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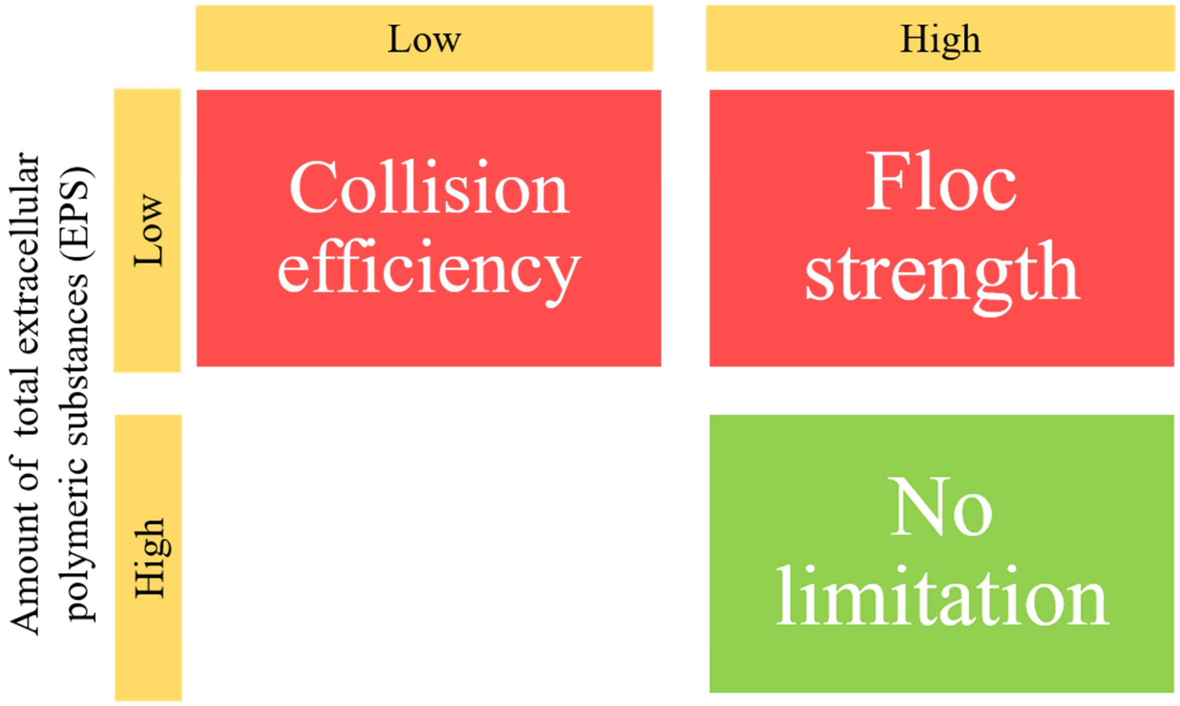
Limiting factors for activated sludge floc formation



Protein (PN) to polysaccharide (PS) ratio of EPS

Amount of total extracellular polymeric substances (EPS)	Low	High
Low	Collision efficiency	Floc strength
High		No limitation

Overcoming floc formation limitations in high-rate activated sludge systems

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Abstract

High-rate activated sludge (HRAS) is an essential cornerstone of the pursuit towards energy positive sewage treatment through maximizing capture of organics. The capture efficiency heavily relies on the degree of solid separation achieved in the clarifiers. Limitations in the floc formation process commonly emerge in HRAS systems, with detrimental consequences for the capture of organics. This study pinpointed and overcame floc formation limitations present in full-scale HRAS reactors. Orthokinetic flocculation tests were performed with varying shear, sludge concentration, and coagulant or flocculant addition. These were analyzed with traditional and novel settling parameters and extracellular polymeric substances (EPS) measurements. HRAS was limited by insufficient collision efficiency occurred because the solids retention time (SRT) was short and colloid loading was high and was predominantly caused by impaired flocculation rather than coagulation. In addition, the collision efficiency limitation was driven by EPS composition (low protein over polysaccharide ratio) instead of total EPS amount. Collision efficiency limitation was successfully overcome by bio-augmenting sludge from a biological nutrient removal reactor operating at long SRT which did not show any floc formation limitations. However, this action brought up a floc strength limitation. The latter was not correlated with EPS composition, but rather EPS amount and hindered settling parameters, which determined floc morphology. With this, an analysis toolkit was proposed which will enable design engineers and operators to tackle activated solid separation challenges found in HRAS systems and maximize the recovery potential of the process.

Keywords:

threshold of flocculation, limit of Stokesian settling, sludge volume index, clarifier, sedimentation, sewage, wastewater

1. Introduction

High-rate activated sludge (HRAS) systems have become a cornerstone in the pursuit of creating a more cost-effective and energy conservative wastewater treatment plant (WWTP). HRAS utilizes a short solids retention time (SRT) and high loading rate, and energy recovery is often maximized by capturing organics for anaerobic digestion and biogas production. To maintain the short SRT and functionality of the system, good solids separation and SRT control is imperative.

Historically, the performance of a clarifier has been assessed based on flux theory, where the main tipping point was driven by sludge loading rate (SLR) rather than surface overflow rate (SOR) (Vesilind 1968). Under normal operation, flux theory was a design parameter that allowed for sufficient capacity. However, ongoing clarifier performance has been found to be predominantly influenced by sludge floc formation behavior and thus driven by SOR (Mancell-Egala et al. 2017b). Therefore, effluent suspended solids concentrations have been dictated by the efficacy of floc formation and the presence of any limitation within the floc formation process. These limitations will often result in unexplained poor effluent quality. This is especially true for HRAS systems, where poor effluent quality has been cited (Rahman et al. 2016). Sludge lost through the effluent is not available for biogas production, thus solids separation is an important variable in the success of a HRAS system. Moreover, downstream processes like partial denitrification-anammox will be affected by elevated solids influx (Agrawal et al. 2018).

With respect to gravitational solids separation in activated sludge, floc formation has two main components: collision efficiency and floc strength. Collision efficiency can be defined as the percentage of total collisions that result in growth of the aggregate (Gregory and O'Melia 1989). Hydrodynamic shear (orthokinetic flocculation) increases the total number of collisions and has been reported to accelerate the flocculation rate (van Smoluchowski 1917). In practice, reactors and clarifiers typically are operated with suboptimal hydrodynamic shear (Parker et al. 2001). Therefore, collision efficiency can be a determining factor for floc formation due to the impairment of the flocculation rate. Beyond hydrodynamic shear, multiple factors can contribute to a collision efficiency limitation, low protein (PN) to polysaccharide (PS) ratio in the EPS (Li and Yang 2007), high organic loading rate (Barbusinski and Koscielniak 1995), SRT (Bisogni and Lawrence 1971), temperature, and unfavorable wastewater characteristics (Roberts 1975). These conditions can often be found in HRAS systems.

Floc morphology is measured by its fractal dimension number, which increases with increasing floc compactness (Meakin 1988). When collision efficiency is low and flocculation rate is sufficiently hampered, small spherical flocs with a high fractal dimension were formed (Aubert and Cannell 1986). When collision efficiency is adequate and the flocculation kinetics can be considered non-limiting, flocs with a lower fractal dimension are typically formed (Gregory and O'Melia 1989). Mechanistically, flocs will break when the tensile energy surpasses the bonding energy (large-scale fragmentation) or will slough small particles from the surface due to tangential shear (surface erosion) (Jarvis et al. 2005). Large, irregularly shaped flocs with a corresponding low fractal dimension undergo large-scale fragmentation while flocs with high fractal dimension are more effected by surface erosion (Yeung and Pelton 1996).

