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## **CONTAMINANTS REMOVAL BY BENTONITE AMENDED SLOW SAND FILTER**

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*Earlier studies have indicated that variability in size, surface texture and charge greatly influence the contaminant removal process in granular media. Based on surface characteristics of montmorillonite, it is anticipated that small addition of this clay would increase adhesion sites for bacterial growth and extracellular polymer production in slow sand filter and thereby enhance its contaminant removal ability. Experiments were performed by permeating groundwater contaminated with pathogens (total coliform and E. Coli) and inorganic contaminants through bentonite amended slow sand filter (BASSF). Surprisingly, the BASSF retained inorganic contaminants besides pathogens. Water-leach tests (pH of water leachate ranged from 2 to 9) with spent BASSF specimen indicated that the inorganic contaminants are irreversibly adsorbed to a large extent. It is considered that the combined effects of enhanced - organic matter mediated adhesion sites and increased hydraulic retention time enables the BASSF specimen to retain inorganic contaminants. It is envisaged that BASSF filters could find use in treating contaminated groundwater for potable needs at household and community level.*

**Keywords:** environment, granular materials, pollution.

### **Introduction**

Slow sand filtration is one of the earliest forms of biological filtration process [1]. Slow sand filtration reduces bacteria, cloudiness, and organic level. The effective size of sand ( $D_{10}$ ) used in slow sand filter lie in the range of 0.15 – 0.35 mm and flow rates range between 0.1 – 0.4 m/h [2]. The contaminant removal by slow sand filter (SSF) is attributed to straining through the filter skin (schmutzdecke) developed at the top few mm of sand, together with slow filtration rate promoted by the fine size of sand [2 – 3].

Stevik et al. [4] had reviewed factors affecting retention of bacteria in porous media. Besides biological and physical straining, adsorption is also considered to play an important role in immobilizing pathogens transported through the porous media. Further, variability in sizes, surface texture and charge of porous media are considered to greatly influence the contaminant adhesion process. Smaller particle sizes expose a larger surface area compared

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to coarse particles and hence provide greater adhesion sites for bacteria removal [5 – 6]. Huysman and Verstraete [7] and Fletcher and Loeb [8] have observed that presence of clay particles in soil media enhances the adhesion of bacteria due to the charge and surface characteristics of the clay minerals. It has been indicated that extracellular polymer production often accompanies bacteria attachment and the polymer mediated interactions can play a role in contaminant removal [4].

Montmorillonite clay exposes large surface area (800 m<sup>2</sup>/g), has very small particle size (< 2 microns) and exposes charged particle surface arising from isomorphous substitution within the clay lattice and presence of broken bonds at particle edges [9]. Based on surface characteristics of montmorillonite, it is anticipated that small addition of this clay would increase adhesion sites in slow sand filter for bacterial growth and extracellular polymer production and enhance the filter's contaminant removal ability. Experiments were hence performed that involved permeation of ground water contaminated with pathogens (total coliform and *E. coli*) and inorganic contaminants through bentonite amended (bentonite content = 10 %) slow sand filter. Results of permeating nitrate spiked solutions through SSF and bentonite amended slow sand filter (BASSF) specimens are also presented.

## Experimental

Naturally occurring bentonite from Rajasthan, India and local river sand (from Karnataka) were used in the construction of BASSF. Bentonite contains montmorillonite as primary clay mineral [10]. The index properties of bentonite and sand are provided in Table 1. The bentonite sample has liquid limit of 184 %, plastic limit of 31 % and contains 48 % clay sized (particle size < 2 micron) fraction. The sand sample is composed of 71.5 % medium fraction (particle size 2 – 0.425 mm) and 28.5 % fine fraction (0.425 – 0.075 mm). The D<sub>10</sub> for the sand sample is 0.1 mm, which is slightly smaller than lower range (0.15 mm) of effective particle size recommended for slow sand filters. To examine the influence of clay addition on the contaminant retention by slow sand filter, sand-bentonite mix composed of 90 % sand and 10 % bentonite (on dry mass basis) was prepared. Table 2 presents the composition of the contaminated groundwater sample from Kolar District in Karnataka that was used to test the efficacy of the BASSF specimen. The groundwater sample shows pathogen and nitrate contamination and was obtained from peri-urban community that disposes human waste using pit toilet system.

