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### 33 **Highlights**

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- 35 • Coagulation process achieved higher turbidity removal but longer settling time.
- 36 • Ballasted flocculation accelerated the settling rate and formation of large flocs.
- 37 • Ballasting agent affected the flocs formation during sweep coagulation mechanism.
- 38 • Quick settling of flocs resulted in the slightly lower removal of suspended solids.
- 39 • The interaction between the ballasted agent and flocs formation is worth exploring.

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### 0 **1. Introduction**

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41 The issue of water scarcity **is already** a reality to more than 2 billion people, especially with growing populations and increasing water usage [1]. Natural organic matter (NOM) and suspended impurities are compromising with the clarity of water known as turbidity, and settling to the bottom and forming sludge beds [2]. Among numerous water and wastewater treatment technologies, coagulation-flocculation process is indisputably the most widely employed technology in water treatment plants [3], [4]. Coagulation-flocculation may be broadly described as the physicochemical process that injects coagulant to destabilize the particles prior to flocculant chemicals to form larger and heavier particle aggregates (flocs) that will settle rapidly for removal by subsequent clarification process [5]–[7].

42 Over the past few decades, land constraint has become one of the major issues in the  
43 construction of conventional wastewater treatment plants [8]. Hence, development of ballasted  
44 flocculation is of great interest to tackle this issue by reducing the system footprint and hydraulic  
45 retention time [9]. Ballasted flocculation involves the injection of a ballasting agent during the  
46 flocculation process to increase the size and density of the flocs [10]. Ballasting agent such as  
47 microsand typically has a high density that could significantly result in a higher settling velocity  
48 compared to non-ballasted flocs [11]. According to previous work done, ballasting agent has  
49 greater mass than the microflocs formed during the coagulation-flocculation process, which  
50 affecting the size of flocs after the aggregation between the former and the latter and increased  
51 settling velocity [12]–[14]. Also, ballasting agent has low surface charge density compared to

52 colloidal particles in water. Hence, the addition of ballasting agent will not chemically disrupt the  
53 interaction between the coagulant/flocculant and colloidal particles [14].

54 Some of the main advantages of ballasted flocculation include reduced system footprint,  
55 elevated start-up time, higher settling rate, and minimized usage of coagulant and flocculant [15].  
56 Furthermore, the quality of water produced has been reported to be equal to or better than the  
57 conventional coagulation-flocculation process [11]. Due to the advantages of the ballasted  
58 flocculation process, it was previously employed commercially for drinking water treatment and  
59 in recent years, has been further used for combined sewerage overflow (CSO) and wastewater  
60 treatment plants [16]. The treatment efficiently reduces hydraulic retention time and increases  
61 overflow rate [17]. For instance, ballasted flocculation enhanced water quality and reduced runoff  
62 volume from the wastewater treatment plant, which prevented the discharge of nutrients, organic  
63 matter and debris into local waterway [8]. As such, it was reported that injecting sand with cationic  
64 polymer could provide a higher phosphorus removal efficiency which is the main cause of  
65 eutrophication and algal growth in the river [18]. This was possible because ballasted flocculation  
66 significantly shortened the time for flocs clarification and sedimentation, allowing the treatment  
67 plant to receive more inflow water for treatment.

68 Some of the recent works reported on ballasted flocculation investigated the characteristics  
69 of flocs formed under various operating conditions [10], [19], [20]. For instance, Young et. al  
70 studied the influence of numerous factors affecting the efficiency of ballasted flocculation  
71 separately [14]. Ghanem et. al studied the mechanism of ballasted flocculation through different  
72 tests: bench scale observations, microscopic observations, density tests, and centrifugal settling  
73 tests [13]. Besides, Sieliechi et. al (2016) used pozzolana as the alternative ballasting agent in their  
study with attention given to the floc compaction in the presence of ballasting agent [12].

