



ROLE OF NON-COVALENT BONDS AND TOPOLOGICAL FACTORS IN IMPROVING CROSSLINKED EPOXY POLYMER PROPERTIES

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Abstract : In this review, we continued our analysis of epoxy curing process. We have worked to sift the literature to propose twelve factors that, either alone or in combinations, play important role in improving properties of epoxy crosslinked networks. These twelve factors can be subdivided into non-covalent bonds and topological factors associated with the crosslinked networks. Few reports also suggest that these factors can interact with characteristics of crosslinked networks like crosslinking density, fractional free volume etc., to bring about rarely reported simultaneous improvement in several properties of the cured materials. Some of the factors also participate in the curing process of other thermosetting resins. Combination of these twelve factors and earlier reported ten parameters from our recent review can broaden the understanding of curing reactions of epoxy and other thermosetting polymers.

Index Terms – epoxy, crosslinking reaction, thermosetting resin, non-covalent bond, topology

Abbreviations- Polyurethane (PU), Crosslinked polyethylene (XLPE), Diglycidyl ether of bisphenol A (DGEBA), Diglycidyl ether of bisphenol F(DGEBF), 9,10-Dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO), Multi wall carbon nanotube(MWCNT), Poly dimethyl siloxane(PDMS), poly(ϵ -caprolactone) (PCL), Di amino diphenyl methane (DDM), Di amino diphenyl sulfone(DDS)

I.INTRODUCTION

Crosslinked polymers such as epoxy, PU, elastomers, and XLPE rely on covalent bonds to create crosslinked networks. These covalent bonds in the networks have high bond energy that provides excellent thermal stability and other valuable properties. However, these covalently crosslinked networks are difficult to reprocess at the end of useful life, leading to persistent polymeric waste that poses environmental risks. Despite these concerns, demand for thermosetting polymers continues to grow, with the global market expected to reach 201 billion USD by 2032, indicating they will remain widely used in the near future [1]. Hence, they continue to be an active area of research.

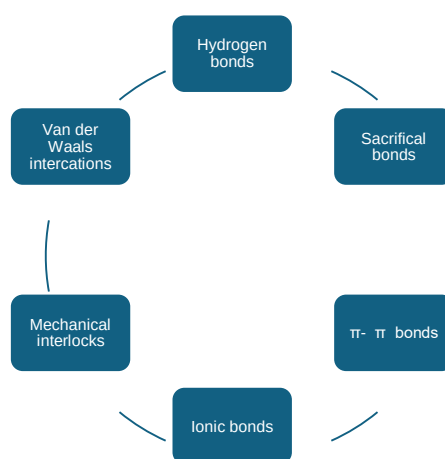
It may be noted that quest to understand relation between crosslinking process and properties of cured materials in classical polymer chemistry continues from past many decades to present time as seen from few examples of fundamental work reports cited here- basic condensation polymerization of multi-functional polymers [2], creep resistance in crosslinked phenolic polymer[3], visco -elasticity studies in rubbers [4], effect of stoichiometry on material properties of epoxy [5], development of mathematical model for very fast crosslinking reaction of polyurethane[6], statistical treatment for network formation reaction beyond gel point in epoxy[7] and mechanisms for kinetics of crosslinking reactions of different epoxy systems[8]. The topic remains active for variety of reasons like studies of nanocomposites, exchangeable bonds containing networks, new techniques like 3D printing, developments using machine learning platforms and artificial intelligence, new bio-based resins and the need for recycling of thermoset resins.

In the last two decades, the need to reduce waste from thermosetting resins has been pushing researchers to find viable solutions. High crosslink density was considered as a major barrier because such thermoset polymers have permanent shape and hence, they degrade when heated. This challenge led to studies on optimizing crosslinking in systems such as epoxy and other resins to help recycling of these resins. Researchers noted that many natural polymers depend not only on covalent crosslinks but also on secondary networks to satisfy application requirements [9,10]. Various non-covalent interactions are possible in natural polymers [Scheme 1]. Further, the hierarchical structures in natural polymers provide strength yet allow reassembly when stimulated,

inspiring work in synthetic hydrogels [11]. Another report shows that development of synthetic crosslinked polymers to reduce covalently bonded crosslinks and simultaneously improve properties like T_g using hydrogen-bonding as non-covalent bonds is possible [12]. The Hydrogen bonding was also found to play a key role in naphthyridine type building blocks containing polymers [13]. This type of study falls under the broader area of supramolecular polymer science.

Two major breakthroughs for recycling thermosetting resins came about 15 years ago. First, the report on covalently adaptable networks that allow bond exchange in crosslinked polymers from Bowman group [14] and then report on design of thermosetting materials that behave like malleable silicate glass that are also called vitrimers [15-17]. Both these approaches allow change in crosslinking density by reprocessing without significant thermal degradation of the used material.

Two more points are relevant for understanding the crosslinking process in epoxy composites for improving properties of cured materials. These are as follows: (i) Recent modeling and experimental studies show that epoxy composites require sufficiently high values for properties like stiffness and toughness to withstand diverse mechanical and impact loads for allrounder performance [18-22]. This highlights that a highly crosslinked matrix alone is insufficient since improvement in toughness usually comes with reduction in tensile strength and elongation. Looking from a little different angle, it is also consistent with extensive use of various fillers and variety of additives in epoxy formulations reported in the literature [23, 24] (ii) It is known that prolonged exposure to stimuli like moisture and high temperature can lead to crosslink scission, new crosslink formation, and surface-microcracking. This will bring about gradual changes in various properties and those that are directly related to the application [25]. Such studies are usually considered under ageing but need detail analysis to prolong the life of cross-linked material, in the context of sustainability issues.



Scheme 1 Various non-covalent bond interactions seen in natural polymers

With this background, we briefly touch upon our recent study of epoxy curing process. We focused on analyzing the epoxy curing process by reviewing reports on different crosslinking reactions and their influence on final properties[26]. This led to characterization of the curing process using ten variables and their interactions within covalently crosslinked networks [26]. These ten parameters include following: raw materials, gel time and vitrification, kinetics, residual stress, relaxation processes, free volume, reaction induced phase separation, interface and interphases, synergy and simultaneous improvements, percolation.