Collision efficiency is not directly quantifiable and is commonly determined by calibrating the collision efficiency parameter within a flocculation model (Lawler 19993). As a macroscopic and experimental metric, the threshold of flocculation (TOF) methodology has been developed to approximate collision efficiency for activated sludge samples (Mancell-Egala et al. 2017a), and TOF has been subsequently linked to clarifier performance (Mancell-Egala et al. 2017b). However, TOF does not mechanistically pinpoint a coagulation versus flocculation limitation. A better mechanistic understanding is needed to effectively overcome and prevent coagulation or flocculation limitations.

The impact of floc strength on the day to day operation of clarifiers is unclear. Floc strength limitations are theorized to only emerge when collision efficiency is adequate. Furthermore, the correlation between poor effluent quality and floc strength limitation is not straightforward. The limitation might emerge infrequently and lead to unexplained spikes in effluent suspended solids (Mancell-Egala et al. 2017b). Moreover, no standardized way to measure floc strength has been proposed, which makes linking the limitation to operational conditions difficult (Jarvis et al. 2005).

Activated sludge floc formation is mediated through EPS, which act as a biopolymer where double layer compression and bridging can take place. Multiple studies have suggested that the structure and composition of EPS is one of the main factors affecting floc formation, citing total amount and the protein (PN) over polysaccharide (PS) ratio being crucial to floc formation (Li and Yang 2007, Liu et al. 2010, Wilen et al. 2003). Floc formation can be artificially induced or improved by the addition of chemicals like metal salts and synthetic polymers (Böhm and Kulicke 1997, Metcalf and Eddy 2003). (Poly)electrolyte chemicals can also be classified by how they interact with colloids. Particle destabilization in the coagulation step can be achieved by adding ferric chloride or a high charge density, polyDADMAC-type polymer. Polyamide type polymers are often linear to maximize their molecular weight, thus minimizing dosage and enhancing the effect of bridging. Branched polyamide polymers are often used to improve floc strength (Bratby 2006). Given these different interactions, different types of (poly)electrolytes could potentially be used to pinpoint coagulation, flocculation, or floc strength limitations in sludge.

Remedies for floc formation limitations currently in use are selectors (Chudoba et al. 1973), flocculation zones (Federation 2005), bioaugmentation (Mancell-Egala et al. 2017b), and addition of chemicals, such as polymers and oxidants (Federation 2005). However, implementation of these techniques might give unsatisfactory results in HRAS systems if the predominant limitation is unknown. An evaluation of research and performance reports showed that comprehensive approaches to pinpoint specific floc formation limitations for activated sludge have yet to be identified. Therefore, the aim of this work was to determine floc formation limitations in three full-scale activated sludge reactors based on polymer addition, EPS characterization, and conventional and novel (TOF, limit of Stokesian settling…) settling parameters. Finally, flocculation limitations that emerged from the analysis were linked to process conditions in order to recommend approaches to overcome and prevent the limitations.

2. **Materials and Methods**

2.1 Activated sludge reactors and sampling

Blue Plains Advanced Wastewater Treatment Plant is one of the largest advanced sewage treatment plants in the world, treating over 1.1 million cubic meters of sewage per day and serving the District of Columbia and parts of Maryland and Virginia in the USA. Samples for this study were obtained from two secondary HRAS systems (HRAS and HRAS+; both with an SRT of 1-2 days). and one biological nutrient removal (BNR) reactor (SRT = 20-30 days). Operational conditions of the two HRAS systems were similar, with the exception that HRAS+ was bioaugmented with BNR sludge**.** This was implemented in 2007 to allow for more nitrogen removal in the high-rate activated sludge system (Bailey Jr et al. 2008). A full detailed description of these reactors can be found in Supplemental A and Mancell-Egala et al. (2017b). The most important operational conditions are summarized in Table 1. Samples from the mixed reactors were collected from June to August 2016. All experiments were performed within a few hours after sampling.

2.1 Conventional and novel settling metrics

Sludge volume index (SVI) and initial settling velocity (ISV) were determined at 3.5 g TSS/L in a Nalgene® two liters settleometer according to standard methods (APHA 2005). The Kinnear limit of Stokesian settling (LOSS) coefficient determined the sludge concentration where flocculent settling transitioned into hindered settling and was measured according to Mancell-Egala et al. (2016). Threshold of flocculation (TOF) measured the minimal sludge concentration required for settleable flocs to form when subjected to two minutes flocculation and settling time, which corresponds to a critical settling velocity (CSV) of 1.5 m/h. Six concentrations from 100 mg/L to 1000 mg/L were prepared. Detailed modus operandi can be found in Mancell-Egala et al. (2017a).

2.2 Polymer types and preparation

Ferric chloride (Fisher Scientific, USA) and polydiallyldimethylammonium chloride (PolyDADMAC) polymer (SNF Polydyne FL-4520, USA) were used as coagulants. PolyDADMAC was a low molecular weight cationic polymer with high charge density (not further specified by manufacturer). PolyDADMAC and $FeCl_3$ were freshly diluted to 0.2% w/w using the company provided stock media on the same day of the experiment.

Two polymers were used for flocculation: (1) a linear cationic polyamide polymer with a high-molecular weight and 10% charge density (SNF Polydyne, Clarifloc SE-1163, USA), and (2) a medium-molecular weight branched cationic polyamide polymer with 10% charge density (SNF Polydyne, Clarifloc C-3220, USA). Linear and branched polymer solutions (0.2% w/w) were prepared and activated on the same day as the experiment by slowly adding the polymer granules in deionized water and stirring the solution at 300 rpm for 30 minutes to activate the polymer.

2.3 Jar test methodology

The standardized jar test (ASTM 1995) was modified to appropriately represent the settling velocity distribution and flocculation behavior of sludge rather than the conventional effluent suspended solids measurement after 30 minutes of settling (Mancell-Egala et al. 2017a). Diluted sludge was poured into a modified Nalgene® 4L graduated cylinder (ø = 10 cm) and mechanically mixed at 245 s^{-1} (500 rpm) for 10 seconds with an IKA Eurostar 60 (IKA, USA) mixer, equipped with two 4-bladed axial flow impellers, after polymer was added. Subsequently, the sludge was agitated at 112 s^{-1} (300 rpm) for 30 seconds to enmesh the polymer within the flocs. When two polymers were added, these two steps were repeated for each polymer. Mixing was throttled down to 22 s^{-1} (100 rpm) for 10 minutes to allow for flocculation. Ten minutes was chosen because this was sufficient for floc formation and breakup to come to an equilibrium in previous studies (Biggs and Lant 2000, Mancell-Egala et al. 2017a, Wahlberg et al. 1994). The graduated cylinder was instantly baffled with a plastic plank after 10 minutes to dissipate kinetic energy and sludge was allowed to settle. After one minute, clamps located five centimeters below the liquid level were opened, and sludge was allowed to rapidly drain into a sample cup within about five seconds. The TSS collected represented the fraction of total TSS that settled slower than 3 m/h. This test was used as the basic procedure for creating the orthokinetic flocculation curve (section 2.3.1), polymer response curve (section 2.3.2), and the settling velocity distribution (Section 2.3.3).

2.3.1 Orthokinetic tests

Orthokinetic tests were used to assess the floc formation at different concentrations under non-rate-limiting conditions. The modified jar test was used at different sludge concentrations ranging from 100 mg TSS/L to 1500 mg TSS/L, thus in the flocculant settling range (below the Kinnear LOSS coefficient). Optimal polymer doses were spiked in these tests after determination using the polymer response curve (see 2.3.2). The control curve was subjected to the same protocol without the addition of polymer to show the individual effect of rapid mixing.

2.3.2 Polymer response curves

A polymer response curve was established to assess the influence of polymer concentrations on floc formation. An orthokinetic curve without the addition of polymer was created prior to the test. The sludge concentration where 20% of the sludge was removed was chosen as the constant sludge concentration to be exposed to different polymer doses. At this concentration, floc formation was deemed sufficiently limited to ensure resolution for the effect of polymer dosage to be observed.

