INTERACTIONS OF CARBON NANOPARTICLES FROM PACKAGINGS WITH COMPONENTS OF FOOD, DRUGS AND BIOLOGICALLY ACTIVE MOLECULES - A REVIEW

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Abstract

Nanomaterials are very important in the field of packaging of food, medicines and dietary supplements. Modern packagings often contain nanoparticles that provide them new feature - nanoparticles are used to activate mainly the packaging inner surface. Carbon occurs in several allotropic forms, such as diamond, graphite (including nanotubes and fullerenes), carbides, and nanocrystalline diamond which is produced in a process of radio frequency plasma activated chemical vapor deposition (RF PA CVD). Variety of allotropic forms of carbon results in different chemical and biological interactions between carbon nanoparticles and the polymer matrix material. Carbon nanopratricles can be used to activate the inner surface of packagings. There is a growing demand for food free of harmful chemicals such as chloramphenicol or toxic food colorings (metanil yellow, auramine, orange II or red aura). The use of nanotechnology in the food packaging sector opens up new possibilities for creating sensors to detect certain harmful analytes. These sensors are easy and quick to use. The basis of their actions is to understand the interactions between nanoparticles and chemicals. Nanoparticles can be utilized to create intelligent high performance packaging materials for contact with food, drugs and biologically active molecules, which will be safe for health of the consumers.

Keywords: *interaction, carbon, nanopractices*

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Introduction

Food can be called “nanofood” when nanoparticles, nanotechnology tools or techniques are used during its production, cultivation, packaging or processing. This does not mean that food is modified at atomic level or is produced by nanomachines. Strategies to apply nanoscience to the food industry are quite different from traditional applications of nanotechnology [1,2]. This is why the definition of food control and functional food is changing.

“Nano” approach can be used to control and manipulate interactions between food ingredients such as proteins, lipids and polysaccharides providing desirable rheological and structural properties of food [2,3].

There are several advantages of materials in the form of nanoscale objects in comparison with micro scale objects. Nanoscale materials act in a different way than typical macroscale materials [2,4]. Recently, innovations in packaging industry turn from the macroscopic to the nano-scale. It really becomes important to develop food products using nanomaterials, chemical and physical properties. Nanotechnology and nanosciences should be used together in food innovations, novel food development, dietary supplements as well as medicines and biologically active molecules [2,3].

This review paper presents recent advances in application of carbon nanoforms in the packaging of food, dietary supplements, drugs and biologically active molecules.

Nanocomposites consisting of carbon nanotubes for the detection of toxic substances in food and dietary supplements

Growing concerns about the safety and security of food, cause that cheaper and faster methods of contaminants detecting are developing to ensure safety of food [5-7]. Chloramphenicol is a well-known veterinary medicine with a broad spectrum of activity for the treatment of infectious diseases in animals [7-9]. Chloramphenicol overdose leads to chronic toxicity resulting in myelosuppression and aplastic anemia [7,10,11]. The use of chloramphenicol in poultry, aquatic and other animals for food production was banned by the United States, European Union, Canada and China [12]. Therefore, the development of sensitive and rapid methods for monitoring of chloramphenicol in food samples is crucial to maintaining food safety. Reports in the literature describe the electrochemical method of detecting low levels of chloramphenicol to 15 nM. Detection is possible through a combination of molybdenum sulfide and multiwalled carbon nanotubes (MWCNTs) - FIG. 1. Detection of this substance is possible in real samples such as milk, powdered milk or honey. The advantages of this method are: simple and ecological process of preparation, fast analysis time, good reproducibility. Negatively charged multiwalled carbon nanotubes are mixed with particles of molybdenum sulfide, forming a grid on the 3D structure. The hierarchical structure of the 3D nanocomposite constructed of molybdenum sulfide and functionalized multiwalled carbon nanotubes (f-MWCNTs) significantly increases synergistic affinity for chloramphenicol. The sensor operates in a wide linear range from 0.08 to 1392 IM. The detection limit is 0.015 IM ±0.003 and exceeds the limits of detection of the previously modified electrodes. Scientists want to target their work towards miniaturization of electrodes for the rapid detection of chloramphenicol in samples on the spot. Combination of nanocomposite molybdenum sulfide and multiwalled carbon nanotubes is highly promising in the analysis of food safety, medicines and dietary supplements [7].

