



# Mechanical and thermal properties of sepiolite strengthened thermoplastic polymer nanocomposites: A comprehensive review

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**Abstract** Sepiolite ( $\text{Si}_{12}\text{Mg}_8\text{O}_{30}(\text{OH},\text{F})_4 \cdot (\text{H}_2\text{O})_4 \cdot 8\text{H}_2\text{O}$ ) is a valuable filler with an enormous capacity to be used in thermoplastic composites, substituting costly reinforcing fillers, such as graphene and CNTs. Sepiolite strengthened polymers nanocomposite materials have encouraged the field of research and ventures because of their strengthening ability and bio-compatibility in polymer composites. Sepiolite shows remarkable characteristics over various fillers due to its higher specific surface area and channel type structure. Numerous investigations were performed to decide different properties of Sepiolite strengthened polymer composites in various applications, for example, tensile strength, flexural strength, impact strength, stiffness, thermal, flammability, thermo-mechanical, and morphological. This review paper focuses on the mechanical and thermal properties of sepiolite strengthened polymer nanocomposites. Generally, it can be determined that the properties of sepiolite loaded thermoplastic polymer composites mainly depend on filler content, matrix, bond interaction, shape, size of sepiolite particles. Further assessment and development are required to expand its utilization in several applications. These comprise the utilization of nano-size sepiolite made synthetically as functionalized filler in thermoplastics.

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## 1. Introduction

Polymer composite is the materials that consist of two or more than two phases of materials, generally consisting of the polymer matrix phase and strengthen or fillers phase. The arrangement improved the properties as compared to the particular component. In general, thermoplastics are mostly preferred to thermosets because they can be easily recycled and thermoformable. Traditional fillers such as glass, aramid, and carbon fibres [1-4], calcium carbonate, and so on [5-8] have encouraging influence in making more substantial and tougher but lightweight materials. Nevertheless, sepiolite is gradually building up an option as compared to these strengthen phase or fillers. Sepiolite exhibits low thermal expansion due to which giving an advantage of better dimensional stability and less shrinkage [9]. Naturally, sepiolite nanoparticles have high surface area morphology, with the dimensions: 0.20–4.00  $\mu\text{m}$  length, 10–30 nm width, 5–10 nm thickness. The shape accepts the penetration of polymeric chains in the internal space channels of sepiolite [9]. It likewise gave better hardness, flexural, tensile, and impact properties contrasted with powder fortified polymer composites [10,11]. The positively charged organic molecules may be attracted by the silanol groups (Si-OH) and negatively charged surface of sepiolite [12]. The coupling capability of sepiolite with the non-polar polymer may be influenced. This problem is frequently overwhelmed with surface modification of sepiolite fillers. Sepiolite was functionalized with amino-silane, epoxy-silane, mercapto-silane grafted molecules, and diamine adsorption by Tartaglione et al. and Peinado et al. [9,13]. Their results indicated that the sepiolite's functionalizing through organic modifier is mainly constrained by the peripheral surface. In particular, the sepiolite filler's hydrophilic nature, particularly organic sepiolite acquired by the adsorption process, is more hydrophobic in contrast to the grafted sepiolite. Ammonium and silane salts have been portrayed to support improved compatibility with the introduced polymer.

Several theoretical models by various researchers have been introduced to improve nanocomposites' properties and to develop relationships between the size, shape and distribution of filler and its agglomerations in polymers. For example, Micromechanical, 2D and 3D modelling of thermal properties for various fillers have been performed by Hassanzadeh et al. [14]. They evaluated the CNTs randomly oriented wavy and

straight distribution of CNTs and compared with experimental data. Also, a series of micromechanical simulations were done to evaluate the influences of volume fraction and diameter of CNTs, temperature and interfacial thermal resistance on the overall thermal behaviour of CNT-polymer nanocomposites. Several worthy conclusion has been reported and may be applied for the sepiolite based nanocomposites. The sepiolite case was utilized as a reinforcing filler in equally micro and nano-sized [15]. The effect of nanoparticle conglomeration on the properties of nanocomposites was investigated by Haghgoo et al. [16]. Several theoretical models and calculations were done in detail to evaluate the various properties, especially electrical, in their research work. In their work, by embracing a two-venture insightful model, the impact of multi-scale fillers on the electrical conductivity and resistivity of CB/CF-built up, polymer multi-scale nanocomposites were researched. The impacts of fibre direction, electrical properties, perspective proportion and polymer likely obstruction on the electrical conductivity of multi-scale nanocomposites were researched. The insightful model outcomes showed great concurrence with current test information and exhibited the ability of the model to measure the impact of multi-scale fillers. The enhancements are associated well with the progressions that multi-scale filler made in the microstructure of the polymer-based composites. The outcomes also demonstrated that the presentation of multi-scale fillers in the polymer-based composite microstructure brought down the percolation limit and improved the conductivity up to significant magnitudes. The effect of nanoclay agglomeration on the creep behavior of polymer nanocomposites was carried out by Hassanzadeh et al. [17]. Mainly silica-based nanoparticle aggregation, specifically on creep behaviour analysis, was performed. A hierarchical micromechanical model established on the Mori-Tanaka (M-T) system was intended. Development of interphase area due to the interfacial contact between the polymer matrix and nanoparticle is featured in the modeling. The contrast between the suggested model results contemplating the viscoelastic interphase indicates a great deal with existing experimentation. When the nano-particles are not well-distributed into the polymer nanocomposites at the high-volume portion, it is essential to ponder the viscoelastic interphase collectively with the nanoparticle accumulation for providing precise esti-

mates. The results disclose that the nanoparticle cluster affects and reduces the polymer nanocomposite creep resistance. An identical dispersal of nanoparticles into the matrix indicates the highest level of nanocomposite creep resistance. The nanoparticle amount fraction, diameter, and interphase qualities on the nanocomposite creep behavior were investigated. Currently, nano-sized fillers have become prevalent as they can improve polymer properties without limiting their processability. Generally, employed nanofillers are carbon nanotubes [18], graphene [19-21], and montmorillonite (MMT) [22,23]. Among the various clays suitable for the fabrication of nanocomposite, sepiolite would be indeed a low-cost nanofiller. Sepiolite is a magnesium phyllosilicate having a formula as  $[\text{Si}_{12}\text{Mg}_8\text{O}_{30}(\text{OH})_4](\text{H}_2\text{O})_4 \cdot 8\text{H}_2\text{O}$ . Sepiolite has a layer of magnesium ions with octahedral coordination and two layers of silica in a tetrahedron. Likewise, in the different phyllosilicates, the octahedron sheets are not constant in sepiolite: Sepiolite presents limitless channels with a cross-section of  $1 \times 0.4 \text{ nm}^2$  in sizes [24-26]. These channels' existence strongly enhances the specific surface area of sepiolite, up to 200–300  $\text{m}^2/\text{g}$ . The vast majority of the previous exploration investigated macro size sepiolite utilization as useful filler in thermoplastics rather than nano size sepiolite.

