

## Coated sand filtration: an emerging technology for water treatment

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### ABSTRACT

The use of oxide coated sand filtration as an alternative to current costly methods for water treatment has been reviewed. The paper highlights the importance of iron, aluminium and manganese oxides, including sand coated with these oxides, for removal of heavy metals and other impurities from water. The literature review showed that oxide coated sand is able to remove heavy metals (both anions and cations) and other impurities (e.g. natural organic matter, microorganisms, sulphate, manganese, etc.) from surface water or groundwater. In this review an extensive list of the use of oxide (iron, aluminium and manganese) coated sand in water treatment by adsorption and filtration processes has been compiled to provide a summary of available information. The information available on the capacity of coated sand for pollutant uptake is very scarce, however some data on the capacity of coated sand for heavy metals uptake have been compiled and presented. The coated sand could be an alternative emerging technology for water treatment without any side effects or treatment process alterations.

**Key words** | adsorption, coated sand, filtration technology, heavy metals, oxides, water

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### INTRODUCTION

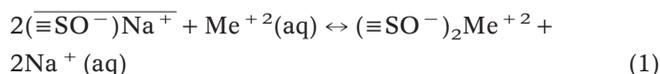
Drinking water resources are faced with an unprecedented risk of contamination, either due to leaching of metals from underground minerals, or the release of large quantities of industrial effluent throughout the world. At present about 20% of the world's population lacks safe drinking water according to UNEP (1999). At the end of 2000 the UN General Assembly declared that 2003 is to be the UN International Year of Freshwater. In fact, without action the world is heading for a water crisis: two-thirds of the world's population could well be living in water-stressed areas by 2025, double the present number (Hayward 2001).

The removal of dissolved impurities in water treatment has gained greater importance due to tightening of water quality standards and the recognition that these impurities may become transport vehicles for undesirable organic and inorganic contaminants, taste, odour and colour-causing compounds, and potentially disease-causing microbial contaminants. The

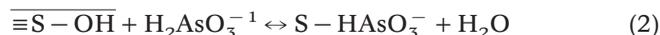
conventional process for treating surface water supplies involves filtration with a stratified bed of graded gravel and sand. In this process the filter grains are usually negatively charged and coagulant dosage is often sufficient to decrease, but not completely neutralize, the negatively charged incoming particles. In such a situation, use of a filtration medium that is neutral or positively charged should lead to more favourable electrostatic medium-particle interactions and possibly to reduce coagulant requirements, better particle removal and more rapid ripening. Furthermore, because the surface charge distribution of influent suspended particles might be quite broad, particle deposition might improve if the surface of the media had both negatively and positively charged regions. Several studies have examined the possibility of modifying filtration particulate media to improve their ability to remove microorganisms and dissolved impurities. These modifications include coating of metallic flocs onto surfaces (Edwards & Benjamin 1989)

and incorporation of metallic peroxides into microporous filters (Gerba *et al.* 1988).

Aluminium and iron oxides are abundant in the natural environment and both have relatively high surface area and variable surface charge. The iron oxide has stronger interactions with dissolved organic matter, iron (Fe), and aluminium (Al) species than does silica and these specific interactions might improve removal of both dissolved contaminants and particles that have organics or coagulant (e.g. polymeric Fe or Al species) adsorbed on their surfaces. Ferrihydrite has been effectively used for the removal of metals, including arsenic, from water and wastewaters (Pierce & Moore 1982). Both electrostatic (i.e. ion exchange) and Lewis acid-base (i.e. metal ligand) interactions are the underlying reasons for the high affinity of heavy metals to ferrihydrites over alkali- and alkali-earth metal cations and anions. In a simplistic way, the ion-exchange type sorption of dissolved heavy metals ( $\text{Me}^{+2}$ ) onto ferrihydrite in an aqueous medium may be presented as follows:



whereas ligand exchange sorption of dissolved anions onto ferrihydrite may be expressed, taking the example of the arsenite ion, as:



where the over-bar denotes the solid phase and  $\overline{\equiv\text{SO}^-}/\overline{\equiv\text{S}-\text{OH}}$  denotes the deprotonated surface site in ferrihydrite. The oxygen atom (Lewis base) may donate a lone pair of electrons to a transition metal ion (Lewis acid). Understandably, ferrihydrites or iron oxyhydroxides would be an attractive sorbent for metal removal because they are likely to be much less expensive compared to polymeric chelating exchangers or activated carbon often used for the same purpose (Clifford 1990).

