

## Review Article

# Nanocomposite Coatings: Preparation, Characterization, Properties, and Applications

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Incorporation of nanofillers into the organic coatings might enhance their barrier performance, by decreasing the porosity and zigzagging the diffusion path for deleterious species. Thus, the coatings containing nanofillers are expected to have significant barrier properties for corrosion protection and reduce the trend for the coating to blister or delaminate. On the other hand, high hardness could be obtained for metallic coatings by producing the hard nanocrystalline phases within a metallic matrix. This article presents a review on recent development of nanocomposite coatings, providing an overview of nanocomposite coatings in various aspects dealing with the classification, preparative method, the nanocomposite coating properties, and characterization methods. It covers potential applications in areas such as the anticorrosion, antiwear, superhydrophobic area, self-cleaning, antifouling/antibacterial area, and electronics. Finally, conclusion and future trends will be also reported.

## 1. Introduction

**1.1. Definition.** A nanocomposite coating is a material composed of at least two immiscible phases, separated from one another by interface region. The material must contain the nanometer scale in at least one dimension in which the major component is called matrix in which fillers are dispersed [1].

**1.2. Classification.** The classification of nanocomposite coatings is based on various methods dealing with (i) type of nanostructured fillers or (ii) type of matrix where filler nanostructures are dispersed.

**1.2.1. Type of Nanostructured Fillers.** There are 3 main groups of nanocomposite coating as follows [1]:

- (i) 0D nanocomposite coatings: the filler are nanoparticles (3 dimensions in nanometer scale).
- (ii) 1D nanocomposite coatings: the filler are nanotubes or whiskers (2 dimensions in nanometer scale).

- (iii) 2D nanocomposite coatings: the filler are nanolayers (1 dimension in nanometer scale).

**1.2.2. Type of Matrix.** Two types of matrix which are organic and inorganic matrix can be found. Thus, there are 4 main groups of nanocomposite coating as follows (matrix/nanofiller):

- (i) Organic/inorganic nanocomposite coatings (O/I nanocomposite coatings)
- (ii) Organic/organic nanocomposite coatings (O/O nanocomposite coatings)
- (iii) Inorganic/organic nanocomposite coatings (I/O nanocomposite coatings)
- (iv) Inorganic/inorganic nanocomposite coatings (I/I nanocomposite coatings).

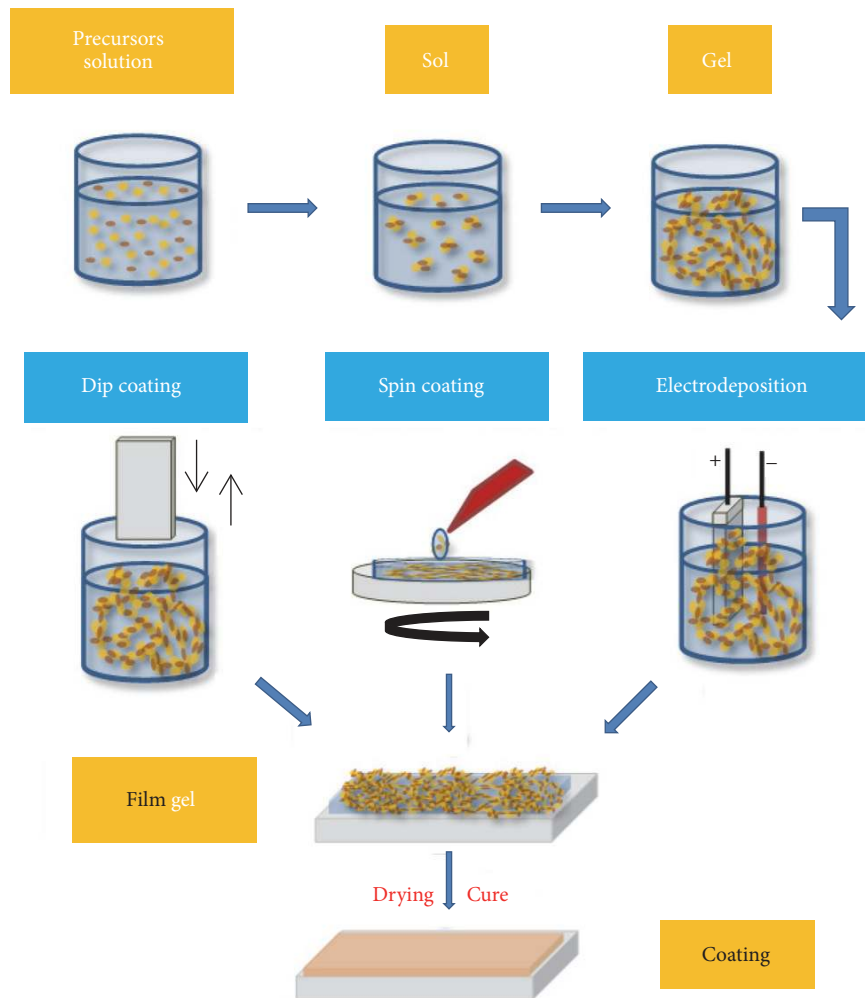


FIGURE 1: Preparation of coatings by sol-gel method [85].

### 1.3. Materials

**1.3.1. Materials for Matrix.** For organic matrix, called polymer-based nanocomposite, the most used polymers for preparation of nanocomposite coating can be listed as follows: epoxy [2–7], polyurethane [8, 9], Chitosan [10, 11], polyethylene glycol (PEG) [12–15], polyvinylidene fluoride (PVDF) [16], PANi [17–19], PPy [20–23], polystyrene [24], polyamic acid and polyimide [25], rubber-modified polybenzoxazine (PBZ) [26], polymers containing reactive trimethoxysilyl (TMOS) [27], pullulan [28], fluoroacrylic polymer [29, 30], ethylene tetrafluoroethylene (ETFE) [31], polyacrylate [3], poly(N-vinyl carbazole) [32], polycarbonate [33], fluorinated polysiloxane [34], polyester [35], polyacrylic [36], polyvinyl alcohol (PVA) [37], polydimethylsiloxane [38], polyamide [39], and UV-curable polymers [40].

For inorganic matrix, such as metal matrix or alloy matrix, these nanocomposite coatings could be prepared by various methods, including chemical vapor deposition (CVD) (see Figure 3), powder metallurgy, physical vapor deposition (PVD) [41–45], thermal plasma spray [46–48], sol-gel [49–52] (see Figure 1), epitaxial growth [53], cold spray

[54–61] (see Figure 2), and electrodeposition [62–67]. Metal matrix composite coatings that dispersed a second phase [68, 69] have attracted extensive attention owing to unique properties such as oxidation and corrosion resistance [69, 70], wear resistance [71, 72], and magnetic properties [72].

**1.3.2. Materials for Nanofillers.** For inorganic nanofillers, the types of nanoparticles are carbides [58], nitrites [59–61, 73–77], borides [55], oxides [4, 5, 62, 65], metallic particles [17, 78], clay [4, 5, 35], CNT [54], and nanodiamond [56, 79]. For organic nanoparticles (organic nanofiller), the most used nanoparticles were PTFE [80–82], PEO [83], PANi [31], or nanocellulose and cellulose nanocrystal [18, 40].

