

**EVALUATION OF THE USE OF METAL OXIDE-COATED SANDS  
FOR THE TREATMENT OF WATER FOR HUMAN CONSUMPTION  
AND NATURAL MINERAL WATERS**

**METAL OXIDE-COATED FILTRATION MATERIALS**

**March 17th 2005**

**Bibliographic study**

## I. INTRODUCTION

Quality limits relative to mineral micropollutant contents in drinking water are becoming increasingly lower and increasingly efficient elimination treatment processes are being implemented in order to meet these requirements.

Some treatments are more aggressive than others and present the drawback of causing the formation of by-products, which are sometimes carcinogenic; this is the case with ozone, for example, which can lead to the formation of bromates in the presence of bromides.

Some of the least aggressive processes used are precipitation-coprecipitation and adsorption on filtration media.

For the precipitation-coprecipitation process, recent studies have shown that sands coated with iron, aluminium, or manganese oxide, hydroxide or oxihydroxide were very good, inexpensive adsorbents which, in some cases, are more effective than the methods usually employed, such as precipitation-coprecipitation or adsorption on activated carbon grains (25, 26, 27, 70, 73, 74, 117, 129, 243, 244, 286)<sup>1</sup>.

Selective adsorption can also retain elements that conventional treatments are unable to eliminate (28, 29, 34, 62, 65, 76, 183, 267, 297).

These treatments are not straightforward filtrations but adsorption treatments.

This phenomenon was demonstrated after having observed that iron and manganese in particular were more effectively eliminated using old filters than filters containing new sand (7, 25, 27, 57, 92, 207, 253, 254) and can be explained, in most cases, by a catalytic action of the oxide deposits on the surface of the sand grains.

This report presents a synthesis of the studies conducted using these media.

The first part of this bibliographic review will examine the methods for preparing and depositing these metal oxides and the second part will look at the selective adsorption of mineral elements, organic matter, organic micropollutants and microorganisms.

## II. METHOD FOR PREPARING AND DEPOSITING METAL OXIDES, HYDROXIDES OR OXIHYDROXIDES ON FILTRATION MATERIALS

In a good number of cases, these deposits are formed naturally. So, for example, when water has a high manganese content, a black layer of MnO<sub>2</sub> is deposited on the surface of the sand grains. This phenomenon was well known by water treatment operators, who had noticed that manganese elimination was much more effective when the sand was coated with this black deposit (95).

Similar deposits form when water has a high iron content or is coagulated using iron or aluminium salt

Due to the interest of these deposits, specific methods enabling them to be obtained have been proposed since the conditions in which they are formed play a significant role in their adsorption capacities (58, 78, 293).

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<sup>1</sup> the numbers refer to the bibliographic references

***For iron oxide deposits:***

- the pH of the water and the iron concentration are very important parameters (17, 25, 73, 160).

- the formation of iron oxide from  $\text{Fe}(\text{NO}_3)_3$  leads to the formation of more amorphous ferric hydroxide than crystallised ferrihydrite (73, 161). The distribution of these two species also depends on the preparation conditions, particularly the temperature (at high temperatures, hematite is predominant) and iron concentrations.

- iron oxides formed under different conditions have different adsorption properties.

- iron oxides evolve depending on the age of the deposit:

lepidocrocite  $\gamma$  Fe OOH  $\rightarrow$  amorphous Fe OOH  $\rightarrow$  goethite  $\alpha$  Fe OOH (271, 273)

- tests have been defined to find out, first of all, the density of the iron oxide layer by measuring the quantities of iron deposited, after acid attack, and, secondly, the deposit's resistance to abrasion along with its specific surface area (160).

- in the application protocol proposed by LO *et al.* (27, 101, 141, 160, 263), the drying temperature plays a very important role on the adsorbent properties of the iron oxide deposited.

- whilst, generally speaking, the deposit is iron oxide (1 to 2 mg Fe per gram of sand), some authors suggest using  $\text{FeCl}_3$  (25).

- some authors have reported that the oxide deposit may slightly increase the diameter of the sand grains (43, 88, 160) and reduce their density (223), which may increase sand losses if water speeds during counter-current washing are not controlled.

***For aluminium oxide deposits:***

- the lower the pH at which the deposit is formed (pH 6 approximately), the higher the adsorption (138).

- as with iron, the preparation is made using a 1N aluminium solution and gradually adding a 4N NaOH solution, with drying being conducted at 70°C.

***For the manganese oxide deposit:***

Merkle *et al.* (187), Bajpai and Chaudhuri (18) have developed a method for applying  $\text{MnO}_2$  to sand, by making  $\text{KMnO}_4$  react with an  $\text{MnCl}_2$  solution in hot basic medium for 48 hours.

