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Application of nanotechnology in wastewater treatment

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Subject review

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Conventional wastewater treatment methods include various physical, chemical and biological processes. The results of such treatment can be limited because of high investment cost or, in some cases, due to poor treatment efficiency. For that reason, new approaches are continuously being developed as a means of supplementing or replacing traditional water treatment methods. The paper provides an overview of development of nanotechnology over time in the sphere of wastewater treatment, and examines the influence of nanomaterials on human health and environment, while also providing a review of future development trends in nanotechnology.

Key words:
nanotechnology, wastewater treatment, nanomaterials, nanoparticles, nanofiltration, nanoadsorbents

Pregledni rad

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Primjena nanotehnologije u pročišćavanju otpadnih voda

Klasični postupci obrade (pročišćavanja) otpadnih voda uključuju različite fizikalne, kemijske i biološke postupke čija primjena može biti ograničena zbog velikih investicijskih troškova ili u nekim slučajevima slabe efikasnosti pročišćavanja. Zbog toga se kontinuirano istražuju novi načini obrade otpadnih voda koji bi poslužili kao nadopuna ili alternativa klasičnim postupcima. U radu je dan osvrt na razvoj nanotehnologije u pročišćavanju otpadnih voda, utjecaj nanomaterijala na zdravlje ljudi i okoliš te prikaz budućih trendova razvoja nanotehnologije.

Ključne riječi:
nanotehnologija, pročišćavanje otpadnih voda, nanomaterijali, nanočestice, nanofiltracija, nanoadsorbenti

Übersichtsarbeit

Emir Zekić, Živko Vuković, Ivan Halkijević

Anwendung der Nanotechnologie bei der Abwasserbehandlung

Die klassischen Verfahren der Behandlung (Reinigung) von Abwässern umfasst verschiedene physikalische, chemische und biologische Verfahren, deren Anwendung aufgrund der hohen Investitionskosten eingeschränkt sein kann oder in einigen Fällen von schwacher Effizienz bei der Reinigung. Deswegen werden kontinuierlich neue Verfahren der Abwasserbehandlung untersucht, die als Ergänzung oder Alternativen zu den klassischen Verfahren dienen würden. Die Abhandlung gibt einen Überblick über die Entwicklung der Nanotechnologie in der Abwasserbehandlung, den Einfluss des Nanomaterials auf die Gesundheit des Menschen und die Umwelt sowie eine Darstellung zukünftiger Entwicklungstrends in der Nanotechnologie.

Schlüsselwörter:
Nanotechnologie, Abwasserbehandlung, Nanomaterial, Nanopartikel, Nanofiltrierung, Nano-Adsorbens

1. Introduction

According to some expert estimates, the world population will rise to 9 billion by 2050 [1]. This will cause problems with water pollution, and with an increasing amount of waste that will accumulate in water bodies. Due to higher consumption of water, and drinking water shortage, a high emphasis will be placed on wastewater recycling. In particular, it will be necessary to treat water to make it fit for particular purposes.

Conventional wastewater treatment methods include various physical, chemical and biological processes the results of which can be limited because of high investment cost or, in some cases, due to poor treatment efficiency (stricter legislation, new compounds in wastewater, higher concentration of heavy metals, etc.). That is why new approaches are continuously being examined to supplement and improve traditional water treatment methods. In this context, nanotechnological approaches are also being considered [1].

Current research in nanotechnology offers the possibility of developing technically and economically viable alternatives to conventional wastewater treatment. This technology is based on the application of materials on the nanometre scale, so that new structures, components and materials can be built at this (atomic) level. The whole theory is actually based on the fact that the properties of materials at the nanolevel are completely different from those at the macro level. Because of its properties, the development of nanotechnology has been quite significant over the past ten years [2, 3].

The United States is at the forefront of nanotechnology research. In this country, research activities in this area are conducted in the scope of the US Environmental Protection Agency (US EPA) and the National Nanotechnology Initiative (NNI). Within its borders, the United States has defined a total of 294,000 contaminated natural locations that need some form of sanitation and environmental protection. These are large and complex areas and most of them will require coordinated work by a large number of participants, as well as the development and implementation of new treatment processes. Among them, a dominant role might be assumed by nanotechnological processes. Currently, various forms of nanotechnology for wastewater treatment are being examined at the level of pilot projects. One of them, involving nano-sized zerovalent iron (nZVI) particles, has already reached commercial level and practical application in the treatment of pollution-affected areas [4].

