Advances in Environmental Technology 2 (2021) 79-90

Advances in Environmental Technology

Iranian Reasearch Organization for Science and Technology (IROST)

Journal home page: https://aet.irost.ir/

Performance of sequential batch reactor coupled to physical system for landfill leachate treatment: A pilot scale design

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ARTICLE INFO

Article history:
Received 29 June 2021
Received in revised form 11 September 2021
Accepted 13 September 2021

Keywords:
Landfill
Leachate
SBR
Filtration
Fly ash

ABSTRACT

Landfills are intended for the management and disposal of municipal solid waste, which produces high volumes of leachate. The complex nature of the landfill leachate leads to various serious problems regarding water quality and human health. Hence, landfill effluents need to be treated before discharge to foul sewers or natural resources to reduce their negative effects and to comply with regulatory standards. The present study focuses on both biological and physical treatments of leachate from a controlled landfill created in Fez, Morocco, using a sequential batch reactor (SBR) coupled with a filtration system. The filtering material was characterized by scanning electron microscopy (SEM). The results obtained show that the latter has a great capacity in the treatment of the effluent. Also, for leachate, high-performance liquid chromatography coupled with ultra-violet (HPLC-UV) indicated the presence of detergent, which led to the formation of foam. Many parameters such as temperature, pH, and cycle time were also considered for their effect on the treatment. The results demonstrated the reliability and the high performance of the developed treatment system as it allowed a total elimination of BOD_5, 98% of the COD, a removal rate of 100% for NH_4, 78% for NO_2-, and 84% for NO_3-. Besides, this treatment system seemed able to eliminate fecal contamination and pathogenic germs. Thus, the present sequential batch reactor proved efficacy for landfill leachate treatment on a pilot scale design, promoting its development for a properly designed and implemented full-scale commercial product.

1. Introduction

The production of household and industrial waste constantly increases [1], leading to serious environmental pollution problems [2]. Leachate is generated in landfills as a result of precipitation, infiltration, compression, and waste degradation with variable quality and quantity [3]. Landfill leachates can generally be defined as a dark liquid with a foul odor [4], containing especially biodegradable and refractory organic materials such as humic, fulvic compounds [5] and heavy metals [6]. The chemical and microbiological

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DOI: 10.22104/AET.2021.5034.1365

composition of leachate is extremely complex and variable, depending on the residual deposit nature, and it is strongly influenced by the age of the landfill [7]. Landfills used for 5 to 10 years are considered young, while those with periods longer than 10 years are considered old [8]. Its composition and formation are strongly influenced by climatic conditions and the dynamics of the decomposition process, such as adsorption, biodegradation, and dissolution [9-12]. The inappropriate treatment of landfill leachate will cause potential long-term damage to human health, the environment on surface or groundwater, animals, and plants [13]. For this reason, the treatment of landfill leachate has been a research challenge for all countries [14]. The treatment of leachate from municipal landfills presents unique problems from a technical point of view, mainly due to the high chemical oxygen demand (COD) (6000–15 000 mg l − 1) and ammonium ion contents (500-3000 mg l - 1), high COD / BOD5 ratio, and the presence of toxic compounds such as metal ions [15]. Landfill leachate management has received special attention in recent years, especially for municipal areas [15]. Recently, many biological [16-19] and physico-chemical processes [20-23] have been developed to treat landfill leachate. The physico-chemical process can remove most contaminants. However, it is expensive and produces secondary pollution, while biological methods are the most common leachate treatment technique because of their simplicity and cost-effectiveness [24]. The main purpose of this study is to test a Sequential Batch Reactor (SBR) for landfill leachate treatment on a pilot scale design to eliminate the chemical and biological load to gain an effluent that meets the rejection standards using biological and physical treatment. The SBR biological process is mainly known as a municipal wastewater treatment process [25] efficient in eliminating pollutant loads such as COD, BOD_5, MES, or even a reduction in nitrogen forms and heavy metals [26]. The physical treatment is based on a filtration column packed with fly ash, which is characterized by a high adsorption capacity thanks to a silico-aluminous matrix contributing to the neutralization of negative charges of organic matter contained in the leachates [27]. Our work concerns a pilot-scale leachate treatment plant composed of two systems: a biological system using an aerobic sequential batch reactor coupled to a physical system in anaerobic conditions using a two-stage packed column filtration with fly ash.