### *Mixed deposits:*

Some authors propose mixed Al, Fe (164) and Fe, Mn deposits (287).

### **III. SELECTIVE ADSORPTION OF MINERAL ELEMENTS**

Sand has a negative charge; prior coagulation of water reduces but does not neutralise all the negative charges of the colloids that it contains and does not therefore improve water clarification.

However, starting with a neutral or positively charged material, the retention of negatively charged particles is better, which has the effect of either reducing the quantity of coagulant required or significantly improving water clarification.

Hence they propose using metal oxide deposits on sand (73, 91),

For the elimination of heavy metals, the conventional methods are precipitation of carbonate, and carbonate hydroxydes, followed by separation by filtration, within a very narrow pH range.

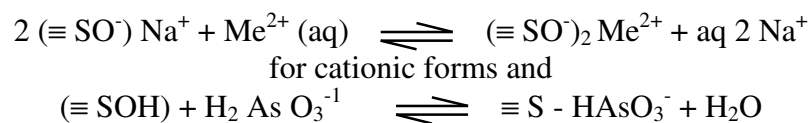
Precipitation is ineffective for  $\text{CrO}_4^{2-}$ ,  $\text{SeO}_4^{2-}$ ,  $\text{AsO}_4^{3-}$ .

However, adsorption is a better method since it permits a wider pH range and a greater efficacy (27, 70, 117, 129, 243).

The pH of the water plays an important role because at a  $\text{pH} < 5$ , the  $\text{H}^+$  ions compete with the cationic elements (56).

Forms which are complexed either to mineral ligands or organic ligands, which are not eliminated by conventional treatments, can also be retained (28, 34, 62, 65, 76). The same is true for certain oxianions (29, 183, 267, 297).

Iron oxides are used to eliminate metal elements (25, 26, 27, 61, 71, 81, 89, 220):



$\equiv \text{SO}^-$  and  $\equiv \text{SOH}$  indicate possible deprotonation at the surface of the metal oxides (209).

The oxygen atom plays the role of Lewis base versus a transition metal (Lewis acid).

Iron oxihydroxide therefore gives equally good results for heavy metals, equivalent to chelating exchangers or activated carbon (52).

$\text{MnO}_2$  and  $\text{FeOOH}$  also have oxido-reduction properties, particularly with arsenic (193), which explains the good adsorption results obtained, even with As (III).

Since iron and manganese oxides cannot be used as they are for water filtration (278) without a special preparation (GEH), sand was coated with these oxides instead to obtain a better mechanical performance (17, 25, 73, 143, 144, 160, 242).

Originally, these oxides were used to recover heavy metals in industry (galvanoplasty, etc.) (17, 26, 73, 226, 242).

These new materials are inexpensive and are available worldwide.

Iron is a very good element for decontamination of the environment (27, 81, 89) since it is both reducing and oxidant, non-toxic and not very expensive. Furthermore, like other oxides and hydroxides, it is an exchanger of either cations or anions and, in some cases, it can even fulfil both roles at the same time (25, 26, 62, 122, 286).

Natural sands combined with glauconite (Albian or Eocene "green sands"), a clay mineral with the formula:  $K, Na (Fe Al Mg)_2 (Si Al)_4 O_{10} (OH)_2$  are used to treat water.

With glauconite (phyllosilicate) coated with manganese oxide, it is possible to eliminate arsenic and manganese.

Adsorption has its limits since it is essential to regenerate the materials, either by acid-base treatment or by oxido-reduction (73, 122, 138, 164). However, the materials can be regenerated up to 50 times without losing their properties (73).

It has even been suggested that water could be filtered through granular magnetite particles (67).

Sands coated with metal oxides can retain either cationic forms or anionic forms (17, 25, 26, 74, 123, 160, 169, 221, 244).

The adsorption and adsorption yields are highly dependent on the pH of the water and the concentration of the element to be eliminated and also on the presence of an interferant (competitive ions) and the size of the sand particles (50, 141).

Table No. 1 gives an idea of the adsorptions obtained, but it is necessary to refer to the original publications for more details.

