

Applications of Clays in Nanocomposites and Ceramics

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Abstract: Clays and clay minerals are common natural materials, the unique properties of which have attracted the interest of the industry, especially because these materials are easily available, cheap, and non-toxic. Clays and clay minerals are widely used in many applications, such as in ceramic production, in the clarification of liquids, pollutant adsorbers, filler in composites and nanocomposites, soil amendments, in pharmacy, etc. This review assesses the development in the area of clay application in nanocomposites and ceramics. The first part of this study covers polymer/clay nanocomposites. Topics of interest include nanofiller sources for polymer nanocomposites, the possible ways of clay modification, polymer/clay nanocomposite classification and their processing, and polymer matrix overview with possible enhancement of nanocomposite properties. Some of the applications have already been commercialized. Approximately 80% of the polymer/clay nanocomposites are destined for the automotive, aeronautical, and packaging industries. The second part of this study describes ceramic materials with a focus on silicate ceramics. Talc and kaolinite represent the main natural raw materials for traditional ceramic applications. Less traditional cordierite, steatite, and forsterite could offer property enhancement and seem to be useful in electronics, electrical engineering, catalysts, solar thermal storage, or medical applications.

Keywords: clay; montmorillonite; nanofiller; ceramics; nanocomposite



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1. Introduction

In the last century, the research in the area of nanotechnology started new technological development. The term “nanotechnology” was used first by the Japanese scientist Norio Taniguchi in 1974 [1]. Nanotechnology works in the area of incredibly small dimensions, in the range of 1–100 nm. A nanometer (nm) is 10^{-9} m, smaller than the wavelength of visible light and a hundred-thousandth the width of a human hair. In some senses, nanoscience and nanotechnologies are not new. Nanostructured and nanocomposite materials are commonly found in nature and living beings (such as bone) [2]. Also, chemists make polymers, which are large molecules made up of nanoscale subunits [3].

“Synthetic” polymer/clay nanocomposites have their origin in the pioneering research at Toyota Central Research Laboratories. In addition, the first practical application of nanocomposites, the nylon/montmorillonite timing belt cover on the Toyota Camry automobile, relates to this company as well. Nowadays, the PA/clay nanocomposites have the largest commercial presence in the application field. The global nanocomposites market size was USD 5.6 billion in 2022 and is likely to reach USD 18.3 billion by 2031 [4]. The market growth is attributed to the rapid increase in the use of nanocomposites in biomedical applications and packaging. Further, rising demand for nanocomposites has been recorded also in the sectors of aviation, sporting goods, and automobile parts [4].

Despite the development in the area of polymer nanocomposites and their expanding use in different applications, research in polymer science and nanotechnology continues.

Especially, it is necessary to understand the principles of nanocomposite behavior at the nanometric level.

Clays and clay minerals have been accompanying humans since the dawn of history. Clays and clay minerals, raw or after modification, have great importance in a wide variety of applications mainly due to their abundant, inexpensive, inertness, stability, reactivity, and environmentally friendly [5,6]. Clays are naturally occurring materials consisting primarily of the various clay minerals content and degree of purity [5,7–9]. Clay minerals belong to the phyllosilicate group with a layered structure and with one dimension in the nanometer range [5,9]. The principal building elements of the clay minerals are two-dimensional sheets of silicon-oxygen tetrahedral and two-dimensional sheets of aluminum- or magnesium-oxygen-hydroxyl octahedral [10]. Individual clay minerals, such as kaolin, clay, bentonite, and vermiculite, differ significantly in their composition and crystal structure which causes different physical and chemical properties (e.g., particle size, surface chemistry, surface area, viscosity, plasticity, absorption, and adsorption) [7,8,11]. In many cases, the clays or clay minerals can be modified to obtain improved mechanical, thermal, structural, or functional properties.

Clays and clay minerals have many industrial applications such as ceramics [12,13], paper coatings [14,15], pesticides [16], paints [17,18], pharmaceuticals [19,20], agriculture [21,22], construction industry [23], ion exchangers, separators, plastics [24–26], cosmetics [27], insulations [28], and electrical applications. Clay minerals can also be used as a good adsorbent for water purification due to lamellar structure, high cation exchange capacity, pore size distribution, and large surface area [29]. Moreover, considerable attention obtained the clay composite or nanocomposite materials which can be incorporated into many areas such as biomedical, construction, automobile, remediation technology, petroleum industry, wastewater, treatment, aerospace, and nanotechnology [30].

In ceramic technology, the most important and most widely used natural raw materials also belong to clay minerals such as kaolinite, illite, montmorillonite, talc, pyrophyllite, and serpentine [31]. Generally, ceramic materials show a combination of useful properties such as high strength and stiffness at very high temperatures, chemical inertness, and low density. Their applications are restricted owing to their brittle behavior. Cordierite, enstatite, and forsterite are silicate materials that form the main components of the ternary system $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$. One of the methods for the synthesis of these ceramic types is the sintering of natural raw materials including clay and clay minerals, especially kaolinite, talc, and a combination of both minerals. The important properties relating to applications of clay minerals in the ceramic industry are plasticity, chemical and mineralogical composition, thermal properties, color, and mechanical strength after firing [32]. Extensive research is also carried out in the field of ceramics. Nanoceramics have emerged as valuable materials in biomedicine and medical technology (orthopedics and bone tissue engineering). In bone repair, nanoceramics serve as nano scaffolds to support and facilitate bone growth. Applications in energy storage, coating systems, environmental technology (water treatment), chemistry, construction, electronics, and batteries have also been reported [33].

This review is divided into two main parts. The first part describes the use of clays and clay minerals in nanocomposites mainly in polymer/clay nanocomposites and their applications. The second part, concerning ceramic materials, provides a summary of clay and clay minerals used in pre-ceramic mixtures, types of final silicate ceramics, and their applications.

2. Polymer/Clay Nanocomposites

2.1. Nanofiller Sources

Nanoparticles have at least one characteristic length scale that is of the order of nanometers and can range from isotropic to highly anisotropic needle-like to sheet-like elements. These nanoelements can lead to ultra-large interfacial areas between the constituents. In addition, the distance between the nanoelements begins to approach molecular dimensions at extremely low loading of the nanoparticles. This large internal interfacial area and the

nanoscopic dimensions between constituents differentiate polymer nanocomposites from traditional composites [34]. Nanofillers can be different types of materials like carbon nanotubes, fullerenes, carbon black, polyhedral oligomeric silsesquioxanes (POSS), silica, and phyllosilicates (montmorillonite, halloysite, and vermiculite). MXene, nanofibers, metals, and their oxides are also used. One of the most common sources of nanofillers is a clay mineral called montmorillonite (MMT).

Montmorillonite

Montmorillonite has become one of the most widely used minerals as nanofillers, because of the versatility of reactions, layered morphology with a high aspect ratio, and large specific area, which offers substantial cation exchange capacities. Additionally, MMT is commercially available.

MMT is a naturally occurring mineral derived from the weathering of volcanic ash. This mineral belongs to the clay minerals of the smectite family. MMT represents 2:1 layered aluminous-silicate. The suggested crystallographic structure for montmorillonite is in Figure 1 [35]. Isomorphous substitutions of Si⁴⁺ for Al³⁺ in the tetrahedral lattice and of Al³⁺ for Mg²⁺ in the octahedral sheet cause an excess of negative charges within the montmorillonite layers. All atoms may also be replaced by Fe³⁺, Ti, Ni, Zn, Cr, and Mn [36]. In many minerals, an atom of lower positive valence replaces one of higher valence. These negative charges are counterbalanced by cations such as Ca²⁺ and Na⁺ situated between the layers.

