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DEVELOPMENT OF BIOGRANULES IN A PILOT-SCALE SEQUENTIAL BATCH REACTOR TREATING ACTUAL TEXTILE WASTEWATER

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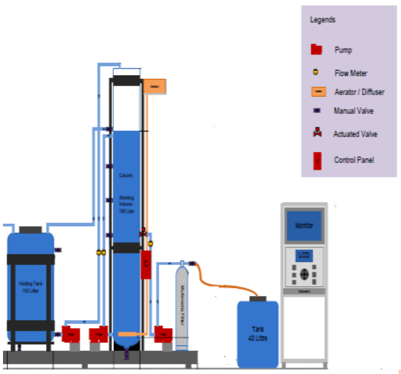
Graphical abstract



Abstract

A pilot-scale sequential batch reactor (SBR) biogranular system for the treatment of actual textile wastewater was developed in this study. The reactor had a working volume of 70 L and was operated according to SBR's sequence for 24-hr cycle, which includes sequential anaerobic and aerobic reaction phases. Wastewater from two textile mills were used as feed, while sewage and pineapple wastewater were used as co-substrate. After operating the system for 60 d, 30% of the sludge had transformed into biogranules and had increased to 67% at the end of the study. The biogranules developed in the reactor have sizes ranging from 0.2 mm to 9.5 mm with a mean settling velocity of 28 ± 7 m/hr and sludge volume index of 73.9 mL/g. At the end of the study, the system yields 92% removal of COD, but the color removal oscillated throughout the development period in the range of 50 to 70%. Although the biogranules development is much faster in lab-scale reactor under controlled environment, the findings indicate the feasibility of developing biogranules in a bigger scale reactor using actual textile wastewater and other high-strength biodegradable wastewater as co-substrate.

Keywords: Biogranules, textile, prototype, color, SBR, pineapple wastewater

Abstrak

Satu sistem reaktor berkelompok berjujukan (SBR) biogranular berskala loji-pandu untuk rawatan airsisa tekstil sebenar telah dibangunkan dalam kajian ini. Reaktor berisipadu 70 L telah dikendalikan mengikut urutan SBR untuk kitaran 24 jam, dengan fasa anaerobik dan aerobik beroperasi secara berselang-seli. Airsisa daripada dua kilang tekstil telah digunakan, manakala kumbahan dan airsisa nanas digunakan sebagai ko-substrat. Setelah beroperasi selama 60 hari, 30% daripada enapcemar telah berubah menjadi biogranul dan telah meningkat kepada 67% di akhir kajian. Biogranul yang terbentuk di dalam reaktor mempunyai saiz diantara 0.2 mm hingga 9.5 mm dengan halaju enapan purata 28 ± 7 m/j dan mempunyai index isipadu enapcemar (SVI) serendah 73.9 mL/g. Pada akhir kajian ini, 92% penyingkiran COD telah dicapai oleh sistem, tetapi penyingkiran warna berubah-ubah di sepanjang tempoh pembentukan granul dalam lingkungan 50 hingga 70%. Walaupun pembentukan biogranul adalah lebih cepat di dalam reaktor berskala kecil dan di dalam persekitaran terkawal, hasil kajian menunjukkan keupayaan pembentukan biogranul dalam reaktor skala yang lebih besar dengan menggunakan airsisa tekstil sebenar dan air sisa mudah biorosot berkuatan tinggi lain sebagai ko-substrat.

Kata kunci: Biogranul, tekstil, prototaip, warna,SBR,air sisa nanas

1.0 INTRODUCTION

Conventional biological processes have been found to be incapable of treating textile wastewater due to the non-biodegradable nature of the wastewater [1]. However, in the past decades, studies have shown that textile wastewater can be treated by the sequential combination of anaerobic and aerobic processes. The anaerobic process is able to decolorize the wastewater, while the following aerobic process further degrades the organics and also removes the amines that are produced during the anaerobic process [2]. These processes have been carried out using either separate anaerobic and aerobic tanks or in a single sequential batch reactor (SBR) with biomass either in the form of suspended flocs or biogranules.

Biogranules are macroscopic combination consisting of dense microbial consortia packed with diverse bacterial species amounting to millions of organisms per gram of biomass [3]. They are formed via a type of immobilisation known as biogranulation and may result from self-immobilisation or immobilisation of microorganism onto seeding agent [4]. Biogranules are categorized as aerobic and anaerobic, which can be developed in a sequencing cycle of feeding, reacting, settling and decanting [5].

There have been various studies carried out at pilot-scale of aerobic granular SBR either using actual wastewater or synthetic wastewater observing significant differences between pilot- and lab-scale results [6, 7, 8]. Environmental factors such as wastewater characteristics, influent substrate concentration and temperature could be precisely controlled at lab-scale as compared to pilot-scale [7]. Studies observed that biogranules development in pilot-scale reactor took six to thirteen months as compared to the lab-scale, which only needs two to four weeks [7, 9]. Undoubtedly, the long start-up period in pilot-scale reactor need more attention from researchers to make the reactor system practical to operate.

Nevertheless, biogranulation study treating actual textile wastewater in an SBR system at a bigger scale is apparently missing. As the characteristics of the textile effluent is well known for its low biodegradability and high variation, many practical aspects of the treatment in developing biogranules need to be explored. Furthermore, while co-substrates such as glucose and acetate are commonly used in lab-scale study to aid the biogranules development and treatment process, their applications in actual plant is costly unattractive. In this study, a pilot-scale SBR biogranular system was developed and tested to treat textile wastewater. The system utilizes the concept of sequential anaerobic and aerobic biological reactions for complete degradation of the wastewater. The use of sewage and pineapple wastewater as co-substrate was explored. This paper describes the experience in developing the biogranules in the pilot-scale reactor using actual textile wastewater.

2.0 MATERIALS AND METHODS

2.1 Seed sludge and Wastewater

Raw textile wastewater from two local textile mills, American & Effird (M) Sdn. Bhd. (AESB) and Ramatex Textile Industrial Sdn. Bhd. (RTISB) were obtained and used in this study. Sewage and its sludge were collected from Indah Water Konsortium's sewage treatment plant at Taman Impian Emas, Johor Bahru. Textile sludge was obtained from RTISB, while pineapple wastewater was collected from Lee Pineapple Company Sdn. Bhd., Johor Bahru.

