Short Communication

Application of nano-silver coated granular activated carbon for inactivation of septic tank effluent

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ABSTRACT

On-site sanitation systems such as cesspools and septic tanks are widely used in most developing countries. These systems primarily aim to collect and treat toilet wastewater or blackwater. Although septic tanks are commonly used in non-sewered areas, their effluents are still rich in pathogens and other pollutants. The practice of direct discharge of septic tank effluents into the surrounding environment in the absence of proper treatment has increased health risks. In order to reduce this problem, a post-treatment unit consisting of nano-silver coated granular activated carbon (NS-GAC) has been developed. The study results revealed that the inactivation efficiency of the NS-GAC unit increased with increasing hydraulic retention times (HRT) from 10, 20 and 30 minutes; however, for economy of scale, the NS-GAC unit could be operated at the 10 minutes HRT to achieve complete removal of Escherichia coli bacteria. The study showed a high feasibility of utilizing the NS-GAC media as a post-treatment unit for pathogen inactivation of septic tank effluent.

Key words | E. coli, granular activated carbon, inactivation, nano-silver, on-site sanitation, septic tank effluents

INTRODUCTION

In developing countries, where on-site sanitation systems are commonly used for wastewater treatment, over 90% of wastewater is discharged without proper treatment (Langergraber & Muellegger 2005). Effluents from on-site sanitation systems, such as septic tanks and cesspools, are allowed to seep into surrounding soils or are discharged into nearby storm sewers or watercourses without further treatment. As these systems contain high concentrations of organic matter and pathogenic indicator microorganisms (AIT 2014), these practices have increased environmental pollution and health risks. For example, access to sanitary toilets is almost 100% in Thailand, but there are increasing incidents of intestinal infectious diseases, which reveals the drawbacks of the present on-site sanitation systems (MoPH 2014). The need for post-treatment units to treat effluent from on-site sanitation systems has been highlighted by Rochmadi et al. (2010) and Khan et al. (2012).

Nanoparticles have been effectively used in the environmental engineering field due to their high charge capacity and surface to volume ratio (Nangmenyi & Economy 2005). The effectiveness of nanoparticles for bacterial decontamination has been widely reported (Xu et al. 2004; Elichiguerra et al. 2005; Gogoi et al. 2006). Silver nanoparticles, in particular, possess antimicrobial effects; their effectiveness has been reported in up to 650 bacterial species (Shahrokh & Emtiazi 2004). Although the exact mechanism of bacterial inactivation by nano silver is not yet clearly understood, cell lysis or the inhibition of cell
transduction by the large surface area of nano silver particles are considered as potential fundamental mechanisms (Prabhu & Poulose 2012). In this study, granular activated carbon (GAC) coated with nano silver (NS) particles was investigated for its efficiency in inactivating fecal microorganisms in septic tank effluents.

**OBJECTIVES**

The overall objective of this study was to determine the pathogen inactivation efficiencies of the NS-GAC unit treating septic tank effluents. The specific objectives were as follows:

1. To evaluate the efficiencies of pathogen inactivation at different concentrations of NS and modes of feeding
2. To determine dimensions of the NS-GAC column unit for various operation periods and hydraulic retention times

**METHODOLOGY**

**Synthesis of NS-GAC**

NS was synthesized by a polyol process (Kilderby et al. 2005), in which silver nitrate (AgNO3, 99.9%) and ethylene glycol (EG) (C2H6O2, 99.5%) were used as oxidizing and reducing agents, and polyvinylpyrrolidone (PVP) [(C6H9NO)n, K30, Mw 40,000] was used as a stabilizer to accelerate the reaction. The reagent concentrations and synthesis procedure were followed according to Lee & Koo (2012) and Wongkiew (2013).

During the NS synthesis, various amounts of AgNO3 (0.25, 0.50, 0.75, 1.00 and 1.25 g) were added per 100 mL of EG and stirred for 10 minutes. To synthesize the NS-GAC without a binding agent, 20 g of GAC were added into the NS solution and mixed continually for 10 minutes. Next, 2 g of PVP was added to the GAC and NS solution, which was then heated to 120 °C for 4 hours and washed with distilled water four times. For the final step, the NS-GAC was heated in an oven at 105 °C for 1 hour to remove excess water and yield the NS-GAC in dry form.