Polymer nanocomposites are the materials carrying natural or artificial fillers with a homogeneous dispersal, usually ranging from 10 to 100 nm in size at least one dimension. It may be mixed physically or chemically through a process of polymerizing [27]. The fillers can be added in the form of particles, continuous layered materials, filaments, or maybe bunches encompassed in a wide-ranging assortment of synthetic and polymers. Composites deliver outstanding properties due to various fillers' utilization, rebound polymers nanocomposites in a high-level-ranking prospective for aerospace, automotive production, electronic engineering, medical and health apparatus end-user goods. Numerous kinds of matrixes, for example, Polyethylene terephthalate (PET) [28], high-density polyethylene (HDPE) [29], polystyrene (PS) [30], polycarbonate (PC) [31], polypropylene (PP), polyamide 6 (PA6) [32-34] have been studied and established by a variety of researchers to enhance the functioning of the polymer-based nanocomposites. In the same way as fillers like MMT, graphene, carbon nanotubes (CNTs), nano clay, natural fillers, sepiolite, silicon dioxide ( $\text{SiO}_2$ ), Titanium dioxide ( $\text{TiO}_2$ ) are inserted in polymers matrixes.

There are very few review papers authored on sepiolite, which focuses on numerous aspects. Bassett et al. [35] described a short review regarding thermodynamics equilibrium constants for kaolinite and sepiolite in terms of Gib's free energy. A review for the case of environmental contamination and analysis of producing sepiolite-based adsorption for removing potentially toxic elements from the water was performed by Wang et al. [36]. Another review paper of Zaini et al. [37] reported a short review of the sepiolite-filled nanocomposites; however, their focus was on the structure of sepiolite, preparation, and application. In contrast, our study focuses on the mechanical and thermal properties of sepiolite strengthened nanocomposites. Muller et al. [38] reported a review of polymer nanocomposites' properties and processing, especially nanocoating. However, they mainly cover various fillers such as single-walled carbon nanotubes (SWCNT), multiwalled carbon nanotubes (MWCNT),

MMTs, with a touch of sepiolite nanocomposites. Their work focus on various properties such as barrier, electrical/electronics, flammability resistance, and microwave absorbing properties and insight into some safety and regulatory aspects. Tian et al. [12] reported the sepiolite nanomaterials structure and its physiochemical properties and functional applications. However, there is no comprehensive review in the development of sepiolite filler filled thermoplastics polymers nanocomposites and their mechanical and thermal properties. This paper is intended to cover sepiolite filler filled thermoplastic nanocomposites in terms of mechanical and thermal properties besides their exploration in various applications.

## 2. Fabrication of polymer clay nanocomposites

### 1. Processing techniques

Several techniques for the fabrication of clay nanofiller and polymers, such as sepiolite-based nanocomposites, were developed, such as melt blending, in situ polymerization and solution blending [39]. The preparation of clay nanocomposites consists of few steps that start with either organoclay or pure clay. Pavlidou et al. [40] researched that melt blending and in situ polymerization techniques are more vital in manufacturing clay nanocomposites. However, Ma et al. [40] claimed that these techniques require a massive amount of organic solvent or high viscosity or thermally unstable polymers at high temperature. Each method exhibits its importance and limitations.

### 2. Melt blending technique

In the melt blending technique, clays are directly mixed in the molten form of the polymer through an internal mixer or twin-screw extruder. This is the widely used method for manufacturing sepiolite nanocomposites [41-48]. Among those, Orozco et al. [48] prepared sepiolite based PP nanocomposites and compared them with carbon nanotubes that were modified and non-modified. Likewise, Xie et al. [49] applied a simple melt blending technique to manufacture sepiolite/PA6 nanocomposites. The nanocomposites were fitted in the twin-screw extruder having a speed of 200 RPM at 210 to 230 °C from the hopper to the die. The confirmation of residence time and high shear stress is essential to exfoliate and disperse individual nanoparticles throughout the polymer matrix successfully.

### 3. Solution blending technique

The solution blending technique includes altogether scattering the layered silicate inside proper solvents, which incorporates the polymer. When the reaction is finished, the solvent would have evaporated, catching the polymer chains intercalated into the interlayers of the clay. Turhan et al. [50] used the solution blending technique to get ready sepiolite nanocomposites. They led a progression of investigations with various concentrations of sepiolite specimens in tetrahydrofuran (THF) solutions. Mohd Zaini et al. [37] reported that this technique is reliable, whereby the silicate can be consistently dispersed at the nano-level within the polymer matrix following by solvent removal.

#### 4. In Situ polymerization technique

This method includes engorging the altered layered silicate utilizing a liquefied monomer or a monomer solution. Polymerization happens among the interpolated sheets when the monomer goes into the galleries of the layered silicate. Heat or radiation can be utilized reliant on the pre-intercalated initiator or catalyst. In situ polymerization is the highly appropriate technique for thermoset-based nanocomposites, which cannot be produced utilizing the melt blending technique [51]. However, the drawbacks of this method may comprise: time-consuming, such as polymerization reaction may take more than 24 h; thermodynamically unstable exfoliation and re-aggregation of the platelets may arise through subsequent treating; this procedure is feasible for resin constructors who can obligate a production line for this persistence. For example, Lu et al. [52] analyzed the modified sepiolite/unsaturated polyester nanocomposites by in situ polymerization. Sepiolite was organo-modified by attaching vinyl-tri-epoxy, holding a double bond on to the surfaces and was utilized as nanofillers. Polyamide 6/sepiolite nanocomposites were made via in situ polymerization by Bilotti et al. [53] This method was also used by Chen et al. [54,55] upon the formulation of sepiolite reinforced polyurethane nanocomposites.

#### 3. Overview and applications of sepiolite

Most polymeric materials are discovered lower in properties when contrasted with metals. Their mechanical and some other properties can be suitably improved at a decently negligible expense by strengthening with nano-size firm filler particles [39,56]. With this philosophy, a polymer's modulus and strength can be adequately enhanced while keeping up its flexibility, low density, and simplicity of handling. It tends to be developed even at commonly low nanofiller content. It has been demonstrated that the aggregation of a small number of picked nanofillers to polymers can improve their mechanical properties [51,57-59], thermal stability, and resistance against chemicals [60]. The prime focus on nanofillers' development to polymers is to improve their mechanical properties [58]. One of the key prerequisites is then fulfilling the equilibrium among the stiffness/strength and toughness as much as possible [61,62].

