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DEVELOPMENT OF MODERN NANOTECNOLOGIES AND COMBINED BIOTOXICITY PROBLEMS

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Abstract

Fast progress in modern nanotechnologies based on use of nanoparticles, nanofibers and nanotubes with different composition, shape and size allows elaboration of materials with superhigh strength, thermal and electric conductivity, acoustical and optical properties. Those materials are already widely used in industry, transportation, aerospace, marine and civil engineering, food processing and medicine. Some examples of nanoreinforces composites, superhydrophobic self-cleaning surfaces, nanodyes and suspensions of nanoparticles are described. The problem of uncontrolled accumulation of some types of nanoparticles in our cells and tissues is discussed within the concept of nanotoxicity. Since the history of permanent observation of human health in connection with nanodust accumulation in the atmosphere, waters and soils is not enough long, the detailed evidences must be documented, systematized and discussed.

In this study a brief systematic review of literature on the biotoxicity problems caused by modern nanotechnologies is given. Production of the nanoparticles, nanofibers and nanotubes for industry, transportation, food processing, as well as utilization of the used materials which properties were modified by the nanotechnologies leads to permanent rise of the nanodust in the atmosphere, soils, river waters, lakes and the sea bottom. Their uncontrolled interaction with flora and fauna could be catastrophic for human health and life on the Earth. Promising ways for the problem solution and perspectives are discussed. Some own results on the protective action of nanodiamonds, silver and some other nanoparticles are presented. A vital necessity of an open access database on known types of nanoparticles, their use in the materials and documented influence of health of animals and humans is discussed.

Keywords: biotoxicity, nanotechnology, microparticles, nanoparticles, carbon nanotubes, nanodiamonds, pollutions, nanodust, cell damage, tissue damage.

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1. Introduction

Nanotechnologies have been significantly developed during the last decade and penetrated into all the fields of modern industry, military and civil life [1]. Many practical implementations use additions or surface coatings with nanoparticles (NP) of ~3–500 nm that can easily enter human cells and tissues via skin, eyes, lungs and with food [2]. The nanotoxicity problem connected with fast progress in novel nanotechnologies has been thoroughly studied [3, 4].

Carbon nanotubes (CNTs) have been widely accepted for reinforcement of composite materials for aerospace engineering and building industry, energy and CO_2 storage, acoustic cloaking and many other purposes [5]. Addition of NPs improves vehicle fuel efficiency and corrosion resistance. The nanocomposite materials for coatings make the cars lighter, stronger, and more chemically resistant than usual metal units. CNTs make sports equipment lighter and stronger. The tennis rackets produced with addition of CNTs bends less during impact, that increases the force and accuracy of the ball flight. The tennis balls treated with NPs can keep bouncing twice as long compared to standard tennis balls. NPs and nanofibers woven in tissues can improve their water and stain resistance, as well as flame resistance. Such tissues have almost the same thickness, stiffness and weight.

Nanostructured nature-inspired self-cleaning surfaces can remove water droplets, dust and bacteria. Nanofilters can remove dust and pollutants from the volumes of air and water, and the CNTs based filters can help in water desalination [1, 2]. Nanoporous water filters of 15–20 nm thickness can efficiently remove NPs, bacteria and viruses. Low-cost portable water treatment systems are promising for improvement of quality of drinking water in developing countries.

Fast, timely and informative medical diagnostics and treatment use lab-on-a-chip units that enable clinical or home testing in real time, which speeds up medical care delivery [6]. Various implants from microchips, vascular graphs and stents to artificial bionic arms and legs use nanomaterial-treated surfaces that improve wear, durability and can resist infections. Medical drags treated with NPs show better absorption inside the body and allows fast delivery to a target tissue or organ. Nanocarriers are also used to transport chemotherapy drugs directly to the cancer cells.

Optic properties of the sunscreens and their ability for light absorption in optical and ultraviolet frequency range can be significantly improved by NPs. The materials can be used for both skin/eye protection and food packaging with reduced ultraviolet exposure. Now many drink bottles are made from plastics with nanoclays that increases resistance to permeation O_2, CO_2 and water. Nanotechnology allows manufacturing chemical sensors which can detect molecular traces of particular chemicals. Such nanosensors are used for accurate identification of specific cells or liquids in human body. Similar sensors are used in the security systems at factories, airports, homes and other secured places.

