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An application-oriented roadmap to select polymeric nanocomposites for advanced applications

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Abstract: Polymeric nanocomposites are the materials that incorporate nanosized particles or fibers into the polymer matrixes. The result of the addition of nanoparticles/fibers is a drastic improvement in properties that may include mechanical, electrical, thermal conductivity or even the acoustical properties of the materials. Polymer nanocomposites have spawned huge interest for the development of high-performance products such as lightweight sensors, windmill blades, thin-film capacitors, high performance batteries, flame retardant products, biodegradable and biocompatible materials suitable for areas ranging from packaging to biomedical applications, etc. The field of polymer nanocomposites has been at the forefront of research in the polymer community for the past few decades and an extremely large number of scientific papers have been published. Nevertheless an application oriented guidelines for selecting the ecofriendly nanocomposites for advanced industrial applications are missing in the state-of-the-art. This review paper summarizes the current state-of-the-art polymeric nanocomposite highlighting prominent nanofillers categorized based on their applications, origins, dimensional characteristics, etc. Each filler type contains a short description of its main feature together with the most relevant and potential applications. To address the global concern about non-degradable wastes, novel green alternatives for each nanofiller type are discussed. Special attention is also given to the biocompatibility properties of the composites. This paper provides a state-of-the-art road map to select multifunctional polymeric nanocomposites from huge varieties for advanced industrial applications.

Keywords: Nanocomposites, Nanofillers, Polymer, Biocompatibility

1. Introduction

Polymer nanocomposites refer to polymer matrixes reinforced with fillers of different shape (e.g., platelets, fibers, spheroids, etc.) that contain at least one dimension in the range less than 100 nm [1]. As the result of comprising different materials, structures, and compositions in the nanocomposites, different properties suitable to various applications can be expected from them [2]. Therefore, multifunctional materials have been one of the main sources of interest for nanocomposite realm. The primal nanocomposites in late 19th and early 20th century such as carbon black, silica and, precipitated calcium carbonate etc. were recognized for their reinforcement capabilities [3]. The interest rose significantly from the 1950s when the first nanoclay composite was described in a patent from the National Lead Company of the USA. However a Toyota patent in 1988 on polyamide (PA) nanocomposites has initiated their wide-spread commercial applications in the industries [4]. The exponential growth of the polymeric nanocomposites owes to the discovery of an allotrope of carbon which is called C60 or Buckyball. This discovery paved the road for discovery of carbon nanotubes. Some of the most major improvements achieved from introduction of nanofillers into polymeric matrixes are: enhancement of mechanical properties [5]; improvements in thermal and electrical properties from the incorporation of carbon nanotubes [6], flame retardant properties in case of the introduction of nanoclays [7] and metal hydroxides [8], thermal stability, gas permeability, etc. The combination of carbon nanotubes (CNTs) has also raised significant interest in the development of shape memory materials due to the electrical properties of these nanocomposites [9]. Shape memory materials have the capability of being deformed into a temporary shape with the help of an external stimulus. The electrical properties of CNTs allow using electrical current to control smart actuators for many different applications. Moreover, the organic origins of some

nanofillers such as nanocellulose, allow the usage of these materials for medical purposes due to their biocompatibility. With the advent of the concepts like the Cyber-Physical Systems, the Internet of Things, the Big Data, E-health monitoring etc. the demand for sensors, actuators, smart electronics, smart materials etc. will be drastically increased. So the polymer based nanocomposites can play ever bigger role in the future for advanced and specialized industrial applications.

A huge number of fundamental researches have been made on polymer based nanocomposites, but the applications of polymer based nanocomposites, application oriented selection of polymer based nanocomposites, eco-friendly selection of nanocomposites etc. are not adequately addressed by the state-of-the-art literature. As application of polymeric nanocomposites is increasing in different industries, the interest of using these multifunctional materials as a viable alternative to traditional materials is soaring sharply. So, the aim of this review paper is to provide a guideline for the selection and application of some important nanomaterials and their corresponding composite materials. In order to provide a better overview of the progress in this field, nanomaterials and composites were categorized based on their geometrical characteristics and applications. Furthermore, green alternatives of the well-known nanocomposites have been introduced to reduce or eliminate the arising pollution from production and waste of the composite materials. Fig 1 categorise the important types of nanofillers and presents the subcategories of each type. Dimensional and shape characteristics are highlighted in different shades of grey; main application related properties of the nanofillers are highlighted in colored dashed lines.

Fig. 1 - Types of nanofillers and the subcategories of each type. Dimensional and shape characteristics highlighted in different shades of grey; main application and properties highlighted in colored dashed lines.

2. Nanofillers

Nanofiller is the common term used for the additives having special structures and compositions, used in a small proportion together with the polymer matrix to generate a large impact on the final properties of the composite materials. New combinations of nanocomposites and hybrids have been developed in the recent past to achieve desired properties for applications such as humidity sensors [10], thermo responsive shape memory polymers [11], piezoresistive materials [12], etc. Besides the different combinations of properties based on the filler type and matrix used, the synthesis and preparation of these materials plays an important role in achieving

desired properties of the materials [13]. In this section, the focus is placed on the combination of different nanofillers and matrixes for some advanced applications; the synthesis and preparation methods will also be briefly mentioned. Nanoparticles or nanofillers have been categorized in six different groups based on their origins: nanoclays, carbon, nano-oxides, metal hydroxides, nanocarbides, organic and other: including metal particles, silica oxides etc. (as shown in Figure 1). The incorporation of these nanofillers in different polymer matrixes can have a great impact on the mechanical, electrical or thermal properties but at the same time that can also have a negative impact on the environment. With the likelihood of these materials to end up in landfills, surface water and wastewater treatment, it is extremely important to study the biodegradability of the composites and to find green alternatives that enhanced the biodegradation of the materials. With the global concern about the waste problems caused by non-degradable plastics and composites, this paper will also present a list of novel alternatives at the end of each nanofiller section which can be greener options to the commonly used nanocomposites. The potential biodegradable polymer matrixes suitable for the green nanofillers such as the aliphatic polyesters: poly (lactic acid) (PLA), Poly(caprolactone) (PCL), poly (p-dioxanone) (PPDO), poly(butyl succinate) (PBS), Poly (hydroxyalkanoates) (PHAs) etc. are presented and discussed in reference :[14].

2.1 Nanoclays

The most widely studied and commercially used nanofillers are nanoclays produced from natural clay minerals. Among different nanoclay types, the most common nanoclay is montmorillonites (MMTs), often called smectite or bentonite. The other nanoclay groups are kaolinite, illite, chlorite and halloysite [1]. Nanoclays are formed by stacked metal silicate layers which can be penetrated by low molecular weight substances, e.g. water, leading to swelling [8]. Depending on the degree of penetration into the interlayer, nanoclays can be divided into: intercalated (expansion of the layers less than 20-30Å), and exfoliated (expansion of the layers between 80-100Å or more) [1]. The best properties are given by the exfoliated nanoclays.

Kaolinite is a member of the nanoclay family which is the most abundant mineral on the earth. Kaolinite is a 1:1 layered dioctahedral aluminosilicate composed of alternating sheets of “SiO₂” and “AlO₆”[15]. MMT/ smectite / bentonite, characterized by a 2:1 sheet-structure, is the most widely used in the preparation of nanocomposites, since even in small quantities inside the polymer matrix, it has a big impact on the mechanical and thermal properties [16]. Illite and especially halloysite have received special attention in the recent years. Halloysite nanotubes (HNTs) are composed of silica and alumina with a structure similar to carbon nanotubes. However their biocompatibility and low price make them attractive replacements to CNT with promising applications in the medical industry e.g. drug delivery, implants, prostheses applications, etc.

Tables 1-4 represent the research studies made by the authors on the state-of-the-art literatures (from 2010 to 2017) on the nanoclay fillers. The polymeric matrixes include various types of thermoplastics, thermosets and elastomers. In order to facilitate the incorporation of these nanofillers into a polymer matrix, most of these clays are chemically modified. More variations of modified clays can be found in the review study done in 2015: [17]. In addition to their constituents, the tables also summarize the properties and applications, and the corresponding preparation method used for synthesis of the nanofillers. Figure 2 summarizes the symbols used in the tables by the authors to refer to the applications and application related properties of the nanofillers. These symbols are used throughout the paper to facilitate the selection of a nanofiller for a specific application or to impart a specific characteristic in the composite materials. This approach will make it easy to select nanofillers and nanocomposites for advanced application from huge number possibilities.

Fig. 2 - Categories for applications and properties of the studied nanofillers and the nanocomposites: the symbols are used by the authors to refer to the applications and application related properties of the nanofillers and the corresponding nanocomposites.

2.1.1 Kaolinite

Same as MMT, kaolinite can significantly enhance the polymer properties even in very small contents. The most common preparation methods for the synthesis of this filler are divided into three main groups. First, in-situ intercalative polymerization (kaolinite is swollen by a liquid monomer solution; the reaction occurs in between the intercalated sheets). Moreover, solution intercalation (a solvent is used to exfoliate the single layers) and, melt intercalation (forces are applied to disperse the sheets in the polymer melt) are also used [18]. Novel approaches for other preparation methods are found in the following tables for various nanofillers. Major applications of kaolinite are focused on thermal properties, specifically to achieve fire retardant polymers while improving the mechanical properties of the nanocomposite. Table 1 summarizes the possible combinations of Kaolinite nanoclay with various polymer matrixes and lists their processing methods, modification agents (to enhance the interaction with polymer matrixes), properties and applications.