## 2. Preparative Methods

**2.1. Sol-Gel Method.** The sol-gel method is suitable to obtain high quality films up to micron thickness and is a complementary of the physical deposition techniques. However, there are limits of sol-gel application to coating on the metallic substrates. This method exhibits several drawbacks involving crackability and thickness limits. Sometimes also

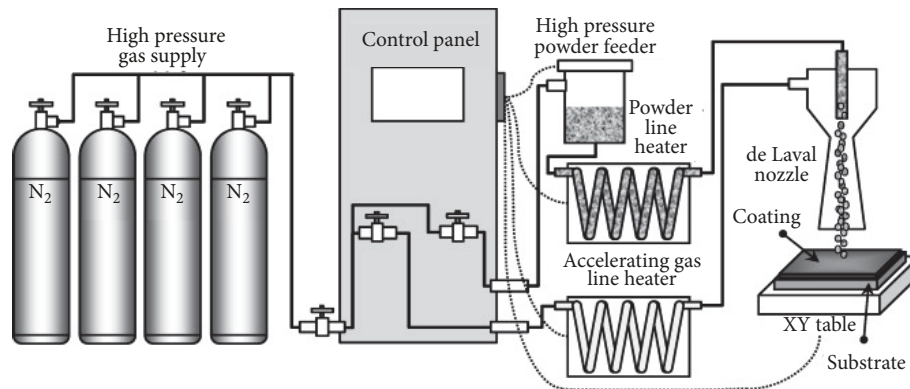


FIGURE 2: Principle of cold spray technique for preparation of nanocomposite [86].

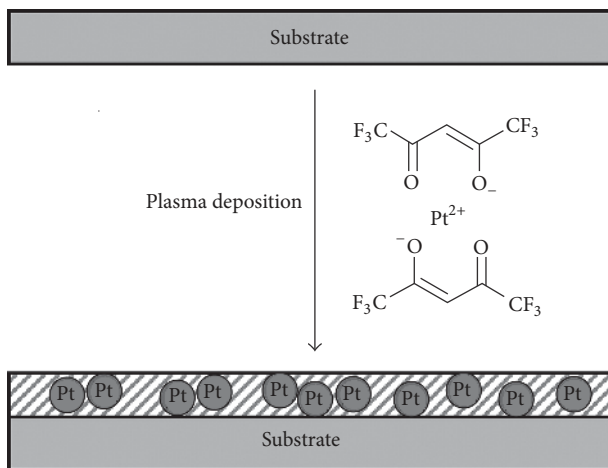


FIGURE 3: Plasma enhanced chemical vapor deposition [91].

the thermal treatment may be critical. Tensile stresses develop during drying and can lead to crack formation if the film is thicker than a critical value.

In case of inorganic matrix, the second phase can be added to sol-gel for inorganic nanofillers, such as the I/I coatings [49–52]. In the case of organic matrix, a well-known approach to generate inorganic nanophases within an organic matrix is to utilize sol-gel chemistry [18]. Inorganic sol-gel precursors such as silicon, titanium, aluminum, and zirconium metal alkoxides are employed in the formulation of nanocomposite coatings. A wide range of oligomers as well as low molecular weight organic compounds are often reported to be used as organic phase precursors. Under controlled conditions, silanes and organic molecules can form coatings containing silica nanoparticles or nanophases. In the presence of a coupling agent, the organic and inorganic phases can be covalently linked. Silica nanofiber formation in a system containing TEOS, methacryloxypropyltrimethoxysilane (MAPTMS), a urethane acrylate resin, and an acrylated phenylphosphine oxide oligomer (APPO) has been reported [40].

The nanofibers were shown to improve the mechanical properties of the organic matrix. By this sol-gel process, Facio and Mosquera [38] also successfully fabricated the nanocomposite coatings containing (i) a mixture of monomeric and oligomeric ethoxysilanes, (ii) a hydroxyl-terminated polydimethylsiloxane, (iii) colloidal silica particles, and (iv) a surfactant (n-octylamine). In addition, the sol-gel method could be used in combination with the electrodeposition for incorporation of inorganic nanofillers into the organic matrix [78] or into inorganic matrix [84].

**2.2. Cold Spray Method.** Unlike the traditional thermal spray (gas-flame, plasma, and detonation spraying), cold spraying allows fabrication of coatings at the lower temperatures than melting points of the sprayed materials. Cold spray technique was conducted at low temperatures, so this method avoids the deterioration phenomenon of the materials such as oxidation and decomposition as well as phase transition during the process. The obtained coatings have low porosity (<1%) and low oxygen concentration. In addition, the coatings have high strength (>280 MPa) and strong adhesion (>70 MPa).

This method is used to produce the nanocomposite coating, which has metallic matrix, such as Cu [54, 55], Al [56, 57], Co [58], or alloy matrix [59–61], and its nanofillers are nitride, carbide, boride, diamond, CNT, or others [54–61]. To fabricate the nanocomposite powders for this cold spray method, the mechanical alloying (MA) should be used with metallic matrix powders and other nanoparticles.

**2.3. CVD Method.** This method usually used for the fabricating of the I/I nanocomposite coatings, which include the inorganic matrix and inorganic nanofillers [87–89]. In order to improve the quality of coating, the aerosol-assisted CVD method can be used [90].

On the other hand, the O/I nanocomposite coatings were also successfully fabricated by using CVD method with platinum (II) hexafluoroacetylacetonate as precursors [91]. This process allows producing a layer of nanocomposite on the organic substrate by a single step which displays both ionic and electric conductivities. The distinct advantages of this method are related to its high quality films and

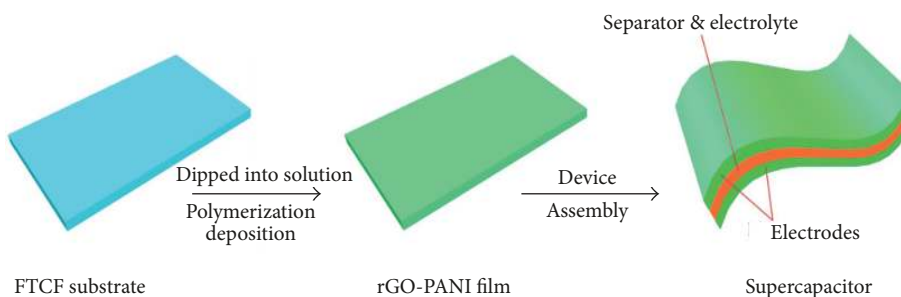


FIGURE 4: Preparation of nanocomposite coating via in situ polymerization method [105].

its facility for any kind of complex substrates with a good reproductivity.