Table 1. Index properties of materials

Sample/Property	Value
Bentonite	
Liquid limit (%)	184
Plastic limit (%)	31
Specific gravity	2.67
% Clay size fraction (< 0.002 mm)	48
% Silt fraction (0.075 mm – 0.002 mm)	52
Sand	
% Coarse fraction (4.75 – 2.0 mm)	0
% Medium fraction (2.0 – 0.425 mm)	71.5
% Fine fraction (0.425 – 0.075 mm)	28.5
Specific Gravity	2.7
90 % Sand + 10 % Bentonite	
Liquid limit (%)	45
Plastic limit (%)	NP (non-plastic)
Specific gravity	2.7
Standard Proctor compaction properties	1.78
Maximum dry density (g/cc) Optimum moisture content (%)	18

Table 2. Composition of contaminated groundwater sample

Parameter	Value
Total coliform (MPN/100 ml)	3200
<i>E. Coli</i> (MPN/100 ml)	600
pH	7.65
Electrical Conductivity (milli-Siemen/cm)	2.17
Total dissolved solids (mg/L)	1409
Calcium (mg/L)	121
Magnesium (mg/L)	52
Potassium (mg/L)	97
Sodium (mg/L)	204
Chloride (mg/L)	388
Nitrate (mg/L)	288
Sulphate (mg/L)	97
Bicarbonate (mg/L)	567

**Permeability.** Permeability controls the rate of flow of water through a porous medium. It is known that addition of clay content to coarse aggregate reduces the permeability of the mix [10]. Therefore a small (10 %) amount of bentonite was added to sand while preparing the BASSF. Constant head permeability test was performed with relatively loosely compacted 90 % sand –10% bentonite specimen as per IS 2720 [11]. The specimens were statically compacted to dry density of 1.3 g/cc at 22 % water content. The dry density of the specimen was 0.27 times lower than the Standard Proctor maximum dry density (MDD) value of the mix (MDD = 1.78 g/cc). Likewise the compaction water content was 4 % higher than the optimum moisture content (OMC) value (OMC = 18 %) of the mix. The specimens were compacted in excess of OMC to ensure saturation of voids. A small hydraulic gradient ( $i = 2$ ) was used in the constant head test. Distilled water was used as the permeating fluid. The sample height and diameter corresponded to 40 mm and 82 mm respectively. The compacted specimen exhibited equilibrium coefficient of permeability of 0.0015 m/h. Using Darcy's equation [12] for saturated flow in porous media:

$$v = ki, \quad (1)$$

where  $v$  is the velocity of flow (m/h),  $k$  is the coefficient of permeability (0.0015 m/h) and  $i$  is the hydraulic gradient ( $i = 2$ ) gives flow velocity of 0.003m/h. The flow rate ( $v$ ) of BASSF is 100-fold smaller than that of slow sand filter (0.1 to 0.3 m/h).

**Contaminant removal by BASSF.** The thickness (height) and diameter of BASSF specimen corresponded to 40 mm and 82 mm respectively. The BASSF specimen was overlain by 15 mm sand layer to trap any suspended solids in the water sample. The specimen was permeated with the contaminated ground water (see Table 2) for 5 – 7 h daily between 10 AM and 7 PM. After the permeation period, the inletvalve of the constant head reservoir was closed. Approximately 2 – 3 mm of water layer was allowed to stand above the specimen during the non-permeation period to prevent drying. Permeation was resumed next morning. The outflow solution collected daily (500 – 700 ml over 5 – 7 h) was analysed for calcium, magnesium, sodium, potassium ions using ICP-OES facility and nitrate, sulphate, chloride ions by ion chromatograph and bicarbonates by automatic titrator. The outflow solutions were examined for total coliform and *E. coli* by multiple tube method [13 – 15]. Permeation of the contaminated water sample was terminated when the flow rate reduced from 100 ml/h to 40 ml/h (cumulative volume permeated = 9850 ml). The reduction in flow rate after passage of the contaminated water for 16 days is attributed to clogging of pores by organic matter.