To synthesize the NS-GAC with a binding agent, 0.5 mL of 3-mercaptopropyltrimethoxysilane (MPTMS) was added to 100 mL of ethyl alcohol (99.9%), and mixed well in a magnetic stirrer hot plate for 2–3 minutes. Thereafter, 20 g of GAC was added to the solution and mixed homogeneously at 80 °C until yielding dry GAC coated with MPTMS (Wongkiew 2013). Finally, 20 g of dried GAC coated with MPTMS was added into varying concentrations of NS solutions, to make the NS-GAC with binder following the aforementioned method.

**Analysis**

Scanning electron microscope (SEM) and Brunauer-Emmett-Teller (BET) analyses were conducted to characterize the physical properties of the NS-GAC. The amount of NS attached on the GAC was measured by inductively coupled plasma mass spectrometry (ICP-MS). *Escherichia coli* was used as a fecal indicator bacteria and analyzed using the plate count standard method (APHA AWWA & WEF 2005).

**Operating conditions**

The inactivation efficiency of NS-GAC was evaluated for both batch and continuous flow experiments by varying the operating conditions, as shown in Table 1.

For each batch operation, 20 g of the NS-GAC and 20 mL of septic tank effluent were put into a 100 mL beaker and mixed at 80 rpm at 25 °C. After each HRT, 1 mL of the mixed liquid sample was taken and analyzed for *E. coli* concentration.

NS doses were calculated based on the average concentration of NS in synthesized NS-GAC, and AgNO3 amounts, which were varied to cover a wide range. The optimum NS dose from the batch tests was selected for the continuous experiment using columns made of acrylic pipe with a 4 cm diameter and 25 cm in length. The experimental setup is shown in Figure 1. Tests were conducted under upward and downward feeding modes at different HRTs using actual septic tank effluent (Table 1(a)). Determinations of *E. coli* breakthrough times were conducted using an actual septic tank effluent with the characteristics shown in Table 1(b), as well as a synthetic effluent, prepared...
by inoculating distilled water with *E. coli* at concentrations of $10^5$–$10^8$ CFU/mL.

**RESULTS AND DISCUSSION**

**Characterization of NS-GAC**

The amounts of NS coated on the NS-GAC particles without and with the binder were found to be 0.1% and 0.2% by weight, respectively. These results show the significance of the binder in priming the GAC surface to bind more NS particles, which are essential for *E. coli* inactivation. The NS-GAC particles with binder were used in the *E. coli* inactivation experiments.

The surface analysis carried out by SEM showed the dominance of pore space on the surface of GAC; however, less pore space and smooth surfaces were observed in NS-GAC, suggesting the uniform distribution of NS particle coating on the GAC surface. Similarly, BET analysis depicted that GAC had a decreased surface area when it was coated with NS. The specific surface areas of GAC...
with and without NS coating were 458.49 m² g⁻¹ and 836.05 m² g⁻¹, respectively. Such a reduction is expected because the NS fills up the GAC pore spaces.

**Batch operation**

*E. coli* inactivation results at various NS doses and HRTs are presented in Table 2. The log reduction of *E. coli* was found to increase with increasing NS doses. HRT, however, had less of an impact on *E. coli* inactivation at NS doses below 2 mg/L. This result is likely due to the need for a minimum NS dose (threshold) to initiate the inactivation; it seems that level is not attained at a lower dose of NS. The maximum *E. coli* inactivation of 7-log was observed at the highest NS dose (3.3 mg/L) for the longest reaction time of 240 minutes.

The NS dose of 2.0 g/L yielded approximately 6-log reduction of *E. coli* at HRT of 240 minutes. Although this inactivation was slightly lower than that obtained at the NS dose of 3.3 g/L, for economic and environmental reasons the NS dose of 2.0 g/L was selected for the continuous-feeding experiment. This dose was equivalent to 2 mg NS/g of NS-GAC.

**Continuous operation**

In the continuous flow experiments, each column was filled with 235 g of NS-GAC, or about 470 mg of NS per column, and the experimental conditions listed in Table 1 were employed. It was found that within the HRTs of 1–240 minutes, *E. coli* was not detected in the effluent of both columns operated with upward-flow and downward-flow modes. These results demonstrated the high efficiencies of the NS-GAC column in inactivating the *E. coli*, which was probably due to the combined effects of NS inactivation, filtration and adsorption of the *E. coli* cells inside the NS-GAC matrix. The high efficiencies of continuous operation relative to the batch test were likely due to the high dose of NS to septic tank effluents. Therefore, relatively lower HRTs of 10, 20 and 30 minutes were chosen in the continuous flow experiment to determine the breakthrough time of the NS-GAC column when treating the septic tank effluents. To avoid long-term clogging and short circuiting, the up-flow mode was selected.

