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## Mechanical properties of LDPE and PS polymer matrix composites reinforced with GNP and CF — A critical review



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#### ABSTRACT

There is always a vital need for more robust, affordable, and multifunctional materials to satisfy the demands of industrial consumers. Therefore, polymer matrix composites (dual and hybrid matrix) have become popular with multiple fillers to meet these needs. Graphene nano-platelet (GNP) and Carbon fibre (CF) are popular among those fillers due to their superior properties, such as good mechanical, thermal, and electrical properties. Low density polyethylene (LDPE), Polystyrene (PS), GNP, and CF are popular and heavily used in the packaging, automotive, and aerospace industries. However, it would be good to look at how these areas have evolved over the last few decades. Hence, this review focuses on a comparison of LDPE and PS as a matrix and GNP and CF as a filler, considering the content that determines the overall performance of blends and composites. The literature was screened for the last few decades. The blends and/or composites produced by a twin-screw extruder were included. A total of 1628 relevant papers were retrieved from all databases. Based on the review, it was deduced that more research should be needed in areas such as the aerospace industry to identify optimum content. Most of the analysis showed that factors such as filler surface area, dispersion, and content affect overall blends and composites' performance in terms of mechanical properties, especially elastic modulus and tensile strength, and other properties. Based on the review, it was realised that using 20 and 30 wt%, 2 and 30 wt%, 2 and 4 wt%, and 20 and 30 wt% filler was the most common combination giving the optimum content for LDPE, PS, GNP, and CF, respectively. EMS and TSH changes of the composites were calculated according to their optimum content. Overall, LDPE and PS are good in packaging areas, but their mechanical properties still need to be improved for use in industries such as automotive, aerospace etc. Due to the advantages of GNP and CF, they are used in different applications, such as electrical devices, medical tools, and automobile vehicles. However, these properties are affected easily by interfacial adhesion, dispersion, and aggregation. Many researchers have searched these parameters and analysed how to prevent the negative effects of these parameters. In conclusion, this review will be helpful for researchers and industrial people to be aware of the state-of-theart of carbon-based composites and the evolution of LDPE, PS, GNP, and CF.

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#### 1. Introduction

Composite materials are defined as two or more phases that constitute the materials, and their properties are distinct from

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each component [1]. Composites generally consist of reinforcement and a matrix, respectively. When the reinforcement is responsible for load carrying, providing strength and thermal balance to the structure, the matrix prevents the reinforcement from mechanical damage, chemical attack, and load placement from one location to another [2-4]. The matrix can be metal, ceramic, or polymer, while the reinforcements can be in the form of discontinuous fibre (short or whiskers), continuous fibre, and particles shown in Fig. 1 [5,6]. There are different kinds of fibrereinforced composites, but the most common one is polymer based because of its outstanding mechanical properties and high

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List of ab	breviations	PS	Polystyrene
		PSN	Pristine silica nanoparticles
ABS	Acrylonitrile-butadiene-styrene	PTES	Phenyltriethoxysilane
AFM	Atomic force microscopy	PVC	Poly (vinyl chloride)
ASTM	American society for testing and materials	RTW	Recycled tyre waste
C10A	Cloisite 10A	s-ZnO	Spherical zinc oxide
C20A	Cloisite 20A	SEBS	Styrene ethylene butylene styrene
CA	Cellulose acetate	SEBS-MA	Styrene ethylene butylene styrene-maleic anhydrid
CF	Carbon fibre	SEM	Scanning electron microscope
CNT	Carbon nanotube	SMA	Styrene-maleic anhydride
CS	Ceratonia silique	SR	Silicon rubber
DMA	Dynamic mechanical analysis	TEC	Thermal expansion coefficient
DSC	Differential scanning calorimetry	TEM	Transmission electron microscopy
DTGA	Derivative thermogravimetry	TGA	Thermogravimetric analysis
EAB	Elongation at break	TPCS	Thermoplastic corn starch
EDM	X-ray dot mapping	TPU	Thermoplastic polyurethane
EDS/EDX	Energy-dispersive X-ray spectroscopy	TSH	Tensile strength
ELS	EVA/LDPE/PSN	TSS	Tensile stress
EMS	Elastic modulus	VTES	Vinyltriethoxysilane
EPP	Waste expanded PP		Ultrahigh molecular weight polyethylene
EPS	Expanded Polystyrene	ULDPE	Ultralow density polyethylene
EVA	Ethylene-vinyl acetate	UTS	Ultimate tensile stress
FMS	Flexural modulus	w-ZnO	Whisker zinc oxide
FSH	Flexural strength	WAXD	Wide-angle X-ray diffraction
			Exfoliated GNP
FTIR	Fourier-transform infrared spectroscopy	xGNP	
GMA	Glycidyl methacrylate	XPS	Extruded Polystyrene
GNF	Graphene nanofibre	XRD	X-ray diffraction analysis
GNP	Graphene nano-platelet	YSS	Yield stress
GO	Graphene oxide	**	
HDPE	High density polyethylene	List of Sym	
HMWPE	High molecular weight polyethylene	E'	Dynamic storage modulus Pa
HNA	ß-hydroxynaphthoic acid	E"	Dynamic loss modulus Pa
IM	Injection Moulding	G'	Storage modulus MPa
ISH	Impact strength	G"	Loss modulus MPa
LDPE	Low density polyethylene	k	Thermal conductivity W/m.K
LDPE-g-M	A low density polyethylene-graft-maleic anhydride	n	Power law index —
LLDPE	Linear low-density polyethylene	T <sub>5%</sub>	5% weight loss temperature C
LVR	Linear viscoelastic region	T <sub>10%</sub>	10% weight loss temperature C
MDPE	Medium Density Polyethylene	T <sub>20%</sub>	20% weight loss temperature C
MMT	Organomontmorillonite	T <sub>30%</sub>	30% weight loss temperature C
MWCNT	Multi walled carbon nanotube	T <sub>50%</sub>	50% weight loss temperature C
Na-MMT	Sodium MMT	T <sub>c</sub>	Crystallisation temperature C
NCaCO <sub>3</sub>	Nano-CaCO <sub>3</sub>	T <sub>e</sub>	Crystallisation end temperature C
NIS	Notched impact strength	$T_{\rm f}$	Final degradation temperature C
PA 12	Polyamide 12	tan δ	Damping factor —
PBAT	Polybutylene adipate co-terephthalate	Tg	Glass transition temperature C
PE	Polyethylene	T <sub>i</sub>	Initial decomposition temperature C
PEEK	Polyether ether ketone	T <sub>max</sub>	Maximum decomposition temperature C
PLA	Poly (lactic acid)	T <sub>max</sub> T <sub>onset</sub>	Reaction start temperature C
PMMA	Poly (methyl methacrylate)		Crystallinity degree %
	Polypropylene	X <sub>c</sub>	
PP ~ MAI		$\Delta H_c$	Crystallisation enthalpy kJ/mol
	Polypropylene-graft-maleic anhydride	$\Delta H_{m}$	Melting enthalpy kJ/mol
PPO	Polyphenylene oxide	η*	Complex viscosity Pa.s
PPr	Recycled copolymer polypropylene	ω	Angular frequency Rad/s
PRCF	Pristine regenerated cellulose fibres		

specific strength (strength-to-weight ratio) [7]. These remarkable properties have replaced aluminium and steel in different fields such as aerospace, military, sports, and electrical industry [8,9].

### 1.1. Properties of polyethylene (PE)

Polyethylene (PE) has become one of the most widely used synthetic thermoplastic polymers, predominantly linear low-

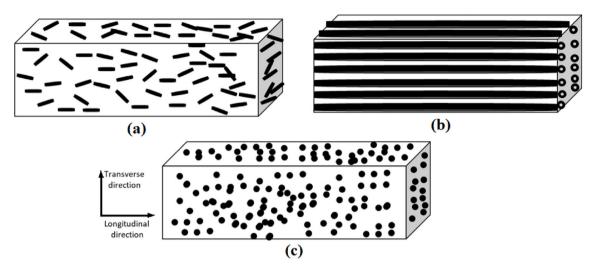


Fig. 1. Classification of composites in filler a) short or whisker, b) continuous, and c) particles.

density polyethylene (LLDPE) and low-density polyethylene (LDPE). This is due to a variety of purposes, including light weight and low cost [10,11]. PE can be distinct in density, chain structures, and crystallinity levels [12]. The mechanical properties of PE including stiffness and yield strength increase with the degree of crystallinity [13].Ultralow density polyethylene (ULDPE), Linear low density polyethylene (LLDPE), Low density polyethylene (LDPE), Medium Density polyethylene (MDPE), high-density polyethylene (HDPE), and ultra-high molecular weight polyethylene (UHMWPE) are the forms of PE, in the order of increasing crystallinity. The density ranges of these different PE types are summarised in Table 1. The density of UHMWPE unexpectedly drops below that of HDPE due to the large molecular weight affecting the packing of polymer chains into the crystal structure [13].

In LDPE the ethylene consists of many side branches, which are usually short [10,14,15]. These kinds of branches keep from preventing knit patterns. As a result, LDPE is comparatively tough, soft, flexible and is widely used in applications which require heat sealing [16]. In addition, it is used dominantly in the packaging industry, and has good chemical resistance. On the other hand, HDPE shows good tensile strength. Unique properties of the different PE types are summarised in Table 2.

The proportions of each type of PE are given in Fig. 2. Ethylene can endure temperatures of up to 80  $^{\circ}$ C for a long time while up to 90  $^{\circ}$ C for a short time [18,19].

#### 1.2. Properties of polystyrene (PS)

In the meantime, PS is also utilised extensively because of its relatively low cost, good mechanical features and excellent durability [20,21]. It is an amorphous thermoplastic polymer with highlighted properties like being hard, colourless, and excellent clarity but somewhat brittle nature [15,22]. PS can be categorised as Regular PS, PS Foam and PS Film [23,24]. Regular PS is a solid with

**Table 1** Approximate density ranges of PE variants [13].

Type of PE	Density Range (kgm <sup>-3</sup> )
ULDPE	0.880-0.910
LLDPE	0.910-0.920
LDPE	0.910-0.940
MDPE	0.925-0.940
HDPE	0.941-0.967
UHMWPE	~0.930

high density which is used to manufacture a plethora of consumer products such as plastic containers and dishware, electrical equipment casings, stationary items and toys [25]. PS foams consist of Expanded Polystyrene (EPS) and Extruded Polystyrene (XPS). These materials exhibit good thermal insulation properties alongside low thermal conductivity, making them ideal candidates for insulating and packaging. Regular PS can be formed into clear PS films, which further subdivide into General, Oriented (OPS) and High impact (HIPS) films [26]. OPS can be used as a cover material, whereas Biaxial OPS was introduced for use in capacitors. Furthermore, BiaxialOPS film preserves clarity while overcoming some of the brittleness of unstretched plastic used in toys, bottles, jars, and other areas such as insulation [27,28].