This study is aimed at brief systematic review of literature on the biotoxicity problems created by modern nanotechnologies with emphasis on promising ways for the problem solution and perspectives.

2. Materials and methods

A variety of NPs of different composition, shape and size are used in modern technologies. They are point-type objects that placed at the transition between bulk materials and atomic or molecular structures and, therefore, possess the physical properties differ from the ones of the corresponding bulk material or atoms/molecules. The particles of size a~0.1–500 nm at least in one direction are considered as NPs (**Fig. 1, *a***). The terms NPs and ultrafine particles (UFPs) are often used synonymously though UFPs can reach also the micrometer (mcm) range. NPs are usually distinguished from "fine particles", ranging at 100–2500 nm, and "coarse particles", sized as 2.5–10 mcm. The nanopores in the filters and nanoporous materials also possess superior properties in their permeability, absorption, optical properties, etc. due to the size and high surface to volume ratio S/V.

The fibers and tubes with cross-sectional dimensions of the d~0.1–100 nm are considered as nanofibers (**Fig. 1, *b***) and nanotubes (**Fig. 1, *c***) while their lengths may vary between L~10–100 nm and L~10–1000 mcm. They are 1D objects which high cross-sectional curvature contribute significantly to thermodynamics on nanoscales. Some of the fibers are multiscales like DNA structure that has a base of <1 nm thickness composed into a spiral tube-like structures with d~100 nm packed into the chromosomes of the size L>10 mcm.

The nanotextured and nanostructured (**Fig. 1, *d***) surfaces of small thickness h~0.1–100 nm can be modeled as 2D or fractal-type 3D objects. The graphene sheet (**Fig. 1, *e***) is an example of the 2D material with zero thickness. The coatings produced by thin film (**Fig. 1, *f***) technologies are also 2D materials. The fullerenes (**Fig. 1, *g***) are considered as 3D objects while they have the surfaces composed as a layer of atoms of zero thickness.

NPs could be accumulated in dry powders, suspended in a basic liquid, dispersed in a gas or incorporated into a solid rigid or soft matrix. The NPs can be isometric with exactly or approximately similar size in any direction, elongated in one direction or flattened (i.e. elongated in two directions). Usually the particles with the aspect ratio (*AR*=length/width=*L*/*W*)<3 are called NPs, while those with *L*/*W*~3−5 are called nanorods (**Fig. 1, *h***). NPs can be fabricated by different techniques in a variety of shapes like nanocrystals (**Fig. 1, *j***), hollow nanospheres (**Fig. 1, *k***) and nanoshells (**Fig. 1, *l***), cubosomes (**Fig. 1, *m***), aggregates or agglomerates, and other complex sructures. Due to high *S*/*V* ratio the interface between the NPs and the solid, liquid or gaseous matrix which are composed by ions, inorganic and organic molecules, plays a specific important role in the nanomaterials (NMs) because the interface crucially changes all chemical and physical properties of the NMs. The interfaces can be described by surface energy, entropy and other thermodynamic parameters like the corresponding volumetric characteristics of the bulk matter at the higher scales [7]. Therefore, high scientific interest to NPs is explained by their intermediate location between the quantum atomic or ionic structures and bulk materials composed by the same molecules. Unlike the nanoscale structures, the physical properties of bulk materials are size-independent.