2.2 Sequential Batch Reactor

The design of the SBR system was based on Muda *et al*. [10]. Figures 1 and 2 show the pilot-scale bioreactor that was used in the study. The system comprised of a holding tank, an SBR column reactor equipped with piping system, air blower (close-loop blower system), air compressor (SWAN), pumps (Kuobao) and valves (Arita). The holding tank of 200 L was used to store the raw wastewater. The SBR column had a working volume of 70 L with an internal diameter of 230 mm and a total height of 2.5 m. The wastewater was pumped into the reactor from the top of the column while air (during aerobic reaction phase) was supplied into the reactor by a fine air bubble diffuser located at the bottom of the column. The decanting of the wastewater was carried out via a valve connected by a flexible hose at the middle of the column. During anaerobic reaction phase, the wastewater was recirculated using a recirculation pump and piping system. A desludging port located at the bottom of the tank was used to maintain the optimum concentration of the biomass. The operation of the system was carried out using a programmable logic controller (PLC). Several monitoring probes i.e. DO, pH, ORP (Horiba Ltd.) were installed in the SBR column, attached to a data logger and control system for continuous monitoring. The pilot-scale reactor system was operated at the Innovation and Commercialization Centre of Universiti Teknologi Malaysia.

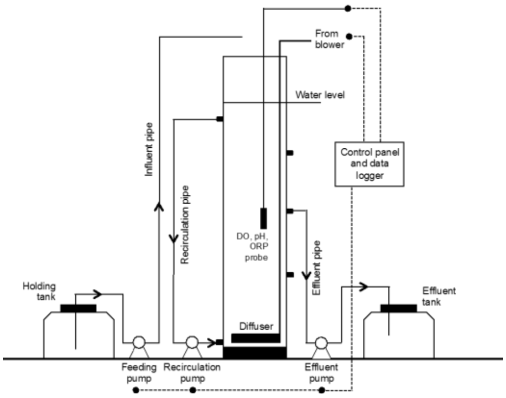


Figure 1 Schematic diagram of the hybrid SBR system

Figure 2 Hybrid Biogranular Sludge Reactor

2.3 Analytical Methods

The influent and effluent from the reactor were analyzed for chemical oxygen demand (COD), color and total suspended solids (TSS). Samples taken from the reactor column during the aeration process were analyzed for mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), granular strength (IC), settling velocity (SV) and sludge volume index (SVI).

Chemical oxygen demand and color (as ADMI) were quantified using HACH Spectrophotometer (DR 6000U). Total suspended solids, MLSS and MLVSS were analyzed according to the Standard Method [11]. The SV was determined as the time taken for the flocs or biogranules to settle at a certain height in a glass column filled with tap water. The SVI assessment was carried out based on the Standard Method [11]. The morphological and structural observations of granular sludge were conducted by using stereo microscope equipped with digital image analyzer (HSZ-EPA 10X22; HSZ-1LST6). The microbial compositions and mineral content within the biogranules were observed qualitatively and quantitatively by Field-emission scanning electron microscope coupled with Energy Dispersive X-ray Analysis (FESEM-EDX; Hitachi SU8020).

Determination of the granular strength was based on Ghangrekar *et al*. [12]. Shear force on the biogranules was introduced through agitation using an orbital shaker at 200 rpm for 5 min. At certain amplitude of the shear force, parts of the biogranules that are not strongly attached within the biogranules were detached. The quantity of the ruptured biogranules was separated by allowing the fractions to settle for 1 min in a 100mL measuring cylinder. The dry weight of the settled biogranules and the residual biogranules in the supernatant were measured. Equation (1) was used to calculate integrity coefficient (IC), which indirectly represents the strength of the biogranules. Smaller IC value indicates stronger biogranule and vice versa.

$$IC = \left(\frac{RG}{SG+RG}\right) \times 100 \quad (1)$$

where

IC	=	integrity coefficient (%)
RG	=	Residual biogranules (mg)
SG	=	Settled biogranules (mg)

Sugar profile of the pineapple wastewater was determined using High Performance Liquid Chromatography-Refractive Index (HPLC-RI). Standard solution of glucose, fructose, sucrose and lactose were prepared. 5g of sample was weighed in 50 mL volumetric flask and topped up to 50 mL with ACN: H_2O (1:1). Standard solutions and samples were injected into HPLC-RI and the concentrations (%w/w) were determined.

2.4 Experimental Procedures

The development of biogranules in the pilot-scale SBR column was carried out according to the procedures developed by Muda *et al*. [10]. The SBR was operated according to the FILL, REACT, SETTLE, DECANT and IDLE sequence for 24-hr cycle. Each cycle initially comprised of 13 min filling, 1380 min reaction, 30 min settling, 9 min decanting and 10 min idle. The reaction phase consists of 255 min of anaerobic phase, followed by 435 min of aerobic phase, and another 255 min of anaerobic phase and 435 min of aerobic phase. Table 1 shows the initial complete cycle of the SBR system.

The wastewater was recirculated at a rate of 3 L/min during the anaerobic phase to ensure continuous contact between the wastewater and the biomass. Aeration during aerobic phase was provided by the diffuser. The DO, pH and ORP were monitored throughout the operation, while the temperature in the reactor was not regulated.

The development process started with the addition of 35 L of mixed sludge (50% v/v sewage and textile) and 35 L of raw textile wastewater mixed with sewage (50% v/v) into the SBR reactor column, making the final volume of 70 L. In the beginning, textile wastewater from AESB with OLR of 0.43 kg COD/m^3/day at alkaline pH of 10, SAV of 0.6 cm/s and settling time of 30 min were employed. Sewage was used as co-substrate and the portion of sewage was then reduced from 50% to 20% in stages within six weeks as an attempt to run reactor solely with textile wastewater towards the end of study. However, reduction of sewage concentration resulted in low organic loading which further delay the biogranule formation. As to increase the organic loading, sewage was replaced with pineapple wastewater after about 90 days, along with the changes from AESB to RTISB's wastewater. Pineapple wastewater was chosen due to its high content of organic's concentration, only 7% v/v of the pineapple wastewater was used to obtain the higher OLRs (0.79-5.65 kg COD/m^3/day). RTISB wastewater were chosen to replace AESB wastewater as the wastewater has wide range of color intensity which further explained in section 3,1. The settling time was also reduced from 30 min to eventually 10 min after about six weeks and then further reduced to 5 min on week 8 and maintained until the end of the study period since short settling time is needed for successful biogranule development [13,14]. The SAV was increased from 0.6 cm/s to 0.8 cm/s after about four weeks. It was then increased to 1.0 cm/s after about 90 days, 1.2 cm/s after 120 days, and finally 1.4 cm/s on day 195th until the end of the study period. The changes on the SAV were made due to foaming problem encountered in the reactor, caused by the constituents in the wastewater.