2.3.3 Settling velocity distribution test

A discrete settling velocity distribution of the sludge was obtained by subjecting the sludge to a range of settling velocities through different settling times: 5 min

(CSV = 0.6 m/h), 2 min (CSV= 1.5 m/h), 1 min (CSV = 3 m/h), and 20 s (CSV = 9 m/h) (Mancell-Egala et al. 2017a). Settling velocity distributions were obtained at the same sludge concentration as the polymer response curves. To assess the impact of shear on the settling velocity distribution, both 22 s^{-1} (100 rpm) or 91 s^{-1} (260 rpm) were applied for 10 minutes as a flocculation step.

2.4 Floc breakage factor

The floc breakage factor determined the sensitivity of the sludge towards increasing velocity gradients. This was captured in a single number by modifying a protocol developed by Leentvaar and Rebhun (1982). Sludge was diluted to concentrations below TOF to minimize the impact of reflocculation during the settling phase and subjected to increasing velocity gradients (22 – 320 s^{-1}) in the same 4 L cylinder as the modified jar tests were performed in. After 10 minutes of mixing, the sludge was baffled and allow to settle for 2 minutes (CSV = 1.5 m/h), where after the effluent was collected for solids measurements and compared to the initial concentration. The two minutes settling condition was chosen to be similar to the TOF method under gravitational flocculation conditions. The floc breakage factor was defined as the slope of a log-log transformation of the $\%TSS_{>1.5m/h}$ as a function of increasing velocity gradient $(\ln(\%TSS_{>1.5m/h})/\ln(TSS_{initial}))$.

2.5 Extraction of extracellular polymeric substances (EPS)

Loosely bound (LB) and tightly bound (TB) EPS fractions were extracted using a heat extraction method modified after Li and Yang (2007). The LB fraction was vortexed for 1 min at 60 °C where after the sludge was centrifuged for 10 min at

4000g, and the supernatant recovered. The pellet was subsequently used to extract the TB fraction with a 30 min incubation at 60 °C and centrifugation for 15 min at 4000g. The extraction was standardized on 25 mg TSS. Both LB and TB EPS were filtered through a 1.5 μm glass microfiber filter (Whatman, USA) and stored at -20 °C. The EPS fractions were analyzed for chemical oxygen demand (COD), proteins (PN), and polysaccharides (PS).

TSS was measured according to standard methods (APHA 2005). COD was determined using Hach® (Loveland, Colorado, USA) kits following the manufacturer's instructions. Protein content was determined using the modified Lowry Protein Assay kit (Thermo Fisher, USA) (Lowry et al. 1951) with bovine serum albumin (BSA) as the standard. Polysaccharide level was determined using the DuBios method with glucose as the standard (DuBois et al. 1956).

2.6 Statistics

Statistical significance between treatments was determined with an unpaired t-test where unequal variances were assumed due to the small sample size. To determine the statistical significance between slopes, three different slopes were calculated using linear regression at the initial linear part of the curve, and an unpaired t-test was performed on the resulting slopes in Microsoft Excel. T-tests with a p-value <0.05 were considered statistically significant.

3. Results

Two HRAS and one BNR system were assessed for their floc formation behavior and subsequent possible limitations. Table 1 gives an overview of the most important performance parameters and operational conditions. A detailed description can be found in the supplemental information.

3.1 Intrinsic settling performance

HRAS showcased the poorest performance in terms of effluent quality of the three reactors assessed, followed by HRAS+ and then BNR. This was echoed by the gravitational (TOF) and orthokinetic flocculation curves (Figure 1). All flocs settled slower than 1.5 m/h below 535, 369, and 295 mg TSS/L for HRAS, HRAS+, and BNR, respectively. Increasing the TSS concentration introduced more collisions, leading to flocs faster than 1.5 m/h, therefore reducing the effluent TSS. The high threshold concentration for HRAS indicated poor intrinsic collision efficiency (Figure 1).

Collision efficiency was improved when HRAS was bio-augmented with BNR sludge, resulting in a lower TOF number for the HRAS+ sludge (Table 1). Shifting from HRAS to HRAS+ significantly increased the SVI while the ISV dropped (Table 1). This indicated a change in hindered settling dynamics. The limit of Stokesian settling (LOSS) decreased when BNR sludge was seeded into HRAS, indicating that floc-floc interactions became more significant at lower TSS.

When orthokinetic flocculation was induced, flocs faster than 3 m/h were observed for all sludge types at the lowest TSS tested (Figure 1). The solids fraction with velocities below 3 m/h decreased steadily with increasing concentrations. A balance between maximum floc formation and ~~floc~~ breakup was achieved and the orthokinetic curve flattened out (Figure 1). Here, BNR sludge produced a higher percentage of flocs travelling faster than 3 m/h

Bio-augmentation of HRAS sludge produced weaker flocs than the HRAS or BNR sludge alone as the floc breakage factor decreased (Table 1). When the sludge was subjected to 91 s^{-1} of shear stress, HRAS+ shifted more significantly than the other sludge types from the 3-9 m/h range to lower velocities (Figure 2). BNR sludge was resistant to the elevated shear, as the floc size distribution hardly changed.

The SVI was significantly higher for HRAS+ compared to HRAS while the ISV dropped (Table 1). This indicated a change in hindered settling dynamics. LOSS decreased when BNR sludge was seeded into HRAS, indicating that floc-floc interactions became more significant at lower TSS. As such, the sludge would enter a hindered settling regime at lower TSS concentrations.

3.2 Extracellular polymeric substances (EPS)

Both HRAS and HRAS+ had similar amount of EPS, whereas a significantly higher amount was determined for BNR (Table 1). However, HRAS had a considerably lower amount of PN/PS ratio in the loosely bound EPS fraction compared to the bioaugmented variant and BNR, which shared a similar composition.

3.3 Polymer response curves

The floc formation response to different concentrations of polymer was assessed at a sludge-specific fixed TSS where 20% of the flocs settled faster than 3 m/h (see 2.3.2). The latter TSS was determined to be 355 ± 19 mg TSS/L, 506 ± 19 mg TSS/L, and 439 ± 46 mg TSS/L for HRAS, HRAS+, and BNR respectively.

HRAS responded to $FeCl_3$ addition at the lowest concentration tested (0.05 g Fe^{3+}/kg TSS), but failed to improve floc formation with increasing dosages (Figure 3B). $FeCl_3$ had no effect on HRAS+ or BNR, indicating that Fe^{3+} particle destabilization played a minor role in the floc formation process (Figure 3E/H). Addition of polyDADMAC only marginally improved floc formation at higher dosages on HRAS and HRAS+, but did induce a significant improvement for CAS. While polyDADMAC increased linearly for HRAS and CAS, a very high dosage (1 g polymer/kg sludge) was required to induce the enhanced floc formation for HRAS+. Bridging effects, rather than charge neutralization, presumably induced the floc formation as polyDADMAC is considered a low molecular weight polymer.

Additon of both linear polymer (LP) and branched polymer (BP) both showed an increase in the formation of flocs that settled faster than 3 m/h (Figure 3A/D/G) for all sludge type tested. As such, flocculation could be improved by inducing polymer-floc bridges. At the maximum dose, the linear polymer was most effective on BNR**,** while HRAS and HRAS+ responded similarly. No significant difference in floc formation was observed between the two flocculants.

Combining 0.5 g polyDADMAC/kg TSS with increasing dosages of linear polymer did not further improve floc formation at low concentrations as indicated by the similar initial slope to linear polymer alone (Figure 3C). However, maximum floc formation was reached at 0.3 g linear polymer g TSS instead 1 g linear polymer/g TSS, indicating a synergistic effect. The initial slope of percent improvement with polymer dose did increase for HRAS+, indicating that less linear polymer was required to achieve the same amount of fast settling flocs (Figure 3F). In the case of BNR, polyDADMAC combined with linear polymer, induced an initial sharp increase in floc formation at a low dosage (Figure 3I). However, the slope quickly flattened out at 54 ± 6 % at 0.1 g linear polymer/kg TSS and remained constant. This was most likely because of steric or electrostatic interference of both polymers.