However, reports on the development of fundamental understanding about the interaction between  
the ballasting agent and polymer flocculant remain scarce. Such understanding could provide  
insight into the impact of the ballasted agent and polymer flocculant on the characteristics and  
formation of flocs. In this context, the objective of this study was to investigate the performance  
of ballasted flocculation in removing turbidity using different dosages of sand and polymer  
flocculant. Accordingly, the impact of ballasting sand on the coagulation-flocculation mechanism  
(formation of flocs) was investigated by observing the flocs formed during the ballasted  
flocculation process. 92

93 **2. Materials and methods**

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95 *2.1 Materials*

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97 The chemicals used in this study: humic acid, kaolin, ferric chloride (FeCl<sub>3</sub>), calcium  
98 chloride (CaCl<sub>2</sub>·2H<sub>2</sub>O), sodium bicarbonate (NaHCO<sub>3</sub>), sodium chloride (NaCl), hydrochloric acid  
99 (HCl) and sodium hydroxide (NaOH) were of analytical grade and purchased from Sigma Aldrich,  
100 Malaysia. The polymer-based cationic flocculant (Zetag 8165) with a charge density of 40-50%  
101 was supplied by BASF, Singapore. The characteristics of Zetag 8165 was summarized in Table 1.  
102 The ballasting agent used in this study was silica sand with a mean diameter of 75 µm.

103

104 **Table 1**

105 Characteristics of Zetag 8165

<b>Form</b>	Powder
<b>Odour</b>	Odourless
<b>Colour</b>	Off white
<b>pH</b>	3.5-4.5

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107 *2.2 Preparation of synthetic water*

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109 All the solutions used in this study were prepared by using ultrapure (UP) water with a  
110 quality of 18 MΩ·cm<sup>-1</sup>. Humic acid and kaolin were used to represent NOM and suspended solids  
111 in water, respectively. The synthetic water was prepared by dissolving 0.1 g of humic acid powder  
112 in 10 ml of 0.1 M NaOH under continuous stirring for 1 hour. A salt mixture (80 ppm NaCl, 200  
113 ppm CaCl<sub>2</sub>·2H<sub>2</sub>O, and 170 ppm NaHCO<sub>3</sub>) was then added to the humic acid solution to mimic the  
114 presence of minerals in the water. Sodium hydrogen carbonate was added to provide carbonate  
115 alkalinity similar to that of natural rivers [21]. UP water and kaolin were then added to alter the  
116 turbidity to 30 NTU. The pH of the prepared synthetic water was then adjusted to 7 by using NaOH  
117 and HCl. The zeta potential of the synthetic water was -29.9 mV.

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119 *2.3 Ballasted flocculation process*

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All experiments were conducted by using a jar test system (Model ZR4-6 Zhongrun Water, China) with 500 ml of prepared synthetic water for each run. The distance between the impeller and the bottom jar is 1 cm with total water depth of 7 cm. All experiments were repeated twice. Two control sets of coagulation processes: coagulation without flocculant and sand, and coagulation with flocculant but without sand, were carried out to provide benchmarking information for understanding ballasted flocculation process. The jar test setting for coagulation without sand and flocculant was rapid mixing at 100 rpm for 1 min and slow stirring at 30 rpm for 30 min. However, the jar test setting for coagulation with flocculant but without sand and also ballasted flocculation were rapid mixing at 200 rpm for 1 min and followed by a slow mixing at 30 rpm for 5 min.

The optimal coagulant ( $\text{FeCl}_3$ ) dosage for all experiments was fixed at 22 mg/L, as determined in the previous study [22]. The dosage of flocculant (Zetag 8165) and sand was varied at 1-4 mg/L and 1-4 g/L, respectively. Coagulant was dosed into the solution instantaneously once the rapid mixing was started, followed by flocculant and sand according to the sequence described by Lapointe & Barbeau [10]. Pictures of the flocs and time for most of the flocs to settle down when entering the slow mixing period was recorded. After the sedimentation period, the supernatant water was then extracted for turbidity and zeta potential analysis.