The results in Figure 2 show the *E. coli* breakthrough time increases with increasing HRTs. Reduction of the performance over time was likely due to adsorption of the solids, including the *E. coli* cells, to the NS-GAC surface, resulting in fewer active NS ions to inactivate the *E. coli*. On the other hand, given the high suspended solids and organic matter concentrations, the actual septic tank effluent had shorter breakthrough times than the synthetic septic tank effluent. For example, at the HRT of 10 minutes, the *E. coli* breakthrough times of the actual and synthetic septic tank effluents were found to be 11 and 15 days, respectively; while at the 20 minutes HRT, the *E. coli* breakthrough times of the actual and synthetic septic tank effluents were 19 and 23 days, respectively. These breakthrough times were relatively longer than those reported by Mthombeni et al. (2012) due to the difference in NS dose and media (resin beads) employed in their experiments. The performance of the NS-GAC unit with respect to other parameters, namely, TSS, COD and BOD removal, were on average 90%, 60% and 50%, respectively.

At present, effluent standards for on-site wastewater treatment systems in Thailand and several other Asian countries do not specify a safe level of fecal indicator microorganisms. However, WHO (1989) suggested that fecal coliforms in effluent to be reused in agriculture should be equal to or lower than 10³ geometric mean no./100 mL.

Data of *E. coli* breakthrough times using the actual septic tank effluent and the WHO (1989) guideline could be applied to calculate the size of a NS-GAC column for treating 125 L/day of septic tank effluent from a single household of 4–5 persons. Obviously the sizes of the

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<table>
<thead>
<tr>
<th>NS dose (g/L)</th>
<th>HRTs (min)</th>
<th>E. coli (log reduction value)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(g/L)</td>
<td>30 60 120</td>
<td>180 240</td>
</tr>
<tr>
<td>0.7</td>
<td>2 2 2</td>
<td>2 2</td>
</tr>
<tr>
<td>1.3</td>
<td>3 3 3</td>
<td>3 3</td>
</tr>
<tr>
<td>2.0</td>
<td>3 3 4</td>
<td>5 6</td>
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<td>2.7</td>
<td>6 6 6</td>
<td>6 6</td>
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<td>3.3</td>
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</table>
NS-GAC column vary with the operation time and HRT. For example, at an operation time of 6 months, the size of the NS-GAC column was calculated to be 16.7 and 27.5 L for HRTs of 10 and 30 minutes, respectively. The choice of NS-GAC column size would depend on economic and practical considerations. Frequent replacement of the NS-GAC column after breakthrough times might not be convenient for the users. If it is decided to replace the NS-GAC once a year (or after an operation time of 12 months), the NS-GAC column would have to be 33.4 or 48.0 L in size for design HRTs of 10 or 50 minutes, respectively.

Moreover, research into appropriate methods of NS-GAC media regeneration needs to be explored. If disposal of the NS-GAC media is required, it should be made in an environmentally sound manner such as through a secure landfill.

Although this study revealed the effective inactivation of E. coli, enteric viruses, which are more resistant, could still be present as a public health threat. Therefore, monitoring for the presence of bacteriophages, an enteric virus indicator, should be done to determine the efficiency of the NS-GAC treatment on enteric viruses. At present, research continues to study the microbial community presence in the NS-GAC column as well as their roles in polishing the septic tank effluents.

Similarly, the engineering and practical implementation need to be further investigated using pilot or full-scale septic tanks to demonstrate the applicability of this technology, including the economic aspects. Full-scale research on the NS-GAC system is under way.

**CONCLUSIONS**

Based on the experimental results obtained from this study, the following conclusions have been made:

1. The synthesized NS-GAC media with the binding agent was found to be effective in inactivating E. coli in the septic tank effluent.
2. The inactivation efficiency of the NS-GAC media increases with increasing HRTs from 10 to 30 minutes.
3. Breakthrough times of E. coli from the NS-GAC unit inactivating actual septic tank effluent were 11 and 19 days for HRTs of 10 and 20 minutes, respectively.
4. Based on the effluent standard for fecal coliforms of $10^3$ geometric mean no./100 mL, the sizes of the NS-GAC units treating septic tank effluent of 125 L/day from a single household for one year were calculated to be 33.4, 38.7 and 48.0 L for HRTs of 10, 20 and 30 minutes, respectively, after which the NS-GAC media needs to be regenerated or replaced.

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REFERENCES

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