Table 1 - Kaolinite nanoclay combinations with polymer matrixes: processing methods, modification agents, properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Thermoplastic	Vinyl alcohol copolymer (EVOH)	Melt intercalation	Dimethyl-sulfoxide, methanol and octadecylamine	Better thermal resistance, glass transition temperature, crystallinity and barrier properties to oxygen.		[19,20]
	Polyvinyl alcohol (PVA)	Melt intercalation	-	Biomedical application: tissue engineering and drug delivery.		[21]
		Preparation via freezing-thawing method	Dimethyl sulfoxide (DMSO) and potassium acetate	Decrease in the swelling ratio as well as in the dehydration rate of nanocomposite hydrogels.		[22,23]
	PA	Melt compounding dried ball- milling mixture	TiO ₂ -coated kaolinite	Enhanced UV anti-aging and mechanical properties.		[24]
	Polystyrene (PS)	In situ emulsion polymerization	Intercalated with dimethylsulfoxide (DMSO)	Slower thermal decomposition. Fire retardant applications.		[25,26]
	Polyarylamide (PAA)	In situ polymerization	Intercalated with dimethyl-sulfoxide (DMSO)/methanol and potassium acetate (KAc)	Salt-resistant super- absorbent.		[27]
	Polyethylene oxide (PEO)	In situ polymerization	Urea as intercalating agent	Improved mechanical properties. Suitable for lithium battery and polymer electrolytes.		[28]

Thermoset	Polyaniline (PANI)	In situ mechano-chemical synthesis	-	Improved thermal stability. Corrosion resistance. Marine applications.		[29]
	Epoxy	Solvent-free method	Modified with silylation of 3-aminopropyltriethoxysilane	Improvement in the water resistance.		[34]
Elastomeric	Natural rubber	Vulcanization in a sulfur curing system.	Chlorinated polyethylene and alumina.	Uses for insulation materials: noise and vibration control of acoustic. Absorbing material, thermal insulation for building construction.		[34]
	Styrene-butadiene rubber	Via melt blending	-	Improvement of mechanical properties and thermal stability.		[32]

2.1.2 Montmorillonite/ smectite / bentonite

MMT is commonly used for fire retardancy purposes. Its combination with rubber matrix has shown a decrease in hydrogen and water permeability [33]. Due to its hydrophilic nature, the dispersion in a polymer matrix can be challenging. Therefore, it is common to find montmorillonite organically modified, with the purpose of improving dispersing capabilities and gelling absorption [34, 35]. Several recent studies include the application of these fillers for other purposes such as self-healing hydrogels with biocompatible properties [17, 36] with significantly improved mechanical properties. Table 2 collects several examples of research studies of different matrixes and MMT/ smectite / bentonite nanoclays published mostly between 2015 and 2018.

Table 2 - Montmorillonite/ smectite / bentonite nanoclay combinations with polymer matrixes: processing methods, modification agents, properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Thermoplastic	PUI	Via in situ polymerization	Hydroxyl-modified	Heat and flame-resistant. Increment of compression strength and modulus.		[37]
	PVA	Solution-Intercalation Method	-	High strength, high stiffness, and corrosion-resistant. Biocompatible.		[38]
	PA	Intercalation through ion exchange process	Amine hydro- chloride	Drug Delivery System. Antimicrobial activity		[41]
	PS	In-situ Polymerization	-	Higher resistance to gas permeability		[44]
	PAA	Via simple UV irradiation polymerization	-	Increase water absorbency of hydrogels. Improved mechanical properties and recoverability.		[36]
	Polymethyl methacrylate (PMMA)	Melt blending	Cation-exchange reaction of Na-MMT with hexadecyl trimethyl ammonium bromide	Better surface roughness for better adhesion, wettability, and printability properties.		[43]
	PEO	Melt intercalation	Intercalated with lithium	Better thermal stability, higher ionic conductivity. Used for ion secondary batteries.		[44]
	PP	Mechanical mixing	-	Waste containment barriers applications. Improved mechanical properties.		[51] 52]
	Polyvinylpyrrolidone (PVP)	Solution mixing	-	Enhanced modulus, higher thermal stability and improved moisture resistance properties. Biodegradability.		[47]

	PTFE	Ultrasonic mixing	Organically modified	Improved permeability, water retention, and proton conductivity of proton exchange membrane for fuel cells.		[34]
	Polyethersulfone (PES)	Solution dispersion and wet-phase inversion methods	Organically modified	Improved the mechanical properties and thermal stability. Removal of pesticides.		[48, 49]
Thermoset	PANI	In situ polymerization	Sodium dodecyl benzene sulphonate (DPM)	Reduce flammability and toxicity. Electrical conductivity.		[4]
	Epoxy	In situ polymerization	-	Moisture absorption minimization: improve aircraft radome longevity.		[50]
		Intercalation	Silicone-intercalated organic MMT	Enhanced damping properties.		[51]
	Liquid crystalline polymer (LCP)	Solvent casting	Organically modified	Enhancement in the degradation temperature. High-frequency electronic applications.		[52]
Elastomeric	Natural rubber	Single-stage mixing	Grafting Stearyl Amine Ethoxylate	Improvements in the mechanical properties.		[33, 35, 53–56]
	Styrene-butadiene rubber	Mixing and curing	Modified with dimethyl, benzyl, one alkyl tail i.e. hydrogenated tallow HT	Lower relaxation rate.		[56]
	Silicone	Solution mixing	-	Improve heat ageing and thermal properties.		[57]

2.1.3 Illite

Compared to MMT which is a smectite clay with small nanolayers, illite has larger nanolayers. The higher aspect ratios enhance the mechanical properties and the thermal stability of the polymer matrixes. Nevertheless, illite has been far less explored as reinforcement agent in polymer nanocomposites, compared to the other nanoclays [58]. The interest in using the nanoclay alternatives has been due to the environmental impact of petroleum-based superabsorbent. From the illitization of smectite, a mixed layer clay: illite/smectite, has been developed for superabsorbent hydrogels which can be of great interest in agriculture, hygiene products, drug delivery, wastewater treatment, etc. [59]. Their Polyethylene Oxide (PEO) based composites, can be used to construct solid polymer electrolytes for rechargeable polymer lithium batteries [58]. More examples of applications and properties are listed in the Table 3.

Table 3 – Illite combinations with polymer matrixes: processing methods, modification, properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.	Year
Thermoplastic	Polyethylene Oxide (PEO)	Exfoliation	Deintercalation of dimethyl sulfoxide (DMSO)	Ability to dissolve salts ensuring a high ionic mobility. Lithium battery applications.		[60]	2016
	Polypropylene (PP)	-	Fluorination of illite and modified with 3-aminopropyltrimethoxysilane and hexadecyltrimethoxysilane	Better thermal stability and flame retardancy, and mechanical properties.		[61]	2012

	High-density polyethylene (HDPE)	Melt vibration with ultrasonic waves	-	Enhanced stiffness, improve UV light absorption, electroluminescence, and drug delivery.		[62]	2004
	Polyvinyl Chloride (PVC)	Melt blending	Aluminate coupling agent	Toughening increased notched impact strength and decreased elongation.		[63]	2015
Thermoset	PANI	In situ polymerization	-	Better processability with colloidal stability, mechanical strength and novel electrical, catalytic properties.		[64]	2011
	Epoxy	Mechanical mixing	Octadecylamine (ODA)	Enhanced thermo-mechanical properties; increase in storage and loss modulus.		[65]	2011

2.1.4 Halloysite

Halloysite is a member of kaolin group, which has attracted interest in many recent studies. Although they are similar to the rest of nanoclays, their structure is much similar to nanotubes. However their production costs are much lower [66]. The main application fields of these nanofillers are: flame retardancy, anticorrosion, dye removal, and drug delivery and recently, stimuli-response. A detailed review dating from 2017 presents more information about this nanofillers (origins, applications and matrix combinations): [67]. The main characteristics of these nanofillers including their combinations with different polymer matrixes, preparation methods, modification agents, properties and possible application areas are presented in the table below.

Table 4 – Halloysite combinations with polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Thermoplastic	PVA hydrogel	Freeze-thawing	-	Substitute for articular cartilage, drug delivery, contact lenses, wound dressing, and artificial organs. Better mechanical properties.		[68]
	PA	Via in situ interfacial polymerization.	-	Higher hydrophilicity: potential use for reverse osmosis (RO) (wastewater treatment).		[69]
	PMMA	Mechanical mixing	Barium-coated	Cytocompatible. Implantation and fixation of prostheses to the bone. Enhancement of mechanical properties.		[70–72]
	Poly(butylene succinate) (PBS)	Solvent casting	Sodium dodecanoate (NaL) Loaded with poly(ethylene glycol)	Increased hydrophilicity.		[73]
	PES	Phase inversion	Immobilized lysozyme	Improved surface hydrophilicity and water flux. Enhanced mechanical strength and good antibacterial activity. Application: reduce fouling in water treatment.		[74]
Thermoset	Polyaniline (PANI)	In situ chemical polymerization	Cyclohexene	Fabrication of a polymer-based green catalyst system.		[75]

	Epoxy	Mechanical mixing	Amine hardener immobilized mesoporous silica	Better waterproofing property. Self-healing. Epoxy coating for carbon steel. Outdoor applications.		[76–78]
	Polyester Resin	Mechanical mixing	Silica nanoparticles	Application: aerospace, automobiles and building materials.		[79]
Elastomer	Natural rubber	-	2-mercaptobenzimidazole (MB)	Antioxidative efficiency. Better mechanical performances (tensile strength, tear strength and heat buildup).		[80, 81]

2.1.5 A green approach to nanoclay filled composites

MMT and halloysite nanoclays have been the most extensively studied nanoclays with biodegradable properties. Polylactic acid (PLA) has received most of the attention as biodegradable polymer matrix, not only for nanoclays but for also for other nanofillers. Although, PLA presents good mechanical properties, its applications are limited in critical areas such as electronics and the automotive industry due to its flammability. As seen in the previous sections, nanoclays have been used as fire retardants for other polymer matrixes, making them potential candidates to transform PLA into a green and improved nanocomposite where flame retardancy is critical. The table below summarizes a series of studies that provide biodegradable nanocomposites using nanoclay fillers that include PLA as well as other polymeric matrixes.

Table 5 – Novel research studies of biodegradable nanocomposites using nanoclay nanofillers.