Over late many years, two or three specific kinds of nanofillers, for instance, nanoparticles, clay, carbon nanofibers (CNFs) [19], carbon nanotubes (CNTs) [63,64], graphene

[65], SiO<sub>2</sub> [66], TiO<sub>2</sub> [67], sepiolite [68], halloysite nanotubes (HNTs) [69], etc. were applied for the improvement of polymer nanocomposites and opened potential applications in the car, avionics, and electronics enterprises [70,71].

Sepiolite is a microcrystalline hydrated magnesium silicate with Si<sub>12</sub>Mg<sub>8</sub>O<sub>30</sub>(OH,F)<sub>4</sub>·(H<sub>2</sub>O)<sub>4</sub>·8H<sub>2</sub>O unit formula. Sepiolite shows unique characteristics over various types of fillers. It has a unique needle-like sinewy structure that liberates when dispersed in water or other polar dissolvable, outlining a monomer fibre or miniature fibre gatherings. Sepiolite moreover has a channel like a micropore channel structure. It uncovers a turn of squares and entries that create toward the fibre. Every essential square is made out of two tetrahedral silica sheets sandwiching a focal sheet of magnesium oxide-hydroxide. Such high-class physiognomies are fit for tough adsorption and high specific surface area [68]. Fig. 1 speaks to the structure of sepiolite fillers along the a-axis and c-axis [72].

Among clays, sepiolite has allured scientist interest since it is effectively accessible, low in cost, high explicit surface area, strong chemical and mechanical stability, and anisotropic molecular shape [73]. The sepiolite essential molecule arises as small fibres 40–150 nm in width and 1–10 μm long. Fibres ordinarily stick all in packs offering 0.1 to 1 μm in diameter. The bundles produce haphazardly arranged aggregates having sizes fluctuating 10–100 μm. The separating of these almost unbendingly pressed aggregates is as yet a challenge for researchers to benefit from the specific sepiolite shape for planning nanostructured materials.

A few essential investigations have been done in Sepiolite/nanocomposites to well control the sepiolite distribution [74,75], and different functionalization therapies of sepiolite have been accounted for to define nanocomposites [12,13,36,76-78]. These treatments/therapies accept the positive effect of sepiolite functionalization on improving the nanofiber dispersion in the polymer matrix and the nanocomposites' useful properties. Generally, interfacial bonding between the polymer and the fillers is vital to be gotten through fillers' surface treatment. Di Credico et al. [75] explained the cited properties and the structural properties of the sepiolite and its hybrid composites. They have detailed that the size and shape of the filler significantly affect the reinforcing of composites and interfacial interaction between filler and polymer. Acidic and salinization treatment was applied in their research work for different loading of fillers in polymers. They tried to develop

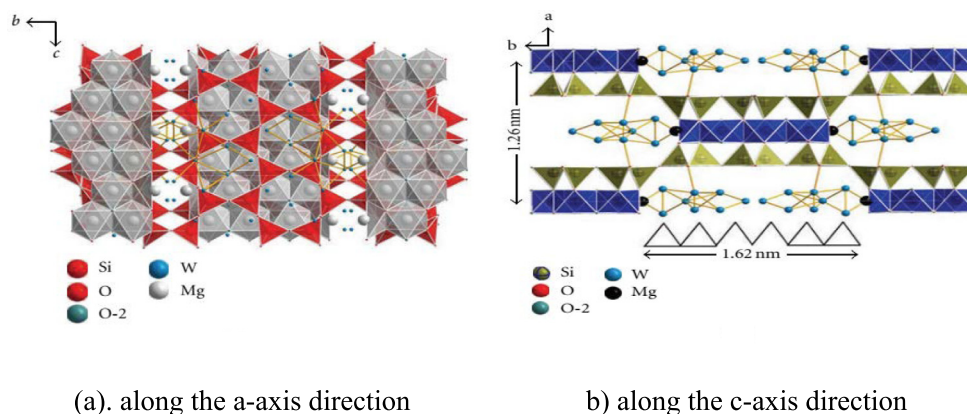


Fig. 1 Structure of Sepiolite [72].



the strong interfacial bonding between sepiolite and its polymer to form a composite and its hybrid nanocomposites. Some of the structural properties of sepiolite are listed in Table 1.

The bonding between the polymer matrix and the strengthening phase is answerable for improving mechanical and thermal properties by confining polymer chains' development, for example, the tensile strength and strain, flexural strength, and strain%, impact strength of polymer composites. The follow-

ing section will discuss various studies related to the mechanical and thermal properties of polymer composites.

Nehra et al. [82] used sepiolite as a strengthening filler to make poly(lactic acid) (PLA)/(styrene-ethylene-butylene-styrene)-g-maleic anhydride copolymer/sepiolite (SEBS-g-MA/Sep) 90/10 (w/w) nanocomposite. It was seen that the tensile modulus upgraded almost 36.0% and 17.0% with the incorporation of 0.5 and 2.5 wt% of sepiolite, correspondingly. Huang et al. [83] developed chitosan (CS)/polyvinyl alcohol (PVA)/sepiolite nanocomposite films by a solution casting technique. They validated that the ascent in sepiolite inclusion strongly improved the mechanical properties of nanocomposite films. Yu et al. [25] arranged polyimide/sepiolite nanocomposite films through an in-situ polymerization measure, including the dispersion of sepiolite. Discoveries uncovered that the addition of 16 wt% of sepiolite improved the tensile strength and modulus up to 41% and 94%, respectively.

Epoxidized natural rubber (ENR) with sepiolite loading has been analyzed by Hayeemasae et al. [68]. The effect of sepiolite consolidation at 100% strain (M100) and at 300% strain (M300) of sepiolite filled ENR composites is addressed in Table 2. It reveals that M100 and M300 both improved with the ascent of sepiolite filling. As more sepiolite particles are incorporated into the ENR, the strain% is decreased, getting step by step more rigid, stiffer, and harder vulcanizates. Such astonishment is truly a direct result of the effect of strain development. Thermal strength for ENR was additionally tried by these specialists recorded in Table 3.