3.4 Orthokinetic tests

Dosing 0.5 g polyDADMAC/kg TSS did not yield any improvement in floc formation at increasing TSS concentrations for HRAS or HRAS+ compared to the control, whereas BNR did achieve a higher production of fast settling flocs (> 3 m/h) per unit of TSS (-123 ± 7 %TSS/g TSS) (Figure 4A/C/E). This reiterated the polymer response curves where similar results were obtained.

Linear and branched polymer had a significantly positive effect on all sludge types. HRAS showcased a similar effect for both linear and branched polymer compared to the control, since their slopes were not significantly different from each other (Figure 4B). However, at 1555 mg TSS/L, branched polymer significantly (p-value = 0.02) outperformed linear polymer in removal percentage as fewer flocs settled than 3 m/h, indicating formation of bigger or faster flocs.

For HRAS+, branched polymer addition had a significantly larger effect on the orthokinetic profile than linear polymer addition due to a steeper slope (p = 0.0001), thus indicating that larger or denser flocs were formed at lower concentrations (Figure 4E). However, this advantage disappeared when the sludge concentration reached 1000 mg TSS/L, resulting in similar maximum removal potential (LP = 6.0 ± 0.8 %, BP = 6.3 ± 0.5 %). Addition of linear polymer caused the formation of faster settling flocs at the lowest TSS tested for HRAS+ compared to HRAS (Figure 4B/E), however both were outcompeted by BNR. Linear polymer outperformed branched polymer at low TSS concentrations for BNR but achieved similar maximum removal potentials.

Combining polyDADMAC with linear or branched polymer did not improve the response of any sludge type tested. Whereas HRAS and HRAS+ was indifferent towards the extra addition of polyDADMAC (Figure 4C/F), fewer BNR flocs settled faster than 3 m/h at lower TSS concentrations (Figure 4I). This indicated an interaction between the coagulant and flocculant, which were also observed in the polymer response curve (Figure 3I).

3.5 Impact of shear & settling velocity distributions

HRAS and BNR sludge types did not experience any impact from increased shear on their settling velocity distribution performed at 351 ± 24 and 472 ± 29 mg

TSS/L, respectively. Bio-augmenting BNR in HRAS made the sludge more prone to breakup, as the number of flocs travelling faster than 3 m/h at 493 ± 31 mg TSS/L dropped from 26% to 7% in HRAS+ (Figure 5).

Addition of 0.5 g polymer/kg TSS significantly increased the fraction of all sludge types settling between 3 and 9 m/h (Figure 5). Linear polymer and branched polymer performed similarly in terms of overall change in settling velocity distribution at low shear. Increasing shear from 22 s^{-1} to 91 s^{-1} did not affect the sludge treated with branched polymer because this type of polymer is designed to increase floc strength. The distribution of HRAS and BNR remained unchanged when conditioned with linear polymer and subjected to 91 s^{-1}. In contrast, HRAS+ gained a significant fraction of slow settling flocs (< 3 m/h), indicating a higher rate of floc breakup. This dissimilar behavior between conditioning with linear or branched polymer implied the formation of weaker flocs compared to other systems, where no such dissimilarity was observed. The addition of polyDADMAC in combination with linear polymer induced a similar effect at the left tail of the distribution compared to linear polymer (Figure 5). Similar observations were made for HRAS+ and BNR in the case of branched polymer combination, while HRAS had a significantly lower amount of fast-settling sludge with that scenario. HRAS produced very fast-settling flocs (> 9 m/h) when conditioned with a combination of polyDADMAC and linear polymer. These flocs were prone to breakup as they deteriorated to (settling < 9 m/h) when a higher shear regime was applied.

4. **Discussion**

4.1 Importance of collision efficiency in solids separation

Coagulation limitation was minor to non-detected in all three reactors as indicated by the ineffectiveness of both $FeCl_3$ and polyDADMAC. The influent of the high-rate activated sludge reactors was pretreated by chemically enhanced primary treatment (CEPT), removing most of the negatively charged particles from the wastewater. Despite the CEPT pretreatment, coagulation could further be enhanced in HRAS, indicating a limitation was still present, as shown by the minor effect (5 to 20% improvement) of $FeCl_3$ and polyDADMAC in Figure 3A.

The minor effect of $FeCl_3$ on HRAS was presumably caused by residual neutralization of influent particles and dewatering return liquid, which introduced a concentrated stream of charged colloids into the reactor. It should be noted that the CSV window (> 3 m/h) used in the orthokinetic tests might be too fast for coagulation, thus any improvement towards coagulation without alleviating the flocculation limitation would not have been captured. In addition, the sludge needs to flocculate further in order to settle out, masking the coagulation process.

The beneficial effect of polyDADMAC on BNR sludge could be explained by the intrinsic flocculation kinetics. BRN’s flocculation kinetics might have been fast enough to see a response in the sludge’s coagulation. Despite this experimental limitation, no difference in floc formation was observed when polyDADMAC was combined with flocculant polymer, compared to just polymer for HRAS and HRAS+

sludge. BNR sludge performed worse presumably due to polymer-polymer interactions. Additionally, as polyDADMAC was able to induce improved floc formation on BNR sludge, a combination with polymer and polyDADMAC resulted in steric interference.

Addition of Linear polymer was less effective for floc formation at very low TSS concentrations (< 250 mg/L) for HRAS than HRAS+ than BNR, compared to their respective controls (Figure 3B/E**).** This indicated that a limited number of total collisions was not able to produce flocs large or dense enough to concur the CSV applied. As such, bridging was more successful for CAS than HRAS, indicating a flocculation limitation. HRAS sludge was fed with CEPT effluent and dewatering solids return and was therefore most likely overloaded with colloids. This resulted in binding spots on the EPS to be occupied by substrate thus hampering bridging. Additionally, the low SVI reported was a symptom that floc-floc interaction was impaired and thus limited by bridging, resulting in ‘pinpoint’ floc formation. While no microscopy was performed in this study, the presence of pinpoint-like flocs was further supported by the high Kinnear LOSS coefficient and ISV, indicating little steric interference between the formed flocs (Mancell-Egala et al. 2017b). Furthermore, bridging impaired particles can form dense and compact structures (Gregory and O'Melia 1989), which has been shown in this and that study. This explained the dramatic increase in fast settling flocs when linear polymer was introduced (Figure 5A). The polymer most likely bridged the dense compact flocs together forming these fast settling flocs.

4.2 Impact of EPS on collision efficiency

Bridging of activated sludge is driven by EPS (Sobeck and Higgins 2002), which is influenced by reactor conditions such as the organic loading rate and SRT. HRAS sludge produced less EPS compared to the BNR system, which was in agreement with previous literature describing a positive relationship between EPS content and SRT (Sesay et al. 2006). Additionally, a positive relationship between substrate utilization rate and EPS amount was found for HRAS systems (Jimenez et al. 2015). The substrate utilization rate was low for the HRAS reactors assessed in this study due to low-strength wastewater, which most likely further contributed to the relatively low EPS content. HRAS sludge had a low PN/PS ratio in the loosely bound fraction of the EPS (Table 1). The repulsive forces between activated sludge cells have been attributed to LB-EPS (Li and Yang 2007), while another study found that LB-EPS was responsible for attractive forces (Liu et al. 2010). However, both agree on the relative importance of LB-EPS compared to TB-EPS in floc formation. A low PN/PS ratio has been accepted as an indicator for poor floc formation (Liao et al. 2001, Morgan et al. 1990), and thus might explain the poor flocculation of HRAS sludge. Most studies to date report a negative correlation between settleability and specific EPS amount (Sheng et al. 2010); however, these studies assess settleability in terms of SVI, a parameter which has been scrutinized to not reflect normal clarifier behavior (Mancell-Egala et al. 2016). These studies supported the results from this study, where the low EPS amounts (90 ± 23 mg COD/g VSS) and low SVI (88 ± 81 mL/g) reported in this study did not result in adequate clarifier performance.

