Research Article

A Review on Polymer nanocomposites: Monometallic and Bimetallic Nanoparticles for Biomedical, Optical and Engineering Applications

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Abstract

Polymer metal nanocomposites have been extensively investigated due to their potential ability to provide materials with novel mechanical, electronic or chemical behaviour for technological applications. Many synthesis methods have been proposed for the metal naocomopsite and their detailed characterizations on polymer metal nanocomposites have been studied. The most recent achievements and trends in the field of polymer monometallic and bimetallic nanocomposites have been reviewed with a focus on creative approaches to synthesis, properties and functions for Biomedical, Optical and Engineering Applications.

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Introduction

Recent advances in nanotechnology have made the nanoscience a field hot area research, making it one of the most research areas of science in the past two decades. Nanocomposites are a new class of materials in which the dimension of the dispersed particles occurred at the nanometer scale. Thus in polymer-metal nanocomposites, metal particles are dispersed in the polymer matrix at the nanometer scale. Polymer metal nanocomposite research has become an attraction and also important due their enhanced properties compared to the bulk materials [1]. Due to their multi-functionality, ease of process-ability, potential for large-scale manufacturing, significantly lighter than metals and ease of synthesis made them as important materials for diverse applications[2]. Some of the properties include catalytic properties, thermal properties, electrical conductivity, optical properties and antimicrobial properties [3–5].

The properties of nanocomposites are influenced by their high surface energy due to their high surface area to volume ratio and relatively small sizes. Synthesis of nanocomposite polymers are easy due to their ease of processing, solubility less toxicity. It is also possible and also because of the possibilities to control the chemical and physical properties via the design of 3D gel structures which provide a powerful strategy to incorporate versatility into engineering gels from the nanometer scale.

The advanced application of metallic nanoparticles require techniques to synthesize nanoparticles that are both cost-effective and eco- friendly. Several processes for metallic nanoparticle synthesis have been developed, however, the synthesis of nanoparticles with precise control over size distribution, shape selectivity and stability remains a challenge [6].

Polymer metal nanocompoites can be prepared by both physical and chemical methods. The physical methods involve vapor deposition, which in principle consist of subdividing bulk precursors to nanoparticles. Chemical procedures start from reduction of metal ions to metal atoms, followed by controlled aggregation of atoms. The mass production of metal nanoparticles by the chemical method is more effective than the physical one. The most common synthesis of polymer hydrogel nanocomposite, have focused on systems utilizing poly (ethylene oxide), poly(acryl amide) or poly(vinyl alcohol) as the polymer. Therefore, hydrogel nano-composites containing these polymers have received greater part of our interest in recent publications and also we are discussing on polymer monometalic, polymer-bimetallic and natural polymer nanocomposite hydrogels. Metal nanocomposite polymer hydrogels may be defined as cross-linked polymer networks swollen with water incorporation of meatl nanoparticles or nanostructures. The polymer is a cross-linked structure to forming a network via chemical or physical interactions as shown in Fig1.

The chemical cross-linking is permanent bonding due to covalent bonds. The physical interactions are non-covalent in nature and often a result of hydrogen bonding, hydrophilic, and ionic interactions. The cross-linked polymer networks are capable of reversible volume change in response to external stimuli such as composition [due to (de)swelling], temperature, and pH.

The nanoparticles present in the polymers can serve either to crosslink the hydrogel, to adsorb or attach to polymer chains, or to add new properties to the hydrogel by simply being entrapped within the hydrogel network. Nanoparticles add unique physical properties to polymer hydrogels such as responsiveness to mechanical, optical, thermal, barrier, sound, magnetic, electric stimulation, etc. These exclusive properties lead to applications in the fields of electronics, optics, sensors, actuators, microfluidics sectors as catalyst, separation devices, drug delivery, food-jelly and many other biotechnological areas. Finally, it is concluded with a brief outline on perspectives for the future challenges that drive the development of novel nanocomposite polymer hydrogels.

