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# Particulate Phases Possibly Conveyed from Nuclear Waste Repositories by Groundwater

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## 1. Introduction

Among the most severe sources of potential concern in terms of natural environment contamination with antropogenic radionuclides, a huge potential is associated to the radioactive waste resulting from the processing of the nuclear fuel used for energy generation purposes and from military and medical actvities. Although it represents about 3% of the total radioactive waste existing nowadays in the world, its total radioactivity amounts to more than 95% of that of the low & intermediate level and high-level radioactive waste taken together (Hu et al., 2010). By the present time, deep geologic disposal is considered to be the safest method for isolating highly radioactive and long-lived waste over millennia (Acero et al., 2010; Kim et al., 2011; Kurosawa & Ueta, 2001), while for the low and intermediate radioactive waste management, near surface storage is, for economic reasons, the preferred option (Dogaru et al., 2010; IAEA, 2004; Niculae et al., 2009).

It is a well-known fact that groundwater represents the most effective agent by which radionuclides stored in repositories could be transferred to the adjacent environment (Altmann, 2008; Baik et al., 2009; Geckeis & Rabung, 2008; Wersin et al., 2011). This is the reason why, each time when the issue of the radioactive waste repositories is addressed, irrespective of the repository type or of its inventory of stored radionuclides, the primary concern is to assess the local groundwater contamination risk.

Radionuclides may be conveyed by groundwater in a dissolved state, but especially in association with particles of various origins that are carried along with water. Those particles may have various structures and chemical compositions, their possible natures being: inorganic, organic or even micro-organisms. The inorganic particles genesis is closely related to the geochemistry of the environment with which groundwater is in contact, while particles of organic and biological nature are, as a general rule, allochtonous.

Particles dimensions may be very different, ranging from those of the colloids, up to microscopic particles, generically designated as "suspended particulate matter". Their reactivity and their ability of binding and conveying contaminants is to a certain extent controlled by their dimension, namely their reactivity is progressively enhanced as the particulate phase is more finely dispersed (Kersting & Zavarin, 2011; Wigginton et al., 2007).

This is also the reason why an increased interest is dedicated to the radionuclides behaviour in the presence of the colloidal matter.

The role played by the colloidal phase is intensely investigated not only for groundwater, but also for terrestrial surface water, namely for streams and lakes (Aleksandrova et al., 2010; Bondareva, 2011; Matsunaga et al., 2004; Monte, 2010; Monte et al., 2009; Ollivier et al., 2011; Semizhon et al., 2010), for estuarine water (Barros et al., 2004; Eyrolle & Charmasson, 2004; Porcelli et al., 1997), for sea water (Bowie et al., 2010; Otosaka et al., 2006; Scholten et al., 2005) or for soil water (Goryachenkova et al., 2009; Maity et al., 2011; Matisoff et al., 2011; Rachkova et al., 2010; Seliman et al., 2010; Xu et al., 2011). The colloid-radionuclide interaction is important not only from the perspective of their migration. For instance, the colloidal systems build-up is utilized for the radionuclides separation (Mansur & Mushtaq, 2011) and there has been noticed that they have interfering effects in the quantitative determination of the radio-isotopes. (Constantinou & Pashalidis, 2010; 2011; Kiliari & Pashalidis, 2010; Kyriakou & Pashalidis, 2011).

The radionuclides migration associated with the particulate phase in general, and with colloids in particular, that flow along with groundwater, is a topic of utmost importance for assessing the safety of the radioactive waste repositories. For assessing that overall process, there are conducted both field observations and experimental simulations in laboratory. The *in situ* investigation and the modeling of the role played by the colloidal matter and by the suspended particulate matter are mainly concerned with the saturated region of the water-bearing structures (e.g., Baik et al., 2010; Grambow, 2008; Kelkar et al., 2010; Laverov et al., 2010; Mal'kovskii, 2011; Malkovsky, 2011; Mazurek et al., 2011; Pourret et al., 2010; Severino et al., 2007; Utsunomiya et al., 2009). The modeling of the radionuclides migration in the unsaturated zone is addressed, as a general rule, by laboratory experiments (e.g., Ku et al., 2009; Massoudieh & Ginn, 2007). The latter range into two broad categories, batch tests and flow-through column experiments. In the first case, a solution spiked with the investigated radionuclide is mixed for a certain time-interval with the solid of interest, then the solution and/or the solid are analyzed (e.g., Anderson et al., 2009; Bradbury et al., 2005; Hu, Cheng, Zhang & Yang, 2010; Lujanienė et al., 2010; Rabung et al., 2005; Singer, Maher & Brown Jr, 2009). The column experiments investigate the radiocolloids migration characteristics, by simulating the groundwater flow conditions (e.g., Bryan et al., 2005; Li et al., 2011; Mibus et al., 2007; Solovitch-Vella et al., 2006).