2. Materials and methods

2.1. Presentation of the installation site

The installation of the pilot was carried out at the first controlled landfill created in the city of Fez, Morocco, under the direction of the company ECOMED that specializes in the construction and the valorization of the controlled landfills. The landfill was located at the Sidi Harazem road in the commune of Ain Bida, 11 km from the city center (Figures 1 and 2). The samples were collected and transported to the laboratory according to aseptic standards.

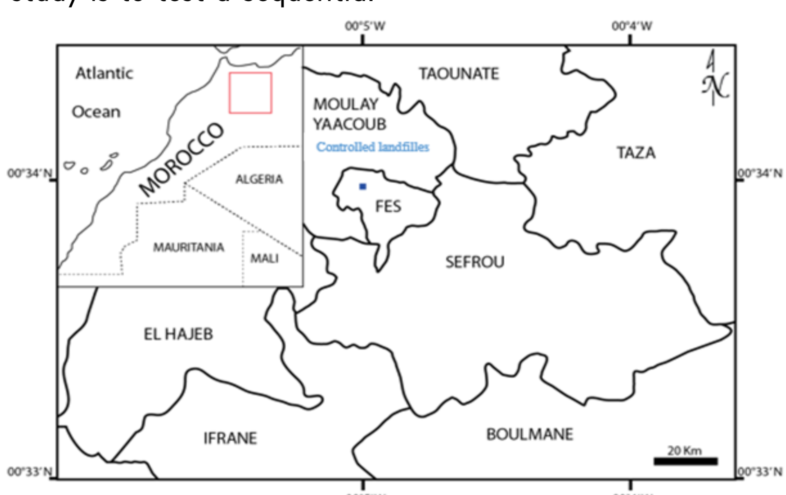

Fig. 1. Geographical location of the study site.

Fig. 2. The pilot at the Fez landfill, A= The Sequential Batch Reactor (SBR), B= Column of storage, C&D= Columns of filtration.

2.2. Biological treatment of leachates in a sequential batch reactor

The leachate treatment is carried out by the sequential batch reactor (SBR) system, which is a biological process using activated sludge [5]. Its operation is more practical than other conventional treatment systems. It has been widely used in wastewater treatment industries as it offers several advantages such as process flexibility, cost-effectiveness, and high biodegradation efficiency [28,29]. Our system was based on the following conduct: an aeration tank with a total volume of $10m^3$, a diameter of 2m, a height of 2.7 m, and a working volume of 6 m^3. The treatment of leachates was at a medium load. Every day a volume of leachates was introduced into the SBR and aerated until the elimination of the biodegradable organic matter. The aeration time was an automated 21 hours, with 13 kg of oxygen per hour. The sludge settling time was two hours, and at the end of the cycle, a quantity of excess sludge was withdrawn. Thus, the volume of the purified effluent was withdrawn and replaced by new raw leachate, and a new cycle begins.

2.3. Physical treatment (Filtration after coupling)

A coupling of the SBR treatment system with filtration using a natural support was performed in this study. The raw leachates treated in the aeration tank (SBR) were conveyed to the filtration column (Figure 2) with the following dimensions:

- Three tanks of 3 m^3 each: one for the storage of leachates after treatment in SBR and the other two for filtration.
- The two filtration tanks are filled with a natural support (Fly Ash), with a particle size of 20 microns.
- Both tanks are lined with a 20 cm layer of coarse gravel and a 15 cm layer of fine gravel. Above these layers, the fly ash is deposited on a layer of 65 cm.
- A hydraulic residence time of 2.36 days for each tank.
- A sprinkler for each filter tank that sprinkles leachate on the column in a homogeneous way.