#### 1.3. Properties of carbon fibre (CF)

CFs are thin lightweight synthetic fibres which mostly consist of carbon atoms aligned parallel to the fibres long axis. These are widely used in producing polymer-matrix composites because of their superior thermal, electrical, and mechanical characteristics and their specific stiffness, strength, and fatigue strength when compared to other reinforcing fibre materials (like natural fibre and aramid) [29]. Apart from high strength and stiffness, CFs also exhibit low thermal expansion, good vibration attenuation, resistance to chemical degradation and corrosion [30]. Because of these characteristics, they are recognised to offer much potential for improving polymer properties. However, because of their intractability, flexibility and poor performance under compression, direct use of CF in engineering applications is restricted. These inadequacies can be avoided by combining CFs with a matrix material, which can handle compressive loading, gives the fibres structure and ensures uniform load distribution [31,32]. Polymer matrices are an ideal solution for this matter since CFs in turn help reduce the brittle nature of polymeric plastics [3,33,34]. The resulting composite parts boast highly directional mechanical properties, light weight and durability [35]. CFs are sub-categorised according to the precursor material used during the manufacturing stage. These are PAN-based, Pitch-based and Cellulosic-based CFs [36,37]. Currently, PAN-based CFs account for more than 90% of the CFs in circulation [38]. Furthermore, although Cellulosic-based CFs were the earliest to be introduced, now they are less prevalent due to low carbon yield and low cost-effectiveness [39]. Table 3 provides an overview of the properties of each of these subcategories.

**Table 2** Characteristics of PE variants [13,17].

Type of PE	Characteristics
ULDPE	High tear resistance, puncture resistance, good optical properties
LLDPE	High tensile strength, impact resistance, puncture resistance, crack resistance
LDPE	High ductility, low strength
MDPE	High resistance to cracking under stress
HDPE	High intermolecular forces and tensile strength
UHMWPE	High toughness, wear resistance, chemical resistance

One of the most pressing issues with CFs is their cost of manufacturing. Given the high demand for CFs in the industry, uncovering cost-effective methods of CF production and optimising current methods has become an active area of research. Since the cost of a CF is directly proportional to its precursor cost, developing inexpensive precursors is the most promising approach towards solving this problem [44].

Furthermore, in light of the emphasis placed on sustainable engineering, research has become accelerated in the field of lignin-based CFs. Furthermore, since lignin is derived from biomass and is abundantly available, it has the potential to addresses the issue of high manufacturing costs associated with CFs as well [30].

#### 1.4. Properties of graphene nano platelets (GNP)

GNPs are nanoscale reinforcements, which can be combined with polymer matrices to produce nanocomposites. The thermal, mechanical and electrical properties of GNP are similar to Carbon Nanotubes (CNT), which is another type of common nanoscale reinforcement [45,46]. However, GNPs provide a superior specific surface area [47] and are easier to work with while having a lower cost of manufacture than CNTs [48,49]. GNPs utilised commercially are in various levels, based on their diameter, which ranges between nanometres and micrometres. There are many improvements in nanotechnology related to using GNPs that alter polymer matrices and improve polymer-based composites' thermal, mechanical, and electrical behaviour [48,50–53]. Some features of the CNT and graphene are illustrated in Fig. 3 [54].

**Table 3** Properties of different types of CF [40–43].

Property	PAN-based	Pitch-based	Cellulosic-based
Density	Moderate	High	Low
Modulus	Moderate	High	Low
Strength	High	Moderate	Low
Electrical Conductivity	Moderate	High	High
Thermal Conductivity	High	High	Low
Cost	Moderate	High	Low

One of the main reasons for using GNP over CNT is their relative ease of manufacturing graphene [55]. The main methods include Chemical Vapour Deposition (CVD), Arc Discharging Methods (ADM), ball milling and graphite intercalation [48]. CVD and ADM have the ability to produce high quality GNPs, they are expensive and time consuming. On the other hand, GNPs can be efficiently produced using ball milling and intercalation techniques [56]. Ball milling involves using mechanical grinding to break the interatomic bonds between graphene sheets. In intercalation methods (also known as electrochemical exfoliation), chemicals capable of releasing gas are made to infiltrate the spaces within graphene sheets. Subsequently, the gases expand and separate the graphene layers, resulting in GNP [57].

#### 1.5. Commercial market of CF and GNP composites

Due to the use of CF composites in many applications such as automotive, wind turbines, sports, aerospace, and marine, the demand for CF composites has recently expanded dramatically. Carbon fibre composites had a market value of about 16 billion dollars in the United States in 2014, and its demand is predicted to rise to 290,000 tonnes in 2024. Such an increase of carbon fibre demand is particularly due to the expansion in the automotive and aerospace sectors over the last two decades [58,59].

In 2016, the use of CF composites in the automotive sector increased dramatically, exceeding 20% and surpassing 25,000 tonnes in demand. Due to the affordable cost of polymer matrices for CF composites and the improved features of emerging CF composites, such as enhanced strength and light weight, this

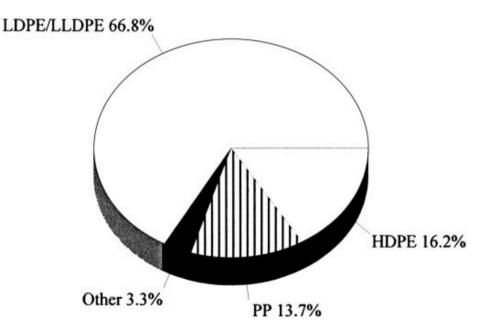


Fig. 2. The main polymers used in the packaging industry [19].

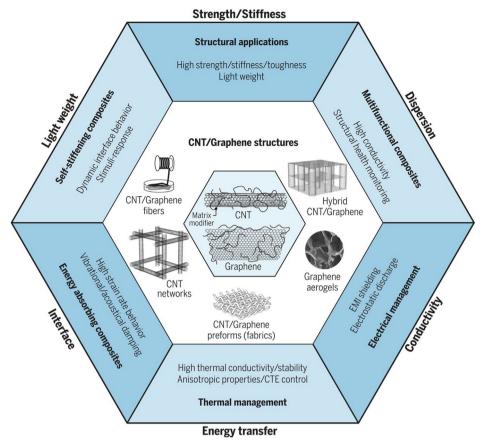


Fig. 3. A schematic illustrating several properties and criteria for CNT and graphene [54].

proportion is predicted to expand to 13% by 2024 [58,59]. Fig. 4 depicts the use of carbon fibre composites in 2016 against what is projected in 2024 in the United States where it clearly indicates that the demand for this material will be increased in all the areas listed.

According to a survey published by Kraus and Kühnel in the United States, polymer matrices had the most extensive utilisation in 2013, approximately 64%, compared to other types of matrices such as ceramics and metals as illustrated by Fig. 5 [60].

According to this data, thermoset polymers were used as matrices in roughly 76% of all polymer types in 2013. In contrast, thermoplastic matrices, including LDPE and PS, were used in around 24% of all polymer types [59,60]. The findings of this analysis back up the idea that thermoplastic matrices be used in combination with CF to solve the limitations in thermoset matrices discussed in the preceding section.

According to market forecasts, the worldwide graphene nanoplatelets market is predicted to reach \$206.0 million by 2025, which was found to worth of \$11.9 million in 2016 (see Fig. 6), where the driving force for this escalated demand is due to the consumer sectors such as energy-power and aerospace. The market is projected to develop because of the growing demand for lightweight composite materials in various consumer industries [61].

Because of the increasing demand for high-performance composites and sustainable energy and power storage systems (as shown in Fig. 7), North America held the highest share of the worldwide market in 2016. Few aviation and automobile manufacturers in the United States have bolstered demand in recent years. Over the forecast period, Asia Pacific is expected to see a surge in demand for graphene nanoplatelets. By 2025, Asia Pacific is expected to overtake North America as the worldwide market leader. Future

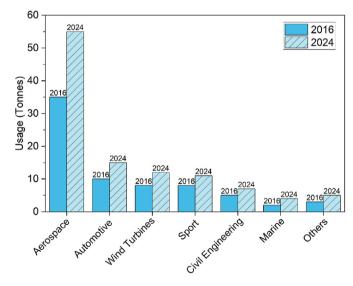


Fig. 4. Comparison of the CF composites market between 2016 and 2024 [58].

industrial expansion in nations such as China, India, and Japan is estimated to contribute to the Asia Pacific's fast growth [61].

The primary goal of this review is to determine the optimum loading ratio (LDPE, PS, GNP, and CF) in terms of EMS and TSH properties that could be used in a variety of applications such as containers, food packaging, flame retardant, aircraft, space shuttle, and water pipes, bottles, and plastic bags in the current literature. The blend(s) (LDPE and PS based) and composites (GNP and CF reinforced) produced using a twin-screw extruder were researched.

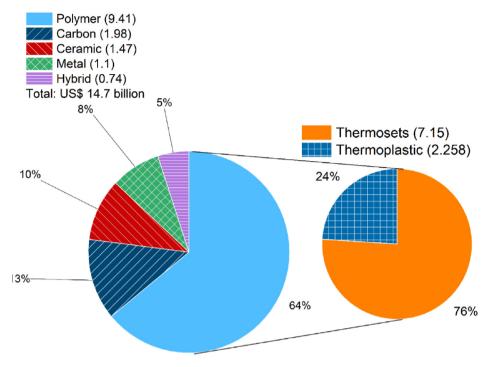
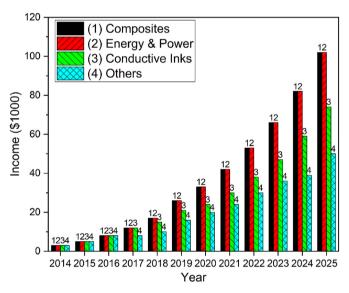


Fig. 5. The income from carbon fibre composite matrices in the United States in 2013 [60].



**Fig. 6.** Income(USD Thousand) of GNP composites in 2014 against projected 2025 in the United States [61].

Following that, the articles were refined according to the research criteria. Then, the previous studies that have already been done in terms of EMS and TSH properties compared. Finally, it is expected to summarize the best possible compositions to enhance the mechanical performance and other properties, such as thermal, electrical, and rheological behaviour.

### 2. Methodology

#### 2.1. Scope of the review

For this study, previous works focusing on mechanical properties were considered. The inclusion criteria were as follows: (a)

includes LDPE and/or PS based and GNP and/or CF reinforced blends or composites; (b) investigates the mechanical properties of blends (LDPE and PS based) or composites (GNP and CF reinforced), and (c) investigates the synergistic mechanical effects of GNP and CF reinforced composites.

Exclusion criteria were as follows: (a) articles not published in the English language; (b) articles for which the full text was inaccessible; (c) book chapters, thesis, dissertations, ideas, editorials, and opinions.

#### 2.2. Focus point

The focus of this research was based on determining the optimum mechanical properties, and the question was: "Which composition of blends (LDPE and PS) or composites (GNP and CF reinforced) shows better mechanical behaviours than others in terms of EMS and TSH?".

#### 2.3. Search strategy

Web of Science, Scopus, and Science Direct were searched for eligible studies until September 2022. No limits on a year of publication were placed for the database searched. The articles were searched and retrieved using different combinations of subject headings terms/subject headings and natural language terms/ phrases, which in all instances encompassed the following integral concepts of the focus question: (a) mechanical effect, (b) twinscrew extruder (c) LDPE and PS blend(s), and (d) GNP and CF reinforcement(s). According to how each database functions, the combinations, and permutations of search phrases were tailored for that database. The optimum search method for each database was chosen based on the following constraints: (a) search syntax variants for each database, (b) character limits for the database, and (c) the number of articles retrieved after the initial search must be smaller than the maximum number of articles the database can retrieve—keywords used for each research given in Table 4.