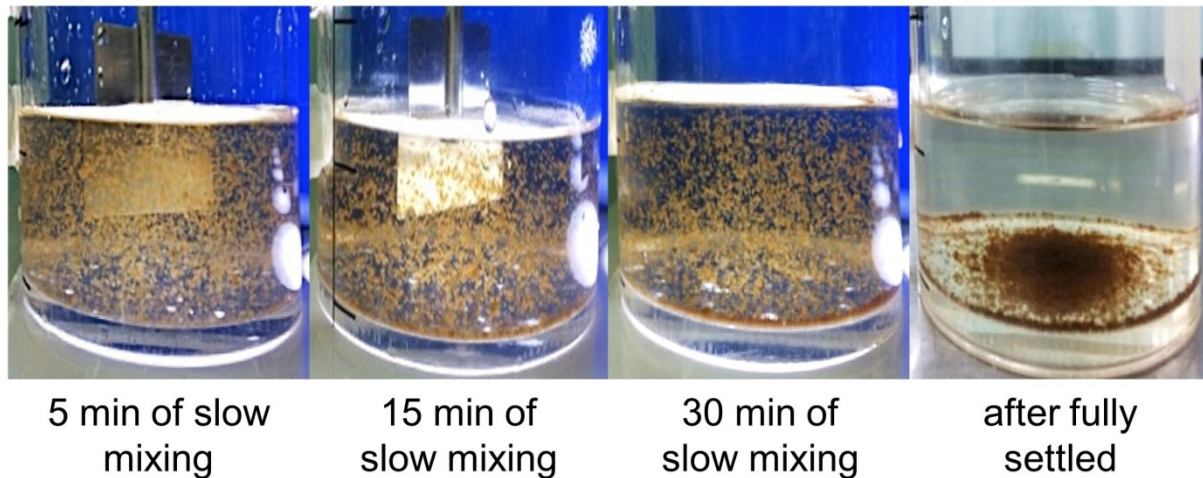
#### *2.4 Analytical methods*

The quality of treated water was measured from the removal of turbidity using Turbidimeter (2100N, HACH, USA). The particle charge before and after the ballasted flocculation processes was evaluated based on the zeta potential value using a Zeta-Sizer (Malvern, UK).

### **3. Results and discussion**

#### *3.1 Performance of coagulation-flocculation process*

150 In order to investigate the impact of sand as a ballasting agent on the formation of flocs,  
151 two control sets of coagulation-flocculation process without the addition of sand were conducted.  
152 For the first control experiment (with coagulant only), it has been observed that the flocs were  
153 gradually formed during the slow stirring period and grew larger with time (Fig. 1).  
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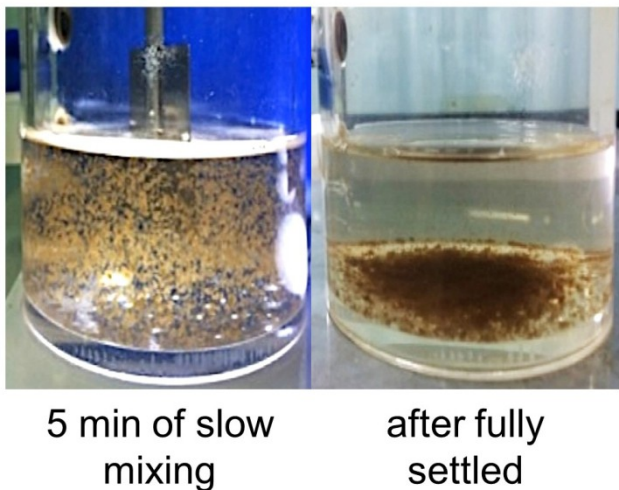