Redox-sensitive manganese and iron hydroxides under oxidising conditions constitute significant sinks of arsenic in aquatic systems (Mok & Wai 1994). Manganese dioxides have been shown to be relatively strong oxidants in the environments, controlling the mobility (and

toxicity) of arsenic in natural waters. But the iron and manganese oxides alone are not suitable as a filter medium because of their low hydraulic conductivity (Theis *et al.* 1992). Therefore, sorptive sand filtration using Fe, Al and Mn oxide coated sand is a relatively new approach for treating drinking water contaminated with toxic metals and other dissolved impurities and would be an effective medium for filtration.

The coating of Fe oxides onto the surface of sand to effectively remove/recover trace heavy metals has been used by many researchers (Edwards & Benjamin 1989; Bailey *et al.* 1992). The coated sand has advantages, e.g. coating chemicals are quite cheap and readily available in all parts of world. Therefore, in water treatment, adsorptive filtration is an essential step in the production of high quality water by depositing suspended solids within or on the medium. This paper describes the advantages of using different versions of oxide coated sand for the removal of various dissolved impurities including heavy metals normally found in surface water/groundwater, and gives some theoretical background on the development of oxide coating on sand.

## THEORETICAL BACKGROUND

### Development of oxide coating on sand

Iron is the best metallic material for environmental remediation because it is a strong reducer, is non-toxic and inexpensive. The Fe oxide has been shown to be an extremely flexible ion-exchange medium, applicable to both anions and cations. In some cases it is even possible to remove anions and cations simultaneously in a single treatment. Despite the high affinity of Fe oxide for metals, it has not been widely used as an ion-exchange/adsorbent medium because its physical properties (it is a bulky, hydrated, amorphous, flocculent material with extremely low hydraulic conductivity) are not conducive to such a process.

The natural greensand effect (NGE) process has also gained acceptance for use in water treatment plants due to its enhanced  $\text{Mn}^{+2}$  removal capacities (AWWA 1994).

The NGE process is 'natural' in that the mineral deposits develop *in situ* within weeks to a month on new, uncoated filter media during normal filter operation. The developed greensand is a granular material composed of the mineral glauconite, which has been coated with manganese oxide. It is a natural zeolite, and has strong ion exchange properties, and will remove iron, manganese, arsenic, sulphide, and many other anions. Like manganese dioxide coated sand, the surface of greensand is strongly oxidizing, and is thus able to remove both arsenite and arsenate. The greensand has its own limitations, i.e. poor natural coating and frequent regeneration to maintain effectiveness.

A few approaches have been developed in the past to overcome these problems. The development of granular form (oxide coated sand) media was an effort to address these problems. For the last two decades the work on oxide (iron, manganese, aluminium) coated sand has drawn the attention of researchers. They have performed a number of studies on the removal of various metals and other pollutants using oxide coated sand media (Edwards & Benjamin 1989; Joshi & Chaudhuri 1996; Kuan *et al.* 1998; Lukasik *et al.* 1999).

The design advantages of the granular forms of iron/manganese/aluminium oxides have been recognized for some time. Griffin (1960) noted that efficient manganese removal did not occur during treatment until the sand grains in the filter had turned black as a result of being coated with deposited manganese oxide. This process was referred to as 'filter ageing'. Dixon (1984) has suggested the use of naturally granular magnetite particles for water treatment. Wangen *et al.* (1982) have put forward a different approach to coat relatively inert quartz grains with iron oxide. They were able to achieve this through rotary evaporation of a sand-ferric chloride mixture. However, the physical integrity of the oxide coating could not be sustained during normal use. Edwards & Benjamin (1989) developed a technique for applying a coating to sand grains with an Fe oxide layer that was stable when exposed to mild abrasion typically encountered during filter backwashing. They used ferric nitrate as the source of iron, resulting in a final product that contained 1–2% iron and that could be used and regenerated in a packed bed design.