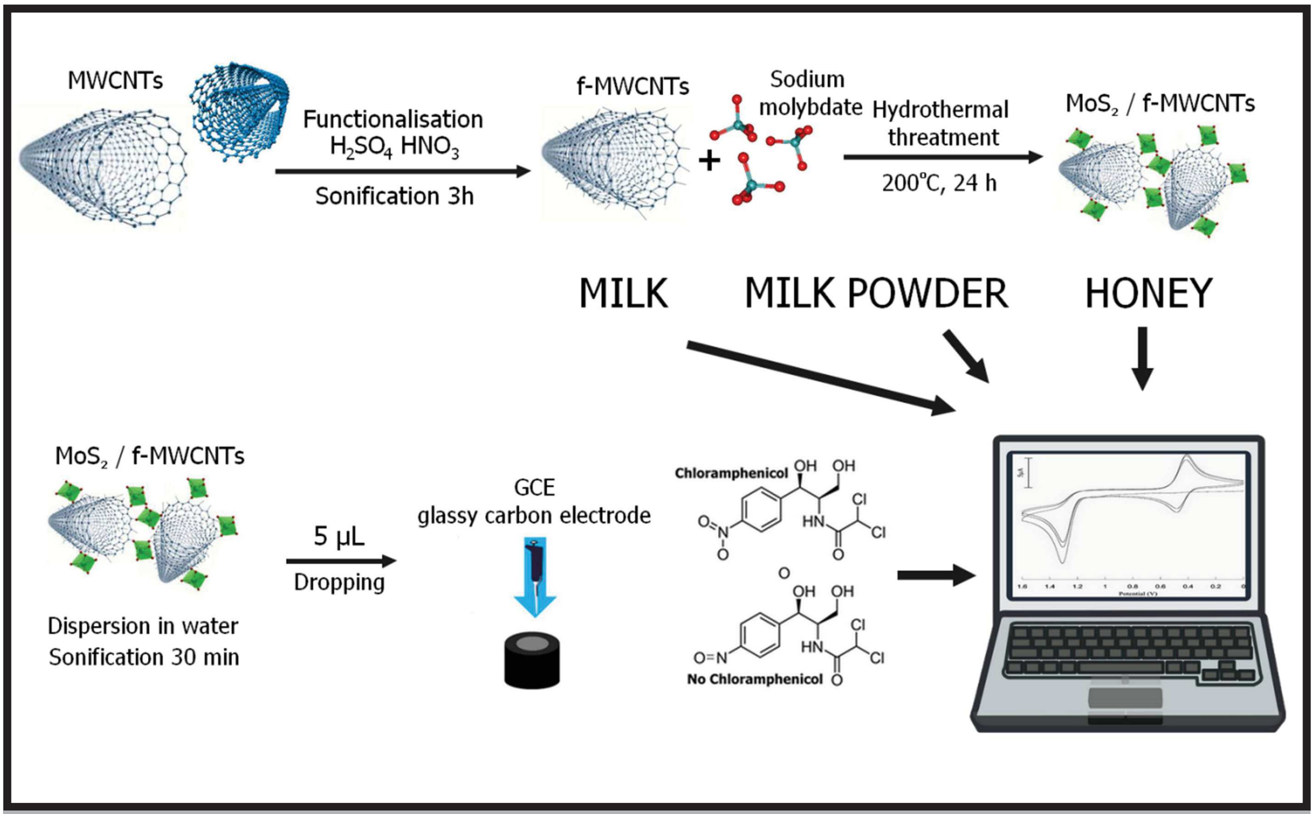

FIG. 1. Determination of chloramphenicol using nanocomposite molybdenum sulfide/multiwalled carbon nanotubes MoS_2/f-MWCNTs, food, and biological and pharmaceutical samples. Developed on the basis of [7].

