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Nanotechnological tools for the groundwater remediation: From lab to the real environments

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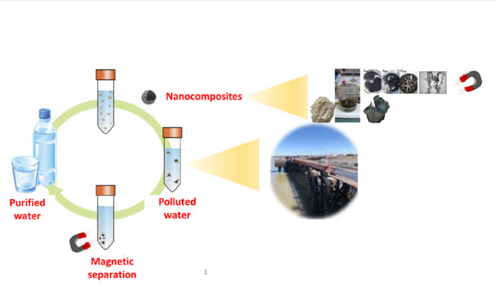
Abstract



The presence of nanomaterials in our everyday life has largely grown during the last years. They have emerged as relevant tools to find solutions to old problems. Among the areas where nanotechnology and nanomaterials have shown value, environmental remediation appears as an important one. The capability to tune the nanomaterial´s properties and characteristics to confer specific properties to eliminate target pollutants, accentuates the potential of these materials. The purification of water for human consumption or industrial process is currently of priority due to the increasing demand and reduction of the available water sources. This point of view article intends to show that the assays performed in the lab are not sufficient to validate large scale processes. Removal of contaminants in real aqueous environments require that due consideration is given to the behavior of the nanomaterial in field processes, because changes in efficiency and lack os stability may result in these conditions.

Keywords: nanocomposites, biomass, iron oxide nanoparticles, hydroxyapatite, arsenic, fluoride

Introduction

Water available for consumption is a very limited resource, essential for urban, industrial and agricultural needs. With the continuous growth of population and economic activities, the use and demand for water is increasing more and more [1]. Therefore, approximately 0.32% of the total water inventory is available for consumption, which represents 112,000 cubic kilometers, of which 90%, that is, 100,800 cubic kilometers, corresponds to groundwater. Furthermore, this amount is unevenly distributed and large regions of the world suffer from water scarcity. For the case of Argentina, see [2]. With the increasing demand for drinking water, groundwater has become the exclusive source of supply for many regions, as it is generally considered to be of better quality than surface water and is often consumed untreated as it does not contain high levels of microbial contamination. However, its quality can be affected by natural (geogenic) contaminants such as arsenic, fluoride and other metals [3].

Currently there are various designs of treatments for contaminant removal, such as electrolysis, ion exchange, chemical precipitation, membrane filtration and adsorption, the latter being one of the most effective and applicable technologies due to its efficiency, low cost and simplicity [4].

The advance of nanotechnology has open new challenges and opportunities to overcame the limitations associated to the water availability. Different kinds of nanomaterials have demonstrated to be suitable for remediation of water by eliminating pollutant of diverse nature. Inorganic nanomaterials appear as most promising due to the properties they expose after their reduction in size; and mainly by their versatility. These nanomaterials have been widely studied as candidates to eliminate different kinds of pollutants from water by two main mechanisms: adsorption, photocatalytic degradation (or the combination of both).

The design of nanomaterials with specific properties to bind pollutants of different origin is a challenge that may derive in the generation of more efficient remediation tools.

This Point of view article deals with the analysis of literature concerning the design of novel nano-based materials to the remediation of water with special emphasis in their perspective to be effectively validated in real environments.

Nano-based materials in water remediation

Nanomaterials promoting water remediation by adsorption mechanisms

Adsorption is a process that involves the enrichment of one or more species (adsorbate), which are initially in the liquid phase, on the surface of a solid (adsorbent) forming an interfacial layer. The term desorption denotes the reverse process. This phenomenon originates from the unsaturation of the surface atoms. allowing the interaction with the molecules of the environment. Based on the forces involved in the process, it is possible to distinguish between three types of adsorption: physical, chemical and ion exchange. Physical adsorption or physisorption is the result of weak interactions between the adsorbate and adsorbent by Van der Waals forces or electrostatic interaction. Since no electrons are shared or transferred, the species retain their chemical nature, making this process totally reversible. The capture of the ions will depend largely on the surface area and the charge development in a given medium. Chemical adsorption or chemisorption is a consequence of bond formation between the species involved, leading to changes in the chemical identity of them. It is generally irreversible, and more specific than the physisorption.

