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REVIEW

Nanosilica—from medicine to pest control

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Abstract Nanotechnology is a broad interdisciplinary area of research, development, and industrial activity that has been growing rapidly worldwide for the past decade. More ambitious uses of nanoparticles are bioremediation of contaminated environments, controlled release of fragrances, biocides, and antifungals on textiles. Silica nanocomposites have received much attention because of its thermal degradation behavior and applications in chromatography, medicine, optics, etc. Nanobiotech takes agriculture from the battleground of genetically modified organisms to the brave new world of atomically modified organisms where rice has been modified atomically. Silica has been widely applied in various industries. Application of gold-coated silica has been used in the treatment for benign and malignant tumor. Surface-modified hydrophobic as well as lipophilic nanosilica could be effectively used as novel drugs for treatment of chicken malaria and nuclear polyhedrosis virus (*BmNPV*), a scourge in silkworm industry. Here, the authors attempt to provide a review to explain the impact of nanosilica on basic biology, medicine, agro-nanoproducts, and use of amorphous nanosilica as biopesticide.

Background

Nanotechnology is a broad interdisciplinary area of research, development, and industrial activity comprising of four broad areas such as nanomedicine, nanofabrication, nanometrology, and nanomaterials (NMs)–nanoparticles (NPs). NPs are 100 nm in size and irregularly shaped and can also exist in fused, aggregated, or agglomerated forms. Nanoparticles with biocidal properties can be used for dressings of wounds and deliver drugs through the skin. Successful experiments have been conducted to grow human nerve cells on circuit boards which pave the way for brain implants to help paralyzed people.

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Research and development is now focusing on applications of NMs on human health (drug delivery, imaging, cancer therapeutics), energy (hydrogen storage, improved efficiency), defence (energetic materials, lightweight armor composites), catalysis, agriculture (increased crop yields, secure packaging, chem–bio-detection), and environment (water filtration, reduced air emissions, remediation, chemical and biological sensing).

Efforts are initiated not only towards the health sector but also in the field of agricultural sector (Ulrichs et al. 2006a). A nanotech research initiative in Thailand aims to atomically modify the characteristics of local rice varieties along with the color of the leaves and stems.

Nanosilica

Silica is one of the most abundant materials on earth (Fig. 1). Naturally occurring silicas, such as quarts sand, rocks, and clays, are used as raw materials in the industries. In order to produce industrial silica products, such as silica gel, precipitated silica, silica sol (colloidal silica), and fumed silica, these primary raw materials are chemically treated to produce direct-silica sources, such as sodium silicate, silicon tetrachloride, and alkoxysilane. It has a wide range of applications in microelectronics, optical communications,

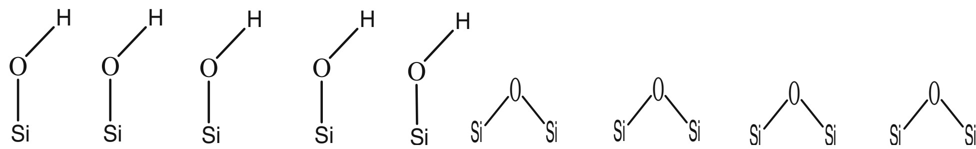

Fig. 1 Hydrophilic (*left*) and hydrophobic (*right*) surface of silica

thin-film technology (Salleo et al. 2003; Che et al. 2003; Suzuki et al. 2004), and industries (Iler 1979). Nanosilica is also used in the pharmacy as a booster agent (Alyushin and Astakhova 1971) and as an enterosorbent (Chuiko 2003). It is well established that the geometries of $(SiO_2)_n$ clusters in the small size range (n>12) are chain like. Thus, these structures consist of some Si atoms that do not have the full fourfold coordination. It is expected that, in a silica nanoshell, dangling bonds exist as they do in SiO_2 clusters and that the sites with dangling bonds may play a role in the binding of Au atoms which in turn acts as a bullet for tumor (Fig. 2; Sun et al. 2004). Amorphous nanosilica can be obtained from various silica sources like shell wall of phytoplankton, different organic sources like epidermis of vegetables, volcanic soil, etc. (Table 1). It can also be obtained from burnt pretreated rice hulls and straw at thermoelectric plants, without the risk of using corrosive substances in the burning process. Transmission electron microscope photographs of nanosilica dispersed in alcohol show uniform particles (Fig. 3, left) of about 20 nm. The “capped” nanoparticle shows strong hydrophobic properties. A scanning electron microscope photograph of the film made of such nanosilica particles is presented on the right of Fig. 3.