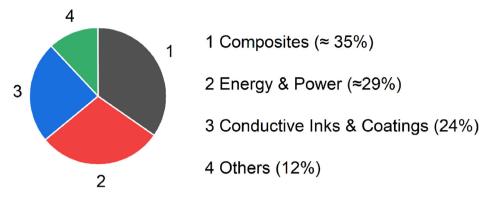


Fig. 7. The illustration of the GNP's global market share by type of application, 2015(%) [61].

There were 544, 741, 179, and 164 (for LDPE, PS, GNP, and CF, respectively) results from the initial online search of several databases. 9, 11, 9, and 14papers (for LDPE, PS, GNP, and CF, respectively) were left for eligibility assessment based on full-text examination after duplicates were removed and independent screening of titles and/or abstracts. Nine, eleven, nine, and fourteen (for LDPE, PS, GNP, and CF, respectively) studies in total satisfied all eligibility requirements and were included in this systematic review (Figs. 8, Fig. 9, Fig. 10, and Fig. 11 for LDPE, PS, GNP, and CF respectively).

## 3. Desired mechanical properties of polymer matrix composites

Various properties must be considered when choosing materials for a particular application. When considering mechanical properties, the elastic modulus (EMS) and tensile strength (TSH) are apparent requirements that must be met [62]. Polymer matrix composites have drawn a lot of attention, mainly because they are more affordable and have higher specific modulus (modulus to density ratio) and strength (strength to density ratio) than traditional metallic alloys [63]. The comparison of polymer, polymer matrix composites, metals, alloys, and other materials in price and density is given in Fig. 12.

When compared to polymer matrix composites, metals and alloys' prices are almost three times higher than that of polymer matrix composites and have much higher density. This situation offers good opportunities for use in different fields, especially aerospace.

The comparison of polymer, polymer matrix composites, metals, alloys, and other materials in terms of TS and density is given in Fig. 13.

Fig. 13 shows that metals and alloys have higher densities that limit their use. The polymer matrix composites are almost in the same range as the metals and alloys in terms of TS but much lower in density, which provides advantages to be used in a vast variety of areas [64]. The comparison of polymer, polymer matrix composites, metals, alloys, and other materials in terms of EMS and density is given in Fig. 14.

When compared to polymer matrix composites, metals and alloys, metals and alloys' EMS are almost the same, but polymer matrix composites have much lower density. While this offers polymer matrix composites myriad opportunities, it restricts the metal and their alloys' usage [65]. The comparison of polymer, polymer matrix composites, metals, alloys, and other materials in EMS and TSH is given in Fig. 15.

They are in the same range compared to EMS versus TSH of polymer matrix composites, metals, and alloys. When density is

**Table 4** Search strategy for databases.

Database	Keyword Research
Science Direct	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND " tensile strength" AND "twin-screw extruder" ("low density polyethylene" OR "LDPE") AND composite\$ NOT ternary
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("polystyrene" OR "PS") AND composite\$ NOT ternary
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("graphene nanoplatelets" OR "graphene nano-platelets" OR "GNP" OR "xGNP") AND composite\$
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("short carbon fibre" OR "short carbon fiber" OR "SCF") AND composite\$
Scopus	* ("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("low density polyethylene" OR "LDPE") AND composite\$ NOT ternary
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("polystyrene" OR "PS") AND composite\$ NOT ternary
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("graphene nanoplatelets" OR "graphene nano-platelets" OR "GNP" OR "xGNP") AND composite\$
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("short carbon fibre" OR "short carbon fiber" OR "SCF") AND composite\$
Web of Science	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("low density polyethylene" OR "LDPE") AND composite\$ NOT ternary
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("polystyrene" OR "PS") AND composite\$ NOT ternary
	"("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("graphene nanoplatelets" OR "graphene nano-platelets" OR "GNP") AND composite\$
	*("elastic modulus" OR "Young's modulus" OR "tensile modulus") AND "tensile strength" AND "twin-screw extruder" ("short carbon fibre" OR "short carbon fiber" OR "SCF") AND composite\$

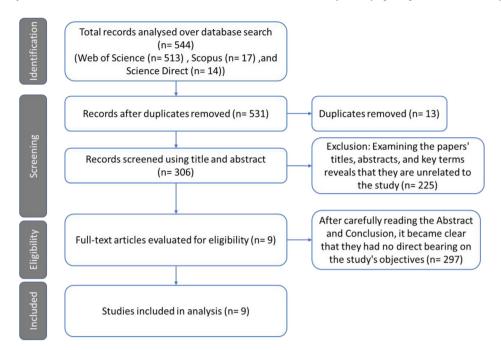


Fig. 8. Procedure used to select articles for LDPE.

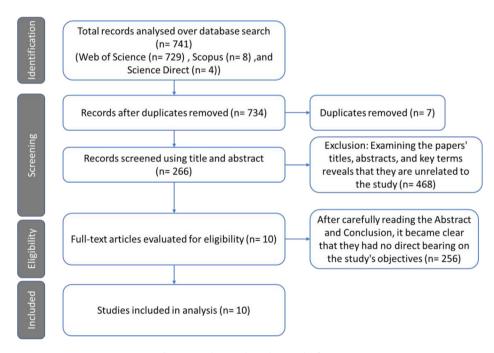


Fig. 9. Procedure used to select articles for PS.

considered a third property, polymer matrix composites are preferable, mainly in the aerospace, automotive and packaging industries. In conclusion, all parameters mentioned above are considered, so it is straightforward to understand why polymer matrix composites are preferable. The following sections will explain the factors affecting the composites in EMS and TSH.

#### 3.1. Elastic modulus (EMS)

The EMS of a material is a measure of the materials resistance to elastic deformation. Materials with a lower EMS are said to be flexible and rubbery, whereas high EMS materials are stiff. It is an extremely important parameter in engineering design due to being considered as the failure limit of a material [66] Polymer matrix composites with high EMS can be helpful for many areas, such as food packaging, automotive, and aerospace [67,68]. Properties of composites frequently fluctuates depending on the conditions used during manufacturing technique, composition, etc [69–71]. EMS can be affected by several reasons, including the fillers' specific surface area, aspect ratio, matrix-filler contact, filler-filler interaction, filler content, and filler dispersion in a matrix [72].

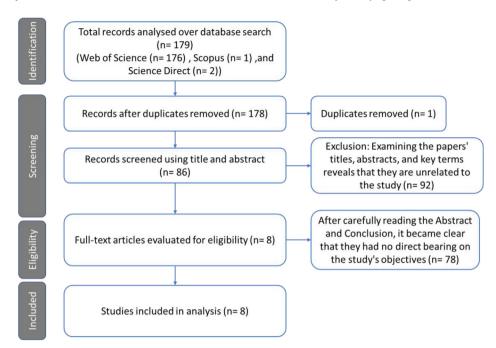


Fig. 10. Procedure used to select articles for GNP.

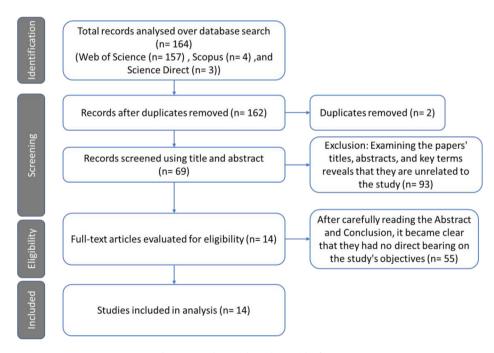


Fig. 11. Procedure used to select articles for CF.

## 3.2. Tensile strength (TSH)

The TSH, also termed the ultimate strength of a material specifies the maximum stress that could be withstood by the material without breaking. TSH can be considered as the absolute upper limit of a materials performance, and care should be taken during design stages to make sure that stresses generated during a component's operation are significantly lower than the TSH [73]. TSH depends on many factors, such as polymer-filler interface adhesion, quality of dispersion, the aspect ratio of the filler, specific surface area, and orientation [72].

Composites have high TSH, which makes them practical in various fields such as automotive, aerospace, and medical [74].

# 4. Parameters affecting elastic modulus (EMS) and tensile strength (TSH) of composites

#### 4.1. Filler surface area

The surface area has a significant role in the mechanical properties of composites. The fillers' surface areas allow interfacial

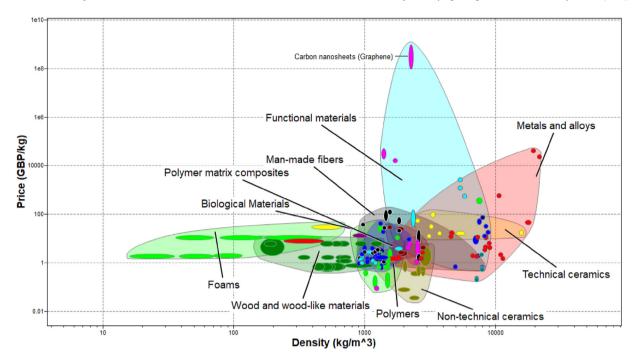


Fig. 12. A comparison of different material groups' prices versus density.

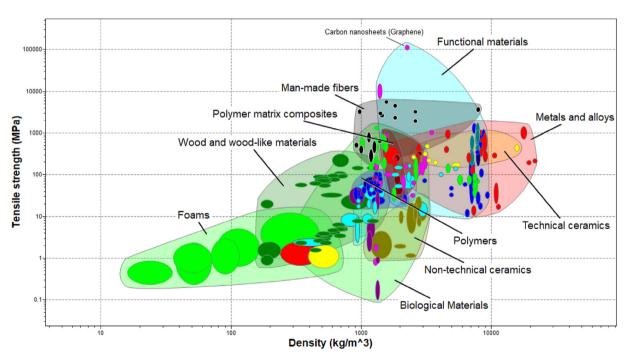


Fig. 13. A comparison of different material groups TS versus density.

contact with the matrix's polymer chains [75,76]. Due to fillers' surface energy, polymeric chains are adsorbed on filler surfaces in this instance [77]. While providing more interfacial contact, fillers with larger surface areas support more polymer chain adsorption on their surfaces, resulting in better adhesion. As previously mentioned, this adhesion between the matrix and filler has a significant impact on the final mechanical properties of the part [78]. In composites, better interfacial contact enables more efficient stress-strain transmission between the polymer and filler [79,80].

Surface area and particle size are directly related to each other. When the particle size reduces, usually surface area increases [81] and a schematic illustration of this is given in Fig. 16. However, the measurement of the surface area of fillers is not a straightforward task and requires the use of different scientific methods. One widely used method is the Nitrogen adsorption method, where Nitrogen gas is allowed to flow over the filler particles and the amount of gas molecules adsorbed onto the filler surface is used as a parameter for the particle size [75,82].

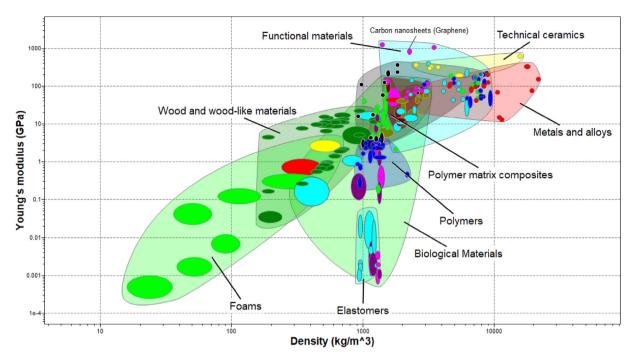


Fig. 14. A comparison of different material groups EMS versus density.