BNR sludge operating at long SRT and low organic loading rate, had a higher EPS content and the highest PN/PS ratio while simultaneously achieving the best effluent quality. This supported that EPS composition is crucial in the floc formation process and is in line with literature showing a positive correlation between PN/PS and SRT (Sesay et al. 2006). Wastewaters with lower COD/N ratios fed to sludge have also been observed to produce more protein rich EPS (Durmaz and Sani 2001), which further explains the favorable PN/PS ratio. Proteins in the EPS are the main source of surface charge and hydrophobic pockets within the sludge and have been linked with enhanced floc formation and bridging. Hydrophobic interactions generally increase with increasing molecular weight of the polymer, hence the poorer performance for branched polymer (medium molecular) compared to linear polymer (high molecular) at low TSS concentrations where collisions are limited (Figure 4H). The limited amount of protein in the HRAS' EPS limit the effectiveness of hydrophobic interactions. This further explains the lower efficiency of linear and branched polymer at low TSS discussed in Section 2.1. Whereas the effect of the PN/PS ratio on collision efficiency is believed to be a continuum, no cutoff has been observed in this study due to a limited number of samples, hence a more rigorous approach testing more PN/PS ratios will be required to assess the transition from bad collision efficiency to good.

4.3 Importance of floc strength on solids separation

The HRAS+ reactors received the waste activated sludge from the BNR reactor. While the practice was originally intended to allow for some nitrification at short

SRT, operators noticed an improvement in effluent TSS quality compared to the identical, non-bioaugmented HRAS. Bioaugmentation of long SRT sludge pushed the LB-EPS PN/PS ratio higher, improving collision efficiency as indicated by TOF (and thus collision efficiency) approaching BNR. Interactions with polymer at low solids concentrations also improved, indicating a greater bridging affinity compared to HRAS sludge and higher collision efficiency. As such, bioaugmentation might have helped to alleviate the flocculation limitation present in HRAS sludge by providing a source of 'fresh' EPS with a composition favorable for bridging to take place. Bioaugmentation did not, however, increase the total EPS of the sludge, strengthening the hypothesis that collision efficiency (and thus TOF) is not dependent on the amount of EPS, but rather the composition. The total amount of EPS remained low, thus not enough EPS might have been available to create a strong floc. In addition, the improved collision efficiency and reduced flocculation limitation of HRAS+ led to flocs that were more sterically hindered as indicated by relatively high SVI, low Kinnear LOSS coefficient, and poor ISV. These conditions correlated with bigger and fluffier flocs as discussed in a previous study (Mancell-Egala et al. 2017a). (Mancell-Egala et al. 2017b) further explored the link between these parameters and floc structure in function reactor operation and found that bioaugmentation caused similarly high SVI, low LOSS and ISV, which corresponded to big and sterically hindered flocs.

Large, fluffy flocs have been known to form when sludge is subjected to high amounts of particulate substrate and when flocculation is not impaired (Wang et al.

2014), which might explain why HRAS+ sludge exhibits these traits while BNR sludge does not. Moreover, floc size has been positively correlated with increased loading rates (Barbusinski and Koscielniak 1995) and irregularly shaped flocs at short SRT (Liss et al. 2002). The formed flocs appeared to be less resistant to shear than their non-bioaugmented counterparts. Indeed, HRAS+ exhibited the highest shear sensitivity and change in settling velocity distribution when exposed to high velocity gradients. Furthermore, the addition of branched polymer was able to increase HRAS+' strength, while having no effect on other sludge types.

Neither the strength of HRAS nor BNR decreased when collision efficiency was artificially improved with (linear) polymer. As such, given that HRAS+ had the same total EPS amount as HRAS, but the same PN/PS ratio as BNR, EPS appeared to have a minor role in determining the strength of the floc. The high LOSS and SVI indicated sludge of increased size and decreased sphericity (Mancell-Egala et al. 2017a), the floc strength of HRAS+ might be determined by these characteristics. A mechanistic understanding of what determines the strength of flocs is scarce and studies on activated sludge are even scarcer, however there is some consensus that strength is negatively correlated to the size and sphericity (expressed as fractal dimension of the sludge) (Jarvis et al. 2005). The flocculation limitation in the form of subpar collision efficiency in high-rate activated could therefore be seen as a 'primary flocculation limitation' that, when overcome, might lead to a floc strength limitation because of the nature of the floc formation process.

4.4 Overcoming floc formation limitations: a toolkit

Limitations within the floc formation process are detrimental to the effluent quality and the overall effectiveness of the wastewater treatment process. Overcoming these limitations is therefore priority. For this reason a tool kit is proposed, which include**s** inducing a stronger feast-famine regime, an improved clarifier design, and bioaugmentation.

Collision efficiency seems to be driven by the nature of the EPS, as such managing EPS seems to be the predominant route to alleviate this flocculation limitation. EPS can be managed by imposing a feast-famine response**,** which is typically induced by anaerobic/anoxic selectors or the novel high-rate contact-stabilization process (Meerburg et al. 2015, Rahman et al. 2016). The former is typically used to control filamentous growth and improve settleability (as measured in SVI) for BNR systems (Chudoba et al. 1973). Alternatively, the feast-famine regime applied in contact-stabilization has been proven to induce an EPS production response, which correlates to improved effluent quality (Rahman et al. 2017).

Last, selection of an optimal SRT would be important to manage floc formation. Meerburg et al. (2016) established that in a high-rate contact-stabilization system, the PN/PS ratio was optimal at an SRT of 1.3 days and decreasing with decreasing SRT. Rahman et al. (2016) found that with a similar configuration, the optimal EPS-to-biomass ratio was set at 0.8 days and decreasing with decreasing SRT. As such, shortening the SRT will result in more net sludge production, but the lack of EPS quality and content might lead to worse effluent quality. This will result in a lower net capture of carbon sludge is lost through the effluent. The long-term role of EPS in the carbon balance at short SRT is still not fully understood. longitudinal study of the impact of EPS on the carbon balance will be of great value towards the optimization of HRAS systems.

When the sludge types were subjected to perfect conditions to allow for orthokinetic flocculation, no significant difference in flocculation behavior was observed. Optimizing flocculation zones to within the clarifiers to maximize orthokinetic flocculation might help with the management of the limitation and mitigate the impact on their performance. This remains challenging because these zones will be subjected to a wide range of inlet flows, changing the velocity gradient and residence time within the flocculation zone, as treatment plants generally can't control their incoming flow (Federation 2005). Bioaugmentation of long SRT sludge into a HRAS system will push the system from a collision efficiency limitation to a floc strength limitation, as given by the lower TOF, higher affinity to polymer, and weaker resistance to shear stress for HRAS+. However, not all treatment plants have access to both sludge types, which makes this solution impractical. A/B plants exhibiting a collision efficiency limitation within their A-step might benefit from this approach. Lastly, polymer addition can also overcome a floc formation limitation. Linear polymer can be useful in mitigating serious collision efficiency limitations. However, polymers are expensive and should, in the authors' opinion, only be used as a last resort.

Floc strength limitation is more erratic in nature because it will only show in the effluent suspended solids when shear forces become too extreme. Managing the loading of particulate COD with primary clarifiers might reduce the size and fluffiness of the floc, but this reduced load might make the optimization of HRAS systems more challenging (Rahman et al. 2016). Reducing shear swings within the inlet zone during wet weather events of the clarifier might also reduce the occurrence of spikes of effluent suspended solids in the clarifier. Dosing of branched polymer to overcome a floc strength limitation is tricky, as there was no consistent influence on effluent suspended solids observed. Lastly, selective retention of strong flocs outside of the reactor with the use of external selectors similar to the ones used in deammonification reactors for the retention of anammox bacteria (Han et al. 2016, Wett et al. 2010) might negate the floc strength limitation. Ultimately, the lack of a current mechanistic understanding of floc strength makes comprehensive approaches to alleviate said limitation challenging.