Bailey *et al.* (1992) developed iron oxide coated sand (IOCS) by increasing the pH of the solution to 11.0 with

NaOH before heating. Szecsody *et al.* (1994) developed coated sand by ageing the acid washed silica sand with iron oxide gel. Stenkemp & Benjamin (1994) prepared coated sand by adding 1.2 ml 10 M NaOH and 80 ml 2.7 M ferric nitrate solution and heating in an oven at 110°C for 14–16 h. Benjamin *et al.* (1996) modified an earlier procedure of iron coating by applying ferric chloride in a strong acidic medium and subsequently heating it in an oven at 550°C for 3 h. This medium was called high temperature medium (HTM), but they concluded that media thus developed have not shown much affinity for anions/cations. Later they modified this medium using ferric nitrate and NaOH and heating at 110°C for 10–14 h. They observed that media developed at later stages were more efficient than HTM. Merkle *et al.* (1997) have also developed many methods using manganese oxide coating on the sand for the removal of manganese from drinking water. Bajpai & Chaudhuri (1999) prepared manganese dioxide coated sand (MDCS) by using  $\text{MnCl}_2$ ,  $\text{KMnO}_4$  and NaOH and heating at 105°C for 48 h for removal of arsenic from groundwater. Lukasik *et al.* (1999) developed coated sand by soaking the sand in a solution of ferric and aluminium chlorides. Lai *et al.* (2000) developed iron-coated sand using ferric nitrate and NaOH at pH  $9.5 \pm 0.1$  after heating in an oven at  $50 \pm 1^\circ\text{C}$  for 96 h. During the first 48 h, the mixture was stirred using a mixer. This iron coated sand medium has shown greater affinity for the removal of lead than copper ions. Vaishya & Gupta (2001a) have developed a mixed oxide coated sand using  $\text{KMnO}_4$ ,  $\text{MnCl}_2$  and ferric nitrate [ $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ] followed by heating in an oven at 60°C for 60 h for removal of arsenic (III) from synthetic water. Kuan *et al.* (1998) have developed aluminium oxide coated sand (AOCS) for selenium removal from water by using  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$  and NaOH followed by heating at 70°C for a day.

There are a number of pollutants which can be removed from drinking water by oxide coated sand either by sorptive filtration or by batch studies. The following section describes various uses of oxide coated sand during batch and column studies for the removal of pollutants. These texts are not exhaustive but every effort has been made to collect and present available information in the literature.

## Removal of heavy metals

The use of Fe-impregnated activated carbon has been studied by Reed *et al.* (2000) for the removal of As(III), As(V), Hg and Pb from drinking water. However, in terms of cost, sand is cheaper and more easily available as compared with activated carbon. Recently Benjamin *et al.* (1996) have used IOCS for removal of heavy metals (i.e. Cu, Zn, Pb), including arsenic, from water. Joshi & Chaudhuri (1996) have used the same process for oxide coating on sand as used by Bailey *et al.* (1992) for arsenic removal from groundwater. Kuan *et al.* (1998) developed aluminium oxide coated sand (AOCS) for the removal of Se(IV) and Se(VI) from water. Bajpai & Chaudhuri (1999) used manganese dioxide coated sand (MDCS) for arsenic removal from groundwater. Khaodhiar *et al.* (2000) used iron oxide coated sand (IOCS) for the removal of copper, chromium and arsenic by an adsorption process. Satpathy & Chaudhuri (1995) have utilized IOCS for the removal of cadmium and chromium from cadmium and chromium plating waste under laboratory conditions. Khaodhiar *et al.* (2000) used IOCS as developed by Edwards & Benjamin (1989) for the removal of copper, chromium and arsenic by an adsorption process. Arsenate adsorption was not affected by the presence of copper and chromate. The presence of arsenate significantly decreased chromate adsorption due to competition for adsorption sites and electrostatic effects. Lai (1994) has reported that iron coated sand can be applied to the conventional rapid filtration process to remove copper ions from water. Lai *et al.* (2000) have used iron coated sand for the removal of lead and copper ions from water.

Vaishya & Gupta (2001b) used iron oxide coated sand (IOCS) for the removal of As(III) during batch studies. They reported that the adsorption of As(III) on IOCS was pH dependent, and the sequence of anions studied competing with respect to the arsenite ion was in the order of  $\text{SO}_4^{2-} > \text{HCO}_3^- > \text{Cl}^-$ . The presence of  $\text{Ca}^{+2}$  ions in the solute significantly improves the removal efficiency. The sorption data were modelled well by the Langmuir and Freundlich isotherms. Thirunavukkarasu *et al.* (2001) have also used iron oxide coated sand and ferrihydrite (FH) in batch studies for the removal of arsenic from drinking water. They obtained approximately 90% removal for an initial As(III) concentration of 325  $\mu\text{g/l}$ . The adsorption

capacity of the IOCS and ferrihydrite used in this study for arsenic was 18.3 and 285  $\mu\text{g/g}$ , respectively.