In the case of food, besides microbiological control it is very important to monitor false, toxic food dyes. This is important because of their potential toxicity and virulence. A hybrid composite, which can be regarded as a sensor for rapid detection of toxic dyes such as metanil yellow, auramine, orange II and aura red has been developed. The hybrid nanocomposite was made with onions nanocarbon-polyoxometalate. The composite allows the detection of trace amounts of toxic dyes used in food, beverages, syrups, and drugs. The water-soluble polyoxometalates belong to the family of anionic metals inorganic oxide, a complex that can be synthesized by simple chemical processes with water [13,14]. This connection allows to identify the toxic chemicals in the smallest amount and can be used in detecting trace amount of toxic dyes (Auramin O and Orange II) used in food. Auramin O is a carcinogen which damages human eye and causes DNA damage. Orange II is toxic azo dye, commonly used in organic light emitting and it affects blood cell. Metanil yellow is used to check the behavior of the composite (FIG. 2). The observation and study of the interaction of carbon nanoparticles with biological material and chemicals gives great possibilities of limiting the amount of toxic substances in food. Hybrid of carbon nano-onion with polyoxometalate nanoparticles may combine the properties of two ideal functional nanomaterials to get a wide range of applications which will accelerate the development of nanoscience and nanotechnology.

The lanthanide polyoxometalate / carbon nano-onion composites have fluorescence properties. In the presence of 1.71×10^{-5} lanthanide–polyoxometalate cluster with different concentration of aqueous solution of metanil yellow, no change in intensity of yellow color was observed; on the other hand in the presence of 3.43×10^{-6} mol/ml^{-1} carbon nano-onion, there was a change in intensity but that was in the micro range (1.03×10^{-2} µmol /ml^{-1}) [14]. FIG. 3 shows change in fluorescence intensity based on varied concentration of the food color with fixed concentration of $[Na_{10}(PrW_{10}O_{36})]_2$-$130H_2O$/CNO nanocomposite [14].

Polydiacetylene (PDA) is a self-assembled polymer with a closely packed and well-aligned conjugated backbone [15-17]. Polydiacetylene monomers in aqueous solution form nano-sized vesicles. To assure specificity to the target analyte vesicle polydiacetylene surfaces are functionalized using a specific probe [17,18]. After binding polydiacetylene vesicle to the target analyte change in color from blue to red due to the physical stress induced by the interaction between the immobilized probe and the analyte is observed. Color of polydiacetylene vesicle indicates the presence of the target analyte. Its concentration can be calculated by determining the degree of color transition [17,19]. The sensors are attractive because the detection of the target analyte is simple. However, the disadvantage is that color change of vesicle polydiacetylene is not enough if the concentration of analyte is too low. It is thus possible to detect the analyte if its concentration is high [17].

Silver nanoparticles formed on the surface of graphene by reduction, can be used to detect different dyes. Experimental results indicate that the silver nanoparticles with graphene can identify different colorants, due to the strong interactions between graphene and dye adsorbed [17].