Radionuclide migration with groundwater colloids through porous media (e.g., Bradford & Bettahar, 2006; Delos et al., 2008; Grolimund et al., 2001; Grolimund & Borkovec, 2001; 2006; Ilina et al., 2008; Kretzschmar et al., 1997; Li et al., 2010; Panfilov et al., 2008; Santos & Barros, 2010), or across fractured rock systems (e.g., Hu & Mori, 2008; Jeong et al., 2011; Kosakowski, 2004; Kurosawa & Ueta, 2001; Malkovsky & Pek, 2009b; Schindler et al., 2010; Tang & Weisbrod, 2009; 2010; Yamaguchi et al., 2008) are intensely investigated topics. At the same time, extensive research is conducted for assessing the humic and fulvic colloids effect on the radionuclides migration across the underground environment (e.g., Bouby et al., 2011; Geraedts & Maes, 2008; Joseph et al., 2011; Lippold et al., 2005; Lippold et al., 2005; Pshinko, 2009; Pshinko et al., 2009; Schmeide & Bernhard, 2010; Singh et al., 2009; Singhal et al., 2009; Yoshida & Suzuki, 2006).

The present work intends to succinctly review and to critically analyze the most recent contributions of the scientific literature in which there are addressed certain issues concerning the part that particulate phases play in the migration of the radionuclides characteristic to the radioactive waste stored in geological repositories, across more or less deep aquifer structures. An attempt is performed to provide an adequate definition of the particulate phases, and the main issues related to the radionuclides speciation are discussed, with particular emphasis on the radiocolloids development and distribution. A topic that is widely addressed in the present work concerns the radioparticles fractionation and their chemical characterization.

## 2. Radioparticles in groundwater

### 2.1 Particulate phases

It is a well-known fact that in a natural subsurface water-rock system, elements are distributed among the three constitutive phases, namely: (1) the solid phase, consisting of the mineral substratum and of the sediments; (2) the aqueous phase, within which elements are considered to occur as dissolved species, and (3) the particulate phase. It is by now fully demonstrated and unanimously accepted that much higher concentrations of contaminants are being carried by the particulate phases, as compared to concentrations of the corresponding dissolved species which are carried in the aqueous solution (Kalmykov & Denecke, 2011; Morel, 1983; Stumm & Morgan, 1996).

The particulate phase conveyed by groundwater is highly dynamic. Particles are continuously produced as a result of physical erosion and of chemical alteration of the mineral substratum, or ensuing to precipitation from super-saturated solutions. They undergo permanent composition changes, and are continuously removed from the water by dissolution, coagulation, and finally by deposition or binding to the solid phase.

Particles existing in groundwater may be both of inorganic and of organic nature (Wolthoorn et al., 2004). As a general rule, the inorganic particles composition mirrors to a large extent the nature of the mineral substratum in contact with water. Specifically, those particles may include solid fragments dislocated from the substratum, clay minerals, Fe(III), Mn(III,IV) and Al(III) oxihydroxide microparticles, silicates, carbonates, complexes and polymers of certain elements, etc. Organic particles are, at least in principle, of allochtonous origin, and they include fragments of degraded organic matter, macromolecules of organic substances which are specific to the soil (ex. humic or fulvic acids), but also organisms which are either alive or decomposing, microorganisms, viruses, as well as their exudates.

Aquatic particulate phases exhibit a continuous particle size distribution. In spite of that, practical reasons require that distinction is made between constituents that are dissolved, and those existing as particles on the one hand, and the various types of particles on the other. From a thermodynamic perspective, "dissolved" refers to a constituent for which a chemical potential can be defined (Stumm & Morgan, 1996).

In terms of dimension, shape and characteristics of transfer across the environment, taken as a whole, the particles range in two broad categories, namely colloids and suspended particles. A distinction between those two categories made by taking into account only the dimension criterion fails to be entirely satisfactory, although it is widely accepted and operationally useful. Opinions fail to be unanimous even as far as the dimensional boundary separating the two categories is concerned, so that according to various authors, the corresponding dimension may be 0.20 μm, 0.45 μm or 1 μm (Malkovsky & Pek, 2009a). In fact, the indicated values represent the pore dimensions of the filtering membranes which are used for separating the particles. The lower boundary of the colloidal particles dimension is accepted to be, as a general rule, $\approx 1$ nm, a circumstance which authorizes, from a certain perspective, the assimilation of colloids to nanoparticles (Geckeis et al., 2011; Wigginton et al., 2007). An important characteristic of the aquatic colloids, useful in distinguishing them from the suspended particles, consists in the fact that their vertical movement is not significantly affected by gravitational settling (Gustafsson & Gschwend, 1997; Stumm & Morgan, 1996).