Kuan *et al.* (1998) used aluminium oxide coated sand (AOCS) for selenium removal from water. Bajpai & Chaudhuri (1999) prepared manganese dioxide coated sand (MDCS) for arsenic removal from groundwater. Hansen *et al.* (2001) used iron oxide coated sand (IOCS) to adsorb strontium from synthetic wastes. They concluded that IOCS can selectively remove strontium from solutions, even when the solutions contain overwhelmingly higher concentrations of potentially competing cations such as  $\text{Ca}^{+2}$ ,  $\text{Al}^{+3}$  and  $\text{Cr}^{+3}$ . Aulenbach & Meisheng (1988) reported that  $\text{CaCO}_3$  coated sand plays a significant role in phosphorus removal, whereas removal of this coating nearly eliminated phosphorus removal. The X-ray diffraction studies showed the presence of  $\text{CaCO}_3$ ,  $\text{CaHPO}_4$ ,  $\text{CaH}_2\text{P}_2\text{O}_7$ , and  $\text{AlPO}_4$  in the coating on the sand. The removal of phosphorus was primarily a physical and chemical process. Szecsody *et al.* (1994) also developed coated sand for the removal of  $\text{Co}^{+2}$  and  $\text{Co}^{+2}$ -EDTA complexes. Ionic  $\text{Co}^{+2}$  exhibited typical cation-like adsorption whereas  $\text{Co}^{+2}$ -EDTA adsorption was ligand-like, increasing with decreasing pH. Adsorption isotherms for all reactive species exhibited Langmuir behaviour, with site saturation occurring at molar values of  $< 0.5\%$  of  $\text{Fe}^{\text{tot}}$ . The dissolution of  $\text{Co}^{+2}$ -EDTA showed that ligand-promoted dissolution was more consistent with the experimental data.

IOCS has been used in laboratory scale studies for metal removal since 1989, and comparisons of uptake potentials for various coated sand media are presented in Table 1. In the literature, sorption potentials have been expressed either in the form of Langmuir monolayer sorption capacity,  $Q_o$  (mg/g), or Freundlich sorption capacity,  $K_F$ , or the ultimate adsorption,  $q$  (mg/g), value can be computed based on  $q$  vs time or  $q$  vs pH graphs. In some references this capacity is expressed in terms of percentage removal for fixed solute concentrations with respect to fixed sorbent doses. Thus, a universal comparison is not possible. However, these data could be used for a qualitative assessment.

Since sorption depends heavily on experimental conditions such as pH, metal concentrations, ligand concentration, competing ions and particle size, this literature

**Table 1** | Uptake of heavy metals onto some reported coated sand media

Coated sand	Sorbate	Uptake capacity (mg/g)	Reference	Remark
MDCS	As(III)	0.068	Bajpai & Chaudhuri (1999)	~At pH = 7.5
	As(V)	0.078	Bajpai & Chaudhuri (1999)	~At pH = 7.5
IOCS	As(III)	0.076	Joshi & Chaudhuri (1996)	~At pH = 7.5
	As(V)	0.083	Joshi & Chaudhuri (1996)	~At pH = 7.5
AOCS	Se(IV)	0.84	Kuan <i>et al.</i> (1998)	~At pH = 7.5
	Se(VI)	0.44	Kuan <i>et al.</i> (1998)	
IOCS	Cu(II)	1.271	Lai <i>et al.</i> (2000)	~At pH = 6.0
	Pb	4.14	Lai <i>et al.</i> (2000)	
IOCS	Cu(II)	0.032	Khaodhiar <i>et al.</i> (2000)	~At pH = 7.5
	As(V)	0.506	Khaodhiar <i>et al.</i> (2000)	~At pH = 7.5
IOCS	Pb	0.083	Theis <i>et al.</i> (1992)	~At pH = 7.0

review has not addressed specific test conditions. The authors recommend that reported sorption capacities be taken as an example of values that can be achieved under a specific set of conditions rather than maximum sorption capacities. The reader is strongly encouraged to refer to the original articles for information on experimental conditions.

### Removal of microorganisms

Lukasik *et al.* (1999) used coated sand for the removal of microorganisms from drinking water. Using 1.0 kg of this medium in the column for the removal of microorganisms, they removed greater than 99% of *E. coli*, *Vibrio cholerae*, poliovirus and coliphage MS-2 from dechlorinated tap water. Neither aluminium nor ferric ions were detected in the column effluents over a long period of time. This showed that the metal hydroxide coating was insoluble. Mansoor & Chaudhuri (1996) also used ferric and aluminium oxides coated sand for virus removal from drinking water.