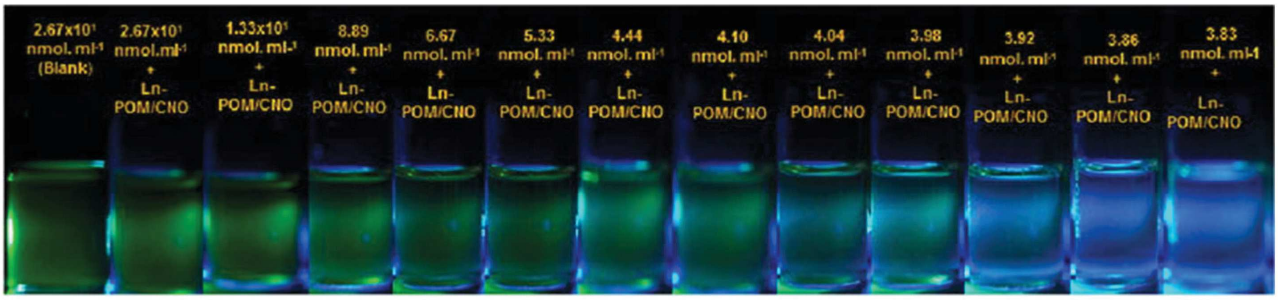

FIG. 2. Changes in fluorescence of different concentration of metanil yellow in 3.43 × 10^{-6} mol/ml^{-1} aqueous solution of the Ln–POM/CNO nanocomposite (lanthanide polyoxometalate/carbon nano-onion). Reprinted with Creative Commons Attribution 4.0 International License permission from [14].

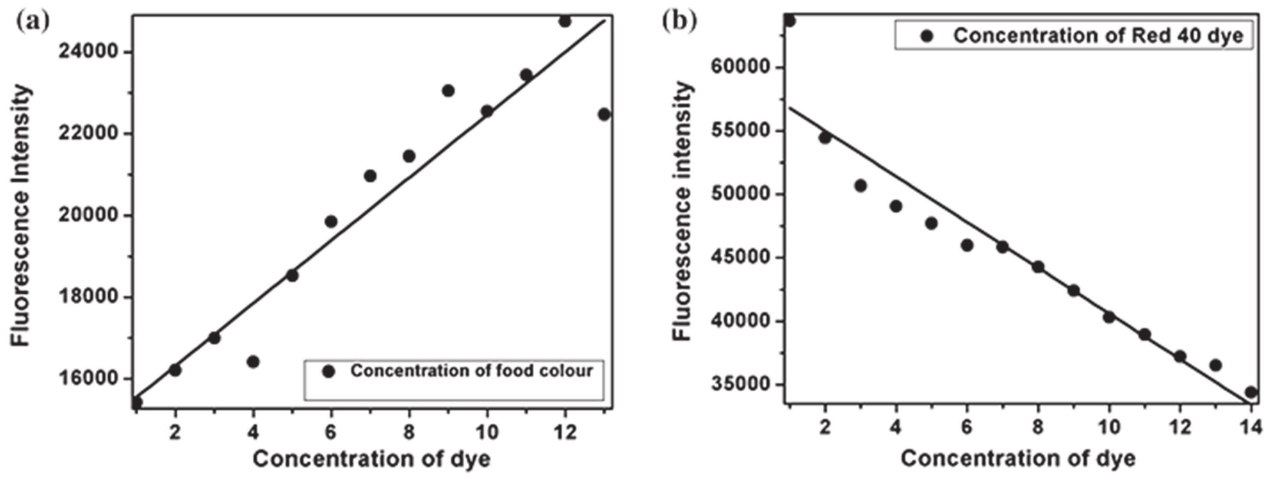

FIG. 3. a) Change in fluorescence intensity of metanil yellow dye (similar behavior for auramine O and orange II food color, data not shown); b) Change in fluorescence intensity for red 40 dye. Reprinted with Creative Commons Attribution 4.0 International License permission from [14].

Nanoparticles in materials that come into contact with food, dietary supplements and biologically active substances

Melamine (1,3,5-triazine - 2,4,6-triamine) is an industrial chemical often used in the production of melamine formaldehyde resins and plastics for coating, commercial filters or laminates. It is a chemical industrial raw material used in the food technology [21,22]. Some of the products such as melamine tableware were used in the catering industry and have become an important material for food contact. Melamine has a high level of nitrogen by weight - 66%, is of low cost, and began to be illegally used in food, in particular milk products, for the creation of the apparent protein content [22,23]. Melamine itself has low toxicity, but in the body it tends to form insoluble melamine crystals in the kidneys, can cause damage of the kidneys and the urinary tract, and even can lead to death [22,24]. Melamine can migrate in food and cause health risks [22,23]. To ensure food safety and protect human health the migration of monomers from a material that is in contact with food has been strictly regulated in many countries.