Table No. 1  
*Selective adsorption of different metals by oxides or oxihydroxides coated sand*

Sand	Element adsorbed	Quantity adsorbed (mg/l)	Reference	Comment
MnO <sub>2</sub>	As (III)	0.068	Bajpai & Chaudhuri [1999] (18)	at pH = 7.5
	As (V)	0.078	Bajpai & Chaudhuri [1999] (18)	at pH = 7.5
FeOOH	As (III)	0.076	Joshi & Chaudhuri [1996] (122)	at pH = 7.5
	As (V)	0.083	Joshi & Chaudhuri [1996] (122)	at pH = 7.5
Al <sub>2</sub> O <sub>3</sub>	Se (IV)	0.84	Kuan et al. [1998] (138)	at pH = 7.5
	Se (VI)	0.44	Kuan et al. [1998] (138)	
FeOOH	Cu (II)	1.271	Lai et al. [2000] (141)	at pH = 6.0
	Pb	4.14	Lai et al. [2000] (141)	
FeOOH	Cu (II)	0.032	Khaodhiar et al. [2000] (126)	at pH = 7.5
	As (V)	0.506	Khaodhiar et al. [2000] (126)	at pH = 7.5
FeOOH	Pb	0.083	Theis et al. [1992] (278)	at pH = 7.0

Adsorption is very rapid (99) but some authors also report slow adsorptions (26, 180, 281). The latter are obtained using high adsorbate/adsorbent ratios and can be explained by multi-adsorptions (26, 54, 234).

In certain cases, the adsorption of divalent metals can lead to the transformation of amorphous iron oxides into spinels with a formula of Fe<sub>3</sub>O<sub>4</sub> (54, 259) due to dissolution-coprecipitation phenomena (284).

The addition of iron II to goethite increases for example the adsorption of Co, Ni, Cu (55). Mixtures are adsorbed in the following order:



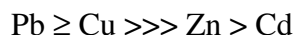
Displacement of Fe by an Me (II) metal occurs in the following order: Ni ~ Zn > Cd

Displacement of Me (II) by Fe occurs in the following order: Cd > Zn ~ Ni > Cd

The presence of metal oxides, and particularly iron oxides, explains the mobility of heavy metals in soils: Cd, Cu, Pb, Zn (177). The age of these metal oxides and the presence of organic matter play a very important role in these migrations.

The phenomena involved are surface phenomena associated with functional oxide groups (245).

Metals are retained by iron oxides in the following order:



### III.1 Selective adsorption of cationic forms

Numerous metals are retained by selective adsorption but some of them have been studied much more than others, particularly divalent iron and manganese.

Manganese oxide MnO<sub>2</sub> generally has higher adsorption capacities than those of iron or aluminium oxides or oxihydroxides (181, 218, 276, 281, 282).

### ***III.1-1 Elimination of divalent iron by selective adsorption***

Iron II is generally eliminated from groundwater by oxidation-flocculation-filtration.

Iron residues in treated water can be significant due to the passage of colloidal iron or iron bound to silica (194, 207, 254) or of incompletely oxidised iron (200), resulting in very rapid plugging of filters.

Adsorption-oxidation-retention leads to better iron elimination (35, 50, 92, 120, 236, 254). The thicker the layer of oxide applied to the sand, the greater the elimination (7, 92, 207, 254).

Adsorption of iron is 20 to 25 times higher on iron oxihydroxide-coated sand than on sand with no coating and this is therefore a very good alternative for the elimination of divalent iron.

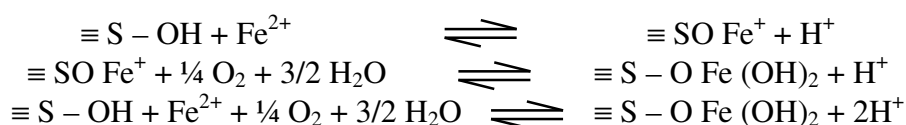
After adsorption of iron II, the latter is oxidised and in turn forms and oxide (254). This explains the very good results obtained in plants using this type of adsorbent filtration (253, 254, 256).

Adsorption of divalent iron is dependent on pH (103, 200) and oxidation, which relies on the oxidant concentration.

The advantage of this system is that the amount of sludge produced is much lower (17, 25, 73, 256).

This phenomenon is now well known (57, 116, 207, 254, 275).

The adsorption of divalent iron has been modelled using Freundlich (255) or Langmuir equations:



The adsorption of iron is accompanied by the release of H<sup>+</sup> ions (22, 274). But if the pH of the water falls, the quantity of iron adsorbed decreases.

Consequently, the alkalinity of water will always be very important:  $\text{H}^+ + \text{HCO}_3^- \rightleftharpoons \text{H}_2\text{CO}_3$

The quantities of oxygen required for this method of deferrisation are in the region of 2 mg/L, values which are comparable to the contents required for biological deferrisation. Furthermore, very often these two phenomena occur at the same time.

### ***III.1-2 Elimination of divalent manganese by selective adsorption***

It has been acknowledged that manganese can be eliminated by adsorption on manganese-coated sand for a very long time.