**Tests with spent filter specimens.** After termination of permeation, the BASSF specimen was dismantled and thoroughly hand-remolded to obtain homogenous sample. This spent filter was stored in a desiccator in the moist state (gravimetric water content = 26 %). Moist samples were used in the water leach tests, while, oven-dried (dried at 60°C) samples were used in determination of organic matter. In the water leach tests, the pH of distilled water was adjusted between 2 and 9 using hydrochloric acid or ammonia solution. Each pH adjusted water sample (volume = 250 ml) was agitated with 12.6 g of moist filter sample (corresponding to dry mass of 10 g) for period of 18 h using mechanical flask shaker. After agitation, the solution was filtered and the filtrate was analysed for calcium, magnesium, sodium and potassium ions using ICP-OES, nitrate and sulphate ions using IC and bicarbonate ions using automatic titrator.

The organic matter (OM) of the spent filter was determined using the Walkley Black method [16]. Organic matter was also determined for the raw 90 % sand – 10 % bentonite mix to obtain base-line OM value for the filter. Polarizing microscope (Model Olympus BX 51) images of spent filter (BASSF and SSF) specimens (magnification = 10 to 20 x) were obtained to examine any growth of biofilms.

**Permeation of SSF and BASSF specimens with nitrate spiked solutions.** To examine the inorganic contaminant retention ability of SSF, an experiment was performed by intermittently permeating 320 mg/L of nitrate solution through SSF specimen. The sand sample used in the preparation of BASSF specimen was used in this experiment. The diameter and thickness of SSF specimen corresponded to 82 mm of 40 mm respectively. The moist (water content = 5 %) sand mass was compacted to dry density of 1.3 g/cc. Nitrate solution (concentration = 320 mg/L) was permeated through the SSF specimen at flow rate of 100 ml/h. The slow, flow rate was achieved by fixing a capillary nozzle to the reservoir outlet. The nitrate concentration used in the test is approximately 10 % larger than the nitrate concentration of the contaminated groundwater sample (nitrate concentration = 288 mg/L).

The experiment was terminated after passage of 10000 ml of nitrate solution as the experiment with BASSF specimen was terminated after passage of similar volume (9850 ml) of contaminated ground water sample. The SSF did not experience any decrease in flow rate after permeation of 10000 ml of nitrate solution.

An experiment was also performed to examine the response of the BASSF to passage of nitrate spiked solution (initial nitrate concentration = 298 mg/L). The experiment was terminated after passage of 3660 ml of nitrate solution as the flow rate decreased from an initial value of 70 ml/h to 28 ml/h. The differences in flow rates developed by the BASSF specimens on permeation

with nitrate solution (70 ml/h to 28 ml/h) and contaminated groundwater (100 ml/h to 40 ml/h) are attributed to their differences in dissolved salt concentrations. The dissolved salt concentration of nitrate spiked solution and contaminated groundwater correspond to 409 mg/L and 1409 mg/L respectively.

The higher dissolved salt concentration of the contaminated ground water reduces the thickness of diffuse ion layers around the clay particles facilitating increased void space for water flow thereby rendering the specimen more pervious with higher flow rate [17].

The outflowing solutions from SSF and BASSF tests were daily analysed for nitrate concentrations using ion-chromatograph. After permeation with nitrate spiked solutions was terminated, the SSF and BASSF specimens were dismantled. The spent specimens were thoroughly hand-remolded, dried at 60°C and used in OM determination by the Walkley Black method [16].

### Results and discussion

Fig. 1 illustrates the reduction in total coliform and *E. Coli* levels upon passage of the contaminated groundwater sample through BASSF. The total coliform (initial value 3200 MPN/100 ml) and *E. Coli* (initial value 160 MPN/100 ml) reduce to values ranging from 0 to 50 MPN/100 ml upon passage through BASSF.

