 decreased, so did the number of phase interfaces. This suggests that the PA66/PA6/POE-g-MAH alloy exhibited reduced vulnerability. Consequently, the tensile strength of the PA66/PA6/POE-g-MAH alloy, with a ratio of PA66 to PA6 of 8:2, showed an increase. To summarize, the tensile strength of PA66/PA6/POE-g-MAH alloys was significantly influenced by multiple interfaces between the PA66 and PA6 phases [3].

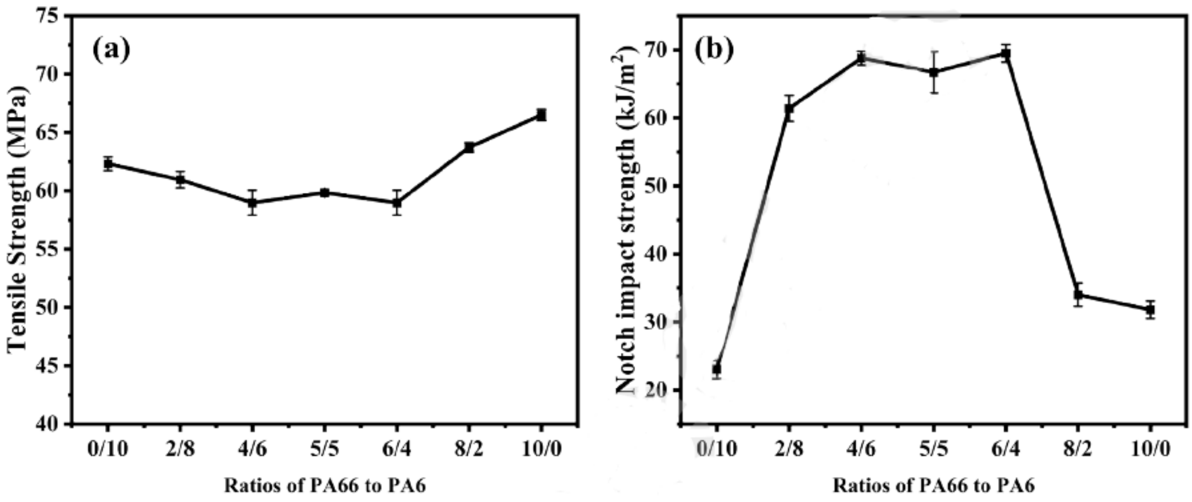

Figure 1. Tensile strength (a) and impact strength (b) of the PA66/PA6/POE-g-MAH alloys with 177 different PA66 to PA6 ratios [3].

The temperature and duration of annealing are two crucial factors that influence the mechanical characteristics of FDA-SNF/PVA. Figures 2a–c demonstrate that the mechanical properties of FDA20 improve with longer annealing times at various temperatures, peaking at 90 minutes. Furthermore, for each specific annealing temperature, the mechanical properties of FDA20 enhance with increased annealing time until reaching a maximum, after which they either stabilize or decline (Figure 2d–f). Generally, the mechanical properties of hydrogels exhibit an upward trend as the annealing temperature rises. The tensile strength of FDA20-60-90 treated at 60 °C measured 2.23 ± 0.31 MPa, significantly higher than that of FD, which was 0.79 ± 0.08 MPa (Figure 2d). As the annealing temperature rises from 60 °C to 120 °C, both the tensile strength and toughness of FDA20, along with its modulus of elasticity, increase significantly (Figure 2d–f). However, upon further increasing the annealing temperature to 150 °C, a decline in the tensile strength and toughness of FDA20 is observed, while the modulus of elasticity continues to rise (Figure 2d–f). This may result from a significant increase in crystal domains and their uneven distribution at 150 °C, which leads to stress concentration, an excessive elastic modulus, and a reduction in tensile strength and toughness. Furthermore, we noted a significant enhancement in mechanical properties with higher concentrations of PVA (Figure 2g). In comparison to the Freeze–Thaw hydrogel (FT), Freeze–Drying hydrogel (FD), and FDAPVA, the FDA-SNF/PVA exhibits distinct and remarkable benefits regarding the combined mechanical properties of robust PVA hydrogels (Figure 2h). More specifically, the tensile strength, toughness, and modulus of elasticity for FDA20-120-90 are recorded at 5.77 ± 0.38 MPa, 6.31 ± 0.32 MJ m^{-3}, and 10.56 ± 0.24 MPa, respectively, which significantly surpass those of FDAPVA (2.49 MPa, 1.58 MJ m^{-3}, and 6.41 MPa). The tensile strength, toughness, and modulus of elasticity of FDA20-120-90 are 7.3, 11, and 19.2 times greater than those of FD, and 48, 94.4, and 101.3 times greater than those of FT. As illustrated in Figure 2i, FDA20-120-90 can bear a load of 2 kg (Video S1, Supporting Information), which is over 4600 times its own weight, and it remains capable of being knotted even after lifting heavy objects, demonstrating that FDA-SNF/PVA possesses exceptional mechanical properties under stretching [4].