Based on the European Union regulation on plastics intended to come into contact with foodstuffs (European Union 10/2011), melamine is subject to specific migration limit of 2.5 mg·kg^{-1}. In view of the proven toxicity of melamine United States Food and Drug Administration (FDA) has established safety limit intake of this substance in an amount of 2.5 ng mL^{-1} for adults, and 1 g of food mL^{-1} of an infant formula [22,25].

Literature describes various methods for determination of melamine such as capillary electrophoresis, high performance liquid chromatography, gas chromatography mass spectrometry, fluorescence, colorimetric method, surface enhanced Raman scattering, electrochemical techniques [22,26-33].

Graphene is a single-layer and two-dimensional material whose properties like rapid electronic transfer and high surface area make it an ideal material for electrochemistry [22,34].

For designing of chemical sensors functionalization of graphene with metallic nanoparticles such as gold, platinum or palladium is used. In particular promising application is to create nanocomposites composed of graphene nanoparticles and gold. Such a combination has been used as the matrix electrodes for amperometric detection or determination of dopamine hydroquinone and catechol [22,35,36]. Gold nanoparticles/reduced graphene oxide (rGO) nanocomposites were synthesized by in situ growth Au nanoparticles on the surface of graphene oxide in the presence of sodium citrate and then reduced by hydrazine.

The obtained Au nanoparticles/reduced graphene oxide nanocomposites were modified on glassy carbon electrode for melamine measurement using hexacyanoferrate as electrochemical reporter. Melamine can be grafted on the surface of the Au nanoparticles by the interaction between the amino groups of the melamine and Au nanoparticles via Au-N bond, which leads to suppression of the peak current of hexacyanoferrate due to poor electrochemical activity of melamine. The degree of suppression is related to the concentration of melamine and can be used for the quantitative determination. In this electrochemical detection system graphene oxide provides a platform for uniform distribution of Au nanoparticles and increases the rate of electron transfer and the sensitivity of this method is higher.

Mechanisms for recognition of nanomaterials by biologically functionalized bacteria that may be relevant in the pharmaceutical industry

The basis of nano biorecognition is a coupling of biomolecular nanomaterials. It is assumed that each nanoparticle having a diameter of about 100 nm can efficiently conjugate 150-200 antibody molecules resulting in more than 300 active sites [37]. Interaction of biomolecules with nanoparticles allows creation of contacts between nanomaterials and target cells (FIG. 4). Functionalized nanomaterials are characterized by higher binding affinity than the free molecule [38]. It was shown that the affinity of the binding constant of an antibody-nanoparticle was eight times higher than the affinity of the free antibody [39]. There is a variety of surface modification strategies of nanomaterials. Generally they can be divided into direct and indirect strategies. In the case of direct strategies, biological molecules can be combined with nanoparticles by physical adsorption or covalent binding. It is recognized that the hydrophobic and electrostatic interactions are the most likely mechanisms involved in the adsorption of proteins. The surface of nanomaterials can be modified by covalent bonding of functional groups: sulfide, amine and carboxyl [40]. In the indirect method biomolecules engage nano-particles by the bridges characterized by high affinity to each other. An example might be the interaction of biotin and avidin. Nanomaterials covered with avidin may interact with biotinylated molecules, and the process is based on the strong affinity of advidin to biotin. Modified antibiotics are also used in the selective isolation of pathogenic Gram-positive bacteria [39]. Vancomycin – a glycopeptide antibiotic – is used to identify Gram-positive bacteria by binding to a peptide (D-Ala-D-Ala) on the cell wall by hydrogen bonding [41,42].

Above interactions can be detected by electrochemical impendance [43], fluorescent microscopy [44-46], immunoassay [47-49] and confocal microscopy [50,39]. This type of research shows how chemical groups can interact and what affects the most important interactions of biological material that in the future can be very important in the development of the food industry, medicines and dietary supplements [51].