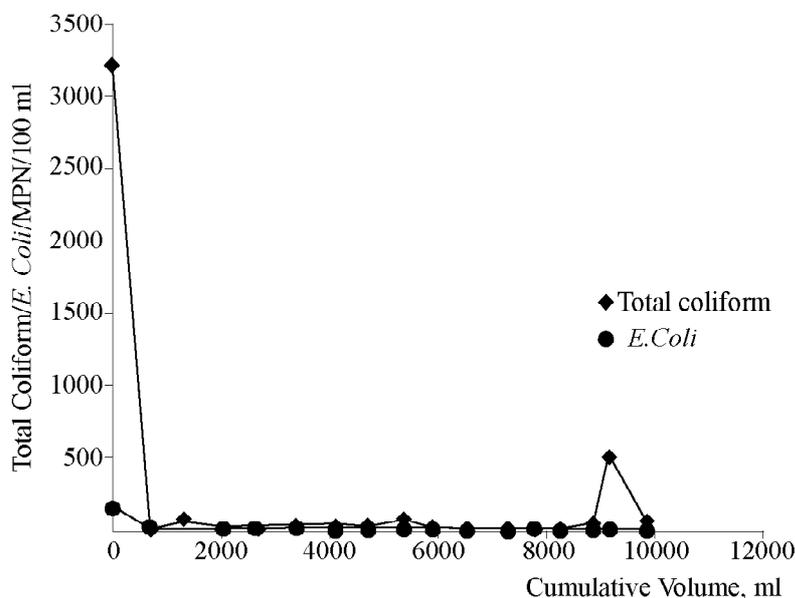


Fig. 1. Total coliform and *E. Coli* removal by BASSF.

2100 mg of sodium, 1192 mg of calcium, 954 mg of potassium and 510 mg of magnesium was permeated by the passage of 9850 ml of contaminated groundwater through BASSF (Table 3). At the end of permeation process, 895 mg of sodium (43 % retention), 738 mg of calcium (62 % retention), 275 mg of magnesium ions (54 % retention) and 567 mg of potassium (59 % retention) were retained by the BASSF (dry mass of BASSF = 275 g). Likewise, 5588 mg of bicarbonate, 3824 mg of chloride, 2838 mg of nitrate and 951 mg of sulfate was permeated by the passage of 9850 ml of contaminated groundwater through the BASSF. At the end of permeation process (see Table 3) 3151 mg of bicarbonate (56 % retention), 2525 mg of chloride (66 % retention), 1912 mg of nitrate ions (67 % retention) and 532 mg of sulfate (56 % retention) were retained by the BASSF (dry mass of BASSF = 275 g).

*Table 3. Ion retention by 275 g of BASSF and SSF Specimens*

Species	Mass permeated	Mass retained	% Retained
	mg		
BASSF Specimen permeated with contaminated ground water			
Sodium	2100	895	43
Calcium	1192	738	62
Magnesium	510	275	54
Potassium	954	567	59
Bicarbonate	5588	3151	56
Nitrate	2838	1912	67
Chloride	3824	2525	66
Sulfate	951	532	56
Nitrate	SSF Specimen permeated with nitrate spiked solution		
	3206	0	0
Nitrate	BASSF Specimen permeated with nitrate spiked solution		
	819	384	47

The results in Fig. 1 and Table 3 demonstrate that addition of bentonite to SSF causes it to retain cationic and anionic contaminants in addition to bacterial contaminants. Interestingly, the BASSF specimen was able to reduce the nitrate concentration of the groundwater sample from an initial value of 288 mg/L to an average value of 92 mg/L and of the nitrate spiked solution from 298 mg/L to an average value of 145 mg/L. In comparison, the SSF specimen was unable to remove nitrate even after the passage of 10 liters of nitrate spiked solution (see Table 3).

After completion of the permeability experiments, dismantling of the spent BASSF specimens did not reveal growth of green algal mass in as was observed by Campos et al. [18] in uncovered slow sand filter beds. The spent BASSF filter specimen (permeated with contaminated groundwater) is characterized by OM content of 4.2 %. Comparatively, raw (unexposed to contaminated groundwater) mixture of 90 % sand+10 % bentonite is characterized by organic matter content of 0.04 %. The spent SSF and BASSF specimens permeated with spiked nitrate solutions were characterized by OM content of 0.7 % and 3.4 % respectively. Lack of substantial OM growth in SSF is possibly responsible for its inability to retain nitrate ions.