**2.4. PVD Method.** This method is used generally for producing of the I/I nanocomposite coating [41–45, 92–97], which includes the inorganic matrices and inorganic nanoparticles. For these coatings, PVD method includes the following: laser ablation [98], thermal evaporation [99], ion beam deposition [100], ion implantation [101, 102], laser-assisted deposition [103], and atom beam cosputtering technique. In case of O/I nanocomposite coatings, Schild [104] has successfully fabricated the nanocomposite coating with organic matrix, by using the aerosol-assisted plasma deposition.

**2.5. Thermal Spray Method.** This method is often used to make nanocomposite coatings with a matrix of metal or alloy. The spray material is a nanosized alloy powder (formed by ball milling [48]) and dispersed in a suspension solution using suspensions (Thermally Sprayed [47] and Suspension Plasma Spray Process [46]) to conduct plasma thermal spraying.

**2.6. In Situ Polymerization Method.** This method was used to fabricate the nanocomposite coatings with organic matrices, which were conducting polymer [17, 18] or other monomers with initiators. The nanofillers were metals or metal oxides (see Figure 7). The polymerization takes place by using electric power (electrodeposition) [19], oxidizing agents [17, 18, 105], or photon (photopolymerized) [40, 106, 107]. The similar methods are emulsion polymerization [108, 109] or latex emulsions [110] for organic matrices (see Figure 4).

**2.7. Electroless Deposition.** This method is usually applied for producing the nanocomposite coatings with Niken matrix and nanofillers are carbide, nitrite, boride, or PTFE [49, 81, 82, 111–117]. In order to improve the hardness, anti-corrosion, and antiwear of coating, the thermal posttreatment at 500–700°C should be applied.

**2.8. Electrodeposition Method.** This method could be used for the fabrication of nanocomposite coatings, which contain organic nanofillers (such as PEO, PTFE [80, 83, 118]) or inorganic matrices [119–123] or organic matrices [19, 22, 32, 78, 124].

In the case of organic matrices, the electrochemical codeposition of nanocomposites has been reported by many

researchers [123–141]. Zhitomirsky had reviewed the various nanostructured organic-inorganic coatings using electrophoretic deposition [140]. The author summarized various organic matrices, such as polyelectrolytes, poly(ethylene imine), which were electrochemically codeposited with metal ions as well as with ceramic nanoparticles. The electrochemical codeposition of carbon nanotubes/conducting polymers has also been reported [126–129, 131, 134]. Other studies involved the electrochemical codeposition of oxide and metal nanoparticles, such as Ni (as matrix) and  $\text{Al}_2\text{O}_3$  (as nanofiller) [130, 136]. Chitosan was also reported as a dominant organic matrix, used for the electrodeposition of nanocomposite coatings [125, 132, 133].

In the case of inorganic matrices, the electrodeposition of nanocomposite coatings can be performed by using the direct current (DC), pulsed current (PC), and pulsed reverse current (PRC) methods [144]. Among these 3 methods, the PC method provides more control on structure and properties of the coatings; thus the obtained coatings had better tribologic and corrosion properties than ones made by DC method [145]. On the other hand, compared with other methods, the electrodeposition technique was much easier in terms of manufacturing processes and lower cost [62]. The main advantages of this technique are related to the uniform distribution of particles, the ability of continuous processing and the reduction of waste materials [63]. Compared to conventional coatings, nanocomposite coatings exhibit higher hardness and heat resistance due to the presence of nanoparticles in the grain boundaries, which can prevent the dislocations movement and recrystallization at high temperature [64]. As a strong and tough metal, Nickel has been widely used as an electrodeposited metal matrix, in combining with nanofillers such as boron nitride [66, 67] and  $\text{Al}_2\text{O}_3$  nanoparticles [62, 65]. Some authors reported that the changing duty cycle and frequency during pulsed electrodeposition can also produce nanocomposite coatings [146].

**2.9. Solution Dispersion.** This method is mainly applied for the preparation of polymer nanocomposite coatings [4, 5, 7, 28], reinforced with nanofillers such as metal oxides, nanoclay, and carbon nanotube (CNT). In this method, beside the use of traditional magnetic/mechanical stirring methods, the ultrasound-assisted (sonication) stirring [4, 5, 28] was used for better dispersion of nanofillers into polymer matrices.

**2.10. Spray Coating and Spin Coating Methods.** These methods are widely used for the preparation of polymer nanocomposite coatings. In the case of spray coating, by using the atomizer, the nanocomposite coatings had better properties [8, 29]. The atomizer could also be used for thermal spray method, for example, atomized spray plasma deposition [147]. In the case of spin coatings [16, 148], it provides uniform thin films to flat substrates. The substrate is rotated at high speed in order to spread the coating materials by centrifugal force. This method is suitable for the preparation of thin-film nanocomposite coatings.

**2.11. Dip Coating.** This technique is widely used in industry; the dip coating technique consists in soaking a substrate in a solution of nanocomposite and pulled up at a constant and controlled speed. The substrates are then covered with nanocomposite as it is removed from the solution. Due to the imposed pulled up rate, the amount of nanocomposite on the substrate surface is also controlled.

There are two pulled out rates of the substrate, which have direct effect on the thickness of the film. At low pulled up rates, there is the capillary regime where the rate of evaporation of the solvent is greater than that of the shrinkage of the plate. This means that the shorter the shrinking speed, the thicker the film. At high pulled up rates, the trend is reversed. In this so-called drainage regime, it is the combination of the adhesion of the solution to the substrate and the gravity that forces the drainage of the solution (Figure 6). This causes thickening of the films as the shrinkage rate increases.

The advantage of this technique is that the preparation of a flat surface is therefore suitable for all forms of coated substrates. The fact that the solution can be reused until evaporation or depletion of the solute also makes this technique particularly convenient, especially for industrial applications.

**2.12. Other Preparative Methods.** There are also several other methods for the elaboration of nanocomposite coatings but they are less popular such as the following: self-assembly (O/I coatings [46]); layer-by-layer assembly (O/O coatings [157]); localized laser heating, solid-state displacement reactions, ball impact deposition (for I/I coatings [158–160], resp.); and atomic layer deposition (for I/O coatings [161–163]).

### 3. Characterization and Properties

**3.1. Effect of Nanofillers on the Microstructure and Morphology of Coatings.** In case of organic matrices, the appropriated dispersion of the nanofillers is crucial to obtain final desired properties. The small size of the nanoparticles exhibits some advantages since it enables penetrating into ultrasmall holes, indentation, and capillary areas in the polymer matrix. For example, the incorporation of nanoparticles into epoxy resins showed an enhancing of the integrity and durability of coatings thanks to the filling up of microcavities and cracks in the coatings [164–167]. Nanoparticles can also prevent disaggregation of the polymer during curing, leading to a homogenous coating. In a recent study, it was recognized that nanoparticles with a higher specific surface area not

only exhibit an increase of the distribution of nanofillers in the epoxy matrix but also enhance epoxy-curing process [4, 5]. For polyester coatings, Golgoon et al. [35] reported that the embedding nanoclay created a denser coating and more uniform and less pores in comparison with the pure polyester coating. The presence of nanofiller cans also perturb the crystallization of polymer. In many case, nanofillers enhance the crystallinity degree and reduce the spherulites file of semicrystallinity polymers.