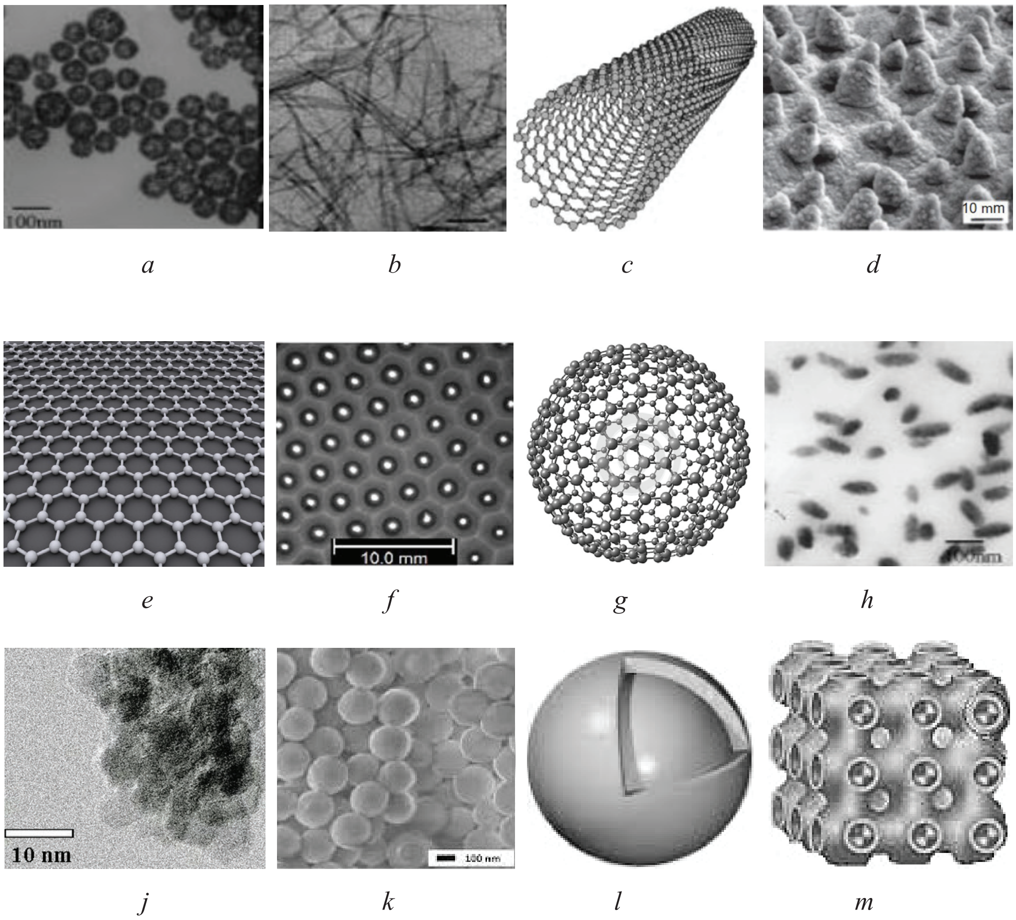

Fig. 1. Variety of nanoobjects: *a* – NPs, *b* – nanofibers, *c* – nanotubes, *d* – nanostructured surface, *e* – graphene, *f* – thin film, *g* – fullerene, *h* – nanorods, *j* – nanodiamonds, *k* – nanospheres, *l* – nanoshells, *m* – cubosomes

Micro- and nanoscale measurements and advanced microscopy methods, such as atomic force microscopy and electron microscopy, have contributed the most to nanoparticle research and measurements of their physical properties. It was shown in numerous experiments that NPs posses unique thermomechanical, electromagnetic, acoustic and optical properties, and many of them are in combinations [8].

Nanotubes have been shown having extremely high mechanical strength, especially CNTs. The single-walled CNTs are very strong in the axial direction with Young's modulus E~0.67–2.2 TPa depending on the type of CNT and its area. The consideration of Poisson's ratio (radial strain) gives E~0.53–2.24 TPa. When the radius of CNT increased from 2.7 Å to 4.1 Å, Young's modulus slightly decreased. For the multiwall CNTs the values E~270–950 GPa and G~11–63 GPa [9] have been obtained, while in [10] the values were E~34.65 GPa, G~0.85 GPa. Any composite materials reinforced by CNTs demonstrated increase in the E and G values in several times. The most strong natural materials in nature are compact bone tissues ($E\sim10^9$ Pa), chitin $C_8H_{13}NO_5$ ($E\sim10^{10}$ Pa), New Zealand flax (Phormium tenax) with $E\sim1.5\cdot10^{10}$ Pa that can be compared to steel ($E\sim10^{11}$ Pa). Therefore, nanotubes and CNT-based composites have unique mechanical strength and resistivity to the loads, crack formation and propagation, and destroy of the material.