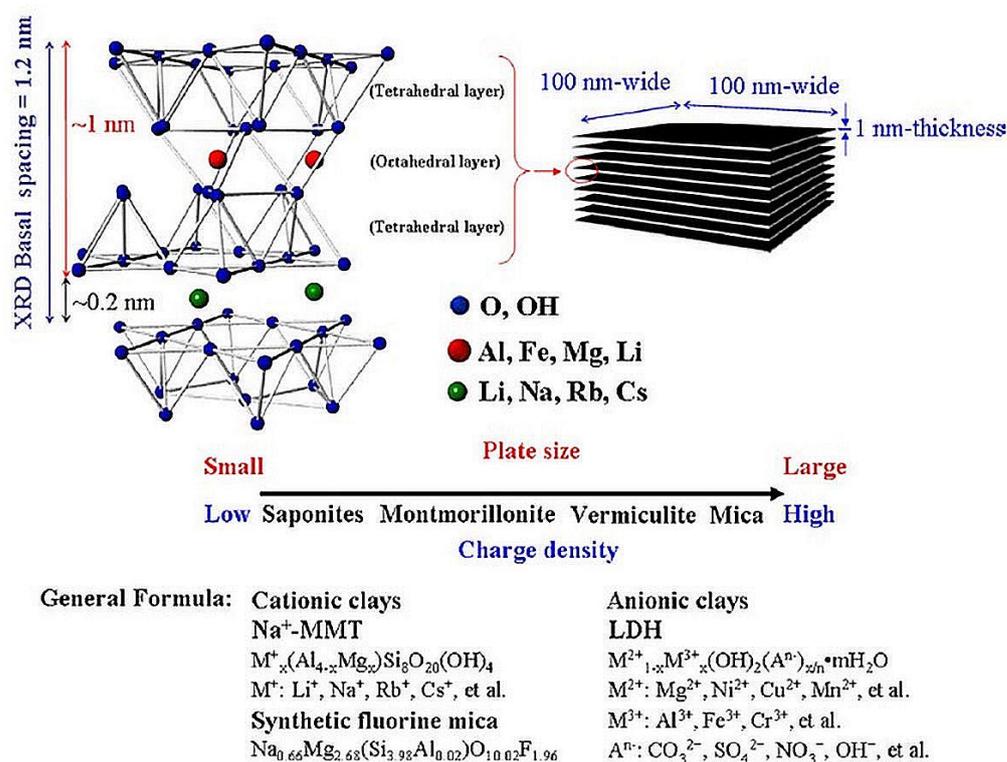


Figure 1. Chemical structure of smectite clay [35].

Due to the high hydrophilicity of montmorillonite, water molecules are usually also present between the layers. Stacking of the layers leads to regular van der Waals gaps called interlayers or galleries. The sum of the single layer thickness and the interlayer represents the repeat unit of the multilayer material, so-called d-spacing or basal spacing.

2.2. Clay Organophilization

The main difficulties for polymer/clay nanocomposites relate to the hygroscopic character of clay and clay minerals. It is relatively simple to disperse clay in water or water-

soluble polymer monomers, but clay dispersed in a high molecular hydrophobic polymer is like trying to mix oil in water. The reason for such behavior is the different nature of these two materials as was mentioned above. Organophilic polymers are not miscible with pristine hydrophilic clay represented in our case by montmorillonite (MMT). This phenomenon is attributed to the higher surface energy of MMT compared to a macromolecular matrix which tends to create a stronger cohesive interaction between clay layers and further hamper the inclusion of polymer chains into the interlamellar region of MMT [37–41]. Therefore, surface modification of MMT plays a very important role in clay incorporation into the polymer matrix. The process leading to nanocomposite is called delamination or exfoliation of montmorillonite to individual sheets. On the other hand, the hydrophilic feature of the MMT surface permits water and other polar molecules to intercalate into the galleries within clay layers [37].

The clay modification process, called intercalation (1 chemical agent) or co-intercalation (2 or more chemical agents), can be defined as the reversible inclusion of a molecule or ion into filler with a layered structure. In the case of layered aluminous silicates (phyllosilicates), intercalation means intercalant penetration into the clay-layered structure. The result of such a successful process is an increase in d-spacing, Figure 2. Next, the changes depend on the type of used intercalant, its concentration, and the length of the chain. The clay affinity to the polymer surface is influenced and the intercalation more compatible montmorillonite-polymer interface can be then developed.

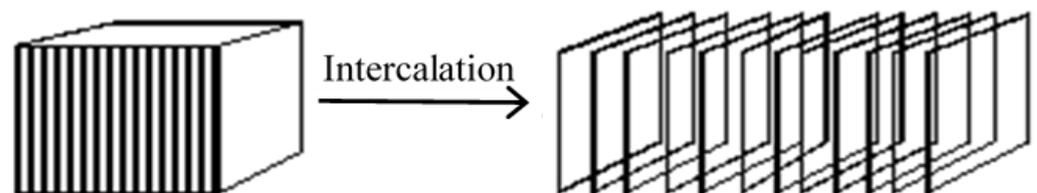


Figure 2. Process of montmorillonite intercalation.

Amino acids were employed as the first intercalants in the synthesis of nanocomposites (polyamide 6-clay hybrids) [42]. Numerous other kinds of intercalant agents have been used in the synthesis of nanocomposites today's. The most popular are cationic surfactants, such as alkylammonium ions, because they can be exchanged easily with the ions situated between the clay layers. All the commercial types of montmorillonites are based on these substances. This kind of surfactant consists of two distinct parts, a positively charged hydrophilic head and a hydrophobic hydrocarbon chain tail. After modification, cationic surfactant molecules attach to the inner and outer surface of clay minerals and thus alter the surface properties of clay minerals from hydrophilic to hydrophobic [43]. In addition, silanes have been used because of their ability to react with the hydroxyl groups situated at the surface and the edges of the clay layers.

Smectite organically intercalated structures were first studied by Lagaly and Weis in 1969. After modification, several arrangements inside the interlayer are possible Figure 3. Lagaly and Weis found two possible arrangements of organic molecules in the interlayer, namely lateral and perpendicular (paraffin) [44,45].

At present three basic methods of intercalation are performed:

1. Ion-exchange method is based on the MMT's ability to sorb some types of cations and to keep them in the charge state [46,47].
2. Ion-dipole method is based on the ion-dipole interaction of an organic intercalant and an interlayer cation [48]
3. Grafting, the formation of a covalent linkage between the clay platelet and the hydrophobic part of the coupling agent [37].

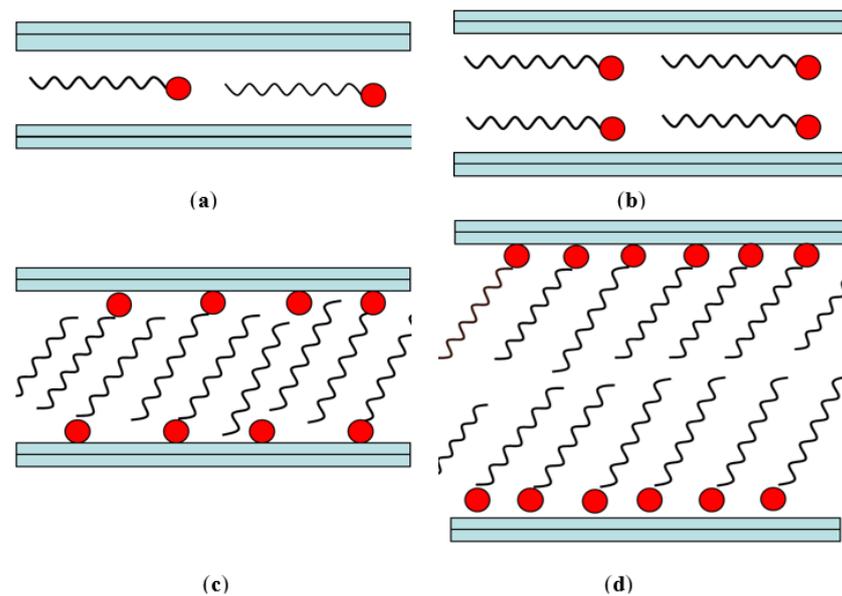


Figure 3. Alkyl chain arrangement in layered silicates: (a) lateral monolayer, (b) lateral bilayer, (c) paraffin (monolayer), and (d) paraffin type bilayer.