In the case of inorganic matrices, the formation of nanocomposite structures is connected with a segregation of the one phase to grain boundaries of the second phase, and this effect is responsible for stopping of the grain growth [79]. Li et al. [118] reported that the presence of nanoparticles (anatase and rutile nano-TiO<sub>2</sub>) reduced the grain size of the nickel matrix. For conventional materials, size of grain of monocrystals varies from about 100 nm to several hundred millimetres. In this case, the number of atoms in grains is much greater than that in boundary regions. For nanocrystalline materials, the grain size is about 10 nm or less [79]. By adding nanocrystal phase into the metal matrix, the volume fraction of grain boundaries might increase. Mitterer et al. [153] formed nitride nanocrystalline phases within a metal matrix, such as TiN in Ni [73], ZrN in Ni [74], ZrN in Cu [75, 76], and CrN in Cu [77]. In these coatings, one metal may be converted into nitride in the nanocrystalline phase and the other may be transported into the growing film unreacted. On the other hand, thermal posttreatment has strong effect not only on the grain size [50, 59], crystallization [154], and morphology [168] but also on the dislocation density and the interactions between nanoparticles and the matrix [46] or the bonding of microstructural phases [47].

#### 3.2. Effect of Nanofillers on the Mechanical Properties of Coatings

**3.2.1. Tensile and Impact Properties.** In case of organic matrices, the incorporation of nanoparticles into the epoxy coating enhanced significantly the impact strength of the epoxy coating [4, 5]. The epoxy coating modified with TiO<sub>2</sub> nanoparticles showed the best enhanced impact strength of all the epoxy coatings. The distinct improvement in their stiffness may be ascribed to the following two mechanisms: (i) the nanoparticles occupy pinholes and voids in the thin-film coating and reinforced interconnected matrix, causing a reduction of total free volume and an enhancement of the cross-linking density of the cured epoxy. As such, the cured nanocomposite coating has reduced chain segmental motions and improved stiffness. (ii) Nanoparticles may act to prevent epoxy disaggregation. By using the nanoindentation method for these epoxy nanocomposites [32], it was found that modification with nanoparticles did not always enhance the stiffness of the epoxy coatings. The presence of nano-SiO<sub>2</sub> in epoxy coatings increased Young's modulus up to 20 times, whereas nanoclay and nano-Fe<sub>2</sub>O<sub>3</sub> modified samples showed ~30% and ~25% decrease, relative to the unmodified epoxy coating, respectively. It was realized that, based on Young's modulus of the nanocomposite coatings, its stiffness depended heavily upon the integrity and internal

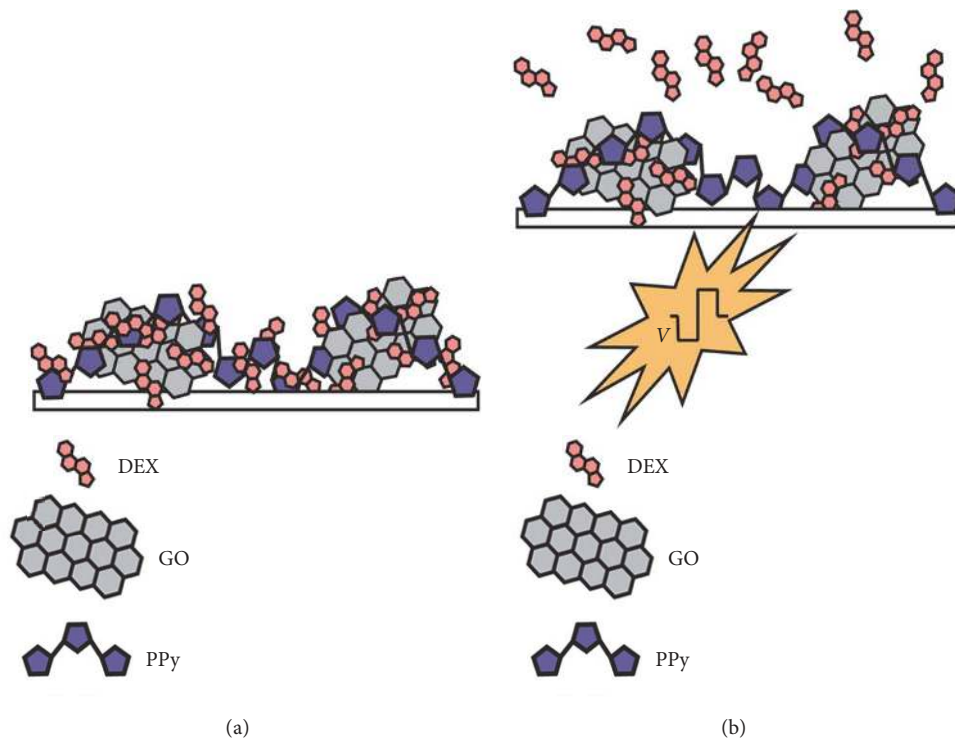


FIGURE 5: Schematic representation of the (a) GO/PPy-DEX nanocomposite and (b) DEX release from the GO/PPy nanocomposite in response to electrical stimulation [124].

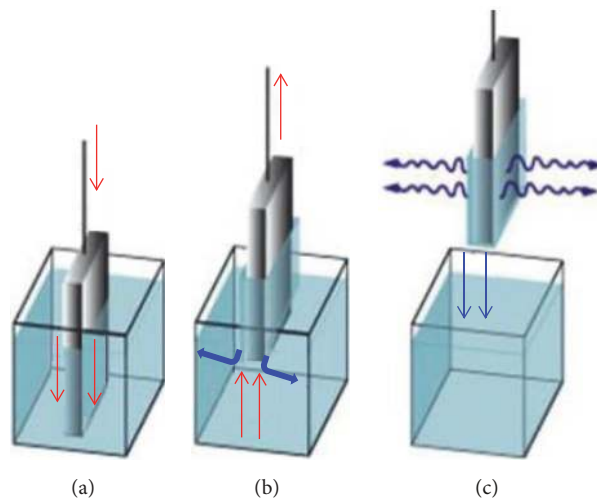


FIGURE 6: The dip coating: (a) submersion of the substrate in the nanocomposite solution, (b) withdrawing at constant speed of the substrate covered with nanocomposite, and (c) evaporation of the solvents.

properties of the coating surface, as under mechanical stress the microvoids between the nanoparticles or between the polymer matrix and the nanoparticles may become the origin of cracks. Ragosta et al. [169] reported that the addition of nano-SiO<sub>2</sub> up to 10 wt.% brought a significant enhancement in fracture toughness and an increase in the critical crack length for the onset of crack propagation. In the case of inorganic matrices, the presence of nanograins in the amorphous matrix improved the protection against impacts of high speed microparticles [2].

