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APPLICATIONS OF NANOFORMULATIONS IN AGRICULTURAL PRODUCTION AND THEIR IMPACT ON FOOD AND HUMAN HEALTH

ZASTOSOWANIA NANOZWIĄZKÓW W PRODUKCJI ROLNEJ I ICH WPŁYW NA ŻYWNOŚĆ I ZDROWIE CZŁOWIEKA

Abstract: In last years, an increasingly growing number of nanoagrochemicals such as nanopesticides, nanofertilizers or plant growth stimulating nanosystems is put into the practice. This is connected with the demand to decrease the dose-dependent toxicity of pesticides by reduction of the amount of applied active ingredients in nanoscale formulations, securing not only their increased aqueous solubility resulting in better bioavailability but also targeted delivery, controlled release and/or protection against degradation. Application of nanofertilizers enables more effective supply of nutrient to plants. The contribution is focused on nanoformulations applied in agriculture enabling controlled release of the active ingredient into weeds and the body of pests and nutrients to plants as well as on benefits of the use of nanoformulations in food industry related to food packaging, food security, encapsulation of nutrients and development of new functional products. The environmental impact of nanoagrochemicals and related health risks are highlighted as well.

Keywords: nanoinsecticides, nanoherbicides, nanofungicides, controlled release and targeted delivery, environmental impact, health risks

Introduction

Nanotechnology, one of the key technologies of the 21st century, can improve current agricultural practices through the enhancement of management and conservation of inputs in crops, animal production and fisheries [1, 2] and to secure "sustainable intensification" of agricultural production resulting in increasing global food production, improved food quality and reduction of waste [3, 4].

Formulations containing nanoparticles (NPs) or encapsulated nanoscale materials with various coatings are suitable for improving the solubility and permeability, *ie* the bioavailability of an active ingredient (AI), reducing the dose of the AI and its dose-dependent toxicity, securing controlled release and targeted biodistribution of the AI and alleviating its impact on the environment [5-7].

Nanoscale herbicides, insecticides and fungicides can help to attenuate environmental pollution, loss of biodiversity and emergence of agricultural pests and pathogens. Moreover, the use of NPs in food sector can contribute to improvement of food safety by pathogen detection, design of smart food packaging systems and enrichment of food with nutraceuticals [2, 8, 9]. The increasingly widespread use of NPs in various areas of anthropogenic activities requires a thorough investigation of their environmental impacts and possible negative effects on human health.

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Nanopesticides

Nanoscale herbicides, insecticides and fungicides were designed for better protection of plants from damaging influences such as weeds, insects or plant diseases in comparison with the application of bulk active ingredients. Methods of preparation of NPs and nanoformulations were summarized in [10]. Encapsulation of pesticides in polymeric core shells of NPs can result in safer and more convenient management of pesticides that promises environmental safety [11]. The control of parasitic weeds with nanoencapsulated herbicides was found to reduce the phytotoxicity of herbicides on crops [12]. The application of nanoherbicides is connected with a reduced environmental impact with simultaneous preservation of herbicidal effectiveness. Foliar application of nanoemulsion systems can be used to increase penetration and uptake of an active ingredient by weeds. Nanoformulations of a herbicide with polymer coating enable its controlled and slower release due to potential interactions between the herbicide and the polymer. CuO NPs were found to be more toxic than bulk CuO or dissolved Cu(II) ions to *Landoltia* (*Spirodela*) *punctata* [13] and *Zea mays* [14]; the activity of antioxidant enzymes increased after the treatment of *Elodea densa* plants with CuNPs [15]; CuO NPs induced also DNA damage in several plants.

