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Research Article

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Using of polydiallyldimethylammonium chloride for removal *Cryptosporidium* from the public recreational water venue

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ABSTRACT

Cryptosporidium outbreaks in recreational water venues have threatened public health, especially in swimming pools. There is still no reliable treatment technique to remove Cryptosporidium oocysts from swimming pools. The performance of polydiallyldimethylammonium chloride (polyDEDMAC)as coagulant on Cryptosporidium removal from pools was evaluated in this paper. Seeding methods of polyDEDMAC and dosage of polyDEDMAC versus oocysts concentrations were tested. Results indicated oocysts removal efficiency for feeding oocysts and polyDEDMAC simultaneously were more than 99% (2 log), and continuously feeding of polyDEDMAC achieved at least 99% (2 log) removals, compared with removal efficiency was 20% by control experiment without coagulation. All of these experiments indicated that the polyDEDMAC should be fed by pump automatically and continuously in order to maximize oocysts removals. In addition, oocysts concentration impacted the system performance. The higher oocysts concentration consumed more polyDEDMAC. Overall, polyDEDMAC is an effective and promising coagulant to improve oocysts removals from swimming pools.

Keywords: polydiallyldimethylammonium chloride, *Cryptosporidium*, recreational water quality

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INTRODUCTION

Polydiallyldimethylammonium chloride (polyDADMAC) is a homopolymer of diallyldimethylammonium chloride (DADMAC). The production reaction of polyDADMAC is shown in Equation (1). The molecular weight of polyDADMAC is typically about thousands of grams per mole, and even up to a million for some products, and the molecular formula is $(C_8H_{16}NCl)_n$[1].It is a high charge density cationic polymer, which makes it well suited for coagulation and flocculation.The pyrrolidine structure is favored.

Cryptosporidium spp.are intracellular parasites that infect human epithelial cells of the small intestinewith diameter of 4-6 µm, commonly found in recreational water bodies[2]. It is geographically widespread which infects many host species, and produces prodigious numbers of oocysts[3]. They are environmentally persistent and very resistant to many disinfectants, including chlorine, which is the major barrier to infectious disease transmission that has been used for the past several decades in the swimming pool water treatment [4]. Typical swimming pools in the United States require at least 1 mg/L (ppm) free residual chlorine [5, 6]. This concentration free chlorine enables *Cryptosporidium* to survive for over 11 days [4, 7]. The use of polystyrene oocysts as an oocyst surrogate has been done by multiple researchers and it was used in this study [8]. Oocysts with diameter of 4.87 µm (Polysciences. Inc) were used as the surrogate since oocysts are virtually identical to *Cryptosporidium* oocysts in size, shape, density, and surface charge in water [8].

Cryptosporidium has caused several large waterborne disease outbreaks of gastrointestinal illness, cryptosporidiosis, and emerged as a parasite of major public health concern in United States, United Kingdom, Australia, etc[9].

Multiple sources have indicated that weaker subpopulations (infants, young children, pregnant women and people with severely compromised immune systems) are more susceptible and could die from cryptosporidiosis [10]. One common infection is by swimming in the swimming pool with human contamination. Infected humans excrete approximately 10^8 to 10^9 oocysts in stool per day [11]. High levels of oocysts in stool make it possible for a single infected person's bowel movement to significantly contaminate beaches and artificial venues such as swimming pools. Numerous waterborne outbreaks of cryptosporidiosis have been linked to swimming pools.

This study developped a novel evaluation procedure for polyDEDMAC coagulation that will produce reliable results applicable in swimming pools. Decisions had to be made regarding whether to add polyDEDMAC as continuous inputs or as slug inputs, and whether or not polyDEDMAC build-up occurs in the system after multiple rounds of dosing causing impaired performance, and whether the concentration of oocysts into the pool system impact the overall polyDEDMAC performance.