**3.2.2. Hardness.** For traditional organic and inorganic coatings, the measurement of microhardness can be used for evaluation of hardness. However, for the nanocomposite coatings, in order to study the effect of nanoparticles on the hardness of coatings, AFM based nanoindentation is found to be more appropriate [4, 56], with the size of AFM tip being about 50 nm. An extra result can be obtained from this nanoindentation is Young's modulus of coating [4].

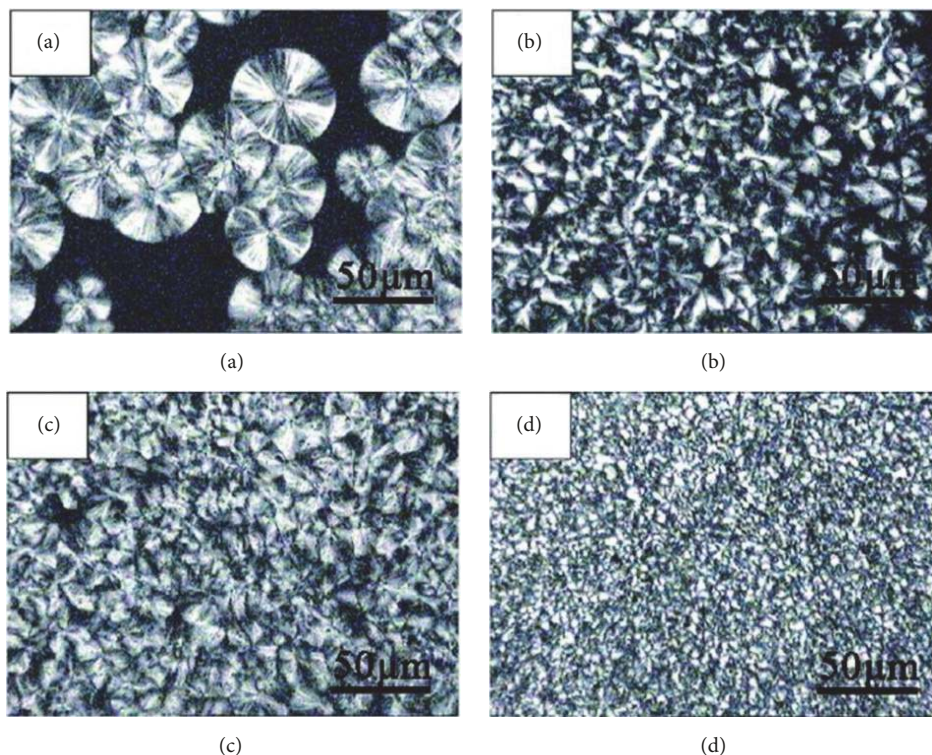


FIGURE 7: Optical micrographs of neat PLA and PLA/MOF (metal oxide framework) composites: (a) PLA, (b) PM0.5, (c) PM1, and (d) PM1.5.

(a) *Organic Matrices.* The addition of solid particles to polymers to improve their mechanical properties is a very common method. These are generally ceramic particles such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{ZnO}$ ,  $\text{TiO}_2$ , and  $\text{CaCO}_3$ . Among them nano- $\text{SiO}_2$  is the most used and studied particle. Sung et al. [9] studied the effects of nanoalumina addition on the scratch behavior of polyurethane coatings. They concluded that the addition of hard particles did not reduce scratch damage as assessed by scratch width measurement. For polyacrylic coating, Sajjadi et al. [36] reported that its scratch resistance is improved by the addition of nanoalumina particles, but it is not greatly changed by the addition of nanodiamond particles. Behler et al. [39] studied the effect of nanodiamond addition to polyacrylonitrile and polyamide (80 wt.% and 40 wt.%) on their modulus and hardness by the nanoindentation test. They observed that the addition of nanodiamond to the matrix could dramatically increase its hardness (~2 times). Maitra et al. [170] produced dilute nanocomposites (0.6 wt.%) of polyvinyl alcohol matrix reinforced with nanodiamond and observed that the addition of the particles could greatly improve hardness (up to 80%) of the polymeric matrix.

(b) *Inorganic Matrices.* Based on the value of hardness, the inorganic coatings can divide into two groups: (i) hard coatings (hardness < 40 GPa) and (ii) superhard coatings (hardness > 40 GPa). Among the various hard materials, there are only a few superhard materials, that is, cubic boron nitride (c-BN), amorphous diamond-like carbon (DLC), amorphous carbon nitride (a-CN<sub>x</sub>), and poly-crystalline diamond [79]. For metallic nanocomposite coatings, in

order to obtain superhardness, normally the plastic deformation should be prohibited; the dislocation movement and grain boundary sliding should also be prevented. Nowadays, researchers believed that the grain boundary diffusion and grain boundary sliding would be needful to improve toughness of nanocomposite coatings. Thus there are two ways for fabrication of the superhard and high toughness nanocomposites [171], as follows.

First, by the combination of two or more nanocrystalline phases, the complex boundaries could be created, then accommodating the coherent strain, thus increasing the hardness of coating. In this direction, Mitterer et al. [153] elaborated hard nanocrystalline phases within a metal matrix. The hardness of these coating systems varied from 35 to approximately 60 GPa. The higher hardness of solid compounds could be obtained by forming high strength grain boundaries, where there was a wide miscibility gap with a certain chemical affinity to each other. In this case, both mechanism modes of dislocation and grain boundary may contribute to the hardness of coatings [172–174], while the metallic matrix could guarantee their toughness.

The second way is to embed nanocrystalline phases in an amorphous phase matrix [87, 93, 168]. In this direction, the promising candidates for the amorphous matrix are the diamond-like carbon (DLC), amorphous carbon nitride, or other hard amorphous materials. Whereas the nanosized nitrides, such as TiN,  $\text{Si}_3\text{N}_4$ , AlN, and BN, could be used as strengthening phases. By embedding nanocrystalline TiC in DLC matrix, Voevodin et al. [103, 154] obtained the hardness of 32 GPa for this nanocomposite coating. Vepřek et al. [155, 156] also embedded the TiN nanocrystals (4–11 nm)

in amorphous  $\text{Si}_3\text{N}_4$  matrix and obtained its hardness of 50–70 GPa. Similarly, Zhang et al. [93] prepared a coating with a hardness of about 40 GPa by embedding TiCrCN nanocrystalline (8–15 nm) in an amorphous DLC matrix.

In another direction, in case of Ni-P electroless coatings, incorporation of other nanoparticles, such as CNT [111, 112], nano- $\text{SiO}_2$  [113, 114], nano-SiC [175, 176], nano-WC [115], and nano- $\text{Al}_2\text{O}_3$  [116], increased their hardness. The hardness of all these nanocomposite coatings also increased after thermal posttreatment at 400°C during 1 h. Similar result was obtained for the thermal sprayed coatings [47]. Ali et al. [47] reported that the thermal posttreatment allows increasing hardness of WC—12 wt.% Co nanocomposite coatings by crystallization and improving the bonding of microstructural phases.