2.2.1. Ion-Exchange Intercalation

A characteristic feature of smectites such as montmorillonite is their ability to sorb certain cations and retain them in an exchangeable state. It means that these intercalated cations can be exchanged by treatment of other cations in a water solution (wet method), Figure 4. The most common exchangeable cations are Na^+ , Ca^{2+} , Mg^{2+} , H^+ , K^+ , and NH_4^+ . Indeed, if the clay is placed in a solution of a given electrolyte, an exchange occurs between the ions of the clay (X^+) and those of the electrolyte (Y^+):

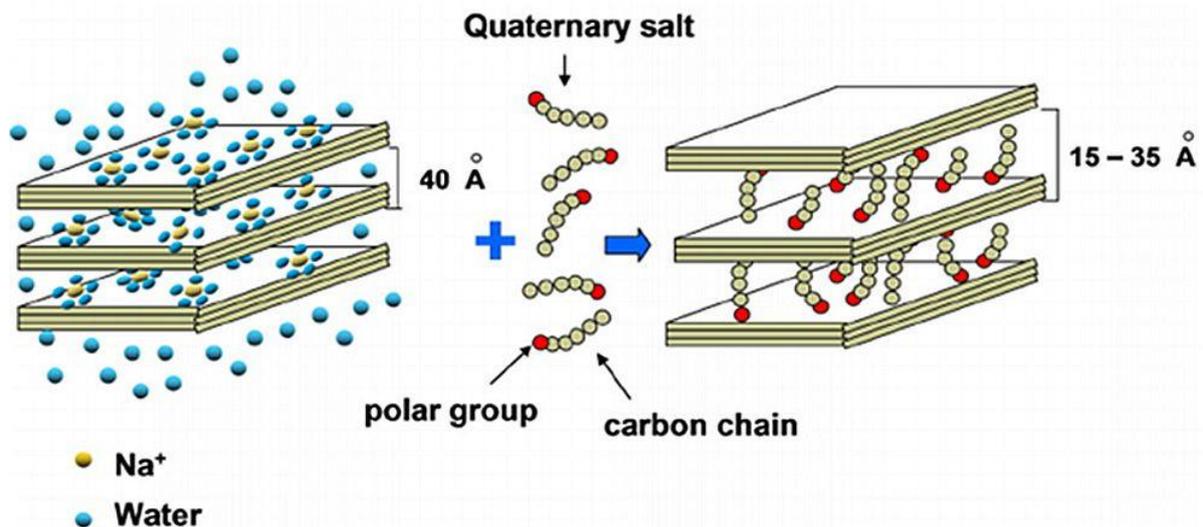
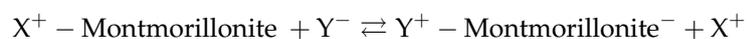


Figure 4. Development of organoclay treated with quaternary salt: ion-exchange intercalation method [49].

For given clay, the maximum number of cations that can be taken up is constant and known as the cation-exchange capacity (CEC). CEC is measured in milliequivalents per gram (meq/g) or more frequently per 100 g (meq/100 g). Cation-exchange capacity measurements are performed at a neutral pH of 7. The CEC of montmorillonite varies from 80 to 150 meq/100 g [50]. In the dependence on the CEC and the character of organocation, different structures can be created in the interlayer space. These structures could be helpful

during the nanocomposite compounding and for the right choice of intercalant. The successful MMT intercalation and following exfoliation process during compounding is a good assumption for enhancement of final materials properties.

2.2.2. Ion-Dipole Intercalation

Ion-dipole method is based on ion-dipole interaction. This method differs from the ion-exchange approach in that the exchangeable cation (partial positive charge) remains on the clay surface, Figure 5. It means that it is not necessary to ablate any product of a chemical reaction. Next ion-dipole approach advantage relates to the non-water environment (dry method). As the intercalants, alkylamine (octadecylamine (ODA), dodecylamine (DDA)), and primary amine can be used. As the compatibilizer the common plastics processing aid, like plasticizers, lubricants, and other modifiers, could also be suitable [51–53]. The intercalate structure depends on the concentration of used organic intercalant, in the guest-guest and guest-host interactions. This technique is also possible to apply to process co-intercalation [54,55].

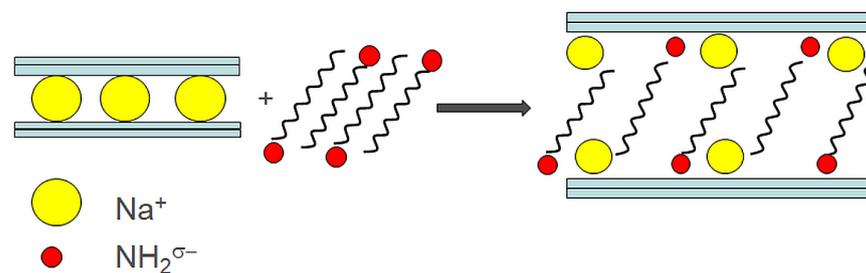


Figure 5. Ion-dipole intercalation method.

2.2.3. Grafting

The grafting approach has attracted scientists over the last 15 years. The method is based on the formation of covalent bonds between the montmorillonite platelet surface and the hydrophobic modifier. This method can improve the stability of organophilized clay surfaces. The most famous modifiers are silanes, therefore this process is called silanization or silylation. Alkoxysilane and chlorosilane are the most common intercalates. However, chlorosilane is not used too often due to its tendency to create HCl during the grafting process [56]. In addition to increased thermal stability, clay treatment by this approach offers irreversible coupling and tightly secures the agent on the clay surface, avoiding release into the environment to cause adverse effects [57]. Organosilanes also serve as a key bridge to enhance the interfacial interaction between the silylated-MMT and the polymer matrix owing to the reduced clay surface energy because of silane grafting, thus enabling better dispersibility of the reinforcing phase in the continuous matrix [37,58].

2.3. Clay Application in Nanocomposites

Generally, nanocomposites represent materials with multiphase ultrafine structures with at least one dimension 10^{-9} m. In the case of polymer/clay nanocomposites, they are formed through the connection of two different materials, organic (polymer) and inorganic (mineral). The next important features of polymer nanocomposites are low content of filler (1%–5%) compared to conventional composites (30%–50%), transparency, and large changes in material properties, like E-modulus, strength, shrinkage, density, chemical and fire resistance, etc.

The polymer nanocomposites could be divided into several groups according to the dimensions of the dispersed nanoscale:

1. Two-dimensional (2D) nanomaterials—2 dimensions in macroscale (layered silicate [59–61], graphene [62,63] or MXene [64,65])—lamellar nanofillers in the form of sheets of one to a few nanometres thick and hundreds to thousands of nanometres long and wide).

2. One-dimensional (1D) nanomaterials—2 dimensions in nanometres and the third is larger (nanofibers or nanotubes, e.g., carbon nanofibres and nanotubes [66] or halloysite nanotubes [67–69]—fibrillar nanoscale).
3. Zero-dimensional (0D) structures—0 dimensions in macroscale (spherical silica [70,71], semiconductor nanoclusters [72] and quantum dots [73], and isodimensional spherical particles) [74,75].

One of the processing problems of nanocomposites is the nanofiller exfoliation during processing. Therefore, polymer/clay systems are divided into four general groups according to the nanofiller exfoliation level (Figure 6):

1. Microcomposite, where the clay acts as a conventional filler. The final material belongs to traditional composite materials.
2. Intercalated nanocomposite consists of a regular insertion of the polymer between the clay layers. The final material belongs to nanocomposites.
3. Intercalated and partially delaminated nanocomposites, an intermediate step between intercalated and exfoliated structure. The final material belongs to nanocomposites.
4. Exfoliated nanocomposite where the filler is delaminated to 1 nm-thick layers. The final material belongs to nanocomposites.