### 2.2 Speciation of radionuclides in the environment

The migration of an element in a natural environment is basically conditioned by the way it speciates in that environment. By definition, the chemical species of a certain element are its specific appearances, defined as electronic or oxidation states, as complex or molecular structures (Templeton et al., 2000). At a given moment, the same element may occur in the hydrosphere under various physical-chemical appearances, including forms associated to the particles in suspension or dissolved forms, such as simple inorganic species, organic complexes, metallic ions adsorbed to the colloidal matter, etc.

Accounting for the radionuclides speciation is a fundamental step toward describing the geo- and bio-chemical processes in which they are involved, and above all for understanding how they migrate through the environment. (Hu et al., 2010; Salbu, 2009; Salbu et al., 2004). The definitions of the terms addressing the elements speciation, recommended by International Union for Pure and Applied Chemistry (IUPAC) (Templeton et al., 2000), have been adapted for the radionuclides by Salbu & Skipperud (2009) as follows:

*Radionuclide species* are defined according to their physicochemical properties such as nominal molecular mass, charge properties and valence, oxidation state, structure and morphology, density, degree of complexation.

The *speciation of radionuclides* is the distribution of a radionuclide amongst defined chemical radionuclide species in a system.

Colloids and suspended particles cannot be considered, under the above definition, species, they are, alternatively, chemical fractions. The *chemical fraction* is a group of chemical entities which have common physical (e.g. size), or chemical (e.g. reactivity) properties, that group being operationally outlined by means of an analytical process. The concerned analytical process is termed *fractionation*. Chemical fractions are not mutually exclusive, i.e. rather than identifying the involved chemical species, they indicate a specific behaviour. In terms of involved experimental techniques, fractionation is much more accessible that the detailed determination of chemical species. For instance, the results of the *in situ* fractionation of water samples derived from The Fen Central Complex (southern Norway), one of the world's largest natural reservoirs of thorium ($^{232}$Th), have shown that radionuclides occur mainly as colloids and chemical species of low molecular mass (Popic et al., 2011).

There is a certain dependence of the chemical reactivity of particles carried by water on their dimension (Wigginton et al., 2007), namely reactivity is, as a general rule, enhanced, as the particle dimension decreases. This behaviour is mainly a result of the fact that with decreasing particle dimension, the ratio between the constitutive atoms and the surface area of the particle increases. This is one of the reasons why a large interest is being recently dedicated to the investigation of how radionuclides are transferred by means of colloidal particles. At the same time, the colloids mobilization and transfer can occur both in a saturated, and in an unsaturated flow regime, this quite important issue requiring a careful consideration for an appropriate management of the radioactive waste repositories safety. The part that suspended particles, and macro-particles in general, play in the radionuclides migration is not a negligible one, yet it is worth considering it only in a saturated flow regime.

## 3. Formation of the radiocolloids

Entities formed by the coupling of colloidal particles with radionuclides and conveyed as such by groundwater, and which commonly are also termed *radiocolloids*, are classified in two large groups, according to their origin: (1) *intrinsic-*, *eigen-colloids* or "true" colloids; and (2) *carrier-* or *pseudo-colloids* (Geckeis et al., 2011; Malkovsky et al., 2009; Malkovsky & Pek, 2009a).

### 3.1 Intrinsic radiocolloids

Intrinsic-colloids are formed spontaneously, as a result of the polymerization of complexes derived from the hydrolysis of metal ions. Under certain environmentally-controlled circumstances, an increased tendency to form such colloids is displayed by Pu, Am, Np, for which the dimensions of the resulting aggregates and their number per unit volume are, as a general rule, proportional to the total concentrations of the concerned actinides in the solution (Murakami et al., 2005).

It is a well-known fact that tetravalent plutonium has a strong predilection to develop polymeric complexes and colloids. There has been however noticed that small polymers such as dimers, trimers and tetramers include mixed oxidation states of Pu (Walther et al., 2009). In natural waters with pH ranging between 6 and 8, Pu(IV) prevalently occurs under the form of true colloids. The latter exhibit a predisposition for getting bound to the rocks surface, a behaviour which with increasing particles dimension, becomes more and more obvious. It was found, for instance, that when the particles dimensions exceed 220 nm, Pu(IV) is virtually quantitatively sorbed on the rock surface (Perevalov et al., 2009). At the same time, equilibrium distribution of Pu(IV) polymers depending on the total Pu(IV) concentration in the solution was analyzed theoretically by Kulyako et al. (2008).

### 3.2 Pseudo-colloids

Pseudo-colloids are formed through the binding of radionuclides to the pre-existing colloidal particles of the groundwater. Any mineral fragment, either crystalline or amorphous (hydrated Al, Fe and Mn oxides), organic compounds (humic and fulvic acids), but also biota consisting prevalently of viruses and bacteria may act as carrier particles for the radio-nuclides. As compared to the first group of radiocolloids, pseudo-colloids are much more abundant in groundwater, and therefore they exert a much more extensive control on the radionuclides transfer.