Because epoxy composites contain several components beyond resin and hardener, understanding both individual effects and interactions of these components are essential for identifying pathways to property enhancement. One discussed case involved use of imidazole functional hyperbranched additive (HBP), that is capable of reacting with the resin as well as the anhydride hardener during curing [27]. This provides a system that has HBP as additional reactant. This HBP containing system can make small yet decisive changes in characteristics of the network namely, crosslink density, fractional free volume, and T_g, to bring about significant changes in properties relative to the control [27]. Furthermore, at 9% additive concentration, an optimal balance of these variables resulted in simultaneous improvements in mechanical and impact properties. This suggests that epoxy curing is a complex process influenced by multiple interacting factors. A natural question is why epoxy composites remain widely studied and commercially successful despite the complexity due to several variables for the epoxy curing process. The answer probably lies in the widely practiced test of measuring T_g as a reliable indicator of adequate curing for a given application. Familiar tests such as heat distortion temperature and DSC-based T_g measurements provide an indirect but reliable measure of degree of crosslinking, and final quality is confirmed through application specific tests.

Continued study of epoxy literature revealed that several non-covalent interactions in epoxy systems must be considered to characterize the crosslinked network for better explanation of property enhancements. In line with the studies on natural polymers, the six types of non-covalent bond interactions are found to play important role in changes in epoxy polymer properties [Scheme 1]. In addition, we propose adding trap-related interactions to this list. The traps can be considered as important types of non-covalent factors used for understanding electrical insulation performance in epoxy, XLPE and other polymers. Yet another kind of influencing parameters identified from literature are long pendant chains, loops, and end groups. In the literature, these are usually described as topological factors [28]. They are known to be different components of any crosslinked polymer structure. In addition, we propose following two as additional topological variables for understanding epoxy curing process : (i) crosslinking density (ii) structural order introduced in the cured matrix. Structural order can be created using different ways like (a) by use of covalently bound mesogenic resin or curing agents (b) using certain reactive groups (c) combining phase separation with use of block copolymer and nanofiller as additives[29]. All these routes help to improve properties. A recent study illustrates this point [30]. The authors of this study developed a new simulation method to capture the effect of structural order for a ternary epoxy system containing a resin, curing agent, and mesogenic comonomer. By varying crosslink density and comonomer content, they observed that mesogenic comonomer help to create molecular order in the crosslinked structure that helps for effective redistribution and

energy distribution. This helps to simultaneously improve toughness, stiffness and thermal stability [30]. More examples are discussed later in the paper in section 3.4.

Combining all the above information, we have considered the variables this described in Table 1 for review of improvement in properties of epoxy and other crosslinked polymers. Their importance is highlighted by a recent example: sacrificial bonds with careful selection of composition enabled fabrication of a polyurethane elastomer with a rare simultaneous improvement in high tensile strength of 98.5 MPa and ultra-high elastic resilience [31]. Overall, improvements in one or more properties arise from interactions between these twelve factors and the evolving network, as described in the following subsections. Examples for ready reference appear in italics (pages 4, 5, 6, 9, 10). It must be noted that proper raw material selection is basic requirement for non-covalent bond interactions. That means, non-covalent bond factors interact with raw material as one of the ten factors proposed earlier [26].

Our objective in this review is twofold. Firstly, collect and analyze the literature on role of various non-covalent bonds in improving properties of epoxy and many other crosslinked polymers like PU, Elastomers, poly benzoxazine, polysiloxane and cyanate esters etc. Secondly, discuss the literature reports on each of the twelve factors listed in Table 1 that are part of the crosslinked networks, and limited to how they contribute to improvement of product properties. We will show that some of these twelve parameters interact within themselves or with topological factors to provide improvement of network properties in some specific cases. Literature in the last ten years has been considered for this paper. The framework for the ensuing discussion of the review is as follows: (i) section 2- role of different non-covalent bonds (ii) section 3- role of topological factors (iii) section 4- Miscellaneous reports (iv) section 5- Conclusions and future scope. We believe that analysis of literature presented here, when combined with our earlier study, provides a new perspective on epoxy crosslinking and offers a broader understanding of the key variables governing the thermosetting polymerization processes.

Table 1 Various topological and non-covalent bond factors for improving composite properties

Non-covalent bond	Topological factors (VN)
Hydrogen bonding	Crosslinking density
Sacrificial bonds	Dangling chains (long pendant groups)
π - π bonds	Loops
Ionic bonds	End groups
Mechanical interlocking	Molecular order of chains in the network
Van der Waals interactions	
Traps for electrical insulation	

2. Role of different non-covalent bonds

Effects of each of the seven non-covalent bond factors on properties of epoxy and other polymer composites are discussed in this section. The reports show that mechanical, thermal and other properties can be improved. Some new concepts are discussed.

2.1 effect of Hydrogen bonds

Hydrogen bonding (HB) is a strong intermolecular interaction formed between a hydrogen atom bonded to an electronegative atom (e. g., O, N, or F) and a nearby electronegative atom. In cross linked thermosets such as epoxy, polyurethane, and siloxane, HB is abundant and contributes to improved various properties including toughness and thermal conductivity (Table 2). HB often works together with network factors such as crosslinking density and fractional free volume, making these systems complex and analytically demanding. This is shown in the recent report by Joo et al [32]. In gas separation membranes based on polymers of intrinsic microporosity, Joo et al. showed that hydrogen bond forming groups play a complementary role in crosslinking. HB groups help regulate the free volume hole size and its distribution, which are key parameters governing physical aging and changes in gas permeation properties [32]. In epoxy-amine systems, the Caillol group demonstrated that HB significantly influences reactivity of the epoxy-amine systems as well as curing kinetics and final properties[33-35]. Further, they proposed an amine reactivity scale independent of functionalized secondary amines[ref 33]. HB can also aid degradation in certain unsaturated polyesters, suggesting potential relevance for sustainable disposal and recycling [36]. Hydrogen-bonds formation toughens and strengthens crosslinked network in benzoxazine and few other types of networks because these bonds can break and reform, allowing them to dissipate energy during deformation. At the same time, hydrogen bonds serve as temporary crosslinking points, increasing the effective crosslink density and thereby enhancing the material's strength [37].