Originally, this retention was attributed to a catalytic action of the MnO<sub>2</sub> deposit.

It was Knocke *et al.* who explained this selective adsorption (131, 134).

The kinetics of adsorption of divalent manganese on MnO<sub>2</sub> are highly dependent on the pH of the water and the quantity of MnO<sub>2</sub> applied to the medium (sand, anthracite, etc.) (187).

Auto-oxidation of  $Mn^{2+}$  by  $MnO_2$  has not been demonstrated, apart from in trials at basic pH values.

Oxidation of  $Mn^{2+}$  requires either chemical oxidation or biological oxidation.

Manganese is therefore eliminated in two stages: first of all, by selective adsorption comparable to that observed for other cations and then, under certain conditions, by oxidation. Hargette and Knocke [2001] (102) studied oxidation by chlorine. Divalent manganese adsorbed on  $MnO_2$  is very rapidly oxidised by chlorine, which makes it possible to always have manganese dioxide with free adsorption sites for binding divalent Mn in water.

The elimination of divalent manganese by filtration was originally obtained on “green sands”, natural sands with which glauconite has been combined. This phenomenon is spontaneous when the sand is coated with  $MnO_2$ . It has been shown that this adsorption slowed down once all the adsorption sites were saturated (132).

It has been shown that this process could last more than a month (131) and that this duration depended on the divalent manganese concentration in the water and the filtration rate. Adsorption occurs in the first 15 centimetres of the filter, including at fast rates (131, 133).

The retention of divalent manganese is not directly dependent on the amount of  $MnO_2$  deposited on the material but, above all, on the number of exchange sites, explaining the crucial role of pH. At pH 6, adsorption is 80% lower than it is at pH 8. The reduction in pH for optimised coagulation of water (pH 5.5-6) used for better elimination of organic matter detrimentally affects manganese retention, unless the pH is increased to 7.5 between decanting and filtration, for example during remineralisation of water.

Studies have shown that chlorine was able to maintain this adsorption over time (53) by regeneration of  $MnO_2$  due to oxidation of adsorbed  $Mn^{2+}$  into  $MnO_2$ .

Chlorine gives good results, even at pH 6.1 whereas at this pH it cannot oxidise  $Mn^{2+}$  into  $MnO_2$  (132, 133).

New studies have mainly focused on the influence of counter-current washing: effect of air, of chlorinated water for washing, accumulation or elimination of deposited  $MnO_2$ .

The addition of air to counter-current washing leads to better “regeneration” of the deposit (5) and the same is true if chlorinated water is used (102).

Counter-current washing eliminates part of the deposit but as there is already the formation of  $MnO_2$ , an equilibrium is reached and the  $MnO_2$  deposit remains constant (102).

### ***III.1-3 Elimination by selective adsorption of lead***

- A number of studies have shown that lead adsorption can be obtained on metal oxides, particularly goethite (81, 104), sand or other materials coated with manganese dioxide (36, 64, 196, 203, 204, 240)
- and on iron or aluminium oxihydroxide deposited on sand (51, 141, 261, 276).
- on granular iron oxihydroxide (278), on activated alumina (265)

Bhattacharjee et al. [2003] (30) demonstrated that polymetallic nodules were very good lead adsorbents. As with other cations, the pH of the water plays an important part. In the case of polymetallic nodules, the optimum pH is 6.



Adsorption has been modelled using Freundlich or Langmuir models (440 ng Pb/g nodule).

Iron oxides give better results than aluminium oxides (261). The pH plays a very important part in these selective adsorptions (265).

For the elimination of lead, other authors have proposed selective adsorption on:

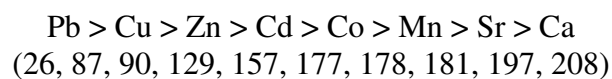
- activated carbon (4, 153, 199)
- peat (109)
- synthetic anionic clay (251)
- biopolymer (252)
- red mud (97)
- fly ash (66)
- natural zeolite (59)
- coral sand (272)

### ***III. 1-4 Elimination of copper, cadmium, zinc, nickel, cobalt and mercury by selective adsorption***

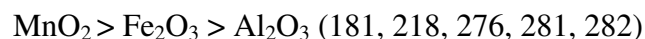
Iron, aluminium and manganese oxides or oxihydroxides are very good metal adsorbents. They explain the migration of heavy metals in soil (281).

In the treatment plant, they fulfil the same role when deposited on sand. Retention occurs in two stages: rapid adsorption at the surface of the oxide then slow diffusion into the oxide particles (14, 15, 17, 25, 38, 73, 85, 126, 145, 163, 189, 216, 253, 262, 265, 282).