Matrix polymer name	Nanoclay type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
PLA	Cloisite 30B (MMT)		Melamine polyphosphate (MPP)	Flame retardant. 3D printing		[82]
Poly (butylene adipate-co-terephthalate) (PBAT) and thermoplastic starch (TPS)	Sepiolite		-	Enhanced thermal and mechanical properties. Lower water absorption rate. Short term applications; packaging, agriculture and hygiene.		[83]
Acrylic acid (AA) monomer	MMT	In situ polymerization	Salep (multi-component polysaccharide)	Porous hydrogel with high swelling capacity. Candidate for agricultural and horticultural applications (reservoir systems)		[84]
Cellulose acetate (CA)/poly-L-lactic acid (PLLA)/	Halloysite nanotube	Electrospinning	-	Used for gel polymer electrolytes (GPEs) for lithium-ion batteries.		[85]
PLA	Halloysite nanotube	Melt blending	-	Increased tensile strength and Young's modulus and storage modulus.		[86]
Poly (butylene succinate-co-adipate) (PBSA) and maleated polyethylene (PEgMA)	Halloysite nanotube and MMT	Melt blending	-	Increased thermal stability with MMT, but not with halloysite nanotubes. Superior rigidity and water uptake rate.		[87]

PVA-PVP	Halloysite nanotube	Crosslinking	-	Application in the dentistry field		[88]
Polyester (PEST)	Halloysite nanotube	Solution casting		Increased storage modulus, tensile and flexural properties as well as fracture toughness, surface roughness and glass transition temperature.		[89]

2.2 Carbon Nanoparticles

The two first known forms of Carbon Nanoparticles were diamond and graphite. However, the seminal researches in 1980s and 1990s lead to the discovery of other allotropes of carbon including fullerene and carbon nanotubes (single and multi-walled: SWCNT and MWCNT). The more recent form is graphene which was recognized around 2007 as a nanofiller for plastic composites. Carbon black has been the most studied so far for the creation of nanocomposites for mechanical and thermal enhancement. However the new forms of carbon nanoparticles are raising unprecedented interest in many areas such as electronics devices, smart materials, electromagnetic shielding, etc. [1]. Similar to the previous section for nanoclays, different studies from the combination of matrixes and carbon particles are presented along with their important modification methods, properties and application areas are extracted from the state-of-the-art literatures and presented in the sub-sections below.

2.2.1 Fullerene

Fullerene presents remarkable properties when synthesized with polymers, providing enhanced mechanical, thermal, electrical, optical, and photovoltaic properties. Nevertheless, the synthesis processes, unlike nanoclays, are still in early stage of development. They are generally used for photovoltaic applications, temperature sensors, and antimicrobial applications among other [90]. Table 6 summarizes the information for Fullerene about the potential polymer matrixes, processing methods, modification as well as properties and applications.

Table 6 –Fullerene combinations with polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Thermoplastic	Thermoplastic polyurethane (TPU)	Prepolymer method	Urethanized fullerene	Improve TPU durability by reducing oxidation. Reduced gas permeability.		[91]
	Poly-ether-ether-ketone (PEEK) composites.	Low-temperature sintering	-	Lower wear rate.		[92]
	Poly (phenylene sulfide) (PPS)	Solution co-blending method	-	Excellent electrical conductivity, increased crystallization temperature, thermal stability and mechanical performance.		[93]
Thermoset	Liquid crystalline polyester (LCP)	-	-	Shape memory behavior.		[94]
	Epoxy	-	-	Electrical insulation.		[95]
Elastomer	Natural Rubber	-	-	Influence on the dielectric properties and increase of coefficient of reflection of microwaves in the 6–12 GHz range.		[96]

2.2.2 CNTs

CNTs are graphene sheets that are rolled to form tubes. At certain loading fraction, so called “percolation threshold”, these fillers provide the matrix electrical conductivity due to the network created. Based on their layer configurations, these nanotubes can be divided in single-walled nanotubes (SWCNTs) and multi-walled nanotubes (MWCNTs) (Fig. 1). Carbon nanotubes due to their special atomic configuration exhibit extraordinary properties such very high stiffness, superior thermal and electrical conductivity, etc. Therefore, they have been interesting for many multifunctional applications especially in lightweight structures. In the recent years, CNTs have been used extensively in nanocomposites hydrogels which have numerous applications for tissue engineering, sensors, solar cells, biomedicine, actuators, biofuels, antennas, solar energy conversion, etc. [97, 98]. Summary of the recent research studies on CNTs for different application areas are presented in Table 7.

Table 7 –CNTs combinations with polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Type of CNT	Properties-applications		Ref.
Thermoplastic	ABS	Melt mixing	TPU	MWCNTs	Increased shape recovery ratio.		[99]
	Aromatic PA	Interfacial polymerization		MWCNTs	Reverse osmosis membrane (water purification).		[100]
	PA 66	Melt compounding		MWCNTs	Multifunctional electrical-mechanical composites. Structural and EMI shielding applications		[101,102]
	PVA–	-	Poly(ethylene oxide) (PEO) and chitosan	MWCNTs	Detection of polar vapors.		[103]
	TPE (styrene–butadiene–styrene (SBS))	Up-scalable extrusion	-	MWCNTs	Increased mechanical modulus, larger deformations and electro mechanical stability. Use for strain sensors.		[104]
	PP	Solution processing and melt blending	-	MWCNTs	Enhancement of modulus, tensile strength and impact strength.		[105]
Thermoset	PANI	In situ polymerization	Graphene	MWCNTs	Extraordinary electrochemical performance. Electrode material for energy storage: super capacitors.		[106]
	Epoxy	-	Graphene	MWCNTs	Improved thermal and optical properties Absorber plates for fresh water productivity.		[107]
	Epoxy	Solution mixing and ultrasonication	-	MWCNTs	Improved friction and scratch resistance-painting and microelectronics		[108]
Elastome	Natural rubber	-	Carbon Black	MWCNTs	Health monitoring, sensors in different dynamic elastomeric parts like tires, valves, gaskets,		[109]

					engine mounts or even human finger motion detections.		
	Styrene-butadiene Rubber (SBR),	-	-	MWCNTs	Force sensor for monitoring automotive tire deformation.		[110]

2.2.3 Graphene and Graphene Oxide

Research studies on Graphene and Graphene oxide based nanocomposites have increased in the recent years. Graphene nanofillers such as carbon nanotubes exhibit exceptional properties. Therefore, they are studied for many novel advanced applications such as polysulfone membranes; thermally interface materials, microelectronics, etc. More and more research studies have been done for water purification purposes, antibacterial materials for medical applications as well as for different sensor applications. Graphene based nanocomposites membranes have received special attention, although most of the recent studies will require further investigations for an ideal functionality [111]. Table 8 presents the research studies on different forms of graphene in combinations with polymer matrixes, processing methods, modification, properties and applications.

Table 8 – Research studies of different forms of graphene in combinations with polymer matrixes: processing methods, modification, properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Type	Properties-applications		Ref.
Thermoplastic	Polyacrylic acid functionalized (PAA)	Facile method	-	Magnetic graphene oxide	Removal of cationic organic pollutants in wastewater treatment. Recyclable and reusable.		[112]
	Polycarbonate (PC)	Solution blending	-		Antibacterial activities against E. coli and S. aureus.		[113]
	PES	Solution blending	TiO ₂	Reduced graphene oxide	Nanofiltration technology, water softening and filtration, improve the hydrophobicity, antibacterial properties, and low toxicity.		[114]
	Polyurethane (PU)	Melt blending	(PEEAMA)	Graphene	Thermo-responsive shape memory properties.		[11]
	PVC	Solution blending	-	Reduced Graphene Oxide Thermally	Improvement of the tensile strength and tensile modulus.		[115]
	Polysulfone (PSU)	Solution blending	-	Reduced graphene oxide	Antibacterial properties against Bacillus subtilis and Escherichia coli. Applications involving human exposure.		[116]
Thermoset	Epoxy	In situ prepolymerization	-	Graphene	Enhanced corrosion resistance, UV stability, and impact resistance.		[117]
	LCP	In situ prepolymerization	-	Graphene oxide	Enhancement mechanical properties and the thermal stability. Better creep stability thermo mechanical behavior.		[118]
	PANI	Nanoemulsion method	ZnO	Graphene oxide	Gas sensing of ammonia.		[119]

Elastomer	Silicone Rubber	Liquid mixing method	Carbon Black	Graphene	Flexible piezoresistive sensors.		[12]
	SBR	Modified Hummer's method	Carboxylated acrylonitrile butadiene rubber	Graphene oxide	Better thermal stability and solvent resistance. Tire manufacture.		[120]

2.2.4 Carbon Black

Carbon black (CB) has been widely used due to its abundancy, low cost, and its self-networking capabilities (self-organizing into three-dimensional continuous networks). This carbonic filler has been used in polymer matrixes such as in tire and manufactured rubber applications, replacement of tire market being the larger sector in world for carbon black demand [121]. Other applications, due to their homogeneous dispersion in polymer matrixes are: super capacitors, batteries, sensors, fuelcells etc. [122]. Its black pigment has also been used for coloring pigments and paints. One of the most interesting application however, same as the other carbon nanofillers, is for electric and electronic application: electromagnetic shielding, thermoelectric energy conversion, electrical switching etc. Unlike CNTs & CNFs however, this filler is not effective for shape memory polymers [123]. For the best electrical properties, CB particles must have small diameters and high surface area, which means that the dispersion in the polymer matrix is highly important. However, the modification of CB in order to simplify the dispersion in the polymer matrix reduces the electrical properties [1]. Table 9 summarizes possible polymer matrixes for CB, thier processing methods, modification as well as properties and applications.