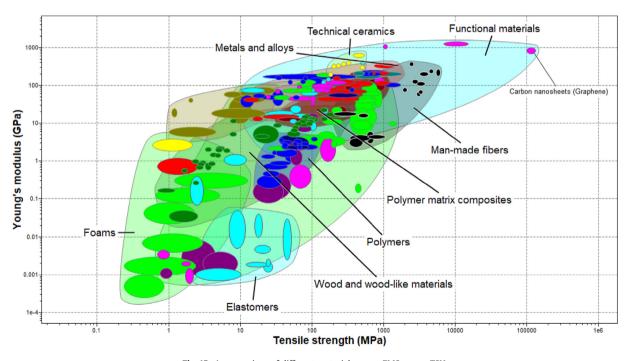


Fig. 15. A comparison of different material groups EMS versus TSH.

#### 4.2. Filler dispersion

Dispersion can be defined as measuring the distance between filler particles or aggregates [84], which is illustrated in Fig. 17. This distance plays a vital role in terms of mechanical properties. The various processing methods often cause varying degrees of dispersion of nanofillers in the matrix, which has varying effects on the mechanical behaviours of polymer composites [85]. For example, when in-situ polymerisation provides a high dispersion rate, the solution dispersion method offers a low dispersion rate

[86]. Highly dispersed fillers provide uniform mechanical properties, whereas filler aggregations result in stress concentrations [87]. Beyond mechanical properties, it was also observed that a good dispersion of fillers result in maximised radiation shielding of composite parts which are designed to withstand radiation attacks [88].

Quantifying the filler dispersion is a key challenge faced during composite manufacturing. Quantitative descriptions can be provided by directly studying micrographs of filler based composites [88]. Going a step further, obtaining microscopic images of

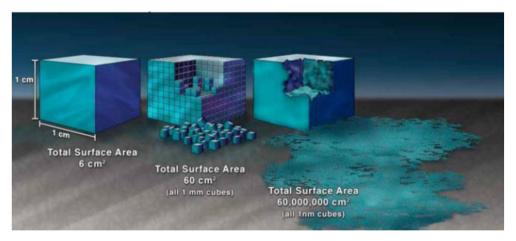


Fig. 16. Schematic illustration of surface area and particle size relation [83].

composite cross sections and defining a dispersion index using statistical methods has also been carried out to comment on the distribution of fillers [89]. Alternatively, the fractal dimension of the filler distribution can be used as a parameter to measure particle dispersion [90,91]. Here, a higher fractal dimension reflects a higher degree of dispersion. However, fabrication of samples for microscopic analysis can be difficult and time consuming, particularly for small fillers. To overcome this issue, small angle x-ray scattering has been used where the density differences within a sample can be obtained, leading to information regarding the filler dispersion [92,93]. However, x-ray scattering methods are not suitable for use in cases where the filler concentration is high, restricting its usage. 3-dimensional methods to characterise filler dispersion have also been proposed to overcome the limitation of two-dimensional micrograph based methods. The authors of [94] proposed a phase contrast based x-ray ultramicroscopy method which uses the difference in refractive index between the filler and the matrix materials. Reconstructed images of CaCO<sub>3</sub> dispersed polypropylene composites using this technique are shown in Fig. 18.

Stating the shortcomings associated with statistical methods and scattering methods, the authors of [95] used macroscopic permitivitty as a parameter for quantifying filler dispersion. A mathematical model was developed to estimate dispersion using permitivitty, which showed good agreement with dispersion values measured using micrographs. The authors of [96] used fluorescently labled CaCO<sub>3</sub> particles as an inorganic filler which were then used to reconstruct the filler distribution within the composite

using fluorescent labeling laser scanning confocal microscope visualisation technology. Fractal analysis was then performed on the visualised filler distribution to study the relationship between the fractal dimension and impact resistance. However, this method cannot be applied to all filler types, and reconstructing the filler distribution can be exceedingly difficult at high filler contents [97].

#### 4.3. Filler content

The filler composition can be a valid variable to achieve desired mechanical properties. Overall composite performance depends on also filler ratio. If the optimum filler content is determined, it provides a more cost-effective and time-efficient way for future studies [98]. An excellent example of the relationship between filler content, particle size, and TSH is given in Fig. 19. It can easily be deduced that there is always an optimum point for all composites, and it plays a crucial role for industries thanks to some advantages mentioned above. For a particular filler content, it can be seen that the TSH increases with decreasing filler size. This can be explained as mentioned in Section 4.1 where smaller particles result in a larger surface area for adhesion with the matrix. The optimal filler content would ensure complete bonding between the fillers and the matrix, and an excessive filler content would in fact weaken the composite due to incomplete bonding [99].

It has also been observed that an increasing filler content up to the optimal point typically increases the strength, hardness and stiffness of the composite material while decreasing polymerisation shrinkage and water sorption [101–103]. Here, polymerisation

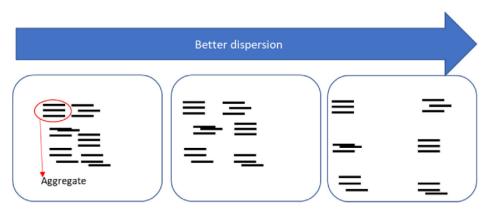


Fig. 17. Schematic illustration of dispersion effect.

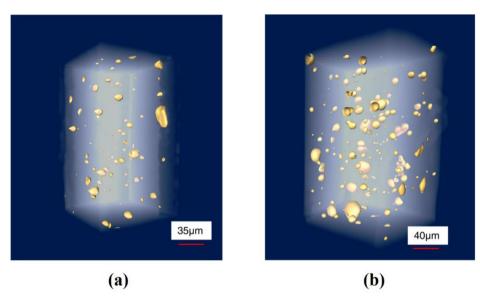


Fig. 18. Reconstructed 3D images of CaCO<sub>3</sub> dispersed PP (a) 1 wt% CaCO<sub>3</sub> content (b) 5 wt% CaCO<sub>3</sub> content [94].

shrinkage refers to the reduction in volume of the matrix polymer due to the solidification reaction. The reduction of shrinkage at higher filler contents has been attributed to the lower amount of monomers available for the polymerisation reaction [104].

## 5. Previous works focusing on LDPE and PS polymers and/or blends and GNP and CF reinforced composites

#### 5.1. PE blends and/or based composites

Al-Oqla et al. [105] investigated how adding olive leaves' fillers with different amounts (10–40 wt% with 10% increment) affects the mechanical behaviour of the LDPE. The composites were prepared and shaped with a twin-screw extruder and compression moulding, respectively. Adding the filler increased the EMS and TSH of the LDPE by 205.9% and 8.2%, respectively, while the EB declined by 9.1%. The enhancements were explained with interfacial forces. When a filler is added, this improves the transfer efficiency between the matrix and the filler. The decrease was explained by brittleness. When the brittle filler is added, this declines EB.

Sabetzadeh et al. [106] determined how the addition of thermoplastic corn starch (TPCS) (10–40 wt% with 10% increment with

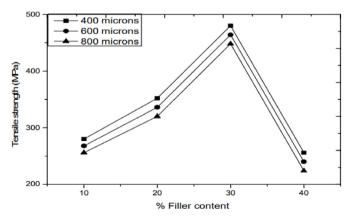


Fig. 19. TSH changes in terms of filler content and filler size [100].

constant 3 wt% LDPE-g-MA) affects the LDPE's mechanical behaviours. The composites were prepared and shaped with a single-screw extruder (screw diameter (d) = 19 mm) and compression moulding, respectively. Adding the filler declined the LDPE's EMS, ultimate TS (UTS), and elongation at break (EAB) by 44%, 33.3%, and 76%, respectively. The compatibilizer effect explained the reason for these deteriorations. When a compatibilizer is added, this creates a crossed-linked structure and results in lower interfacial forces between the filler and the matrix.

Khattab et al. [107] investigated how adding vapour-grown CNF (0.1, 0.25, 0.5, 0.75, 1, 1.5, 2, 2.5, and 3 wt%) affects the LDPE's thermal as well as mechanical properties. The composites were manufactured and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the filler increased the EMS, TSH, and hardness of the LDPE by 34.5%, 13.6%, and 7.6%, respectively, while the EAB declined by 19.4%. The reason for the improvement was explained that adding the filler could hinder crack growth and result in mechanical enhancement.

Sabet and Soleimani [108] examined how the addition of CNT (1, 3, 5, and 10 wt%) influences LDPE's mechanical and electrical behaviours. The composites were manufactured and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the CNT increased the LDPE's EMS and TSH by 90.9% and 361.5%, respectively. The reason for the enhancements was explained by stress transfer. When the CNT is added, this provides better stress transfer efficiency and results in higher mechanical performance. In addition, the addition improved the k of the LDPE by 100%. This enhancement was due to the percolation network. When the filler is added, it creates the network and enhances the k marginally.

Awad [109] studied how the addition of nanoclay (2, 4, and 8 wt %) affects the thermal and mechanical behaviour of the LDPE. The composites were manufactured and shaped with a twin-screw extruder and hot pressing, respectively. Adding the filler increased the matrix's EMS, TSH, FMS, FSH, and ISH by 38.6%, 16.8%, 19.4%, 11.6%, and 77.5%, respectively, while the EAB declined by 17.6%. Although the reason for these improvements was not explained, the deterioration was explained by agglomeration. When the nanoclay is added, this leads to worsen the ductility. After adding the filler, the G' and  $T_{50\%}$  of the LDPE increased by

10.9% and 2.6%, respectively. Interfacial relationships explained the reason for these enhancements. When the nanoclay is added, this improves the interfacial forces between the filler and the matrix.

Ghani et al. [110] investigated how the addition of tyre dust (5-25 wt% with 5% increment) influences the mechanical and thermal properties of the LDPE. The composites were manufactured and shaped with a twin-screw extruder and compression moulding, respectively. Adding the filler increased the LDPE's EMS by 10.4%, while the TSH and EAB decreased by 15.3% and 30.6%, respectively. While stronger interfacial forces explained the improvement, the deterioration was the incompatibility between the filler and the matrix. The  $T_{5\%}$  of the LDPE increased by 4.5% after adding the filler. This enhancement was also explained with better interfacial forces.

Diallo et al. [111] investigated how adding cellulose filaments (10–40 wt% with 10% increment) affects LDPE's mechanical behaviours. The composites were prepared and shaped with a twinscrew extruder and injection moulding, respectively. The addition of the filaments increased the EMS and TSH of the LDPE by 100% and 32.4%, respectively, while the EAB declined by 53.8%. Intrinsic filler properties explained the reason for the improvements. When a filler having high mechanical properties is added, this improves the mechanical properties of the composites. The reason for the decrease was explained with also the filler's inherent behaviour. In addition, the ISH of the LDPE rose by 30.1% with the addition of the filaments. This enhancement was associated with the interfacial forces, when the filler is added, this increases the interfacial forces, resulting in better ISH (Impact Strength).