Mechanical, thermal, thermo-mechanical, and morphological properties of sepiolite filled in PA11 nanocomposite created by two unique procedures, i.e., melting compounding and in situ polymerization, were newly inspected by Herrero et al. [84]. Mechanical properties examined through the diverse filling of sepiolite and created through melt compounding (EX) and in situ polymerization (IS) technique has been meant in Fig. 2. It has been evident from all the graphs that with the

**Table 1** Structural properties and applications of sepiolite [79-81].

Properties	Applications	Remarks
Acicular shape (needle-like)	Foundry industry Ceramic industry	It improves mechanical strength Work as an anti-cracking agent
White colour Brown colour (3% carbon content)	Coating, paint Cosmetic Fluid fertilizer Drilling fluid	Used as a thickening agent, it impedes the sagging of paints, reduces the amount of pigment required Serves as a clarifying agent. Not easily flocculated due to its particle shape. Sepiolite mud viscosity, together with its gel strength, is not affected by the change in electrolyte content.
Specific surface area (200–450 m <sup>2</sup> /g)	Polymer composites	Support good mechanical and thermal resistance. Improves processing.
Excellent thermal stability	Polymer composites	Enhance Mechanical, thermal and flammability properties
High viscosity	Construction	-Reduces friction between the building structure and soil.
Non-toxic	Medical industry	Use for slow-release systems such as tablet manufacture.
High porosity	Medical industry	To absorb bacteria and toxins
Good flame retardancy	Foam industry	Use as fire extinguishing powder, thereby preventing fire spread.
High absorption (water:75–120%, Oil: 60–95%) and adsorption capacity	Environment Floor sweep compound, Tobacco Industry	Water and wastewater purification -Absorbing oil and grease spills.
Moderate hardness (2–2.5 Mohs)	Construction	Used as cigarette filters Processing aid used in the production of fibre-reinforced cement composites
Good plastering ability and plasticity	Road construction	An essential additive in bitumen emulsions.
Low cation exchange capacity (20–30 mEq/100)	Pharmaceutical and Chemical industry	Absorb and adsorb drugs, pesticides, herbicides with less active ingredients.

**Table 2** Tensile modulus of sepiolite filler filled ENR composite [68].

Sepiolite content (phr)	Tensile Modulus (MPa) (M100)	Tensile Modulus (MPa) (M300)
0	0.85	2.03
1	0.86	2.12
2.5	0.94	2.35
5	1.05	2.45
7.5	1.07	2.54
10	1.15	2.65

**Table 3** Thermal parameters of sepiolite loading ENR composites [68].

Sepiolite content (phr)	Temperature at 10% weight loss (°C)	Temperature at 50% weight loss (°C)
0	350.2	401.1
2.5	351.1	402.4
5	353.9	403.8
10	369.3	4.1

ascend of sepiolite in PA11 mechanical properties have been improved alongside huge ductility up to 8 wt% of sepiolite

Sepiolite can be utilized as an ideal carrier of catalyst, decolourize vegetable oils, as an animal feeding additives, antibacterial materials, biological and biomedicines, energy and anode material, thermal energy storage material, electronic conductors, sensing material, ceramic and cement material, strong functional construction materials, flame retardant and thermal insulation material, hybrid pigments, colloidal agents, papers, storage and slow proclimation of nitric oxides [12].

#### 4. Properties of thermoplastic polymer/sepiolite composites

Mechanical, thermal, and thermo-mechanical properties of the thermoplastic filled sepiolite nanocomposites may be influenced by various reasons, for example, the interfacial interaction between matrix and fillers, fillers concentration, dispersion, the capability of filler to nucleate, the filler reinforcing ability, and the efficiency of loading transfer between the filler and polymer matrix [85].

##### 4.1. Polypropylene/Sepiolite composites

Polypropylene (PP) is one of the most routinely used polymers today in light of its ease and adjusted properties. Mechanical, thermal, and thermo-mechanical properties were studied by various Sepiolite researchers embedded in the PP matrix, and PP blended with another polymer matrix. Several attributes

of sepiolite reinforced PP composites' properties were explored, for example, impacts of various filling, surface treatments, shapes, and sepiolite sizes. Generally, the integration of sepiolite has been demonstrated to improve the mechanical and thermal properties of the PP composites [45,86,87]. It has been seen that the functionalization method does perhaps the covalent bonding between the hydroxyl groups of earth minerals and polymer lattice checked by the increment of mechanical properties [87]. Sepiolite has a specific nucleating impact on the PP polymer matrix.

Besides, in PP-composites, Sepiolite creates a solid orientation of polymer crystallites. A direct relationship is seen among nucleation and orientation, which can be used as a straightforward tool to assess sepiolite dispersal. This limitation can be consequently used to describe the effectiveness of sepiolite on nucleation and, eventually, on sepiolite dispersion/adhesion in the matrix of the polymer. Simultaneously, a considerable improvement of the term oxidative conduct is brought about by the presence of sepiolite [45].

Wang et al. [88] researched the impact of organo-sepiolite (O-Sep) on the mechanical, morphological, and thermal properties of PP/ABS polymer blend. Their rheological results demonstrated that organo-sepiolite increase the viscosity, storage and loss modulus of polymer blends. Simultaneously the mechanical testing uncovers that O-Sep (at suitable content) improved the impact strength, tensile and flexural modulus of PP/ABS/O-Sep polymer blends. The results likewise demonstrate that PP's toughness can be enhanced by incorporating O-sep and ABS PP. This work presents another valuable technique to develop the PP/ABS polymer blends by combin-

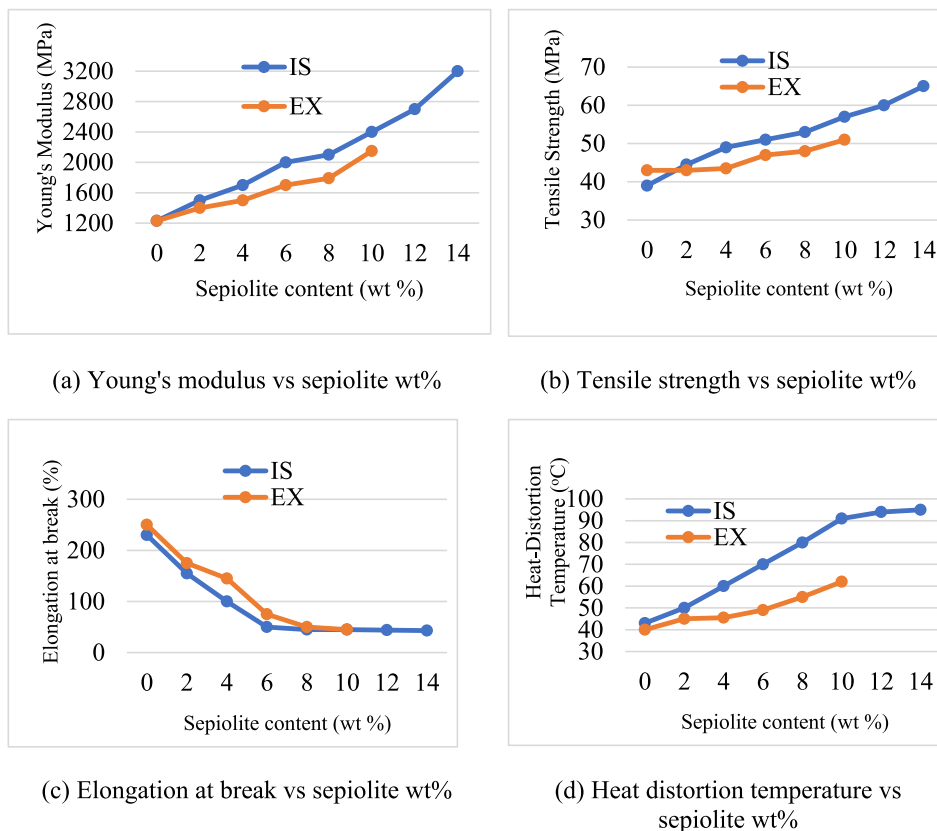


Fig. 2 Mechanical properties vs sepiolite content wt% for various proportion [84].

ing O-Sep as a compatibilizer. The nucleating agent may offer a way for PP's toughness.