Finally, ion exchange is a fast and reversible process, in which ions of the adsorbent are displaced by those of the solution [5].

The adsorbent-adsorbate interaction forces are modulated by the environment in which they are immersed, and thus the environment has a significant influence on the efficiency of the adsorption processes. The conductivity of the medium, temperature, pH, in addition to the nature of the adsorbent and the adsorbate are variables of great incidence [6].

Nanomaterials employed in water remediation that function by adsorption mechanisms have diverse structure and nature. Magnetic nanoparticles (MNPs) deserves special interest regarding their surface reactivity, easy handling and separation from the aqueous media. The selected coating of MNPs may improve their adsorptive properties conferring specificity for certain contaminants groups. Recent works deals with the use of low-cost green materials to generate a stabilization and a synergy with the magnetic phase [7].

Derivatives of minerals such as clays, or zeolites, other inorganic minerals like hydroxyapatite, and some biomass residues have been selected and tested as adsorbents of inorganic contaminants in model water samples. Figure 1 summarizes different combinations that lead to nanocomposites from magnetically responsible iron oxide nanoparticles with biomass and mineral components.

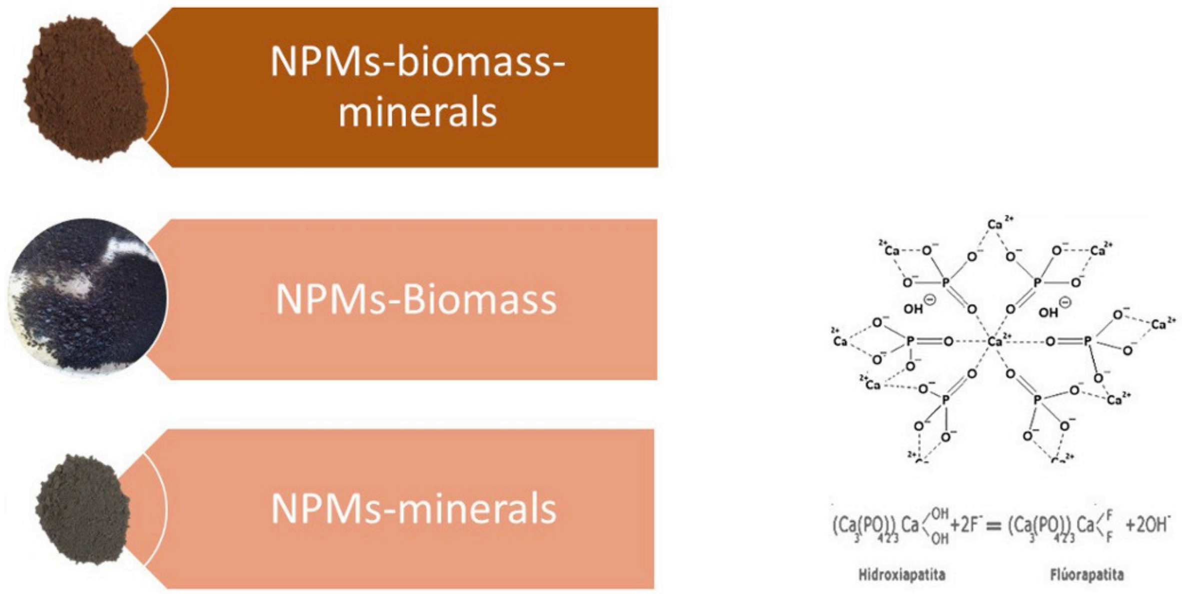


Figure 1. Schematic representation of different combinations of raw materials to obtain magnetic nanocomposites.