Table 9 –Carbon Black combinations with polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Thermoplastic	PC	Melt blending	MMT	Enhanced tensile strength, better thermal stability. Homogeneous dispersion of carbon black.		[124]
	PMMA	In situ suspension polymerization	Methyl methacrylate coated	Enhancement of thermal and mechanical properties. Uses: ultra-lightweight proppants in the shale oil and gas exploitation industry.		[125]
	PVA	Solution casting	Polystyrene sulfonic acid (PSSA)	Improved dielectric constant and mechanical properties. Flexible energy storage device applications, charge storage capacitors, electrostriction artificial muscles and drug delivery.		[122]
	Polyimide (PI)	In situ polymerization	Chloride modified. Intercalated with poly(amic acid) (PAA)	Higher refractive index. Optical applications of thin films.		[126]
Ther	PANI	Hydrothermal synthesis	Manganese dioxide (MnO ₂)	Excellent electrochemical performance. Supercapacitor materials uses.		[127]
Elastomeric	Silicone rubber (SR) based	Mechanical mixing	Ionic liquid-modified carbon black	Good piezoresistive properties. Flexible piezoresistive sensors and wearable electronic devices.		[128]

2.2.5 Graphitic Carbon Nitride

Graphitic carbon nitride (g-C₃N₄), in contrast to the other carbon nano fillers, has electron-rich properties, basic surface functionalities and, physical and chemical properties. With a similar structure as graphene, it has been largely investigated in the field of photocatalysis and heterogeneous photocatalysis [129]. Recent studies are also considering using g-C₃N₄ for proton exchange membranes for fuel cells. The addition of this nanofiller enhances the mechanical properties by increasing the tensile strength of the membranes [129]. G-C₃N₄ has a layered structure, same as graphene, montmorillonite and LDH, however with the advantage of being cheaper. In addition, it is easier and faster to prepare. Studies suggest that it could be a superior alternative to LDH in terms of flame retardancy, improved thermal stability, as well as better mechanical properties and UV shielding [130]. Its incorporation into thermoplastic polyurethane nanocomposites can be a successful alternative to reduce fire hazards [131]. More examples of applications are presented in the Table 10.

Table 10 – Carbon Nitride combinations with polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Thermoplastic	Sulfonated poly(ether ether ketone) (SPEEK)	Melt blending	Graphitic carbon nitride	Photocatalysis and heterogeneous chemical catalysis. Nanosheets for methanol fuel cell application.		[129]
	PVA	Melt blending	Graphitic carbon nitride	Good mechanical, thermal, and anti-swelling properties of the membranes. Water-selective hybrid membranes.		[132]
	Thermoplastic polyurethane (TPU).	Melt blending	Aluminum hypophosphite (AHPi)	Improved thermal decomposition. Flame retardant.		[130]
	PES	Melt blending	Graphitic carbon nitride	Osmosis membrane.		[133]
Thermoset	Polyaniline (PANI)	Melt blending	Graphitic carbon nitride	Photo catalyst for solar energy conversion, hydrogen production and environmental pollution purification.		[134]
Elastomer	Silicon	Ion beam assisted magnetron sputtering	Graphitic carbon nitride	Wear resistance. Subcutaneous joint replacement. In vivo biocompatibility.		[135]

2.2.6 Nanodiamonds

Nanodiamond also known as ultra-dispersed diamond or ultrananocrystalline diamond has initially studied in 1960s. This round nanofiller has been very interesting for many industrial applications due to superior hardness, thermal and physical properties [136,137]. Nanodiamond required for industrial applications are produced in large quantities by detonation method. Non-toxicity and biocompatibility of these nanofillers have been confirmed extensively [138]. Therefore, they are one of the ideal candidates for biomedical applications such as reinforcement agents for bone surgery and tissue engineering. Table 11 summarizes the information from the extensive research study for Nanodiamond reinforced polymer matrixes, processing methods, modification agents as well as properties and applications.

Table 11 – Nanodiamond reinforced polymer matrixes: processing methods, modification as well as properties and applications.

	Plastic name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Thermoplastic	Polyvinyl alcohol (PVA)	electrospinning	-	Increased tensile modulus and strength. biological applications		[139]
	Poly-L- (lactide-co-ε-caprolactone)	grafting - from approach of anionic polymerization	-	Increased elastic modulus. Tissue engineering		[140]
	PA 66	In situ reactive extrusion	-	Improved mechanical properties		[130]
Thermoset	Epoxy	liquid-liquid extraction method	Graphene oxide	Enhanced mechanical properties, fracture toughness, and thermal stability.		[134]
	Epoxy	In situ polymerization	-	Friction, tribological, and mechanical properties		[137]
Elastomeric	Nitrile butadiene rubber	In situ polymerization	Mercapto - terminated silane agent	Improved mechanical properties		[135]

2.2.7 A green approach of carbon nanocomposites

Applications of CNTs are increasing significantly. However, it is important to considerate the environmental impacts of the CNT inclusion into polymer matrixes. In fact, many studies have shown that they could increase the persistence of polymers in landfills, lakes and surface waters [141]. There are different studies that show both the cytotoxic and non-cytotoxic effects of CNT filled nanocomposites towards different microorganisms with important role in polymer degradation. Their inclusion in matrixes such as PLC, led to a significant decrease of the biodegradability rate [141]. Nevertheless, other hybrid nanocomposites using CNTs have shown improved biodegradability properties [142–144]. CNTs have also shown a significant improvement of Polyhydroxyalkanoates (PHA), improving its Young's modulus, increasing thermal stability and electrical conductivity. Its addition to PHA maintains its biodegradability, however it has a negative effect on the biodegradability rate [66]. The example shown in Table 12, proposes the use of 3-hydroxybutyrate and 3-hydroxyhexanoate PHBHHx, a PHA with mechanical properties and biocompatibility. These nanocomposites can be of great interest in the packaging industry, medical, agricultural and many others.

Graphene oxide also has its share of results in terms of biodegradability: in combination with chitosan biopolymer, the presence of the graphene oxide increased the inactivation of the microorganisms in charge of biodegrade them [145]. Combinations with carbon nitride and modified silver chromate nanoparticles, show to be enhance the photocatalytic degradation, an alternative method that is considered an environmentally friendly solution for pollutants decomposition using solar light energy [146]. A review from 2016 collects a variety of examples of biopolymer–graphene nanocomposites together with the available processing methods [147]. In the same context of photocatalytic degradation, carbon nitride has been investigated under combinations with titanium dioxide and cadmium [148]. Graphite has been proven to be degradable in the presence of *Penicillium funiculosum* in combinations with Poly(3- hydroxybutyrate) (PHB) matrix, despite a higher incubation period [149]. Less information is available about carbon black in terms of biodegradability behavior in polymer nanocomposites with biodegradable matrixes. Table 12 summarizes the information from the research studies of biodegradable nanocomposites using carbon nanofillers.

Table 12 - Novel research studies of biodegradable nanocomposites using carbon nanofillers.

Matrix polymer name	Carbon nanofiber type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
PES	Fullerene	Solution casting	-	High modulus and impact resistance. Applications such as biomarkers, drug delivery dental applications		[150]
PLA	Fullerene	Electrospinning and electrospraying	Zinc Oxide (ZnO)	Photocatalytic and self-cleaning properties. Antibacterial activity against Staphylococcus aureus.		[151]
Polyaniline (PANI)	MWCNTs.	In situ polymerization	Carboxymethyl cellulose	Enhanced electrical and electrocatalytic properties. Used for electroactive membranes – electrochemical sensors.		[142]
PLLA	MWCNTs.	Pressure-induced flow (PIF) processing		Light-weight high-strength foams with good electrical conductivity and EMI shielding properties.		[144]
Carboxymethyl cellulose (CMC)	MWCNTs	Ultrasonication	-	Hydrogel showing biocompatibility using rat fibroblast cells. Used in transdermal drug delivery.		[143]
Copolymer of PLA-PCL (Poly-ε-caprolactone)	MWCNTs	Boron Nitride		Improved damping behaviour (dissipation of energy inside the material) with higher concentrations of CNTs.		[152]
Cellulose nanofiber (CNF)	MWCNTs	Vacuum-assisted self-assembly technique	-	Flexible dielectric paper. Improved mechanical strength. Used in electronics and electric power systems.		[153]
PHBHHx	MWCNTs	Solution mixing and evaporation	-	Films for human mesenchymal stem cell (hMSC) differentiation. Used in bone tissue engineering.		[154]
PLA-PBAT	Graphene	Melt - mixing	-	Enhanced electrical permittivity, dielectric properties and electromagnetic interference shielding effectiveness.		[155]
Amyloid fibril	Graphene	-	-	Exceptional mechanical and electronic properties. Reversibly change shape properties towards humidity stimuli. Uses: biosensors for quantifying the activity of enzymes.		[156]
PCL	Graphene	Solvent precipitation technique	Ag	Increased modulus and electrical conductivity. Antibacterial effects, Use in tissue engineering and fracture fixation devices.		[157]

PANI	Carbon nitride	In-situ polymerization	ZnO	Improved photocatalytic properties.		[158]
Natural rubber (NR)	Carbon black		Cellulose nanowhiskers	Enhancement of electrical and mechanical properties. Uses for EMS, chemical and stress sensors, actuators, electrostatic dissipation etc.		[159]
Poly (butylene succinate) PBS	Carbon Black	Melt compounding	-	Improved mechanical and electrical properties.		[160]
PLLA	Carbon Black	Melt compounding	-	Manipulation of filler network for decrease in the electrical percolation threshold.		[161]
Polyvinyl alcohol (PVA)	Nanodiamond	electrospinning		Increased tensile modulus and strength. biological applications		[139]

2.3 Nano Oxides

Nano oxides are particle shaped nanofillers including Oxygen in their structure. The most commonly used nano oxides are silica (SiO_2), alumina (Al_2O_3), and titania (TiO_2). Due to the hydrophobic properties of the particle surface, these fillers often require heat treatments or chemical reactions in order to facilitate a homogeneous dispersion in the polymer matrix. These nanofillers are used in a broad range of applications. Silica and titania has been also widely researched for polymer electrolytes for fuel cells (see Table 13). The composite membranes present good ionic conductivity as well as good water retention at low to medium temperatures [162]. However, unlike Silica and alumina, titania is used in photocatalytic, optical, and photovoltaic applications [1]. A maximum addition of 2% of silica in food and beverage materials is approved by the FDA, which makes this nanofiller attractive for packaging applications [163]. Its addition to films can improve the tensile strength without compromising the permeability and biodegradability. In many applications, silica has been combined with MMT, since it provides better dispersion and exfoliation in MMT nanocomposites [66,164,165]. Other hybrids combine silica and chitosan, for fuel cells [162], biomaterial for bone regeneration [166],[167], among others. More examples are shown in the Table 13.