Reports in the literature describe the effect of resveratrol in the core of biopolymeric nanoparticles and its impact on the antioxiant and antitumor properties. Resveratrol is a naturally occurring polyphenolic phytoalexin produced by a number of different plants, such as grape, berry, mulberry, cranberry, peanuts [52,53]. Recently the positive impact of resveratrol on human health, such as an antioxidant, was highlighted [54].

Despite such benefits it is very difficult to use resveratrol in pharmacy as a supplement or as a functional food product due to its weak solubility in water and chemical instability and low bioavailability [53,55,56].

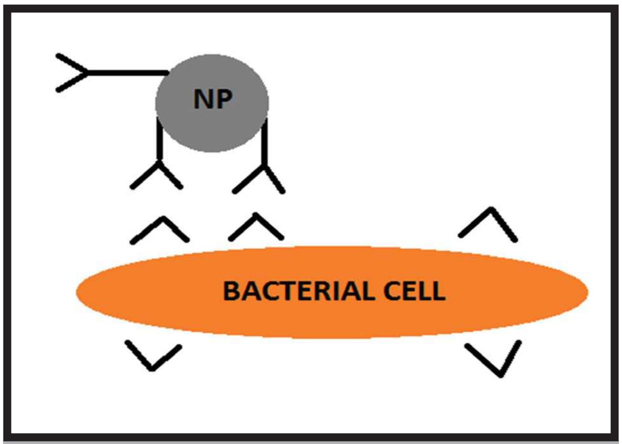

FIG. 4. Nanomaterials coated with an antibody conjugated to a bacterial cell. Developed on the basis of [39].

The interaction of polymer nanoparticles with cell membrane

Knowledge of the influence of the structure of nanoparticles and their interaction with cell membranes is important for understanding the effects of nano-toxicity to human and animal health and the environment and to optimize formation of nanoparticles for biomedical applications. Reports in the literature describe the interaction of the nanoparticles with the polymer of 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine lipid cell membranes [57]. In addition, alignment of chain segments from the polymers with that of hydrocarbon chains in the interior of the membrane facilitates the complete immersion of the nanoparticles into the cell membrane. These results highlight the importance of knowledge of the topology of the surface and structure of the polymer molecules that influence adsorption on the membrane and subsequently, induce the possible transport into the cell [57]. Effect of nanoparticles on a cell is significant, because the penetration into the cell may result in toxic effects. This can be done by endocytosis, a direct diffusion or breaking of the membrane. It is very important to construct nanoparticles with a surface morphology that does not evoke harmful effects on living organisms and at the same time would fulfil their function. Evaluation of toxicity of nanomaterials is still difficult to determine in living organisms due to the limitations of experimental techniques [57,58]. There are several factors that affect adhesion of nanoparticles to the cell membrane such as particle size, surface structure and chemical composition. Polyethylene and polystyrene are produced in huge quantities annually and are used in abundance in industry, as well as found in the environment. Simulations were based on a coarse-grain models in order to examine the trend how polystyrene nanoparticles penetrate cell membranes. The polymer particles smaller than the thickness of the membrane can more easily penetrate to hydrocarbon interior of the lipid bilayers [57,59].

Some studies have shown that the absorption of polystyrene nanoparticles inside the cell membrane might change its mechanical and structural properties. It has been shown that irreversible adhesion can be initiated by introducing the free side groups of the hydrophobic polymer in the interior of the membrane [57].