**3.3. Effect of Nanofillers on Thermal Property of Coating.** In the case of organic matrices, generally the incorporation of nanometer-sized inorganic particles into the polymer matrix can enhance thermal stability by acting as a superior insulator and mass transport barrier to the volatile products, generated during decomposition [177]. Adding the nanoparticles into polymer matrix modifies its glass transition temperature ( $T_g$ ). An increase in  $T_g$  has been reported in few cases for polystyrene and other polymers [178–180], whereas few other reports have demonstrated a reduction of  $T_g$  when nanofillers were added to polymers, such as the alumina/poly(methyl methacrylate) (PMMA) nanocomposites [181]. Therefore, the relationship between the interaction mechanism and  $T_g$  is still an open question. In a recent research [5], the DSC tests revealed that all of the epoxy nanocomposite coatings, except the one modified with the  $\text{Fe}_2\text{O}_3$  nanoparticles, showed an increased glass transition temperature value. The incorporation of nano- $\text{TiO}_2$  led to an increase of the  $T_g$  value of the epoxy coatings from 58°C to even 170°C. The TGA test also showed that, except for nano-ZnO, the thermal stability of the modified epoxy nanocomposites was not superior to that of the neat epoxy resin at low temperature. During high temperature periods, nano- $\text{SiO}_2$ , nano- $\text{Fe}_2\text{O}_3$ , and nano-ZnO enhanced the thermal stability of the epoxy matrix. Among the epoxy nanocomposites, the one modified by nano-ZnO showed the best thermal stability. By using DSC and DMTA, Zabihi et al. [182] also found that  $\text{Fe}_2\text{O}_3$  nanoparticles improved the crosslink density of epoxy, as evidenced by the increase in the glass transition temperature. In case of inorganic matrices, for thermal sprayed coatings, the nanocomposite structure had higher thermal shock resistance than the conventional structure [183, 184].

**3.4. Effect of Nanofillers on the Anticorrosive and Antiwear Properties of Coatings.** In the case of organic matrices, it is reported that epoxy based coatings [4] containing metal oxides nanoparticles significantly improved the corrosion resistance of the coated steel, with the  $\text{Fe}_2\text{O}_3$  and halloysite clay nanoparticles being the best. The EIS measurements indicated that the incorporation of nanoparticles increased the coating resistance and the charge transfer resistance while reducing the coating capacitance and the double layer capacitance. Zhang et al. [185] reported that incorporation

of nano- $\text{SiO}_2$  into epoxy reduced its friction and wear at low nanofiller content (~2 vol%). For polyester coatings, Golgoon et al. [35] reported that the embedding nanoclay led to the absence of pores and flaw in coatings, as well as creating a protective barrier against corrosive electrolytes penetration; thus the corrosion resistance of nanocomposite coatings was higher than the pure coatings. They also reported that, with the presence of nanoclay, the wear rate of nanocomposite coating was much lower than neat coating because of relatively denser structure in the presence of nanoparticles.

In the case of inorganic matrices, for Ni-P electroless coatings, the incorporation of nanoparticles, such as nano- $\text{SiO}_2$  [186], nano- $\text{Al}_2\text{O}_3$  [116, 117, 187], nano- $\text{CeO}_2$  [188], and CNT [112], increased their anticorrosion in NaCl and  $\text{H}_2\text{SO}_4$  solutions. The presence of nanoparticles in Ni-P matrix also reduced their wear rate, as reported for CNT [112], nano- $\text{Al}_2\text{O}_3$  [116], and nano- $\text{SiO}_2$  [113, 114]. For electrodeposited coating, incorporation of hard SiC nanoparticles into metallic matrix led to having better corrosion resistance and excellent wear resistance [152, 189–193]. Similar results are also obtained for magnetron sputtered coatings. Wei et al. [194, 195] reported that the presence of metal carbides nanocrystallites in amorphous matrix exhibits the low-friction behavior within a wide load range. The presence of nano- $\text{Al}_2\text{O}_3$  in the Ni matrix also increased its anti-sand-wear performance for steel substrate [119].

**3.5. Nanoparticles in Nonwetting Nanocomposite Coatings.** Surfaces are considered as superhydrophobic when water contact angle (WCA) is greater than 150°. The use of superhydrophobic coatings to remove water on metal surfaces is gaining a lot of attention due to the special benefits of (i) self-cleaning surfaces, (ii) protection against freezing or snow (anti-icing) on the surface of the metal in the wintry winter weather, (iii) antifouling properties, (iv) barrier to atmospheric corrosion for metals in the marine environment, and (v) minimizing some of the unwanted effects on the surface of metals in outdoor conditions [196–200]. Depending on the type of material, various superhydrophobic coatings have been successfully developed in different scales. Manca et al. [196] reported self-cleaning and antiglare coating on glass plates using modified nanosilica particles with sol-gel technique.

He et al. [197] used myristic acid to successfully treat the hydrophobic surface for aluminum anodization, thereby improving corrosion resistance in seawater. Kako et al. [200] found that changes in surface morphology (from hydrophobic channels to superhydrophobic surfaces) could alter the ability to cling to or slip snow particles. Sarkar and Farzaneh [201] argued that ice shovels were removed from the surface of the superhydrophobic material, while that of ice deposited on bare aluminum surfaces was large at  $369 \pm 89$  kPa. Kulinich and Farzaneh [202, 203] also commented on similar results. Li et al. [204] have introduced hydrophobic polydimethylsiloxane (PDMS)/nanosilica overlays to reduce ice deposition. The authors found that both coatings exhibited super hydrophobicity, in terms of both multiscale and low surface energy.