Fouad et al. [89] researched enhancing PP's flammability with joint action of PA6 and Sepiolite. The characterization techniques utilized were loss mass calorimetry and thermogravimetric analysis (TGA). As per their TGA results, the utilization of maleic anhydride joined polypropylene (MA-g-PP) as a compatibilizer raced to a significant expansion in thermal stability noticeable in the air. Such development was related to the arrangement of a char layer, avoiding the PP's degradation thermo-oxidative. Excitingly, this char layer's thermal resistance was enhanced by adding 5 wt% of sepiolite, prompting a critical increment of time to ignition and decrease of heat discharge rate up to 53% during mass calorimeter trial of PP/PA6 polymer blend. It happened out because PA6 nodules, dispersion rate, and size were the key-boundaries that allowed improved thermal resistance to form a durable char layer. The adequacy of the char layer created during the ignition was seen to rely upon the amount of both Sepiolite and PA6. In compatibilized polymer blends, the presence of appropriately distribution of PA6 allowed for achieving cohesive charring because of the fact of interlinked 'islands-in-the-sea' carbonaceous areas. The sepiolite nanoparticles improved the thermal resistance with good flame retardant properties. Since then, polymer blending has permitted improving PP's fire properties over a careful selection of the nanoparticles/compatibilizer pair. The idea leads to more commonly employed polymers as a modern economical route for thermoplastic fabrication having flame retardancy properties.

Pappalardo et al. [90] reported that PP comprising of sepiolite nanoparticles was previously proved that the combination of 0.5 wt% of O-Sep permits the acquisition of remarkable fire properties. While in their research, it was established that such variety also confirms the mechanical properties enhancement. Bilotti et al. [91] studied the effect of sepiolite on its mechanical properties, dispersion, and crystallinity in polymer nanocomposites. Manchanda et al. [92] researched the sepiolite/PP nanocomposites and their influence on morphological, mechanical and thermo-mechanical properties. Their research presented the influence of natural sepiolite nanofibers and PP's properties in the lack/existence of compatibilizers includes (maleic anhydride grafted-polypropylene (PP-g-MA)). The twin-screw extruder was utilized for the manufacturing of composites. The morphology and scattering of Sepiolite in PP composites portrayed using SEM and transmission electron microscopy (TEM) uncovered that the dispersal of Sepiolite in PP blend was prevalent in the presence of PP-g-MA. TGA was used to look at the Influence of Sepiolite on PP's thermal properties in the lack of PP-g-MA. Mechanical properties assessed according to ASTM systems exhibited a ~ 65% increase in flexural modulus, 60.0%-70.0% ascent in tensile modulus and 10.0%-20.0% upsurge in impact strength on adding of 10.0 wt% sepiolite in PP. The compatibilizer consideration has also improved the flexural modulus close to 85.0% as related to neat PP.

Nanocomposite consisting of Sepiolite and PP/PLA were studied by Prera et al. [93]. Morales et al. [94] researched the PP/sepiolite DMA properties established on sepiolite surface treatment. Polymer nanocomposites involved in organically modified sepiolite nanoparticles and their dispersion was explored by Hirayama et al. [95]. Maria et al. [96] analysed the isostatic polypropylene (iPP) based sepiolite nanocompos-

ites and their copolymers. The impact of the amount of comonomer, the quantity of filler, and the kind of branching picked up on the final properties of iPP nanocomposites were examined.

All of the above investigations, clearly evident that sepiolite and its functionalization have greatly affected and enhanced the mechanical, thermal, and thermo-mechanical properties of PP and PP blended with various polymers depending upon the dispersion, size, and compatibility between the matrix and filler. Also, the choice of compatibilizer and matrix pair impacts the properties of thermoplastic-based polymer nanocomposites.

#### 4.2. Polyethylene/sepiolite composites

Polyethylene (PE) lies in the class of recyclable semicrystalline thermoplastic having great formability and low density, that can be categorized upon density, for example, low-density polyethylene (LDPE), ultra-high molecular weight polyethylene (UHMWPE), medium-density polyethylene (MDPE) and high-density polyethylene (HDPE). The sepiolite incorporation usually improves the modulus and ultimate tensile strength at the cost of strain% of PE composites [44]. Storage and loss modulus were reported higher, owing to enhanced restraintment in polymer chains by sepiolite in nanocomposites without compatibilizers compared to compatibilizer (PE-g-MA) utilizing ten wt% of sepiolite filler [97]. The increase of 26% in LDPE storage modulus was reported by Carrero et al. [98] using silica as a catalyst having 2.8 wt% sepiolite's filler.

Singh et al. [99] contemplated the thermal and mechanical properties of HDPE/sepiolite nanocomposite in the presence/absence of (HDPE-g-MA) compatibilizers. According to their investigation, Sepiolite content extensively improved the mechanical properties, viscosity, and melt strength with a minor diminishing in drawability at ten wt% of sepiolite content. For example, a 40% and 50% surge in tensile and flexural modulus was achieved at ten wt% sepiolite incorporation. Thermal stability is improved in the presence of sepiolite. Thermal and mechanical properties further advancement was accounted for the addition of compatibilizer. However, a high-level molecular weight compatibilizer usage showed better results than a low-level molecular weight compatibilizer. The lower molecular weight compatibilizer has a more prominent grafted maleic anhydride level, diminishing the chain length and average molecular weight of the HDPE matrix that indicates lower mechanical properties.

Samper et al. [100] analyzed the impact of compatibilization on polyethylene-thermoplastic starch blend in the presence of sepiolite for environment-friendly films and packaging applications. As per their investigation, sepiolite improved mechanical properties. However, silane-treated sepiolite is poor than that found with traditional copolymers such as PE-g-MA. It is imperative to mention that sepiolite is easily available, lower in low cost, and a biocompatible additive that can be altered to customized functionalities. It might denote a fascinating subject in the packing industry.