Studies developed in aqueous model solutions demonstrated that nanocomposites with improved capabilities may be achieved by combining the raw materials above mentioned. The functionality resulting in these nanocomposites allows the elimination of the most critical pollutants present in groundwater. Table 1 compares the performance of different nanocomposites registered in model solutions employing aqueous solution of As and F^-, as representative pollutants of groundwater matrix. The removal efficiency is expressed as mg pollutant g^{-1} adsorbent.

Table 1. Efficiency of different nanocomposites to eliminate As and F- from aqueous solutions.

Nanocomposite	As *	F-**	Reference
NPMs	0.180	ND	[8]
Zeolite	ND	ND	-
NPMs-zeolite	0.176		[8]
Hydroxyathithe	ND	12.1	[9]
Hydroxyapatite-biomass	ND	10.9	[9]

*Initial concentration: 200µg/L.
**Initial concentration: 3-80mg/L.

The initial concentrations were selected based on the content of these species in real groundwater samples. From these data it is clear that the magnetic phase is responsible for the As capture inducing mainly the formation of coordination linkages between Fe and As species. Electrostatic interactions are not favored due to the surface charge exhibiting the nanocomposites at the pH of the aqueous solution [8]. Concerning F^- removal, hydroxyapatite is the best option. The capacity of nanocomposites without HAp for fluoride removal is limited. However, zeolite-NPMs nanocomposites were satisfactory surface modified with Al and Ca moieties that generate greater affinity for F^-. However, the leaching of metallic moieties was evidenced after certain treatment time [10].

The stability of nanomaterials is not frequently tested in studies of water remediation, even though stability is a highly relevant characteristic.

Nanomaterials promoting water remediation by photocatalytic degradation and other combined mechanisms

Nanoscale semiconductors, such as TiO_2, CdS, ZnO and ZnS, have been extensively studied in photocatalysis applications from water splitting to pollutant degradation. Hence, their suitability to photodegradate organic pollutants (dyes, hydrocarbons, pharmaceuticals) in water has been extensively demonstrated by robust literature [11]. Photocatalytic degradation, one of the advanced oxidation processes (AOPs), is well known for its advantages being an energy-efficient method and relatively green procedure.

Various properties of nanosystems, such as size and morphology, strongly impact on the performance as photocatalysts. Hexagonal ZnO nanorods have a higher surface area and stability in comparison to spherical nanoparticles, leading to a more efficient photo-oxidation process [12]. For further improvement of photocatalytic activities of ZnO, metals, non-metals, and carbon materials can be deposited on ZnO to modify their optical characteristics and to improve electron-hole pair separation, which is essential for enhancing the photocatalytic process [13]. Studies have shown that the overall photocatalytic efficiency can be improved by depositing platinum to reduce electron-hole pair recombination and enhance visible light absorption. [14,15].

The application of these nanomaterials for the remediation of groundwater deals, mainly, with the elimination of organic matter affecting the conditions of water for human consumption. In this regard, the efficiency of ZnO, TiO_2 in the nanosize have been demonstrated against organic pollutants as dyes, aromatic polycyclic compounds and other; as well as against different types of bacteria [16,17].

However, the information concerning semiconductors based nanomaterials to eliminate inorganic species affecting the water quality is restricted. For instance, few articles deal with the use of ZnO-TiO_2 nanomaterials for the elimination of nutrients (mainly N and P species) from water. It is worth mention that nutrients involving N species such as nitrites, nitrates, ammonium; as well as phosphates species cause serious damages related to the eutrophication of water sources [18]. Own studies were developed by combining experimental assays and DFT theoretical calculations, demonstrating satisfactory yields in the removal of ammonium from water samples employing ZnO nanoparticles. The achieved findings revealed that non-electrostatic interactions took place but instead of that hydrogen and coordination bonds were the preferential interactions between ZnO NPs and ammonium. These data further indicated other abilities, besides photocatalytic degradation, of semiconductor nanomaterials to function as water remediators.