Table 13 –Silicon dioxide (SiO_2) combinations with polymer matrixes: processing methods, modification as well as properties and applications

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Filed	Ref.
Thermoplastic	PES	Via sol-gel non- solvent induced phase separation	P(PEG-PDMS-KH570)	Antifouling performance. Membrane for water treatment.		[165]
	Poly(lactic-co-glycolic) acid (PLGA)	Evaporation	PO_2	Biodegradability and biocompatibility. Uses: bone regeneration.		[168]
	Polypropylene (PP)	Melt blending	Graphene Oxides	Significantly higher tensile and impact strength.		[169]
	Shape memory foams (SMF)		Tungsten, Aluminium oxide	Toughness and strain at break. Microelectromechanical systems, actuators and biomedical devices and medical implants.		[170]

	Poly(MMA-co-TMSPMA)	Fragmentation chain transfer (RAFT) polymerization	-	Significant improvement of mechanical properties. Uses for bone scaffold materials.		[171]
Therm	Siloxane polymer	Organic-inorganic hybrid method	KH570, Al ₂ O ₃ -TiO ₂	Films with excellent hydrophobicity.		[172]

2.3.1 Alumina

Alumina nano particles offer high thermal properties, chemical stability, high creep resistance, hardness, strength and, high melting point. In combinations with natural rubber, alumina and kaolinite particles, have been used so far for similar purposes: reinforcement for the mechanical ,thermal and electrical properties [173]. Alumina has also been investigated in combinations with graphene for novel electrical and dielectric properties, making this nanocomposites a promising approach for microwave communication of substrates, patch antennas, electromagnetic interference shielding electrostatic dissipation as well as radar cross section reduction for aerospace and automobile sector [174]. More recent investigations suggest the combination of alumina with PEO-PPG blends for polymer electrolytes in zinc batteries, as a replacement of liquid solvents in order to reduce the risk of fire hazard, leakage, waste disposal limitations , and mechanical strength [175]. Table 14 summarizes the Alumina (Al₂O₃) combination with polymer matrixes, their processing methods, modification, properties and applications.

Table 14 – Alumina (Al₂O₃) combinations with polymer matrixes: processing methods, modification, properties and applications

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Thermoplastic	Low-density polyethylene (LDPE) as	Pre-mixing and melt-extrusion	-	Lower DC conductivity. Insulation of high voltage direct current cables.		[176,177]
	Polyethylene (PE) o	Dip-coated and electron beam irradiation.	(PVDF-HFP) crosslinker (TTT)	High thermal resistance and electrochemical stability. Uses: lithium secondary battery.		[178]
	PEO	-	Polypropylene glycol (PPG) as [Zn(CF ₃ SO ₃) ₂]	Conductivity enhancement. Satisfactory electrochemical stability. Uses: zinc batteries.		[175]
	PVA	Cyclic freezing/thawing method.	-	Thermal and water dual-responsive shape memory properties. Enhanced mechanical properties. Uses in the biomedical and aerospace field.		[179]
Thermoset	Epoxy	Gelcasting, sintering and vacuum infiltration methods	-	High strength and thermal conductivity		[180]
Elastomer	Silicon rubber	Mechanical mixing	poly(dopamine) (PDA)	Enhanced thermal conductivity		[181]

2.3.2 Titania

Titania or Titanium Dioxide is an attractive metal oxide especially due to its photocatalytic properties. It has been widely used in electronic devices because of its low cost and structural stability. Combined with graphene

oxide, Titania has been investigated for flexible pressure sensors in combination with PVDF, showing good dielectric properties, increased wettability and reduced bio-fouling (accumulation of microorganisms on wetted surfaces) [182]. In the Table 15, other potential applications are listed, such as in textile application with antimicrobial and self-cleaning properties along with the other information collected from the state-of-the-art literatures.

Table 15 - Titanium Dioxide (TiO₂) combinations with polymer matrixes: processing methods, modification, properties and applications

	Plastic name	Preparation method	Modified/mixed with	Properties-applications	Filed	Ref.
Thermoplastic	Nylon	-	ZnO Cotton	Fabric with good abrasion resistance, dye ability and comfort Antimicrobial properties of hospital clothing.		[183]
	PBT	Melt blending	-	Self- cleaning properties under UV-A radiation. Possibility of producing textile products.		[184]
	PEO	Solution casting	NaClO ₄	Sodium ion batteries.		[185]
Thermoset	Epoxy	Solution blending	Ag	Used as bactericidal surface coating in industrial or healthcare.		[186]
Elastomer	Silicone rubber	Ultrasonic treatment and mechanical mixing	-	Highly transparent LED packaging materials.		[187]

2.3.3 Magnesia

Similar to Titanium Dioxide, Magnesium Oxide or Magnesia possess features as photocatalytic and antimicrobial activity. Non-toxicity, biocompatibility, thermal and electrical insulation, UV blocking ability are also among other interesting properties of this nanofiller. These features have made MgO attractive for applications such as bactericide, packaging material (in combinations with chitosan), as well as material for toxic waste remediation [188]. Recent studies propose using MgO in LDPE blends as novel insulation material for high voltage distribution of renewable energy [189]. A uniformly dispersed coating could enhance the thermal degradation significantly, by increasing it with approximately 100 °C. Table 16 presents the summaries of the research study of the state-of-the-art literatures about Magnesium oxide (MgO) about the polymer matrixes that can be combined with MgO, the processing methods of the combinations, modification agents of the nanofiller along with their important properties and applications.

Table 16 - Magnesium oxide (MgO) combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
LDPE	Melt compounding	Vermiculite	Long-term antibacterial effect. Biocompatible. Uses in health care and medical products, syringe, catheters, pharmaceutical bottles, PE bags, health foils etc.		[190]

Polyindole (PIn)	-	TiO ₂	Electrically conductive; excellent optical and electrical properties. Uses in solar cells, gas sensors, and short-wavelength light-emitting devices rechargeable batteries etc.		[191]
Polypyrrole	In-situ polymerization	Ammonium Persulphate	Reduced DC conductivity. Uses in insulating material for electronic insulation: Printed Circuit Boards (PCB), capacitors, precoat for PCB holes, etc.		[192]
Thermoplastic polyurethane (TPU)	Pulsed laser ablation	-	Photoluminescent properties. Green photoluminescence emission. Uses in biomedical or photovoltaic devices.		[193]
CTAB-modified geopolymer matrix	Chemical precipitation method [10]	Cu ₂ O	Removal of methylene blue (MB) dye dyes from water by adsorption and photodegradation. Removal of organic pollutants from water or atmosphere.		[194]

2.3.4 Zinc oxide

Food and Drug Administration (FDA) has recognized ZnO as safe enough to be used as additive in amplification of foods [179]. In the area of nanocomposites, Zinc Oxide has been used as an antibacterial agent, UV absorber, and vulcanizing agent for elastomers [8]. New studies propose using bacterial cellulose biopolymer, in combination with ZnO to enhance the antibacterial properties, and transforming the nanocomposite into an excellent dressing for burn wounds [195]. A state-of-the-art summary of the research study about ZnO is presented in the Table 17.

Table 17 – Zinc oxide (ZnO) combinations with polymer matrixes: processing methods, modification, properties and applications

	Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications	Filed	Ref.
Thermoplastic	Linear low density polyethylene (LLDPE)	Melt blending	-	No production of space charges and enhanced DC breakdown strength. Uses: cable insulation application		[196]
	PLLA	Simple casting method.	Hydroxyapatite (HA)	Orthopaedic tissue engineering applications. Biomaterial for bone implantation.		[197]
	PVC	Ultrasonic irradiation and solution dispersion	PVA	Environmentally friendly manufacturing of nanocomposite films. Biomedical applications for wound healing devices, transparent electronics, light emitting devices, etc.		[179]
Thermoset	Epoxy	Mechanical mixing	-	Avoid dielectric breakdown or tracking failure in electronic devices.		[198]
Elastomer	Polydimethylsiloxane (PDMS)	CVT synthesis	-	Sensors for biomedical devices, optical displays		[199]

2.3.5 A green approach of biodegradable nano oxide composites

Nano oxides show degradability properties combined with biodegradable polymer matrixes when buried underground. In blends with poly butylene succinate (PBS) - chitosan (CS), both Silica and Alumina have shown signs of degradability, even more compared to graphene particles. However there are signs of lower weight loss compared to the pure PBS-chitosan blend [200]. TiO₂ and polypropylene PP nanocomposites show efficient photocatalytic degradation behavior [201], as well as in blends with ϵ -caprolactone / DL-lactide (ϵ -CL/DL-LA) copolymerspoly [202]. Table 18 summarizes the state-of-the-art information from the research studies of biodegradable nanocomposites using nano oxide.

Table 18 - Novel research studies of biodegradable nanocomposites using nano oxide.

Matrix polymer name	Carbon nanofiber type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
PVA	Silica (SiO ₂)	In situ polymerization	Chitosan	Excellent mechanical properties, low permeability for oxygen and water. Uses: food packaging.		[163]
Polybutadiene rubber	Silica (SiO ₂)	In situ polymerization	Chitosan	Improved flexibility, toughness, and thermal insulation. Uses in wound dressing, drug delivery, and tissue engineering.		[203]
PLA	Zeolites (NaAlO ₂ , SiO ₂)	Melt blending	-	Lower water and oxygen permeability. Improved mechanical properties and antimicrobial activity. Uses in food packaging.		[204]
Guar gum	Silica (SiO ₂)	In-situ deposited through sol-gel technique	-	Absorption capacity of cationic dyes/metal ions, cationic malachite green from mixture of dye solutions. Uses in wastewater management.		[205]
ϵ -caprolactone (PCL)	Silica (SiO ₂)	Crosslinking	-	Shape memory polymer, UV triggered. Improved mechanical properties. Uses in biomedical applications.		[206]
Cellulose acetate (CA)	Alumina (Al ₂ O ₃)	Melt blending	-	Improved dielectric properties. Uses in electronic devices, e.g. embedded passives.		[207]
Cellulose	Titanium Dioxide (TiO ₂)	Phase inversion technique.	-	Photocatalytic performance for phenol degradation in aqueous solution. Used for wastewater treatment.		[208]
Starch, amino acid (lysine), polypropylene glycol (PPG)	Zinc oxide (ZnO)	Solution casting	-	Increased thermal stability and mechanical properties. Shape memory properties. Uses: food packaging		[209]
PLLA	Magnesium oxide (MgO)	Casting method	-	Improved osteoblast adhesion while maintaining mechanical properties. Orthopedic tissue engineering applications.		[197]

2.4 Hydroxides

Hydroxide nanofillers have been intensively investigated for the replacement of toxic flame-retardant additives, since they have the capacity to reduce the toxicity of the fumes emitted to low or inexistent. The applications where it is of main concern are in energy storage devices (lithium batteries) as well as in electrical and communication cables, furniture, etc. The most common hydroxides fire retardant additives are Aluminum Hydroxide (ATH) and Magnesium Hydroxide. Boehmite and Layered Double Hydroxides (LDHs) increase thermal stability in polymer nanocomposites, however their efficiency is not as good as in the aforementioned hydroxides [210]. The sections below will discuss the most common hydroxide nanofillers with examples of different applications and alternative combinations.