5. Conclusion

This study identified and differentiated limitations within the activated sludge formation process. The major conclusions were:

- HRAS systems at short SRT receiving a high colloid loading exhibited a primary collision efficiency limitation. This limitation was mainly driven by the low PN/PS ratio in the LB-EPS fraction rather than the total EPS amount.
- Overcoming insufficient collision efficiency while subjecting the sludge to high-rate conditions highlighted a second floc formation limitation: poor floc strength. This did not seem to be correlated with EPS composition, but rather low EPS amount in conjunction with structural properties of the flocs as

measured by the hindered settling parameters SVI, ISV and the Kinnear LOSS coefficient.

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Tables

Table 1. Full-scale reactor and clarifier performance as well floc and settling characteristics and composition for the high-rate activated sludge system (HRAS), bioaugmented HRAS (HRAS+) and nutrient removal system during the period of the study.

	HRAS	HRAS+	BNR	
Reactor performance (n>20)				
SRT	1.46 ± 0.41	1.32 ± 0.33	30 ± 21	*d*
Effluent TSS	33.1 ± 12.4	23.8 ± 28.9	7.0 ± 4.5	*mg TSS/L*
Reactor rates (n > 20)				
Influent organic loading rate	2.40 ± 0.73	2.61 ± 0.73	0.27 ± 0.08	*kg COD/kg TSS/d*
Waste liquor loading rate*	0.11 ± 0.07	0.31 ± 0.2	-	*kg TSS/kg TSS/d*
Soluble P loading rate	4.4 ± 1.4	9.4 ± 3.6	2.2 ± 1.0	$kg\ P/m^{3/}d$
Ferric dosage rate	31.7 ± 2.3	35.8 ± 3.4	-	$g\ Fe^{3+}/m^{3}/d$
Polymer dosage rate	0.05 ± 0.01	0.07 ± 0.02	0.1 ± 0.02	*g polymer/kg TSS/d*
Bioaugmentation rate	-	0.32 ± 0.09	-	*kg TSS/kg TSS/d*
Clarifier rates (n>20))				
Surface overflow rate	24 ± 2	25 ± 3	11 ± 1	$m^{3}/m^{2}/d$
Sludge loading rate	110 ± 29	73 ± 23	23 ± 4	$kg/m^{2}/d$
Floc formation parameters (n>3)				
TOF	535 ± 139	369 ± 60	295 ± 12	*mg TSS/L*
Floc breakage factor	-0.6 ± 0.3	-0.9 ± 0.3	-0.2 ± 0.2	*% TSS/gTSS*
LOSS	1706 ± 539	801 ± 259	1287 ± 307	*mg TSS/L*
ISV	3.37 ± 1.24	1.36 ± 0.95	2.29 ± 1.05	*m/h*
SVI_{30}^{**}	88 ± 81	154 ± 60	122 ± 46	*mL/g*
EPS characterization (n=3)				
Total EPS	90 ± 23	93 ± 6	135 ± 10	*mg COD/g VSS*

PN/PS Total EPS	1.63 ± 0.38	2.19 ± 0.96	2.00 ± 0.13	*mg BSA/mg glucose*
LB-EPS	8 ± 1	6 ± 1	16 ± 2	*mg COD/g VSS*
PN/PS LB-EPS	0.76 ± 0.85	1.85 ± 1.47	2.03 ± 0.76	*mg BSA/mg glucose*
TB-EPS	82 ± 22	87 ± 6	118 ± 8	*mg COD/g VSS*
PN/PS TB-EPS	1.98 ± 0.57	2.23 ± 0.74	2.01 ± 0.35	*mg BSA/mg glucose*

*** waste liquor is mixture of dissolved air flotation underflow and belt filter press filtrate which was high in colloidal particles.**

**** SVI30 was measured at 3.5 g TSS/L**

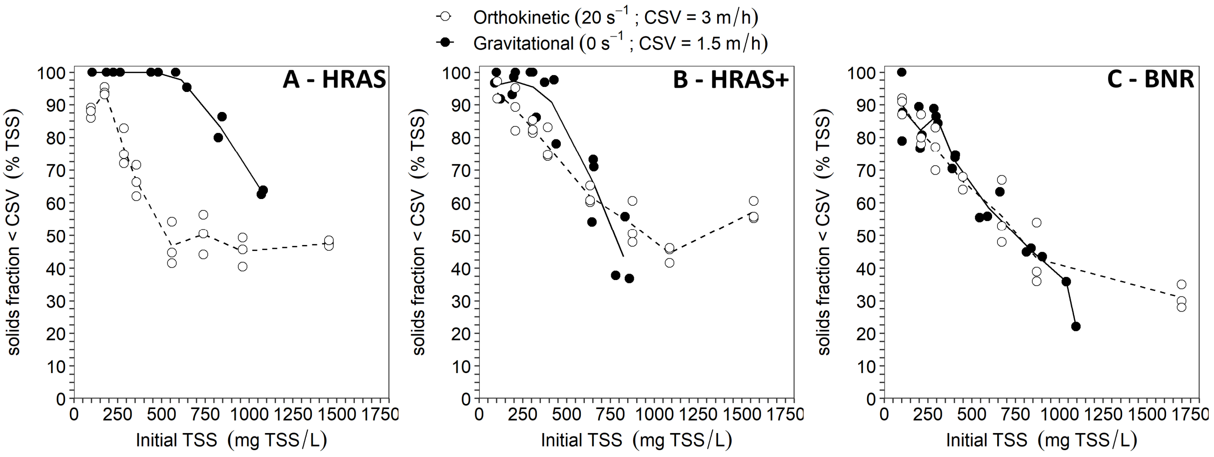
Figure 1. Orthokinetic and gravitational flocculation results for HRAS (A), HRAS+ (B), and BNR sludge (C) showing the effluent solids fraction with settling velocity lower than the applied critical settling velocity (CSV) in function of initial TSS concentration. Gravitational flocculation tests were performed at a cutoff CSV of 1.5 m/h, while orthokinetic flocculation tests were done at 3 m/h.

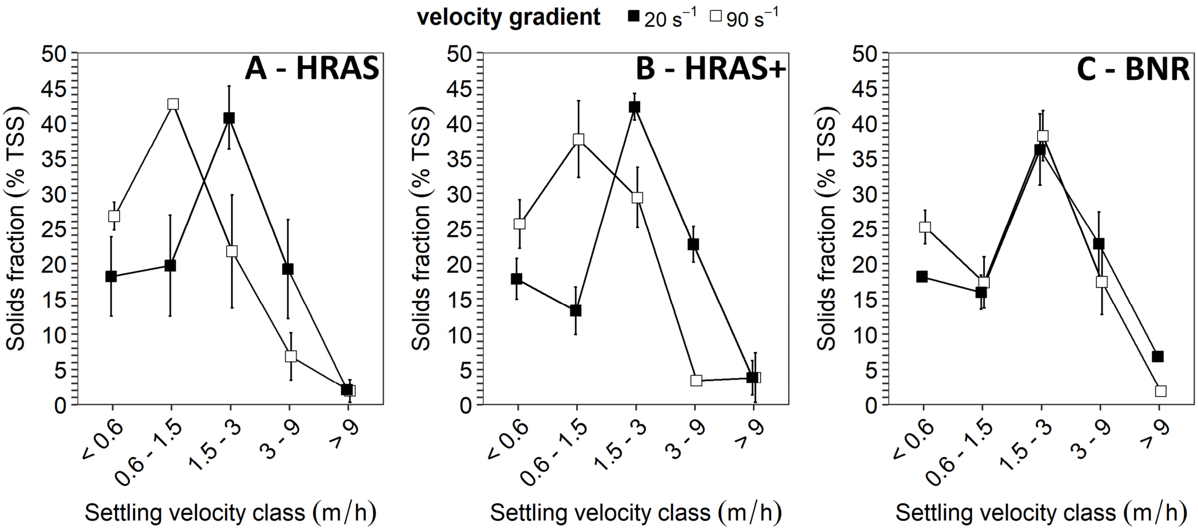
Figure 2. Settling velocity distribution at 22 s-1 (solid rectangles) and 91 s-1 (open rectangle) for HRAS (A) bioaugmented HRAS (B) and BNR sludge (C) expressed as a TSS sludge fraction (%) of initial sample. The test was performed at 351 ± 24, 493 ± 31, and 472 ± 29 mg TSS/L for HRAS, HRAS+ and BNR respectively and without addition of polymer (n=3).