Nanosilica for medicine and drug development

The newly emerging area of inorganic particles entrapping biomolecules has already exhibited its diversity and potential applications in many frontiers of modern material science including sensors, biosensors, optical materials, biocatalysts, electrochemistry, immunochemistry, and materials for use in environmental sciences (Avnir et al. 1994). Particulate carriers of colloidal dimensions and below, such as liposomes, polymeric nanoparticles, microemulsion droplets, etc., have already attracted considerable attention as drug carriers for achieving controlled delivery of drugs at specific body sites (Kreuter 1994; Yokoyoma and Okano 1996). Nanoshells have a core of silica and a metallic outer layer and can be injected safely in animal models and preferentially concentrate in cancer lesion sites. Further, nanoshells can carry molecular conjugates to the antigens that are expressed on the cancer cells themselves or in the tumor microenvironment. An exciting new application of gold-coated silica nanoshells has been found in treating benign and malignant tumors (Hirsch et al. 2003a, b; Brongersma 2003). Silica (SiO_2) core of 100 nm, coated with about 20 nm of gold, absorbs near infrared light (NIR) and converts it to heat, causing irreversible thermal cellular destruction. When the nanoshells were incorporated into human breast cancer cells in a test tube and exposed to NIR, 100% of the cancer cells were killed. Besides the applications in cancer, it can also be incorporated into temperature-sensitive hydrogels to synthesize a new type of composite material that collapses on laser irradiation. Nanosilica is characterized by a concentration-dependent bioactivity with respect to different cells (microorganisms, erythrocytes, gamete, etc.) because it possesses highly adhesive properties to a cellular surface and can affect membrane structures. In addition, it can be considered as a promising carrier of organic compounds for development of new drugs in medicine and biotechnology due to its specific properties (Chuiko 2003) and can be used as a component of tourniquet preparations (Chuiko 2003).

Nanosilica A-300 (5−15 nm in diameter) containing approximately two OH per square nanometer per surface

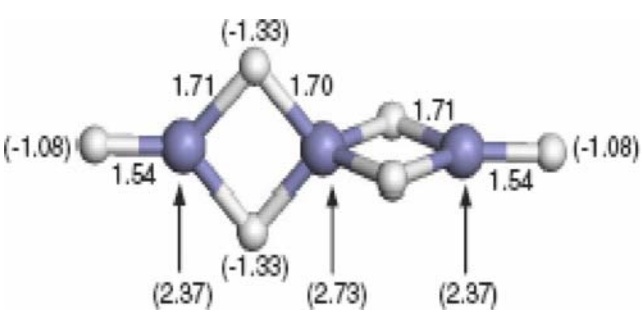

Fig. 2 Geometry and charge distribution of $(SiO_2)_3$. The *numbers in parentheses* refer to charges, while other numbers refer to bond lengths in ampere. The *balls* with *light color* are for O atoms. The *arrows* specify the charges on Si atoms. (Sun et al. 2004)

Table 1 Different polymorphs of silica

Crystalline	Sources	Amorphous
Quartz	Natural	Biogenic origin (diatomaceous earth, rice husks)
Tridymite		
Cristobalite		Volcanic origin (vitreous silica)
Coesite	Artificial	Chemically prepared silicas (precipitated or pyrogenic)
Stishovite		Ground silica glass
Porosils		

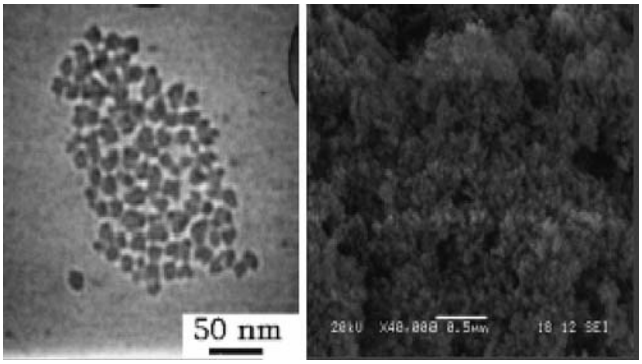

Fig. 3 TEM (*left*) and SEM (*right*) micrographs of nanosilica (Li et al. 2006)