EXPERIMENTAL SECTION

Experiment Setup

A 5,000 L swimming pool was built with filtration system and chemical control system. Pool water can be pumped through the filter (either granular filter or precoat filter), shown in Fig. 1. The sand filter was made from transparent polyvinyl chloride (PVC) pipe. It utilized an integral media support cap (Leopold, ITT) as support for filter media as well as backwash flow distribution. The filter had a diameter of 15 cm and the sand depth of 30 cm. The effective size of the sand was 485 μm. The hydraulic loading efficiency(HLR) for the sand filter was 35 m/h, which is a typical high-rate filter loading rate used in swimming pools. All chemicals and oocysts were fed using peristaltic pumps. Coagulant (polyDEDMAC) and oocysts were fed into the pipe ahead of the pump and pre-filtration for a rapid polyDEDMAC mixing. Streaming current meter, turbiditimeters, particle counters were installed to measure the surface charge of the water, turbidity and particle concentration. On-line data can be record and download from a computer.

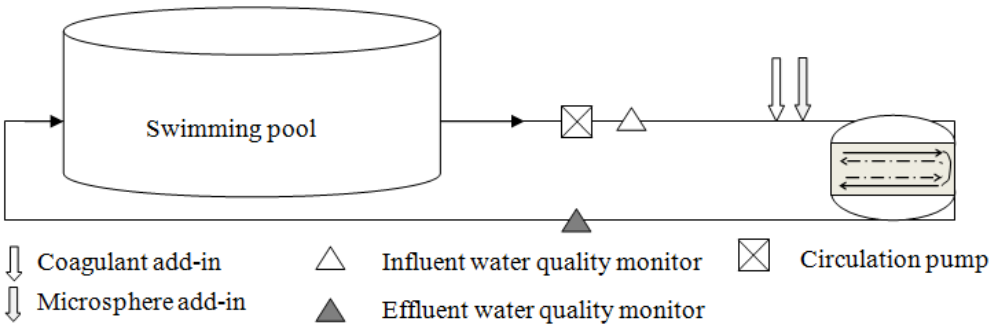

Fig. 1 Swimming Pool Setup

Experimental Approach

Order of feeding polyDEDMAC and oocysts

Three scenarios are possible in practice and were evaluated to produce reliable results, "adding polyDEDMAC first", "adding oocysts first", and "adding polyDEDMAC and oocysts simultaneously". The recommended dosage of polyDEDMAC and 10^7oocysts (1.8 #/mL) were seeded for each experiment. The experiment with adding polyDEDMAC and oocysts simultaneously were conducted in one turnover time (8 hr), which was named as "normal" experiment. Samples were collected at 0.5, 1, 2, 4 6, and 8 hr, respectively. Oocysts were seeded and samples were taken after seeding one recommended dosage of polyDEDMAC for the "coagulant first" experiment over 8 hrs. Oocysts were seeded prior to polyDEDMAC addition for 30 mins, and the polyDEDMAC was feed for 8 hr and samples were taken over this time for the "oocysts first" experiment.

Feeding modes of coagulant and oocysts

"Slug feeding of coagulant" and "continuous feeding of coagulant" were evaluated. The experiment with slug feeding was conducted by adding the polyDEDMAC with times per day. One recommended dosage of coagulant was fed in 8 hrs, the amount of 10^7oocysts (1.8 #/mL) was seeded and samples were taken after the coagulant addition over the next 8 hrs. "Slug feeding" experiments were conducted approximate 64 hrs. The experiment with continuous feeding coagulant was conducted by continuously feeding 1.56 mg/L/8hrs polyDEDMAC by coagulant pump, which was just like "normal" experiment.

Oocysts concentration versus polyDEDMAC dosages

Multiple experiments with different polyDEDMAC dosages (from 0.03 mg/L to 1.56 mg/L) and oocysts concentration (the amount of 10^5, 10^7 and 10^8oocysts, with concentration of 1.8 $\times 10^{-2}$ #/mL, 1.8 #/mL, and 18 #/mL, representatively) were performed.