Choudhury et al. [112] studied how the addition of coir fibre (10. 20, and 30 wt%) affected the LDPE/LLDPE (50:50) blend's thermal and mechanical behaviour. The milk pouches were shredded and washed with hot aqueous NaOH and cold water, before being vacuum dried. The coir fibres were first washed and vacuum dried, followed by immersion in aqueous NaOH. Afterwards, the fibres were washed with acetic acid and vacuum dried. The composites were fabricated by melt mixing and two-roll milling the milk pouch polymers, LDPE/LLDPE and coir fibres. The composites were prepared and shaped with a twin-screw extruder and a two-roll mill, respectively. The addition of the filler increased the blend's EMS, TSH, and toughness by 73.1%, 73.9%, and 2400%, respectively, while the EAB declined by 53.8%. The reason for the enhancements was explained by distribution. When the filler is added, this is distributed homogeneously, improving the stress transfer efficiency, and resulting in improvements. The intrinsic filler feature explained the reason for the deterioration. When a brittle filler is added, this increases the brittleness of the composites. The reaction start temperature (Tonset) of the blends rose by 5.4%, while the Xc declined by 31.9%. The reason for this increase was explained as the heat deflection. When a filler has a high heat distortion temperature (HDT), this improves the thermal stability of the composites. However, the addition caused short-range order in the polymer chain and led to lower crystallinity.

Key details of the above previous works are summarised in Table 5 below. The "—" symbol in the optimum content column represents that EMS and TSH are negative. In addition to that, all changes were calculated according to the optimum content. These are valid for all summary tables.

#### 5.2. PS blends and/or based composites

Gao et al. [113] tried to find how Nano-CaCO<sub>3</sub>(NCaCO<sub>3</sub>) fillers (1.3, 2.1, 4.4, and 7.4 vol%) affect the PS-based polymer composites' (PC) mechanical behaviour. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of filler increased the EMS of the PS by

15.4%, while the TSH declined by 6.4%. Interfacial forces explained the reason for enhancement. When the filler is added, this improves the interfacial forces between the filler and matrix and results in improvement. However, the reason for the decrease was not explained.

Vaziri et al. [114] determined how adding nano-silica (0.1, 0.2, 0.75, 2, 5, and 10 wt%) affects the mechanical behaviours of PS. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. Adding the filler rose the PS's EMS, TSH, FSH, and EAB by 2%, 7.9%, 4.4%, and 10.4%, respectively. Interfacial relations explained the reason for these improvements. When the filler is added, this advances interfacial forces and leads to better stress-strain transfer. However, the reason for the EAB rise was not mentioned. Generally, it is expected that when the high modulus filler is added, this decreases the EAB.

Chen et al. [115] studied how the addition of the PS (10–50 wt% with 10% increment) influences the PP's mechanical, thermal, and rheological behaviour. The all-PS additions mildly crystallised end temperature (Te) of PP 1.2%. However, the filler ratio increase declined the PP's crystallisation temperature (T<sub>C</sub>). This decrease was evidence that the addition of PS impedes the PP's polymer's chain movement to create PP crystals. SEM results demonstrated that the amount of PS composition (10-40 wt%) rose the PP/PS blends' mean diameter, and the shape of the PS particles was spherical or rounded. A separate noncontinuous phase structure was produced when the PS content reached 50 wt%. Thermal stability and the complex viscosity  $(\eta^*)$  of the PP/PS blends (up to 20 wt% PS) were relatively higher than the blends containing more than 20 wt% PS. When the PS percentage was 20 wt% or less, the tensile strength fell dramatically with increasing PS content. The TSH of the PP70/PS30 blend rose marginally when the PS ratio was 30 wt% compared to the PP80/PS20 blend, but it then declined dramatically when the PS ratio exceeded 30%. The mechanical behaviour of the PP/PS blends was remarkably affected by the PS

Gao et al. [116] investigated how the addition of PP (10–90 wt% with 10% increment) affects PS's mechanical behaviours. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of filler increased the PS's EMS and TSH by 27.1% and 15.3%, respectively. However, the reason for these enhancements was not explained.

Parameswaranpillai et al. [117] investigated how adding PP-g-MAH (2, 5, and 10 wt%) impacts the PP/PS (10–90 wt% PS with 10% increment) blends' morphology and dynamic mechanical behaviours. DMA was used to measure the blends' G', glass transition temperature  $(T_g)$ , and viscoelastic behaviour. The morphology and concentration of the blends were analysed with SEM and FTIR, respectively. PS (40 wt%) was the dispersed phase, like huge islands in the PP (60 wt%) matrix phase. The addition of PP-g-MAH (2–5 wt%) decreased the particle size of all blends compared to without the compatibilizer but 10 wt% PP-g-MAH increased the particle size. PP/PS (90/10) blends'  $T_g$  was the greatest at (106 °C) and increasing the PS ratio decreased the  $T_g$  of the blends. The neat PS'  $T_g$  was the lowest at 100 °C. All samples of SEM graphs should have been put to understand different blends' morphology.

Akbari and Bagheri [118] investigated how adding nano clay (1.5, 3, and 5 wt%) impacts the neat PS's micromorphology, deformation mechanism, and mechanical behaviour. The composites were prepared and shaped with a twin-screw extruder (L/D=20) and injection moulding, respectively. The addition of the clay increased the EMS of the PS by 2.2%, while the YSS and the EAB declined by 8.8% and 2.3%, respectively. The reason for the improvement was explained by dispersion. When the clay is dispersed homogeneously, this improves mechanical properties. The reason for the decrease was not mentioned.

**Table 5**Summary of the previous studies focused on LDPE matrix composites.

Author/s (Year) [Ref. No]	Fibre Type	Optimum Content(wt%)	Density g/cm <sup>3</sup> ) and MFI (g/10 min)	Matrix	Production Method	EMS Changes (%)	TSH Changes (%)	Other Findings
Al-oqla et al. (2021) [105]	Lignocellulosic olive leaves	20	0.919-0.65	LDPE	A twin-screw extruder and Compression moulding	218.8	9.8	The addition of lignocellulosic olive leaves decreased the EAB of the matrix by 8.3%.
Sabetzadeh et al. (2004) [106]	Thermoplastic starch	-	0.924-0.047			-44	-33.3	The addition of the starch declined the EAB and ISH of the matrix by 59.1% and 55%, respectively.
Sabet and Soleimani (2014) [108]	CNT	10	0.921-0.89			90.9	361.5	The addition of CF improved the k of the LDPE by 100%.
Ghani et al. (2012) [110]	Tyre dust	15	0.925-2.6			10.4	-15.3	The addition of the filler decreased the LDPE's EAB by 30.6% while T <sub>5%</sub> increased by 4.5%
Khattab et al. (2012) [107]	Vapour-grown CNF	3	0.920-0.27		A twin-screw extruder and Injection moulding	34.6	13.6	The addition of the filler increased the hardness of the LDPE by 7.6%, while the EAB declined by 19.4%.
Yaras et al. (2021) [11]	Carbonation sludge	20	0.925-0.075			32.7	-14.9	The addition of the sludge increased the G' of the matrix by 49.3%.
Diallo et al. (2019) [111]	Cellulose fibre	30	0.920-1			100	32.4	The addition of the filaments declined the EAB of the LDPE by 53.8%, while the ISH rose by 30.1%.
Awad (2021) [109]	Nanoclay	8	0.925-0.5		A twin-screw extruder and Hot pressing	38.6	16.8	Adding the filler increased the matrix's FMS, FSH, and ISH by 19.4%, 11.6%, and 77.5%, respectively, while the EAB declined by 17.6%. With adding the filler, the G' and T50% of the LDPE increased by 10.9% and 2.6%, respectively.
Choudhury et al. (2007) [112]	Coir fibre	30	0.922-0.55—0.919- 1	LDPE/LLDPE (50:50)	A twin-screw extruder and A two-roll mill	73.1	73.9	The $T_{onset}$ of the blends rose by 5.4%, while the $X_c$ and EAB declined by 31.9% and 53.8%, respectively.

Nayak and Mohanty [119] studied how the addition of cloisite 20A (C20A) (1–7 wt% with 2% increment) influences the neat PS' mechanical, thermal and rheological behaviours. The composites were prepared and shaped with a twin-screw extruder (31.8/20 mm ((rear/front)) and injection moulding, respectively. The addition of the filler increased the EMS, TSH, FMS, FSH, and ISH of the PS by 10%, 16.5%, 6.6%, 9.7%, and 51.8%, respectively, while the EB declined by 16.5%. These advances were explained by the polymer chains' mobility and exfoliation. When the filler is added, this restricts the polymer chains' movement and leads to enhancements. In addition, exfoliated filler contributes the same effect. However, the reason for the decrease was not explained.

Kaseem et al. [120] examined how adding wood particles (10–50 wt% with 10% increment) influences the PS's mechanical and rheological behaviours. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the filler increased the PS's EMS and TSH by 228.9% and 263.2%, respectively, while the ISH and EAB declined by 33.3% and 44.7%, respectively. Interfacial forces explained the reason for these improvements. When the filler is

added, this improves the stress-strain transfer efficiency and leads to enhancements. The reason for the decrease was explained by stress concentration. When the filler is added, this creates more stress concentration regions and declines IS.

Tiwari et al. [121] studied how the addition of Cloisite 10A (C10A) (2, 4, and 6 wt%) changes the PS/Polyphenylene oxide (PPO) (80.51 wt%)/(19.49 wt%)'s mechanical and thermal behaviour.

The composites were prepared with a twin-screw extruder and an internal mixer. The addition of the clay increased the EMS of the blend by 32%, while the TSH and the EAB declined by 50% and 40%, respectively. The reasons for both the advance and the deterioration were not explained.

Parameswaranpillai et al. [122] studied how the addition of CF (0.05, 0.1, 0.3, 0.5 and 1 wt%) affects the PP/PS (80/20 wt%) with SEBS. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of CF increased the blend's EMS and TSH by 28.2% and 3.7%, respectively, while the EAB and IS declined by 102.6% and 16.7%, respectively. While the improvements were explained with distribution, the deteriorations were intrinsic filler's properties. When the filler is

added, this leads to improvements with the help of better distribution.

On the other hand, the decrease is related to the CF having high mechanical properties rose, the brittleness of the composites, and resulting in deterioration. The E' of the blend increased by 17.6% with the addition of the CF and the polymer chains' motion explained this. When the filler is added, this restricts the polymer chains' movement and leads to enhancement. However, the reason why E" did not change was not explained.

Key details of the above previous works are summarised in Table 6 below.

#### 5.3. GNP reinforced composites

Mittal et al. [124] examined how adding GNP (2 and 4 wt%) affects the PE's mechanical, thermal and rheological behaviours. The composites were prepared and shaped with a twin-screw extruder (d=0.4~mm) and injection moulding, respectively. The addition of the GNP increased the EMS and TSH of the PE by 28.4% and 18.2%, respectively, while the EAB declined by 3.4%. The reason for the improvements was explained by dispersion. When the filler is dispersed homogenously, this improves stress-strain transfer efficiency and leads to enhancements. However, the reason for the decrease was not explained.

Dul et al. [125] determined how the addition of GNP(2–30 wt % with 2% increment) affects the mechanical behaviours of the acrylonitrile—butadiene—styrene (ABS). The composites were prepared and shaped with a twin-screw extruder and compression moulding, respectively. The addition of the filler increased the ABS's EMS by 218%, while the EAB declined by 96.4%. Interfacial relations explained the reason for the advancement. When the filler is added, this improves the interfacial interaction between the filler and the matrix and results in better mechanical properties. However, the reason for the decrease was not explained.