can effectively adsorb high-molecular proteins of synthetic and natural origin (Chuiko 2003). It interacts strongly with red blood cells (RBCs) changing both interfacial water bound by cells (e.g., their membranes) and water structures in the bulk. Nanosilica composed of bioactive particles can affect the RBC state causing strong hemolysis at relatively low concentration of silica $C>0.1$ wt.% (Gerashchenko et al. 1994, 1996, 2002; Diociaiuti et al. 1999; Blitz and Gun'ko 2006). It was found that the shape of RBCs change on interaction with unmodified and modified silicas from discocytes to spherocytes through echinocytes and then to shadow corpuscles depending on concentration. A strong distortion of the membrane (e.g., on interaction with silica nanoparticles) leads to loss of its flexibility and resiliency. RBCs swell and increase in size in comparison with the spherocyte that leads to membrane break. Eliminated hemoglobin can also be detected; i.e., hemolysis of RBCs occurs (Fig. 4). However, the perforated cellular membrane remains uniquely as a whole and forms a so-called shadow corpuscle. It is likely that nanosilica aggregates promote this process. The agglutination of RBCs can be enhanced due to interaction with aggregates of silica nanoparticles which prevent the electrostatic repulsive interaction of negatively charged cells. This is due to strong interaction of silica nanoparticles with proteins integrated into the RBC membranes (Chuiko 2003).

The hemolytic effect of nanosilica can be reduced by modification of immobilized polymers (e.g., poly(vinyl pyrrolidone), poly(vinyl alcohol)) or through the use of binary nano-oxides such as silica–alumina and silica–titania (Blitz and Gun'ko 2006). Control of the hemolytic effect of nanosilica is vital in medicinal preparations (Chuiko 2003). Other types of nanosilica induce proinflammatory signaling in endothelial cells. Researchers in the Rochester group have compared crystalline (Min-U-Sil) silica to amorphous nanosilica. They found that the nanosilica was much less potent than Min-U-Sil silica. Proteins, including lysozyme, horseradish peroxidase, catalase, and trypsin, adsorb strongly to SiO_2 (sizes ranging from 9 to 40 nm) nanoparticles. These proteins undergo a partial loss of structure and generally a significant loss in enzyme activity (Norde and Anusiem 1992; Kondo et al. 1993; Bower et al. 1998; Czeslik and Winter 2001).

Molecular components of subcellular organelles and membranes are highly curved. These curved surfaces may result in the stabilization of proteins, nucleic acids, and other biological macromolecules with significant secondary and tertiary structure. Adsorption of chicken egg lysozyme on nanosilica of various diameters has been studied to see the effect of nanoparticle size on the structure and function of the adsorbed protein molecules. The size of the nanoparticle perhaps influences adsorbed protein structure and function which may be due to the contributions of surface curvature (Alexey et al. 2004). The performance of a dental composite depends on filler type, resin composition, filler–matrix bonding, and cure conditions (Wendt 1987; Pallav et al. 1989; Ferracane et al. 1998; Watts and Hindi 1999; Lim et al. 2002). The degree of polymerization conversion has been increased *via* heat treatment, leading to moderate strength increases (Loza- Herrero et al. 1998). Recently, nanosilica-fused whiskers were incorporated into resins, resulting in strength increases (Xu 2000; Xu et al. 2000). Nanosilica particles were fused onto the whiskers at 800°C. Wear scar depth, diameter, and volume decreased with increasing filler level and improved the composite wear resistance. Novel nanosilica-fused whisker composites were developed with in vitro wear resistance higher than that of conventional glass-particle-filled composites and similar to that of dental amalgam.

Nano—as a catalyst

Due to the small size of the nanoparticles, the proportion of their constituent atoms at the surface will be higher than in their bulk. For example, gold as a bulk has a face-centered cubic structure but nanoscale gold particles of 3–5-nm diameter have icosahedral structure that have more amount of kinks and edges than a cube. This is an important issue