Several other countries have also invested a lot in nanotechnology research. Among them is Japan which invests in similar projects through its research program (Japan Science and Technology Agency"s Core Research for Evolutional Science and Technology – JST"s CREST). South Africa has developed its National Strategy for Nanotechnology in 2006, and has innovation centres focusing on water treatment. Other countries belonging to this group are Brazil, Saudi Arabia, India, China, and some other developing countries [2]. The goal is to develop economically viable water/wastewater treatment methods to ensure proper sanitary conditions for a long period of time.

However, despite growing research efforts, most nanotechnological procedures have not yet reached the stage of practical application. The main reasons are possible negative impacts on living organisms and the environment. While, for now, only one technology (zerovalent iron nanoparticles) has reached commercial use, other technologies are still being explored. The research is mainly of a scientific nature and it takes time to explore how certain technologies can be implemented, what types of pollution can they be used for and, generally, whether they are actually suitable for practical application. When discussing the properties of nanoparticles and the area of their application, it can be seen that most papers on this issue focus on the structure of nanoparticles, i.e. on the synthesis of atoms and molecules within nanostructures, Figure 1. However, in recent years, a growing number of papers have been published on their toxicity and environmental impact.

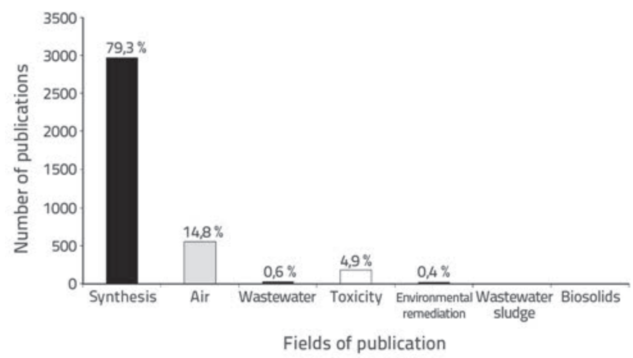

Figure 1. Number of publication in different disciplines of nanoparticles, until December 2008 [5]

The following main topics could be selected from the relevant literature survey [2]:

- New materials for membrane filtration,
- Nanomaterials for catalysis and photocatalysis,
- Nanomaterials for water disinfection,
- Nanomaterials for adsorption of pollutants,
- Nanoscale zerovalent iron.

The term "nanotechnology" was first mentioned in the late 19th century (1867) when James Clerk Maxwell published his first observations about this technology and presented possibilities of manipulating individual molecules [6]. At the beginning of the 20th century, with the development of ultramiscroscopes, nanostructures of a size of 10 nm could for the first time be observed and investigated (Zsigmondy 1914). In 1959, Professor Richard Feynman presented to the world a revolutionary theory. In his work "There"s Plenty of Room at the Bottom" he predicted various practical applications of nanotechnology [6]. However, his vision became feasible only in the 1980s or, more precisely, in 1981, when IBM experts developed the first "scanning tunnelling microscope (STM)", which enabled not only observation of certain atoms and molecules, but also their manipulation and construction of various nanostructures [6]. The first such (artificial) configuration of the atoms was made by

IBM scientists. Using xenon atoms (35 in total), they managed to form the letters "IBM", Figure 2.

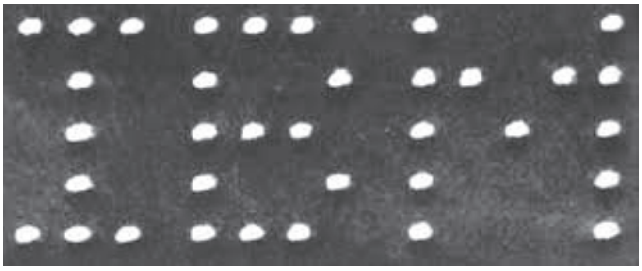

Figure 2. "IBM" formed by means of 35 xenon atoms, [7]

Interestingly, despite the great interest in this technology, its accurate definition is still lacking. It is generally accepted that nanotechnology implies management of those materials and particles of at least one dimension in the range of 1-100 nm [4, 8]. Such materials and particles are called nanomaterials or nanoparticles. However, the above definition is incomplete. The American National Nanotechnology Initiative defined two additional conditions, which implied that:

- nanomaterials are characterized by their unique physical, chemical and / or biological characteristics, different from their equivalent on the macro level
- it must be possible to configure and control nanomaterials at the atomic (nano) level [4].