According to the reported information, it is noted that some inorganic pollutants such as heavy metals can be found in groundwater environments combined with other organic pollutants such as the polycyclic aromatic hydrocarbons (PAHs), pesticides, etc. In such cases, simultaneous removal mechanisms have been reported by means of semiconductor based nanomaterials. One of them is related to the preparation of phenanthroline/TiO_2 nanocomposites rich in oxygen vacancy defects exhibiting multiple functions as pollutant detection, adsorption, and degradation [19].

Other researches deal with the removal of heavy metals from aqueous effluents, which may also be present in combination with some organic compounds. In this case, semiconductors nanomaterials may be used for removal by advanced oxidation processes. In a second step, the ionic reduction of heavy metals can be induced by photocatalysis, typical of metal oxides such as TiO_2, ZnO, ZrO, Fe_3O_4, etc. [20]. These materials function as heterogeneous photocatalysts having a relatively narrow band gap width between the conduction band (CB) and the valence band (VB), in the presence of light of the appropriate wavelength (usually <370 nm) the photons are absorbed by the oxide, exciting the electrons to CB an crating a hole in VB. The existence of the electron-hole (e^-/h^+) pair enables redox-type reactions [21]. As mentioned, in water OH and superoxide radicals are formed. These last are responsible for the reduction of heavy metals to lower oxidation states which, in some cases, are less toxic for the environment; such is the case of Cr(VI) to Cr(III) [22,23]. In fact, it has been shown that the reduction of heavy metal ions is more effective when it is carried out simultaneously with the removal of organic contaminants [24,25].

These results may substantiate the use of semiconductors nanomaterials (mainly ZnO and TiO_2) for the remediation of groundwater in rural environments where sewerage is precarious or almost inexistent.

Effect of water matrix in the efficiency of nanomaterials in water remediation

Removal of pollutants from aqueous sources depends crucially on experimental conditions that have to be necessarily taken into account. For instance, in the case of adsorption mediated pollutant´s elimination, the adsorption time affects the process in different ways depending on the adsorbate particle size, the amount of active preferential sites on the surface, as well as changes in the chemical composition on the surface of the adsorbent material [26]. The pH plays a key role in the adsorption of ionic species, as it modifies the rate of the process as well as it can induce changes in the active sites of the adsorbent, modifying the selectivity against different adsorbates. [27]. Temperature is another important variable, in the case of adsorption mechanism. If the adsorption process is exothermic the adsorption efficiency will be lower as temperature increase, due to a decrease in the intensity of attractive forces between surface-active sites and adsorbate particles [28].

In general, after a survey of the current literature, one can find that most of the studies tending to provide insights regarding the strategies, mechanisms, etc. to water remediation are carried out under ideal conditions of pH, conductivity, temperature; even more normally, pollutant´s concentrations employed for these assays are not in line with those found in real environments. Therefore, model studies, as are normally defined, exhibit moderated utility when the aim is finding concrete solutions to a real problem.

Simulated water samples

A recommended practice is to use in the laboratory water samples replicating the conditions found in the aqueous environment to be remediated. For instance, magnetic nanoparticles (MNPs), based on iron oxide (magnetite) and ferrogel of gelatin and MNPs, were employed to remove heavy metals and nutrients from water samples. In a first step, an exhaustive adsorption study was conducted in batch using model solutions aiming to adjust the adsorption conditions, exploring the capability of these nanomaterials to remove Cu, NO_3^-, and NO_2. Adsorption studies using water simulating the real samples were also performed. In this case the simulated samples were prepared in laboratory by adjusting the conditions and composition in order to replicate an aqueous sample from the Bahía Blanca estuary. The components were ionic salts such as $NaNO_3$, $CaCl_2.2H_2O$, $MgSO_4.7H_2O$ Na_2CO_3, K_2HPO_4, $FeCl_3$, $CuSO_4.5H_2O$, $ZnSO_4.7H_2O$. The solution was adjusted to pH around 6.44 and conductivity of 4.98 mS. Both nanomaterials, MNPs and ferrogels, demonstrated an efficiency between 60 and 80% for the removal of heavy metals and 85% for NO_3^- and NO_2. [29]

In another study marine water was also simulated aiming to determine the ability of microplastics to accumulate heavy metals. In this case replicated marine-water was obtained from the commercially available "Marinium" that was diluted in distilled water by magnetic stirring [30]. The composition was mainly given by Na, Mg, Ca, Sr and K chlorides, bromides, sulfates and fluorides; to this milieu known concentrations of heavy metals were added [31].