NPs have very high surface area for the heat transfer because approximately one molecule of five in their composition lies at the surface. Simple molecular packing in a spherical shape problem solution shows that in the NPs of d~30 nm only ~5 % of its molecules are on the surface. In the NPs with d~10 nm and d~3 nm this ratio is ~20 % and ~50 % accordingly. That is why different types of nanoparticles offer different levels of thermal transfer enhancement. CNTs have a thermal conductivity $\lambda>3000$ W/mK, carbon nanofibers $\lambda\sim10-1000$ W/mK, and copper nanotubes $\lambda\sim400$ W/mK [11]. Addition of NPs to basic technical and biological fluids enhances their thermal properties as coolants [1, 12], especially CNTs and cellulose nanofibers [11, 13]. Thermal parameters of CNTs and CNT-based nanomaterials are reviewed in [14].

Numerous experimental measurements revealed superior electric properties of nanomaterials. Electric conductivity in solid nanoparticles, nanofibers and nanotubes is determined by electron transport, while in the viscoelastic NPs, cubosomes, suspensions of NPs, nanoemulsions and liquids with nanobubbles the ion transport is also significant. Electron transport in nanotubes, nanowires and nanoribbons is considered as 1D (are called molecular wires) due to their high aspect ratios (with thickness ranged from d<1 nm to 4 mcm and lengths L up to several microns) [1, 14]. This characteristic of nanotubes makes them excellent candidates for use as. The electron transport in molecular wires can be metallic or semiconducting, depending on their diameter and chirality. Since all carbon atoms in CNTs lie entirely on the surface, leaving the tube hollow, the electron conduction occurs either ballistically (without scatter) or diffusively (with scatter). CNTs have an electric conductivity $\sim10^6-10^7$ S/m, carbon nanofibers $\sim(2-8)\cdot10^6$ S/m, and copper nanotubes $\sim6\cdot10^7$ S/m [13, 14].

One of the classics of magnetism French physicist Louis Neel noticed that although the atomic spins in antiferromagnetic materials fully compensate each other in small particles of nanometer size this compensation may be incomplete because of the unequal number of atoms of two magnetic sub lattices particle acquires a nonzero magnetic moment. The reality even surpassed the expectations: at the nanoscale ferromagnetism becomes almost universal property [1, 15]. For instance, bulk gold as magnetically indifferent material in the form of golden nanoparticles demonstrate magnetic properties that is used in medicine for safe treatment of cancer tissues with embedded golden NPs by external magnetic fields [16]. Magnetic moment can be tuned with the nanoparticle cluster size, and the retained superparamagnetic properties are independent of the nanoparticle cluster size. The silica nanosphere coating enables straightforward covalent functionalization of magnetic NPs.

The lattices of NPs, nanofibers and nanotubes can influence light propagation, reflection, diffraction and interference [1, 11]. Therefore, the nanomaterials have various optical properties like ability of ideal white light generation with nanooptics, sunlight absorption with efficiency >40 % that is used in the solar panels. Highly concentrated solutions of TiO_2 and other metal oxides are used as inks for 3D-nanoprinter printed nanostructures using the sol-gel phase transitions. The sol-gel ink solidifies in air and retains its shape. Being warmed up 700 °C, it possesses liquid properties. After the printing, the ink dries almost immediately in the air, even when there are very small gaps between the slices in the 3D-structure. The ink is automatically distributed by the robotic arm and the designed structure allows quickly printing of micro-sized sensors and photonic crystals. The materials based on the CNTs have especially high light absorption properties similar to the 'absolute black body' in physics. The transparent materials have been obtained from the nanofibers of cellulose.

The nanostructured surfaces and lattices of NPs or nanofibers influence the acoustic wave propagation and reflection and, therefore, have unique sound absorption, generation and modification properties [1, 11]. The dark conductive surface of the materials coated by CNTs can be easily heated by a laser or electricity, which in turn causes changes in air pressure around the nanotubes, that is distinguished by human ear as sound. The phenomenon is called nanophotoeffect or thermoacoustic effect. A variety of sounds can be extracted from vertically arranged "grasses" of nanotubes that evaluated by eye as black velvet. High quality sound can be created when these surfaces are irradiated with laser light, fluctuating in the range of acoustic frequencies. These physical properties can be used in the production of light and flexible loud speakers, because the "grasses" can be used to create invisible wireless speakers suitable for installation in walls, windows, computers and cars.