Experiments have been conducted which were aimed at establishing the conditions under which Fe(III), Cr(III) and Zr(IV) build up pseudo-colloids together with colloidal silica. There was thus noticed that in a Fe(III) solution with a concentration of $\approx 1 \times 10^{-7}$ M, under pH conditions normally met in the hydrosphere, $Fe^{3+}$ cations and mononuclear $Fe(OH)_n^{3-n}$ hydroxo complexes mostly occur, and pseudo-colloids may form by the binding of Fe(III) species to colloidal silica (Davydov et al., 2003). In Cr(III) $\approx 1 \times 10^{-6}$ M synthetic solutions, when pH $>4$, in solution there are prevailing the $Cr(OH)^{2+}$ and $Cr(OH)_2^{+}$ species, with chromium displaying an obvious predilection toward forming pseudo-colloids with silica (Davydov et al., 2006). At the same time, in a solution of $\approx 1 \times 10^{-13}$ M concentration, Zr(IV) occurs under hydrated form as $Zr(OH)^{3+}$ and $Zr(OH)_2^{2+}$. At pH 2–12 Zr(IV) participates in formation of stable pseudocolloid particles (Davydov et al., 2006).

By using the surface complexation model, several investigators have modeled the radionuclides adsorption on the surface of the colloidal particles to form pseudo-colloids. For instance Batuk et al. (2011) interpreted in this way the sorption behavior and speciation of U on silica colloids, Degueldre & Bolek (2009) modeled plutonium adsorption on hydrous metal oxide solids, Del Nero et al. (2004) interpreted Np(V) sorption on amorphous Al and Fe silicates, and uranyl ions on Al-hydroxide (Froideval et al., 2006). Sorption of Np(V), Pu(V), and Pu(IV) on colloids of Fe(III) oxides and hydrous oxides and $MnO_2$ was studied over wide ranges of solution pH and ionic strength by Khasanova et al. (2007). The surface complexation model assumes that the adsorbing ion forms a surface complex with the adsorbing site, similar to the formation of a dissolved complex.

A box model has been proposed in order to interpret the kinetics of the radionuclides uptake on suspended particulate matter (Barros & Abril, 2005; 2008). At the same time, there has been investigated the kinetics of the Cs(I) sorption on hydrous silica (Pathak & Choppin, 2006), and $Am^{3+}$ on suspended silica as a function of pH and ionic strength in the presence of complexing anions, humic acid and metal ions (Pathak & Choppin, 2007).

Living organisms with dimensions similar to colloids, like for instance pathogenic bacteria or viruses, are present in groundwater naturally, to form a distinct group named biocolloids (Bekhit et al., 2009). Since they are living organisms they migrate in the subsurface porous medium, being subject to a complex of biological, physical and chemical processes. Those micro-organisms act as pseudo-colloids, since their surfaces are often negatively charged, thus having the ability to bind and carry radionuclides through the subsurface environment (Johnsson et al., 2008; Luk'yanova et al., 2008; Seiler et al., 2011; Singer, Farges & Brown Jr, 2009; Wilkins et al., 2006; 2010).

### 3.3 Colloids generated by engineered barriers

Besides the two already mentioned colloid groups, Malkovsky et al. (2009) and Malkovsky & Pek (2009a) distinguish an additional third group, which they designate as "primary colloids". The latter are colloidal particles derived as a result of groundwater leaching the isolating materials utilized in the storage of low and intermediate level radioactive wastes, as well as of high-level radioactive wastes in geological disposal.

Action taken in order to prevent, as much as possible, the contamination of the geological environment with radionuclides stored in a radioactive waste repository, irrespective whether the latter is located at the ground surface or in the underground, makes use of the so-called "*barriers*". The broadest meaning for barrier is "a physical obstruction that prevents or delays the movement of radionuclides or other material between components in a system, for example a waste repository" (IAEA, 2003). Such barriers may be either natural (i.e. geological), or constructed, in that latter case being called "*engineered barriers*". In most instances there are used "*multiple barriers*", namely "two or more natural or engineered barriers used to isolate radioactive waste in, and prevent migration of radionuclides from, a repository" (IAEA, 2007). Such barriers include the glass or ceramic matrixes to which the liquid waste is usually converted (Anderson et al., 2009; Curti et al., 2009), the steel canisters used for isolating the waste, the buffering backfill materials placed between the containers and the walls of the repository cells. The most adequate filling material for this purpose is bentonitic clay (Akgün et al., 2006; Ferrage et al., 2005; Galamboš et al., 2009; 2011; Gaucher et al., 2004; Pérez del Villar et al., 2005), since it has a low permeability and its coefficient of radionuclides diffusion is quite small (Arcos et al., 2008; Bradbury & Baeyens, 2011; Hu, Xie, He, Sheng, Chen, Li, Chen & Wang, 2010; Missana & García-Gutiérrez, 2007; Wang et al., 2005). When in contact with groundwater, all those barriers can release colloidal particles that in terms of both their chemical, and their mineralogical composition, are not characteristic to the concerned geological environment (Cadini et al., 2010; De Windt et al., 2004; Filby et al., 2008; Kunze et al., 2008; Wieland et al., 2004).