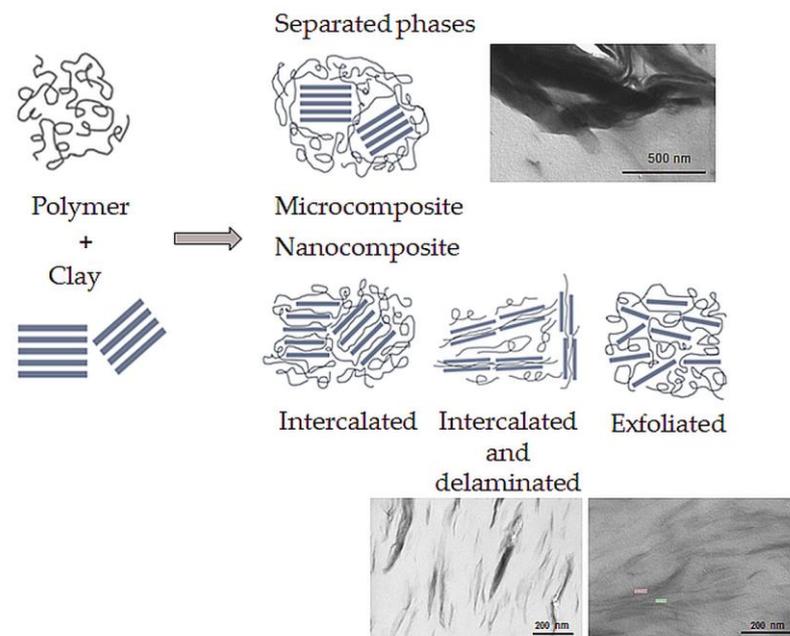


Figure 6. Polymer/clay nanocomposite classification.

2.3.1. Polymer/Clay Nanocomposite Processing

The final step of nanocomposite preparation is the organoclay mixing with polymer. Early experiments with clay-filled polymers required processing that was not commercially friendly, but this situation has changed. A primary difficulty is the proper dispersion of the filler in the polymer matrix. Without good dispersion and filler distribution, the high surface area is compromised, and the aggregates can act as defects, which limits their properties [2]. Several strategies have been considered to prepare polymer/montmorillonite nanocomposites (Figure 7) [76]:

1. In-situ polymerization method, intercalation of a suitable monomer followed by polymerization. The first method used to synthesize polymer/clay nanocomposites is based on polyamide 6.
2. Solution method, intercalation of dissolved polymer from a solution. The drawback of this method is the requirement of a suitable solvent. It has been shown that intercalation only occurs for certain polymer/solvent or monomer/solvent pairs [77].

Nanocomposites based on high-density polyethylene [78], and polyimide [79] can be synthesized by this method.

- Melt intercalation method, mixing the clay (usually organoclay) with the polymer matrix above its softening point in either static or flow conditions. The polymer chains spread from the molten mass into the silicate galleries to form either intercalated or delaminated hybrids according to the degree of penetration [56]. This process was first reported by Vaia et al. [80] in 1993. This method is relatively easy and allows for the use of current processing equipment for nanocomposite technology. Traditional processing techniques could be used for melt intercalation, like a two-roll mill, twin-screw extruder (PA, PP, PE, and PVC), injection molding, blow molding, and thermal spraying [2].

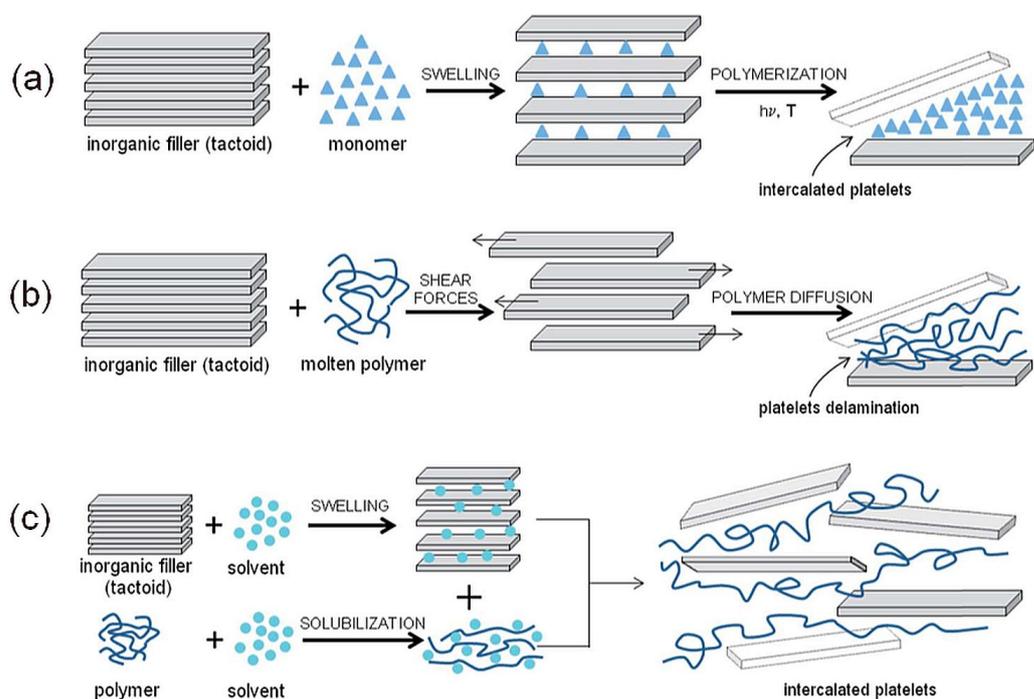


Figure 7. Illustration of (a) in situ polymerization, (b) melt intercalation, and (c) solution intercalation [76].

Highly polar polymers such as Nylon [81–83] or polyimides [84,85] are more easily intercalated than non-polar polymers such as polypropylene because polar polymers have a higher affinity for the polar clay galleries. In situ polymerization monomer intercalates directly into the organically modified clay galleries and the monomer either can adsorb onto the layer surface or can be anchored by free radical techniques. Melt intercalation involves mixing the clay and a polymer melt with or without the shear. The success of melt intercalation is surprising, given that the gallery spacing is only about 2 nm and the radius of gyration of the polymer is significantly larger than this. Even more surprising is that the speed of melt intercalation is faster than that of the self-diffusion of polymers and scales with the inverse of the molecular weight. The results of molecular dynamics and experimental studies indicate that the stronger the clay/polymer interaction, the lower the intercalation rate. In addition, layer flexibility seems to control the mechanism of intercalation [2].

2.3.2. Polymer Matrix

Many different polymers have already been used to produce polymer/clay nanocomposites. Both thermoplastics and thermosets can be successfully utilized for the preparation of nanocomposites. Intensive research is carried out in rubber mixtures too. The first and the most studied thermoplastics for the synthesis of polymer-clay nanocomposites has been polyamide 6 [86–91]. Polyamide 6/clay hybrids were discovered by Toyota re-

searchers in the early nineties [92–95] and nowadays they are used in automotive parts. The teams dealing with these materials were concentrated around Usuki A., Kojima Y., Okada A. [92–96], Azuma H. [97], Fukusima Y. [98], Wu T. [99], Devaux, E. [100], Tanaka G. [101], and Utracki L. A. [87,89,90].