2.4. Analytical methods

2.4.1. Physico-chemical analysis

The temperature and pH were detected by a pH meter; the electrode is of type X 22 Felt. Dissolved O2 (mg/l) was continuously monitored and measured by an oximeter, type WTW OXI 315i. The COD, NH_{4+} - N, NO_{2}- , NO_{3}- , TKN, PO_4, $SO_{4,}$ and BOD_5 were measured according to standard methods described by Rodier [30]. These analyses were carried out three times for each sample.

2.4.2. Microbiological analysis

The microbiological analyses of the leachate samples were carried out immediately after collection or after treatment to avoid any possible change in the microbial concentration. These analyses were carried out three times for each sample in order to obtain the average. The analyses were carried out according to the method described by Rodier [30].

2.4.3. HPLC-UV analysis

A high-performance chromatography analysis was carried out to analyze the presence of detergents in the raw leachate. The sample was filtered with 0.45 µm filters before analysis by HPLC (UV–vis). Separation of the compounds was performed on a Wakosil C18HG (5 µm, 4.6 × 150 mm) at a temperature of 40 °C. The elution was carried out in gradient mode using a binary solvent mixture composed of water acidified with 0.2% phosphoric acid (solvent A) and methanol/acetonitrile 50/50 (solvent B). A linear gradient was run from 96% (A) and 4% (B) to 50% (A) and 50% (B) during 40 min; it was changed to 40% (A) and 60% (B) for 5 min. And during 15 min, it was changed to 0% (A) and

100% (B), after reequilibration for 12 min to initial composition. The mobile phase flow rate was 1 ml/min, and the injection volume of the sample was 20 µl. All compounds of our sample were identified by comparing their retention times with the standard (Triton) reference detergent analyzed.

2.4.4. Morphological analysis

The morphological analysis of the fly ash was performed by a scanning electron microscope (SEM).

2.4.5. Statistical analysis

One-way analysis of variance (ANOVA) tests were performed to determine the significant effects of the SBR in the removal of COD, NH_{4+}, NO_2-, NO_3-, TKN, PO_4, SO_4, and BOD_5. All the tests were performed in GraphPad Prism8 software.

3. Results and discussion

3.1. Biological treatment of leachates in a sequential batch reactor

Table 1 summarizes the main physical and chemical properties of the leachate; the leachates from the controlled landfill of the city of Fez did not meet the rejection standards. The COD values obtained during this study reached up to 17575.75 mg/l, with an average of 12619.29 mg/l. The BOD_5 values of the leachates studied were 10500 mg/l with an average of 10250 mg/l. Also, there was a high concentration of ammonium NH_4 up to 235.76 mg/l, which is an indicator of pollution caused by urban waste. In addition, a slightly high concentration of SS was noted; this could be explained by the high organic and mineral load, which was due to the nature of the waste. Besides, the pH of 8.35 was slightly basic. These values are not in the range of Moroccan standards of the general limit values for discharges into surface and groundwater (Table1).

Table 1. Results of leachates physico-chemical analyzes.

	Maximum values	Minimum values	Average values	General limit values for discharges into surface and groundwater
pH	8.85	8.41	8.60	5.5-9.5
Temperature	12.8	11.2	12	-
Electrial conductivity (ms/cm)	45.8	44.8	45.3	-
SS (mg/l)	3550	1024	1808.5	100
COD (mg d'O2/l)	17575.75	6606.06	12619.29	500
BOD_5 (mg d'O_2/l)	10500	1000	10250	100
NH_4 (mg/l)	235.76	13.64	182.88	-
NO_3^- (mg/l)	6.57	4.80	5.74	-
NO_2^- (mg/l)	8.80	8.28	8.59	-
PO_4 (mg/l)	3.87	1.31	2.44	2
SO_4 (mg/l)	412.91	217.08	304.32	600

During the SBR treatment, we noticed the emission of the foam. For this, silicone was added as an anti-foaming agent to prevent the loss of biomass. Normally, the presence of the foam is due either to the existence of detergents or the presence of filamentous bacteria in high concentration (*Microthrix parvicella*) [31]. Due to the complete absence of filamentous bacteria, the detergents were analyzed by HPLC in the raw leachates. The chromatogram obtained (Figure 3) shows that detergents of the order of 16.26 g/l are present in comparison with the chromatogram of a Triton reference detergent analyzed under the same conditions.