The effect of engineered barriers in terms of radiocolloids production is an outstandingly important research topic. It is worth mentioning that especially bentonite barriers in contact with weakly mineralized groundwater generate a highly concentrated colloidal phase which is liable to carry radionuclides (Albarran et al., 2008; 2011; Kalmykov et al., 2011; Kurosawa & Ueta, 2001; Missana et al., 2008; Sabodina et al., 2006; Tertre et al., 2005; Vilks et al., 2008).

### 3.4 The colloids stability

The colloids stability in groundwater is primarily controlled by the processes through which they agglomerate; at their turn, those processes are ruled by the colloid surface charge and by the solution composition, namely by its pH and ionic strength. (Geckeis et al., 2011; Schelero & von Klitzing, 2011). Accordingly, the groundwater chemistry plays a fundamental part in controlling the stability of the colloidal particles. Those particles ability of remaining in suspension in an aqueous environment depends on the interactions that are established between them when they reach close to one another. Colloids become stabilized through the formation of an electric double layer strong enough for preventing agglomeration. Yet this layer may be destroyed and the colloidal particles consequently coagulate and leave the system, along with the increase in ionic strength. There has been noticed that a reverse correlation exists between the colloids concentration in the solution and the ionic strength of the latter (Deepthi Rani & Sasidhar, 2011; Loux, 2011).

## 4. Fractionation and radioparticles charcterization

Analytical tools and detection methods used to characterize radioparticles in groundwater may be categorized as a function of the parameter to be determined as follows: (1) size fractionation; (2) size distribution; (3) surface area characterisation; (4) chemical and radiochemical analysis (May et al., 2008). Common size fractionation methods include ultrafiltration, tangential/cross-flow ultrafiltation (TFF/CFF), centrifugal-ultrafiltration (e.g., Gimbert et al., 2005; Liu et al., 2006; Pourret et al., 2007) and field-flow fractionation (FFF). Normally, those techniques are followed by the chemical analysis of the separated fractions. They will be discussed in detail in the following sections, since they are, taken together, the most frequently used experimental investigation approaches.

The size distribution of radioparticles can be investigated by a wide variety of techniques, such as laser light scattering (LLS) (e.g., Dreissig et al., 2011), diffuse light scattering (DLS) (e.g., Lahtinen et al., 2010), laser-induced breakdown detection (LIBD) (e.g., Baik et al., 2007), or photoelectron spectroscopy (Laverov et al., 2010). For the same purpose, atomic force microscopy (AFM) and transmission electron microscopy (TEM) (e.g., Doucet et al., 2005; 2004) are used as well.

The specific surface area (SSA) of the particles is the parameter describing the interdependence between the particles dimensions and their chemical or mineralogical composition. The SSA determination is frequently conducted by means of the Brunauer-Emmett-Teller (BET) gravimetric method. In an indirect way, information about the particles surface area may be obtained by means of AFM or TEM measurements.

In order to characterize the radionuclides speciation, a series of investigators have resorted to sequential chemical extraction experiments, conducted in accordance with pre-established work protocols (Bondareva, 2011; Bondareva & Bolsunovskii, 2008). As a general rule, ensuing to a scheme of sequential extraction of radionuclides from colloidal matter of groundwater, the following products will result: (1) water-soluble; (2) exchangeable, by using as reactant a 0.5 M $Ca(NO_3)_2$ solution at pH 5.5; (3) associated with carbonates, in the presence of 0.1 M $NH_4Ac$ solution, pH 4.8; (4) associated with organic matter, with 0.1 M NaOH solution, pH 10; (5) amorphous oxides, by using a mixture of 0.18 M $(NH_4)_2C_2O_4$ and 0.1 M $H_2C_2O_4$ solutions, at pH 3.5; (6) the residue digestion by means of HF (Novikov et al., 2009; Novikov et al., 2009).

### 4.1 Suspended particulate matter (SPM) fractionation

The SPM analysis techniques have been developed and are frequently conducted for surface, estuarine or sea water samples, but they are equally utilized as well for the analysis of radionuclide-contaminated groundwater (Katasonova & Fedotov, 2009; Stepanets et al., 2009).