The affinity of metal oxides for metals is as follows:



Oxides can be ranked in the following order:



These selective adsorptions have often been modelled using Freundlich or Langmuir equations (55, 137, 197, 214).

Gray demonstrated that the adsorption of  $\text{MnO}_2$  was inversely proportional to the ionic radius of the elements adsorbed (93).

Oxides or oxihydroxides can also adsorb complexed heavy metals (17, 25, 43, 122, 135, 273).

Specific adsorption studies have been reported in the literature:

- nickel (28, 283)
- cadmium (26, 38, 61, 162, 222, 242, 261)
- copper (38, 73, 126, 141, 142)
- zinc (258, 283)
- cobalt (171, 180, 273)
- mercury (229)

### ***III. 1-5 Radioactive cationic elements***

These mainly concern uranium, plutonium and radioactive strontium (3).

#### **III. 1-5.1 Uranium**

Uranium has been introduced into the environment by extraction of ore for nuclear power stations or for military applications. It is found in surface water and in some groundwater. It can also have a natural origin and in this case it is very often accompanied by arsenic.

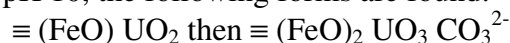
It is a radio-element with a very long radioactive half-life, which is always accompanied by its isotopes (20).

Uranium U (VI) is the stable valence in water.  $\text{UO}_2^{2+}$  is therefore the most commonly found cation for this element.

Iron oxides (ferrihydrite, goethite, hematite) are the most widely used oxides for retention of this element (19, 20, 112, 170, 198, 230, 289, 291).

The pH plays a very important role (20, 21): for pH values under 4 and over 10, adsorption does not occur.

At acidic pH, it is the  $\text{UO}_2^{2+}$  form that is present and since the  $\text{H}^+$  ions compete, adsorption does not occur. At pH 4 to pH 10, the following forms are found:



bound to iron oxides fixed on the sand. At pH values of over 10, the  $\text{UO}_2(\text{CO}_3)_2^{2-}$  form is no longer obtained and turns back to a solution.

#### **III. 1-5.2 Plutonium**

The adsorption of plutonium VI is obtained either on manganese dioxide (257), or on iron oxihydroxide (124, 185).

#### **III. 1-5.3 Strontium**

Radioactive strontium is discharged into the environment by nuclear power stations.

It is iron oxihydroxides that are used to eliminate it (12, 41, 47, 101, 190, 191).

Ahmad reports that the presence of calcium and magnesium may markedly reduce the adsorption of strontium (25).

### **III.2 Selective adsorption of anionic forms**

It is oxianions that are concerned.

The elements included are arsenic, selenium, antimony, chromium, phosphates and silica.

#### ***III. 2-1 Elimination of arsenic by selective adsorption***

The quality limit for arsenic in water for human consumption having been reduced from 50 to 10  $\mu\text{g/l}$ , numerous studies have been conducted regarding elimination of this compound.

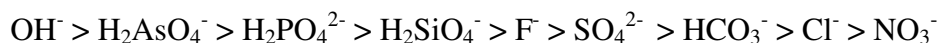
Elimination of arsenic by precipitation-coprecipitation is only effective if the arsenic is at valence V.

Furthermore, balanced concentrations are close to 10 µg/l, which means that the standard is not always complied with.

Numerous natural mineral waters have a high arsenic content and since any modification in the mineralisation of water is prohibited, precipitation-coprecipitation treatments using iron salts are therefore impossible to use. Selective adsorption has been one of the methods proposed for this type of water. For drinking water, the use of these treatments is mainly due to the fact that some adsorbents used can guarantee retention of tri and pentavalent forms (119).

Several media have been proposed, particularly iron, aluminium and manganese oxides and oxihydroxides (40, 84, 85, 174, 175, 219, 270) and activated alumina (235, 237).

For this adsorbent, we can rank the elements by decreasing affinity:



Once again, pH plays a very important part.

Fluoride and phosphate ions compete with arsenic; trivalent arsenic is not eliminated.

### III. 2-1.1 Activated carbon (98, 154)

The adsorption durations are short. Some authors have suggested pre-treating activated carbons to coat them with metal oxide or oxihydroxide (225, 229).

In this case, the material is much too expensive since the same results can be obtained with sand coated with these oxides (25, 73, 121, 122, 138, 164).

### III. 2-1.2 Iron oxihydroxides (73, 114, 122, 126, 279, 288)

These oxides can be either amorphous iron oxides compressed and formed into granules (GEH), or sand coated with these oxides (45, 183, 246).