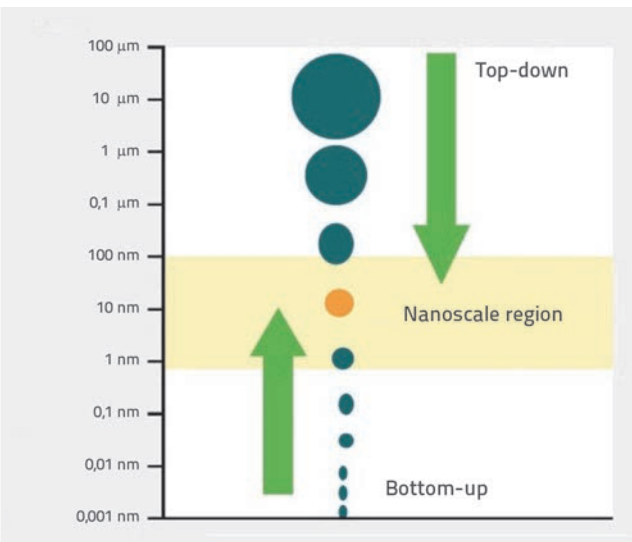

Figure 3. Top-down and bottom-up approaches for synthesis of nanomaterials, [9]

When it comes to synthesising or structuring nanomaterials, two main approaches can be applied [2]:

- "bottom-up" approach - in this approach, nanomaterials and nanostructures are made of individual atoms and molecules linked by chemical bonds, thus forming a somewhat larger and more complex nanostructure
- "top-down" approach - nanostructures are made of larger entities without configuration control at the atomic (nano) level. Synthetic techniques according to this approach include various physical breaking methods, laser radiation, nano-lithography, etc.

It should be noted that the approach used for structuring nanomaterials plays a key role in determining their main properties, stability, morphological characteristics, adsorption ability, degree of catalysis, etc. As mentioned before, nano-sized materials have unique properties, completely different from the equivalent structures on the macro-level. The most important feature is the large surface to volume ratio, which is why they are suitable for different forms of water treatment (adsorption, photocatalysis, membrane processes, etc.). Other important properties are related to different behaviour and motion of electrons (quantum effects). Thus nanostructures have completely different optical, electrical and magnetic properties, greater reactivity with neighbouring (polluting) atoms, faster chemical processes, etc. [2, 10].
All these characteristics of nanomaterials make this technology attractive in terms of eliminating contaminants and enabling wastewater treatment. Several different nanostructures have been defined with regard to treatment method. In this respect, nanotechnological wastewater treatment processes can be divided into three main groups:

- treatment and remediation
- sensing and detection
- pollution prevention [4].

The greatest emphasis has currently been placed on the treatment and remediation of wastewater. Various nanomaterials are at different stages of research, and each one has its own unique functionality. Some nanoparticles destroy contaminants (oxidation in the presence of nanocatalysts), while other separate and isolate these contaminants (nanomembrane filtration). Carbon nanotubes (Figure 4) have been recognized for their ability to adsorb dioxins. In this regard, they are much more efficient than the conventional activated carbon process [11].

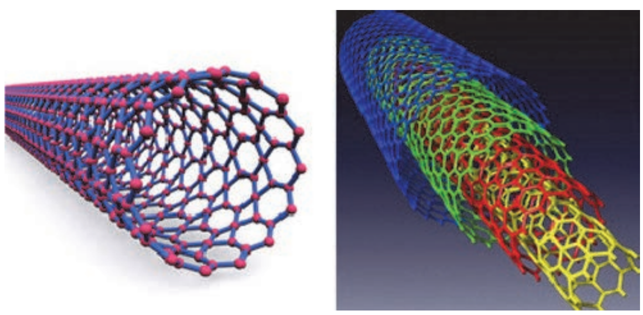

Figure 4. Single-walled and multi-walled carbon nanotubes, [12, 13]

As already mentioned, most nanomaterials and nanotechnological processes are still in the phase of scientific research. Only some of them are commercially available. Additional laboratory tests and pilot tests are required to gain a clearer understanding of these processes, and to fully define possibilities for their broader application. With each new technology there is concern about its potential impacts, which have not as yet been fully explored. In terms of maintenance, a possible problem for now is the elimination of accumulated contaminants from nanomaterials, and the possibility of their reuse. Further work is needed to minimize the shortcomings of this technology, and to take full advantage of its potential in the field of environmental protection.