3.0 RESULTS AND DISCUSSION

3.1 Characteristics of Wastewater

The characteristics of the textile wastewater used in this study are shown in Table 2. The wastewater from RTISB has higher color intensity ranging from 600 to 4500 ADMI as compared to AESB (500 to 900 ADMI). However, RTISB's wastewater had lower COD value of 600 to 1000 mg/L as compared to AESB (950 to 1750 mg/L). The pHs of the wastewater from both mills were alkaline ranging from about 8 to 12. The sewage used in the study had COD of 200 to 600 mg/L and color between 100 to 200 ADMI. The pineapple wastewater had a very high COD (55,000 – 75,000 mg/L), but colorless with low pH between 3.2 and 4.5. Sugar profile of pineapple wastewater shows the presence of glucose and fructose with 0.9%w/w and 0.4% w/w respectively.

The mixture of sewage and textile wastewater from AESB gave a COD and color in the range of 660 to about 1500 mg/L, and 170 to 900 ADMI, respectively. A 7% v/v of pineapple wastewater mixed with RTSB textile wastewater yield COD and color in the range of 1760 to about 13,000 mg/L, and 500 to 4,500 ADMI, respectively with pH ranging from 6 to 7.8. The high variations in the COD and color are mainly due to the variations in the characteristics of the textile wastewater throughout the study period. Due to these variations, the influent color ranged from about 170 to 4500 ADMI and the loading rates fluctuated in the range of 0.28 to 5.65 kg COD/m^3·day. The high variations of the actual wastewater signify the challenges dealing with the actual condition as compared to those of lab-scale study.

Table 1 The initial complete cycle of the hybrid biogranular SBR system

Phase	One complete cycle (≈24 hours)		
Filling	13 min		
		Anaerobic	Aerobic
React	1st phase	255 min	435 min
	2nd phase	255 min	435 min
Settling	30 min		
Decanting	9 min		
Idle	10 min (in between each anaerobic and aerobic phase)		
Total Cycle Length	**1437 min**		

.

Table 2 Characteristics of wastewater and co-substrates

Parameters	American and Effird	Ramatex	Sewage	Pineapple	[a]Textile + Sewage (20-50% v/v)	[b]Textile + Pineapple (7% v/v)
COD (mg/L)	950-1750	600-1000	200-600	55000-75000	660-1480	1760-13190
Color (ADMI)	500-900	600-4500	100-200	50-100	170-900	500-4500
pH	10-12	8-10.5	7-8	3.2-4.5	8-10	6-7.8
Total suspended solid (mg/L)	14-77	50-210	39-68	1000-2810	15-120	60-800
Organic loading rate (OLR) (kg COD/m^3/d)	-	-	-	-	0.28-0.63	0.79-5.65

[a]American & Effird; [b]Ramatex Textile

3.2 Biogranules Development

The profile of the biomass concentration (MLSS) and TSS in the effluent are given in Figure 3. The MLSS concentration was inconsistent in the first six weeks, believed to be caused by the changes in sewage mixing ratio and settling time. The settling time was reduced sequentially from 30 to 5 min to ensure that only biogranules are retained in the reactor. The MLSS was slightly fluctuating in the following weeks until 105th days in the range of 1.8 to 3.3 g/L. Upon the changes from AESB to RTISB's wastewater and the addition of pineapple wastewater into the

greater than 2 mm had SV of more than 100 m/hr, while those of smaller size had SV ranging from about 10 to 20 m/h. The mixture of these sizes resulted in the average SV of 28 ± 7 m/hr. While this value is much lower than those reported in the lab-scale study (80 ± 8 m/hr) by Muda *et al*. [10], it correlates with study reported by Zheng *et al*. [16] (18 – 31 m/hr). The mean SVI value of 73.9 mL/g, although higher than the one achieved in the lab-scale study (69 mL/g), indicates good settling reactor, a drastic increase in biomass concentration (from 1.8 to 9.3 g/L) was observed.

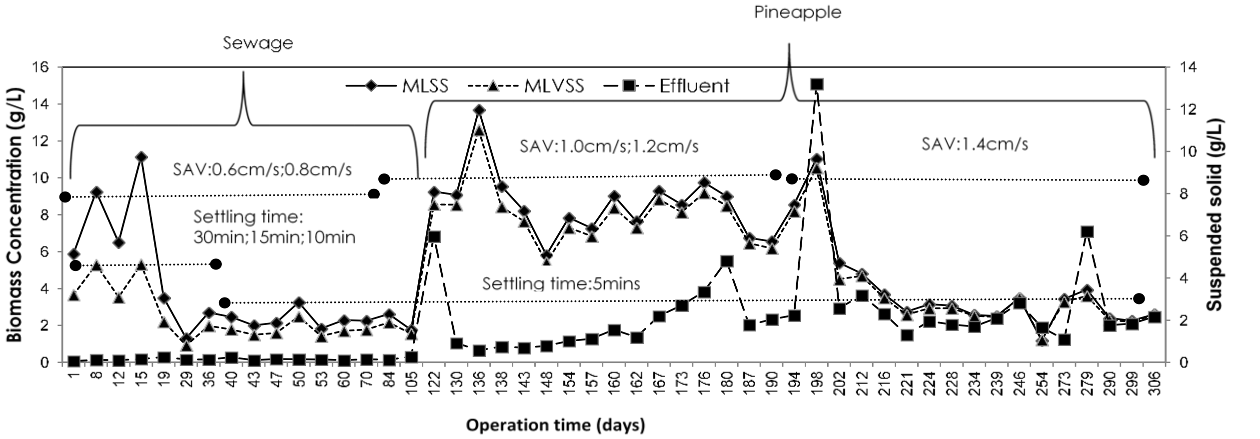


Figure 3 Profile of biomass concentration in reactor and TSS in the effluent

This could be due to the presence of non-reducing sugar, carbohydrates and protein from the pineapple wastewater, which act as a better medium of growth for the microbes [15]. During this phase, the MLSS fluctuated between 8.6 and 9.3 g/L for about 90 days. However, when the SAV was increased from 1.2 to 1.4 cm/s to enhance the density of the biomass, a peak of biomass concentration and a substantial loss of biomass from the reactor was observed within the same day. This was caused by the resurgence of the sludge that was accumulated at the bottom prior to the increase of the SAV. This reduced the MLSS from 11.0 g/L to 5.4 g/L in 5 days. The MLSS did not improve in the following weeks and eventually reduced to 2.6 g/L on the 306th day of operation.