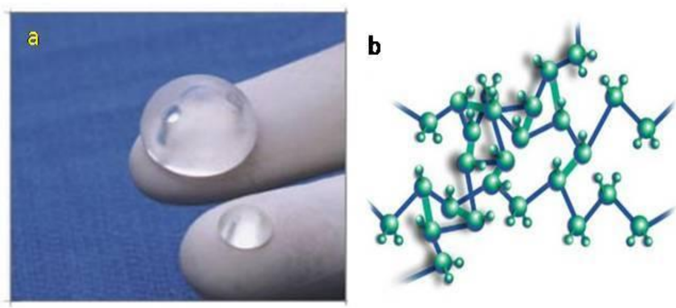

Figure 1 Structure of Hydrogel **:** (a) Polymer hydrogel and (b) cross linked polymers

Synthetic Methods for Polymer-Metal Nano-Composites

During the synthesis of nanocomposites, one must consider the properties of the polymer matrix as well as the stability of the nanoparticles and more importantly, the prevention of particle aggregation [7].

The control of nanoparticle morphology is an important aspect, since the morphology extremely influences the material performance. As a long term goal the development of synthesis schemes are able to control the particle size, shapes and composition independently from one another which is very important, in order to allow the tuning of nanocomposite properties. Metal–polymer nanocomposites can be obtained by two different approaches, namely, *ex situ* and *in situ* metohods.

Ex-situ method

Different approaches were used to prepare polymer-metal nanocomposites. In the older method, polymerization of organic monomer and the formation of metal nanoparticles are performed separately, and polymer matrix and metal nanoparticles are physically or mechanically mixed to form polymer-metal nanocomposites. The metal nanoparticles are not homogeneously dispersed in the polymer matrix (8).

In-situ Method

Recently in-situ synthesis is in use for the preparation of polymer-metal nanocomposites. This method is based on the reduction of metal ions that are dispersed in polymer matrices or the polymerization of the monomer dispersed with metal nanoparticles (9). Simultaneous polymerization-reduction approach is also another version of the *in-situ* synthesis. The polymerization of the organic monomers is carried out in parallel with the formation of the metal nanoparticles.

In the *in situ* methods, metal particles are generated inside a polymer matrix by decomposition (e.g., thermolysis, photolysis, radiolysis, etc.) or chemical reduction of a metallic precursor dissolved into the polymer. In the *ex situ* approach, nanoparticles are first produced by soft-chemistry routes and then dispersed into polymeric matrices. The *ex-situ* techniques for the synthesis of metal/polymer nanocomposites are frequently preferred to the *in situ* methods because of the high optical quality that can be achieved in the final product.

Bimetallic Nanoparticle Hydrogels

Bimetallic nanoparticles, composed of two different metal elements are of superior interest than monometallic nanoparticles, from both the scientific and technological point of view, for the improvement of the catalytic properties of metal particles [11-13]. This is because bimetallization can improve the catalytic properties of the original single-metal catalysts and create a new property, which may not be achieved by monometallic catalysts. These effects of the added metal component can frequently be explained in terms of an ensemble and/or a ligand effect in catalyses. Sinfelt and coworkers have vigorously studied inorganic oxide-supported bimetallic nanoparticles for catalysis and analyzed their micro-structures by an EXAFS (Extended X-Ray Absorption Fine Structure) technique [14-16]. These supported bimetallic nanoparticles have already been used as effective catalysts for the hydrogenation of olefins and carbon-skeleton rearrangement of hydrocarbons [17]. The alloy structure can be carefully examined to understand their catalytic properties. In contrast, colloidal dispersions of bimetallic nanoparticles have not been well examined [18].