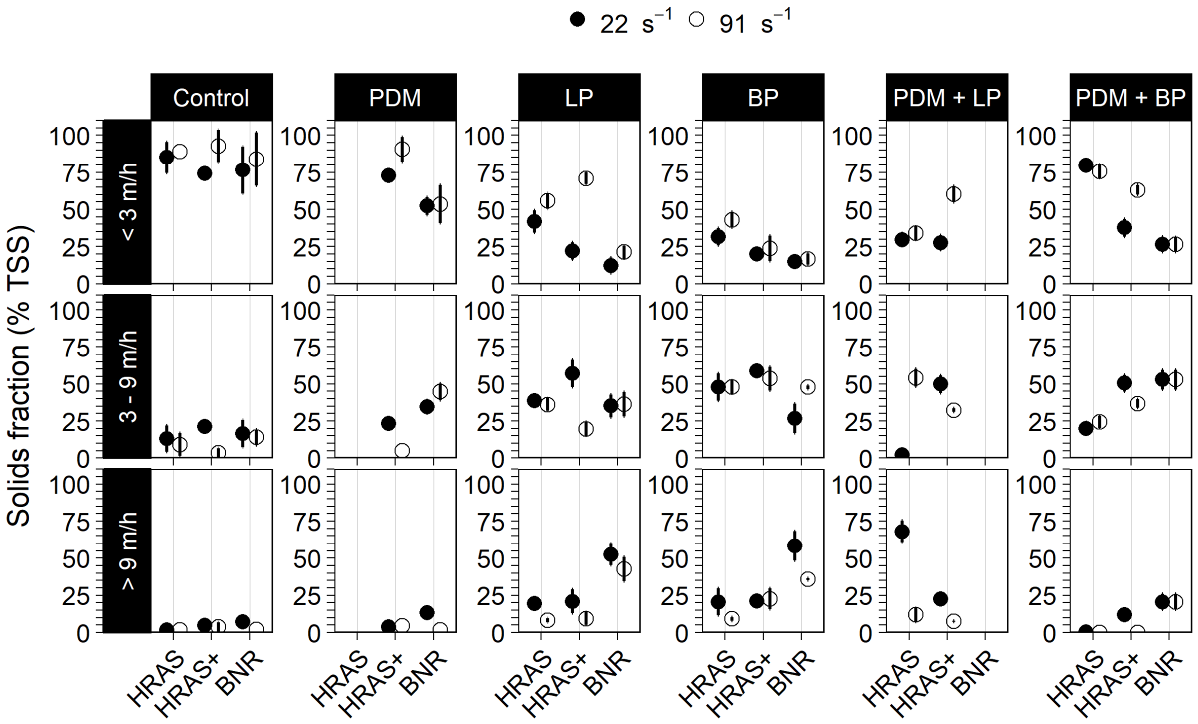
Figure 3. Polymer response curves showing the improvement in effluent quality relative to the control experiment (without polymer addition) in function of polymer dose for HRAS (A-C), bioaugmented HRAS (D-F), and BNR sludge (G-I). Coagulants used were ferric chloride ($FeCl_3$) or polyDADMAC (PDM) (A, D, G). Flocculants used were linear polyamide polymer (LP) or branched polyamide polymer (BP) (B, E, H).Also a combination of 0.5 g PDM/g polymer + LP (C, F, I) was also evaluated. The tests were performed at 355 ± 19 mg TSS/L, 506 ± 19 mg TSS/L, and 439 ± 46 mg TSS/L for HRAS, HRAS+, and BNR respectively.

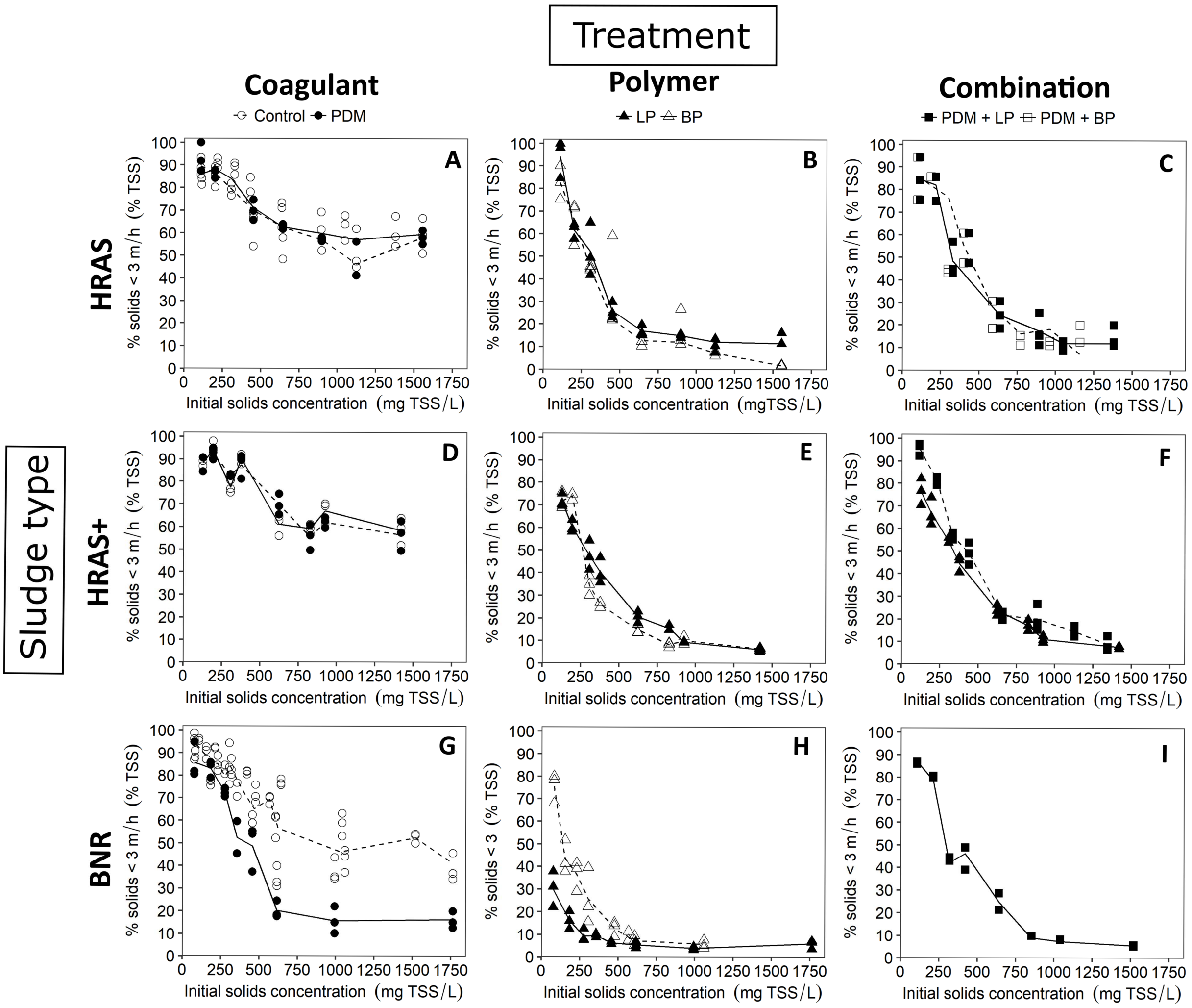
Figure 4. Orthokinetic flocculation curves with and without polymer addition, showing the remaining sludge fraction with settling velocity < 3 m/h in function of initial MLSS concentration after orthokinetic flocculation at 20 s-1 for 10 minutes for HRAS (A-C) HRAS+ (D-F) and BNR sludge (G-I). Polymer dosage was 0.5 g polymer/ kg TSS for PolyDADMAC (PDM), linear polymer (LP), branched polymer (BP) and 0.5 g polymer/ kg TSS for both polymers when a combination of the latter was used. No polymer was added in the control experiments