Depending on pH, it is possible to adsorb arsenic at valence III and valence V (174, 220). And at a pH of close to neutrality, the two valences are retained (115).

Sulphates interfere very slightly (6).

As for phosphate ions, these interfere very strongly with adsorption but their content in water means that this factor does not really need to be taken into consideration (111).

Trivalent arsenic is less well adsorbed than pentavalent arsenic (105, 156, 260).

As far as iron oxihydroxides are concerned, uncertainties still remain for certain parameters:

- lifetime of filters treated in this way (46, 83, 125)
- phenomenon explaining the Red-Ox transformations occurring in the filters when arsenic is adsorbed (As III and As V).

It has been shown that in aerobic water, complete oxidation of As III is obtained (80, 239).

This oxidation has been demonstrated experimentally (72, 127).

Certain authors trigger oxidation due to bacterial phenomena (100, 294, 295) or the presence of algae (268).

The catalytic action at alkaline pH of certain clays or metal oxides has also been demonstrated (82).

Some authors have demonstrated the coexistence of As III and As V (1, 139, 202).

In certain cases, phenomena of bioreduction of As V into As III have also been reported (31, 42, 130, 147, 206, 264).

As a result of these uncertainties, arsenic is very often subjected to prior oxidation by ozone, for example, before being eliminated by selective adsorption on iron oxides (17, 75, 105, 122, 127, 183, 184, 215, 285).

### III. 2-1.3 Aluminium oxihydroxides coated sand

These materials do not permit retention of trivalent arsenic. Fluorides and phosphates interfere (172, 174).

### III. 2-1.4 Manganese oxides, MnO<sub>2</sub>

These oxides fulfil a dual role: firstly, their adsorption capacity is greater than those of iron and aluminium, Mn > Fe > Al and, secondly, manganese dioxide adsorbs arsenic III and oxide into arsenic V. Adsorption is therefore the same for both arsenic valences (18, 23, 68, 69, 106, 173, 193, 195, 205, 210, 211, 213, 247, 269, 285).

### *III. 2-2 Elimination of selenium by selective adsorption*

Selenium present in water can also be found at two different valences: valence IV which is eliminated by precipitation-coprecipitation and valence VI which is not eliminated by this method.

Since the quality limit is set at 10 µg/L, it is not always possible to reduce concentrations below this level by precipitation-coprecipitation using iron salts.

Here, selective adsorption is again a process of choice to eliminate this element.

Oxides or oxihydroxides coated sand give good results.

### III. 2-2.1 Iron oxide or oxihydroxide

Iron oxides or oxihydroxides eliminate Se IV and Se VI. The pH value plays an important role (138, 248).

### III. 2-2.2 Aluminium oxihydroxide

Aluminium oxihydroxide coated sand has been used to eliminate selenium IV and VI from water.

The elimination of Se IV is better than that of Se VI (138).

### ***III. 2-3 Elimination of antimony by selective adsorption***

Antimony is retained by selective adsorption in exactly the same conditions as arsenic. As with arsenic, Sb III is less well adsorbed than Sb V.

Manganese dioxide oxidises Sb III into Sb VI.

### ***III. 2-4 Elimination of chromium by selective adsorption***

Chromium is found at two valences in water.

Cationic chromium III is very easily eliminated by precipitation-coprecipitation in the form of carbonate or hydroxycarbonate.

Hexavalent chromium is not eliminated by this method and it must first be reduced into trivalent chromium.

Only selective adsorption permits retention of hexavalent chromium.

#### **III. 2-4.1 Iron oxihydroxide**

These adsorbents have been used for chromium (44, 101, 126, 133, 242).

Iron oxihydroxides can retain 99% of hexavalent chromium. Regeneration is performed using sodium hydroxide (17).

Studies on adsorption competitions have shown that the presence of arsenic V reduces the adsorption of chromates (126).

#### **III. 2-4.2 Manganese dioxide**

At pH values of between 4 and 7, manganese dioxide is capable of oxidising chromium III into chromium VI (23, 24, 79, 128, 283) and of selectively retaining chromium VI.

### ***III. 2-5 Elimination of phosphates by selective adsorption***

Phosphates behave in a similar way to arseniates (As V). The elimination methods are completely identical: precipitation-coprecipitation with iron or aluminium salts or selective adsorption on iron, aluminium or manganese oxides or oxihydroxides.

Iron and aluminium oxides or oxihydroxides have been very widely used (16, 249).

For synthetic solutions, elimination rates are more than 90% for water during potabilisation. For waste water, the yields are in the region of 80% and 70%.

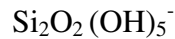
Studies on the adsorption of phosphates on iron oxides or oxihydroxides have been carried out at the time of addition of orthophosphates to mains water in order to reduce the corrosivity potential of water with respect to lead.