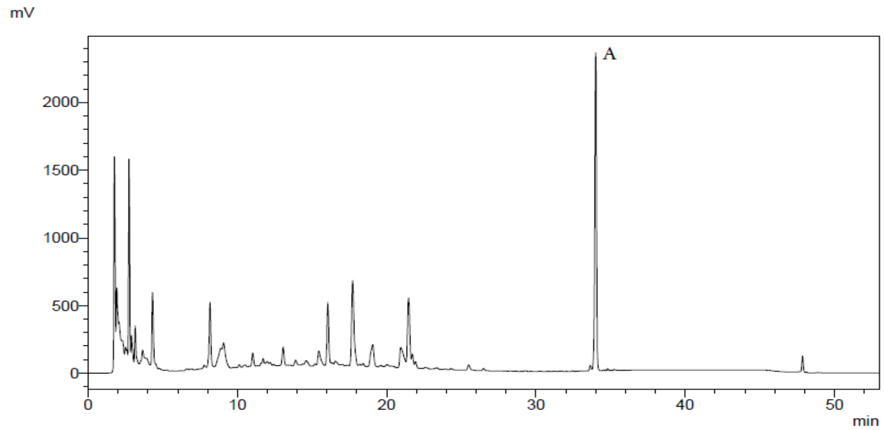

Fig. 3. Chromatogram of leachate with detergent peak at t = 35 min A = Triton.

3.1.1. Variation of pH, dissolved oxygen, and temperature in a sequential batch reactor

The main parameters that influence the removal mechanisms of nitrogen and organic matter are pH, dissolved oxygen (DO), and temperature [32] since biological treatment is based on organisms that are sensitive to these parameters. PH is also a very important parameter that affects the efficacy of the SBR treatment. It affects the metabolic activity of microbial substrates [33], as well as the performance of the activated sludge. Figure 4 shows that the pH after the treatment varies between 7.3 and 8.2.

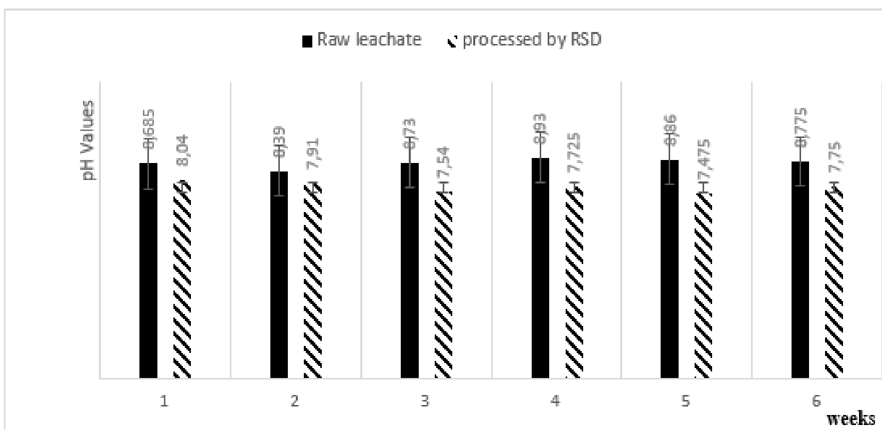

Fig. 4. Variation of the pH in the sequential batch reactor.

The concentration of dissolved oxygen is considered the most important control parameter in the nitrogen removal process. Excessive dissolved oxygen levels lead to unnecessary energy consumption due to high aeration and can affect anoxic processes. In the case of a low concentration of dissolved oxygen, there will be an inhibition of the growth of nitrifying bacteria. The nitrification process is the biological transformation of ammonia to nitrate that depends on the availability of oxygen [34]. During the SBR treatment, the dissolved oxygen concentrations ranged from 2.6 to 3.8 mg O_2/l (Figure 5), which explains the decrease in nitrogen forming during treatment, such as ammonium ion nitrification.