Monometallic Nanoparticle Hydrogels

Hybrid materials can be obtained by combining metal based nanoparticles such as gold and silver with polymer hydrogels. There is a slight effect on the mechanical properties of the resulting nanocomposite hydrogels by the incorporation of metal nanoparticles as long as the interactions between polymer and nanoparticles are weak. In this case, phase transition, thermosensitivity, and viscoelasticity of the polymer gel remain unchanged, and the properties of the nanoparticle such as improved electrical conductivity, response to optical stimuli, improved antimicrobial properties, etc. are added to the gel. Hydrogels based on Poly(acrylamide)

(PAM) may show sensitivity towards external stimuli such as pH, light, temperature, solvent, pressure and magnetic fields [19-24]. Poly (acryl amide) (PAM) based gels such as poly (N-isopropyl acryl amide) (PNIPAM) are more important in many aspects. The PAM polymers chains are more flexible than the PNIPAM polymer, which leads to differences in elastic recovery, hysteresis, tensile strength, and elongation [25]. Metal nanoparticles can be prepared by two distinct ways. The subdivision of bulk metals (a physical method) and by the growth of particles starting from metal atoms, which are obtained from molecular or ionic precursors (a chemical method) (Fig. 2)

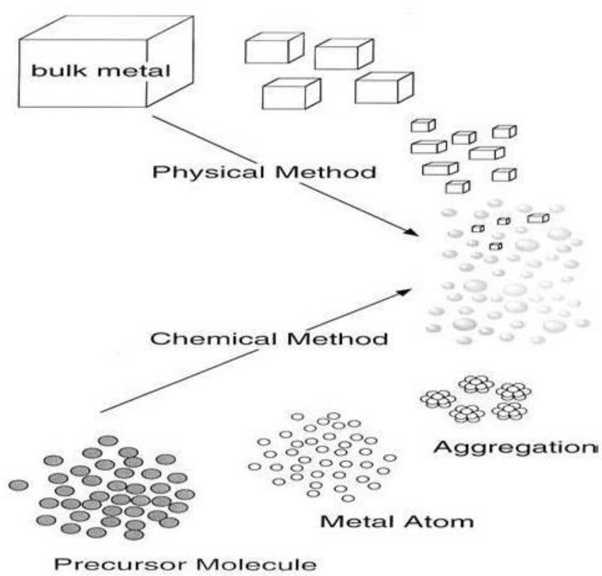

Figure 2 Schematic illustrations of preparative methods of metal nanoparticles

Characterization methods of polymer-metal nanocomposites

Characterization methods of polymer-metal nano-composites provide the information on sizes, shapes and physicochemical properties of the nanoparticles.

UV visible spectroscopy

Colour is the most immediate observable property for certain metal nanoparticles dispersions. This colour effects based upon the surface plasmon resonance or particle plasmon resonance. Noble metal nanoparticles such as Au, Ag and Cu have characteristic colours that are related to their particle size, shape, and particle distance and on the matrix material .For these metals, observation of UV-visible spectra will be a useful complement to other methods in characterizing the nanomaterials [44]. For example, information on the formation of bimetallic nanoparticles can be derived from this technique [28]. The UV-visible spectra of bimetallic nanoparticles are not simply the sum of the two monometallic nanoparticles[26]. If two separate metallic phases are present, the absorption spectrum would be characterized by double peaks due to the SPR of single metal nanoclusters provided the two metals have characteristic SPR[27]. Comparison of spectra of bimetallic nanoparticles with the spectra of physical mixtures of the respective monometallic particles can confirm the formation of bimetallic nanoparticles (Fig 3).