156 **Fig. 1.** Development of flocs with coagulant

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158 This observation could be explained by sweep coagulation mechanism as supported by zeta  
159 potential value in Table 2. As explained in the previous study, iron-based coagulant hydrolysed to  
160 form positively charged ferric hydroxides that partially neutralized the negatively charged particles  
161 and bound them together to form flocs [6], [23]. During the slow stirring period, small flocs swept  
162 across the solution and captured the residual suspended solids particle. At the same time, the flocs  
163 collided with each other and coalesced into larger aggregates, as indicated by the growing size of  
164 flocs in Fig. 1. A similar observation was reported by Ang et al. [22] where the flocs became larger  
165 and heavier due to collision between positively charged ferric precipitates and negatively charged  
166 suspended solids particles through the sweeping mechanism. Both phenomena resulted in the  
167 removal of more suspended solids and formation of larger flocs for subsequent removal via  
168 sedimentation. The time taken for the majority of the flocs to settle was more than 5 min, as  
169 tabulated in Table 2.

170 In comparison, the addition of flocculant in the second control set reduced the settling time  
171 of the flocs to around 2 min with the formation of larger flocs. Fig. 2 shows the development of  
172 flocs with the addition of flocculant in the coagulation-flocculation process. It could be observed

173 that the flocs in Fig. 2 are larger compared to the flocs without flocculant in Fig. 1. This is because  
 174 the flocculant acted as a binder which bound the suspended flocs together, further increased the  
 175 entanglement of flocs during the stirring period [24]. Therefore, this resulted in two consequences;  
 176 lower turbidity removal and shorter settling time. It was postulated that larger flocs had less  
 177 exposure to residual suspended solids in the solution due to its lower surface area and shorter  
 178 suspension period in the solution. Hence, the turbidity of the treated water with flocculant (2.36  
 179 NTU) was subsequently lower compared to without flocculant (0.69 NTU) as presented in Table  
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182  
 183 **Fig. 2.** Development of flocs with coagulant and flocculant

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 185 **Table 2**

186 Turbidity and zeta potential of control sets (with coagulant only, with coagulant and flocculant)

Synthetic water condition	Settling time (s)	Turbidity (NTU)	Zeta Potential (mV)
With coagulant only	> 300	0.69	-18.6
With coagulant and flocculant	> 120	2.36	-18.7

187 *3.2 Effect of flocculant dosage*

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189 The flocculant used in this study was Zetag 8165, which is a positively charged polymer  
 190 with zeta potential of 52.43 mV at pH 7. The success of the coagulation-flocculation process is  
 191 heavily reliant on the destabilization of the negatively charged suspended solid particles with a  
 192 high oppositely charged coagulant and flocculant [25]. The cationic polymer flocculant played a  
 193 significant role in the formation of flocs by destabilizing the repulsion forces between the particles  
 194 and interacted with the particles to form larger agglomerates [26].

195 Table 3 presents the turbidity of the supernatant under the influence of different flocculant  
 196 dosage (1-4 mg/L) and sand dosage fixed at 1 g/L. From Fig. 3, it was observed that the turbidity  
 197 removal increased from 35% to 90% with the dosage of flocculant increased up to 2 mg/L.  
 198 However, a further increment of flocculant dosage (>2 mg/L) did not help with the removal of  
 199 turbidity where the removal efficiency declined slightly (1-2 %). Indeed, as tabulated in Table 3,  
 200 the final turbidity of the supernatant solution under these dosages are within in the same range (3-  
 201 3.5 NTU). Hence, it could be concluded that the optimal dosage of flocculant in this case was 2  
 202 mg/L.