Throughout the development period, the SV and SVI oscillated (Figure 4) as compared to the steady development in the lab-scale study reported by Muda *et al*. [10]. As expected, the higher SV results in lower SVI values indicating good settling characteristics of the biogranules. Formation of patchy flocs during low OLR and the presence of biogranules coexisted with the fluffy flocs with pores and filaments at high OLR at certain days were noticed in the reactor. The adaptation of biogranules in treating raw wastewater with fluctuating characteristics might be the reason for oscillation of SV and SVI and the TSS in the effluent.

The SVs that correspond to the size of the biogranules (taken after 60 days of development) are shown in Table 3. The biogranules with size ofproperty considering the value is still less than 100 mL/g [17].

Figure 5 shows the changes in the formation of the bioflocs and biogranules during the biogranules development. In the initial stage of the study, the biomass in the reactor was mainly in the form of flocs. Small biogranules were observed from the 15th day onwards. After two months of continuous running, biogranules were found in sizes ranging from 0.2 to 9.5 mm. Despite operating at low OLR

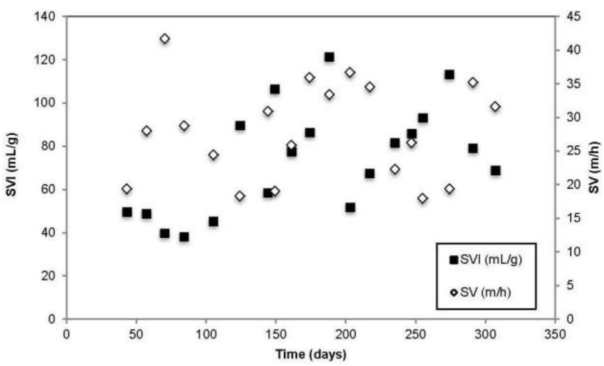


Figure 4 Profile of settling velocity and sludge volume index during the development period

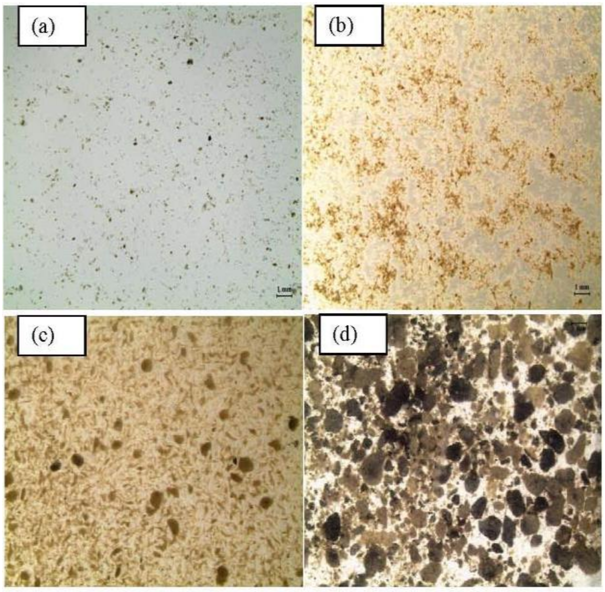


Figure 5 (a) Sludge particles during the initial stage of the experiment; (b) bioflocs at the start of experiment; (c) co-existence of flocs with biobiogranules; and (d) developed biogranules at the 60 days of the experiment. Pictures were taken using a stereo microscope with magnification of 6.3X (scale bar = 1mm)

Table 3 Settling velocity of biogranules according to size (after 60days)

Size (mm)	m/hr
0.20 -0.30	9.4
0.30-0.43	10.8
0.43-0.60	15.1
0.60-0.80	16.6
0.80-1.18	21.9
1.18-2.36	60.1
2.36-4.75	117.3
4.75-6.30	134.9
6.30-9.50	189.2
≥9.50	224.8

(0.28 - 0.63 kg COD/m^3/d) and alkaline pH of 10, approximately 30% of the sludge had successfully formed biogranules. However, there was no increase in the MLSS in spite of having the biogranules. As the experiment continued, more biogranules were visible but flocs were still present inside the reactor. At the end of the study, 67% v/v of the biomass was in the form of biogranules. The biogranules with size of 0.2 mm to 0.4 mm were found to be dominant in reactor, while the bigger biogranules were found to have diameter of about 10 mm. At SAV of 1.4 cm/s, the biogranules average diameter of 2.7 ± 3.0 mm obtained in this study is bigger than those achieved by Muda *et al*. [10] (2.3 ± 1.0 mm at SAV of 1.6 cm/s).

The granular strength of the biogranules was determined based on the integrity coefficient (IC) as mentioned earlier. Development of biogranule with adequate mechanical strength is required for a successful SBR operation. The smaller the value of IC, the greater the strength of biogranule withstanding the breakage due to shear force of the aeration. Even though the IC value does not signify absolute shear strength, it is expected to represent relative strength of biogranules against hydraulic shear, which biogranules often experiences during reactor operation. Figure 6 shows the profile of IC of the developed biogranules. According to Ghangrekar *et al*. [12], biogranules with integrity coefficient of less than 20% were considered as high strength biogranules. Granular strength was initially determined after the formation of matured biogranules, which was after 2 months, giving IC value around 14%. With an initial value of 14%, the IC was reduced to about 8% at the termination of the experiment. There is a sharp increase in IC value to 30%, which might be due to the increase in SAV. After 190 days, the IC value start to reduce gradually indicating the increase in the strength of the bond that binds the microorganisms together within the biogranules. During the aeration phase, those biogranules, which could not withstand the shear force undergo rupture and the microbes within the biogranules were loosely bounded to each other. Generally, the developed biogranules possessed higher strength as the IC value remained below 20% most of the time.