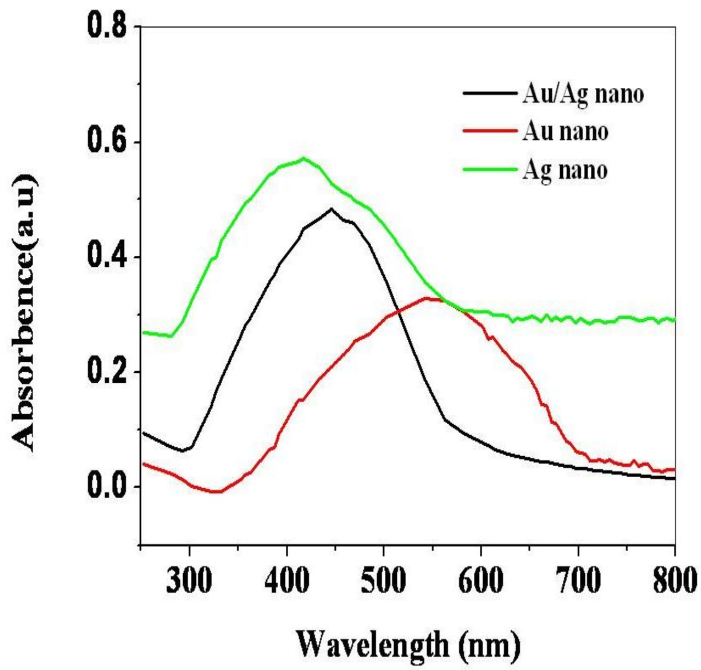

Figure 3 UV spectra of nanocomposites

Infrared spectroscopy (IR spectroscopy)

IR spectroscopy is one of the methods most extensively used for the investigation of polymer structure as IR band characteristics of the polymers are readily observed [10]. It is very helpful in the characterization of nanocomposites prepared by the polymerization of the monomers as the formation of the polymer in the nanocomposite is confirmed by this method. To prevent agglomeration of nanoparticles during polymerization and to achieve their compatibility with the polymer, the surfaces of some nanoparticles had been coated with oleic acid. IR spectroscopy gives evidence of the surface coating and is used to investigate the surface chemistry of the nanoparticles as polymer-stabilized nanoparticles give IR bands characteristics of the polymer (Fig 4).

X-Ray diffraction (XRD)

X-ray diffraction (XRD) is a method of choice to investigate the solid structure of metal nanoparticles and morphology of polymer in polymer-metal nanocomposites. The phase changes with increasing diameter of nanoparticles and the crystal structure can be determined with XRD. The polymer and the metal nanoparticles can be identified by their characteristics peaks in XRD patterns. This technique can investigate the formation of bimetallic nanoparticles as opposed to a mixture of monometallic nanoparticles, since the diffraction pattern of the physical mixtures which may consist of overlapping lines of the two individual monometallic nanoparticles and is distinct from that of the bimetallic nanoparticles. Information on the morphology of the polymer constituent of the composite could be derived also from XRD (Fig 5).

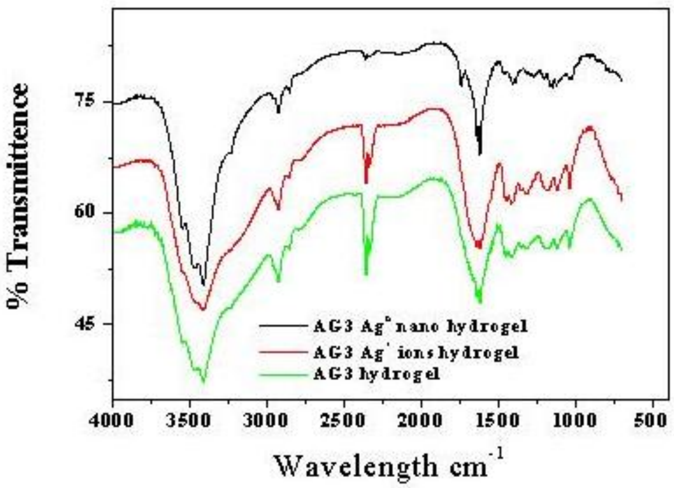

Figure 4 FTIR spectra of polymer nano-composites

The technique can reveal the degree of crystallinity in the polymer component of the nanocomposites as sharper peaks with increased relative peak intensity implies a more ordered arrangement shown by the XRD spectrum of metal nanocomposite. The broad reflection in the range of $2\theta = 15 – 35$ indicates the a low order of crystallinity and is attributed to the nanocomposites while sharper reflections at $2\theta > 35$ are attributed to gold nanoparticles [28].

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis finds application in the characterization of polymer-metal nanocomposites as it provides information on the thermal stability of the nanocomposite relative to the polymers. During composite formation, stabilizers (polymers/surfactants) make organic coating on the nanoparticles surface and TGA can be used to evaluate the amount of stabilizer chemisorbed on the surface of the nanoparticles [10].