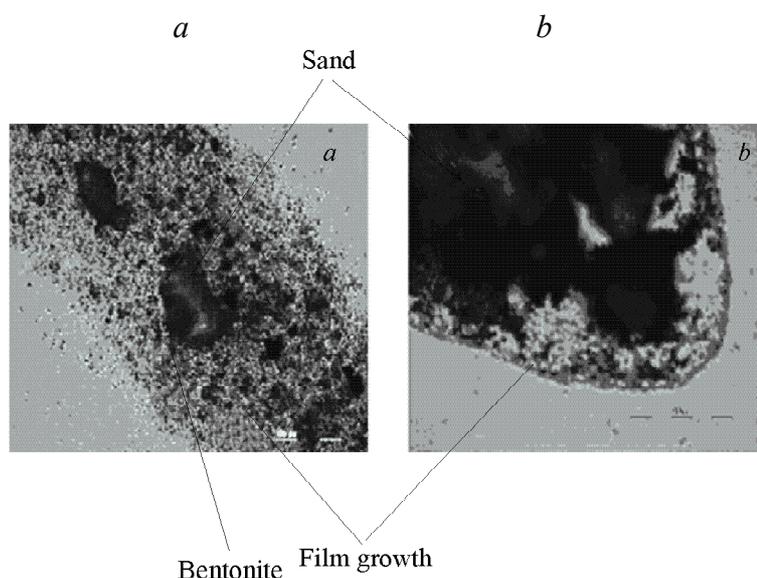
Water-leach tests (pH of water leachate ranged from 2 to 9) with spent BASSF specimen showed that maximum of 5 to 12 % of retained cationic species (at pH = 2) and 0 to 18 % of anionic species (at pH = 9) is released (Table 4) indicating that the inorganic contaminants are irreversibly adsorbed by BASSF specimen to a large extent.

*Table 4. Leaching of ions from 100 g of spent BASSF at different pH values*

Species	Mass retained (mg)	Mass leached (mg) at different pH					% Retained at different pH				
		Leachate pH					Leachate pH				
		2	4	6	7	9	2	4	6	7	9
Sodium	326	16	13	13	11	13	95	96	96	97	96
Calcium	268	31	7	3	3	5	88	97	99	99	98
Magnesium	100	12	2	0.6	0.9	0.8	88	98	99	99	99
Potassium	206	16	15	16	15	17	92	93	92	93	92
Bicarbonate	1146	29	101	80	95	203	97	91	93	92	82
Nitrate	695	0	0	0	0	0	100	100	100	100	100
Sulfate	193	9	7	4	5	19	95	96	98	97	90

The spent BASSF and SSF specimens show growth of thin films in the polarizing micrographs (Fig. 2, a, b). Despite growth of biofilms occurring both in BASSF and SSF specimens, the former alone was capable of retaining inorganic contaminants. The ability of BASSF to retain inorganic ions is attributed to the inclusion of bentonite in the filter material. Bentonite particles increase the adhesion sites for bacterial biofilms leading to enhanced extracellular polymer production as evidenced by the higher OM contents of the spent BASSF specimens (3.4 to 4.2 %). It has been suggested

that such films are composed of polysaccharide polymers that can bind contaminants by hydrogen and Coulombic bonding [ 4,19]. Further, the presence of micropores in bentonite would increase the hydraulic retention time of contaminants inside the BASSF specimen. The combined effects of increased OM mediated adhesion sites and increased hydraulic retention time possibly enables the BASSF specimen to retain inorganic contaminants besides microorganisms. The composition and distribution of the cell biomass developed in the sand-bentonite mix is under investigation.



*Fig. 2. Polarizing microscope photographs of spent filter specimens: BASSF (a), SSF (b).*

Clogging of micropores by accumulation of cell biomass along with other enzymatic secretions apparently reduced the flow rate during passage of groundwater and nitrate spiked solution in the BASSF specimens. In order to minimize clogging of pores by organic growth, the impact of reducing the bentonite content and dry density of BASSF is being examined. It is envisaged that BASSF filters could find use in treating contaminated groundwater for potable needs at household and community level.

### **Conclusions**

Small addition of bentonite to slow sand filter enables it to retain inorganic contaminants in addition to microbial contaminants as demonstrated by experiments performed with BASSF specimens permeated with contaminated

groundwater and nitrate spiked solution. The SSF specimen in absence of bentonite addition was unable to retain nitrate ions on passage of nitrate spiked solution. The BASSF specimens developed OM contents of 4.2 and 3.4 % respectively on permeation of contaminated groundwater and spiked nitrate solution. The SSF specimen developed negligible OM content of 0.04 % on permeation with spiked nitrate solution. It is believed that lack of substantial OM growth hampered the retention of nitrate ions by SSF specimen. Comparatively, the combined effects of increased OM mediated adhesion sites and hydraulic retention period possibly enabled the BASSF specimen to retain inorganic contaminants besides microbial contaminants.

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