Polyvinylchloride [8,102,103] nanocomposite processing was studied, in addition. Polyvinylchloride (PVC) is an important commercial polymer. It is one of the most versatile and oldest thermoplastics. It is a material that offers several positive aspects like low cost, recoverability, facile processing, and excellent electrical and chemical resistance. Commonly, PVC is available in two broad categories: flexible PVC (plasticized one), and rigid PVC (unplasticized one). The PVC products range from piping and siding, door and window profiles, blood bags, and tubing, to wire and cable insulation and more. The first results dealing with PVC/MMT nanocomposites were presented at the international conferences ANTEC'01 and ANTEC'02 in the USA [104,105] by Kalendova et al. In this study, particularly the suspension type of PVC was employed for the polymer/clay nanocomposites development. The formulation for the PVC mixtures consists of 74% of PVC, 24% of plasticizer, and 2% of stabilizer. The natural type of montmorillonite and the organophilized one were tested. The melt intercalation was employed to produce nanocomposites. One of the important questions was how long alkyl chains should be used for the exfoliation of silicate layers. Therefore, alkylamines with different alkyl chain lengths were tested as the organic compatibilizer. Especially shorter alkyl chains were tested as suitable chemical modifiers of MMT. Firstly, Na⁺ montmorillonite was ion-dipole intercalated with dodecylamine (DDA, 12 C) and octylamine (OA, 8C) molecules. The structure of octadecylamine (ODA, 18 C) ion-dipole intercalated into MMT was described by Pospisil et al. previously [106]. The material structure was determined based on X-ray diffraction and molecular simulation results. Molecular mechanics and classical molecular dynamics were carried out in the Cerius2 modeling environment. Based on calculated values of interaction energies between two guest layers in the interlayer space of montmorillonite and XRD patterns, a probable intercalant molecules arrangement in the MMT interlayer was designed. It was confirmed by Pospisil [106], Figure 8, that ODA ion-dipole intercalated into MMT allows the existence of supramolecular structure which is presented by bilayer arrangement alkyl chains perpendicular or slightly oblique to the aluminosilicate layer. The results are consistent with the studies carried out by Lagaly [43,107–109] in the field of ion-exchange intercalation. In addition, the Octylamine (OA) ion-dipole intercalated was also not able to provide the bilayer arrangement in the MMT interlayer space. From the obtained data it follows that montmorillonite intercalated with short alkylamine chains suggests a disadvantage for polymer/MMT nanocomposite production.

In the past, the most important requirement for new synthetic polymers was their resistance to climate change [110]. Recently research has undergone substantial changes, due to the ecological problems caused by non-degradable polymer litter. These facts resulted in the research in the field of biopolymers. Currently, biodegradable polymers like polylactic acid (PLA) and Polyhydroxybutyrate (PHB) are studied successfully [111–115]. Poly(lactic acid) or polylactide (PLA) has a leading position in the market of biobased polymers and is one of the most promising sustainable alternatives to petroleum-based polymers [116]. PLA is not only bio-based but compostable and biodegradable through hydrolysis by microorganisms [117]. Recently, new techniques to produce high molecular weight PLA with relatively good properties, have led to the expansion of the product portfolio from the biomedical area to short service life applications (agriculture and packaging).

Nonetheless, predisposition to degradation, poor thermal resistance, and unsuitable mechanical and barrier properties limit industrial application for long-term performance products in automotive and electronic industries. For items such as these, resistance to degradation is required, unlike for disposable applications [118]. Further, high price is also an obstacle to greater expansion of PLA on the market. To address one of these limitations, nanoscale structured layered clay particles can be incorporated into PLA conferring strength, increasing its gas barrier properties [113,119,120], and ultimately

allowing the nanocomposite to compete with conventional plastics in a wide range of applications [121–123]. A very important role of PLA is in short-term food packaging (bags). For long-term food storage, PLA offers insufficient barrier properties. Commercial products of multilayer packaging films consist of several layers created from different polymers. From the viewpoint of circular economy, separation and recycling of such multilayered material is impossible. An alternative to the traditional approach could be multilayered films from one material type, where the barrier layer could be based on the polymer/clay nanocomposites. Such films should offer uncomplicated recyclability. Kalendova et al. [113] describe the role of different modified montmorillonites in the barrier properties of PLA/clay nanocomposites. The air, CO₂, and water vapor permeability, along with morphology were investigated. The mixtures with Cloisite 10A and 30B give similar results with enhancement of more than 70% WVT after 24 h. The WVT data evaluated after 60 days show a drop in WVT, but still, the improvement is around 30% compared to pure PLA film. Further, the CO₂ and air permeability show an improvement of more than 50% compared to pure PLA film, Table 1. The best barrier properties were exhibited by the sample with Cloisite 10A and Cloisite 30B. The barrier properties improvement is connected probably with the nanofiller arrangement in the polymer matrix, Figure 8, and filler aspect ratio. A large aspect ratio of nanoplatelets creates a more dramatic reduction in permeability, which was explained by Choudalakis G. and Lu C. [124,125].

Table 1. CO₂, air permeability for PLA/Cloisite compositions [113].

Composition	Permeability Q _(CO₂) [m ² Pa ⁻¹ s ⁻¹] × 10 ⁻¹⁶	% Q _(CO₂) to Pure PLA [%]	Permeability Q _(Air) [m ² Pa ⁻¹ s ⁻¹] × 10 ⁻¹⁶	% Q _(Air) to Pure PLA [%]
PLA	2.28	0	3.94	0
PLA/10A	0.71	−69	1.57	−60
PLA/20A	0.84	−63	1.95	−51
PLA/30B	0.80	−65	1.35	−66
PLA/Na+	5.56	144	5.80	47

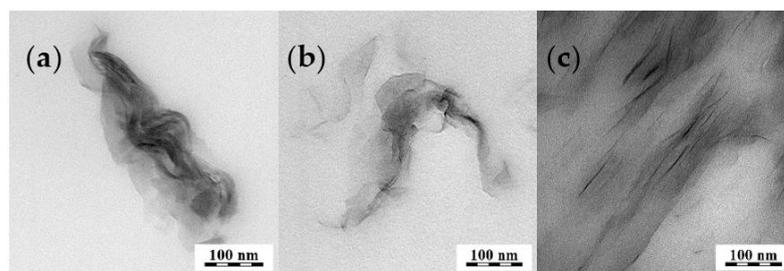


Figure 8. TEM: (a) PLA/Na+ 5%, (b) PLA/10A 5%, and (c) PLA/30B 5% [113].

In the field of thermosets, epoxy resins, and unsaturated polyesters are studied intensively. The reason for this is that the reactants of epoxy systems have a suitable polarity to diffuse between the clay layers and form a delaminated nanocomposite upon polymerization. One of the most important phenomena is the self-polymerization of epoxy resin in organophilic smectite clays due to the presence of alkylammonium ions [126,127]. This phenomenon was studied by Pinnavaia et al. at Michigan State University [128,129].

Intensive research has been conducted on polyurethanes [130–133]. The non-polar polymers, polypropylene, and polyethylene nanocomposites have also been studied in recent years, but these non-polar materials are more difficult to prepare [134–138]. It can be concluded that polar systems are studied more intensively compared to non-polar polymers.

Finally, it could be concluded, that generally polar thermoplastics such as polyamide, polymethylmethacrylate [139,140], polystyrene [141,142], polyimide [143], polyethylene oxide [144], polyethylene terephthalate [145], polyvinylchloride [8,102–105], and polylactic acid [123–125] provide good prospects for the preparation of nanocomposites. The non-

polar polymers, polypropylene, and polyethylene nanocomposites have also been studied in recent years, but these non-polar materials are more difficult to prepare [134–137]. It is still difficult to reach a well-exfoliated structure of the nanofiller in a polyolefine polymer matrix, despite the achievements in their research. To reduce the problems with the compatibility of PE, PP, and MMT, a new strategy is investigated. The non-polar polymer could be incorporated polar part of the polymer chain to gain better intercalation and exfoliation of layered clays in polymer [146,147]. Finally, it could be concluded that polar systems are studied more intensively compared to non-polar polymers.

