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Research paper

Purification of greywater by a moving bed reactor followed by a filter including a granulated micelle-clay composite

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ABSTRACT

Reuse of grey water (GW) enables to reduce fresh water consumption, but a treatment is required to prevent potential transmission and propagation of pathogenic organisms. This study presents results on the removal of pathogenic bacteria from GW as well as reduction of turbidity, TSS COD, and BOD by a novel treatment system. Compared to previous studied methods, three new elements are presented in the current treatment of GW: (1) A granulated complex of micelles of the organic cation octadecyltrimethylammonium (ODTMA) with montmorillonite was employed in filtration of GW. This complex was efficient in purifying GW due to its large surface area, positive charge and existence of hydrophobic domains. The granulated complex enabled flow when present exclusively in the filter; (2). A moving bed reactor for decomposition of part of the organic matter in the GW. This pretreatment stage, prior to the micelle-clay filter, was also efficient in removing pathogenic bacteria; (3) A regeneration stage of the micelle-clay filter conducted by passing either dilute solutions of Na-hypochlorite or HCl through the micelle-clay complex, or by heating the complex. Incubation of GW for either two weeks or one day in the pretreatment stage yielded a 10- and 7-fold enhancement in the volume filtered, which did not contain fecal coliforms, i.e., 300 and 210 L for 40 g of complex, respectively. The capacity of purified volume per gram of the complex increased further several-fold (>23 L/g) for filters filled exclusively with granules. Regeneration of the complex in the filter further enhanced the capacity.

1. Introduction

Reuse of grey water (GW) enables savings in fresh water consumption. Grey water from sinks, showers and baths (the focus of this study) amounts to 150–300 L per household per day (Christova-Boal et al., 1996; Alfiya et al., 2012). Use of inadequately treated GW can be a potential health hazard (Maimon et al., 2010; O'Toole et al., 2012; Gross et al., 2015). In particular, pathogenic microorganisms pose a main health hazard (Dixon et al., 1999; Chen et al., 2013) as illustrated by recent studies in which risk assessment of GW due to presence of pathogens was examined (Haas et al., 1999; Ottoson and Stenstrom, 2003; Winward et al., 2008a, 2008b; Barker et al., 2013; Maimon et al., 2014). For above reasons disinfection of grey water is generally considered an essential step in the purification process of GW (Winward et al., 2008a, 2008b; Maimon et al., 2014). Treatment of GW by filtration was described by Dalahmeh et al. (2012) who employed pine bark, activated charcoal, polyurethane foam and sand as filter materials; they concluded that activated charcoal and bark were the most efficient agents in the removal of anionic detergents and bacteria from GW. A filter containing a positively charged micelle-clay complex mixed with sand was much more efficient than activated carbon in removal of indicator bacteria (Brook et al., 2015). Winward et al. (2008b), who used moving bed reactor (MBR) and other technologies, concluded that the MBR was the most efficient and robust technology. Bani-Melhem and Smith (2012) successfully used a submerged MBR for removal of anionic surfactants and coliforms. Friedler et al. (2005) combined biological treatment using a rotating biological contactor (RBC) with a sedimentation stage, a sand filtration stage and a hypochlorite disinfection stage. Also, RBC's combined with sand filtration and UF membranes (Friedler et al., 2006) and combined with UV disinfection (Gilboa and Friedler, 2008) were tested for treatment of GW. Constructed wetlands have been frequently employed for GW treatment (e.g. Vymazal, 2007; Winward et al., 2008b). Gross et al. (2007) and Benami et al. (2013) employed a recirculating vertical flow constructed wetland (RVFCW) whereas Travis et al. (2012) examined the use of an anaerobic pretreatment stage before treatment of GW by RVFCW. Santos et al. (2012) tested purification of GW by a hydraulically, self-cleaning mesh filter followed by UV disinfection.

Shtarker-Sasi et al. (2013) demonstrated efficient removal of microorganisms by columns filled with a mixture of powdered micelle-clay complex and sand. Brook et al. (2015) employed this system for treatment of GW from showers and sinks. The micelle-clay complex was

Abbreviations: CEC, cation exchange capacity; GW, grey water; ODTMA, octadecyltrimethylammonium.

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formed by an interaction of the organic cation, (ODTMA) with sodium bentonite. The size of the micelles, composed of several tens to several hundred molecules, is in the nanometer range, whereas the clay platelets have a thickness of around one nanometer and a typical area of around 1 μm^2 (Mishael et al., 2002). The ODTMA-bentonite was constructed to have an excess of positive charges of half of the CEC of the clay mineral. The material characteristics of the micelle-clay complex differed from those of a tested organo-clay complex of the same composition, which was formed by adding ODTMA as monomers. The micelle-clay complex was shown to be highly efficient in the adsorption of anionic herbicides whereas the organo-clay complex was not. It was shown that GW with a total suspended solids (TSS) content of >20 mg/L was devoid of TSS after passage of 4 L through the filter (Brook et al., 2015). Assuming a typical size of *Escherichia coli* of $0.5 * 2$ μm (Miao et al., 2003), which is equivalent to a volume of 0.4 $(\mu m)^3$, it follows that per liter the mass of 100 million of such bacteria is <0.1 mg/L. On average this is two orders of magnitude less than the mass of the other materials in the grey water. Hence, removal of organic material prior to filtration by the micelle-clay complex might significantly enhance the filter's capacity for removal of bacteria.

In the present study, results are presented on a two-stage treatment configuration for GW, consisting of a moving bed reactor (Odegaard, 2006; Biswas and Turner, 2012) for decomposition of a part of the organic matter in the GW followed by a filter containing a micelle-clay complex. A granulated micelle-clay filter was used and its performance with and without sand was examined. The amount of the active material in this novel granulated micelle-clay filter could be enhanced at least 10-fold in comparison to previously used filters. Finally, the micelle-clay complex was subjected to several regeneration procedures and their effect on the removal capacity of the complex was examined.