2. Main nanotechnological processes for wastewater treatment

2.1. Nanofiltration

Membrane filtration plays an important role in removing various types of contamination and enables high level of water purification. Until recently, its biggest problem was a substantial investment cost (about 70 % of the total investment cost refers to membranes). As the price is lowering, the membrane wastewater treatment process becomes more and more popular in the market, mainly due to its high efficiency in the removal of solid waste materials, monovalent and divalent ions, various pathogens, etc.

Nanofiltration (with reverse osmosis, RO) is a high-pressure membrane treatment process. But unlike the RO, it requires a much lower drive pressure (7 to 14 bar), and so allows lower energy consumption. Centrifugal pumps are most often used for the pressure and circulation of wastewater within the nanomembrane. The plant consists of a large number of modules, with different membrane configurations within each module. In nanofiltration, the usual length of the module varies from 0.9 to 5.5 m, and the diameter ranges from 100 to 300 mm [14]. The modules are installed on the stand and can be arranged either horizontally or vertically (Figure 5). For vertical installation, a smaller number of connecting pipes and fittings are required, and the footprint is smaller.

Nanofiltration produces water that meets highly stringent requirements in terms of water reuse. Since this process is highly efficient in the removal of organic and inorganic substances, bacteria and viruses, the need for subsequent disinfection of water is minimal. Typical removal rates of some compounds and contaminants are shown in Table 1.

Table 1. Efficiency of nanofiltration in the removal of contaminants, [14]

Contaminant	Unit	Removal efficiency
Total dissolved solids	%	40 – 60
Total organic carbon	%	90 – 98
Colour	%	90 – 96
Hardness	%	80 – 85
NaCl	%	10 – 50
Sodium-sulphate	%	80 – 95
Calcium-chloride	%	10 – 50
Magnesium-sulphate	%	80 – 95
Nitrates	%	80 – 85
Fluorides	%	10 – 50
Arsenic	%	< 40
Atrazine	%	85 – 90
Proteins	log	3 – 5
Bacteria	log	3 – 6
Protozoa	log	> 6
Viruses	log	3 - 5

Nanotechnology offers a wide range of solutions for membrane materials, including [2]:
- Ceramic membranes for nanofiltration
- New polymeric membranes with anti-fouling coating (Organic brush-like coating, Membrane impregnated with nanoparticles)
- New composite membranes:
- Thin film composite membranes
- Metal/metal oxides + polymer
- Carbon nanotube + polymer
- Zeolites + polymer
- Aquaporin + polymer

According to Pendergast and Hoek (2011) [17], all membrane types used in nanofiltration can be grouped in three categories:
- nanostructured ceramic
- organic-inorganic membranes
- biologically inspired membranes.


Figure 5. Horizontal and vertical arrangement of modules, [15, 16]

They also developed methods for evaluating certain nanomaterial properties, such as permeability, strength, practical application, etc. They concluded that biologically inspired membranes possess the greatest potential for improvement but are far from commercial use. On the other hand, zeolite membranes have limited possibilities for further development but are closest to commercial use. Figure 6 shows that no type of membrane exists in the optimal (upper right) quadrant, but this could change over time when the biologically inspired membranes technology reaches maturity [2].

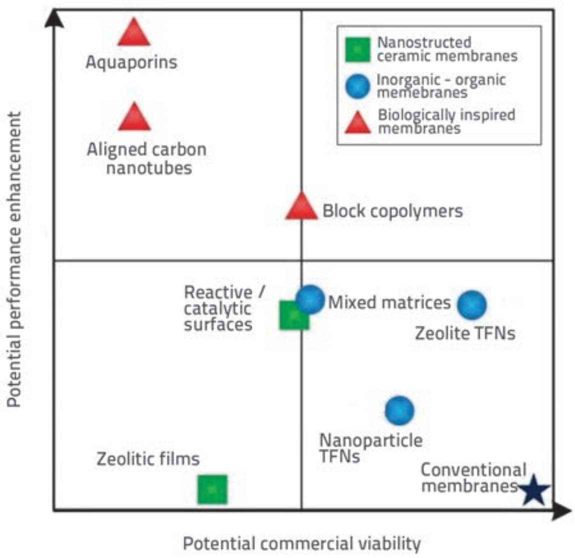

Figure 6. Comparison of potential performance and commercial viability of nanomembranes, [2]