Table 2: Effect of hydrogen bonding on crosslinked network and properties of materials

System	Key result	Reference
epoxy	Hydrogen bonding was identified as the dominant factor controlling thermal conductivity (TC) in amorphous epoxy networks, if hydrogen bonding molecules are used in significant amounts. The highest TC value reported was 0.4 W/(m. K).	38
epoxy	Two types of epoxy systems were evaluated by simulation, with and without HB. Further, the simulation study evaluated topological factors(VN) like free volume distribution, crosslinking density, and modulus as the property for these systems. Furan ring epoxy molecules provide more hydrogen bonding sites in the network containing hydrogen bonding while it was absent in the other two epoxies with hydrocarbon ring. As a result of HB, Furan based system shows lower fractional free volume and lower crosslinking density but higher modulus. It has relatively lower Tg than the other two samples. <i>This work is one of the few studies showing how hydrogen bonding parameters interact with VN factors of crosslinking density and free volume content.</i>	39

Polyurea	Supra molecular polyureas were prepared by copolymerizing diisocyanate with ureidopyrimidinone (UPy) groups bearing diamines and covalent diamine/triamine monomers. The UPy groups introduce strong, reversible quadruple hydrogen bonds, while triamines create permanent crosslinks for strength and solvent resistance. Films with 9.7% and 14.6% UPy showed high toughness (74.17 and 124.17 MJ m ⁻³) and remained transparent. The 14.6% UPy material was recycled five times with retained properties. <i>This demonstrates that hydrogen bonding can enhance both toughness and recyclability.</i>	40
PU	This work uses a diol and amine with hydrogen-bonding groups and a diisocyanate to form a polyurethane network with a non-covalent crosslinker. The material shows high tensile strength of 50 MPa, 300 MJ/m ³ toughness, and over 96% recycling efficiency after six cycles. Higher hydrogen-bondable content produces smoother recycled surfaces by reducing wrinkles.	41
PU	A series of PU polymers were synthesized using two polyols (poly(tetramethylene ether)glycol and poly(carbonate diol)) as soft segments and MDI/Butanediol as hard segments. Increasing the hard segment content strengthened microphase separation and improved tensile strength and dielectric constant. The optimized sample showed the best performance, with the distinct phase separation, a dielectric constant of 10.50 at 10 ² Hz, and a tensile strength of 18.33 MPa. Hydrogen bonding from polyol contributed to property enhancement.	42
Epoxy	A combination epoxy-amine system and semi-interpenetrating polymer having exchangeable ester bonds and hydrogen bonding groups in the network was studied. Hydrogen bonding helps in impact energy dissipation while crosslinks in epoxy provide adequate rigidity. Further, use of excess amine generates tertiary amine that catalyzes ester bonds exchange and also increases hydrogen bonding. This allowed high tensile strength of 123 MPa and very high impact strength of 52.3 kJ/m ² .	43
Bio-based epoxy	Epoxy composite wherein bio-based vanillin-derived hardener forming supramolecular hydrogen bonds along with exchangeable imine covalent bonds was reported. It showed better mechanical and thermal properties, lower activation energy for bond exchange, and good recyclability and self-healing. The vitrimer matrix retained over 86 % of lap shear strength after multiple re-bonding cycles and recovered carbon fibers maintained 92 % of tensile strength in reuse. Density functional theory calculations confirmed role of hydrogen bonding in reducing activation energy for bond exchange.	44
Nitrile-Butadiene rubber (NBR) composite	The composite of NBR with hindered phenol filler (HPF) has application in damping materials. Excellent hydrogen bonding was detected between cyano groups and HPF in the composite by simulation study. Further, it was found that at NBR/HPF ratio 100/68, the composite showed the maximum loss factor and highest modulus. This improvement also coincides with the smallest fractional free volume content for the composite. <i>There is thus interaction between hydrogen bonding parameter with free volume characteristic of the crosslinked network and mechanical properties.</i>	45

2.2 Role of sacrificial bonds in improving properties of composites

The sacrificial bonds are expected to be weaker than covalent bonds, but they meet the important requirement of protecting the network of covalent bonds from impact of stimuli. There are three types of sacrificial bonds commonly seen in the literature [46]. The first type is sacrificial hydrogen bonds. The second type are metal-ligand coordination bonds, and the third type are ionic bonds (discussed later). The first and second type of bonds are discussed here.

We will discuss examples of epoxy systems showing use of sacrificial bonds first and then discuss other systems in the next paragraph. Zhang et al synthesized adduct between different Imidazole compounds[IM, 2MI,EMI] and bismaleimide[BDM] by Michael addition between bismaleimide and IM or 2MI or EMI and used as latent curing agents and flame-retardant synergists for DOPO modified epoxy [47]. They showed excellent storage stability with a pot life over 40 days. Upon heating, they underwent retro Michael reaction to release BDM and free imidazole, enabling fast imidazole-initiated epoxy curing. The C-N bond between BDM and IM serves as sacrificial bond in the retro-Michael reaction. The regenerated BDM then homopolymerizes and gets integrated with the epoxy network. This improves thermal stability and flame retardation via significant char formation. Use of peculiar structure of graphene based stacked-cup low-cost carbon nanofibers allow the composite to respond to impact in two ways: (i) pi-pi interaction between layers in the side walls to splay and absorb impact (ii) combine the uncoiling action of cup-stacks while doing crack-bridging and avoid damage to structural integrity if graphene sheets[48]. These mechanisms allow use of only 0.68% CNF to improve toughness by 40-112% in different combinations of tri and tetra functional epoxy resins. Further, these composites show good electrical conductivity required in some applications. Use of cyano group (CN) containing tung oil was designed based on inspiration from natural systems [49]. It works as a renewable toughener for epoxy due to sacrificial hydrogen bonding between CN groups and the hydroxyl groups in the matrix [49]. There was simultaneous improvement using sacrificial hydrogen bonding as shown by significant improvement of impact strength, elongation at break, and toughness to reach 76.7 kJ/m², 25.0%, and 7.6 MJ/m³, respectively (control sample showed the values of 21.1 kJ/m², 5.7%, and 3.1 MJ/m³, respectively). Li et al developed a compact 3D tannin modified graphene/MWCNT hybrid epoxy network. The spectroscopic evidence confirmed pi-pi interaction between MWCNT and tannin. This pi-pi interaction also worked as sacrificial bonds needed for toughening [50]. With only 0.01 wt% filler, the epoxy showed notable enhancements in tensile strength (23.3%), Young's modulus (44.6%), elongation at break (41.2%), impact-resistance factor K_{IC} (114.6%). This loading is 10–15 times lower than previously reported levels required for

similar improvements in tensile strength and impact-resistance. The composites also exhibited excellent electrical performance with an ultralow percolation threshold of ~ 0.076 wt%.