2. Materials and methods

2.1. Materials

Bentonite was purchased from Tolsa–Steetley, UK. The organic cation ODTMA was purchased as a bromide salt from Sigma-Aldrich (Sigma Chemical Co., St. Louis, MO). Quartz sand (0.8–1.5 mm particle size) was provided by Negev Minerals LTD, Israel. Non-woven, polypropylene geotextile filter was manufactured by Markham Culverts Ltd., Papua - New Guinea. The following chemicals were used: Sulfuric acid (H_2SO_4; 95–97%, MERCK, 1.00731, Germany), Potassium dichromate ($K_2Cr_2O_7$, Sigma, P5271, USA), Silver Sulfate (Ag_2SO_4, Sigma, S-7638, USA). Mercury(II) Sulfate ($HgSO_4$, Riedel - de Haen, 31013, Germany). Ferroin indicator (Sigma S-46270), $Fe(NH_4)2(SO_4)2 * 6H_2O$ (Sigma S-203505). De-ionized water was used to prepare all solutions. Growth media for enumeration of bacteria were obtained from Difco (Kavacik/Beykoz - Istanbul, Turkey) and HyLabs (Park Tamar, Rehovot).

2.2. Methods

2.2.1. Granulation procedure

Granulated complexes of ODTMA-clay were prepared as described (Nir et al., 2015). The amounts used were multiples of 10 g/L clay and 12 mM of the cation. The stages of preparation were: (1) Incubation of the (salt of) organic cation in tap water at temperatures between 38 °C and 50 °C followed by addition of the clay; (2) Removal of water in a filter press to a final water content of about 50% and placing the cake in an oven at 60 °C to reach 42–45% water; (3) Granulation by a two stage machine, first to obtain the desired granule size followed by spherodizing; (4) Drying of the granular complex in an oven at 60 °C to 3% humidity; (5) Sieving the granules to particle sizes between 0.3 and 2 mm.

2.2.2. Grey water sampling protocol

Grey water was collected from the effluent of showers and sinks in student dormitories. Grey water was collected into 200 L barrels, periodically transferred to 20 L canisters and stored at 4 °C until the filtration experiments. In case of the two-stage filtration step, part of the GW was placed in the bioreactor used for pretreatment.

2.2.3. Moving bed reactors

The polypropylene moving bead reactors with a water holding capacity of 150 L were operated with an operating volume of up to 130 L of GW. The reactors contained 30 L of biofilm carriers (K1, Kaldnes Ltd., Norway) with a specific surface area of 500 m^2/m^3. Through a manifold at the bottom, compressed air was driven into the reactor causing a dispersion of the beads up to a height of approximately 2/3 of the water table in the reactor. Retention time of the GW in these batch-operated reactors varied from one day to one month. Each of the moving bed reactors was operated with matured biofilms on the carriers as a result of exposure to GW for several weeks to months. Experiments were initiated after discharge of the water in the reactors and refilling them with the GW to be examined.

2.2.4. Filtration experiments

Two sets of filtration columns (40 cm length, 5 cm diameter) were prepared, with geotextile coverings at the inlet and outlet. Measured weights of complex (ODTMA-clay, 40 or 50 g were mixed with approximately 1.2 kg of washed quartz sand, for each column. Approximately, a 2 cm layer of sand was added to the upper and lower ends of the column in order to fill it completely. Additional experiments employed smaller filters (20 cm long, 1.6 cm diameter). The columns were connected to a peristaltic pump (Cole-Palmer Masterflex L/S, Vernon Hills, Illinois, USA) with Tygon tubes. Each treatment was conducted in duplicate. Tap water was added to the columns at a rate of 10 mL/min in an upward direction in order to eliminate air pockets and channeling. Passage through geotextile served as a pre-filtration stage. Grey water was transferred to 200 L barrels from where it was pumped into duplicate filter columns at a flow rate of 40 mL/min (or 5 mL/min in the case of the smaller filters). Samples of 500 mL were collected before and after the geotextile filter and from the effluent of each of the filters after passage of a measured volume of greywater. Samples were immediately stored at 4 °C until microbiological testing, which was usually within one day. Preservation of grey water samples for COD analysis was done by acidifying samples with sulfuric acid to pH < 2, followed by storage at −20 °C for a maximum period of two weeks. TSS and turbidity samples were stored at 4 °C until testing.

2.2.5. Bacterial counts, COD and BOD measurements

Counts of total bacteria, coliforms, fecal coliforms, total chemical oxygen demand (COD) and biological oxygen demand (BOD) were performed by a certified laboratory (Aminolab laboratory, Ness Ziona 70400, Israel) in accordance with the Standard Methods (APHA et al., 2005).

2.2.6. Regeneration

Regeneration was conducted with small columns (1.6 cm, 20 cm). Three procedures were employed: (i, ii) Washing with 275 mL of a 0.5 % Na-hypochlorite solution, or HCl (0.05 M) at a flow rate of 2 mL/min followed by 250 mL of tap water. For second regeneration the concentrations of washing solutions were doubled. Hereafter, the column was placed in an oven at 60 °C for drying the complex. (iii) Heating for 3 h at 120 °C.

3. Results and discussion

3.1. Preliminary tests of purification efficiency

Grey water collected from showers and sinks of a student dormitory was passed through a geotextile filter followed by a filter containing 50 g of a granulated micelle (ODTMA)-bentonite complex mixed with 1000 g sand at a 1:20 w/w ratio. The flow rate was 40 mL/min.