2.2. Nanomaterials for catalysis and photocatalysis

Metal nanoparticles and metal oxides have proven to be very good catalysts in oxidation reactions. They exhibit a strong catalytic activity through which pollution molecules are oxidized forming less toxic substances, or converted into ecologically acceptable final products [2]. The main reasons for these properties of nanoparticles are:
- very small particle size, i.e. a large surface to volume ratio
- high reactivity directly related to nanoparticle size.

Nanocatalysts can effectively be used for chemical oxidation of organic and inorganic pollutants in water in advanced oxidation processes (AOP) [2]. These processes are based on formation of highly reactive radicals that react easily with pollutant molecules. The application of this process is often limited because of the extremely high costs of providing required energy (UV lamps, ozonators, ultrasonicators, etc.) [2].
Photocatalysis is the most significant oxidation process. This is a chemical reaction change that is induced by adsorption of a photon whose energy is greater than the energy needed to overcome the interstitial of two electron shells (valentine and conductive) of a semiconductor. When the photon illuminates the catalytic surface, the electron (negatively charged particle) is transferred from the valentine shell to the empty conduction shell and leaves a "hole" behind it with a positive charge. This "e-h" pair ("electron-hole") creates highly reactive radicals that bind the molecules of pollution and thus break them down [18].

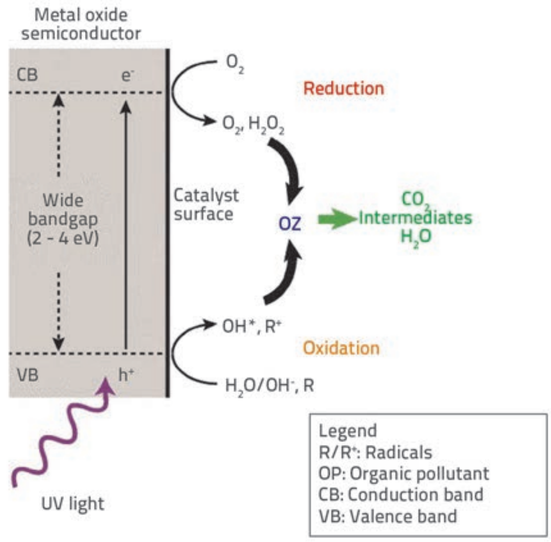

Figure 7. Photocatalysis on the surface of semiconductor as a nanocatalyst, [18]

However, there are several technical challenges that have to be met to enable broader practical application of this process, including
- optimization of catalysts in the exploitation of available light energy
- more efficient separation of nanocatalysts after treatment and re-application
- improvement of selective properties during chemical reactions.

However, the biggest drawback of this technology is the high operating cost of providing the required light energy (UV radiation), which is why this technology is still not considered to be economically viable. The research has therefore concentrated on the exploitation of (natural) solar energy for photocatalytic processes. Apart from being free, the advantage of solar energy is that it can also be used in open-air processes. The exploitation of renewable energy in these processes opens up new opportunities for the development of technologically efficient and economically viable wastewater treatment technologies [18].

2.3. Nanomaterials for water disinfection

In addition to having excellent adsorption and catalytic properties, some nanomaterials have proven to have great antimicrobial activity as well. Such materials include chitosan, silver nanoparticles, titanium dioxide, fullerene nanoparticles, carbon nanotubes, etc. All these nanomaterials are mild oxidants and are relatively inert in water, and are therefore note expected to create harmful by-products. There are several ways of applying the nanomaterials in water disinfection processes:

- direct action on (bacterial) cells in the sense of preventing electron passage through the membrane
- break through the cell membrane
- oxidation of some cellular components
- Hydroxyl radicals (within the action of nanoparticles as photocatalysts)
- the formation of dissolved metal ions that can cause damage to cellular components [19].