Chatterjee et al. [126] examined how the addition of GNP (0.05, 0.1, and 0.5 wt%) affects the mechanical behaviours of polyamide 12 (PA12). The composites were produced by a twin-screw extruder and mixed with neat PA12 via a spinning process. The addition of GNP increased the EMS and YSS of the PA12 by 369% and 143.1%, respectively. However, the reason for the improvements was not explained.

Jun et al. [127] investigated how the addition of GNP (5–20 wt% with 5% increment) influences the electrical, mechanical, and thermal properties of PP. The composites were prepared and shaped via a twin-screw extruder and compression moulding, respectively. The addition of GNP increased the EMS and TSH of the PP by 52.7% and 19.5%, respectively, while the EAB declined by 901%. The reason for this increase was related to improving stress

**Table 6**Summary of the previous studies focused on PS matrix composites.

Author Name(Year) [Ref. No]	Fibre Type	Optimum Content (wt%)	Density (g/cm <sup>3</sup> ) and MFI (g/10 min)	Matrix	Production Method	EMS Changes (%)	TSH Changes (%)	Other Findings
Gao et al. (2009) [113] Vaziri et al. (2011) [114]	NCaCO <sub>3</sub> Nano silica	2.1 0.75	1.040-8 1.050-13	PS	A twin-screw extruder and Injection moulding	15.4 2	-6.4 7.9	Adding the filler increased the PS's FSH and EAB by 4.4% and 10.4%, respectively.
Akbari and Bagheri (2016) [123]	Organoclay	3	1.040-11			4	_	The addition of the fille increased the PS's EAB by 16%.
Nayak and Mohanty (2009) [119]	C20A	5	1.050-8			10	16.5	The addition of the fille increased the PS's FMS FSH, and ISH by 6.6%, 9.7%, and 51.8%, respectively, while the EAB declined by 16.5%.
Kaseem et al. (2017) [120]	Wood particle	30	1.050-7			228.9	263.2	Adding the filler decreased the PS's ISH and EAB by 33.3% and 44.7%, respectively.
Parameswaranpillai et al. (2016) [122]	CF	0.3	1.050-6.5			28.2	3.7	Adding the filler decreased the PS's ISH and EAB by 16.7% and 102.6%, respectively, while the E' increased by 17.6%.
Parameswaranpillai et al. (2015) [117]	PS	60	1.050-6.5	PP		39.9	-10.5	Adding the filler increased the PS's FMS by 43.3%, while the FSF EAB, and IS declined by 5%, 36.3%, and 81.8%, respectively.
Gao et al. (2010) [116]	PS	50	1.050-8.5			27.1	15.3	-
Chen et al. (2021) [115]	PS	30	1.040-10		A twin-screw extruder and Compression moulding	23.1	-10.2	The addition of the fille increased the PS's FMS and EAB by 4.8% and 10.4%, respectively, while the FSH and ISH declined by 5.2% and 30.6%, respectively.
Tiwari et al. (2008) [121]	PPO	4	_	PS	A twin-screw extruder	32	-50	The addition of the fille decreased the PS's EAE by 40%.

transfer efficiency from the matrix to the filler with the existence of the GNP. The  $T_{5\%}$  of the PP increased by 11.5% with the addition of the GNP. The distribution of the heat explained the reason for this improvement. When the GNP is added, it provides better heat distribution in the composites, resulting in higher thermal degradation temperatures.

Yang et al. [128] studied how the addition of GNP (1, 3, 5, 8, and 10 wt%) affects the mechanical and thermal behaviour of poly (arylene ether nitrile). The composites were manufactured and shaped with a twin-screw extruder (d = 14 mm) and injection moulding, respectively. The addition of GNP increased the neat polymer's EMS, EAB, FMS, FSH, and ISH by 9.5%, 8%, 11.8%, 13.3%, and 18.3%, respectively, while the TSH declined by 66.7%. Increasing interface forces explained the improvement. When the GNP is added, this improves the interfacial relationship between the matrix and the filler. The reason for the deterioration was explained that the addition of the GNP declined the  $X_{\rm C}$  of the polymer and resulted in a decrease in the EAB.

Ghani et al. [129] investigated how adding GNP (0.1, 0.3, 0.5, and 1 wt%) affects the mechanical behaviours of PLA. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the GNP increased the TSH and the EAB of the PLA slightly by 17% and 0.4%, respectively, while the EMS decreased by 21.7%. The enhancement was explained by dispersion as well as interaction. When the GNP is added, this addition provides better dispersion and a much more efficient interaction between the matrix and the filler. They also stated that the deterioration was related to the polymer chain motion. When the GNP is added, this leads to the relaxation of the polymer chains and results in lower EMS values.

Inuwa et al. [72] determined how adding GNP (3 wt%) influences polyethylene terephthalate's thermal and mechanical behaviours. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the GNP increased the polyethylene terephthalate's EMS, TSH, FMS, and FSH by 154.5%, 60.7%, 22.2%, and 3.2%, respectively, while the EAB decreased by 1.3%. These improvements were explained by the interaction between the filler and the matrix. When the GNP is added, the addition provides better interaction. The worsening was associated with the polymer chain motion. When the filler is added, it restricts the motion and results in a decrease. The Tonset and Tmax of the matrix increased slightly by 2.3% and 1.6%, respectively, with the addition of the GNP. The reason for the enhancements was associated with stability. When the filler is added, it prevents the gaseous products from going through the matrix and results in thermal improvement.

Pour et al. [130] determined how the addition of GNP (1, 3, and 5 wt%) affects the mechanical, thermal, and morphological properties of the polycarbonate (70 wt%)/ABS (30 wt%). The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the GNP increased the matrix's EMS, TSH, FMS, FSH, and EAB by 29.4%, 15%, 32.8%, 11.6%, and 94%, respectively. These enhancements were explained by an interaction between the filler and the matrix and dispersion. When the GNP is added, this provides better interaction and dispersion and results in improvement. The ISH of the matrix declined by 91.4% with the addition of the filler. This decline was explained by rigid filler. When the rigid filler is added, it decreases the ductility and ISH. The addition of the filler increased the matrix's T<sub>50%</sub> by about 7%. However, the reason for this enhancement was not explained.

Wijerathne et al. [131] analysed how the addition of GNP (1, 3, 5, 8 and 10 wt%) affects polycarbonate's (recycled) thermal and mechanical behaviours. The composites were prepared and shaped with a twin-screw extruder (d=24 mm) and injection moulding,

respectively. The addition of the GNP increased the polycarbonate's EMS and TSH by 21.7% and 5%, respectively while the EAB decreased by 24%. These enhancements were explained by polymer chains' limitation. When the filler is added, this restricts the chain movement and results in mechanical advancements. The other reasons can be related to the interfacial adhesion and good distribution. The reason for decline was explained by content. When the low amount of the filler is added, this cause ductile behaviour of the composite. However, when the high amount of the reinforcement is added, this leads to the brittle behaviour of the composite.

Key details of the above previous works are summarised in Table 7 below.

#### 5.4. CF reinforced composites

Yang et al. [132] examined how the addition of CF (10 wt%) affects Polyether ether ketone (PEEK)'s mechanical behaviours. The composites were prepared with a twin-screw extruder (d = 20 mm). The addition of the filler increased the PEEK's EMS, TSH, FMS, and FSH by 162.9%, 165.7%, 153.3%, and 164.8%, respectively, while the EAB declined by 97.9%.  $X_{\rm c}$  explained the reason for these improvements. When the filler is added, this leads to an increase in  $X_{\rm c}$  and results in mechanical enhancements. However, the reason for the deterioration was not explained.

Cho et al. [133] evaluated how the addition of CF (5, 10, 20, and 30 wt%) affects the mechanical and thermal behaviours of polyketone (PK). The composites were prepared and shaped with a twin-screw extruder (d = 16 mm) and injection moulding. respectively. The addition of the filler increased the PK's EMS and TSH by 520% and 87.5%, respectively, while the EAB declined by 98%. Interfacial relations explained the reason for these advancements. Adding the filler provides better stress-strain transfer between the filler and the matrix and improves mechanical properties. The reason for the decrease was explained by brittleness. When the brittle filler is added, this increases the structure's brittleness and results in a more brittle structure. The PK's melting enthalpy change ( $\Delta H_{\rm m}$ ) decreased by 47.6% after adding the filler. The reason for this advancement was explained by restriction. When the filler is added, this limits polymer chains' motion and results in the lower enthalpy change.

Ren et al. [134] examined how the addition of CF (0.5, 1, and 3 wt %) affects the mechanical and thermal behaviour of the UHMWPE/LDPE (30:70 wt%) polymer blends. The composites were prepared with a twin-screw extruder (d = 18 mm). The EMS and TSH of the blends improved by 15.3% and 37.5%, respectively, with the addition of the CF. The intrinsic filler's property explained the reason for these enhancements. When the filler having high mechanical properties is added, this contributes to better mechanical performance. The E' of the blend increased by 9.8% with the addition of the filler. This enhancement was explained by limiting the mobility of chain rises the E'.

Vivekanandhan et al. [135] determined how the addition of CF (5, 10, 15, 20, and 30 wt%) influences the poly(trimethylene terephthalate)'s mechanical and thermal behaviours. These composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of the filler increased the matrix's EMS, TSH, FMS, FSH, and ISH by 600%, 120%, 450%, 116.2%, and 35.1%respectively. The reason for these improvements was explained by interfacial adhesion. When the filler is added, this leads to better adhesion between the filler and the matrix and results in greater mechanical performance. The G' of the polymer increased by 583.3% with the addition of CF, while the tan  $\delta$  decreased by 84.6%. Polymer chains' motion explained the reason for these changes. When the filler is added, this leads to restricting

**Table 7**Summary of the previous studies focused on GNP reinforced composites.

Author Name/Year/ Ref.	Fibre Type	Optimum Content(wt%)	Matrix	Production Method	EMS Changes (%)	TSH Changes (%)	Other Findings
Mittal et al. (2016) [124]	GNP	4	PE	A twin-screw extruder	29.4	18.2	In the nanocomposites, there was no change in the polyethylene diffraction signals with the addition of GNP. The EAB of the composite increased 3.4% with the addition of 4 wt% GNP.
Dul et al. (2018) [125]		30	ABS		218	-	The addition of GNP decreased by 96.4% EAB of the ABS and tensile energy to break with 97.1%. The percolation threshold of the ABS was observed at 7.3 wt% GNP.
Chatterjee et al. (2013) [126]		0.5	PA 12		369	_	The addition of the filler increased the YSS of the matrix by 143.1%.
Jun et al. (2018) [127]		20	PP	A twin-screw extruder and Injection moulding	52.7	19.5	The addition of GNP EAB declined by 901%. The $T_{5\%}$ of the PP increased by 11.5% with the addition of the GNP.
Yang et al. (2014) [128]		3	Poly (arylene ether nitrile)	, ,	9.5	-66.7	The addition of GNP increased the neat polymer's EAB, FMS, FSH, and ISH by 8%, 11.8%, 13.3%, and 18.3%, respectively.
Ghani et al. (2021) [129]		0.3	PLA		-21.7	0.4	The addition of the GNP slightly increased the EAB of the PLA by 17%.
Inuwa et al. (2016) [72]		3	PET		154.5	60.7	The addition of the GNP increased the FMS and FSH of the polyethylene terephthalate by 22.2% and 3.2%, respectively, while the EAB decreased by 1.3%. The T <sub>onset</sub> and T <sub>max</sub> of the matrix increased slightly by 2.3% and 1.6%, respectively, with the addition of the GNP.
Pour et al. (2016) [130]		3	Polycarbonate/ABS (70:30)		29.4	15	The addition of the GNP increased the matrix's FMS, FSH, and EAB by 32.8%, 11.6%, and 94%, respectively. The ISH of the matrix declined by 91.4% with the addition of the filler. The addition of the filler increased the matrix's $T_{50\%}$ by about 7%.
Wijerathe et al. (2022) [131]		5	Polycarbonate		21.7	5	When the addition of GNP increased $T_{max}$ by 2.2%, decreased $T_g$ by 1.2%.

polymer chains movement capability and results in better performance.