2.3.3. Polymer/Clay Nanocomposite Advantages

Nanocomposites offer performance similar to conventional polymeric composites (30%–50% of reinforcing material) only with very low filler loading (1%–5%) [148]. Generally, the nanocomposite advantages are connected with reduced weight for the same performance (lower density of composites), transparency, greater tensile and flexural strength for the same dimension of plastic part, improved gas barrier properties for the same film thickness, minimal loss of ductility (nanoparticles do not create large stress concentrations), increased dimensional stability (reduced shrinkage), flame retardant properties, and high chemical resistance [55,57,148–152]. Observed enhancement in different types of properties (thermal endurance, electrical conductivity, barrier properties, and chemical resistance) makes these materials prime candidates for packaging, membranes, automotive applications, and coating. The next interesting application could occur in the pharmaceutical industry. In the case of polymer-clay nanocomposites, particularly thermoplastic clay nanocomposites, the properties are determined by their morphologies, that is, clay layer aspect ratio, surface energy, interfacial adhesion, and dispersion in the polymer.

Some studies have focused on the degradation of polymer/clay nanocomposites were proceeded. They are connected not only with the influence of different additives on the behavior of material after the ending of a lifetime, but they deal also with the possibilities of extending the material lifetime. The influence of hydrolysis-inhibiting additives on the degradation and biodegradation of PLA nanocomposites was duplicated by Stloukal et al. [153]. As the inhibition additives carbodiimide-based ones were used, which can avoid hydrolytic degradation during processing. Aromatic carbodiimide effectively influenced the PLA stability during thermal degradation and a positive effect was also noted for nanocomposites. Surprising findings from the obtained data indicated that the nanocomposites containing carbodiimide appeared to be stabilized during melt processing. Next, the tested additive significantly retarded biodegradation.

On the other hand, a few disadvantages associated with nanoparticle incorporation into the polymer were observed. The nanocomposites usually exhibit lower toughness, impact resistance, and abrasion resistance, compared to the pristine polymer. Newly published studies register improvement of abrasion resistance in the presence of a silane coupling agent [154] or a combination of organically modified MMT with carbon black [155,156]. The next disadvantage is the higher viscosity of the melt and usually non-uniform distribution [157].

3. Ceramic Materials

3.1. Ceramics Based on Clays

Generally, ceramic materials can be classified into two large groups according to their properties and applications: traditional (based on clay and silica) and technical or advanced (based on carbides, nitrides, borides, pure oxides, and many others) ceramics Figure 9 [8,31,158,159]. Ceramics based on clays include the materials for which the presence of a predominant amount of clay raw materials is decisive, especially for their shaping and sintering. Silicate ceramic is one of the oldest used ceramic materials. The main components are natural raw materials (e.g., clays, talc, kaolinite) in their crude or modified state [160]. Silicate ceramic can be modified by synthetic oxides which ensure the improvement of final properties. Silicate ceramics are possible to divide into several groups according to their chemical composition. Ceramics based on aluminosilicates (representa-

tion of individual oxides in system $\text{Al}_2\text{O}_3\text{-SiO}_2$, ceramics based on magnesium silicates ($\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$), and ceramics with low thermal expansion ($\text{Li}_2\text{O-Al}_2\text{O}_3\text{-SiO}_2$) [161].

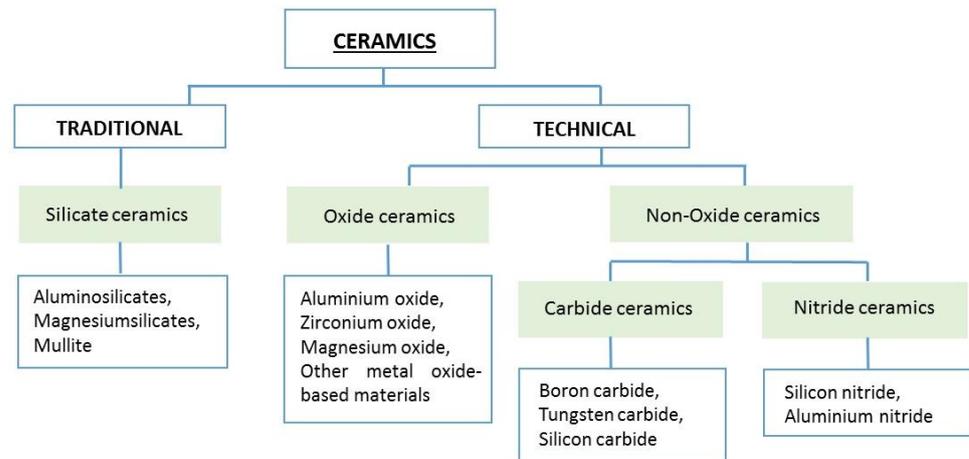


Figure 9. Types and examples of ceramics according to their chemical composition.

Silicate Ceramics

The ternary system of $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$, shown in Figure 10, includes steatite (MgSiO_3), forsterite (Mg_2SiO_4), and cordierite ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) ceramics.

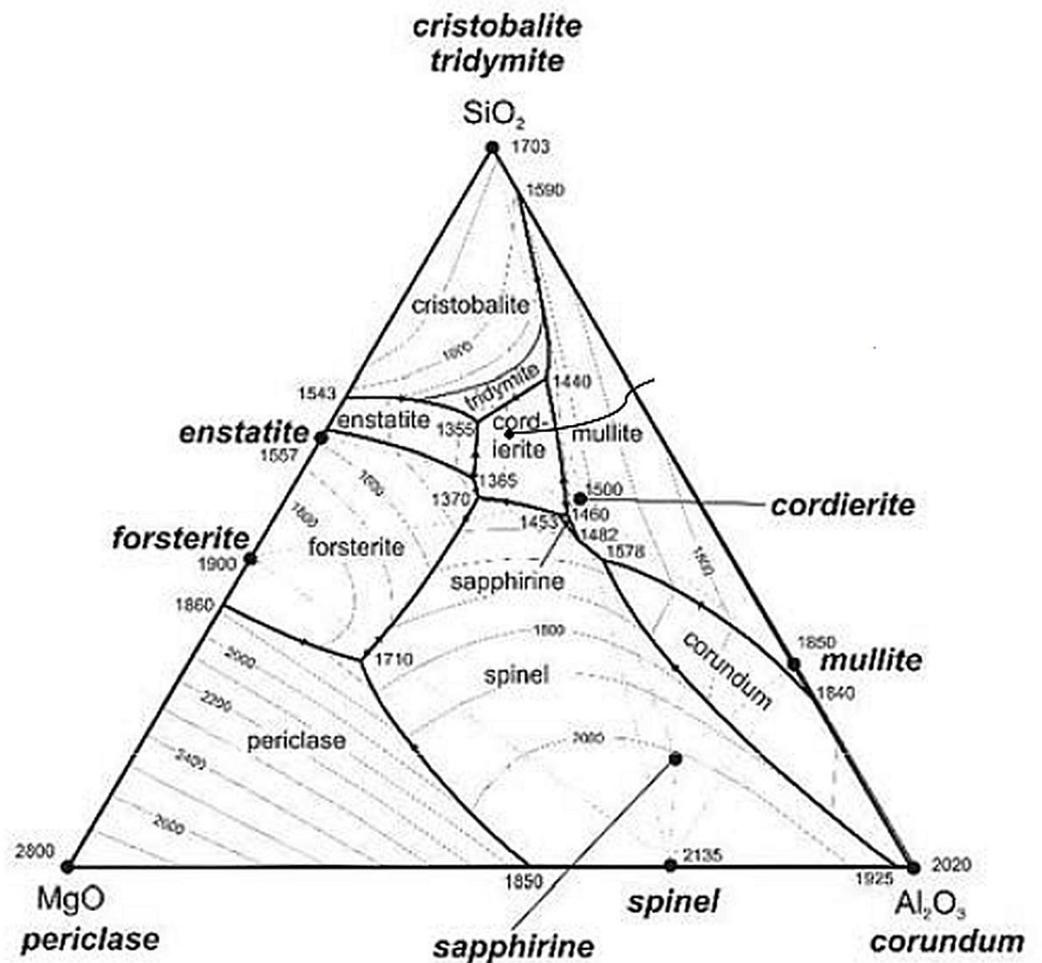


Figure 10. The phase diagram of the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ system [162].