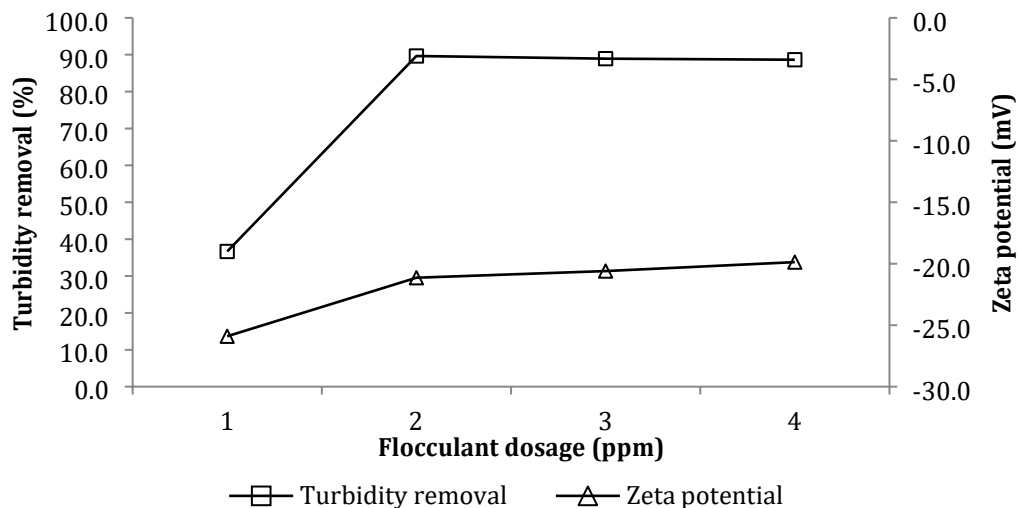
203  
 204 **Table 3**

205 Effect of flocculant dosage on settling time, turbidity, and zeta potential

Flocculant dosage (mg/L)	Settling time (s)	Turbidity (NTU)	Zeta Potential (mV)
1	4.01	19	-25.9
2	3.21	3.1	-21.1
3	3.55	3.32	-20.6
4	3.86	3.41	-19.9

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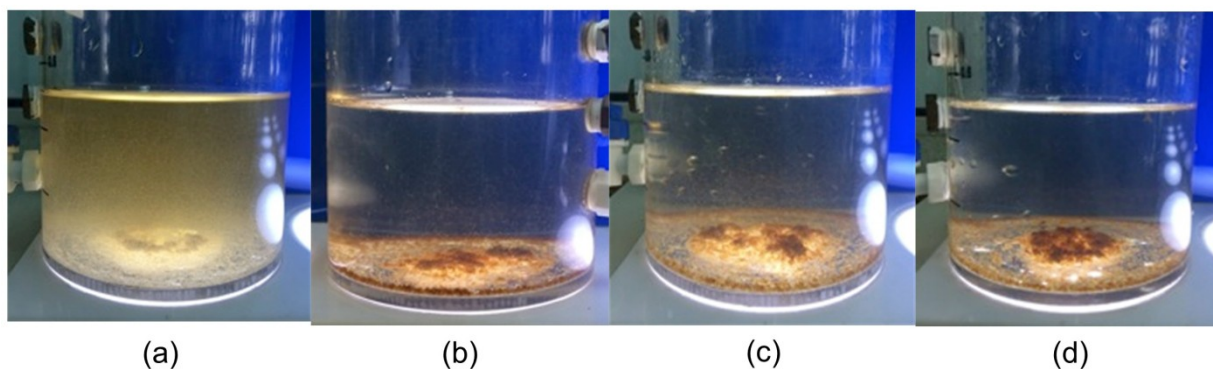
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208 **Fig. 3.** Effect of different flocculant dosage on turbidity removal and zeta potential

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210 Fig. 4 provides direct visual evidence on the quality of the supernatant after ballasted  
 211 flocculation at different flocculant dosages. It is visible that with 1 mg/L dosage of flocculant, the  
 212 supernatant appeared turbid with most of the suspended solids remained in the solution and only  
 213 small number of flocs settled at the bottom (Fig. 4(a)). On the other hand, the supernatant solutions  
 214 for flocculant dosage of 2-4 mg/L were much clearer with large flocs settled at the bottom (Fig.  
 215 4(b)-(d)). Such observation can be explained by the interaction between the flocculant and flocs  
 216 during the ballasted flocculation process. In this case, the zeta potential was used to understand  
 217 the interaction between the flocculant and suspended solids particles that eventually can be utilized  
 218 to explain the observed performance [27].