Li, S. and Li, D [136] investigated how the addition of CF (4 and 8 wt%) affects the UHMWPE/charcoal powder (70:30 wt%) blend's dynamic mechanical and tensile behaviours as well as morphology. These composites were prepared with a twin-screw extruder (d = 0.4 mm). The addition of CF increased the TSH and EMS of the blend by 23.6% and 24.2%, respectively, while decreased EAB by 9.2%. Polymer chains' motion explained the reason for these enhancements. When the filler is added, this limits polymer chains' movement and results in better mechanical properties. However, the reason for the decrease was not explained. The G' of the blend increased by 150% with the addition of the filler, while the tan  $\delta$  decreased by 50%. This rise and decline resulted from stress-strain transfer improvement between the matrix and the reinforcement and limiting the movement of the chains.

Liang et al. [137] determined how the addition of CF (5–20 wt% with 5% increment) affects mechanical and thermal behaviours and crystallinity of poly(butylene succinate). The composites were prepared and shaped with a twin-screw extruder (d = 21.7 mm) and injection moulding, respectively. The addition of the CF increased the polymer's TSH, EMS, and ISH by 140.9%, 496.1%, and 103.6%, respectively, while the EB decreased by 499.2%. EMS's improvement was explained by restricting the polymer chains' movement, while the TSH increase was the efficiency of the interface between the matrix and the reinforcement. In addition, increasing the loading ratio by being higher than 15 wt% caused agglomeration and increased the number of stressed concentrated regions. As a result, this leads to straightforward crack propagation because it requires lower energy. However, the reason for the EAB's decrease was not explained.

Lin et al. [138] investigated how adding CF (10, 15, and 20 wt%) influences the PEEK's wear and mechanical behaviours. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. While the TSH and EMS of the PEEK increased with the addition of CF by 78.6% and 281.1%, the EAB decreased by 19.8%. However, the reason for these changes was not explained.

Wang and Ying [139] examined how the addition of CF (5, 15, and 25 wt%) influences the tensile, thermal, and rheological behaviours of PP. The composites were prepared and shaped with a twin-screw extruder (d = 0.4 mm) and an injection moulding, respectively. The addition of CF increased the PP's EMS and TSH by 272.9% and 119.1%, respectively. The reason for these advancements was not explained. The E' and E" of the PP increased by 57.9% and 17.7% with the addition of the filler. Polymer chains' motion explained the reason for these improvements. When the filler is added, this limits polymer chains' movement and leads to better properties.

Phua and Ishak [140] examined how adding CF (40 wt%) affects the polycarbonate's thermal and mechanical behaviours. The composites were prepared and shaped with a twin-screw extruder (d = 31.2 mm) and injection moulding, respectively. The addition of CF increased the polycarbonate's EMS, TSH, FMS, and FSH by 350%, 43.9%, 1033.3%, and 87.5%, respectively. These rises were explained by improving the interaction between the filler and the matrix. The E' of the polycarbonate increased by 480% with the addition of the CF. Polymer chains' motion explained this enhancement. When the filler is added, this limits the chains' mobility and leads to advancement.

Junaedi et al. [141] investigated how the addition of CF (15 and 35 wt%) affects the mechanical behaviours of the PP. The

composites were manufactured and shaped with a twin-screw extruder and injection moulding, respectively. The addition of CF rose EMS and TSH of the PP by 535.6% and 41.6%, respectively, while the EB decreased by 483.7%. However, the reason for these changes was not explained.

Li [142] determined how the addition of CF (10–40 vol% with 10% increment) influences the tribological and mechanical behaviours of the polytetrafluoroethylene. The composites were prepared and shaped with a twin-screw extruder and injection moulding, respectively. The addition of CF increased the EMS and TSH of the

polytetrafluoroethylene by 140% and 148%, respectively, while the EB declined by 99.6%. However, the reason for the changes was not explained.

Zhong et al. [143] determined how the addition of CF (10 wt%) influences the mechanical and wear properties of the PEEK. The composites were prepared and shaped by a twin-screw extruder and injection moulding, respectively. While the addition of the CF increased EMS, TSH, and ISH of the PEEK by 154.1%, 47.8%, and 35.4% respectively, the EAB declined by 18.72%. However, the reason for these changes was not explained.

Table 8
Summary of the previous studies focused on CF reinforced composites.

Author Name (Year) [Ref. No]	Fibre Type	Optimum Content (wt%)	Matrix	Production Method	EMS Changes (%)	TSH Changes (%)	Other Findings
Yang et al. (2021) [132]	CF	10	PEEK	A twin-screw extruder	162.9	165.7	Increasing $X_c$ increased by EAB (97.9%), EMS (157.1%), FMS (153.3%) and FSH (164.8%). Heat temperature increased by $X_c$ (21–35) with 300 °C.
Ren et al. (2007) [134]		3	LDPE/UHMWPE (70:30)		15.3	37.5	The addition of CNF decreased by the X <sub>c</sub> of UHMWPE/LDPE 20.3% with 0.5CNF wt%. The CNF did not considerably affect the damping factor and T <sub>g</sub> . The CNF addition increased the thermal conductivity coefficient (k) by 39.5%.
Vivekanandhan et al. (2012) [135]		30	Poly (trimethylene terephthalate)		600	120	The G' of the polymer increased by 583.3% with the addition of CF, while the tan $\delta$ decreased by 84.6%. The FMS and FSH of the neat polymer were increased by 450% and 116.2% with the addition of CF, respectively. The addition of CF increased by 35.1% of the polymer's ISH.
Li, S and Li, D (2014) [136]		4	UHMWPE/charcoal powder (70:30)		24.2	23.6	The G' of the blend increased with increasing the CF ratio by 150%, while $\tan \delta$ decreased by 50%.
Cho et al. (2019) [133]		30	PK	A twin-screw extruder and Injection moulding	520	87.5	The EAB and $\Delta H_m$ decreased by 98% and 47.6%, respectively, with the addition of CF, while k increased by 300%.
Liang et al. (2015) [137]		15	Poly (butylene succinate)		496.1	140.9	The addition of the CF increased the ISH of the polymer by 103.6% respectively, while the EAB decreased by 499.2%,
Lin et al. (2011) [138]		20	PEEK		281.1	78.6	The EAB of the PEEK decreased with the addition of the CF by 19.8%.
Wang and Ying (2012) [139]		25	PP		272.9	119.1	The E' and E" of the PP increased by 57.9% and 17.7% with the addition of the filler.
Phua and Ishak (2010) [140]		40	Polycarbonate		350	43.9	Adding CF increased the FMS and FSH of the polycarbonate by 1033.3% and 87.5%, respectively. The E' of the polycarbonate increased by 480% with the addition of the CF.
Junaedi et al. (2018) [141]		35	PP		535.6	41.6	The EAB of the PP decreased by 483.7% with the addition of the filler.
Li (2010) [142]		30			140	148	The EAB of the PP decreased by 99.6% with the addition of the filler.
Zhong et al. (2010) [143]		10	РЕЕК		154.1	47.8	While the addition of the CF increased ISH of the PEEK by 35.4%, the EAB declined by 18.72%.
Li et al. (2014) [144]		20	Polyamide 6		64.1	50.3	The EAB declined by 80.7% with the addition of the filler.

Li et al. [144] analysed how the addition of CF (10 and 20 wt%) affects the mechanical behaviours of polyamide 6. The composites were prepared and shaped by a twin-screw extruder (d = 21.7 mm) and injection moulding, respectively. The EMS and TSH of the polyamide 6 rose with the addition of CF by 64.1% and 50.3%, respectively, while the EAB declined by 80.7%. These changes were explained by adding the CF improved interfacial efficacy between the filler and the matrix. As a result of this, the EMS and TSH were enhanced. Also, increasing the filler ratio limits the polymer chains' motion and results in cracks. These cracks lead to a decrease in the EAB of the composite.

Chen et al. [69] determined how the addition of recycled CF (5–25 wt% with 5% increment) influences the mechanical and thermal behaviours of poly(butylene terephthalate). The composites were prepared by a twin-screw extruder (d = 30 mm). The addition of the recycled CF increased EMS, TSH, FMS, and FSH of the poly (butylene terephthalate) by 481.8%, 248.8%, 500%, and 169.8%, respectively. Interfacial forces explained the reason for these advancements. Adding the filler improves the interfacial forces between the filler and the matrix and leads to better mechanical properties. The HDT of the poly (butylene terephthalate) rose by 307.7% after adding the filler. The reason for this enhancement was explained by mechanical improvement, especially FSH, by referencing the literature. However, the mechanism of this improvement was not mentioned.

Key details of the above previous works are summarised in Table 8 below.

#### 6. Discussion

GNP and CF are types of widely popular fillers in the composites manufacturing while polymer materials such as LDPE and PS are among the widely used matrix materials. However, there are still a lot of issues that need to be resolved to reach the maximum potential of these nanocomposites. For instance, twin-screw extruders are commonly used in industry to produce composites particularly due to their good mixing performance [145]. This production technique can affect dispersion, particle size, surface area, exfoliation, and distribution behaviour of composites adversely depending on content [72,146]. However, it is difficult to control these parameters during manufacturing. The easiest and most effective way can relatively be to control the content ratio to reach optimum mechanical properties. Therefore, a systematic

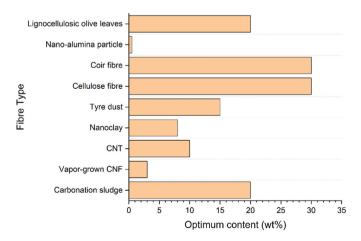


Fig. 20. Optimum content for different fillers with LDPE.

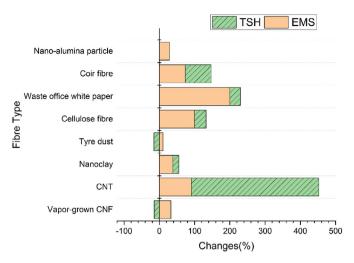


Fig. 21. EMS and TSH changes of LDPE with different filler.

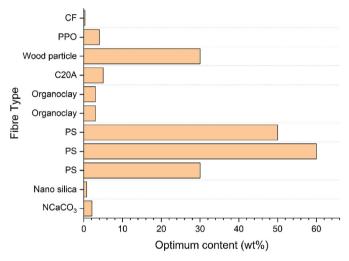


Fig. 22. Optimum content for different fillers with PS.