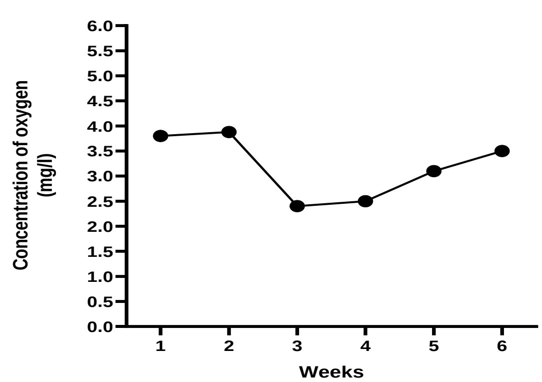

Fig. 5. Variation of dissolved oxygen in the sequential batch reactor.

The temperature determines the speed and rate of the reactions of biochemical degradation. The higher the temperature is, the faster the reactions [33]. Bacterial growth increases when the temperature increases [35]. Temperature is a necessary variable that influences nitrification and the rate of growth of nitrifying microorganisms in the temperature range between 5 and 30 °C. In this interval, an increase of 10 °C promotes an increase in the nitrification rate by a factor of two to three [36]. According to Figure 6, the temperature of the SBR treatment in this period varies between 29 °C and 35 °C.

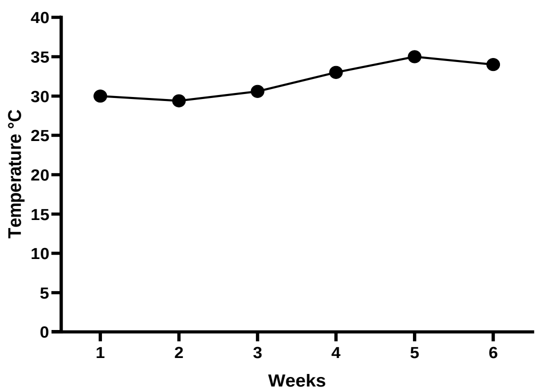

Fig. 6. Variation of the temperature in the batch sequential reactor.

3.1.2. The effect of cycling per day on the efficiency of the removal of organics and nitrogen in the SBR

The results of the removal of COD, BOD5, TKN, NH_{4+}, NO_2-, NO_3-, PO_4, and SO_4 under various cycles per day are shown in Figure 7. According to Figure 7, the abatement rates are very similar; it can be seen that the effluent at each treatment point in the bioreactor exhibits similar or very close biodegradation by the microorganisms. Due to a biomass adapted (strong acclimatization) to the effluent, this very close decomposition develops even with two or more cycles per day. The statistical study (one-way ANOVA) confirmed that the effect (factor 1) of the SBR operating cycles (one and two cycles per day) on the removal of COD, NH_{4+}, NO_{2-}, NO_{3-}, TKN, PO_4, SO_4, and BOD_5 had no significant effect between a treatment with one cycle and two cycles per day. But for NO_{3-} and SO_4, there was a slightly significant impact ($P = 0.05$).

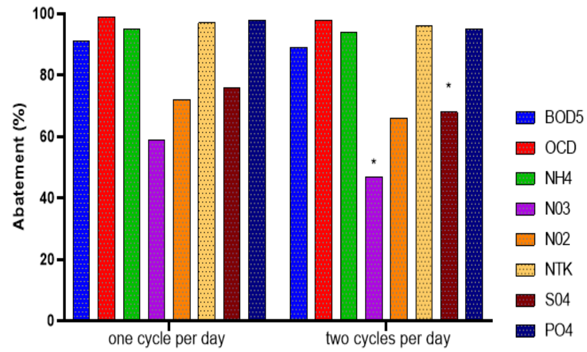

Fig. 7. Rate of reduction of the parameters after treatment of the leachate by the SBR with one and two cycles per day.