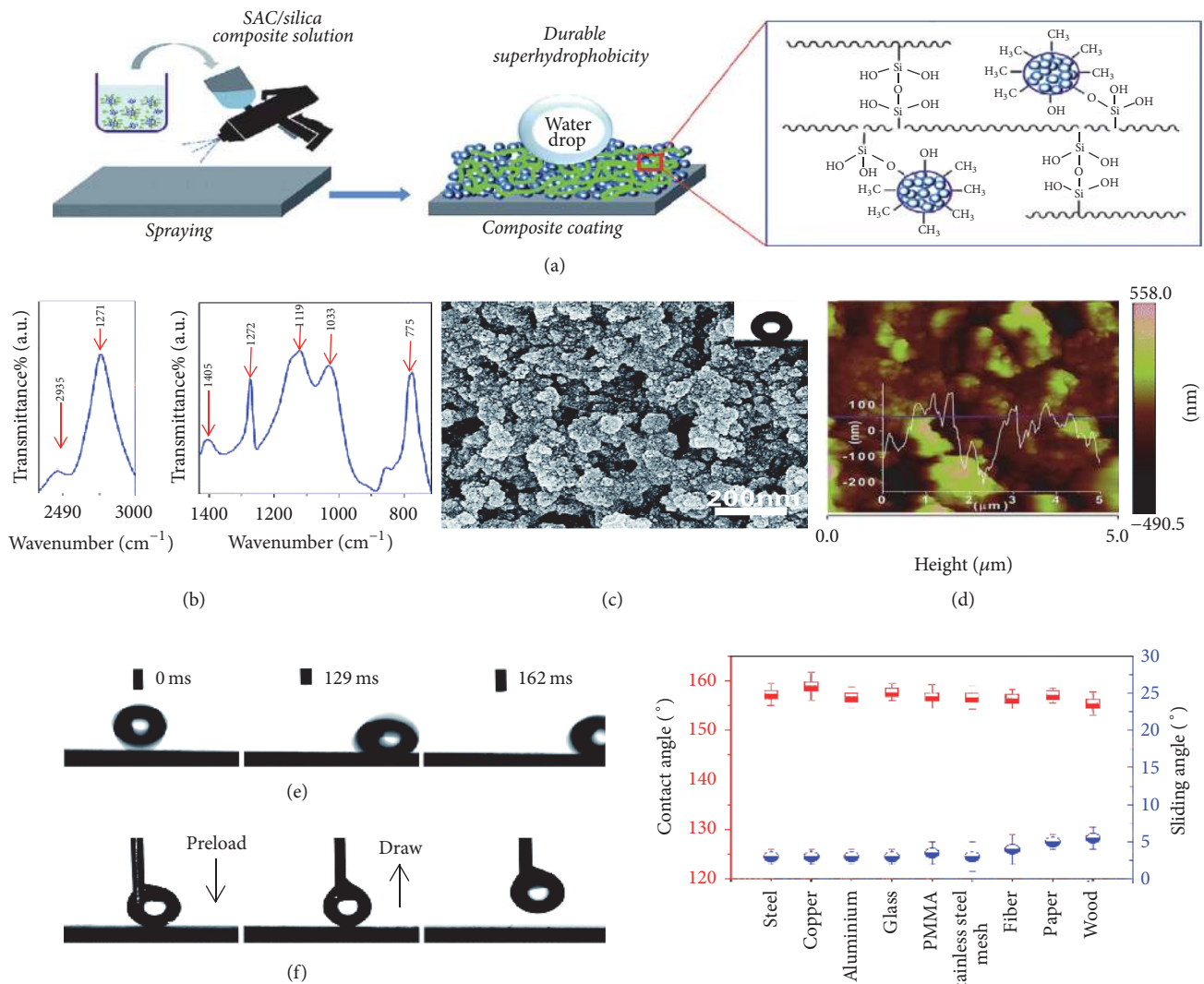


FIGURE 8: Synthesis of the composite superhydrophobic coating. (a) Schematic of the fabrication of the superhydrophobic coating and the possible bonding structure in the composite coating. (b) FTIR spectra of the composite coating. (c) SEM image of the composite coating on a glass slide. (d) AFM height image of the superhydrophobic surface. Inset: AFM section images of the positions marked by the line. (e) Time-resolved images of a 9.0 μL water droplet spreading quickly on the composite coating within 162 ms (tilting angle < 3°). (f) The contact, deformation, and departure processes of a water droplet on the composite coating. (g) The water contact angles and sliding angles of the composite coating on various substrates [142].

The hydrophobicity is usually improved by increasing the surface roughness [205, 206] and the hydrophobic substrate requires an adequate surface roughness [207]. Therefore, techniques for making hydrophobic coatings are divided into two steps: (i) creating of a rough surface at microscale for host materials and (ii) creating nanoscale architecture surface (see Figure 9), followed by the deposition of low surface energy materials on the surface. The most common low surface materials are fluorinated and silane compounds [207]. There are many ways to increase the surface roughness: mechanical stretching, physical methods (laser, plasma), chemical etching, lithography, sol-gel processing, electrochemical reactions, or even chemical deposition in CVD vapor phase.

It should be noted that when the roughness increases, the angle of contact with water will increase or decrease

depending on the surface being hydrophobic or hydrophilic [205]. Furthermore, Zhu et al. [208] argued that both the nanostructure and microstructure of the surface contribute to its hydrophobicity, as observed in the nature (the lotus leaf).

Recently, nanoparticles have been used to obtain superhydrophobic surface by turning surface roughness. In that method, the nanoparticles are applied to a fine surface or microroughness to produce a surface roughness at the submicrometer scale. Surfaces that have been sanded continue to be chemically treated or covered with a low surface energy material to increase hydrophobic effect [209–211]. In another way, the nanoparticles are dispersed in the polymer solution and sprayed onto the smooth surface (such as the glass in the self-cleaning paint) [208, 212–215] as shown in Figure 8. Recently, Milionis's group [216–220]

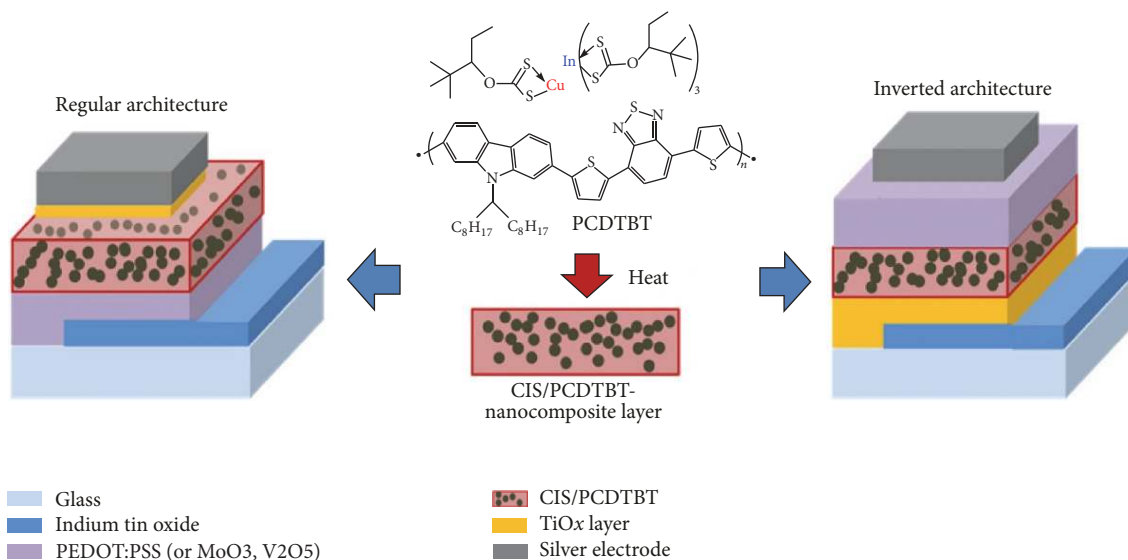


FIGURE 9: Schematic illustration of the regular and inverted device architectures and the in situ route using metal xanthates as precursors for the formation of metal sulfide nanocrystals directly in polymer matrices [143].

developed some nonwetting (superhydrophobic) coatings for specific applications, particularly for underwater-saturation or tribological purposes using nanosilica or carbon nanotube dispersed in various polymer matrices.