In a polyurethane system, Liu et al. designed novel biobased materials by covalently attaching UPy groups to double bonds in a PU elastomer [51]. The UPy segments form hydrogen bonded aggregates that act as reversible sacrificial units, breaking and reforming under stimulus to significantly enhance modulus, strength, and elongation. These hydrogen bonded aggregates also retard full relaxation during stretching, giving the elastomer shape memory behavior. Overall, the sacrificial bonds with UPy groups provide an effective mechanism to tune modulus and elongation under stress. Chen et al [46] prepared semicrystalline polyesters containing sacrificial bonds (SB) created by the coordination of dangling terpyridine (TPY) ligands with transition metal ions. These coordination bonds form supramolecular cross-linking points as sacrificial bonds, in a double-cross-linked system. Tensile testing showed that stronger coordination bonds provide a larger Young's modulus, but not for strength and toughness, which increase first and then decrease after achieving a maximum value at an optimal SB strength. Li et al designed metal-ion coordination based tunable sacrificial bonds whose strength depends on metal ion content [52]. Further, they used this strategy in a PDMS elastomer with a 2,6 pyridine dicarboxamide ligand that anchors Fe^{2+} through a strong pyridine-iron bond. The weaker coordination of Fe^{2+} with the additional carboxamide groups in the ligand breaks and reforms easily (serving as sacrificial bonds), enabling high stretchability and self-healing.

2.3 role of π - π bonds in improving properties of crosslinked polymers

This type of interactions is not only seen in clear epoxy systems but also in fiber or filler reinforced composites used for applications like load bearing, and it also interacts with crosslinked density and free volume of network. The report of Tang et al is an example where π - π stacking interactions were intentionally engineered to improve strength and toughness in an epoxy composite system. The composite makes use of rolling of micron size silica spherical shape filler particles that helps for high toughening (358% higher than control) as well as 49.8% enhancement in strength over the control [53]. *This report is an example where π - π bonding parameters interact to improve two properties of a network.*

We will now discuss some reports on epoxy composites and in the second paragraph about other polymeric systems. The π - π interaction between dopamine and carbon fiber surface improved interfacial shear strength (IFSS) by 70% when carbon fiber surface modified with poly dopamine and GO was used to make the composite [54]. He et al reported a SBR composite made with surface modified nano silica. They used resveratrol (RES) for covalently grafting onto nano silica to produce RES functionalized silica filler (SiO_2 -RES), which was used as a functional filler in SBR composites. The grafted RES enabled dynamic π - π interactions with SBR chains, reducing the Payne effect and improving filler dispersion. As a result, SiO_2 -RES provided much higher rubber reinforcement efficiency than unmodified or silane modified filler [55]. Jiang et al evaluated how sizing concentration alters the surface morphology of carbon fibers and the interfacial and mechanical properties of CF/epoxy composites [56]. XPS confirmed increased surface functional groups and higher π - π^* transition intensity in the C1s spectrum after sizing. These changes provided higher interfacial shear strength and improved mechanical performance compared to untreated CF/EP composites. In their next paper, Jiang et al reported an epoxy-polyamine adhesive for superior underwater adhesion performance [57]. Various molecular interactions like hydrogen bonding and π - π bonding were proposed to explain the superior properties like a strength of 3.87 MPa after 50 days of immersion. Further, after the adhesive underwent 50 thermal shock cycles, it exhibited a marginally lower strength of 3.59 MPa [57]. A recent review discussed how the mechanical and impact resistance properties of epoxy-glass fiber composites were improved by adding reduced graphene nanofiller made using coal-based graphene oxide [58]. It stated that the π - π interaction between filler and polymer matrix is a major reason behind these improvements. Li et al made comparative study of two epoxy resins (with phenyl or biphenyl type pendant groups), to analyze the effect of length of pendant groups having π - π interactions ability within the network [59]. The resins cured with DDM showed that coefficient of thermal expansion decreased relative to control sample and Tg for network containing shorter length phenyl pendant group showed more increase in Tg than the other network. This was attributed to π - π interactions between chain segments that restricted chain movement and helped closer packing [59].

Xu et al studied high dielectric constant thermally stable cyanate ester composites having lower dielectric loss [60]. A special nanofiller was made by decorating reduced graphene oxide nanosheet with the ionic liquid monomer having epoxy and vinyl as functional groups. The π - π interaction between aromatic rings on reduced graphene and ionic liquid allows uniform adhesion. The vinyl groups were then polymerized by free radical process to get a polymeric coating on the graphene. The modified nanofiller was used in cyanate ester composite to get lower dielectric loss at higher frequencies simultaneously with a very low percolation threshold of 0.4 wt% and very high dielectric constant of 340. *The non-covalent π - π bonds thus influences material properties to show very low percolation threshold and give unprecedentedly high dielectric constant value.* In an interesting study, a novel strategy was used to improve insulation performance. A 1:1 blend of PI and PEI achieved an exceptionally high breakdown strength of 550 MV/m at 200 °C due to strong π - π interactions between their aromatic rings, which promoted dense molecular packing. This dense packing sharply lowered void density and free volume [61]. Although this system does not use crosslinked materials, the example is chosen as the value of dielectric breakdown strength at high temperature is exceptionally high and difficult to achieve in covalently crosslinked XLPE or epoxy. *The results show that π - π interactions can couple with network characteristics such as free volume to significantly enhance electrical insulation performance even at high testing temperature.*

2.4 Role of ionic bonds in the improvement of composite properties

Use of special raw materials containing ionic groups is required to get advantages of ionic bonds for performance improvements as seen from examples in Table 3. Ionic bonds are helping new applications needing self-healing (Table 3).

Table 3. Effect of ionic bonds on properties of crosslinked polymers

System	Key results	Reference
epoxy	The epoxy system using 1 ethyl 3 methylimidazolium chloride ionic liquid, which introduces cation–anion electrostatic interactions within the epoxy network was characterized. The ionic liquid acts as a crosslinking agent. At 5 wt %, ionic interactions significantly increase stiffness (~7 times improvement) compared with amine cured epoxy (6.95 GPa vs. 1.1 GPa for TETA hardener). Tg was ~100 °C for composite of). With DDS hardener, modulus was 2.96 GPa and Tg 175 °C. This difference in modulus is attributed to presence of ionic bonds originating from presence of rigid, planar, and aromatic imidazolium ring into the polymer network, which restricts segmental motion more effectively than even the biphenyl type linkages in DDS hardener. This paper also shows several roles that are played by IL depending upon concentration used.	62
PU	Hang et al studied the influence of variation in ionic liquid (IL) segment content on Tg, rheology, modulus and % elongation of the polyurethane homologous series containing a new chain extender (1,4-butanediol containing imidazole-type ionic liquid). The interaction of IL moieties significantly promoted physical crosslinking in PU. The polymer containing 34.4 wt% IL diol showed around 34 °C higher Tg and nearly 100% higher elongation than the control PU polymer.	63
PU	A non-covalent, ionically cross-linked polyurethane with excellent self-healing and recyclability has been developed for flexible conductive materials. Strong physical crosslinks formed through anion–cation electrostatic interactions (also help self-healing), while dynamic ionic bonds dissipate strain energy, giving the material high mechanical strength. A high tensile strength of 20.5 MPa along with 97% self-healing efficiency at 60 °C/1 hr was reported.	64
Elastomer based on ionic liquid group containing pendant chains	Liu et al. studied how chemical composition, microstructure, and especially side chain length affect PIL based elastomers. ATFTIR confirmed the microstructures and revealed H bonding and ion–dipole interactions acting as physical crosslinks. By adjusting polymerization conditions and side chain length, properties such as ionic conductivity, self-healing, and Tg were tuned. Increasing the imidazolium side-chain length from 1 to 9 greatly improved stretchability, from 124.58% to 1336.17%.	65

2.5 role of mechanical interlocking in improvement of composite properties

This kind of non-covalent interaction is reported in limited reports, and most of them are fiber reinforced composites the examples are discussed in Table 4. The surface treatment usually improves IFSS and /or ILSS (Inter laminar shear strength) and mechanical properties and it helped in carbon fiber recycling in one case (Table 4).