The simulation of effluents from an urban discharge plant was also done, by the enrichment of aqueous samples with nutrients species such as ammonium and phosphates. Concentrations around 40-100 mg L^{-1} of NH^{4+} were employed adjusting the pH between 8-9 [32].

Substantial changes in nanomaterials stability can occur under simulated conditions. Therefore, it stability must be evaluated in experiments simulating the aqueous media, measuring also the possible leaching of nanomaterial´s components. Figure 2 depicts the sequential steps that should be followed in trying to close the lab investigation to real application in the water remediation field.

Figure 2. Representation of the steps involving in the validation of (nano)materials for water remediation.

Real water samples

The validation of novel technologies developed in the laboratory may be achieved by studying the behavior of such (nano)materials in the real environments where they will be employed. As it was earlier mentioned, significant surface modifications can occur leading to differences in the expected efficiency of those materials.

Even though the focus of this contribution is on the groundwater, it is useful to show the incidence of different media of nanomaterials. Table 2 summarizes data on pollutant removal by adsorption from marine, surface and ground water. Different kinds of pollutants are found depending on the type of water; hence diverse interaction involving in different elimination mechanism can occur.

Table 2. Efficiency of different nanomaterials in real water remediation ny adsorption

Water source	Nanomaterial	Pollutant	Efficiency (% of pollutant removal)	Reference
Groundwater	Nanocomposites zeolite-iron oxide	As	94	[8]
Groundwater*	Nanocomposites zeolite-iron oxide	As	61	[8]
Groundwater	Nanocomposites hydroxyapatite-magnetite	F-	56	[33]
Marine water	Gum Arabic co- ated iron oxide nanoparticles	NO_3^-, NO_2^- Pb_2+	29 44 36	[29]
Marine water	Ferrogels	NO_2^- Pb_2^+	56 47	[29]
Groundwater	Hydoroxiapatite-biomass nanocomposites	F-	77	[9]
Surface water**	Iron oxide biomass Nanocomposites	As Ca^{2+} Na^+	81 14 9.2	[34]
Surface water***	Iron oxide biomass Nanocomposites	As Ca^{2+} NO_2- and NO_3^-	73 29 82	[34]

*Continuous remediation process
** Water samples collected in Sauce Grande river
***Water samples from El Divisorio brook

Another point deserves attention is the design of the experiments able to reliably represent the remediation process in a domestic, industrial or rural environment. It is important to highlight that the information listed in Table 2 mostly refers to batch assays where the nanomaterial was incubated with the real water sample by the ´preset time according to previous basic or simulated assays. Any modification in the remediation process, which can be required in each specific case, necessary leads to differences in the pollutant removal efficiency. This issue is reflected in the data included in the Table as *. In such case the assays performed in entry 1 were replicated by employing a lab-made flux system, as shown in Figure 3, where the water sample is pumped in a continuous flux to a vessel containing packed adsorbent nanomaterial.