### Removal of natural organic matter

The removal of natural organic matter (NOM) from water is of utmost importance as its presence increases the rate of growth of disease-causing microorganisms. McMeen & Benjamin (1997) compared iron oxide coated olivine (sand) (IOCO) with uncoated olivine and plain sand as a medium for slow sand filtration. The results of their 16-month pilot study showed that NOM removal was better than with uncoated olivine sand medium. Also, the NOM removal capacity of the IOCO medium was not exhausted in a manner similar to rapid rate filtration. The increased NOM removal was credited to enhanced bioactivity in the IOCO medium.

### Removal of sulphate

Meeussen *et al.* (1999) used a goethite coated quartz sand column for the removal of sulphate. Based on the results of modelling studies they concluded that transport behaviour of sulphate in a goethite system is very sensitive to

relatively small amounts of adsorbed sulphate initially present.

### Removal of $Mn^{+2}$

Knocke *et al.* (1991) removed Mn(II) with oxide coated filters. The kinetics of Mn(II) sorption by  $MnO_x(s)$ -coated filter media were a function of solution pH,  $MnO_x(s)$  surface concentration, and free chlorine concentration of filter applied water. In absence of free chlorine, Mn(II) removal was by adsorption only; no evidence of auto-oxidation of Mn(II) was noted. Merkle *et al.* (1997) also used synthetic manganese oxide coated anthracite and sand filter media for the removal of  $Mn^{+2}$  from drinking water and hazardous effluents by filtration.

### Disposal of metal-loaded sand

The excessive presence of Ca, especially of  $Ca(OH)_2$ , in cement is capable of immobilizing almost one atom of As(III) per Ca atom by formation of the sparingly soluble phase  $CaAsO_2OH$ . Since cements are also very efficient at removing dissolved carbonate as  $CaCO_3$ , enhanced solubilization of As from cement-conditioned materials is unlikely to be significant until sufficient carbonate has been added to neutralize the intrinsic high pH of the cement resulting from  $Ca(OH)_2$  and Ca-rich C-S-H (Stronach *et al.* 1997). Therefore, As(III)-laden sand can be used as a normal building material, especially in superstructure or in heavy engineering works, to dispose of it as cementitious solidification. The US EPA also recognizes cementitious solidification as 'the best demonstrated available technology' (BDAT) for land disposal of most toxic elements (Landrett 1986). Palfy *et al.* (1999) also reported that embedding of calcium and ferric arsenates/arsenites in the cement matrix could reduce the original arsenic solubility of 6430 mg/l from untreated waste to 0.823 mg/l for solidified waste.

To meet WHO (1993) guidelines for drinking water, the coated sand meets almost all selection criteria for an emerging technology (ET). To achieve the status of an innovative technology and ultimately become an available alternative technology (AAT), more pilot plants and field

studies are required. However, water treatment by coated sand has not yet received adequate attention despite a few good studies in this area so far. Therefore concerted efforts are yet to be made to explore the possibility of employing the coated sands for the treatment of contaminated water in small and medium treatment plants. It is necessary to explore cost effective alternative technologies or sorbents for the removal of metals and other impurities from contaminated water by sorptive filtration techniques. The cost is an important parameter for comparing the sorbent materials. However, information on cost is seldom reported, and the expense in preparation of individual sorbents varies depending on the degree of processing required and local availability.

### CONCLUSIONS

It is evident from the above discussion that oxide coated sand has many advantages for water treatment especially in removal of dissolved impurities. The results of pilot scale and laboratory studies are positive for its further use. Also, some selected sand pre-coating processes are quite simple and can be prepared economically for commercial operations. The removal of cations and anions can take place simultaneously at neutral pH. It has also been observed that coated sand can withstand mild abrasion during filter backwashing. The coated sand fulfils almost all required conditions of a prospective emerging technology for water treatment. Thus inexpensive, effective, easily prepared materials like coated sand may be an alternative to activated carbon or ion exchange resins. Due to robustness and convenience in preparation, oxide coated sand media may become viable and cost effective alternatives as filter media for treatment of contaminated drinking water. Coated sand has the advantage of local availability with little processing being required. Quartz sand, which is a by-product of weathering of rock, and coating chemicals are cheaply and abundantly available all over the world. Therefore this innovative medium may become popular in future for the treatment of drinking water on a cost comparison basis.

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