3. Results and discussion

NPs are widely used for surface patterning and coating, modification of the composite materials, their reinforcement, or as drug-delivery particles for individual medical therapy, contrasting agents in visualization and medical diagnostics. NPs are used in display technologies, because their reflectivity depends on their orientation according to an applied electric field. Noble metal NPs and nanorods are promising as theragnostic agents for individualized therapies of different diseases. Theragnostics is a novel strategy that combines therapeutics and diagnostics with enhanced personalized efficacy by using patient- and disease-specific agents for diagnostic imaging with high molecular specificity and sensitivity [17]. Some types of anorods can absorb electromagnetic energy in the near-infrared (IR) frequencies and produce heat. Therefore, the nanorods selectively accumulated by specific tumor cells of a patient could be locally heated just by visual light that is safe for body tissues and serve for overheating the tumor cells in individual cancer therapy. For instant, golden nanorods are promising as contrast agents for biological imaging and photothermal heating [18].

Nanorods prepared from semiconducting materials can also be used for energy harvesting as light emitting nano-devices. For instance, ZnO nanorods can serve as nano-sources of near-ultraviolet (UV) radiation; they demonstrated tunable photoluminescence due to Stark effect mediated by external electric-field [19]. ZnO nanorods have also been used in different nano-scale electronic devices like ultraviolet photodetectors, Schottky diodes/rectifiers, field effect transistors, and ultra-bright light-emitting diodes LED [20].

Inorganic NPs are widely used in different fields of industry. Titanium dioxide (TeO_2) NPs exhibit antibacterial properties [21] and used for the wastewater treatment [22] and food safety [23]. TiO_2 coated packaging materials have clear antimicrobial properties [24]. Since UV is helpful in surface disinfection and post-harvest disease control of fresh produce, a combination of coating and UV activation is uses in the food technology.

Magnetic particles (iron, nickel, cobalt, gold, etc) with d~5–500 nm, magnetic beads with d~0.5–500 mcm, and magnetic nanobeads with d~of 50–200 nm are used in catalysis including nanomaterial-based catalysts, biomedicine (tumor heating), tissue specific targeting, magnetically tunable colloidal photonic crystals, microfluidics, magnetic resonance imaging, magnetic particle imaging, data storage, environmental remediation, nanofluids, optical filters, defect sensors, and caution sensors.

NPs of smaller size can easier change their physicochemical properties compared to larger particles and can create the opportunity for increased uptake and interaction with biological cells and tissues. Toxicity is often concerned with the production of reactive oxygen components like free radicals which will result in oxidative stress, inflammation, and consequent damage to proteins, membranes and DNA. Smaller NPs can easily gain access via skin, eyes and respiratory tract to the blood stream and from there they will be transported to the all internal organs, muscles and tissues. The high dose and long residence time of NPs in the vital organs like lungs, liver and kidneys can gradually lead to their dysfunction.

CNTs were shown to inhibit cell growth and cause the death of kidney cells [25]. Exposing the abdominal cavity of mice to long CNTs lead to inflammation of the abdominal wall [26].

Whereas 500 nm TiO_2 particles have only a small ability to cause DNA strand breakage, 20 nm particles of TiO_2 are capable of causing complete destruction of super-coiled DNA, even at low doses [27]. The laboratory mice which were given 2–5 nm TiO_2 nanoparticles showed a significant but moderate inflammatory response.

Magnetic NPs accumulated in the cells, organs and tissues which are at the influence of external magnetic fields $\vec{B}$, could be concentrated in the regions with high gradients $\nabla\vec{B}$ by the ponderomotive forces $\vec{F} = \chi_m \left(\vec{B}\nabla\right)\vec{B}$, where χ_m is the magnetic susceptibility of the material.

Gold NPs and nanostructures are the most extensively investigated ones owing to their versatility and unique optical, electronic and catalytic properties, biocompatibility, non-immunogenicity, antimicrobial properties, and facile surface chemistry [28–30]. Due to their biological comparably, easy synthesis and high stability, different gold particles have been studied for their practical uses. Various types of gold nanoparticle are used in many industries, such as electronics and medicine (for drug delivery and image contrasting), though their potential in nanotheragnostic applications. Experimental studies on mice shown the gold nanoparticles $AuNP_{20}$ and $AuNP_{50}$ were accumulated in liver, spleen, kidney, heart, blood and brain within a 30 days period [31].