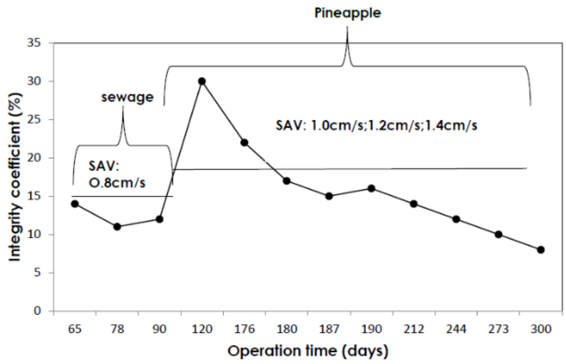


Figure 6 The change in integrity coefficient (representing the granular strength) during the biogranule development period

The microstructure and mineral with metal content of the mature biogranules were analysed using Field-emission scanning electron microscope coupled with Energy Dispersive X-ray Analysis (FESEM-EDX) are shown in Figure 7 and Table 4. The FESEM images illustrate the dominancy of rod shape bacteria compactly clumped to each other on the cross-section surface of mature biogranules. As can be seen in Figure 7c, the biogranules exhibit a filamentous structure upon addition of pineapple wastewater whereas the biogranules fed with sewage had a non-filamentous coccoid bacterial structure (Figure 7a). Glucose presence in pineapple wastewater had contributed to the filamentous structure of biogranule [18]. Difference type of microorganisms may predominate at different type and concentration levels of substrate eventually influencing the structural and morphology of the developed biogranules [19]. However, in both biogranules, rod shape bacteria dominant by tightly linked and embedded to one another and form a rounded shape on the surface, covered with extrapolysaccarides (EPS) (Figures 7b and 7d). Besides, there are numerous cavities presence among the clumped bacteria expected to provide smooth mass transfer of substrates or metabolite substances in and out of the biogranules.

As can be seen from the Table 4, aluminium (Al), iron (Fe), magnesium (Mg), sodium (Na), calcium (Ca) and cobalt (Co) are the common minerals found in both biogranules. In biogranule A, zinc (Zn), potassium (K), chromium (Cr) and nickel (Ni) were found, while in biogranules B, none of these minerals were found except copper (Cu) was found to be incorporated in the biogranule. However, there is not much difference in both biogranules in terms of the concentrations (wt%) except for Ca and Co, which is apparently higher in biogranule B (pineapple-textile fed biogranule) as compared to biogranule A (sewage-textile fed biogranule). This may indicate the contribution of the inorganic elements in the granulation process. Sobeck and Higgins [20] stated that the presence of Ca^{2+} and Mg^{2+} promotes equivalent floc properties based on the divalent cation bridging theory. According to Ren *et al*. [21], biogranule-rich Ca^{2+} displayed more compact granular structure and higher strength as compared to biogranule without Ca^{2+} accumulation, which correlates with the biogranule strength at the end of this study. Metal requirements of the variety of microorganisms present in the reactor system may influence the uptake of metals into the biogranules. The presence of metal and minerals in the biogranules had also proved the capacity of biogranules to act as biosorbent. Large surface area, high porosity and high settling properties of biogranules help in absorbing the metals found in wastewaters [22].

Table 4 Comparison of mineral content of sewage-fed biogranule with pineapple wastewater-fed biogranule

Elements	Biogranule A(wt%)	Biogranule B(wt%)
C	68.05	71.45
O	23.40	21.90
Al	4.40	1.85
Fe	1.60	0.30
Zn	0.20	0.00
Mg	0.20	0.50
K	0.15	0.00
Na	0.05	0.45
Ca	0.15	1.30
Cr	0.35	0.00
Mn	0.00	0.00
Co	0.05	1.45
Ni	1.45	0.00
Cu	0.00	0.70

The time taken to form the biogranules in this pilot-scale study was almost 10 months as compared to 2 months, obtained in the lab-scale [10]. The delay in achieving a full granulation process is anticipated, to be caused by the unstable wastewater characteristics, low OLR (0.28-0.63 kg COD/m^3/d) at the beginning of the study with the use of sewage as co-substrate and low SAV (0.6-0.8 cm/s). The use of low SAV at the beginning of the study was to reduce the foaming problem caused by the wastewater content, which had led to the outflow of the biomass from the top of the reactor.

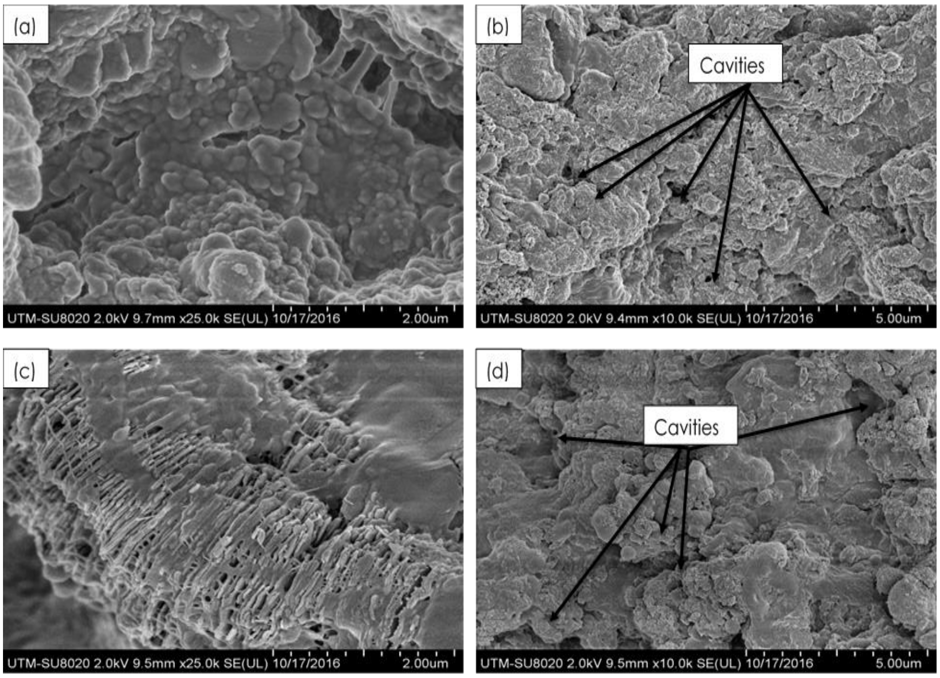


Figure 7 FESEM microstructure observation on mature biogranules. (a) Coccoid bacteria tightly linked to one another in sewage-fed biogranules at a magnification of 25.0k. (b) Cavities that appear between bacteria clumped inside the sewage-fed biogranules at magnification of 10.0k (c) Filamentous structure of pineapple wastewater-fed biogranule at a magnification of 25.0k (d) Cavities and rod shaped bacteria embedded on the pineapple wastewater –fed biogranule at a magnification of 10.0k

3.3 Removal Performances during Biogranules Development Stage

The removal of COD and color throughout the development stage was also monitored in this study. At this stage, the performance of the system was not expected to be high as the system has not being optimized. The removals of COD for wastewater from AESB mill are shown in Figure 8. At the initial stage of treating AESB's wastewater, the removal of COD was 86%. After a few days, it began to drop and became inconsistent for the remaining period achieving average removal of COD of 64%. In treating AESB's wastewater, the highest removal of COD was 92%, while the lowest was 35%. The removals of COD in treating RTISB's wastewater were higher; attaining mean removal of 92% (Figure 9). The removal ranged from 98% and 81%, which were higher than the removal of COD from AESB's wastewater.