Problems connected with plant fungal diseases can be alleviated by design and preparation of effective nanofungicides that can control fungal diseases by specifically inhibiting or killing the fungi that cause the diseases. Several metals (*eg*, Ag, Cu) or metal oxides (*eg*, CuO, ZnO) NPs were found to be effective fungicides. Well-dispersed and stabilized AgNPs solution can act as an excellent fungicide due to good adhesion on bacterial and fungal cell surface [16], and biosynthesized AgNP-based biopesticides can be used in the future as nanoweapon against phytopathogens [17]. Also the antifungal activity of CuNPs is connected mainly with NPs adhesion to the bacterial cell surface because of their opposite electrical charges, resulting in a reduction reaction at the bacterial cell wall [18]. Hexaconazole NPs (100 nm) stabilized by polyethylene glycol (PEG) were more potent than bulk hexaconazole and were found to be a safe nanofungicide [19]. The controlled release of carbendazim from nanoformulations of PEG-based functionalized amphiphilic copolymers was observed, and the efficacy of these nanoformulations against plant pathogenic fungi *Rhizoctonia solani* expressed by ED_{50} values varied from 0.40 to 0.74 mg/dm^3 [20]. However, in the case of application on nanofungicide formulations in field, possible interactions of nanofungicides with non-target organisms affecting directly or indirectly the maintenance of soil fertility could be always considered [21].

Nanoinsecticides are agents of chemical or biological origin that control insects in more effective manner than the bulk insecticides. The insecticidal efficacy of liposome-based formulations was described also by Hwang et al [22] and Kang et al [23], and the chitosan coated nanoliposomes exhibited sustainable slower release of the entrapped core material (etofenprox and alpha-cypermethrin) due to thicker coating layer [24]. Pyridalyl nanosuspension prepared using sodium alginate was 2- and 6-fold more effective as stomach poison against *Helicoverpa armigera* than the technical product and the commercial formulation, respectively [25]. Also some nanosized inorganic materials (*eg* Al_2O_3, Fe(0), TiO_2, ZnO, Ag, SiO_2) were found to be good insecticides (in detail see in [10]).

A comprehensive survey on nanoherbicide, nanofungicide and nanoinsecticide formulations and their biological effects was published by Jampilek and Kralova [10].

Nanofertilizers

Fertilizers play a pivotal role in increasing the agricultural production by up to 35-40%. Nanofertilizers represent an efficient means to distribute fertilizers in a controlled fashion with high site specificity, thus reducing collateral damage. They can reduce the dosage of nutrient, ensure its controlled slow delivery resulting in the increased efficacy of the fertilizer and thus overcome the problem of eutrophication. In nanofertilizers, the nutrients can be capsulated inside nanomaterials, coated with thin protective polymer film and delivered as particles or emulsions of nanoscale dimensions [26]. Fertilizers formulations of chitosan NPs for controlled release of NPK [27, 28], hydroxyapatite NPs for the supply of P nutrients to crops [29] and fertilizer loaded nanoclay/superabsorbent polymer composites [30] were used. Foliar application of nano-potassium and nano-calcium chelated fertilizers on *Ocimum basilicum* had beneficial effect on production characteristics of plants [31]; calcium phosphate nano gel fertilizer composites were found to increase the germination in *Oryza sativa*, *Arachis hypogea* and *Amaranthus spinosus* plants [32].

A significant improvement of some production characteristics of *Pennisetum americanum* L. plants due to application of ZnO NPs was observed by Tarafdar et al [33], while Pandey et al [34] reported about the enhanced activity of some enzymes after application of Zn nanofertilizer. Nano-Al_2O_3 enhanced root elongation of *Arabidopsis thaliana* [35] and *Lemna minor* [36] and increased the quantum yield of photosystem II [37]. Production characteristics of plants significantly increased also after the application of iron fertilizers [38, 39]. Nano-iron oxide was also found to facilitate the photosynthate and iron transferring to the leaves of peanut [40]. The single-walled carbon nanotubes (SWCNTs) were found to activate seed germination and enhance growth of different organs of several plants [41]. A beneficial effect on the growth of plants after application of multi-walled carbon nanotubes (MWCNTs) was observed as well [42-44]. Using nanotechnology, intelligent nanofertilizers can be designed in which extremely thin layer of polymer would be placed over the fertilizer granules and extremely tiny biological sensors situated in this polymer would be able to detect "signals" sent naturally by plant roots, which would essentially tell the polymer when to dissolve and release the nutrients into the soil [45].