In addition, the side groups, as well as the nature of chain entanglements, can also influence the interaction with the membrane and subsequent uptake of the nanoparticles [57]. The cell membrane 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine is a phospholipid and composed of a polar hydrophilic phosphate heads and two hydrophobic hydrocarbon tails. The plasma membrane consists of two layers, wherein the molecules of 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine are arranged so that the interior of the hydrocarbon tails form the membrane and the polar main groups form a surface exposed to an aqueous medium. The polar head groups are highly soluble and can prevent polymer nanoparticles being absorbed into the more favorable hydrophobic region. However, the dangling end of the chain can readily pass through the hydrophilic barrier and be irreversibly adsorbed in contact with the upper part of the membrane of hydrocarbon chain of 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine. Cell membranes comprise other molecules such as sterols and proteins having different functions in the membrane. Presumably presence of all of these biomolecules can play the role in influencing the interaction of the nanoparticles with cell membranes [57].

Carbon nanotubes and polymers

Surfactants with amphiphilic nature are extremely attractive for nanotube dispersions. There are two mechanisms of nanotube dispersion in polymers: non-wrapping and wrapping. These two mechanisms differ in the strength of adsorption between the nanotube and the polymer. Wrapping occurs when a strong single polymer layer helically wraps nanotube [60].This is considered a very strong interaction, because it affects the electrical properties of nanotubes. In non-wrapping interaction polymers - nanotube is low due to van der Waals forces. Electrical properties are not changed in the latter case. Atomic force microscopy (AFM) observations showed that presence of polymer layers (polyvinylpyrrolidone, polystyrene) on the surface of the nanotube [61].

In 2008 Maity et al. showed that the wrapping is actually possible by the use of poly-N-vinylcarbazole (PNVC) and they produced nanocomposite of single walled carbon nanotubes and multiwalled carbon nanotubes. Monomers of poly-N-vinylcarbazole contain two aromatic rings and give a strong affinity to the surface of the nanotubes in a similar way as surfactants. The authors assessed the interaction between the nanotubes and the polymer with a Raman spectroscopy [62].

In 2003, Zheng et al. found that polythymine (T) wrapping was an enthalpically driven spontaneous process with energies favoring the interaction of polymer-nanotube instead of nanotube-nanotube binding. Using modeling, the adsorption mechanism was again thought to originate from π-π interactions between the nanotube and the nucleic acid, which was further promoted by the extreme solubility of the phosphate backbone in the aqueous solution [63]. In 2005 Dror et al. described two different polymers: Gum Arabic (GA) and alternating copolymer of styrene and sodium maleate (PSSty) to disperse nanotubes. In addition to both being amphiphilic and charged, the latter provides electrostatic repulsion, the polymers differed in two ways. GA is a highly branched polysaccharide while PSSty is a linear copolymer of alternating hydrophobic and hydrophilic units [64]. In its aggregated state, nanotubes are not as electrically or thermally conductive and cannot provide mechanical support due to low percolation. To solve this problem, there are two chemical approaches to modifying nanotubes to make them more homogeneously dispersed in solution: covalent and non-covalent modifications. Covalent modifications involve attaching different functional groups to the surface of nanotubes, however, these processes typically involve harsh treatments with acids as the initial step can lead to destruction of the nanotube's structure and therefore deteriorate nanotube's properties [60]. Non-covalent modifications include the use of amphiphatic molecules such as surfactants or polymers for coating the nanotubes, which stabilize the environment around them. This strategy allows to keep many important properties of nanotubes. To this end, various types of surfactants (anionic, cationic, ionic), and the polymers and their ability to spread have been tested. Recently, researchers attempt to combine these two types of molecules together in a dispersion of nanotubes [60].

Modification of the surface of carbon nanotubes and their interactions with organic pollutants

Carbon nanoparticles have unique properties and potential for different applications. Surface properties of carbon nanoparticles affect the interaction of organic pollutants. The decreased diameter of carbon nanotubes results in increased surface area and leads to enhanced adsorption of pyrene, ofloxacin (OFL) and norfloxacin (NOR) [60,65,66].