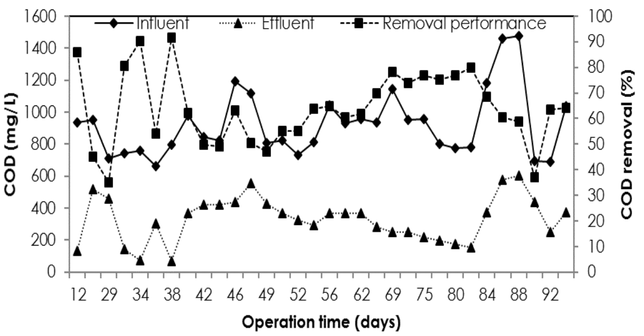


Figure 8 Profile of COD removal for American & Effird mill wastewater

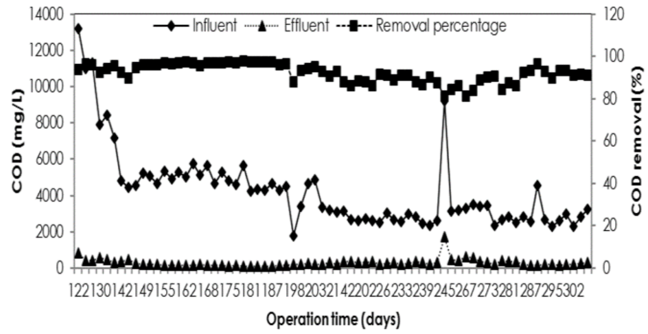


Figure 9 Profile of COD removal for Ramatex wastewater

The color removals for AESB and RTISB are illustrated in Figure 10 and 11, correspondingly. During the treatment of AESB's wastewater, the highest removal of 69.2% was recorded. Notably, there was an increase in effluent color value compared to influent during certain days resulted in lowest percentage removal of -116.1%. Nevertheless, there was improvement in color removal for RTISB wastewater with highest of 88.8% and mean removal percentage of 50.6% achieved. On the whole, the removal efficiency of color fluctuated throughout the development period.

Poor color removal in treating the AESB's as compared to RTISB's wastewater could be due to type of dyes used by the former [23,24]. However, it is difficult to determine the categories of the dyes used, as the mills use 50 to 60 different types of dyes of different colors. It was reported that the byproducts from the cleavage of reactive azo dye namely Reactive Black 5, Reactive Violet 5 and Direct Black 8 are couldn't be further degraded in aerobic conditions [25,26,27]. There is possibility of presence of these types of dye in AESB wastewater which resulted in poor color removal. According to Ong *et al*. [28] and Pandey *et al*. [29], the rate of dye degradation is affected by the increase in organic loading and possibly through the addition of co-substrate. The addition of organic loading rate increases electron donor substrates such as glucose, sucrose, acetic acid that eventually doubles the amount of electron donor transferring to the N=N bond and give rise to color removal. In this study, an increase in the reduction of COD and color was noticed after the addition of pineapple wastewater. Presumably, the presence of glucose and fructose in pineapple waste boost the metabolic activities of the microorganism in the biogranules. This apparently improved the COD and color removal as compared to the addition of sewage as the co-substrate.

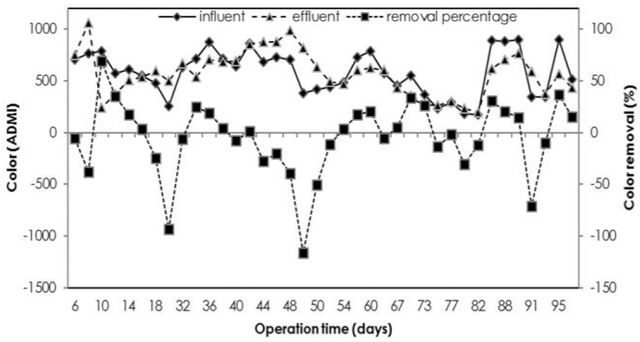


Figure 10 Profile of color removal for American & Effird wastewater

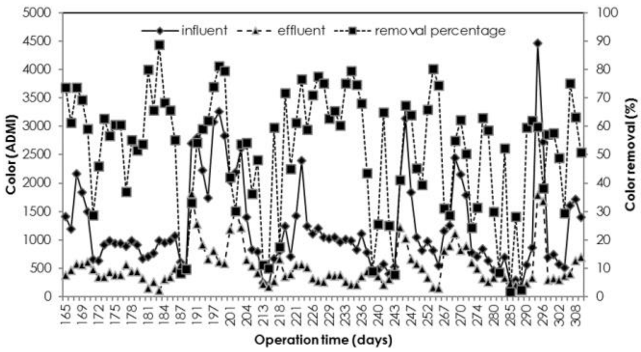


Figure 11 Profile of color removal for Ramatex wastewater

Table 5 summarizes some of the studies conducted in treating actual textile wastewater by using SBR-biogranulation technology. The differences in the performance of removal may be due to the types of dyes, biomass used, reactor system and the operational conditions used in the study. A few studies used synthetic wastewater and specific microbes during their start-up to cultivate the biogranules in order to treat the textile wastewater [30, 31, 32]. This study is apparently the only study conducted using large-scale SBR system.

As can be seen, the removal performance obtained from this study is considerably good as compared to other studies, despite of not having fully grown biogranules, having low biomass concentration, and operating at unoptimized condition under high variation of influent characteristics.

using pineapple wastewater are its acidic property and high organic content. Only a small volume of the pineapple waste need to be used to increase the OLR and at the same time neutralizes the pH of the wastewater.