Nanotechnology in food industry

Important areas of nanotechnology in food sector are, for example, food safety (through the use of nanosensors for pathogen detection), smart packaging and valorisation of food products by nanoencapsulation/nanodelivery of food ingredients (*eg* flavours) [8, 46]. Nanostructured lipid carriers may improve the bioavailability and stability of bioactive compounds, consumer acceptability, functionality, nutritional value and safety of food systems, prolong shelf-life and provide controlled release of encapsulated materials [47]. Novel functional foods developed using nanotechnology may have physiological benefits or reduce risks of diseases. Vitamin D3 in whey protein isolate NPs was found to be suitable for use in clear or non-clear beverages as enriching agent [48].

The function of packaging is to protect the packed food and to maintain its integrity and quality. The package should hinder gain or loss of moisture, prevent microbial contamination and act as a barrier against transfer of oxygen, carbon dioxide and aromatic compounds. The packaging material itself should not promote deteriorative food quality changes or endanger the health of the consumer of the packed food as a consequence of uncontrolled migration of any chemical substances from packaging into food [49]. The use of NPs in food packaging can improve protection of foods, for example, by reducing permeation of gases, minimizing odour loss and increasing mechanical strength and thermal stability. Nanoscale food packaging materials may extend food life, improve food safety, alert consumers that food is contaminated or spoiled, repair tears in packaging and even release preservatives to extend the life of the food in the package [50]. Intelligent packaging is an emerging technology that uses the communication function of the package to facilitate decision making to achieve the benefits of enhanced food quality and safety [51]. Current nanocomposite technologies suitable to enhance the mechanical and barrier properties of synthetic polymers and biopolymers for food packaging, the development of intelligent packaging with enhanced communication function focusing mainly on oxygen, humidity and freshness indicators and nanostructured coatings that enhance the barrier properties of packaging films were summarized in a review paper of Mihindukulasuriya and Lim [52].

Environmental impact of nanoparticles

Most hazardous chemicals applied in agriculture exert unwarranted toxicity and lethal effects on non-target organisms, develop physiological resistance in target and cause adverse environmental effect. By the use of green and efficient alternatives for the management of insect pests in agriculture, toxic effects on the environment could be alleviated [53, 54]. As favourable could be considered formulations that target the pests specifically and in turn, prevent pollution, for example, the “gut buster” (*ie* the encapsulated product) breaks open only when it comes in contact with the alkaline environment like the gut of the insects [55]. The application of such smart pesticides results in more precise, controlled and effective use of pesticides enabling potential reduction of the overall quantities of applied pesticides [56].

On the other hand, some NPs, for example, AgNPs, are toxic because they can destroy bacteria and other microorganisms, and their entry in the environment represents a risk that useful bacteria and aquatic organisms will be destroyed as well [57]. Consequently, although AgNP-based biopesticides could revolutionize the agricultural sector in the future, increased attention must be given to the impact of risk factors associated with their usage on the environment.

The toxicity of NPs depends on the shape, size, surface area and surface charge of NPs as well as on several abiotic factors such as pH, ionic strength, water hardness, presence of organic matter. The most frequent mechanisms of toxic effects exhibited by NPs are damage of membranes, generation of reactive oxygen species (ROS) and genotoxicity. Increased attention must be devoted to toxicological investigation of non-biodegradable materials due to risks connected with their accumulation and persistence in soil, plants and mammals, which may subsequently result in various pathological processes [58, 59]. The most important regulations related to the application of NPs especially in relation to nature, environment and health are discussed in [10].

Health risks of nanoparticles

NPs can enter the human body in several ways: (i) via the lungs where a rapid translocation through the blood stream to vital organs is possible, including crossing the blood brain barrier; (ii) absorption by the intestinal tract; and (iii) absorption by the skin [60]. Toxic effects of nanomaterials are closely connected with physicochemical properties of NPs such as particle size and size distribution, agglomeration state, shape, crystal structure, chemical composition, surface area, surface chemistry, surface charge, and porosity [61].