RESULTS AND DISCUSSION

Orders of Seeding Oocysts and polyDEDMAC

In swimming pools, three possible scenarios are existed referring to *Cryptosporidium* contamination, such as oocysts releases into the pool while no coagulant residual exists in the pool (corresponding to the experiment procedure adding oocysts first), or there is coagulant residual in the pool when oocysts are released (corresponding to adding coagulant prior to oocysts), or oocysts contamination occurs during coagulant active addition (corresponding to adding oocysts and coagulant simultaneously). The order of adding polyDEDMAC and oocysts might impact the overall removal. Control experiments were conducted without coagulant addition, which showed 20% oocyst removals from the pool. Fig. 2 shows the percent removal and log removals of *Cryptosporidium*oocysts referring to the three scenarios. The removal efficiency, 99.5% (2.3 log), was achieved by feeding polyDEDMAC and oocysts simultaneously. Adding polyDEDMAC first averaged 94% removal (1.3 log). The average removal efficiency was only 65% (0.5 log), for 'adding oocysts first' experiment.

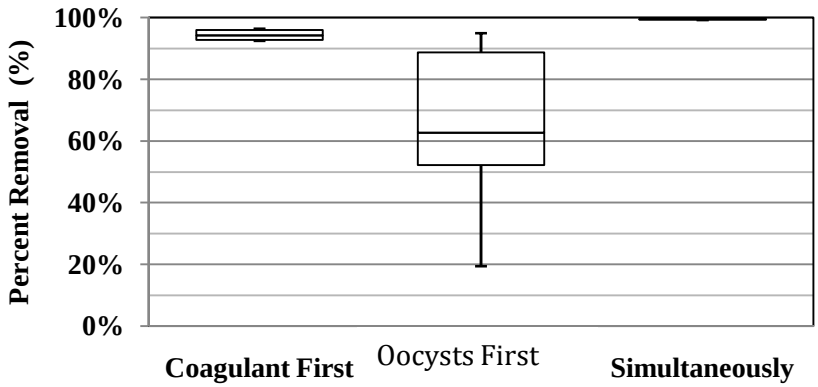

Fig. 2 Performances of the Three Scenarios Referring to Sequence of Adding 10^7Oocysts (1.8 #/mL), 1.56 mg/L polyDEDMAC, 30 cm Sand, and 37 m/h Filtration Rate

("coagulant first" – seeding of oocysts as well as collecting samples after feeding 1.56 mg/L polyDEDMAC for 8 hrs; "Oocysts first" – seeding oocysts 30 mins prior to polyDEDMAC addition, followed by feeding polyDEDMAC for 8 hr and taking samples over this time; "Simultaneously" – feeding oocysts and polyDEDMAC simultaneously.)

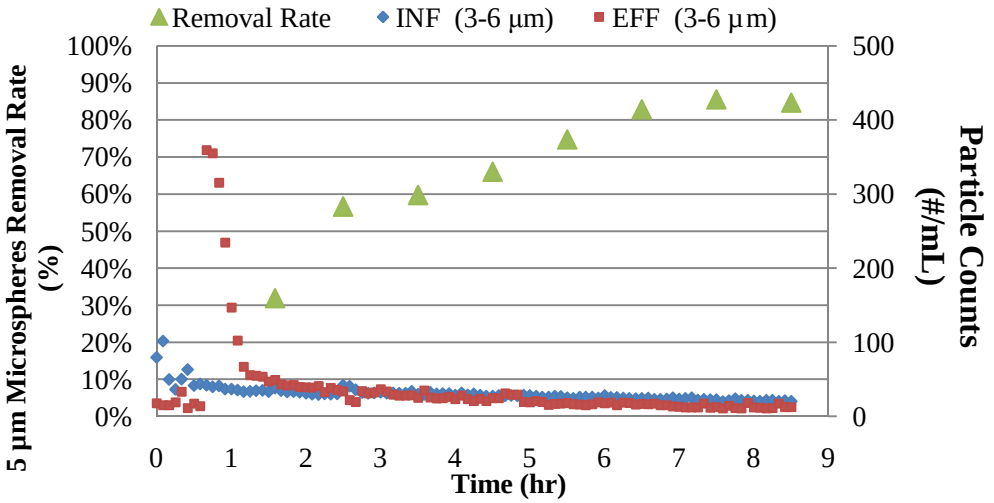

Fig. 3 "Oocysts First" Test, Oocysts Removal and Filter Influent and Effluent Particle Counts, 30 cm Sand, and 37 m/h Filtration Rate