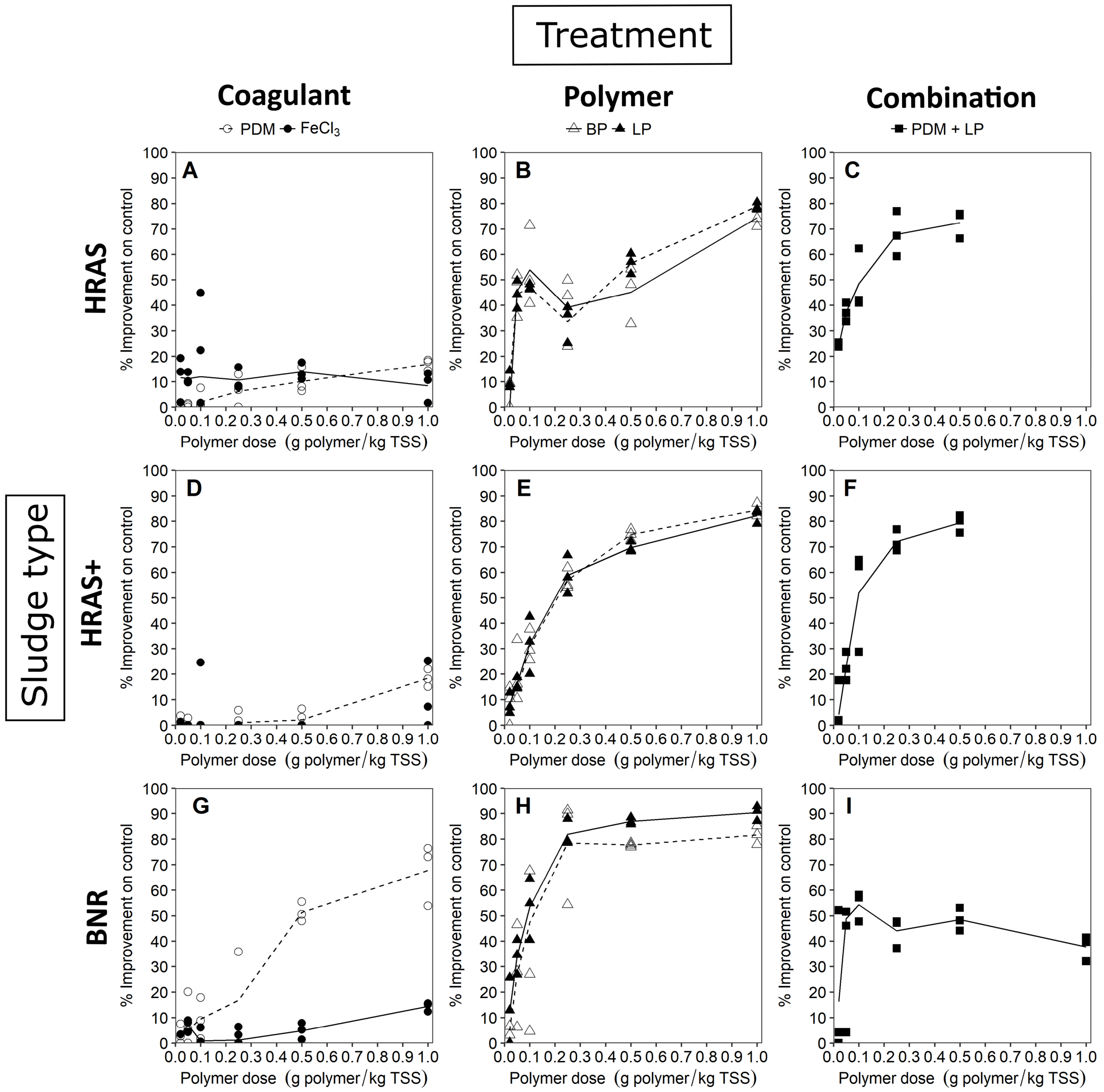
Figure 5. Impact of orthokinetic mixing intensity on settling velocity distributions expressed in three fraction (< 3 m/h, 3-9 m/h and > 9 m/h) with and without polymer addition for HRAS, HRAS+ and BNR sludge. Differences in settling fractions between the velocity gradient of 22 s-1 (solid) and 91 s-1 (hollow) indicate floc breakage. PolyDADMAC (PDM), linear polymer (LP) and branched polymer (BP) were added at 0.5 g polymer/g TSS or at 0.5 g polymer/g TSS each when a combination of polymers was used, except for the control where no polymer was added. Test were performed at 351 ± 24, 493 ± 31, and 472 ± 29 mg TSS/L for HRAS, HRAS+ and BNR respectively (n=3).


Orthokinetic (20 s^{-1} ; CSV = 3 m/h)
Gravitational (0 s^{-1} ; CSV = 1.5 m/h)
A - HRAS
B - HRAS+
C - BNR
solids fraction < CSV (% TSS)
Initial TSS (mg TSS/L)
0
10
20
30
40
50
60
70
80
90
100
0
250
500
750
1000
1250
1500
1750


velocity gradient
■ 20 s⁻¹ □ 90 s⁻¹
A - HRAS
B - HRAS+
C - BNR
Solids fraction (% TSS)
0
5
10
15
20
25
30
35
40
45
50
< 0.6
0.6 - 1.5
1.5 - 3
3 - 9
> 9
Settling velocity class (m/h)


● 22 s⁻¹ ○ 91 s⁻¹
Control
PDM
LP
BP
PDM + LP
PDM + BP
< 3 m/h
3 - 9 m/h
> 9 m/h
Solids fraction (% TSS)
0
25
50
75
100
HRAS
HRAS+
BNR


Treatment
Coagulant
Control
PDM
Polymer
LP
BP
Combination
PDM + LP
PDM + BP
Sludge type
HRAS
HRAS+
BNR
A
B
C
D
E
F
G
H
I
% solids < 3 m/h (% TSS)
% solids < 3 (% TSS)
0
10
20
30
40
50
60
70
80
90
100
0 250 500 750 1000 1250 1500 1750
Initial solids concentration (mg TSS/L)
Initial solids concentration (mgTSS/L)


Treatment
Coagulant
Polymer
Combination
PDM
FeCl$_3$
BP
LP
PDM + LP
Sludge type
HRAS
HRAS+
BNR
A
B
C
D
E
F
G
H
I
% Improvement on control
0
10
20
30
40
50
60
70
80
90
100
Polymer dose (g polymer/kg TSS)
0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0

Overcoming floc formation limitations in high-rate activated sludge systems

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Highlights

- Collision efficiency and floc strength were main limitations in floc formation
- Protein (PN) to polysaccharide (PS) ratio as differentiating feature
- Low PN/PS ratio with low EPS amount led to collision efficiency limitation
- Improved PN/PS ratio with low EPS amount led to floc strength limitation
- Bioaugmentation of BNR sludge to high-rate systems improves collision efficiency