#### 4. Applications

Table 1 presents the summary of materials, preparative methods, and applications for 4 different types of nanocomposite coatings. As can be seen in this table, the most used type of nanocomposite coatings is organic/inorganic (O/I) type (matrix/nanofiller) with various applications, such as anticorrosion, antiwear, superhydrophobic application, self-cleaning, antifouling/antibacterial application, and electronics.

In the case of anticorrosive and self-healing coatings, a specific type of nanofiller is often incorporated in their formulation, denoted as nanocontainer. Nanocontainer is of a nanosized volume and contains the active substances (inhibitor-loaded nanocontainer and healing agent-loaded nanocontainer). As designed, the nanocontainer could prevent the direct contact between these active agents and coating matrix. As the nanofillers, nanocontainers were well dispersed in the coating matrix; then they can release these healing agents under critical conditions, such as mechanical rupture, changing pH, and light change. Nanocontainers included the halloysite clay nanotubes [221–223], zirconia nanospheres [224], nanosilica [225–227], nanoceria [228, 229], and supramolecular nanocontainers [230, 231].

#### 5. Conclusion and Future Trend

We tried to trace here an overview of the nanocomposite coatings in both basic fundamental and last recent developments

in design, preparation, and applications of the nanocomposite coatings. With a rapid growth rate of the nanotechnology and related fields, nanocomposites coatings today become smarter, cheaper, and more functional. The domains of application of nanocomposite coatings are thus expected to be larger in the future, dealing with drug delivery systems, anticorrosion barrier coatings, antibacterial coatings, self-scratch repair, fire retardant coatings, reflective coatings, and screen effect coatings.

The nanocomposite coating today not only serves as a protection for the materials but also plays other roles due to the presence of multifunctional nanofillers. Two most popular examples can be cited, and they are antibacterial coatings and smart coatings which are used for sustainable energy fields. In the first case, the nanofillers based silver nanoparticles and their related products are very promising in the next decades. In the future, we will face many risks and challenges, especially energy problems, and the research on the sustainable energy conversion is expected to explode, in terms of both theory and experiment, and the nanocomposite coating will not stand out of this trend, for example, self-cleaning or “easy-to-clean” coatings, coated on building, protective substrates and on glass, can help save energy and water in facility cleaning while insulant nanocomposite coatings help to save the energy loss saving billions of dollars for maintaining homes in winter, especially in North America where the winter is cool and long (see Figure 5).

The smart coatings play also an important role in the development of new generations of sustainable energy where the multifunctional coatings can be used for the elaboration of devices in photovoltaic solar, solar thermal energy, and solar-to-fuel conversions process.

TABLE I: Summary of materials, preparative methods, and applications for nanocomposite coatings.

Matrix	Nanofillers	Preparative method	Application	Reference
Epoxy resin	<i>Organic/inorganic nanocomposite coatings</i> Oxides, clay	Ultrasonic stirring, solution dispersion	Anticorrosion	[4-7]
Polyester	Clay	Electrostatic method	Anticorrosion Antiwear	[35]
Perfluoroacrylic polymer	ZnO	Spray atomization	Superoleophobic	[29]
Fluoropolymer	CNT, graphene	Drop casting suspensions	Superhydrophobic	[30]
Pullulan	Clay	Ultrasonic-assisted	High oxygen barrier	[28]
PANI	Clay	Electrodeposition	Anticorrosion	[19]
Fluorinated polysiloxane	TiO <sub>2</sub>	Blending ambient-cured	Superhydrophobic self-cleaning	[34]
Ethylene tetrafluoroethylene	PANI/CNT	Ultrasonic stirring, air spray	Self-cleaning, antiwear, anticorrosion	[31]
Polydimethylsiloxane	SiO <sub>2</sub>	Ultrasonic stirring, brushing	Superhydrophobic	[38]
Polydimethylsiloxane	Al <sub>2</sub> O <sub>3</sub> /Clay	Ultrasonic stirring, spraying	Superhydrophobic	[149]
Polymers with reactive trimethoxysilyl groups	SiO <sub>2</sub>	Ultrasonic stirring, spin coating/dip coating	Superhydrophobic	[26]
Polyacrylate	SiO <sub>2</sub>	Dip coating	Superhydrophobic and oleophobic	[107]
Epoxy resin	ZnO/PANI	Dip coating	Antifouling and antibacterial	[150]
Polycarbonate	CNT	Ultrasonic stirring, casting	Infrared sensor	[33]
Poly(3-hydroxy-2,3-dimethylacrylic acid)	TiO <sub>2</sub>	Spin coater	Color filters for liquid crystal displays	[148]
Polyvinyl alcohol	ZnO	Sol-gel	Biosensor	[37]
Polyvinylidene Fluoride	Au	Ultrasonic stirring, spin coating	dielectric	[16]
Hexafluoroacetylacetonate precursor	Pt	Plasma chemical deposition	Ion- and electron-conducting	[91]
Monomers (N-isopropylacrylamide and/or dodecyl methacrylate)	SiO <sub>2</sub>	Evaporation-induced self-assembly, dip coating	Environmentally responsive coatings	[151]
PANI	<i>Organic/organic nanocomposite coatings</i> Nanocellulose	In situ polymerization	Electrochromic	[152]
Ni-P	<i>Inorganic/inorganic nanocomposite coatings</i> Al <sub>2</sub> O <sub>3</sub>	Electroless deposition	Anticorrosion Anti-wear	[116, 117]
Co	CeO <sub>2</sub>	Electrodeposition	Anticorrosion	[122]
Ni	TiO <sub>2</sub>	Electrodeposition	Anticorrosion	[118]
Ni, Cu	TiN, ZrN, ZrN, CrN	Magnetron cosputtering	Superhard	[73-77, 153]
Diamond-like carbon	TiC	Laser-assisted deposition	Superhard	[103, 154]
Diamond-like carbon	TiCrCN	Magnetron sputtering	Superhard	[93]
Si <sub>3</sub> N <sub>4</sub>	TiN	PVD	Superhard	[155, 156]
Ni-P	<i>Inorganic/organic nanocomposite coatings</i> PTFE	Electroless deposition	Anticorrosion Anti-wear	[81]
Ni-P	PTFE	Electroless deposition	Antibiofouling	[82]

## Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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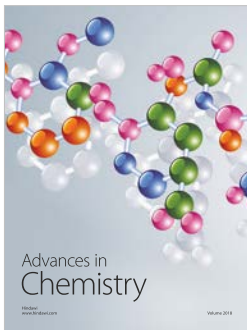
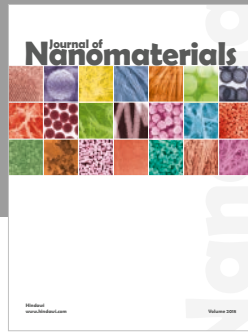
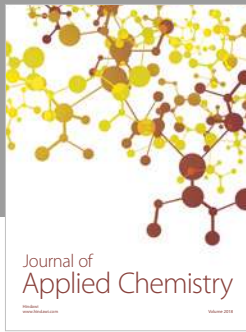
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