In corroded systems, it has been shown that the addition of phosphates led to a reduction in the quantity of free bacteria in the water (9).

The explanation put forward is that phosphates are adsorbed on iron oxide deposits and change the surface polarity (150, 217), modifying the adherence of bacteria and therefore reducing biofilms (9, 10, 151, 188, 217, 241).

### ***III. 2-6 Elimination of silicates by selective adsorption***

Silica is omnipresent in water; in surface water, the silica content ranges between 1 and 20 mg/L and in groundwater, between 7 and 45 mg/l. It was long believed that silica was found in the form of monosilicic acid, but recent NMR studies have shown it to be balanced with the dimer (63).



The monomer, dimer and polymer proportions are dependent on the pH of the water.

At the pH values found in natural water, the dimer corresponds to 99% of polymerised silica.

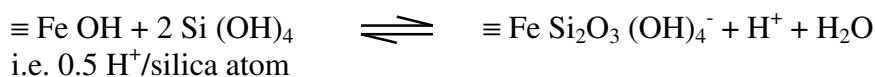
Dimers are adsorbed very well on iron or aluminium oxides or oxihydroxides.

Polymerised silica has a very high affinity for iron or manganese oxides.

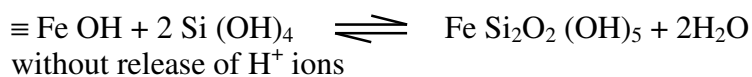
The polymer with 35 silica atoms is adsorbed 1,000,000 times more than the monomer (37, 232).

With iron oxides, we have:

- for low silica contents:



- for water with a high silica content:



Silica is used to protect iron against corrosion.

Robinson *et al.* [1992] (232) demonstrated that when silicate was added, the anti-corrosion effect was only detected if chlorine was added before the silica and that, otherwise, no effect was observed.

The explanation given is that chlorine oxidises divalent iron, producing trivalent oxides that adsorb silica, which limits corrosion.

By adding silica before chlorine, the divalent iron reacts with the former to create a soluble ferrous silicate, which is then oxidised with chlorine to produce a soluble ferric silicate.

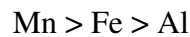
### ***III. 2-7 Elimination of sulphates by selective adsorption***

Meeussen *et al.* [1999] (186) use sand coated with goethite to eliminate sulphates.

## **IV. SELECTIVE ADSORPTION OF ORGANIC MATTER**

Iron, aluminium and manganese oxides and oxihydroxides have been used to retain organic matter. Of the organic matter present in water, it is humic acids which react the best (43, 96, 182, 280).

Manganese oxides give very good results:



Oxide or oxihydroxide-coated sands give better results than new uncoated sands (182, 286).

## V. ELIMINATION BY SELECTIVE ADSORPTION OF MICROORGANISMS

In the study of the adsorption of phosphates on iron oxides, we have already cited their effects on biofilms.

Negatively charged microorganisms are adsorbed on positive iron oxides (9, 233). After the addition of phosphate, the positive sites are saturated and the deposits have negative charges, no longer permitting adhesion of microorganisms and the development of the biofilm. The result is a reduction in the number of bacteria in the water. (10, 151, 188, 241). Le Chevalier et al. [1993] (152) demonstrated that bacteria adsorbed on iron oxides were more resistant to the biocidal action of chlorine or chloramines.

Sands coated with iron or aluminium oxihydroxide have been used to retain microorganisms (2, 48, 77, 164, 165, 292).

Lukasik et al. [1999] (164) give reduction levels of 1 to 2 logs for *Escherichia coli*, *Vibrio cholerae* Poliovirus I and bacteriophage MS2.

In some studies, iron oxides give better results than aluminium oxides for bacteria and viruses (2). Whilst Walker and Montemagno [1999] (290) demonstrate the superiority of aluminium oxihydroxides for the retention of *Cryptosporidium* oocysts.

Mansoor and Chaudhuri [1996] (176) used sands coated with iron oxihydroxide to eliminate viruses present in drinking water.

## VI. OTHER SELECTIVE ADSORPTION MEDIA

### VI.1 Sand coated with calcium carbonate and other carbonate materials

Aulenbach and Meisheng [1998] (13) used sand coated with  $\text{CaCO}_3$  to eliminate phosphates from water.

Analysis by X ray diffraction demonstrated the presence of  $\text{CaCO}_3$ ,  $\text{CaHPO}_4$ ,  $\text{CaH}_2\text{P}_2\text{O}_7$ .