Guastavino et al. [101] researched the enhancement of insulation for LDPE nanocomposites electrical properties by utilizing various nanofillers. The result demonstrates that the dispersal of a mixture of nanoparticles with distinct geometry

and morphology, Sepiolite and MMT, could be an exciting method to improve the electrical properties of insulation in polymers.

Zhang et al. [102] performed research on LDPE/sepiolite nanocomposite in the absence/presence of modified sepiolite by joining poly-pentaerythritol diphosphonate dichloride-hexamethylenediamine (PSPHD). As per them, a fantastic interfacial modification result has been accomplished between PSPHD/sepiolite and LDPE polymer matrix. A particular increase in tensile strength has appeared in tensile testing. DMA demonstrates that PSPHD-sepiolite/LDPE nanocomposites storage modulus is impressively better than that of perfect LDPE and sepiolite/LDPE structures. Mir et al. [103] performed research on the Influence of Sepiolite upon thermal and rheological properties of LDPE/starch blend. Results demonstrated that the most remarkable tensile properties were seen in composites involving 2 phr sepiolite and 15 phr starch. It has been accounted for that silane has upgraded the dispersion of filler and interaction among the polymer and filler interphase. Besides, the outcomes suggest that sepiolite acts as a proficient filler to improve the thermal and rheological properties. Garcia et al. [104] did an investigation on different fillers geometry and its impact on the LDPE matrix thermal properties

The effect of ethylene glycol dimethacrylate (EGDMA) as a cross-linking mediator in LDPE/sepiolite was examined by Ghazali et al. [105]. It has been stated that added EGDMA to the sample slightly enhances the tensile strength (9% rise) and Young's modulus in a co-agent presence. At the same time, a reduction in thermal constancy was observed in LDPE/sepiolite nanocomposites. Garofalo et al. [106] discussed the morphological, rheological, thermal, and mechanical properties of PE and PP-based recycled plastic by utilizing sepiolite and various fillers. The flame retardant modified sepiolite nanofibre in LDPE has also been studied by Cheng et al. [107]. The impact of nanofiller's drawing and content upon mechanical and morphological properties in polyethylene and sepiolite nanocomposites was studied by Alonso et al. and Khan et al. [108-110].

#### 4.3. Polyamide/Sepiolite composites

Amongst all kinds of polyamide, PA6 has been utilized for sepiolite based composites highly. PA6 has broadly employed thermoplastics in various industries such as electrical, automobile, and electronic, owing to the excellent mechanical properties. Nevertheless, the crack sensitivity, dimensional instability, and high moisture sorption reduced structural parts applications. Sepiolite inclusion had been introduced into the polymer matrix to address these drawbacks. [42]. Furthermore, the sepiolite surface may be modified to improve its interaction with the chains of PA6 to improve the composite properties compared to the sepiolite of the unmodified surface. Typically, the incorporation of sepiolite in the PA6 and PA11 matrix enhances the mechanical, predominantly tensile and flexural strength and improves thermal properties [111-113].

Sepiolite (2 to 10 phr) was incorporated in PA6/ethylene vinyl acetate (EVA) blended polymer at the ratio of 80/20 to improve the mechanical properties of the blend [112]. The increase in modulus and flexural strength was reported up to 6 phr incorporation of sepiolite with a slight decrease in impact

strength. The optimum value has been reported at 4 phr of sepiolite, while the explanation for the reduction in impact strength was described due to lower interfacial adhesion or large particle size. Various alteration of Sepiolite for PA6 composites has been introduced by García-López et al. [46].

Sepiolite base poly(amide-imide) nanocomposite preparation and properties were studied by Aida et al. [111]. According to their results, the best thermal and mechanical properties were at 3 phr of sepiolite due to good homogeneous dispersion, while increasing sepiolite beyond this limit leads to agglomeration. At 3 phr of sepiolite, the tensile modulus was described to rise from 2476 to 3001 MPa, while a slight reduction in elongation at break and tensile strength has been informed.

Walter et al. [114] described a stiffening mechanism in amorphous polyamide bio-nanocomposites utilizing various nanofillers such as sepiolite and different clays of montmorillonite. According to Walter et al.'s investigation, it is hypothesized that dynamic network formation via polymer-filler hydrogen bonding interactions offers a tertiary stiffening mechanism that works in the rubbery plateau region, i.e., at a temperature above  $T_g$ . The increase in thermal and mechanical properties in polyamide with sepiolite filler has also been reported in Saeed et al.'s research work [115]. Sepiolite-based PA6 nanocomposites thermal stability has been explained by Cristina et al. [78] owing to the establishment of hydrogen bonding between (C-H-O) sepiolite and the polyamide chains. The impact of sepiolite orientation and concentration on morphological, mechanical, and thermal properties of bio-based polyamide 4.10 has been studied by maria et al. [116] for structural application in automotive industries. Hence, it proves the improvement in thermal and mechanical properties with sepiolite incorporation in the polyamide matrix and its blends.

Polyamide 11 (PA11) is a polyamide, bioplastic, and apart/member from the nylon group of polymers made by 11-amino undecanoic acid polymerization. It is made from castor beans by Arkema under the trade name Rilsan. PA11 is associated with oil and gas, aeronautics, car, material industry, electronics, and equipment of sports, frequently in the tubing, wire sheathing, and metal coating [117]. Focal points of PA11 incorporate low water ingestion in nylon group, significant chemical resistance, and the capacity to acknowledge high filling of fillers. Impediments of PA11 are a considerable expense when contrasted with different nylons and slight heat resistance.

Before, much consideration has been given to polymer nanocomposites [118-121]. The primary fruitful investigation of PA6 has been grounded in writing. As far as anyone is concerned, considerably less thought has been given to PA11 and its nanocomposites. PA11 ordinarily more flexible and has better dimensional consistency and preferred impact properties over regular short-chain polyamides. It is easy to process through extrusion, blow-moulding, roto-moulding, and injection moulding. PA11 has a broad chance of utilized temperatures (- 40° C to + 130° C) with extraordinary flexural modulus up to 1200 MPa and strain% to 300-400%. PA11 resins are incredibly lightweight, having a density of 1.03 g/cm<sup>3</sup> [117]. PA11 matrix can accommodate countless kinds of fillers and have the valuable properties of impact, tensile, and thermal [112,122]. In this way, thinking about these fantastic properties, PA11 expansion in other reused polymers to shape a matrix can assume an indispensable part in rela-



tively expanding the overall properties of impact, stiffness and tensile strength.