219



220

221 **Fig. 4.** Floc formations of different flocculant dosage after settling; (a) 1 mg/L (b) 2 mg/L (c) 3  
 222 mg/L (d) 4 mg/L

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224           With the addition of coagulant only (as discussed in the previous section), the zeta potential  
225 of the supernatant was around -18.6 mV. After the addition of 1 mg/L of flocculant and 1 g/L of  
226 sand, the final zeta potential was -25.9 mV, which was more negative compared to the control set.  
227 It could be attributed to insufficient of flocculant dosage to weaken the charge repulsion and bind  
228 the particles together [28]. Consequently, most of the suspended solids were still present in the  
229 solution and resulted in a turbid supernatant, as shown in Fig. 4(a). Some flocs were formed in this  
230 ballasted flocculation process, but the size and amount were obviously smaller and lesser  
231 compared to other flocculation scenarios. The settling time for the majority of the flocs was around  
232 4 seconds, which was not much different from the rest.

233           The zeta potential values for the ballasted flocculation process with dosages 2-4 mg/L were  
234 within the range of -20 mV. The more positively zeta potential charge showed that the cationic  
235 polymer flocculant played a role in the formation of flocs by weakening the repulsion charge  
236 (negatively charge) on particles and bound the particles together to form larger flocs [26]. The  
237 settling time for most of the flocs during the slow mixing period was within 3-4 seconds. It was  
238 considerably fast compared to the control sets without flocculant and sand (>300 seconds) and  
239 with flocculant only (>120 seconds). The incorporation of sand in the flocs had increased both its  
240 size and density, hence reduced its settling time [29]. This reflected the benefit of ballasted  
241 flocculation, which can significantly shorten the settling time for flocs sedimentation and  
242 clarification. However, the improvement in settling time had compromised the final quality of the  
243 supernatant, where the final turbidity for ballasted flocculation was around 3 NTU, slightly higher  
244 than 1 NTU when only coagulation process was employed. This is because the flocs embedded  
245 with sand in ballasted flocculation appeared to be much larger, thus reducing the collision rate  
246 with the residual turbidity due to faster settling rate and lesser contact frequency between the flocs.  
247 Fig. 4 shows that the size of the flocs grew larger with the flocculant dosage, corresponding well  
248 with the postulation where the larger flocs had lesser chance to capture the residual turbidity and  
249 resulted in lower turbidity removal.

250           In previous studies, it was reported that  $\text{FeCl}_3$  coagulation process removed the NOM and  
251 suspended solids by sweep coagulation mechanism [6], [30]. Under this mechanism, the coagulant  
252 formed high positive charged hydroxides that captured the negative charged impurities. During the  
253 slow stirring period, the flocs swept across the solution, grew larger by enmeshing the other flocs,

254 residual NOM and suspended solids present in the solution. Subsequently, the flocs became  
255 heavier due to the incorporation of sand and settled faster during the sedimentation period [10].  
256 This is as in evidence by the large dark brown flocs settled at the bottom in Fig. 4 (b)-(d).