Two different approaches are utilized in order to determine the concentrations of heavy metals and radionuclides bound on the SPM, one which is direct, and the other indirect. The direct determination method consists in separating the suspensions on filtering membranes of various porosities, followed by subsequent digestion and quantitative assessment of the contaminants from the separated material, by means of an adequate spectrometric technique (Blo et al., 2000; Nordstrom et al., 1999; Ödman et al., 1999; 2006; Ollivier et al., 2011; Yeager et al., 2005). Through the indirect method, both the filtered and the unfiltered water samples are analyzed in parallel, and the resulting concentration difference is considered to represent the concentration of the element bound on the SPM (Cidu & Frau, 2009; Cortecci et al., 2009; Gammons et al., 2005; Pokrovsky & Schott, 2002).

In a comparative study, Butler et al. (2008) have demonstrated that congruent results were obtained when the two methods were applied in parallel. Potential artifacts induced by filtration, such as contamination and/or adsorption of metals within the membrane have been investigated for different membrane materials, metals, etc. (Hedberg et al., 2011).

#### 4.2 Ultrafiltrafiltration

Ultrafiltration became the usual technique for separating colloidal particles from any type of natural water. The separation is performed by using a filtering membrane of a nominal size, often reported in a molecular size cut-off, in dalton (Da) units. Dalton is non-SI unit accepted for being utilized in the International System of Units whose values in SI units must be obtained experimentally. One dalton unit is equivalent to one atomic mass unit (amu) and is used in ultrafiltration in order to determine the approximate size of particles for which a rigorous molecular mass cannot be indicated.

Single filtration is frequently used for radioparticles separation (Caron & Smith, 2011; Novikov et al., 2009). Many investigators resort however to sequential (cascade) filtration, which involves the utilization of a series of cells with mixing, where each cell contains a membrane and a drainage system connected to measuring devices (Bauer & Blodau, 2009; Dreissig et al., 2011; Eyrolle & Charmasson, 2004; Graham et al., 2011; Pourret et al., 2010; Stepanets et al., 2009). In most cases, before starting the ultrafiltration operation, the SPM is separated by filtering the water samples on filtering membranes of 0.45 μm or 0.22 μm (e.g., Graham et al., 2008; Novikow et al., 2009; Singhal et al., 2009).

Tangential-flow ultrafiltration (TFF/CFF) is a common method for size fractionation in natural waters which has also been applied for colloids separation (e.g., Andersson et al., 2001; Buesseler et al., 2009; Goveia et al., 2010; Hassellöv et al., 2007; Ohtsuka et al., 2006). The main advantage of TFF is its use as a preparative fractionation method that allows for processing of large volumes of sample - even water samples reaching, each one, up to several hundreds of liters. It is also well known that the size distribution of colloids in natural waters can easily change due to aging, changes in pH, ionic strength or redox conditions (Hedberg et al., 2011). The processes that can potentially alter the size distributions of the colloids include coagulation, adsorption to surfaces, hydrolysis and precipitation. In addition to these processes, associated trace constituents are also affected by sorption processes, solution complexation and redox precipitations (Katasonova & Fedotov, 2009; Salbu, 2009).

#### 4.3 Field-Flow Fractionation

Several recently published reviews emphasize the efficiency of the Field-Flow Fractionation (FFF) techniques for separating and estimating physical parameters of different materials: biopolymers, biological cells, microorganisms, and colloidal and solid particles (Bouby & Geckeis, 2011; Dubascoux et al., 2010; Kowalkowski et al., 2006; Qureshi & Kok, 2011; Stolpe et al., 2005; Williams et al., 2011).

Particles separation by means of the FFF techniques is achieved by a combined action of the non-uniform flow velocity profile of a carrier liquid and a transverse physical field applied perpendicularly to this carrier. Carrier liquid flowing along the channel forms a nearly parabolic flow velocity profile across the channel. The sample to be investigated is dissolved or suspended in a carrier fluid and is pumped through a thin, not filled, channel. At the present time, FFF comprises a family of separation devices with a great number of sub-techniques used mainly for the separation and characterization of particulate species in the size range from $10^{-3}$ μm to $10^{2}$ μm. Highly popular among those sub-techniques is the flow field-flow fractionation (Fl-FFF). The version of the manufactured separation system for which the channel conveying the carrier liquid had an asymmetric shape (As-Fl-FFF) proved to be the most efficient, as it enabled nanoparticles ranging form 1 nm to 100 μm to be separated. As a general rule, an Inductively Coupled Plasma Mass Spectrometry (ICP-MS) is used as on-line detector, which allows reaching low detection limits, high sensitivity, large dynamic range and ability to simultaneously measure a large number of elements (Table 1).