Scanning Electron Microscopy (SEM)

Hydrogel nanocomposite samples (surface or cross-sections) were coated with a thin layer of palladium-gold alloy after mounting on a double sided carbon tape. The morphological variations of silver nanoparticles were by using scanning electron microscope [28].

Transmission electron microscopy (TEM)

Transmission electron microscopy (TEM) was used to find out the size of nanoparticles inside the hydrogel nanocomposites. Size analysis of metal nanoparticles was carried out using electron microscope. Solid samples were studied in dilute aqueous suspensions while in the case of sols, droplets of the undiluted material were placed on Formwar-coated copper mesh grids (diameter 2 mm). The average particle diameter and size distribution of the samples were determined by using TEM images. The nanoparticles formed inside the gel networks are spherical in shape could be found [44].

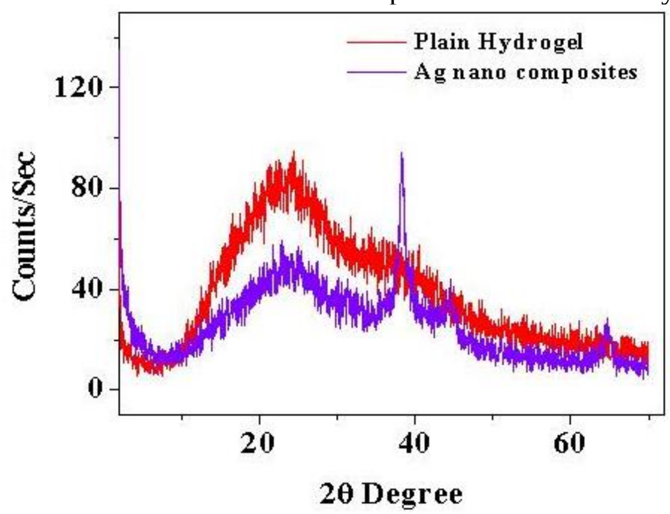

Figure 5 X-Ray diffraction of polymer nanocomposite

Nanocomposite Polymer Hydrogels for Biomedical Applications

There are many examples of nanocomposite hydrogels being used as biomaterials in drug delivery, tissue engineering, and general biomedical applications. For a rational design of biocompatible and nanocomposite hydrogels, the structure and properties of the hydrogels must be considered because the nanoscale morphology of the hydrogel surfaces and interfaces will affect cell adhesion, migration, proliferation, and gene expression.

Antibacterial Activity

To examine the antimicrobial activities of the nanocomposite hydrogels, they were tested with nutrient agar media and absorbance count method.

Disc Method

Nutrient agar medium was prepared by mixing peptone (5.0 g), beef extract (3.0 g), and sodium chloride (NaCl) (5.0 g) in 1000 mL of distilled water and the pH was adjusted to 7.0. Finally, agar (15.0 g) was added to the solution. The agar medium was sterilized in a conical