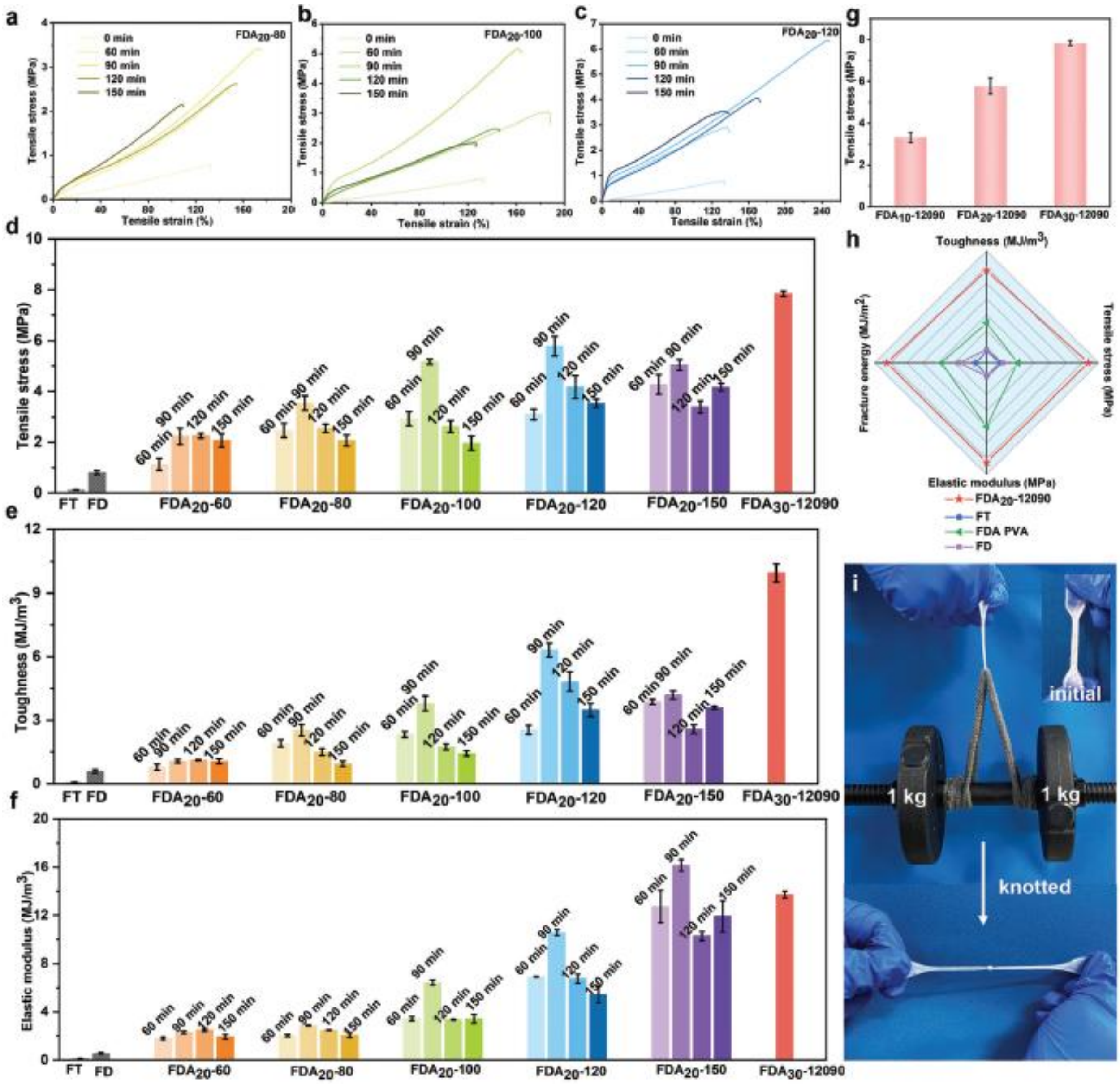

Figure 2. (a–c) Tensile stress–strain curves of FDA at different annealing temperatures and annealing times from 0–150 min: a) 90 °C, b) 120 °C, c) 150 °C; (d–f) Bar charts showing different mechanical properties of SNF/PVA composite hydrogel: d) Tensile strength, e) Toughness, f) Elastic modulus; g) Histograms of the tensile strength of FDA with different initial PVA concentrations (10, 20, and 30 wt%) after annealing at 120 °C for 90 min; h) Comparison of the mechanical properties of FT, FD, FDAPVA, and FDA20-120-90; i) Photographs demonstrating the excellent mechanical strength of FDA20-120-90, which can support two dumbbells (total of 2 kg), weighing >4600 times its own weight. (FDAX-YZ, X, Y, and Z represent the mass fraction of PVA in H_2O solution, annealing temperature, and annealing time, respectively). Data in (d–g) are means ± standard deviation (SD), n = 3 [4].