| Method | Analyt | Detection method | Details | References |
|---|---|---|---|---|
| As-Fl-FFF | Cs, Eu, Th, U | ICP-MS | Interaction of bentonite colloids with metals in presence of HA | Bouby et al. (2011) |
| As-Fl-FFF | minor elements | UV detector, ICP-MS | Analysis of colloids released from bentonite and crushed rock | Lahtinen et al. (2010) |
| As-Fl-FFF | U | UV detector, ICP-MS | U complexation by groundwater dissolved organic C | Ranville et al. (2007) |
| As-Fl-FFF | Cs, La, Ce, Eu, Th, U | ICP-MS | Quantitative characterization of natural colloids | Bouby et al. (2008) |
| Fl-FFF | U(VI) | ICP-MS | U(VI) sorption to nanoparticulate hematite | Lesher et al. (2009) |
| Fl-FFF | $^{57}Fe$, $^{65}Cu$, $^{127}I$, $^{184}W$, $^{88}Sr$, $^{238}U$ | ICP-MS | Chemical and colloidal analyses of natural seep water | Cizdziel et al. (2008) |
| As-Fl-FFF | | LIBD, ICP-MS | Characterization of aquatic groundwater colloids | Baik et al. (2007) |
| As-Fl-FFF | | | Cm(III) complexation behavior of isolated HA and FA derived from Opalinus clay | Claret et al. (2005) |
| As-Fl-FFF | As, Cd, Sb, Se, Sn, Pb | ICP-MS | Soil leachate | Dubascoux et al. (2008) |

Table 1. Some field-flow fractionation procedures and detection method used to charactherize colloid particles.

A large number of recent works contributing to the development of the FFF techniques significantly widened their applicability range in particle size analysis (Ahn et al., 2010; Baalousha et al., 2006; 2005; Dubascoux et al., 2008; Gascoyne, 2009; Isaacson & Bouchard, 2010; Otte et al., 2009; Pifer et al., 2011).

### 4.4 Passive sampling techniques

For a given element, the overwhelming majority of its species are unstable chemical forms that occur under precarious equilibria. As a general rule, these equilibria are disturbed during the routine operations of collection, transport and storage of the samples, this fact resulting in most cases in erroneous information about the considered system. This is the reason why recently, in the elements speciation analysis, passive sampling techniques have received increasingly large acceptance Vrana et al. (2005).

In the most general meaning, passive sampling is that particular sampling technique which relies on the free transfer of the analyte from the sampled environment to a receiving phase in a sampling device, by the effect of the difference between the chemical potentials that the analyte has in the two environments. The transfer of the analyte from one environment toward the other continues until equilibrium is reached within the system, or until the sampling is stopped by an operator. In the first case, it is said that the passive sampling device operates in an equilibrium regime, while in the second one, it is said that it operates in a kinetic regime. In both situations, the sampling occurs without the involvement of any source of energy other than the indicated difference of chemical potential.

Among the passive sampling techniques, the diffusive gradients in thin films (DGT) technique, introduced by Davison & Zhang (1994), is highly ranked as a consequence of its ability to determine labile species in natural waters, sediments and soils. The DGT technique theoretical background relies on the Fick's first law of diffusion. For aqueous systems determinations a passive sampling device is used, which consists of a plastic piece in the shape of a piston, on which two gel discs and a filtering membrane are installed. The first gel, impregnated with binder material, is used for retaining the analytes. The second gel disc has a pre-determined porosity and its role consists in maintaining a constant concentration during the analyte diffusion between the solution and the binder material. The typical binder material is the Chelex-100 resin, while the material used for the diffusion control is the acrylamide/agarose hydrogel. In the end, the binder gel is eluted and the resulting solution is analyzed by means of ICP-MS French et al. (2005); Garmo et al. (2008; 2006), multi-collector ICP-MS (Malinovsky et al., 2005), thermal ionization mass-spectrometry (TIMS) (Dahlqvist et al., 2005), or directly through the gel analysis by laser-ablation ICP-MS (Pearson et al., 2006; Warnken et al., 2004). A comparative study between the DGT techniques and ultrafiltration has been conducted by Forsberg et al. (2006).

By simultaneously immersing several devices which have diffusion gels of various thicknesses or porosities, there is possible to obtain information about the nature of the complexes which are present in various categories of natural waters, and also about the kinetics of the geochemical reactions in which those complexes are involved (Zhang & Davison, 2000; 2001), including in porewater (Leermakers et al., 2005; Wu et al., 2011). DGT has been successfully utilized in order to monitor the radionuclides migration (Chang et al., 1998; Duquène et al., 2010; Gao et al., 2010; Gregusova & Docekal, 2011; Li et al., 2007; 2009; 2006; Salbu, 2007).