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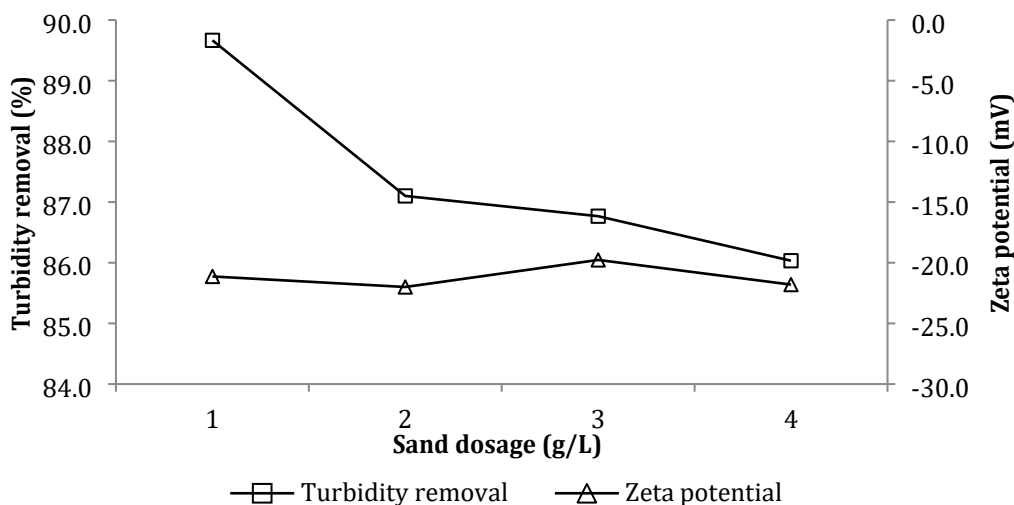
### 258 3.3 Effect of sand dosage

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260 Following from the previous section, the optimal flocculant dosage was fixed at 2 mg/L  
261 while the sand dosage varied from 1-4 g/L. The incorporation of polymer flocculant assisted the  
262 sand to attach to the flocs due to the binding property of the polymer [11]. Fig. 5 shows that the  
263 turbidity removal decreased slightly from 90 % to 86 % with the dosage of sand. Such trend was  
264 also observed by Sieliechi et al. that a slight deterioration of the residual turbidity was noted with  
265 the increased dosage of pozzolana particles as ballasting agent [12].

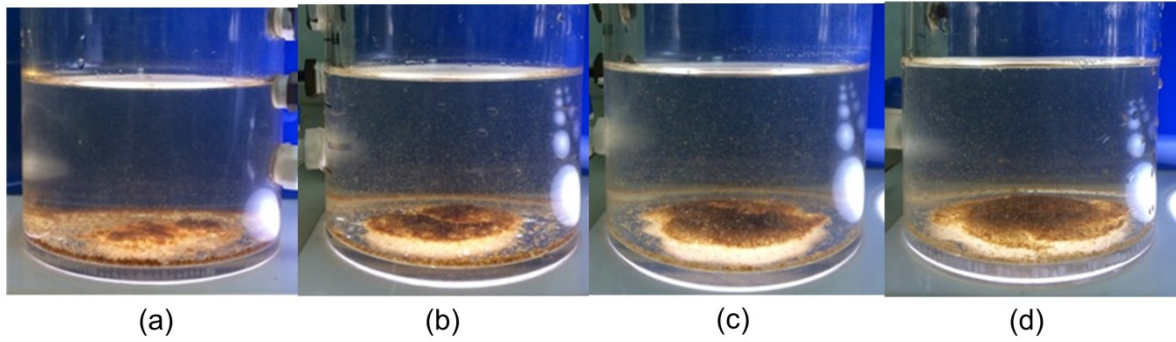
266 The increased dosage of sand resulted in the formation of smaller, isolated, and heavy flocs.  
267 As observed in Fig. 6, the flocs settled at the bottom of the jars shows an interesting pattern. The  
268 flocs formed for the sand dosage of 2-4 g/L appeared to be smaller, unlike the flocs in Fig. 6(a),  
269 which were in large chunk form. In addition, a greater number of flocs formed at sand dosage 2-  
270 4 g/L compared to sand dosage at 1 g/L, in which the flocs covered larger area of the jars. This  
271 observation indicates the role and impact of sand in the formation of flocs that would directly  
272 influence the turbidity and suspended solids removal.