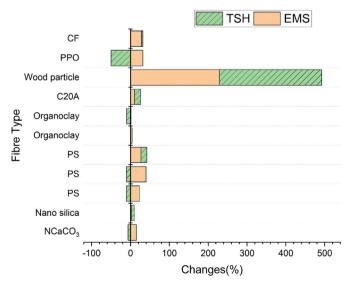


Fig. 23. EMS and TSH changes of PS with different fillers.

review was conducted to analyse the effect of polymers and/or blends' (LDPE and PS) and reinforcements' (GNP and CF) content ratio on the EMS and TSH.

One of the limitations of this study can be just focusing on one production technique (melt intercalation) because there are two more common techniques: solvent casting and in-situ polymerisation. The in-situ polymerisation technique can offer better dispersion, distribution, and exfoliation behaviour than melt intercalation, which is relatively better than solvent casting. The mechanical properties of composites can also be enhanced further with the functionalization route with the polymerisation method [147]. The melt mixing method was chosen because of its widespread use in industry. In the following sections, LDPE, PS, GNP and CF will be analysed regarding the mechanism behind the EMS and TSH changes. The other limitation can be GNP's price. GNP is not so much expensive compared to CNT but still expensive. When the optimum content figures are analysed, it is clear that the maximum amount used was 30 wt% GNP.

#### 6.1. Low density polyethylene (LDPE)

A comparison of the optimum contents and the EMS and TSH changes associated with LDPE were given in Figs. 20 and 21, respectively.

It can be seen that the optimum content of the filler varies depending on the used filler type. The optimum content effect on the EMS and TSH are various. The addition of filler generally changes the EMS and TSH of the blends and composites. When the filler is added, this can increase EMS and/or TSH. This addition usually increases EMS, which is also related to the intrinsic filler properties, dispersion of filler and filler, and matrix adhesion [128]. When a high modulus filler is added, the modulus is typically enhanced. On the other hand, adding filler generally increases the EMS but not the TSH. For example, when the vapour-grown CF was added to LDPE, it increased EMS by 3% but decreased TSH by 1.7%. This decrease was explained by crack formation. When the filler is added, this can create new micro-crack regions and results in lower TS [148]. While the filler's intrinsic modulus explained the advancement, the decrease was explained by poor interfacial relation [110], which was supported by Khattab et al. [59]. The typical optimum content was 20 and 30 wt% among the filler. It can

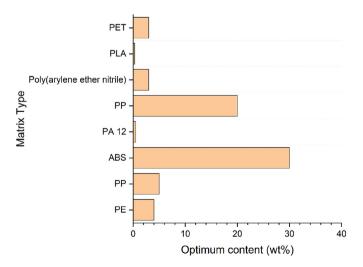


Fig. 24. Optimum content for different fillers with GNP.

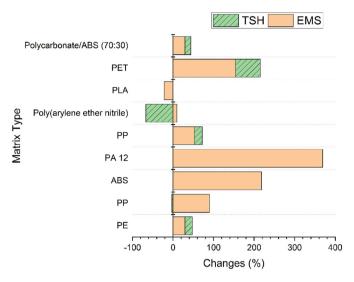


Fig. 25. EMS and TSH changes of GNP with different fillers.

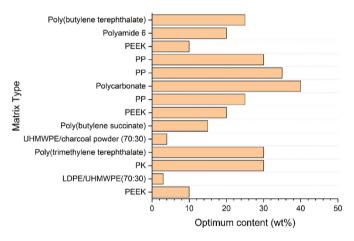


Fig. 26. Optimum content for different fillers with CF.

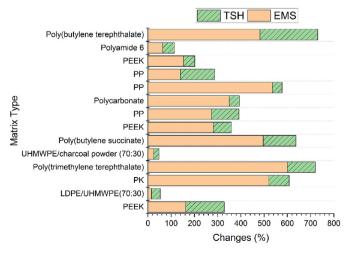


Fig. 27. EMS and TSH changes of CF with different fillers.

be concluded that the mixture of materials should be optimised by content ratio to reach optimum mechanical properties.

#### 6.2. Polystyrene (PS)

A comparison of the optimum contents and the EMS and TSH changes associated with PS were given in Figs. 22 and 23 and respectively.

The optimum content of filler was diverse. Unlike LDPE, the typical optimum content of the filler with PS matrix was between 2 and 6 wt%. It can be deduced from Fig. 22 that the filler affected the EMS and TS of the PS at different rates. While some fillers just changed the EMS (i.e., organoclay) or the TSH (i.e., nano silica), other fillers affected both EMS and TSH. While EMS increase was explained by adhesion between the matrix and filler, the TSH variations were associated with intrinsic filler properties [114,123]. Adding wood particles provided the most contribution among the filler and increased EMS and TSH by 228.9% and 263.2%, respectively. These advancements were explained by good dispersion and adhesion between the filler and matrix [120].

#### 6.3. Graphene nano-platelet (GNP)

A comparison of the optimum contents and the EMS and TSH changes associated with GNP were given in Figs. 24 and 25, respectively. The optimum content of GNP with the matrix was varied, and the typical optimum content was around 4 wt% among the filler. It can be deduced from the above figure that the filler affected the EMS and TSH of the composites at different rates. While some just changed the EMS (i.e., ABS), others affected both the EMS and TSH (i.e., PET). While EMS improvement was explained by dispersion of the filler and stress-strain transfer efficiency between the filler and matrix, TSH (i.e., poly (arylene ether nitrile)) deterioration was attributed to  $X_{\rm c}$  variations [126,128]. The addition of PET showed the most contribution among the filler and increased EMS and TSH by 154.5% and 60.7%, respectively. This was explained by good dispersion and restriction of the polymer chain between the filler and matrix [72].

#### 6.4. CF (carbon fibre)

A comparison of the optimum contents and the EMS and TSH changes associated with CF were given in Figs. 26 and 27, respectively. The optimum content of CF with the matrix was diverse and was between 20 and 30 wt%. It can be deduced from the above figure that the filler affected the EMS and TSH of the composites at different rates. Adding the filler increased all matrixes' EMS and TSH by at least 15.3% and 37.5%, respectively. While the CF contributed to improving the poly (trimethylene terephthalate) matrix's EMS and TSH the most with increases of 600% and 120% respectively, the lowest was LDPE/UHMWPE (70:30) matrix's EMS and TSH with increases of 15.3% and 37.5% respectively. The most outstanding contribution was explained by good dispersion and intrinsic filler properties, while the lowest was interfacial adhesion between the filler and matrix [134,135].

#### 7. Conclusions and future work

This work summarises the current state of composites which consist of CF and GNP reinforced PE and PS matrices. It is necessary to make crucial research accessible and useable for researchers to achieve high mechanical properties such as EMS. The mechanisms that affect the properties of composite materials, such as filler surface area, dispersion, and content, are described in this review. It is necessary to increase the EM and TSH of currently available

polymers, especially LDPE and PS, using fillers with high mechanical properties; GNP and CF have been demonstrated to achieve this. Notably, even for one polymer, such as LDPE, there are significant differences among the data sets.

However, tendencies can be noticed when comparing the same polymer with a different filler or the same filler with a different polymer matrix. While one type of starch increased the EMS of LDPE by 218.8%, the other decreased by 44%. One type of particle increased the EM of PS by 228.9%, while the other rose by 2%. While GNP rose by 154.5% of the EMS of the polymer, it declined by 21.7% of the other polymer. CF increased by 600% of the polymer while it rose by 15.3% of the blend. Dispersion of filler directly affects the mechanical properties of composites. The proportion of fillers in the composite affects the EMS and TSH values; both values can be increased beyond the original pure polymer or blend value, which was previously unthinkable. For example, CF, when the CF (35 wt%) was added to the PP matrix, it increased by 535.6% of the matrix's EMS. When the CF (30 wt%) was added to the same matrix, it rose by 140%. In both examples, a twin-screw extruder and injection moulding were used.

The typical optimum content depends on both matrix and filler types. The typical optimum content was 20 and 30 wt% among the LDPE filler and between 2 and 6 wt% for PS. The optimum content of GNP with the matrix was varied, and the typical optimum content was around 4 wt% among the filler. The optimum content of CF with the matrix was diverse and was between 20 and 30 wt%.

Adding fillers in LDPE affected only mechanical but also thermal properties. The affected properties were  $T_{5\%}$ ,  $T_{50\%}$ ,  $T_{onset}$ , and G'. For example, when the carbonation sludge was added to LDPE, it increased the G' of the matrix by 49.3%.

Adding fillers in LDPE affected only mechanical properties of PS. These were FMS, FSH, and ISH, in addition to EMS and TSH. For example, when the C20A was added in PS, this rose ISH of the matrix by 51.8%.

Most of the GNP addition affected not only mechanical but also thermal properties. These were  $T_{5\%}$ ,  $T_{50\%}$ ,  $T_{onset}$ , and  $T_{max}$ . For instance, when the GNP was added to PP matrix, it increased  $T_{5\%}$  of the matrix by 11.5%.

Adding CF in different matrixes affected not only the mechanical properties but also thermal properties and crystallinity. These were  $T_g$ , E', E'',  $\Delta H_m$ ,  $\tan \delta$ , and  $X_c$ . For example, when the CF was added to PP, it increased by 57.9% of the matrix.

Several key issues associated with CF and GNP reinforced PE and PS were identified in this review. At the forefront is the high manufacturing cost of CF, which is restraining their widespread application in modern engineering applications. Therefore, optimising, and innovating CF manufacturing processes has become an active field of research and will continue to be so in the upcoming years. Another key challenge is the characterisation and control of the filler content within a composite. Factors such as filler surface area, size, distribution, and percentage have a strong influence on the final properties of the composite part. However, due to their small length scales, quantifying and visualising the filler content accurately is not straightforward. Understanding the influence of the filler presence within the matrix is a key requirement for reducing manufacturing costs and process optimisation. Furthermore, these optimal filler-matrix ratios are highly material dependent, hence a broad range of material combinations need to be analysed to make informed decisions. Therefore, more research needs to be conducted on filler distribution visualisation, statistical analysis of micrographs and the relationship between fillers and mechanical properties. Furthermore, given the recent drive towards sustainability, much work is being carried out on substituting synthetic engineering materials with bio-sourced alternatives. One such example is Lignin-based CFs, which currently show

limited mechanical properties. Research needs to be conducted to improve the mechanical properties of these CFs such that in the future they can be readily substituted for conventional engineering materials.

The final aim of industries is the large scale commercialisation of these composite materials, where the constituents are chosen to maximise the physical properties. To achieve this, continuous research is necessary to overcome the previously mentioned challenges and to produce high quality CF/GNP reinforced PE/PP parts.

#### **CRediT authorship contribution statement**

**Bilal Gayretli:** Data curation, Formal analysis, Investigation, Methodology, Software, Writing — original draft, Writing — review & editing. **Rajinth Shanthar:** Formal analysis, Investigation, Methodology, Writing — original draft, Writing — review & editing. **Tahsin Tecelli Öpöz:** Conceptualization, Data curation, Methodology, Writing — original draft, Writing — review & editing. **Chamil Abeykoon:** Conceptualization, Data curation, Formal analysis, Investigation, Project administration, Supervision, Writing — original draft, Writing — review & editing.

#### Declaration of interests

The authors declare that there is no conflicts of interest.

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