The most significant antimicrobiological mechanisms are shown in Figure 8.

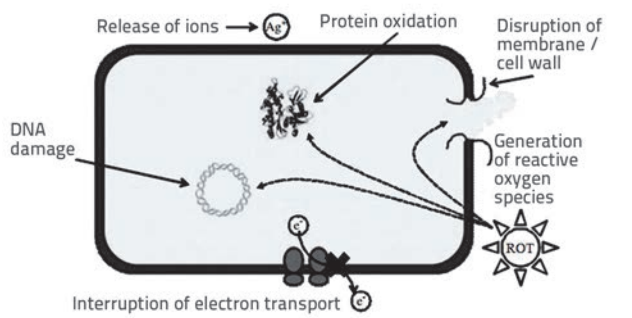

Figure 8. Various mechanisms of antimicrobial activities exerted by nanomaterials, [19]

However, there are some limitations regarding the use of nanotechnology in wastewater disinfection processes. For some nanomaterials to be effective in removing different types of microorganisms, they must be in direct contact with the cell membrane of bacteria, viruses, etc. Therefore, some nanomaterials (carbon nanotubes) need to be strongly connected to the reactive surface. Also, the deficiency of nanotechnology in disinfection processes is that there is no residual, i.e., subsequent antimicrobial activity in wastewater (such as in the case of chlorine use) [2]. The great advantage of conventional disinfection procedures is currently low cost. Nanotechnological processes will become more competitive after standardisation of the production of nanomaterials and following reduction of total labour costs.

2.4. Nanomaterials for adsorption of pollutants

Nanoparticles possess two important characteristics that make them very good adsorbents. These are the large specific surface of nanomaterials and surface multifunctionality or the ability to easily chemically react and bind ~~to~~ different adjacent atoms and molecules (Figure 9). These characteristics make nanoparticles not only effective adsorbents for various contaminants in wastewater but also allow for long-term stability, as this also results in adsorbent degradation (with the addition of catalytic properties of nanoparticles) and improves the adsorption efficiency.

With the discovery of carbon nanotubes (Iijima, 1991), a new carbon-based adsorption material was introduced to the world. Compared to the best known such material - activated carbon - carbon nanotubes possess approximately the same large specific surface, but their great advantage lies in the structure of nanomaterials and a much better arrangement of carbon atoms. In addition, nanomaterials possess unique mechanical, electrical, chemical, optical and many other characteristics that allow them to have much better adsorption properties for some contaminants (heavy metals and organic pollutants). This is why they are called the "material of the 21st century" [11].

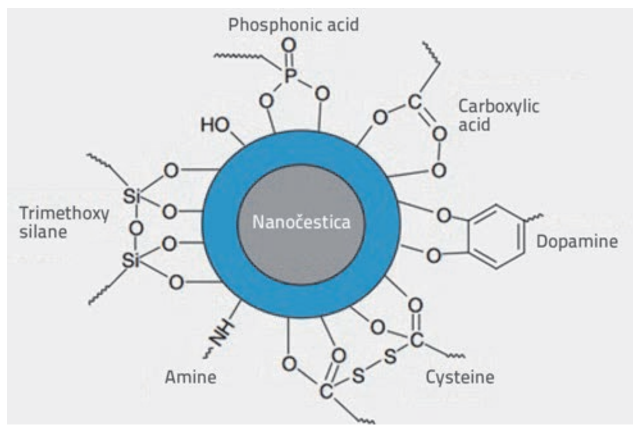

Figure 9. Multifunctionalities of metal oxide nanoparticle surface, [9]

Besides carbon nanotubes, metal based nanoparticles also have adsorption characteristics. The most common metal oxides used as adsorbents are iron oxides (Fe_xO_y), silicon (Si), titanium (Ti) and tungsten (W). They are mainly used for adsorption of heavy metals and radionuclides (unstable nuclides). The adsorption process is based on the electrostatic interaction of dissolved metals in wastewater and the nanoadsorbent surface. Changing the pH of the solution can significantly affect the strength of this interaction. Thus, the surface of the nanoadsorbent may be:

- acidic, with positive charge attracting anions
- basic, with a negative charge attracting cations from waste water [20].