2.4.1 Aluminum Hydroxide

Aluminum Hydroxide is the most extensively used as fire retardant agent on the market due to its low costs [8]. Their incorporation within polymer blends not only increase the flame retardancy but also reduces the smoke emissions toxicity [211]. Other advantages of incorporating Aluminum Hydroxide in polymers include higher toughness, low density and corrosion resistance. It is common to see Aluminum Hydroxide used in PCV and PE wires, as well as in combinations with silicones, nanoclays and boron compounds [211]. Recent studies propose a more efficient alternative to Aluminum Hydroxide, by incorporating Magnesium–Aluminum–Zirconium Hydroxide into poly(4-vinylpyridine-co-butyl methacrylate) (PVBm) matrixes, for the preparation of composite coatings [210]. Table 19 presents the important information about Aluminum hydroxide, its possible combinations with polymer matrixes, processing methods, modification, properties and applications.

Table 19 –Aluminum hydroxide ($\text{Al}(\text{OH})_3$) combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications	Filed	Ref.
Cellulose aerogel	In-situ sol-gel synthesis	AmimC	Improved improve flame retardancy. Uses in tissue engineering, waste-water treatment, thermal insulation, etc. Biocompatibility, biodegradability.		[212]
AAm and AMPS	In situ copolymerization	-	Hydrogel with good mechanical properties and self-healing properties. Biomedical applications in healing tissue engineering.		[213]
Poly(1,4-butylene adipate) (PBA)	Solution mixing	(layered double hydroxide (PBA/m-LDH)) m-LDH	Improved crystallization behaviour. Biodegradable.		[214]
Polyurethane foam (PU)	Solution mixing	-	Improved flame retarding. Uses in nanocoatings and conformal coatings, alternative to halogenated systems.		[215]

2.4.2 Magnesium hydroxide

Magnesium Hydroxide (MH) has the same working principle as Aluminum Hydroxide, however it has been far less used in polymer blends. Due to the higher degradation temperatures (approximately 100 °C higher than ($\text{Al}(\text{OH})_3$)), it performs better when incorporated in polymers with higher thermal stability [8]. Despite its low cost, the required content and mass to deliver the desired fire retardancy properties, often leads to poor dispersion and aggregation. Therefore, a large number of studies have focused on improving their dispersion in the polymer matrix by modifying the MH nanoparticles, e.g. with oorganic phosphate [216]. Table 20 presents the state-of-the-art information collected about Magnesium hydroxide.

Table 20 – Magnesium hydroxide (Mg(OH)₂) combinations with polymer matrixes: processing methods, modification , properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications	Filed	Ref.
ABS	-	Organic phosphate	Improved flammability due to enhancement of the strength of the char layer. Uses in car industry, pipes, telephone parts etc.		[216]
PE	-	Hexyl and phosphoric moieties	Better thermal stability and surface modification at nano scale.		[217]
Polystyrene	Melt compounding	Organcoclay (Organically modified montmorillonite)	Enhanced thermal and fire retardant properties.		[218]

2.4.3 Boehmite

Boehmite is Aluminium Oxide Hydroxide [AlO(OH)], generally used as nanofillers for polymers to enhance mechanical properties and other novel properties such as corrosion and wear resistance coatings [219] suppressing flammability of PU foams, proton exchange membranes, smart hydrogels etc. [220]. The preparation methods for Boehmite can be divided in four groups: in situ synthesis, template synthesis, melt intercalation (melt blending) techniques and intercalation of polymer from solution [221]. Table 18 presents examples of some relevant properties and applications that these nanofillers can offer to different polymer matrixes. More details about the combinations of matrixes and Boehmite can be found in a review study published in 2018 : [222]

Table 21 –Boehmite [AlO(OH)] combinations with polymer matrixes: processing methods, modification , properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications		Ref.
PLA	Melt processing	PCL	Improved mechanical, thermal, and rheological properties. Biodegradability. Medical applications, packaging, agriculture.		[223]
PMMA	In situ polymerization	Silane modified nano-boehmite	Improved thermal properties. Excellent copper absorption in aqueous solutions.		[221]
Siloxane-modified methacrylic	In situ polymerization	-	Better sun light protection, hydrophobicity and durability. Used for stone materials coatings (monuments, artifacts, etc.).		[224]
Silicone rubber	Melt compounding	-	Better erosion, arcing resistance and tracking. Used for outdoor insulators.		[225]

2.4.4 Layered Double Hydroxide

Layered Double Hydroxides (LDHs) are inorganic clays containing positively charged metal hydroxides together with multivalent anions, achieving thus, neutrality. These compounds are Zn/Al, Mn/Al, Fe/Al, Li/Al and Mg/Al. The highly tunable properties, low cost, thermal stability and chemical versatility make them attractive for hybrid nanocomposites using carbon nanofillers: CNTs, CNFs, and graphene [226]. These hybrids present unique properties, particularly for thermal stability. For instance, graphene and LDH nanocomposites are used as magnets, supercapacitors, water treatment, and medical applications. They are

mostly incorporated in different polymers in order to improve fire retardancy properties and to enhance graphene's properties in this direction [226]. Table 22 compiles the information about the polymer matrixes, processing methods of composites, modification agents of the LDHs nanofillers, the composite properties and applications for LDHs.

Table 22 – LDHs combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	LDH type	Preparation method	Modified/mixed with	Properties-applications		Ref.
Polyethylene terephthalate (PET)	MgAl	In situ polymerization	Modified with sulfanilic acid salt (SAS)	Improved thermomechanical properties, crystallization behaviour and gas barrier properties. Uses in electronic-packing materials, food and beverage packaging.		[227]
PP	MgAl	One-step melt mixing	Ethylene vinyl acetate copolymer (EVA)	Increased storage modulus and improved thermal stability.		[228]
PA	ZnAl	In situ polymerization	-	Thin film membranes with increased hydrophobicity for removal of natural organic matter from drinking water.		[229]
PS	Li/Al	In situ polymerization	-	Improved chemical and mechanical stability in water. Defluorination of water		[174]

2.4.5 A green approach of biodegradable hydroxide nanocomposites.

Table 23 presents different examples of LDH included biodegradable matrixes. In addition to the most commonly investigated biodegradable polymers, other ecofriendly materials like poly(hydroxyalkanoate)s (PHAs), have also been presented due to their excellent biodegradability, biocompatibility as well as good mechanical properties. A variation of this material, poly(3-hydroxybutyrate-co-4-hydroxybutyrate) (P(3,4)HB) improves the initial high crystallinity and lowers the melting point (<170°C) which improves the initial brittleness, and the thermal stability and processing temperatures. With the addition of CoAl LDH, the thermo-mechanical properties can be further improved [230]. Other types of LDH have also shown to improve the mechanical properties: e.g. Mg/Al-LDH for PBS [231], Zn3Al -LDH for PLA [232]. Boehmite nanoplatelets have not only shown improvement of mechanical properties, but also increased the biodegradability rate of biobased Poly(propylene sebacate) (PPSe) [233].

Table 23 - Novel research studies of biodegradable nanocomposites using nano hydroxide.

Matrix polymer name	Nano-hydroxide type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Magnetic alginate beads (LDH-nMABs)	Mg-Al-LDH nanoflake	-	-	Biodegradable, hydrophobic and magnetic sensitivity. Used for water fluoride removal.		[234]
PLA	Stearate-Zn3Al LDH	Solution casting method	-	Significant improvement for elongation at break as well as an increased rate of degradation in soil.		[232]
PBS	Mg/Al-LDH	In situ polymerization	-	Superior mechanical properties.		[231]

P(3,4)HB	CoAl LDH	Melt intercalation	Modified with sodium stearate	Better thermal stability, enhanced storage modulus. Plastic in food packaging industry, tissue engineering, agricultural applications.		[230]
PPSe	Boehmite (BM) nanoplatelets	In situ polymerization	-	Enhanced mechanical properties and biodegradability. Shape memory properties at body temperature. Biomedical applications, such as stents, in human body		[233]
Blends of PLA and PCL	Boehmite (BAI) nanoparticles	In situ polymerization	-	Enhanced mechanical properties. Uses in medicine, engineering, packaging industry, and agriculture.		[223]

2.5 Carbides

Nano carbides are compounds of carbon and metal or semimetal. They present high melting temperatures and are usually used as refractory materials. Due to the similarities in the crystalline structure with diamond of silicon and boron carbide, they present the highest hardness, providing the polymer matrix better wear resistance [1]. The most commonly used nano carbides are silicon (SiC), boron (B₄C), titanium (TiC) and zirconium carbide (ZrC). SiC has high mechanical thermal and electrical properties, and they exhibit great biocompatibility and its hardness makes it attractive for erosion resistance coatings[235], [236]. As can be noticed from the examples in the Table 22, it has been used for improving the dielectric properties, however they are used in hydrogels for water decontamination [236]. B₄C showing similar properties as SiC, has been also intensively used to improve wear resistance in polymers. Additionally, B₄C has neutron absorbing properties rendering it suitable for nuclear applications [1]. In contrast to other nanofillers, Carbides are not commonly included in different polymer matrixes; however their applications have been explored for different coatings to improve wear resistance (PU, epoxy, see Table 22). TiC has been used is electrochemically inert and due to catalytic activity towards I₃ reduction, it has been investigated in combinations with poly(3,4-ethylenedioxythiophene) (PEDOT) for dye-sensitized solar cell applications [237]. With a similar effect as graphene, in combinations with PANI, it can improve the performance of electrode material for supercapacitors [238]. More examples are shown in Table 24 below.