Thermal, Mechanical, and morphological properties of sepiolite-based PA11 nanocomposites have been analyzed by Herrero et al. [84]. The blends were manufactured by two different techniques, including in situ polymerization and melt compounding. According to them, improvement in thermo-mechanical properties was noticed with rising sepiolite content regardless of the manufacturing approach. However, the PA11/sepiolite nanocomposites prepared through in situ polymerization showed better tensile strength, heat distortion temperature and Young's modulus, owing to a higher dispersal of sepiolite filler in nanocomposites. Hu et al. (2006) investigated the mechanical properties of PA11 and its clay nanocomposites at several loading, as listed in Table 4. They determined that the mechanical properties of PA11 nanocomposite gradually rises with the rise of clay content.

PA12, PA11, and PA6 nanocomposite filled with MMTs assessment are recorded in Table 5. The organoclays used to analyze the properties were fabricated by a cation exchange reaction among sodium MMT and quaternary alkylammonium chlorides [124].

PA6 nanocomposites Izod impact strength for the entire concentrations of clay is unresponsive. However, nanocomposites dependent on PA11 and PA12 reduces severely with rising clay contents; this behaviour is agreed to stem princi-

pally from the Izod ductile area to brittle transition temperature regarding the ambient testing temperature for the three separate kinds of nanocomposites [124]. The properties of the PA11 nanocomposite mentioned above are very good. Combining PA11 with recycled polymers with low-cost fillers such as sepiolite would be a great notion to vie with several manufacturing materials in the automotive and aerospace industries.

#### 4.4. Polymethyl methacrylate/sepiolite

Polymethyl methacrylate (PMMA) is a bio-inert acrylic resin having inadequate thermal and mechanical properties. Hence sepiolite is utilized as a filler and introduced in the matrix to enhance mechanical and thermal properties [125,126]. The strong enhancement in fracture toughness of PMMA/sepiolite composites was observed at 50 wt% incorporations of sepiolite. This may be attributed to the capability of sepiolite filler to prevent the cracking and so improved the fracture resistance of sepiolite based polymer nanocomposites. Similarly, significant improvement in thermal properties was reported by Vahabi et al. [126] when sepiolite in combination with zirconium oxide ( $ZrO_2$ ) and MMT in the existence of ammonium polyphosphate (APP) was investigated in the PMMA matrix. The improvement and mechanism of PMMA/sepiolite composite have been investigated by several researchers [125,127-132].

#### 4.5. Poly(vinyl chloride)/sepiolite

Poly(vinyl chloride) (PVC) is one of the most noteworthy typically used thermoplastic polymers. Generally, PVC is delivered in two broad formations: inflexible or plasticized polymer and flexible or plasticized plastic. The Influence of Sepiolite on thermal stability, morphology, interactions, and optical behaviour on the PVC matrix was examined in the research of Turhan et al. [50]. Overall, results showed that thermal stability and some other properties were improved

**Table 4** Mechanical properties of PA11 and its nanocomposite [123].

Samples formulations	DMA Modulus (GPa)	Tensile Modulus (MPa)
PA11/Clay (95/5)	1.84	861.80
PA11/Clay (98/2)	1.41	752.60
PA11/Clay (99/1)	1.31	622.40
Neat PA11	1.08	612.60

**Table 5** PA12, PA11 and PA6 nanocomposites mechanical properties [124].

MMT's wt%	Modulus [GPa]	Yield Strength [MPa]	Elongation at break [%]	Izod Impact Strength [J/m]
PA12 Nanocomposites				
0.0	1.50	40.9	286	161.1
1.7	1.80	44.9	245	106.9
2.9	2.00	46.7	246	78.2
4.4	2.30	48.4	131	69.9
6.5	2.70	50.5	62	40.0
PA11 Nanocomposites				
0.0	1.30	41.3	312	116.2
1.8	1.60	44.3	251	75.5
3.1	1.90	47.1	264	38.0
4.0	2.0	48.9	206	33.4
5.7	2.30	51.4	65	31.0
PA6 Nanocomposites				
0.0	2.70	69.7	129	43.9
1.5	3.50	84.0	NA	34.7
2.9	4.20	85.2	31.0	34.4
4.6	4.70	90.7	16.0	37.0
6.6	5.80	69.7	3.6	36.5

by incorporating sepiolite in the PVC matrix. Generally, no other paper has been found on sepiolite/PVC composite except the above mentioned, and it needs further investigation and exploration.

#### 4.6. Polycarbonate/sepiolite

The mechanical, vapour transmission and ionic migration resistance properties of polycarbonate (PC)/sepiolite-based membrane were studied by Oliveira et al. [133,134]. According to the results, slightly thermal and mechanical properties were improved. Tensile strength was improved  $55.1 \pm 1.8$  MPa, whereas it was  $54.3 \pm 4$  MPa for neat PC matrix. Similarly, the glass transition temperature ( $T_g$ ) was improved from 132.7 to 135.2 °C. The water vapour transmission testing uncovered that the melt state process shows a critical part in the clay-matrix interaction as related to specimens made by the casting process, lessening water penetration and meaning that the rendering of the casting process for the manufacturing of polymer membranes to a melt extrusion processing wants further intensive and explicit advancement.

The properties of sepiolite reinforced nanocomposites having a matrix of polycarbonate/poly(acrylonitrile-butadienestyrene) in the existence of polystyrene-maleic anhydride copolymer utilized as a compatibilizer were investigated by Bicheng et al. [135]. It was found that with rising filler loading up to 7.5 wt% in the matrix, the mechanical and thermal properties followed the trend of increasing. At five wt% nanofiber loading, the formulated composite presents the tensile strength of 52.10 MPa, the Vicat softening temperature of 124.0 °C, and the tensile elastic modulus 764.40 MPa.

#### 4.7. Polyurethane/sepiolite

The mechanical properties of thermoplastic styrene-ethylene-butylene-styrene copolymers (SEBS) and polyurethanes (PU) with the incorporation of various fillers were studied by Jaudouin et al. [136]. The silicates that were organically changed and used were Cloisite® 30B, an MMT adjusted with a ternary ammonium salt with hydroxyls as end groups, and Nanofil®2 modified by long chains of hydrocarbon and benzyl groups. Sepiolite, a characteristic clay with stringy morphology, was likewise used. According to the research work results and conclusion, the tear strength's best development was attained using sepiolite at a level of 1.0 wt%, but at the minor cost of strain%. The modification of sepiolite structure by thermal treatment and the effect on the properties of PU adhesive was explored by Torro et al. [137]. Two different temperatures that are 550 and 1000 °C were utilized to modify the sepiolite structure and then incorporated in solvent-based PU adhesive. The high-temperature application to sepiolite removes the pores, and the structure of sepiolite collapses. According to the results, the mechanical and thermal properties of untreated sepiolite-based PU composite were better than the treated ones, which is due to the better interaction between sepiolite and PU adhesives.