Table 5 Treatment of raw textile wastewater using SBR biogranulation technology

Wastewater	Biomass	[a]Reactor configuration/phase	Removal performance	References
Real textile wastewater - cotton and polyester	Sludge	Working volume: 5 L Height: 60 cm F:1-1.5h;R:25h;S:0.5-1.0h;D: 0.5 h	$COD_{Soluble}$: 55% (aeration: 20-30 h); $COD_{Soluble}$: 62% (aeration: 45-50 h); $COD_{Soluble}$:80-95% (aeration: 22-25 h)	[33]
Combined domestic and textile wastewaters- synthetic and real textile wastewater	Mixed microbial consortium	Working volume: 2 L Height: 30 cm Internal diameter:10 cm *HRT:12hr;* F:0.25hr;A:10.75hr; S and D: 0.75 hr; I: 0.25 hr *HRT: 8 hr;* F:0.25hr;A:6.75hr;S and D: 0.75 hr; I: 0.25 hr	BOD: 73% TSS: 63% COD: 63% MLSS: 5 g/L - 5.5 g/L	[30]
Textile dye industry wastewater	Mixed white rot fungi	Working volume: 2 L F:1hr; R:20hr;S:2hr; D: 0.75hr ; I: 0.25 hr	Color: 71.3% COD: 79.4% MLSS: 5.75 g/L	[31]
Real textile wastewater	Textile sludge with four dyes degrading bacteria	Workingvolume:1L Height: 90 cm; Internal diameter: 6 cm; F: 5 min; An:170 min; A: 170 min; S/D/I: 5 min	Color: 61% COD: 46% Biomass: 12.9±0.8 g/L (MLSS) 11±0.6g/L (MLVSS)	[32]
Real textile wastewater	Mixed sludge (i.e. sewage and textile)	Working volume: 88 L Height: 2.5 m Internal diameter: 23 cm F:13 min; An: 510 min; A:870min;S:5min; D: 9 min; I: 10 min;	COD_{total}: 76% $COD_{soluble}$: 90% Color: 50% Biomass: 2.61 g/L (MLSS) 2.59g/L (MLVSS)	This study (2016)

[a]F-Fill; R-Reaction; An-Anaerobic; A-Aerobic; S-Settling; D-Decant; I-Idle

3.4 Challenges in Developing Biogranules

Development of biogranules in a pilot-scale reactor is rather challenging than doing it in a lab-scale reactor. The main differences between the two scales are the reactor volume and influent wastewater quality [7]. As the size of the reactor increases, the use of co-substrates such as acetate and glucose become costly prohibitive; hence, other alternative need to be used. In this study, sewage was initially used as the co-substrate but it was later replaced with pineapple wastewater. Sewage did not provide sufficient organics to support the growth of the biomass and the development of the biogranules. The advantages of

The use of actual textile wastewater also contributes to the difficulty in developing the biogranules. As compared to synthetic wastewater which is more stable, actual textile wastewater highly varies according to the type of dyes used in the manufacturing process. Obviously, this will reduce the rate of biogranules development. In actual situation, a holding or stabilizing tank will be needed to minimize. Furthermore, appropriate SAV is crucial to provide uninterrupted operation of the system. Applying high air flow rate(L/m) in order to increase SAV might result in outflow of biomass from reactor due to the foaming characteristic of wastewater content while operating system at low air flow rate will result in poor circulation of sludge in reactor delaying the formation of biogranule.

4.0 CONCLUSION

In this study, the development of biogranules has been carried out in a pilot-scale reactor treating actual textile wastewater. Sewage and pineapple wastewater were sequentially used as the co-substrates. Approximately 67% v/v of flocs was transformed to biogranules in the reactor at the end of development stage of about 10 months. Using SAV of 1.4 cm/s, the biogranules had an average diameter of 2.7 ± 3.0 mm, mean settling velocity of 28 ± 7 m/hr and sludge volume index of 73.9 mL/g. At the end of the development period, the reactor system was able to remove 92% of COD and 50% of color. Due to the low OLR and the highly unstable wastewater characteristics, longer period was required for the biogranules to develop in the pilot-scale reactor as compared to the lab-scale study. The use of suitable co-substrate, optimum OLR and SAV is expected to shorten the start-up period of granulation in pilot scale reactor.

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References

[1] Rajaguru, P. K. K., Palanivel, M. and Subburam, V. 2000. Biodegradation of Azo Dyes in Sequential Anaerobic-aerobic System. *Applied Microbiology and Biotechnology.* 54: 268-273.

[2] Franciscon, E., Mendonça, D., Seber, S., Morales, D. A., Zocolo, G. J., Zanoni, M. B., Grossman, M. J., Durrant, L. R., Freeman, H. S. and Umbuzeiro, G. A. 2015. Potential of a Bacterial Consortium to Degrade Azo Dye Disperse Red 1 in a Pilot Scale Anaerobic-aerobic Reactor. *Process Biochemistry.* 50(5): 816-825.

[3] Weber, S. D., Ludwig, W., Schleifer, K. H. and Fried, J. 2007. Microbial Composition and Structure of Aerobic Granular Sewage Biofilms. *Applied and Environmental Microbiology*. 73(19): 6233-6240.

[4] Liu, Y. and Tay, J. H. 2004. State of the Art of Biogranulation Technology for Wastewater Treatment. *Biotechnology Advances*. 22(7): 533-563.

[5] Ibrahim, Z., Amin, M. F. M., Yahya, A., Aris, A. and Muda, K. 2010. Characteristics of Developed Biogranules containing Selected Decolourising Bacteria for the Degradation of Textile Wastewater. *Water Science and Technology*. 61(5): 1279-1288.

[6] Ni, B. J., Xie, W. M., Liu, S. G., Yu, H. Q., Wang, Y. Z., Wang, G. and Dai, X. L. 2009. Granulation of Activated Sludge in a Pilot-Scale Sequencing Batch Reactor for the Treatment of Low-Strength Municipal Wastewater. *Water Research*. 43(3): 751-761.

[7] Liu, Y.Q., Moy, B., Kong, Y. H. and Tay, J. H. 2010. Formation, Physical Characteristics and Microbial Community Structure of Aerobic Biogranules in a Pilot-Scale Sequencing Batch Reactor for Real Wastewater Treatment. *Enzyme and Microbial Technology*. 46(6): 520-525.

[8] Jungles, M. K., Figueroa, M., Morales, N., Val del Río, Á., da Costa, R. H. R., Campos, J. L., Mosquera-Corral, A. and Méndez, R. 2011. Startup of a Pilot Scale Aerobic Granular Reactor for Organic Matter and Nitrogen Removal. *Journal of Chemical Technology and Biotechnology*. 86(5): 763-768.