Availability of surface to absorb contaminants on single walled carbon nanotubes could be higher than that of activated carbon with the relative effect of blocking the pores. For flat structural molecules such as benzene, flat surface makes more contact with the carbon nanotubes, so adsorption is improved with increasing diameter of nanotubes [60,67]. Adsorption is also influenced by other factors such as molecular structure, functional groups and morphology of carbon nanoparticles. Functional groups such as -OH, -COOH, and -C=O from carbon nanoparticles may be intentionally created by the method of oxidation [60,68].

The functional groups of the carbon nanoparticles can interact with water by hydrogen bonding leading to the formation of water clusters, which reduce the availability of carbon nanoparticles to interact with the surface of the organic impurities [60,69]. Increased interaction leads to the formation of bonds between functionalized carbon nanoparticles and organic polutants. Multiwalled carbon nanotubes with -OH groups and multiwalled carbon nanotubes with -COOH groups may form a hydrogen bond with the 2-phenyphenol and enhance its adsorption [60,70]. The increase in adsorption of organic impurities is affected by the grafting of functional groups on the surface of carbon nanotubes. The adsorption capacity can be increased by adsorption of β-cyclodextrin on the surface of the carbon nanotubes. Such carbon nanotubes have a higher affinity for Pb (II) and 1-naphthol because hydroxyl groups and internal hydrophobic core in the cavity β-cyclodextrin can form complexes with metal ions and organic contaminants [60,71]. The main interactions between organic contaminants and carbon nanoparticles are hydrophobic, electrostatic, hydrogen bond, and π-π interactions. These interactions and their strength are influenced by the surface properties and morphology of carbon nanoparticles and the molecular size, structures, and functional groups of organic contaminants. For a given carbon nanoparticles, various mechanisms may simultaneously control the sorption progress of organic contaminants on carbon nanoparticles, while the sorption controlling mechanisms may depend on different environmental conditions. Effects and comparison of adsorption of various organic pollutants on the carbon nanoparticles may provide important information on the relationship and the different mechanisms of action. Research should be directed toward the assessment of the dispersibility of carbon nanoparticles in a variety of environmental conditions on the characteristics of sorption of organic pollutants [60].

Promising intelligent packagings in the food and pharmaceutical industries

The biopharmaceutical industry is developing rapidly, but every innovation must be safe. Many companies are working to develop packages that will warn of contamination of packaged food and respond to changes in environmental conditions. There are examples of nanotechnology application in the food, medical and pharmaceutical industries; this is realised by the use of nanocapsules with flavor enhancers and the nanoparticles having the ability to bind and remove chemicals from the food [72]. Intelligent packaging is a promising application of nanotechnology innovation to develop antimicrobial packaging applicable in the medical and pharmaceutical industries. Intelligent packaging can response to the environmental conditions or warn the consumer about the contamination and the presence of pathogens. Researchers are trying to develop a package that could in time evolve preservatives to extend the shelf-life of the food product. These solutions are most exciting innovation in the food industry worldwide. Nanocrystals embedded in the plastic material help to form a molecular barrier to prevent oxygen transport. The current technique allows to preserve the freshness of the beer for 6 months and some companies are working on extending the freshness of it by using this technology for 18 months. Coatings with nanoparticles could create a kind of sensor to detect pathogens in food. Such a sensor would detect the presence of pathogens by changing the color of the packaging to alert consumers that food has become contaminated or food began to spoil. Nanosize natural biopolymers such as polysaccharides can be used to encapsulate vitamins, prebiotics and probiotics. In the food industry one of the major problems is time-consuming and laborious process of food quality control analysis. Innovative devices and techniques can be developed to facilitate the preparation of food samples for analysis. From this point of view, the development of nanosensors for the detection of microorganisms and impurities can be used in food and pharmaceutical industry [72].

Conclusion

Nanotechnology and nanosciences have great potential for use in the food, chemical, medical and pharmaceutical industries. This gives an opportunity to raise awareness of the mechanisms of action of nanoparticles with food ingredients and drugs, as well as packaging of food and dietary supplements. Nanoparticles as active packaging components can be used for antimicrobial agents encapsulation, adsorption and chemical conjugation.

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