The thermal properties of sepiolite-based PU nanocomposites were analyzed by Chen et al. [54]. According to the results, PU nanocomposites' thermal stability was enhanced by modified sepiolite. The onset of decomposition temperature for PU

nanocomposites with a sepiolite content of 3.0 wt% was almost 20.0 °C better than neat PU. Sepiolite was incorporated in PU to investigate extremely effective recyclable oil absorption by Qiu et al. [138]. Sepiolite was investigated for the influence of crystallinity and shape memory properties in PU composites [139].

The thermal and mechanical properties of Sepiolite in PU were studied by Chen et al. [79]. According to the observations, the tensile strength and strain% for the nanocomposites upsurge with the incorporation of sepiolite related to the neat polyurethane. Similarly, the nanofillers incorporation results in more excellent thermal stability. The Influence of modified Sepiolite on PU was reported by Chen et al. [55]. Preparation and properties investigation of Sepiolite and PU foams has been written by Wang et al. [140]. The effect on flame retardant and smoke suppressant properties of PU and Sepiolite was investigated by Pan et al. [141]. The PU/sepiolite nanocomposites were analyzed for the purification of polluted water [142]. The mechanical properties and synthesis of epoxy-based PU and Sepiolite were done by Anna et al. [143]. In contrast, the effect of Sepiolite on the structure and properties of PU nanocomposites was studied and discussed by Lu et al. [144]. All of the above research work done for various purposes and application showing the significant influence and properties of sepiolite based PU nano composited utilized for various applications.

#### 4.8. Ethylene-vinyl acetate copolymer/sepiolite

The Influence of Ethylene-vinyl acetic acid copolymer (EVA)/sepiolite on the wax crystal and rheological properties of Daqing raw petroleum was examined by Zhang et al. [145]. The natural nano-sepiolite was changed to accomplish dispersibility exceptionally in the oil stage, and afterwards, EVA/nano-sepiolite was made by melt blending. The impacts of un-doped/doped different oil samples on the gel point, pour point, yield point, and Daqing raw petroleum's viscosity were examined. The changes of wax crystal morphology at a low-level temperature were seen by polarizing optical microscopy (POM). The outcomes show that in light of the heterogeneous nucleation component and outrageous scattering in the oil stage, EVA/nano-sepiolite significantly influences the dropping pour point. It makes EVA a better local concentration in the oil phase, debilitating the wax crystal structure and upgrading Daqing raw petroleum's low-temperature rheology. Diverged from unadulterated EVA, EVA/nano-sepiolite can likewise diminish the pour point, gel point, viscosity, and yield value of Daqing raw petroleum at the ideal additive intensity 200 ppm. It can also reduce the pour point 2.0 oC and further lessening the gel point 0.93 oC; at 15 oC, the viscosity decrease rate was 20.0%, and the yield value at 15.0 °C was decreased 82.0%.

The thermal, mechanical, and flame retardant properties of unmodified and modified sepiolite incorporated in EVA were investigated by Hossein et al. [146]. According to them, the modified sepiolite's thermal, mechanical, and flame-retardant properties were better than the unmodified one. It was likewise detailed that the sepiolite guard's carbonyl group of EVA from further debasement. Most excitingly, a concurrent ascent in toughness and ductility was observed in the produced composites.

The comparison of various fillers, including sepiolite incorporation in EVA, was investigated by Isci et al. [147]. The mechanical properties of sepiolite in the matrix of PA6/EVA were investigated by Fauzi et al. [112]. The concentration of sepiolite was from 2 to 10 phr while PA6/EVA ratio was 80/20. According to them, modulus and flexural strength were enhanced up to 6.0 phr of sepiolite loading. In contrast, PA6/EVA/sepiolite composite's impact strength was reduced steadily as sepiolite content rose. The optimum properties of flexural and impact strength were achieved at 4 phr of sepiolite. Similarly, the thermal and mechanical properties of EVA/sepiolite were investigated and discussed by Hussein et al. [47]. The improvement in adhesion of filler in the EVA matrix through UV was investigated by Ladete et al. [148].

#### 4.9. Polystyrene/sepiolite

Morales et al. [149] worked upon the thermodynamic compatibility upon sepiolite and poly(vinylidene fluoride)- polystyrene polymer blends. The investigation on the manufacturing of mesoporous (MCM-41) from naturally available sepiolite and its catalytic activity of cracking waste polystyrene plastics was carried out by Jin et al. [150]. The formation of altered sepiolite-g-polystyrene sulfonic acid nanohybrids through radiation-induced graft polymerization was finished by Shaista et al. [151]. As indicated by them, the grafting yield ascends with the assimilated portion, and the system allows the controlled grafting of styrene on sepiolite in dichloromethane. The usage of sepiolite in polystyrene for magneto-optical Faraday activity in straightforward FeCo-sepiolite/polystyrene nanocomposites was explored by Fernandez et al. [152].

### 5. Conclusion and future prospects

The advancement of sepiolite strengthened polymer has been a functional area of exploration for research throughout the most recent couple of decades because of its reinforcing capability in thermoplastic polymers. The most widely studied area among the polymer utilized includes PP and PE composite for mechanical and thermal properties. The researchers' most concerning area consists of the interfacial adhesion, polymer chains restrictions, and polymer deformation affecting the thermal and mechanical properties. Generally, the incorporation of sepiolite enhances the thermal and mechanical properties because of its excellent morphology, loading content of sepiolite, type of surface treatment, and hybridization with more fillers. One of the fundamental reasons prominent to deterioration of mechanical and thermal properties is the weak interaction between filler and polymer matrix in some polymers and bad dispersion of sepiolite that happens ahead of the optimal filler incorporation. The impact of size, shape, and quality of sepiolite must be explored more to enhance the properties.

Furthermore, investigation of sepiolite in polyesters such as rPET and its blends with other polymers are almost non that needs to be examined. Besides, the application of Sepiolite in PA11 and polyesters needs further exploration. Excitingly, the usage of sepiolite in polymer matrix offers an extra advantage of environment-friendly material as it can found naturally. Additionally, the size and shape of the sepiolite may be directed to manufacture nanocomposites having preferred

properties. Moreover, the channel-like structure and filling of sepiolite into a polymer matrix can increase numerous useful properties, and the investigations of the tribological properties of sepiolite filler filled thermoplastic composites could be a leading area of exploration in upcoming research work, particularly for automotive applications, for example, brake pads, interior parts, electrical switches, and gears. Moreover, research of sepiolite incorporation in green polymers, for example, PLA and PHA matrices, for the application in biomedical and other fields should be done. The studies on nano-size sepiolite incorporation in PET, rPET, and its blending polymers are very limited in the literature to enhance flammability, thermal and mechanical properties; hence this could be one more attractive area of research for exploration by academics and entrepreneurs.

### Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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