Calcium contained in cements, especially  $\text{Ca}(\text{OH})_2$ , is capable of retaining one arsenic atom per calcium atom, with the formation of  $\text{CaAsO}_2\text{OH}$  (266).

Cement also eliminates calcium from water due to an increase in pH and precipitation of  $\text{CaCO}_3$ , particularly in the first days of use of constructions.

The same is true with phosphates, for which, at alkaline pH, there is the formation of apatite  $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ .

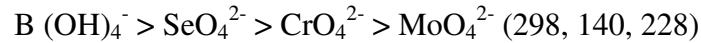
USEPA recommends cement to trap certain toxins before discharge into the environment (146).

Palfy et al. [1999] (213) particularly recommend using cement to eliminate arsenic.

### VI.2 Use of hydrocalumite and ettringite as a selective adsorbent

These products are obtained by alkanisation with lime of airborne ash from incineration plants.

Hydrocalumite  $\text{Ca}_4\text{Al}_2(\text{OH})_{12}(\text{OH})_2 - 6\text{H}_2\text{O}$  and ettringite  $\text{Ca}_6\text{Al}_2(\text{OH})_{12}(\text{SO}_4)_3 - 26\text{H}_2\text{O}$  precipitate and form solids with a high adsorbent capacity (298). Ettringite can form in cracked cement crossed by water. Hydrocalumite and ettringite, which are capable of retaining B, Se, Cr, Mo in alkaline medium, can also be formed by hydration of Portland Cements (277).



Myneni *et al.* (1997) studied the selective adsorption of arsenic (201). For all these adsorptions, the pH values must be high and greater than 10.5.

This type of treatment can therefore only be used on industrial effluents.

### VI.3 Apatite

Apatite  $\text{Ca}_5(\text{PO}_4)_3\text{OH}$  is a very good material for selective adsorptions (32).

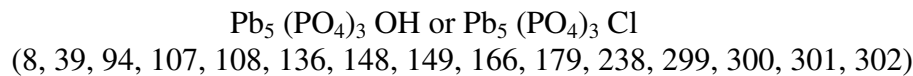
It is often obtained from calcined bonemeal (110, 173) or natural minerals.

In many cases, this product is used to decontaminate soil.

Apatite can fulfil three main roles.

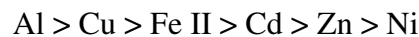
① Cation exchanger from calcium:

the main publications concern lead, with the formation of hydroypyromorphite or chloropyromorphite.

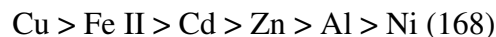


Studies have shown that apatite could eliminate lead by selective adsorption but some metals could interfere. These interferences were dependent on the lead content of the solution.

For high lead contents, the interference is as follows:



whereas for low lead concentrations, we have:

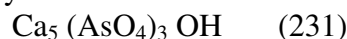


Numerous studies mention adsorption on apatite of other mineral micropollutants:

Eu (136) - Cd (49, 60, 94) - Cu (49) - Zn (49, 94, 224) - U (11, 86) - Cr (296) - Ni (250).

② Oxianion exchanger from the phosphate group

Arsenic is the most widely studied element.



Once again, the adsorption of other oxianions is reported:

- antimony (155)
- chromium (296)
- phosphates themselves (118).

③ Anion exchanger from the hydroxyl group

Fluorine is the most widely used compound (fluorapatite):



$\text{Ca}_5(\text{PO}_4)_3\text{F}$  (167, 299).

Other halogenides can also be adsorbed: Br, Cl.

#### **VI.4 Activated carbon impregnated with sulphide**

Activated carbon is impregnated with hydrogen sulphide acid. The sulphides bound to the activated carbon selectively adsorb the toxic metals by the formation of sulphide.

This has been applied to nickel and beryllium (158, 159).

Hydrogen sulphide is used to regenerate the activated carbon.

#### **CONCLUSION**

The beneficial effects of sands coated with metal oxide or oxihydroxide have long been known. The formation of these deposits was spontaneous.

It was only around ten years ago that the adsorption mechanisms were revealed and properly studied, leading to the preparation of these new materials.

This enthusiasm for the use of these filtration media is becoming increasingly widespread with:

- the setting of quality limits for mineral toxins in natural mineral waters. In fact, in this type of water, mineralisation must not change after treatment and only selective adsorption systems can therefore be used.
- the quality limits for mineral toxins are increasingly strict and cannot be achieved using conventional chemical precipitation-coprecipitation treatments.
- the low cost of these materials worldwide.
- filtration operations are stages which demand less technical knowledge on the part of personnel than that required for chemical precipitation-coprecipitation operations since they involve passive systems in which human error is not a potential problem.

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