Table 24– Silicon carbide (SiC), boron carbide (B₄C), and titanium carbide (TiC) in combinations with polymer matrixes: processing methods, modification, properties and applications.

Matrix polymer name	Carbide type	Preparation method	Modified/mixed with	Properties-applications		Ref.
Polycarbosilane (PCS) & PVP	SiC	Melt blending	Layered carbon	Excellent dielectric loss properties tunable dielectric and broadband electromagnetic wave absorption. Used for electromagnetic shielding in wireless devices.		[239]
PS	SiC	Melt blending	-	Enhanced thermal and optical properties. Used in microwave dielectrics, power energy storage materials,		[240]
Cross-linked polyethylene (XLPE)	SiC	Melt blending	-	Improved DC breakdown strength, increased real and imaginary permittivity, suppression of space charge		[241]

Polyurethane (PU)	B ₄ C	Mechanical mixing	-	Enhanced erosion wear resistance in silt erosion conditions. Used for coating in order to avoid erosion in turbine blades.		[235]
Epoxy	B ₄ C	Mechanical mixing	PbO	Improved mechanical and thermal properties by ultrasonic irradiation.		[242]
Epoxy	B ₄ C	Mechanical mixing	Clay	Increased Young's modulus, decrease of tensile strength, better thermal stability. Used for neutron shielding application.		[243]
Polyphenylene sulfide (PPS)	TiC	In situ polymerization	-	Enhanced tribological and mechanical properties.		[244]
Polyimide (PI)	TiC	In situ polymerization	PVP	Enhanced mechanical properties and electrical breakdown strength.		[245]
Ppolypyrrole (PPy)	TiC	In situ polymerization	PVA	Enhanced mechanical properties. High-power supercapacitor material.		[246]
Regenerated bacterial cellulose (RBC)	TiC	-	-	Enhanced bactericidal activity. Used for wound healing and tissue regeneration applications.		[247]
Polypyrrole (PPy)	TiC	Heterogeneous nucleation process.	-	High-rate (up to 400 C-rate) charge-discharge capability and cycling stability of Li-ion batteries.		[248]

2.5.1 A green approach of biodegradable carbide filled nanocomposites

Table 25 collects two relevant studies for biodegradable versions of nano carbide reinforced nanocomposites. No studies were found for biodegradable options for boron and titanium carbide nanoparticles.

Table 25 - Novel research studies of biodegradable nanocomposites using carbides.

Matrix polymer name	Carbide nanofiber type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
PLA	SiC	Solution-blending method	Surface-modified SiC (S-SiC)	Enhanced impact strength; more ductile fracture.		[249]
Gum karaya (GK) hydrogel grafted with PAA	SiC	In situ graft copolymerization method		Increased strength and adsorption rate capacity of cationic dyes from contaminated water.		[236]

2.6 Organic nanofillers

Many industries have started to search for alternatives to synthetic nanocomposites by using natural nanofillers that can provide polymers not only features such as enhanced mechanical properties, light weight, low cost, but also the advantages of recyclability, biocompatibility, renewability and non-hazardous properties [250]. The important information about some of the most important organic nanofillers are collected for the recent literatures and presented in the sections below.

2.6.1 Cellulose

Cellulose, a linear macromolecule, is the most abundant polymer on Earth. Cellulose originates in plants, algae, certain type of bacteria, tunicates, etc. In addition, recent research provide methods of using waste paper for the extraction of cellulose nanofibers [251]. This bio-based nanofiller can be used as reinforcement for multiple polymer matrixes in order to achieve higher stiffness, strength, and biodegradability. The process of extraction of cellulosic fiber, either mechanically or chemically induced, produces nanomaterials called cellulose nanofibrils (CNFs) and cellulose nanocrystals (CNCs) respectively. While CNFs present high aspect ratio flexible filaments, CNCs occur as rod-like acicular nanoparticles [252]. Cellulose nanocrystals (CNCs) have been recently proposed also as an alternative to synthetic nanofillers such as carbon nanotubes. CNCs have hydrophilic properties, which makes them ideal to combine with hydrophobic polymer matrixes. Some of the applications of these fillers include pharmaceutical, biomedical implants, drug delivery, batteries, super capacitors, and antimicrobial membranes, etc. Recent studies have started to focus also on the light scattering properties of CNCs [253]. The study presented in [250] provides a summary of almost forty different polymer matrixes CNC based nanocomposites and, the corresponding rheological properties. The important information on CNCs from the recent literatures are collected and presented in Table 26.

In order to produce CNCs and CNFs filled composites, melt processing approaches such as extrusion, hot pressing, and injection molding have been widely used. . As CNFs present higher probability of entanglement of the former in such processing methods, a reduced percolation threshold could be expected. Polymers such as poly(ethylene oxide) (PEO), poly(ethylene glycol) (PEG), Polylactic acid (PLA) as well as polypropylene (PP) have been extensively used in combination with these nanofillers. Different processing techniques of other polymer matrixes are presented in the following study: [252]. However, alternative processing such as casting and evaporation have proven to create the most efficient and well-performing cellulosic nanocomposites [252].

Table 26 – Cellulose nano-crystals (CNC) combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Poly(soybean acrylate) (P1) and poly(soybean methacrylate)	Solution mixing method	hydroxyl –OH and carboxyl –COOH groups via thiolene	Stimuli responsive material (water-triggered modulus switching). Applications: packaging.		[254]
Triblock copolymer (PEO–PPO–PEO)	Melt extrusion	-	Thermal stability, improved mechanical properties, hydrophobic properties.		[255]
Polydimethylsiloxane (PDMS)	In situ polymerization	PVA	Light Management in Optoelectronic Devices Organic light-emitting diodes Light absorption enhancement in thin-film solar cells.		[253]
NR in latex form	Hand lay-up method	-	Higher tensile strength		[256, 257]

2.6.2 Nanolignin

While cellulose is an insoluble substance constituting the cell walls of vegetable fibers, lignin is an organic substance that binds the cell fibers and vessel elements in side woods. Lignin nano particles has been used to reinforce natural rubber or synthetic polymers in order to increase thermal stability, strength, Young's modulus [258], and anti-oxidant and microbicidal properties [259]. Recent research proposes using nano-ligning for partially replacing carbon black in natural rubbers to improve their mechanical and thermal properties, thermo-

oxidative stability and providing a faster vulcanization. The benefit of these nanofillers over carbon black is that lignin is less dense, non-electrically conductive and lighter in color [260] . More information about Nanolignin is resented in Table 27.

Table 27 – Nano-lignin in combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Hydrogels: (graphene, polyacrylic, l amide-based, lignosulfonate, pectin etc.)		-	Removal of various pollutants from water due to high absorption properties.		[261]
Natural rubber	Co-precipitation of colloidal lignin-cationic polyelectrolyte complexes and rubber latex	Poly (diallyldimethylammonium chloride) (PDADMAC)	Accelerated vulcanization. Enhanced thermal stability and mechanical properties.		[260]

2.6.3 Chitin

Chitin is another abundant polymer found in nature, which makes it ideal for many biomedical applications. Chitosan is a deacetylated product of chitin and used in combination with other polymer matrixes. The corresponding composites present better mechanical strength, water absorption capacity, biocompatibility, biodegradability, etc. Moreover, extensive research studies have been done in using these nanofillers in tissue engineering, replacing or restoring defective tissue [262]. In order to improve their antibacterial properties, modifications with most effective antimicrobial agents such as silver nanoparticles have also been proposed [263]. Xylan chains have also been added to chitosan nanocomposites, due to their antibacterial and antioxidant properties [264]. Table 28 presents the Chitin's possible combinations with polymer matrixes, the processing methods, modification, properties and applications.

Table 28 – Chitin in combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications		Ref.
Sodium hydrogen carbonate	Magnetic stirring	PLGA nanoparticles	Higher elastic modulus. Thermosensitive hydrogels. Antibacterial, antifungal, mucoadhesive, analgesic haemostatic, biodegradable, biocompatible properties.		[265]
PVA hydrogel	Solution casting method.using gluteraldehyde as a crosslinking agent	ZnO	Increased antimicrobial activity (ZnO). Tensile strength depending on cross linking agent. Applications: packaging sheets, membrane filtration, biomedical material for controlled release, tissue regeneration etc.		[266]

2.6.4 Nanostarch

Starch nanocrystals can be found abundantly in plants and it has been used for the preparation of biocompatible films. Nano starch has antibacterial properties against Staphylococcus aureus and Pseudomonas aeruginosa. In addition, Nanostarch fillers shows other remarkable properties such as swelling reduction, barrier and blood compatibility properties which makes it ideal for biomedical applications such as wound

healing [267]. The use of nanostarch is currently limited due to the high cost of their extraction and processing. Preparation methods with different matrixes have been successful in the case of NR or styrene butadiene rubber latex, , a powder form for commercial sale has not been yet achieved to be used for melt processing techniques [8]. Table 29 presents the summary of the current research studies on the Nanostarch.

Table 29 – Starch nanocrystals combinations with polymer matrixes: processing methods, modification, properties and applications

Matrix polymer name	Preparation method	Modified/mixed with	Properties-applications		Ref.
PVP	Solution casting method.	-	higher tensile strength and Young's modulus Reduced oxygen permeability, antibacterial activity. Wound dressing applications.		[267]
Plasticized starch matrix	Solvent-casting method.	-	Better mechanical properties. Reduced moisture absorbtion. Hydrophilic properties. Shape memory properties. Biomedical application.		[268]
Plasticized starch matrix	Solvent-casting method.	-	Enhanced mechanical properties. UV resistance. Thermal resistance. Applications: sustainable packaging.		[269]

2.7 Other nanofillers

Many other nanofillers have captured the attention in recent research, e.g. Polyhedral oligomeric silsesquioxane (POSS), which provides high thermal stability, flame retardancy, better mechanical properties, low dielectric properties, etc. It can be part of green nanocomposites when combined with polymers such as cellulose, providing improved wettability, better UV aging and UV resistance/shielding [270]; It has been part of novel composites membranes combined with chitosan, used for guided bone tissue regeneration [166]. Recent combinations include PA matrix, for improved thermal stability and surface hydrophobicity [271], PMMA for tuning properties of transparency [272]; Hybrid copolymers of N-isopropyl acrylamide (NIPAm) for thermally-responsive hydrogel behavior [273], etc. Silver is another widely used nanoparticle, generally preferred due to its antimicrobial properties against bacteria, fungi and viruses. Surprisingly, in the form of nanoparticles, presents even better properties than particles with larger diameter [274]. Furthermore, Gold nanoparticles therapeutics, sensor, therapeutic agents, drug delivery in biological and medical applications, imaging, electronics, etc. [275]. Table 30 presents some applications of the nanofiller reinforced composites.