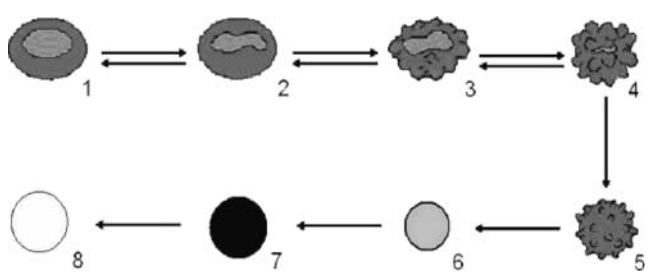

Fig. 4 Scheme of RBC transformation from a discocyte to a shadow corpuscle on interaction with nanosilica: discocyte (*1*), echinocyte I–IV (*2*–*5*), spherocyte (*6*), and shadow corpuscle (*7*–*8*) shown with consideration for their relative sizes (Gun'ko et al. 2007)

as catalyst because it is now known that catalyst primarily happens on steps and edges of the surface where the atoms are comparatively loosely bonded. In fact, it has been observed that nanoscale gold particles have novel catalytic properties (Haruta 2003). Nanoencapsulation of enzymes by inorganic materials for the in vivo use of sustained drug release as well as drug targeting is also expected to have potentiality in enzyme therapeutics (Wiseman 1985). The inorganic particles containing enzymes exhibit excellent storage stability of enzymes (Akbarian et al. 1997). Most of the works reported so far are on the entrapment of enzymes by ceramic materials made of silica gels (Avnir et al. 1994).

Nanosilica—a novel nanobiopesticide

Increasing environmental hazards and enhanced resistance towards insecticides, along with the restrictive use of many chemical pesticides and a limited production of newer, safe chemicals, have prompted active research in biological control. Even today, biodiversity ensures discovery and extraction of new drugs not only for use as human or animal medicines but also as biopesticides for agricultural and domestic use. In the present context, amorphous nanosilica is a promising new venture. Insect pests use a variety of cuticular lipids for protecting their water barrier and thereby prevent death from desiccation. These nanosilicas get absorbed into the cuticular lipids by physiosorption and thereby cause death of insects purely by physical means. Application of nanoparticles on the leaf and stem surface does not alter either photosynthesis or respiration in several groups of horticultural and crop plants. They do not cause alteration of gene expression in insect trachea and are, thus, qualified for approval as the nanobiopesticide in this particular category. Use of amorphous silica is considered to be safe for human by World Health Organization and US Department of Agriculture.

Surface charged modified hydrophobic (Fig. 1.) nanosilica (~3–5 nm) could be successfully used to control a range of agricultural insect pests and animal ectoparasites of veterinary importance (Ulrichs et al. 2005, 2006b, c, d, e, f). Naturally occurring amorphous silica is used by poultry industries and is considered to be safe for human consumption by different regulatory agencies worldwide. Surface-modified hydrophobic as well as lipophilic nanosilica could be effectively used as novel drugs for treatment of chicken malaria. The application of the higher doses of the nanosilica depletes the cholesterol level completely and impedes the process of recovery of the chickens. These particles were used as drugs to mop up the excess amount of the host serum cholesterol lipids which is used by the malarial parasite mainly for their intraerythrocytic growth. The number of schizonts depletes and thereafter the number of trophozoites becomes lesser and as a result total parasite burden is reduced significantly. Nanoparticles are being preferentially harnessed because they offer a greater surface area and circulate more easily and in lepidopteran system (Ulrichs et al. 2006g); they are removed within 24 h from the body. Lawry (2001) surmised that particles significantly smaller than micron order would be less harmful in the hemocoel. Furthermore, Hui-peng et al. (2006) pointed out that lipase treatment, the only viable option for controlling BmNPV, interferes in hormonal balance and cannot be applied to premolting stage. Plant-stem-derived nanosilica is capable of reducing certain classes of lipoproteins (in native form) present in the silkworm larval hemolymph. Here, too, infection by nuclear polyhedrosis virus (*BmNPV*), a scourge in silkworm industry, enhances the level of certain lipids, part of which is reduced by nanosilica.

The chicken and silkworm model for drug development might be used successfully for controlling other insect, animal, and human diseases. In particular, controlling human malarial parasite growth both in vitro and in vivo might be possible (Sharma et al. 1996; Panteghini et al. 2001; Majumder et al. 2006, 2007; Ulrichs et al. 2007). Malaria and other parasites, including virus, often induce an increase in host lipids which the invaders use to their own advantage. This nanosilica treatment might be used together with other arsenals on all sorts of virus which take advantage of enhanced host lipids. Like nanosilica, other nanoparticles should be tested against agricultural, stored grain, household pests. It could be useful for the control of insects, flies, ecto and endo animal parasites, fungal organisms, worms, etc. Amorphous silica gel is another desiccant that pest control operators feel safe to use in homes that have infestations of ticks, bed bugs, and other unwanted creatures hiding in wall voids, attics, cracks, and crevices. The utility of nanosilica is diverse and certainly calls for intense research and application.

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