Table 4. Mechanical interlocking for improving properties of composites

System	Key result	Reference
epoxy	Mechanical interlocking and pi-pi interaction help to get synergistic improvements. Basalt fiber coated with polydopamine or polyethyleneimine, followed by decoration with carboxyl functional MWCNT. The epoxy composite with this modified fiber showed improvements of 64% in IFSS, 37% in ILSS and 29% in tensile strength.	66
epoxy	This study uses sizing of polydopamine coated graphene oxide onto carbon fibers which create a roughened, nano-textured surface. It promotes mechanical interlocking with epoxy matrix and improves ILSS and IFSS values of composite by 23.82% and 16.85% respectively, as compared to control sample.	67
Bio-based epoxy composite	Thiol end-capped biobased hyperbranched polymer (HBP) containing degradable imide bonds was used for surface treatment of carbon fiber fabric. The treated fabric, and a biobased toughener were used for curing bio-based epoxy resin. The composite showed simultaneous improvement with high tensile strength of 869 MPa, high flexural strength of 866 MPa and high ILSS of 53.6 MPa. Further, the degradable bonds allowed recovery of carbon fiber fabric for recycling.	68
Epoxy carbon fiber	This review summarizes recent advances in surface and structural modification of CFRPs to improve interfacial properties using interlocking. Amine treatment of carbon fiber (with polyamine D400) was reportedly found better than acid treatment to improve matrix-fiber interfacial strength by 53%. Adding combination of nanofiller and carbon nanofiber could improve ILSS by 150% and impact strength by 102% in one cross-reference quoted in the review.	69
epoxy	Carbon fibers modified with amine functional POSS nano filler produce a rougher surface on high strength carbon fibers with tensile modulus more than 500 GPa. Characterization showed that surface roughness increased from 10.3 to 18.1 nm, and the O/C ratio increased significantly from 0.060 to 0.232, confirming the successful	70

	surface modification. Furthermore, compressive strength and ILSS improved by 31 and 20%, respectively.	
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2.6 role of Van der Waals interactions in improvement of composite properties

These forces are considered to play small but useful role in adhesion nanofiller filled composites and fiber reinforced composites. Some interesting reports are discussed in Table 5.

Table 5. Using Van der Waal forces for improving properties of composites

System	Key result	Reference
Epoxy-carbon fiber	Using a new plasma surface treatment method for carbon fibers to remove contaminants showed more than 30% increase in interfacial shear strength between carbon fiber and epoxy resin in anhydride cured composite. In addition to presence of useful functional groups formed on the surface of fibers, the better adhesion strength was attributed to enhanced van der Waals bonding after surface cleaning and activation.	71
Epoxy-IPDA-CNT composite	This is simulation work on this epoxy with functionalized CNT (hydroxyl or carboxyl). Compared to control CNT filler without functional group, the composite of carboxylic acid functionalized CNT showed higher degree of crosslinking near the resin-filler interface, as seen from calculated higher value of radial density distribution of oxygen in modified CNT. This inference was also supported by calculation of Radial distribution function (RDF) for N attached to cured epoxy matrix and hydrogen atom attached to oxygen in modified CNT.	72
Epoxy-graphene composite	A home-made Python algorithm was used to establish van der Waals bonds both between graphene layers and between graphene and the epoxy matrix. This allowed realistic simulation of interfacial load transfer. The results demonstrate that increasing the number of graphene layers and volume fraction leads to notable improvements in tensile strength and elastic modulus.	73

2.7 role of traps in improving quality of electrical insulations

Synthetic polymers usually have good electrical insulating properties due to low electrical conductivity. The crosslinked polyethylene (XLPE) is a well-known crosslinked polymer that has been used in high voltage insulation for past many decades. Epoxy or PU based composites are also used for making accessories for high voltage cables. The recent literature on insulation properties of polymers like XLPE, epoxy and PU has been reviewed recently [74]. One useful concept used to explain the good insulation in epoxy and PU under high voltage exposure is evoking the presence of traps in the crosslinked matrix. The formation of the charge in localized states (associated with a certain potential energy level) in the material is named trap[74]. Two types of traps are described commonly. The deeper traps have large difference in potential energy level with respect to conduction band (more than 1 eV) while shallow traps have lower value for this difference ($< 1\text{eV}$) [75]. As power consumption is increasing fast at global level, improving quality of solid insulation, understanding trap levels and their distribution in solid insulation is important to ensure reliable power supply. The topic of improving insulation properties in solid polymers has been recently reviewed [76]. Some more examples are discussed in Table 6.

Table 6. Role of traps in improving dielectric strength of polymer composites for electrical insulation

System	Key results	Reference
epoxy	Zhang et al. prepared alumina filled epoxy composites with varying filler contents and evaluated their insulation performance. At 3% alumina loading, the trap level and trap density reached maximum values, indicating stronger charge trapping capability at the interface. The breakdown strength increased from 27.6 to 35.8 kV/mm. The authors proposed this composite for use in 200–500 kV HVDC high speed mechanical switches.	77
Epoxy	Recent general review related to traps for filler containing insulations, role of fillers in traps formation and charge transport in different insulations like epoxy or cellulose insulating paper used for oil filled transformers.	78
PEI/PAI blend	Uniform dispersion of PAI in the PEI continuous phase created self-assembled nanostructure at the interface between the two polymers. This morphology helped to create deep charge traps, resulting in a substantial reduction of leakage current density by more than one order of magnitude. An optimized blend composition of PEI/PAI 80/20 showed 22.3% higher dielectric strength than PEI control. This makes it possible to design all-polymer dielectric material for energy storage	79
Solid polymers	In the most recent theoretical analysis of trap formation, this paper explains the trap formation due to three-body reaction of two water molecules and one oxygen molecule by a slow process that follows third-order kinetics. It also explains a commonly observed $\sim 3.6\text{ eV}$ trap level seen in many polymers.	80

3. Role of topological factors in improving crosslinked material properties

There are reports on topological factors helping property improvement in crosslinked polymers. Five factors discussed here are crosslinking density, long dangling chains, end groups and loops created during crosslinking (these are considered together) and molecular order of chains in the network.