(*seeding 1.8 #/mL oocysts 30 mins prior to 1.56 mg/L polyDEDMAC addition*)

Removals were above 99% feeding polyDEDMAC and oocysts simultaneously over the 8 hrs. But removals decreased from 98% to 92% over time when feeding polyDEDMAC first. The oocysts removals were increased over

time for feeding oocysts first experiment, shown in Fig.3. The effluent particle counts (3-6 µm) was significant higher than the influent particle counts (3-6 µm) in the first 1 hr after feeding oocysts, shown in Fig.3.

The Mode of Feeding of polyDEDMAC

Experiments adding coagulant with continuous inputs or slug (pulse) input were conducted. Adding the recommended dosage of polyDEDMAC and waiting for the pool run without polyDEDMAC feeding is called "slug feeding",oppositely is "continuous feeding". Fig.4 shows the removal efficiency for continuous and slug feeding. Oocysts removal of 99.5% (2.3 log) was achieved by continuously feeding polyDEDMAC and oocysts simultaneously. While only 74% (0.6 log) was achieved by slug feeding. The mechanism of "slug feeding" is similar to the "polyDEDMAC first". The differences between these two experiments operations were that "coagulant first" experiment was only conducted in 2 turnovers (16 hrs), with feeding of polyDEDMAC for 8 hrs, and seeding oocysts and collecting samples during the next 8 hrs. The "slug feeding" experiments were conducted over 8 turnovers (64 hrs). The same as "coagulant first" experiment, polyDEDMAC was fed for 8 hrs and samples were collected in the next 8 hrs. Two samples were collected in the following 8 hrs, 2 hrs samples and 8 hrs samples since stop feeding of polyDEDMAC in each period. Removal efficiency decreased over time by slug feeding. The removal efficiency at the eighth hour since stopping feeding of polyDEDMAC was typically less than that at the second hour. All these results indicated the polyDEDMAC should be fed continuously to maximize the removals of *Cryptosporidium*oocysts from the pool.

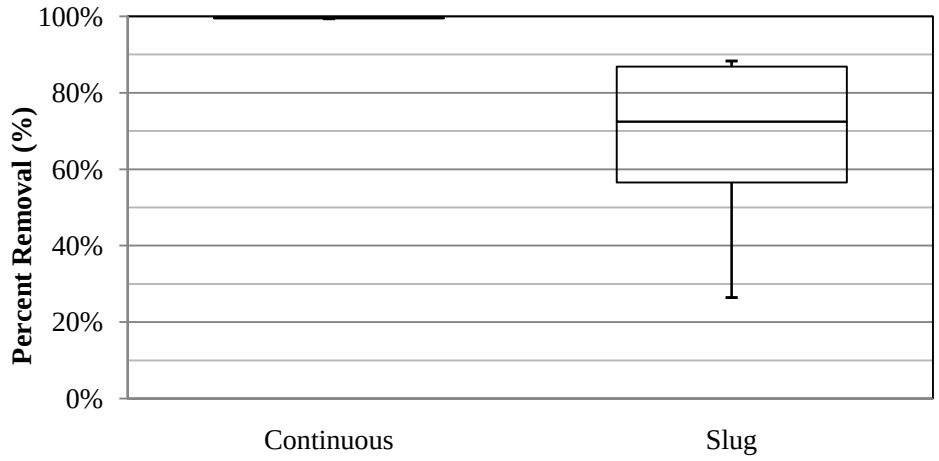

Fig. 4 Performances of Continuous Feeding and Slug Feeding, 10^7Oocysts (1.8 #/mL), 1.56 mg/L polyDEDMAC, 30 cm Sand, and 37 m/h Filtration Rate

("Slug" ─ 1.56 mg/L polyDEDMAC was fed in 8 hrs, and samples were taken after the polyDEDMAC addition after 2 hrs and 8 hrs delay; "Continuous" ─ feeding oocysts and polyDEDMAC continuously and simultaneously)