The mechanical properties of PEO and Liquid Metal (LM) electrolyte, pure PEO, and PEO and Solid Organic (SO) electrolyte were tested. Young's modulus of PEO and LM electrolyte is 3.76times that of PEO and SO electrolyte (Figure 3). Generally, a high Young's modulus means good stiffness and elasticity, which can also serve as a parameter for the effectiveness of inhibiting sodium dendrites [2].

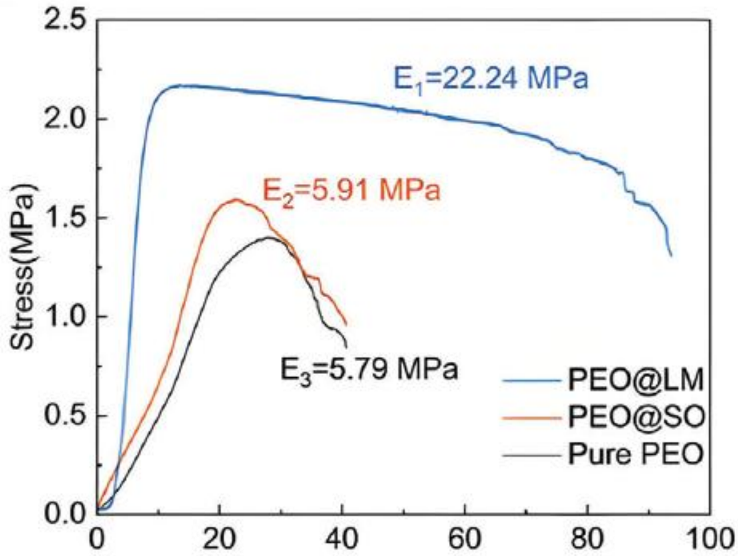

Figure 3. Stress-strain curve of PEO and LM, PEO and SO, and pure PEO electrolyte [2].

5. Challenges and future perspective

Achieving super toughness in polymers involves significant challenges due to inherent trade-offs between thermodynamic stability, kinetic activity, and external field responsiveness. Traditional cross-linked elastomers provide stability but limit segmental motion, whereas dynamic covalent polymers offer responsiveness but often lack sufficient thermodynamic stability. Photo-healing systems can suffer from non-uniform filler distribution, affecting tensile strength and photothermal efficiency, while intrinsic systems must balance chain mobility for healing with structural stability under prolonged exposure. Elastomer toughening may reduce modulus and tensile strength, and rigid particle incorporation requires ductile matrices and uniform dispersion, which are difficult to achieve simultaneously. Grain refinement and core-shell structures demand advanced processing and careful design, and biomimetic layered nanocomposites face interfacial weaknesses due to modulus mismatch.

Super toughness arises from multiscale mechanisms. At the molecular level, Cu(II) coordination bonds and hydrogen bonding enhance bond strength, conjugation, and π–π interactions, improving photothermal efficiency, stability, and toughness. Polymer chain mobility and crystalline domain formation in hydrogels dissipate energy and improve stiffness. At the microscopic scale, phase interfaces, elastomer debonding, crazing, core-shell architectures, and microfibril cooperativity all contribute to energy dissipation and crack resistance. At the nanoscale, well-dispersed SiO_2 nanofibers, reduced graphene oxide as nucleation sites, and liquid metal nanoparticles enhance load transfer, crystallinity, and mechanical properties.

Overall, overcoming these challenges requires carefully designed polymers that exploit multiscale mechanisms to dissipate energy, control crack propagation, and maintain mechanical resilience, enabling the realization of super tough polymer systems.

6. Conclusion

The development of super tough polymers represents a paradigm shift in the design of advanced functional materials, combining exceptional energy absorption, fracture resistance, and structural integrity. This work demonstrates that super tough behavior is not an intrinsic polymer property but an emergent feature achievable through multiscale material design. At the molecular level, dynamic interactions—including Cu (II)-mediated coordination, hydrogen bonding, and π–π stacking—enhance bond strength, network stability, and reversible stress dissipation. Microscale structural features, such as elastomer domains, phase-separated interfaces, crazing, and core–shell architectures, further facilitate controlled crack propagation and efficient energy dissipation. Nanoscale reinforcements, including SiO_2 nanofibers, reduced graphene oxide, and liquid metal nanoparticles, optimize load transfer, crystallinity, and mechanical robustness. Experimental studies on systems such as FDA-SNF/PVA hydrogels and PA66/PA6/POE-g-MAH alloys confirm that careful control of composition, annealing conditions, and network architecture can produce materials with tensile strength, toughness, and elastic modulus surpassing conventional systems by orders of magnitude. Collectively, these findings underscore the importance of hierarchical structuring and multilevel energy dissipation in achieving super tough polymers. This approach provides a versatile framework for designing next-generation polymeric materials with unprecedented mechanical performance, high resilience, and functional adaptability for structural, electronic, and biomedical applications.

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