2.5. Nanoscale zerovalent iron (nZVI)

Remediation of wastewater using nanotechnology is also gaining a greater interest. Compared to conventional materials and technologies, nanoparticles can provide great material and energy savings due to their prominent properties. Their nanostructure allows them to act as colloids, and so they can easily be injected into the soil and associated with contamination, regardless of location and depth. Key characteristics that nanoparticles must have in terms of remediation of contaminated groundwater are:

- high reactivity with contaminants
- high mobility within porous medium
- appropriate life span
- negligible harmful effects.

According to available research results, the zerovalent iron (nZVI) is considered to be the most interesting nanomaterial due to its low production costs, positive environmental effects, and high reactivity with contaminants. This nanotechnology has found extensive application in contaminated groundwater treatment, and has already reached the commercial level of application in the world. In addition, it has already proven to be a highly efficient technology for the removal of various organic and inorganic pollutants, including chlorinated solvents, pesticides, nitroamines and nitroaromatics, organophosphates, inorganic anions, arsenic, uranium, numerous metals, etc. [21].

Depending on the mobility of the contaminant, the nZVI injection method can be tailored for the treatment of:

- mobile contamination
- immobile contamination (Figure 10).

Thus, for the treatment of mobile contaminant plumes, low mobility nZVI is typically used for the formation of a "reactive treatment zone". The nanoparticles are sequentially injected and adsorbed to native aquifer material, forming a physicochemical filter. Mobile nZVI is typically selected for the treatment of a static contaminant body and is injected upstream for direct treatment [21].

A legal framework has to be put in place for the application of the described nanotechnology. This framework has already been defined in North America and in some European countries. A broader application of this technology relies on clear understanding of all processes and behaviour of the nZVI in the underground water system. Great emphasis is placed on its impact on the environment. The same experts who proposed this technology as an efficient method of water treatment also point to possible negative environmental effects. So far, little research has been done on the subject of its potentially harmful influence. Additional research and knowledge in this field is needed to accurately predict behaviour of nanoparticles in natural environment.

This detailed knowledge combined with the expected reduction in total costs will enable this nanotechnology to become more competitive, as compared to other conventional methods [21].

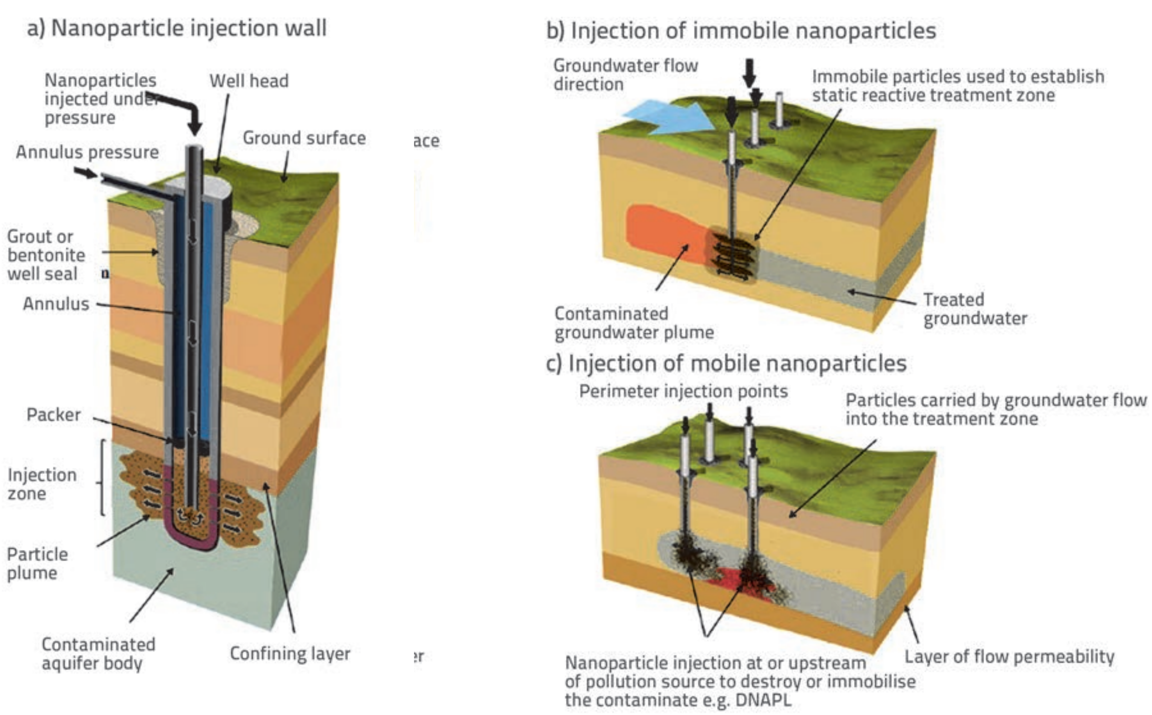

Figure 10. In-situ remediation technology with nZVI, [15]