Table 2 shows the overview of clay minerals used in pre-ceramic mixtures and types of silicate ceramics. These types of ceramics can be synthesized from clay minerals kaolinite and talc. The main constituents of talc are SiO_2 and MgO and therefore talc represents a very convenient material for the synthesis of steatite and forsterite ceramics. Excellent electrical properties, high mechanical resistance, low dielectric loss, and high-temperature resistance allow for the use of steatite ceramic in electronics and electrical engineering. Steatite is also an attractive material used in medicine for its good biocompatibility, high hardness, and resistance [163–165]. Steatite ceramic is possible to synthesize from natural raw material containing magnesium silicate (talc) together with auxiliary fluxes (feldspar or barium carbonate) and the addition of clays [163,166,167]. Sintering at around $1400\text{ }^\circ\text{C}$, the crystalline phase of enstatite (approximately 70%) originating from mineral talc and a vitreous phase (approximately 30%) from flux are transformed into steatite ceramic [164,168]. Clay in the pre-ceramic mixture facilitates the molding and processing of the ceramic mass [163]. During the calcination of talc, the formation of a vitreous phase occurs as a side effect that changes the dielectric properties of the final ceramic [164]. The type of used flux affects the dielectric properties of ceramic [167]. Enstatite is a low-temperature modification of MgSiO_3 transformed into protoenstatite (PE) during heating and clinoenstatite during cooling of PE [169]. Purified, milled, or calcined talc, after heat treatment, changes to proto-enstatite (high-temperature form) [170]. Terzić et al. (2014) [164] tried to activate talc using mechanical grinding. The authors found that grinding had a positive influence on the decreased sintering temperature of talc for steatite ceramics. Goerriot et al. (1998) [171] dealt with enstatite ceramics prepared from talc for biomedical applications (mainly dental or bone restorations). The pre-ceramic mixtures composed of talc and kaolin waste were studied by Araujo et al. (2022) [163] to prepare steatite ceramics with improved dielectric properties. Dolomite together with talc and clay stabilized with polyacrylic acid led to the synthesis of cordierite ceramic [172].

Forsterite is the main crystalline phase of forsterite ceramics generated during the firing of materials with a high content of talc [173]. Forsterite ceramic has found applications in many fields of electronics, communications, ceramic-metal seals, and refractories because of its low electrical conductivity, high melting temperature, good chemical stability, excellent insulation properties, and bioceramics [174–176]. The traditionally used synthesis method for forsterite ceramic consists of firing powders of talc together with magnesium oxide [174,175,177–179] and/or magnesium carbonate [178–182]. It is important to keep the MgO/SiO_2 at the molar ratio of 2:1 corresponding to the theoretical value of pure forsterite. During the synthesis of forsterite is very difficult to eliminate the undesirable secondary phases such as enstatite and periclase. For the formation of pure single-phase forsterite ceramic, several hours of milling followed by their sintering at high temperatures ($1200\text{--}1600\text{ }^\circ\text{C}$) is used [174,175,178,180,181]. Mustafa et al. (2002) [182] improved the sintering and mechanical properties of forsterite through the addition of Al_2O_3 . After the addition of different amounts of Al_2O_3 to the synthesized forsterite, the spinel-forsterite mixture was created. On the other hand, the Al_2O_3 addition higher than 15% led to the formation of enstatite in final ceramics. Sadeghzade et al. (2015) obtained pure forsterite-diopside nanocomposite powder (from talc, MgCO_3 , and CaCO_3), applicable in tissue engineering applications, by mechanical alloying and subsequent sintering [183].

Another type of silicate ceramics is cordierite ceramics. The main crystalline phase of this ceramic is cordierite. Cordierite is a significant material in many industrial applications mainly due to its low coefficient of thermal expansion, low dielectric constant, excellent thermal shock resistance, high refractoriness, good mechanical properties, and chemical resistance. These properties result in its use in high-temperature applications, refractory material, packing material, as an electrical insulator, filters, and exhaust catalyst support [158,184–186]. Cordierite ceramics can be prepared from synthetic materials containing individual oxides MgO , Al_2O_3 , and SiO_2 , hydroxides of their salts but also from natural materials such as talc, kaolin, gibbsite dolomite, spinel, mullite, forsterite, various clays, and from their mixture at $1340\text{--}1450\text{ }^\circ\text{C}$ [158,185,186].

The basic pre-ceramic mixtures to produce cordierite consist of talc, kaolinite, and gibbsite [187–189] and/or talc, kaolinite, and alumina [184,190–192]. The mixture of submicron particles of kaolinite and $Mg(OH)_2$ was used by Kobayashi et al. (2000) in the preparation of dense cordierite ceramics [193]. These ceramics have the potential to be substrate materials for circuit boards and as thermal shock resistance ceramics. Another mixture of ball clays, talc, alumina, and siliceous sand enabled the creation of cordierite ceramic with controlled microstructures [194]. Almeida et al. (2018) [195] obtained the cordierite ceramics via solid-state sintering method from talc, kaolin waste, and MgO. Pavlikov et al. (2011) [190] replaced kaolin in some cordierite pre-ceramic mixtures with another clay mineral pyrophyllite. Goren et al. (2006) successfully synthesized the cordierite ceramics without any other secondary phases by sintering at higher temperatures (1350 and 1400 °C) mixture of talc, diatomite, and alumina [185]. Goren et al. (2006) chose as the starting materials besides talc fly ash, fused silica, and alumina for the preparation of cordierite ceramics [196]. Kumar et al. (2000) synthesized cordierite ceramic by sintering talc, fly ash, and calcined alumina. The authors showed that fly ash can be used as a substitute for clay in cordierite ceramic for refractory applications [197]. Microcellular cordierite ceramic with improved 14 hermos/mechanical properties was obtained from a mixture of different materials containing talc, polysiloxane, alumina (as filler), and expandable microspheres (thermoplastic poly(methyl methacrylate) shell). Three steps were chosen for the ceramics preparation, namely foaming of pre-ceramic mixtures, cross-linking, and subsequent sintering [198]. The combination of commercially available and commonly used clay minerals talc and kaolinite and untraditionally vermiculite as precursor for synthesis of cordierite ceramics utilized studies by Valášková et al. (2009), Valášková and Simha Martynková (2010), Valášková et al. (2014) [199], and Valášková et al. (2018) [200]. Acimovic et al. (2003) [201] produced cordierite ceramics based on kaolin, quartz, and sepiolite and talc, kaolin, silica, and feldspar. Sepiolite represents hydrated magnesium silicate, and it can replace talc in pre-ceramic mixtures. Microstructure, porosity, bulk density, and thermal conductivity were characterized by Li et al. (2015) [202] for cordierite foam ceramics prepared from kaolin, attapulgite, and MgO. In the study of Gökce et al. (2011), steatite (mixture of calcined talc and Al_2O_3 and $BaCO_3$) and cordierite (calcined talc and kaolinite and Al_2O_3) ceramics were synthesized separately. After the sintering of both types of ceramics, the cordierite powder was added to the steatite ceramic. Cordierite acted as reinforcement material in the steatite matrix. The authors evaluated the influence of cordierite content and sintering temperature on the properties of final cordierite/steatite ceramics.

The next possibility, how to obtain cordierite ceramic, is a modification of clay minerals in various ways e.g., with organic compounds or cations or inorganic cations. The influence of different organic modifying agents (octadecylamine, hexadecyltrimethylammonium, and hexadecylpyridinium) and two sources of vermiculite (Brazil and South Africa) was investigated on the mineral phase composition and porosity of cordierite/steatite ceramics [200]. The use of organically modified vermiculite in the pre-ceramic clay mixtures as a pore-forming additive led to the crystallization of indialite (high-temperature phase of cordierite) at the expense of other mineral phases and the creation of porous cordierite/steatite ceramics [200–203]. Vermiculite matrix with anchored CeO_2 nanoparticles was used in pre-ceramic mixtures (with talc and kaolinite) for preparation of photocatalyst CeO_2 /cordierite/steatite ceramics [204,205]. Valášková et al. (2015, 2019) and Valášková et al. (2013) [204–206] investigated the characterization of zircon-cordierite ceramics created from zirconium-vermiculite precursor in pre-ceramic clay mixtures. Zircon-cordierites exposed a lower porosity and higher hardness in comparison to basic cordierites. Another cordierite-mullite composite ceramic is a potentially suitable material for solar thermal storage. Authors prepared this type of ceramic composite from raw materials talc, kaolin, andalusite, potassium feldspar, albite, and Al_2O_3 [207].