Table 30 –polymer nanocomposites filled with other nanofillers: processing methods, modification, properties and applications

Matrix polymer name	Nanofiller	Preparation method	Modified/mixed with	Properties-applications		Ref.
poly(ϵ -caprolactone) (PCL)	Silver	Solution casting method.	Reduced graphene oxide	Fracture fixation devices and tissue engineering.		[157]
Chitin	POSS	Solvothermal synthesis	-	tissue engineering, enzyme immobilizations, waste water treatment		[276]
PMMA	BaTiO ₃	Polymerization and dispersion via heat	-	Display technologies.		[277]

		treatment				
Poly(dimethylsiloxane)	Gold	Thermal polymerization		Microfluidic and optofluidic applications		[278]

2.7.1 A green approach of biodegradable nanocomposites with other type of fillers.

Table 31 presents some of different types of nanofillers that have not yet been described in the previous sections but has high potential as biodegradable nanocomposites. The selected ones focus mainly on medical applications such as bone tissue engineering and cancer cell growth inhibition since there is an increased interest and demand of medical solutions to bone and cancer problems. These studies also present biodegradable and recyclable properties in line with the scope of this paper.

Table 31 - Novel research studies of biodegradable nanocomposites using other type of fillers.

Matrix polymer name	Nanofiller type	Preparation method	Modified/mixed with	Properties-applications	Field	Ref.
Poly(butylene succinate-co-terephthalate) (PBST) nanocomposites	Fibrous attapulgite (ATP- magnesium aluminum silicate clay)	In situ polymerization	-	Enhanced mechanical properties compared with only silica nanoparticles.		[279]
Gellan and xanthan polymers	Bioglass nanoparticles (SiO ₂ -CaO-P ₂ O ₅)	In situ polymerization	-	Tunable bone tissue engineering scaffolds in terms of mechanical stiffness, bioactivity and bacteriostatic properties.		[280]
PANI	Porous silicon nanoparticles (PSiNPs)	Surface initiated polymerization	-	Superior biodegradability and biocompatibility in vitro and in vivo. Uses: cancer cell growth inhibition.		[281]
Poly(propylene fumarate) (PPF)	Tungsten disulfide nanotubes	In situ polymerization	SWCNTs and MWCNTs	Better mechanical properties. Used for bone tissue engineering.		[282]
PBST	Fibrous attapulgite (ATP)	In situ polymerization	-	Enhanced mechanical properties.		[279]
Poly(butylene succinate) (PBSu)	Strontium hydroxyapatite nanorods	In situ polymerization	-	Enhanced mechanical properties, enzymatic hydrolysis and apatite formation. Osteoblast compatibility. Tissue engineering applications.		[283]

3. Summary and discussion

The paper presents a comprehensive list of nanofillers, list of compatible polymer matrixes for each of the nanofillers, novel polymeric nanocomposites and their preparation methods etc. from an application in advanced application view point. While nanofillers could enhance different properties in the polymeric matrix and produce multifunctional materials, they critically affected by the effectiveness of preparation method. Acquiring a proper dispersion and distribution of fillers define the properties and application of the produced

composites. There are different parameters in non-uniform dispersion of nanofillers such as high interfacial energy, high aspect ratio, incompatibility of the hydrophilic fibers and hydrophobic nature of polymer matrixes, etc. Moreover, a high concentration of fillers inside the matrix has a negative impact on micro structural defects. Most nanofillers presented in this paper could enhance one or a group of properties including mechanical, thermal, electrical, and tribological properties. The different properties and other relevant notes regarding each nanofillers group are discussed below.

Nanoclays increase the tensile, flexural modulus and yield strength of the polymeric composites. However, they reduce the maximum strain of thermoplastics, while increasing the elastic modulus, strength, and toughness of thermosets. Within elastomer matrixes, nanoclays decrease the relaxation rate but improve aging properties. It is also noteworthy to mention that they could act as an alternative to carbon black since they exhibit similar properties in lower contents. However, the particular influence of nanoclays is their improving influence on the thermal stability of the polymer matrix, making them an excellent choice for many fire-retardant applications. Nevertheless, studies that have been conducted on medical applications acknowledge them an attractive option of tissue engineering materials, drug delivery devices, and hydrogel with antimicrobial activity, etc.

Carbon, the second studied group of nanofillers presented a big impact on the mechanical properties of the polymer matrix by increasing toughness, young modulus and tensile strength of the corresponding composites. Carbon black as the oldest member of this group has been known for more than a century, however, the new members of this group attracted science and industry in the recent decades. CNTs are the most studied nanofillers in the recent years, however, new studies show that graphene can also provide a comparable enhancement in mechanical properties. One of the most important properties of carbonic nanofillers is their electrical properties. This property makes these fillers suitable for polymers with shape memory properties and various applications in electronic industries. In addition, the extraordinary thermal properties of Graphene fillers have already made them indispensable in future microelectronics devices. Nanodiamonds exhibiting the superior properties of diamond in nanoscale have also proved themselves influential in enhancement of mechanical, tribological, thermal properties.

Ceramic nanoparticles such as nano oxides provide comparable thermal properties as nanoclays. Membranes containing Silicon Dioxides can be also used for water treatment. Other films containing the same filler have also exhibited hydrophobic properties. Titanium dioxide, presents self-cleaning properties which makes them attractive for biomedical applications, hospital clothing, and surface coatings for medical devices etc. Moreover, hydroxide nanoparticles have been extensively studied in the last decade. Although this paper presented three examples, there are many other hydroxides used for nanocomposites, such as: yttrium hydroxide $Y(OH)_3$, layered double hydroxide (LDH) of different metals, etc. Most of them, are used for fire retardancy applications. There are also novel research studies that use boehmite for other applications such as sun light protection coatings, outdoor insulators, etc. In addition, carbides are used in coatings for high temperature applications, since they have very high melt temperatures. Silicon carbide is used to increase wear resistance, while boron carbide has been studied for neutron absorption nanocomposites, ideal for nuclear applications.

Organic nanofillers are of huge interest at present due to their biocompatibility and their potential for medical applications. A good number of studies have been conducted in the recent past to increase the biodegradability of the polymer composites to eliminate their negative environmental impacts. An example is the recent discovery of *Ideonella sakaiensis* 201-F6 bacterium, which has the ability to use PET as its main carbon and energy source for growth, transforming this polymer into a possible biodegradable alternative [284]. PET is one of the most popular synthetic polymers for the production of containers for liquids and foods as well as textiles. Although recyclable, this plastic it has been accumulating, and its resistance to biodegradability has raised serious concerns due to its effect on the growing risk of fauna and flora, mainly in marine environments. This paper gives examples of multiples biodegradable nanocomposite alternatives: PLA, PHB, aromatic copolyesters such as PBAT, polyethers, PA, PU, chitin, chitosan, PC, and PHA. In addition, there are many other biodegradable polymers as green alternative for future polymer nanocomposites such as

PLC, PGA, PVL, Poly(-decalactone), Polyhydroxyvalerate (PHV), Poly(-malic acid) (PMLA), Poly(ortho esters) (POE), Poly(amide-enamines), Polyanhydrides, Polysaccharides (naturally occurring starches and cellulose), Polycyanoacrylates, Polyketals, Polyphosphoesters, Polyacetals, Polyphosphazenes (PPHOS), Poly(imino-carbonates), Poly(1,4-dioxan-2-one) (PDO) [285].

4. Conclusions

Polymeric nanocomposites are one of the most interesting and promising topic in the area of polymer science, chemistry, nanotechnology, etc. In fact, the advent of the nanofillers opened new doors to new applications and technologies not existed before. Moreover, the new composites are replacing many traditional materials in different industrial sections. The multifunctional properties introduced by these novel materials are key features in many advanced products. Industrial applications from medical micro components to aerospace products are redefining their design to benefit from the development of nanocomposites. However, it seems that many achievements might have been missed among these huge amount of data and research studies. This review papers endeavored to categorize some part of this significant progress with respect to their applications in the different industrial sections and give a guideline for the easy selection of nanocomposites for industrial applications. The most prominent nanofillers are introduced based on their geometrical characteristics and enhancement features. Their applications in the different polymers and preparation methods are discussed. However, application of many of these composites might have undesired negative environmental impacts. To produce and dispose nanofillers and nanocomposites in environmentally safe ways are vital. Therefore, this paper focused particularly on green and biocompatible composites to allow the readers to find alternatives to conventional nanocomposites for providing minimal environmental impact in connections with the applications of nanocomposites into industrial products.

In conclusion, new technologies are emerging and processing nanomaterials and composites showing novel properties and improved performances. Nanocomposites are potential materials to meet the emerging demands arising from scientific and technologic advances. Processing methods for different types of nanocomposites are available, but some of these provide challenges which give the opportunities for further researchers and developments. A good number of applications of nanocomposites already exist, but more widespread applications of nanocomposites are possible in the future provided some technical challenges are overcome; health, safety and environmental concerns related to nanocomposites are addressed. The release of nanoparticles into the environment is a big concern for health and safety. Hence there is an increasing need for research for the emission control of nanocomposites and nanoparticles. The green or environmentally friendly nano composites seem to be the future of nanomaterials and their world-wide applications in the industrial products. So the future research should focus on the development of green nanofillers, nanocomposites and their novel applications.

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