3. Environmental and human health effects of nanomaterials used in wastewater treatment

Contemporary nanotechnology research in the sphere of wastewater treatment should not solely be based on the potential for improving properties of nanomaterials used in wastewater treatment, their efficiency, or standardization. A very important factor that is highly significant for broader commercial use of these products in the future is their impact on human health and the ecosystem. In almost all forms of application of this technology, the most critical part is the presence of nanomaterials in wastewater, because of their potential environmental impact. Additionally, once nanoparticles come to nature, their interaction with chemical substances in the environment can often have negative consequences.

Although no exact data are currently available, it is well known that largest quantities of nanomaterials that appear in the environment will eventually end up in soil, while smaller amounts of nanoparticles are present in water and air. One of the largest "sources" of nanomaterials in the soil is the sludge generated in wastewater treatment plants. After wastewater treatment, total pollution (including nanoparticles) accumulates in sludge, which is then transported and handled in various ways. A major problem in the treatment phase is that various types of polyelectrolytes are added to the wastewater for the purposes of flocculation and sedimentation of particles, and they directly affect the above-mentioned properties of nanoparticles: reactivity, dispersion, mobility, etc. The sludge is then used for various purposes and it ends up in the environment where it is either deposited at landfills, used as soil improver in agriculture, etc. This domain of wastewater treatment has so far been completely ignored and, in the future, proper sludge disposal methods will have to be considered, in line with environmental protection principles [2].

Basically, some studies on the impact of nanomaterials on the ecosystem have so far been made, and some satisfactory results have been achieved. However, numerous nano-toxicological studies are still in the initial phase and a lot of work is needed to make accurate conclusions about the effects of certain nanomaterials on human lives and the environment. More than that, this issue might be a limiting factor for commercial use of some promising nanotechnologies.

4. Conclusion

Although some nanotechnologies have proven to be very effective and promising in laboratory conditions in terms of their wastewater treatment potential, the commercial applicability varies considerably depending on the type of nanotechnology. In some cases, practical application has already been reached, while in other cases additional research is needed to minimize or eliminate all negative impacts. There are three main challenges to the mass production of nanomaterials:

- technical challenges on the use of particular nanotechnologies
- economic cost-effectiveness in terms of production and operating costs
- impacts on human life and the environment.

This paper focuses exclusively on the possibilities of nanotechnology with regard to wastewater treatment. However, nanoparticles possess another important feature that could make them a generally acceptable method: it is their ability to detect and prevent contamination. Because of their size and other characteristics, they have the ability to act as sensors for some targeted contaminants in wastewater. This property will be extremely important in terms of detecting pathogens or some newly discovered substances in water/wastewater.

Finally, future research should address the issue of compatibility of nanotechnology with the existing wastewater treatment methods. In developed countries, where a complete sewerage network with wastewater treatment plants is already in place, it can not reasonably be expected that the existing facilities will be abandoned in the oncoming period. Therefore, it will be important to define how to incorporate specific nanomaterials into the system, with minimum changes to the existing infrastructure. Meanwhile, independent nanotechnology processes could be applied in undeveloped or developing countries that have not as yet defined infrastructure and wastewater treatment methods [14].

Given the degree of development and future prospects for improving efficiency and economic viability, the following three types of nanomaterials can be regarded as the most promising ones:

- nanoadsorbents
- nanomembranes
- nanocatalysts.

The above-mentioned challenges regarding commercialization of nanomaterials are important, but many of them, including technical characteristics, cost of production, environmental impacts, etc. may be considered as temporary impediments only. To overcome these challenges, close co-operation between all interested parties is necessary. It can be expected that this advanced technology, with careful management aimed at avoiding undesirable consequences, can make a huge contribution to this field, and establish itself as a good wastewater treatment solution.

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