3.1 Role of Crosslinking density

Crosslinking density is an indicator of how much crosslinking has occurred in the epoxy reaction mass and experimentally, it is estimated from modulus data. The properties of crosslinked material usually require sufficiently high crosslinking density but interference from factors like hydrogen bonding can cause exceptional results as discussed in previous subsections.

We will discuss few reports on epoxy in this paragraph and then about other polymers in the next paragraph. Huang et al reported relationship between the crosslinking density and the thermo-mechanical properties of plant oil-based epoxy resins from tung oil composites. Their results show that the tensile strength, Young's modulus, and glass transition temperature increase with an increase in crosslinking density. The elongation at break and tensile toughness shows nonlinear downward trends as the crosslinking density increases [81]. Zhou et al proposed improved composition for reducing brittleness in epoxy for steel bridge deck application coating, by using epoxy functional hyperbranched polymer (HBP)[82]. Further, study was made on variation in epoxy crosslink density through adjustments in curing agent concentration, incorporation of HBP, and optimization of curing conditions. Employing a multi-objective optimization strategy, this research showed enhanced toughness while minimizing strength degradation. The optimized formulation comprised 45.21% curing agent, a curing temperature of 60.45 °C, and 5.77% HBP content to get optimized tensile strength and 200 % elongation [82]. Li et al reported simulation study on adhesion of ultra-thin polymer films based on epoxy resin networks with four different cross-linking densities on the substrate [83]. It was found that, with increasing the cross-linking density, the polymer sticks to the substrate more tightly. Mechanical properties of interface also improved with film of higher crosslinking density. Frank and Wiggins compared networks formed from DGEBA and DGEBAF resins cured with 3,3' DDS and 4,4' DDS[84]. The mechanical properties and free volume analysis showed that using excess resin reduced crosslink density, but improved chain packing and lowered moisture and solvent uptake. Further, the use of excess resin increased etherification and produced a tightly packed network which was different from the one made for stoichiometric control.

Park et al investigated the use of a crosslinker as an adhesive layer for binding two poly dimethyl siloxane (PDMS) layers to tune the tensile properties of precured PDMS[85]. By modifying the interfacial crosslink density, especially in thin layers, they aimed to tailor the mechanical behavior of the bonded material. The method involved depositing a crosslinker on precured PDMS with low crosslink density, where chain mobility remained high, followed by post curing. This enabled greater chain interdiffusion and higher interfacial crosslinking, resulting in a network of higher crosslink density at the interface and improved tensile performance. Getting good damping in highly crosslinked polymer is difficult. Blending of different thermoset polymers can broaden the damping temperature range; however, this reduces mechanical properties due to inadequate compatibility between the matrix materials. The report of Han et al uses interpenetrating polymer network (IPN) by polyborosiloxane and phenyl silicone rubber (PBS). PBS forms a physically cross-linked network via dynamic B-O bonds, contributing wide damping temperature range [86]. PSR forms a secondary network through covalent cross-linking to provide excellent mechanical properties. This synergistic combination results in interpenetrating network composite with excellent damping and mechanical properties. Tyng et al developed a material for photoelectronic device encapsulation[87]. A series of organic-inorganic polysiloxanes were synthesized using a special end capper to study how crosslink density affects refractive index [87]. Refractive index increases with crosslink density at first and then decreases in the range of 1.410-1.425. The 10% (v/w) end capper containing sample showed the lowest refractive index(selected for application test), while the 40% (v/w) end capped sample shows the highest value (1.4204). X ray analysis also confirms that increased crosslinking densifies the network, especially in crystalline regions. Designing silicone hydrogels revealed that two opposite trends related to properties are seen while making the product by crosslinking[88]. If more free pendant polysiloxane chains are allowed, they are beneficial because they increase oxygen permeability coefficient (Dk) and equilibrium water content (EWC) of the material. On the other hand, increasing crosslinking density reduces amount of pendant groups and leads to decrease in both Dk and EWC values. Hence careful optimization is necessary to obtain suitable products. Verheyen et al tested a high molecular weight silicone rubber with five different peroxide curing agents to analyze the effect of peroxide on crosslinked polymer properties[89]. In general, Higher amount of peroxide crosslinker produces higher crosslink density. Also, properties such as hardness, resilience, stress at 100% elongation increase with crosslink density. However, the Ultimate elongation decreases with increasing crosslink density. Briesenick et al found that hybrid siloxane/silsesquioxane materials with bulky aromatic groups often show incomplete crosslinking, which limits their thermal and mechanical performance. They proposed a new approach of post polymerization crosslinking strategy[90] wherein they placed the phenyl based silsesquioxane crosslinker additive at chain ends, to create crosslinks after film formation. The crosslinked films using new method reached thermal stability up to 453 °C and maintained 99% transparency at 200 °C with minimal yellowing as compared to the control. Dynamic mechanical analysis showed that post crosslinking produced elastic materials with a rubbery plateau up to 200 °C, reflecting improved rigidity due to aromatic substitution. Naphthyl substituted silsesquioxanes post-curing additives can offer further improvements, but their steric hindrance limits the amount that can be used. *Thus, a new crosslinking strategy helps improve properties and allows use of limited amount of bulky naphthyl groups.* More detailed studies on differences in conventional and new methods of crosslinking are desired.

3.2 Role of Dangling chains

Dangling chains in epoxy usually result because of hinderance to formation of longer chains in curing reaction. They are formed because of a different reaction mechanism operating to create secondary reaction via chain transfer or because of deviation from stoichiometric ratio. Presence of dangling chains is known to influence network properties like Tg.