The same class of techniques also encompasses the diffusive equilibration in thin films (DET). In this latter case, the sampling device includes only a single layer of gel. This gel layer is maintained in contact with the environment to be analyzed, until equilibrium is reached between the analyte concentration in the environment, and the corresponding concentration in the gel. The technique is mainly utilized in sedimentary environments (Dočekalová et al., 2002). The total content of the analyte in the gel mirrors the ability of the concerned species to penetrate the gel, being controlled by its dimension. In the case of elements associations with colloidal matter, by combining information provided by DET on the equilibration, with information provided by DGT on the species dynamics, a much more appropriate description is obtained in terms of elements speciation (Fones et al., 2001; Gao et al., 2006; van der Veeken et al., 2008; Vandenhove et al., 2007).

In spite of being quite simple to handle, the DGT/DET techniques require a very cautious approach in terms of interpretation. One must take into account that the elements behaviour is not identical with respect to the binder gels, and that equilibration is largely controlled by the pH and the ionic strength of the solution. In addition, potential artifacts may be introduced during the devices preparation, elution and determination processes.

## 5. Conclusion

All the energy generation, industrial, medical, or military activities which utilize radioactive substances are producing low and intermediate level, and high-level radioactive waste, which needs to be isolated from the biosphere in order to protect the future generations from the hazards potentially induced by the associated radioactivity. As a function of the radioactivity level and of the half life length of the radioisotopes existing in their inventory, either near surface disposal facilities or deep geological repositories are assigned to the storage of that waste.

It is necessary that a radioactive waste repository location is selected very carefully in terms of its hydrogeological environment, since it is a well-known fact that groundwater is the most important vector involved in the transfer of the contaminants. The radionuclides migration across the geosphere takes place as water-dissolved constituents, but especially bound to the particulate phases carried by groundwater. Generally speaking, elements in a "dissolved" state speciate, i.e. they are distributed among forms defined by electronic states, oxidation states, isotopic compositions, as well as complex or molecular specific structures, while when they occur as particles, it is said that they belong to chemical fractions. A chemical fraction is a group of chemical entities which have common physical (e.g. size), or chemical (e.g. reactivity) properties, that group being operationally outlined by means of an analytical process.

Many authors rightly believe that the part played in the radionuclides transfer by particulate phases in general, but especially by the colloidal matter, is so important, that modeling approaches which do not take it into account are unrealistic. In an aqueous environment, radiocolloids occur as intrinsic-colloids and pseudo-colloids. The first category is specific to several transuranic elements which under certain conditions (e.g., pH, ionic strength), possess the capacity of forming structures with colloidal properties. The second type of colloids forms through the attachment of the radionuclide to particles pre-existing in groundwater. Those particles are spontaneously formed in the aquifer structures, but they might also derive from engineered barriers that are built in order to stop radionuclides from migrating out of the repositories. In this respect, bentonitic clays used as a buffer are the most important source of colloidal particles.

In order to identify and to characterize the particulate phases-radionuclides associations, a set of specific separation and determination methods is resorted to. Among the separation techniques, the filtration/ultrafiltration is the most frequently used, while for the colloidal matter characterization, much more efficient are the techniques belonging to the field-flow fractionation category. The latter have the advantage that they may be hyphenated with quantitative determination techniques, among which the inductively coupled plasma mass spectrometry is the most widely used. Recently, into the radiocolloids analysis domain there additionally included the "diffusive gradients in thin films (DGT)" and "diffusive equilibration in thin films (DET)" techniques, which appear to be outstandingly promising for the study of the radionuclides migration across the environment.

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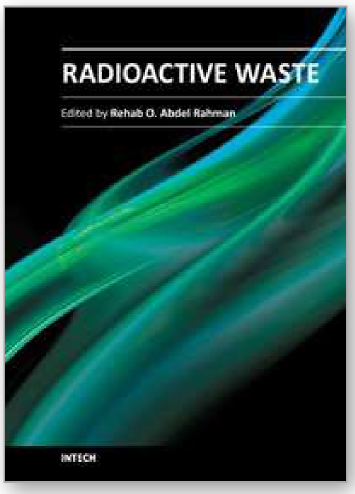


**Radioactive Waste**

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The safe management of nuclear and radioactive wastes is a subject that has recently received considerable recognition due to the huge volume of accumulative wastes and the increased public awareness of the hazards of these wastes. This book aims to cover the practice and research efforts that are currently conducted to deal with the technical difficulties in different radioactive waste management activities and to introduce to the non-technical factors that can affect the management practice. The collective contribution of esteem international experts has covered the science and technology of different management activities. The authors have introduced to the management system, illustrate how old management practices and radioactive accident can affect the environment and summarize the knowledge gained from current management practice and results of research efforts for using some innovative technologies in both pre-disposal and disposal activities.

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