Small sized (1-100 nm) NPs derived from Ag, Cu, Al, Si, carbon or metal oxides were found to easily cross the blood-brain barrier and/or produce damage to the barrier integrity by altering endothelial cell membrane permeability [62]. According to Cho et al [63], metal oxide NPs induce unique inflammatory footprints in the lung with a subsequent increase of the risk of asthma attacks and caused DNA damage in the human lung epithelial cell line A549 [64, 65]. A comprehensive critical review paper about applications of AgNPs and human health was presented by Ahamed et al [66]. Disruption of the mitochondrial respiratory chain by AgNPs resulting in increased ROS production and interruption of ATP synthesis and leading to DNA damage was observed [67]. AgNPs exhibited also geno- and cytotoxic effects in human mesenchymal stem cells (hMSCs) at high exposure concentrations [68] and reduced the cell viability of alveolar macrophages and lung epithelial cells [69].

Zero-valent iron NPs induced oxidative stress resulting in the damage of lung cells [70]. After exposure to SWCNTs, pulmonary granulomas were observed in rats [71], and functionalization of SWCNTs played an important role in their cytotoxicity [72]. Dermal exposure to unrefined SWCNTs led to dermal toxicity due to accelerated oxidative stress in the skin of exposed workers [73]. On the other hand, the diameter of MWCNTs was found to be a critical factor for inflammogenicity and the subsequent mesothelial carcinogenesis [74, 75]. Oxidized MWCNTs were found to induce massive loss of the viability of human T cells through programmed cell death at doses of 400 ng/dm^3, which corresponds to approximately 10 million carbon nanotubes per cell [76].

Conclusion

The application of nanoscale science and nanotechnology in agricultural and food industry/production demonstrated to have a great potential in providing various innovations and improved solutions. In this contribution, the most important and frequent applications of nanotechnology in agricultural and food production are summarized. NPs and/or controlled release and targeted delivery nanoformulations are broadly used for agrochemicals (*eg*, nanopesticides, nanofertilizers) and were primarily designed to reduce the amount of applied active ingredients by means of their enhanced bioavailability and protection against degradation, which finally resulted in a decrease of dose-dependent toxicity for non-target organisms and environmental burden. The application of nanotechnology in the areas such as food packaging, food security, detection of pathogens and contaminants by using nanosensors and indicators, encapsulation of nutrients and development of new functional products is growing rapidly. Nanoscale food packaging materials help to extend food life. Nano-size materials change their physical and chemical properties in comparison with bulk materials and can become toxic when reaching nano-size. Therefore increased attention must be devoted to the impact of risk factors associated with their usage on the environment and possible adverse effects on non-target organisms and mammals, especially humans.

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ZASTOSOWANIA NANOZWIĄZKÓW W PRODUKCJI ROLNEJ I ICH WPŁYW NA ŻYWNOŚĆ I ZDROWIE CZŁOWIEKA

Abstrakt: W ostatnich latach coraz bardziej rośnie liczba nanochemikaliów stosowanych w rolnictwie, takich jak: nanopestycydy, nanonawozy lub stymulujące wzrost roślin nanosystemy. Jest to związane z potrzebą ograniczenia, zależnej od dawki, toksyczności pestycydów poprzez zmniejszenie ilości stosowanych składników aktywnych w nanopreparatach, zapewniając nie tylko ich zwiększoną rozpuszczalność w wodzie, co gwarantuje lepszą biodostępność, ale również kontrolowane uwalnianie i/lub ochronę przed degradacją. Zastosowanie nanonawozów umożliwia bardziej efektywne dostarczanie składników odżywczych roślinom. W pracy przedstawiono nanozwiązki stosowane w rolnictwie, umożliwiające kontrolowane uwalnianie składnika czynnego do chwastów i szkodników oraz składników odżywczych dla roślin, jak również korzyści wynikające z użycia nanozwiązków w przemyśle spożywczym związanym z pakowaniem żywności, bezpieczeństwem żywności, kapsułkowaniem składników odżywczych i rozwojem nowych produktów funkcjonalnych. Omówiono również wpływ nanoagrochemikaliów na środowisko i związane z nimi zagrożenia dla zdrowia.

Słowa kluczowe: nanoinsektycydy, nanoherbicydy, nanofungicydy, kontrolowane uwalnianie i ukierunkowana dostawa, wpływ na środowisko, zagrożenie dla zdrowia