We will discuss the report on epoxy composite followed by reports on other polymer systems. Yamamoto et al studied composites of epoxy- amine (DDM) at excess amine content[91]. They reported that excess amine created pendant groups which help to generate heterogeneity in the network along with low molecular weight epoxy chains. The X ray analysis in combination with simulation work showed that excess amine made the phenyl groups stay closer as compared to configuration seen in sample with stoichiometric composition. This compactness helped to improve tensile strength but exact reasons for this observation were not understood as per the authors since both crosslinking density and Tg reduced as fraction of amine was increased. An IPN between epoxy and PU was proposed to improve damping range in traditional polyurethane elastomers to use them for a broader range of

temperature variation [92]. This work proposes a cooperative molecular motion of “short rigid–long flexible” dangling hybrid segment and the non-freezing Bisphenol A units of aromatic main chain that causes intermolecular friction. Consequently, the damping temperature range gets widened, and the loss factor also increases. Zhao et al simulated the effect of partially replacing Diethyl toluene diamine (DETDA) with n butyl amine (having pendant butyl chain) in a DGEBA–DETDA system [93]. When a small fraction of DETDA was replaced, the pendant butyl groups aggregated, occupied free volume, and locally entangled the rigid matrix. This reduced free volume, brought segments closer, and the pendant group effectively acted like reinforcement. This gave a 25% increase in modulus and a 9% increase in elongation at 2% butyl amine. At higher butyl amine levels, the pendant groups not only filled free volume and instead loosened the matrix.

Wang et al reported bio based aqueous PU systems (WBPU) containing either UPG polyol (without dangling chains) or OPG polyol (with dangling chains) [94]. The PU made with UPG polyol showed higher activation energy and a longer prepolymer reaction time (240 min), indicating lower isocyanate reactivity. On the other hand, dangling chains reduced dispersion stability: OPG based WBPU required 6.5 wt% DMPA solvent (vs. 5.5 wt% for UPG) and formed larger particles. Further, it also showed higher hydrophobicity, extensibility, and lower thermal stability and tensile strength. Jin et al prepared PU elastomers with different pendant group contents in order to improve damping property [95]. Increasing pendant group content reduced tensile strength, while elongation first rose and then declined. Longer pendant chains enhanced hydrogen bond reinforcement. The optimized sample (around 15% pendant groups) showed T_g of -29.3 °C, a 63.9 °C damping range, a 0.91 damping factor, and 407.29% elongation at 2.94 MPa tensile strength. Ye et al found that the presence of elongated dangling chains impedes the agglomeration of rigid regions within PU foams, thereby enhancing properties of the crosslinking structure [96]. The optimized sample exhibited excellent balance of mechanical, thermal insulation, and damping properties, with a compressive strength of 1.34 MPa, and a maximum damping factor of 0.40. This kind of material has potential use in aerospace and transportation industries in terms of structural support, thermal insulation and vibration damping [96]. Yu et al conducted simulation study of different PU polymers to analyze influence of dangling chains on four types of relaxations: β , soft phase α , the hydrogen bond driven I-process, and MWS interfacial polarization [97]. The I-process occurs at a temperature between α and MWS. β relaxation temperature is unaffected by dangling chain length or polarity. Longer dangling chains lower T_g, favor faster segmental motion, suppress the I-process, and shift MWS to higher frequencies. Polar ester groups raise T_g, retard segmental motion, and increase fragility. Higher hard segment content restores the I-process and slows MWS, reflecting hydrogen bond interactions and PU micromorphology. Radi et al reported that for polysiloxane networks, imperfections such as loose or dangling chains lower crosslink density, reduce elastic recovery, and increase swelling [98]. They studied these defects using a model PEG 2000 sol-gel network synthesized by tuning ratios of bis and mono tri ethoxy silyl propyl urethane PEG precursors to introduce pre-defined number of dangling chains. Step crystallization DSC and dynamic light scattering probed network structure and micelle formation, while ²⁹Si NMR, sol fraction, and swelling tests showed that more dangling chains enlarge mesh size and increase water uptake. Yanagi et al used metathesis polymerization route to introduce dangling chains in the graft copolymer of isotactic PP and EPDM [99]. The metathesis at both in-chain double bonds and those in 1,2-butadiene derived pendant vinyl groups generated polyolefin with dangling chains. Partial phase compatibilization gave far better performance than simple i-PP/EPDM blends, increasing tensile absorption energy nearly 50-fold (2.8 to 140 MJ/m³), highlighting their potential as polyolefin elastomers.

3.3 role of end groups and loops in improvement of composite properties

It is interesting that end groups that are usually present in very low weight fractions of the total weight of the crosslinked molecule can influence the properties, similar to that in very high molecular weight linear molecules. The characterization of end groups is usually difficult and more so for crosslinked polymer. Similarly, the loops are considered as part of finer network structure and difficult to analyze [28, 100]. Nevertheless, few reports on simulation work indicate that loops can influence network properties, and some other reports mentioned property improvement with end groups.

We will discuss the reports on epoxy systems first followed by reports on other systems in the next paragraph. Simulation study of Zihan et al. suggests that formation of loops can help to improve mechanical strength in the epoxy-amine system [101]. This simulation study also proposed a parameter called ring efficiency for improving network properties. The report involved comparison of crosslinked networks formed by di functional DGEBA, trifunctional p-amino phenol epoxy resin and tetra functional DDM based epoxy resin. It showed that trifunctional epoxy resin formed network with better network efficiency than tetrafunctional epoxy. DGEBA based network was the last in terms of network efficiency to improve properties. A simulation on general system of two multifunctional monomers ($A_2 + B_3$) was conducted by Wang et al [102]. They used an advanced version of Network disassembly spectrometry (NDS) developed earlier to simultaneously measure the primary and secondary loops in end-linked polymer networks for the first time. Semi-batch monomer addition decreases primary and, to a lesser extent, secondary loops.

Rangamani et al synthesized a series of sequence-defined PU macromers (SD PUM) with different end groups for vanillin-based PU positional isomers [103]. These oligomers were crosslinked to form thermosetting networks using multivalent thiol cross-linkers. The results show that the networks' thermomechanical properties depend strongly on both the regioisomer in the SD PUM backbone (meta vs para connectivity) and the structure of macromer end group. The T_g was found to decrease with increase in the flexibility of end groups and viscosity was influenced by connectivity in regioisomers. Chang et al reported a new route to synthesize low dielectric constant (< 3.0) polyimide (PI) for use in high-speed signal transmission [104]. This work reduces dielectric constant without affecting thermal or mechanical performance by introducing reactive end groups after PI synthesis, enabling controlled crosslinking after film formation. This preserves backbone packing, maintains processability, and allows tunable crosslink density. *The resulting PIs show simultaneous improvements by introducing reactive end groups: lower dielectric constants—2.52 (2.58 for control) at 29 GHz and 2.52 (2.57 for control) at 38 GHz—with a slightly reduced dissipation factor and unchanged thermal stability.*

Mixtures of standard epoxy resin and butylated epoxy resin (CAS number 71033-08-4) are used in various applications in sealants and adhesives. The butylated epoxy resin (BER) is used as flexibilizer. The simulation study was made recently to understand the effects of changing terminal epoxy groups on the formation of network and its properties. The calculations used structural skeletons of DGEBA and BER mixed in different ratios and cured with amine hardener [105]. It was found that topology and thermal, mechanical properties are significantly influenced by this change in resin structure. T_g can decrease in a range of about 0-60K depending upon amount of BER and coefficient of linear expansion increases with higher BER content. A new parameter of confinement index was proposed to indicate extent of loosening of the network that takes place due to addition of BER.