Own experiments on the influence of nanodiamonds (NDs) of d~4–6 nm on rats revealed anticancer and radio protective influence [32, 33]. The rats with experimental Guerin's carcinoma were collected in several groups. The rats from the groups I and II were given NDs added to their usual diet (1 ml of diluted 5 % aqueous suspension) during 10 days prior to the X-ray cancer therapy. The rats from the groups I and III were treated by a therapeutic dose of X-ray radiation (5.8 Gy). The real (ε′) and imaginary (ε″) parts of the dielectric permittivity of the red blood cells from the blood samples of the rats have been measured by the microwave dielectric spectroscopy in a range of temperatures $0<T<48$ °C. The results of X-ray therapy of cancer were better in the rats received NDs and X-ray treatment (**Fig. 2**). Uptake of NDs give the $\varepsilon'(T)$ curve located closer to the control group results that the one for the group taken the X-ray therapy without NDs diet.

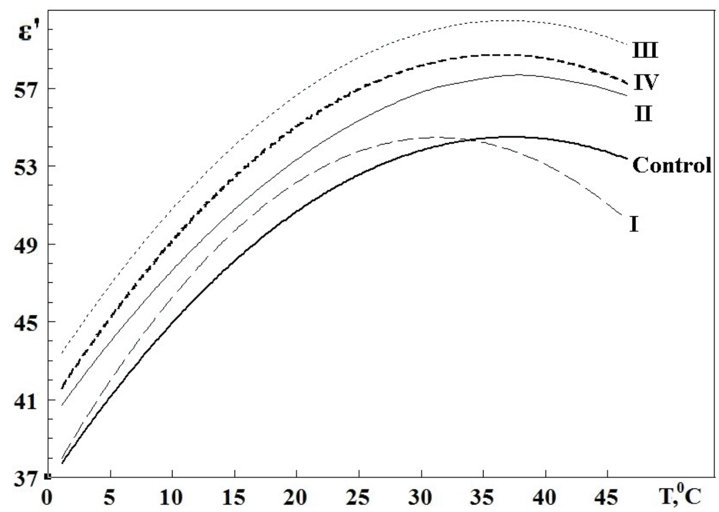

Fig. 2. Dependence ε′(T) for the groups I-IV of rats compared to the control group (from [32, 33])

Therefore, the toxicity problems caused by different NPs are of great importance for future development of nanotechnologies and healthcare. A series of toxic effects of CNTs, metal, oxide and some other NPs have been observed.

4. Conclusions

In the past decades a burst of experimental and theoretical research in nanosciences and new achievements in nanotechnologies has been observed. Nanotechnologies have played an important role in making various devices progressively smaller, more efficient and multi-functional. Among the most widespread are the paints for any surfaces from buildings till human body, self-cleaning surfaces, flexible and transparent solar panels with high solar conversion efficiency, skin "tattoos" that monitor salts and other metabolite levels and alert athletes or people suffering of diabetic, hypertension and other diseases. Shoes or clothes with embedded sensors collect data on muscle activity as a person moves, trains or proceed with sport movements. Nanosized drug carriers deliver chemicals to cancer cells, and the circulating drugs can be externally-activated and controlled. Energy-harvesting integrated systems (in textiles, shoes, human bodies) can harvest solar, electric or mechanical energy to charge electronic devices (pacemakers). Nano-treated surfaces of a food packaging can detect spoilage, contamination and has a tracking system to alert the producer and retailer.

Since the nanomodified materials are exposed to water, salt, oxidative, temperature influences, the NPs can be reattached, transferred and accumulated in the environments. Production and utilization of nanomaterials have also high risks of water, air and soil contamination by the NPs while efficient methods of their extraction, processing and modification in safer forms are not developed yet. The toxic influences reported in literature include oxidative influences and oppressive changes in the vital organs (liver, kidneys, lungs, brains, heart).

The main conclusion of the presented literature review is an urgent need for collecting all the evidenced of harmful toxic as well as positive health/treatment improving influence of NPs and nanostructured materials. It can be done via a database open for future statistical analysis of the data, generalizations and decision making based on expert conclusions or promising artificial intelligence technologies.

Conflict of interest

The authors declare that there is no conflict of interest in relation to this paper, as well as the published research results, including the financial aspects of conducting the research, obtaining and using its results, as well as any non-financial personal relationships.

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