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274

275 **Fig. 5.** Effect of different sand dosage on turbidity removal and zeta potential



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278 **Fig. 6.** Floc formations of different sand dosage after settling; (a) 1 g/L (b) 2 g/L (c) 3 g/L (d) 4  
279 g/L

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281 **Table 4**

282 Effect of sand dosage on settling time, turbidity, and zeta potential

Sand dosage (g/L)	Settling time (s)	Turbidity (NTU)	Zeta Potential (mV)
0	-	30	-29.9
1	3.21	3.1	-21.1
2	3.37	3.87	-22.0
3	3.81	3.97	-19.8
4	4.73	4.19	-21.8

283

284 Table 4 presents the zeta potential of the supernatant solution after the ballasted  
285 flocculation process. Generally, the zeta potential values fall in the same range regardless of the  
286 sand dosage. The result revealed that sand has no chemical interaction with the coagulant or  
287 flocculant as mentioned by Young et al. [14]. Instead, sand interacted physically with the flocs and  
288 resulted in the formation of flocs with a marked difference in size and quantity for the various  
289 dosage of sand. From the preliminary study, sand could not be embedded into the flocs without  
290 the dosing of flocculant. It was due to the binding property of flocculant that the sand could be  
291 incorporated into the flocs [11].

292 Based on the zeta potential value in Table 4, it could be deduced that sweep coagulation  
293 was the main mechanism for the removal of turbidity and suspended solids. This mechanism

294 utilizes the presence of premature flocs to sweep across the solution and captures more impurities  
295 before settling down for subsequent removal [22]. The aggregation of larger flocs from premature  
296 flocs was due to van der Waals attraction that facilitated the sweep coagulation mechanism [31].

297 The addition of excessive sand ( $> 1$  g/L) increased the presence of sand in the solution and  
298 thus enabled more sand to be embedded into the premature flocs. These flocs were small yet heavy  
299 due to the incorporation of sand. Consequently, the flocs tended to settle quickly, especially during  
300 the slow mixing period. With the diminishing of suspended flocs to sweep across the solution, the  
301 removal of residual particles was also reduced. Furthermore, the premature flocs also had lesser  
302 chance to collide with each other to form larger flocs as shown in Fig. 6(a). This is in good  
303 agreement with the result reported by Gasperi et al. that the addition of excessive sand dosage  
304 saturated the flocs and reduced particulate removal [17]. Hence, this explained the increased in  
305 final turbidity of the supernatant with sand dosage more than 1 g/L.

306 The settling time for the flocs is reported in Table 4. The settling time did not record any  
307 significant difference for all sand dosages. The slightly longer settling time (4.73 s) with 4 g/L of  
308 sand could be attributed to the remaining light premature flocs that did not have the chance to  
309 embed with the sand or collide with other flocs to form heavy agglomerates. Nonetheless, the time  
310 difference was too small to have any significant impact.

311 Overall, this study shows that the incorporation of ballasting agent has the potential to  
312 interrupt the efficiency of existing water and wastewater treatment (coagulation/flocculation)  
313 process. It was also noticeable that sweep coagulation mechanism required the flocs to sweep  
314 across the solution for a longer period in order to capture more suspended solid particles. The  
315 addition of sand increased the density of the flocs significantly and rapidly settled the flocs. With  
316 fewer flocs available to sweep across the solution, removal of impurities also declined. This  
317 indicated that the interaction between the ballasting agent and flocs should be considered during  
318 the design of ballasted flocculation process.

319

#### 320 **4. Conclusion**

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322 The results presented in this study provided understanding about the interaction between  
323 sand and flocs in the ballasted flocculation process. The optimal dosage for this study was 2 mg/L  
324 of flocculant and 1 g/L of sand that gave rise to 90% of turbidity removal. The settling time for the

325 ballasted flocs was much shorter (3 seconds) compared to non-ballasted flocs due to the  
326 incorporation of sand that caused the premature flocs to become heavy and settle quickly.  
327 However, the turbidity removal efficiency was slightly compromised due to presence of lesser  
328 suspended flocs to enmesh the residual turbidity during sweep coagulation stage. The impact of  
329 ballasting agent on the flocs formation and impurities removal is worth further investigation for  
330 other different coagulation-flocculation mechanisms with different types of coagulants and  
331 flocculants.

332

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334

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