Table 1 Applications of polymer metal nanocomposites

Polymer matrix	Nanoparticle	Applications	Reference
PolyAcrylamide	Gold-Silver	Antimicrobial, wound dressing	28
Chitosan	Copper	Pharmaceutical, biomedical	29
Poly Styrene	Silver	Inorganic polymer nano composite	30
PEO-PPO	Gold-Silver	Multimetallic colloids	31
PTFE	Silver	Electrical insulating	32
THEMA	Bismuth	Coating	33
PVA	Gold-Silver	Optical	34
TMSPA	Gold	Electrical	35
PVB	Gold-Silver	Bio sensor	36
PDMA–*b*–PMMA–*b*–PDMA	Silver	Optical	37
PVP	Silver	Optical	38
Chitosan	Silver	Antimicrobial	39
PolyAcrylamide	Silver	Antimicrobial	40
Poly(styrene-co-acrylic acid)	Silver	Antimicrobial	41
Chitosan	Silver	Antimicrobial	42
Polylactide	Silver	Biomedical	43
(AAm) N-vinyl-2-pyrrolidone	Silver	Biological	44
PAM-PVA	Silver	Antimicrobial, wound dressing	45
Chitlac	Silver	Bioactive biomaterials	46
PNIPAAm	Iron	Biocompatibility	47
PAM	Zinc	Optical	48
PNIPAAm	Iron	Drug delivery	49
Polyimides	Silver	Electrical Conductivity	50
Poly(methylhydrosiloxane)	Silver	Coating	51
PGMA-co-PNIPAM	Gold	Nano reactors	52
P(AMPS)	Nickel	Nano catalyst	53
Chitosan	Silver	Antibiotics	54
Chitosan	Silver , Gold , Pt, Pd	Morphological studies	55
poly(HEMA–PEGMA–MAA)	Silver	electronics, biosensors and drug delivery	56
PVP	Silver	Noninvasive biomonitering	57
PNIPAAm	Iron	Remote controlled drug delivery	58
copoly(TEAMPS/VP)	Silver	Humidity Senor	59
PVA	Silver	Polarizing filters/ flexible films	60
PANI	Silver	Ethanol sensor	61
PMMA	Silver	Thermal stability	62
PEG	Iron	Drug delivery, hyperthermia therapy	63
Polyaniline	Gold	Sensors	64
Poly isoprene	Silver	Latex gloves	65
PET	Silver	Band pass filter	66
Polypyrrole	Platinum	Fuel cell	67
PVC	Copper	Biotechnology	68
Polyaniline	Copper	Sensors	69
PTFE	Silver	Optical filler/sensor	70
PEDOT: PSS	Gold-Silver	Electrochromic device	71
Bisphenol A	Silver	Capacitor	72
Polydimethylsilaxane	Gold	SERS	73
PANI-PSS	Platinum	Fuel cell	74
Polysulphon	Platinum	Sensors	75

flask at a pressure of 15 lbs for 30 min. This mixture was transferred into sterilized Petri dishes in a laminar air flow chamber. After solidification of the media, bacillus culture (50 µL) was spread on the solid surface of the media .To the inoculated Petri dish, one drop of nanoparticles solution (20 mg/10 mL distilled water with alkali treatment) was added using 50-µL tip and incubated for 2 days at 37^0C in the incubation chamber [44].

Absorbance Count Method

The effect of bacterial growth of bacillus in mineral salts medium (MSM) was studied in the presence of nanoparticles (Ag, Au, Au-Ag nanoparticles)(28). This medium was prepared by the following composition: NH_4NO_3 (1.5 g), KH_2PO_4 (2.5 g), K2HPO$_4$ (0.5 g), NaCl (1.0 g), $MgSO_4$ (1.5 g), $MnSO_4$ (0.01 g), $FeSO_4$ (0.05 g), and $CaCl_2$ (0.05 g) were added to 1000 mL of distilled water and the pH was adjusted to 7.0. Then, yeast extract (0.01%) was added for bacterial growth. After that the MSM medium was sterilized and 50 mL of solution was transferred into a sterilized 250-mL conical flask. Afterward, 100 µL bacillus bacterium was added into the media. Finally, 100 µL of nanoparticles solution (10 mg/5 mL distilled water) or its equivalent nanoparticles suspension was added, and the optical density of the bacterial medium was measured using a UV-vis spectrophotometer at 600 nm.

Conclusions

The advances in nanotechnology have proved to find excellent approaches for the stabilization and production of controlled nanostructures using various polymeric systems. From stimuli-responsive sensors and actuators to microfluidics, pharmaceutical, and biomedical devices, the potential impact for nanocomposite hydrogels to influence the lives of the general public continues to grow. The large amount of recent reports has explained some of the physics and chemistry behind the unique properties of these hydrogel materials. Responsive hydrogels that change properties and function as response to external stimuli such as artificial muscles are often inspired by natural systems. The development of synthetic and fabrication technologies for fundamental understanding of nanocomposite hydrogels will continue with a greater emphasis on designing sophisticated multi component and complex materials that can be tailored to very specific applications. Emerging new techniques strongly support the systematic characterization of nanocomposite gels which, in return, drives research forward and impacts the rational design of materials.

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