[9] Isanta, E., Suárez-Ojeda, M. E., del Río, Á. V., Morales, N., Pérez, J. and Carrera, J. 2012. Long Term Operation of a Granular Sequencing Batch Reactor at Pilot Scale Treating a Low-Strength Wastewater. *Chemical Engineering Journal*. 198: 163-170.

[10] Muda, K., Aris, A., Salim, M. R., Ibrahim, Z., Yahya, A., van Loosdrecht, M. C., Ahmad, A. and Nawahwi, M. Z. 2010. Development of Granular Sludge for Textile Wastewater Treatment. *Water Research*. 44(15): 4341-4350.

[11] Federation, W. E. and APH Association. 2005. Standard Methods for the Examination of Water and Wastewater. *American Public Health Association (APHA):* Washington, DC, USA.

[12] Ghangrekar, M. M., Asolekar, S. R. and Joshi, S. G. 2005. Characteristics of Sludge Developed under Different Loading Conditions during UASB Reactor Start-up and Granulation. *Water Research*. 39(6): 1123-1133.

[13] McSwain, B. S., Irvine, R. L. and Wilderer, P. A. 2004. The Influence of Settling Time on the Formation of Aerobic Biogranules. *Water Science and Technology*. 50(10): 195-202.

[14] Qin, L., Liu, Y. and Tay, J. H. 2004. Effect of Settling Time on Aerobic Granulation in Sequencing Batch Reactor. *Biochemical Engineering Journal*. 21(1): 47-52.

[15] Hemalatha, R. and Anbuselvi, S. 2013. Physicohemical Constituents of Pineapple Pulp and Waste. *Journal of Chemical and Pharmaceutical Research.* 5(2): 240-242.

[16] Zheng, Y.M., Yu, H.Q. and Sheng, G.P. 2005. Physical and Chemical Characteristics of Granular Activated Sludge from a Sequencing Batch Airlift reactor. *Process biochemistry*. 40(2): 645-650.

[17] Kushwaha, J. P., Srivastava, V. C. and Mall, I. D. 2013. Sequential Batch Reactor for Dairy Wastewater Treatment: Parametric Optimization; Kinetics and Waste Sludge Disposal. *Journal of Environmental Chemical Engineering*. 1(4): 1036-1043.

[18] Tay, J. H., Liu, Q. S. and Liu, Y. 2001. Microscopic Observation of Aerobic Granulation in Sequential Aerobic Sludge Blanket Reactor. *Journal of Applied Microbiology*. 91(1): 168-175.

[19] Schwarzenbeck, N., Borges, J. M. and Wilderer, P. A. 2005. Treatment of Dairy Effluents in an Aerobic Granular Sludge Sequencing Batch Reactor. *Applied Microbiology and Biotechnology*. 66(6): 711-718.

[20] Sobeck, D.C. and Higgins, M.J. 2002. Examination of Three Theories for Mechanisms of Cation-Induced Bioflocculation. *Water Research*. 36(3): 527-538.

[21] Ren, T. T., Liu, L., Sheng, G. P., Liu, X. W., Yu, H. Q., Zhang, M. C. and Zhu, J. R. 2008. Calcium Spatial Distribution in Aerobic Biogranules and Its Effects on Biogranule Structure, Strength and Bioactivity. *Water Research*. 42(13): 3343-3352.

[22] Adav, S. S., Lee, D. J., Show, K. Y. and Tay, J. H. 2008. Aerobic Granular Sludge: Recent Advances. *Biotechnology advances*. 26(5): 411-423.

[23] Solís, M., Solís, A., Pérez, H. I., Manjarrez, N. and Flores, M. 2012. Microbial Decolouration of Azo Dyes: A Review. *Process Biochemistry*. 47(12): 1723-1748.

[24] Al-Amrani, W. A., Lim, P. E., Seng, C. E. and Ngah, W. S. W. 2014. Factors Affecting Bio-decolorization of Azo Dyes and COD Removal in Anoxic–Aerobic REACT Operated Sequencing Batch Reactor. *Journal of the Taiwan Institute of Chemical Engineers*. 45(2): 609-616.

[25] Panswad, T. and Luangdilok, W. 2000. Decolorization of Reactive Dyes with Different Molecular Structures under Different Environmental Conditions. *Water Research.* 34(17): 4177-4184.

[26] Libra, J. A., Borchert, M., Vigelahn, L. and Storm, T. 2004. Two Stage Biological Treatment of a Diazo Reactive Textile Dye and the Fate of the Dye Metabolites. *Chemosphere*. 56(2): 167-180.

[27] Sponza, D. T. and Işık, M. 2005. Reactor Performances and Fate of Aromatic Amines through Decolorization of Direct Black 38 Dye under Anaerobic/Aerobic Sequentials. *Process Biochemistry*. 40(1): 35-44.

[28] Ong, S. A., Toorisaka, E., Hirata, M. and Hano, T. 2005. Treatment of Azo Dye Orange II in a Sequential

Anaerobic and Aerobic-Sequencing Batch Reactor System. *Environmental Chemistry Letters*. 2(4): 203-207.

[29] Pandey, A., Singh, P. and Iyengar, L. 2007. Bacterial Decolorization and Degradation of Azo Dyes. *International Biodeterioration & Biodegradation*. 59(2): 73-84.

[30] Nawaz, M. S. and Khan, S. J. 2013. Effect Of HRT On SBR Performance for Treatability of Combined Domestic and Textile Wastewaters. *Journal of the Chemical Society of Pakistan.* 35(2): 527-532.

[31] Sathian, S., Rajasimman, M., Radha, G., Shanmugapriya, V. and Karthikeyan, C. 2014. Performance of SBR for the Treatment of Textile Dye Wastewater: Optimization and Kinetic Studies. *Alexandria Engineering Journal*. 53(2): 417-426.

[32] Kee, T. C., Bay, H. H., Lim, C. K., Muda, K. and Ibrahim, Z. 2015. Development of Bio-biogranules using Selected Mixed Culture of Decolorizing Bacteria for the Treatment of Textile Wastewater. *Desalination and Water Treatment*. 54(1): 132-139.

[33] Abu-Ghunmi, L. N. and Jamrah, A. I. 2006. Biological Treatment of Textile Wastewater using Sequencing Batch Reactor Technology. *Environmental Modeling & Assessment*. 11(4): 333-343.