Table 2. The overview of pre-ceramic clay mixtures and types of silicate ceramics.

Pre-ceramic Mixtures					
Clay Minerals	Other	Type of Ceramics	Sintering Temperature (°C)	Applications	Ref.
Talc	-	Enstatite	1275, 1350, 1375	Machinable prosthesis	[171]
Talc, clay	BaCO ₃	Steatite	1240–1380	-	[167]
Talc	BaCO ₃	Steatite	1380	-	[170]
Talc, montmorillonite Talc, kaolinite Talc, montmorillonite Talc, kaolinite, Acid treated talc	Na ₂ CO ₃ Na ₂ CO ₃	Steatite	1300	-	[165]
Talc, clay	BaCO ₃ , boric acid	Steatite	1000–1200	High temperature electrical applications	[166]
Talc, kaolin	-	Steatite	1200, 1250, 1300	Material in electrical insulation	[163]
Talc, clay	Dolomite, polyacrylic acid	Steatite	1275, 1300	-	[172]
Talc	MgO	Forsterite	1200, 1300, 1400, 1500	Biomedical applications	[177]
Talc	MgCO ₃	Forsterite	1000	-	[180]
Talc	MgCO ₃ , NH ₄ Cl	Forsterite	1000	Bioceramics (bone tissue engineering applications)	[181]
Talc	MgCO ₃ , NH ₄ Cl	Forsterite	1000, 1200	-	[173]
Talc	MgO	Forsterite	1200	-	[174]
Talc	MgO	Forsterite	1200–1500	-	[175]
Talc	MgO	Forsterite	1000, 1200	-	[176]
Talc	NH ₄ F	Forsterite	-	-	
Talc	NH ₄ Cl	Forsterite	-	-	
Talc	MgCO ₃	Forsterite	1000, 1200	-	[178]
Talc	MgO	Forsterite	-	-	[182]
Talc	calcined MgCO ₃	Forsterite	1400	-	
Talc	MgCO ₃	Forsterite	1200, 1300	Refractory ceramics	[179]
Talc	MgO	Forsterite	-	-	[183]
Talc	MgCO ₃ , CaCO ₃	Forsterite/diopside	1200	Tissue engineering	
Talc, kaolin, Talc, kaolin Talc, kaolin, pyrophyllite Talc, pyrophyllite Talc, kaolin, Talc, pyrophyllite	Mg Al ₂ O ₃ Al ₂ O ₃ Al ₂ O ₃ Mg, Al ₂ O ₃ Mg, Al ₂ O ₃	Cordierite	1000–1380	-	[190]
Talc	Al ₂ O ₃ , BaCO ₃	Steatite	1320	-	[184]
Talc, kaolinite	Al ₂ O ₃	Cordierite	1250	-	
Talc, kaolin waste	MgO	Cordierite	950, 1050, 1150, 1250, 1350	Refractory and insulating materials	[195]
Kaolinite	Mg(OH) ₂	Cordierite	1350	Substrate material for circuit boards, thermal shock resistance ceramics	[193]
Talc	Diatomite, Al ₂ O ₃	Cordierite	1300, 1350, 1400	-	[185]
Talc	Fly ash, fused silica, Al ₂ O ₃	Cordierite	1200, 1300, 1350, 1375	-	[196]
Kaolin, attapulgite	MgO	Cordierite	1200	Thermal insulator	[202]
Kaolinite, talc, vermiculite		Cordierite/steatite	1300	-	[199]

Table 2. Cont.

Pre-ceramic Mixtures					
Clay Minerals	Other	Type of Ceramics	Sintering Temperature (°C)	Applications	Ref.
Kaolinite, talc, vermiculite, organo-vermiculite		Cordierite/steatite	1300	-	[200]
Kaolinite, talc, vermiculites	Al ₂ O ₃ , Al(OH) ₃	Cordierite	1300	-	[187]
Kaolinite, talc, vermiculite	Al ₂ O ₃	Cordierite	1300	-	[191]
Kaolinite, talc, vermiculite, organo-vermiculite		Cordierite/steatite	1300	-	[203]
Talc, kaolinite, vermiculite	MgO, Al(OH) ₃	Cordierite	1300	-	[192]
Talc, kaolinite, ball clay	Al ₂ O ₃ , silica sand	Cordierite	1300	Industrial manufacture of porous ceramic materials	[194]
Talc, kaolinite, CeO ₂ /vermiculite	-	Cordierite/CeO ₂	1300	Photocatalysts	[204,205]
Talc	Polysiloxane, Al ₂ O ₃ , expandable microspheres	Cordierite	1300	-	[198]
Talc, kaolinite	Al(OH) ₃	Cordierite	1260	-	[189]
Kaolin, sepiolite Kaolin, talc	Quartz SiO ₂ , feldspar	Cordierite Cordierite	1250, 1300, 1350	Application in foundry	[201]
Talc	Fly ash, Al ₂ O ₃	Cordierite	1350	Refractory application	[197]
Talc, kaolin	Feldspar, albite, andalusite, Al ₂ O ₃	Cordierite/mullite	1340–1420	Thermal storage materials	[207]

4. Conclusions

The presented study focuses on one part of nanotechnology targeted to polymer science, concretely to the field of polymer/clay nanocomposites. These hybrid organic-inorganic materials show an interesting enhancement of properties compared to traditional micro-composites with a low level of nanofiller loading obviously in the range of 1%–5%. The most favorite source of nanoelements is montmorillonite (MMT), a layered aluminosilicate whose morphology is created by stacks of individual platelets with 1 nm thickness. Although clay nanocomposites were discovered in the 1990s, interest from the scientific community remains high. Some of the applications have already been commercialized. Approximately 80% of the polymer/clay nanocomposites are destined for the automotive (General Motors, Chevrolet, Nissan), aeronautical, and packaging industries [208]. The key drivers for the use of polymer/clay nanocomposites in the automotive industry are reduction in vehicle's weight, improved engine efficiency (fuel saving), reduction in CO₂ emissions, and superior performance (greater safety, increased comfort, and better drivability) [208]. The next perspective is presented by packaging applications. The tested materials based on PLA showed common improvement in the range of 30% [113]. Other commercial applications include cables, furniture, and domestic appliances. Further, applications like in selective catalyzers, conductive polymers, filtration of toxic materials, drug delivery systems, and energy storage, are expected. Although nanocomposites present a series of advanced properties, their production is considered low in comparison with other materials, due to the production costs.

The second part focused on ceramic materials prepared from clay minerals in pre-ceramic mixtures. The kind, quantity, and combination of clay minerals in the pre-ceramic mixtures, as well as different preparation methods or their chemical modifications significantly influence or alter the resulting properties of ceramics after their sintering. Clay

minerals such as talc and kaolinite represent the main natural raw materials for traditional applications mainly for creating silicate ceramics. Silicate ceramics like cordierite, steatite, and forsterite are used in many industrial applications (especially in electronics and electrical engineering) and offer cost savings mainly due to using natural raw materials in comparison to other types of ceramics. Further, silicate ceramics have a great versatility of applications including refractory, insulating, packing materials, filters, catalysts, and thick films and as glass-ceramics, forsterite, and bioceramics. Moreover, ceramic composite materials have been increasingly used in various fields including catalysts, solar thermal storage, and medical applications. Steatite, forsterite, and cordierite-based ceramics may offer new properties to industry applications.

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