3.4 Role of molecular order in improvement of properties of epoxy crosslinked network

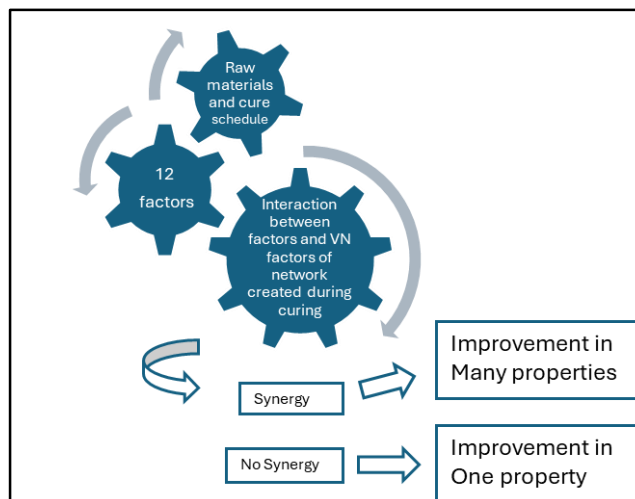
The term molecular order refers to degree of segmental alignment, orientation, or local structural order within the polymer matrix for influencing epoxy material properties in this discussion. Even though epoxies are typically amorphous, they can exhibit short-range order, orientation-induced microdomains, or locally aligned network segments. Such molecular order can help mechanical, thermal properties and thermal/electrical conductivity in epoxy and other composites. An example is impressive thermal conductivity of 0.76 W/(m.K) without using thermally conducting filler in a liquid crystalline epoxy resin which also shows high Tg of 133 °C [106]. Another example of very efficient ordered structure is a network created by reactive brush-coil block copolymer (BP) that reacts with epoxy matrix. The BP reacts using epoxy functional block while allowing the flexible siloxane blocks to form softer core. Further, the additive at just 1 % level gets efficiently distributed in the UV curable 3D epoxy printing ink. The cured 3D material showed simultaneous improvement in modulus as well as impact strength at such a low additive content because of uniform distribution of BP in an orderly manner in the matrix[107]. Table 7 gives more information on the interesting role of molecular order in improving cured epoxy resin properties.

Table 7 Molecular order effect on improving crosslinked epoxy properties

Key results	Reference
Huang et al. developed an epoxy with azomethine bonds that induce molecular order during curing. They found that increasing the curing temperature (120–160 °C) accelerates reaction kinetics, but temperatures above the optimal ~120 °C outpace liquid crystal (LC) formation, reducing crystallinity. The optimally cured resin showed ~14.4% LC content, higher fracture toughness (0.93 kJ/m ²), and improved thermal conductivity (0.32 W/m.K). <i>This shows that molecular order can enhance multiple properties simultaneously.</i>	108
Liquid crystalline epoxy resins containing two types of epoxy groups (those situated at chain end and those that are internal part of the chain) were Synthesized and compared for change in properties after curing. Changing the epoxy group position showed significant effect. Non-terminal epoxy group resins showed a Tg reduction of more than 70 °C compared with terminal analogues, enabling the formation of elastic LC networks.	109
Deng et al modified cured network of epoxy-amine systems by incorporating small amount of maleic anhydride (0.5%) and the double bonds were then used to add bulky aromatic groups that produce deep traps, improve dielectric strength (from 61 to 73 KV/mm), reduce dielectric constant by 30 % and increase tensile strength marginally by 8 %. This confirmed that grafting was successful and modifier groups were well spaced and well dispersed without agglomeration, which also ensured distribution of deep traps in the matrix and helps for better dielectric strength.	110
Two tri-block copolymers [PDMS-PCL-PS] with different contents of polystyrene(PS) were used at 1-10% level for toughening. The surface microstructure was more ordered for composite containing lower PS content. This block copolymer (BP) formed core-shell like structure while the other BP with higher PS formed vesicular microstructure in the cured matrix. The material containing BP with high PS content showed simultaneous improvement in tensile and impact strength at 3.75% BP content, which was relatively more than the other BP that showed only improvement in tensile strength. The crosslinking density and Tg was little than the control in both the cases.	111

4. Miscellaneous reports

So far, we have discussed above the various non-covalent interactions within the groups and atoms present inside the crosslinked matrix. Another type of interaction, the interfacial interaction is also frequently reported. This factor is more relevant for composites that contain nanofillers. The very large surface area of nanofillers can bring about significant improvement in composite properties when they are properly dispersed in the matrix. Many reports discuss improvements in mechanical, adhesive, anti-corrosive, flame retardation properties and synergy effect [112-115]. This is consistent with our proposed approach of including interface as an important parameter to understand epoxy curing process[26]. The discussion in all above sections is summarized in Scheme 2.



Scheme 2 Interactions during curing process of epoxy and consequent effects on product properties

5. Conclusions and future scope

Conclusions are as follows:

- 1) twelve factors- seven non-covalent and five topological- were examined across various epoxy networks. These factors can interact with network features such as crosslink density and free volume, enabling simultaneous improvement in multiple properties.
- 2) many of the factors discussed here are also valid in other crosslinking polymers like PU and polysiloxanes, elastomers etc. This widens the scope of the framework proposed here for describing the epoxy curing process, to other crosslinking reactions.
- 3) Interfacial interactions are also reported between matrix and additives like nano fillers or nano fibers present in the composites and they help to improve properties.
- 4) active role of non-covalent bonds in improving properties of epoxy networks shows that this subtopic is well connected to the broad topic of supramolecular polymer chemistry.

Future scope: The twelve variables discussed here complement and extend scope of our earlier work [26]. Interactions among these twelve variables often lead to simultaneous improvements in multiple properties, but they also require deeper analysis to capture the dynamics of the curing process. In addition, better understanding of characteristics like different types of loops and dangling chains as well as their relations with properties of materials is needed. Long term exposure to external stimuli may alter network characteristics, which warrants detailed study [25]. Many of the twelve variables also influence the curing of other polymers such as PU, polysiloxanes, and cyanate esters, making this a research area for these materials. The new developments and concepts in highly active and broader topic of supramolecular chemistry of polymers can be borrowed to further widen the understanding of how to improve the properties of epoxy and other thermoset resins.

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