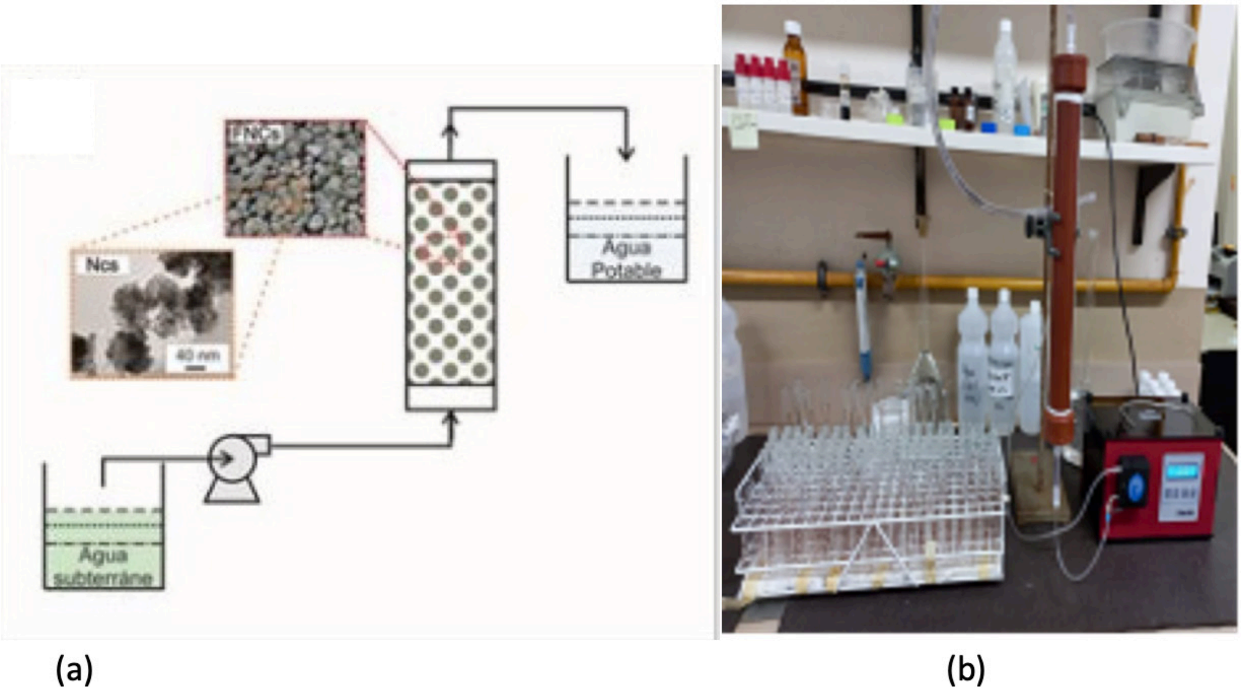


Figure 3. a. Schematic representation of a continuous in flux system to remediate groundwater; b. Photograph of a lab-made continuous system.

Special attention is further required to check the stability of the nanomaterial in continuous systems. The decay of the efficiency is not the only issue that has to be careful considered, but also the possibility of leaching of components of the nanomaterials. Continuos reutilization of the nanomaterial requires that enough information has to exist in order to predict the number of reutilization cycles or the volume of fresh water that the system will be able to remediate.

Concluding remarks

This point of view article aimed to concretely show the impact of nanomaterials in the water purification area. Even when the literature is abundant, this article focuses on the precautions required to translate the experiments in the lab to field processes. The knowledge of the relationship between nanomaterial´s properties and remediation mechanisms is mandatory and data concerning the stability of the nanomaterials in the media where they will be employed are also required. The need to analyze the behavior of nanomaterials in media and conditions where they actually will be employed appears as a key issue. Therefore, a sequence of basic assays using model aqueous solutions, followed by assays in simulated water should be suitable to lastly develop remediation assays in real water samples. Finally, the need to a multidisciplinary focus is highlighted in relation to the design of a suitable and efficient process which will be specific for the water source, and most important, for the site where the remediation will be implemented.

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Bio

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She's a researcher in the area of chemistry of materials, orienting her work to the design of nanomaterials with specific properties to be applied in the biomedical and environmental remediation fields. She received her PhD in Chemistry from National University of South in 2006, performing her studies in PLAPIQUI (UNS-CONICET), Bahía Blanca. After her postdoctoral studies, she moved to INQUISUR(UNS-CONICET, Bahía Blanca) as CONICET Assistant researcher. Currently she is the head of the Applied Hybrid Nanomaterials (NanoHiAp) group and is Associated Professor of Department of Chemistry in UNS.