Oocysts Concentration

The removals of *Cryptosporidium* were also depending on the oocyst concentration in the source water. Multiple experiments were conducted in multiple oocysts concentrations and multiple polyDEDMAC dosages in order to determine whether the concentration of oocysts seeded into the pool system impact the overall oocysts removals.PolyDEDMAC was fed from high dosage to low dosage in order to discover the dosages corresponding to 99%, 95% and 90% oocysts removals. Fig.5displays the removal efficiency at 99%, 95% and 90% for the oocysts with different magnitude versus the polyDEDMAC dosage. Results indicated oocysts concentration impacted the overall percentage of oocysts removals. The relationship between polyDEDMAC dosage and oocysts concentration should be stoichiometric, which was indicated by the coefficient of determination (R^2) in Fig. 5.

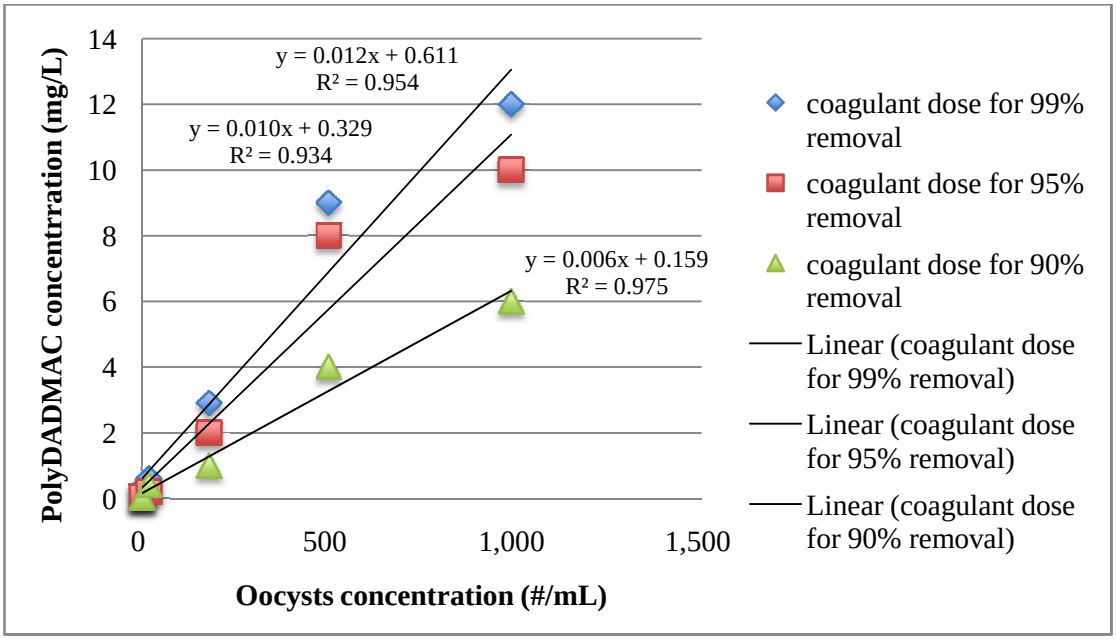

Fig. 5 PolyDEDMAC dosage versus Oocysts Concentration at OocystsRemoval efficiency of 90%, 95%, and 99%, 30 cm Sand, 37 m/h Filtration Rate

CONCLUSION

The oocysts removal efficiency for feeding oocysts and polyDEDMAC simultaneously were over 99% (2 log), compared with 94% removal (1.3 log) for "feeding polyDEDMAC first", 65% (0.5 log) for 'adding oocysts first'. Continuously feeding of polyDEDMAC achieved over 99% (2 log) removals, compared with 74% (0.6 log) by slug feeding. All of these experiments indicated that the polyDEDMAC should be fed by coagulant pump continuously in order to maximize oocysts removals. Oocysts concentration impacted the system performance. The higher oocysts concentration needed the higher polyDEDMAC dosage. However, extended feeding of polyDEDMAC led to polyDEDMAC accumulated in the system and reduced removal efficiency under the experimental condition. While a real-world pool would be expect to have a continuous supply of bather providing a natural bather load did not led to this fact.

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