3.2. Physical treatment of leachates by filtration - fly ash

According to the morphological observation of fly ash, the latter consists of spherical particles, which are responsible for the nature of the porosity since the interstices created between them form irregular pores. As shown in Figure 8, the fly ash has an irregular morphology of particle size and shape with different particle shapes and sizes.

3.3. Coupling of SBR treatment system and filtration

Table 2 shows the results of the physicochemical analyses before and after SBR and filtration treatment, which are compared with the raw leachate to determine the abatement rate. The results are presented as mean values of the raw leachate and the mean value after SBR treatment coupled with the filtration treatment over the study period. According to the physicochemical analysis, the leachates of the controlled dump of Fez do not meet the standards of rejection according to COD, BOD_5, form nitrogenous, orthophosphate and SS, which are characterized by very high concentrations. After leachate treatment by sequential aeration in the SBR, there is a decrease in the concentration of the parameters. Regarding the COD after the SBR treatment, the concentration for the BOD_5 is 1742.28 mg/l, and there is a decrease in the concentration up to 80 mg/l.

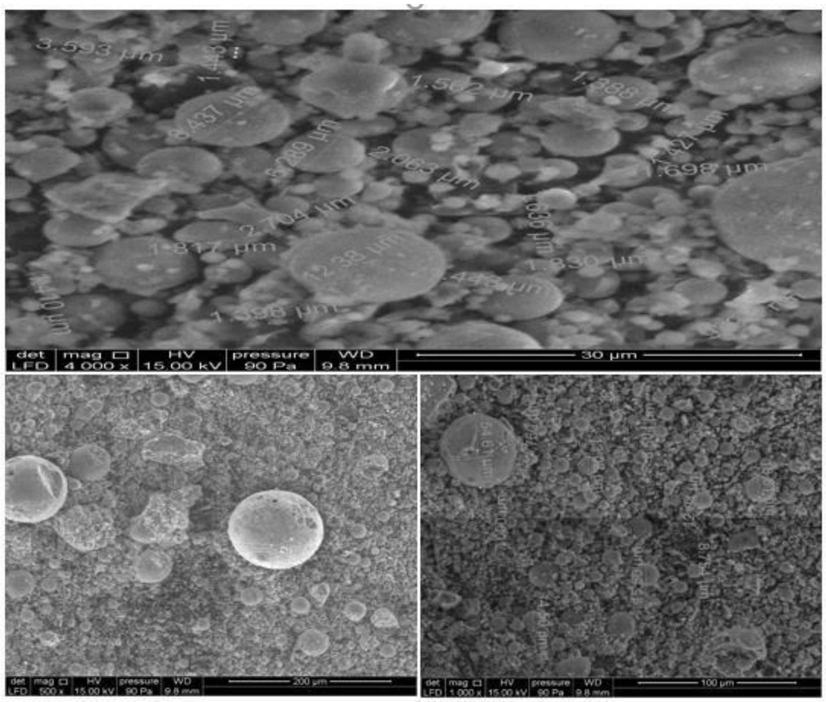

Fig. 8. Observations of fly ash structure at scanning electron microscopy (SEM), (A) Magnification 30x, (B) Magnification 100x, and (C) Magnification 200x.

These results can be explained by the performance of the sludge used in the SBR, which degrades the biodegradable organic matter present in the leachate, as well as by the presence of a purifying biomass. When comparing the present results to other studies, the results showed that SBR was similar to the data reported by O. Ouidiane and Mohamed (2016). The SBR showed efficacy in the treatment of domestic wastewater; it led to an efficiency of 96% in terms of COD removal and 98% for BOD_5 [26]. On the other hand, according to H. Yazdani et al. (2020), the SBR contributed strongly to the elimination of COD in sanitary wastewater [37]. The results in Table 2 show almost total denitrification (99%) through the filter column. This can be explained by the anaerobic conditions of the column and the presence of denitrifying bacteria in the biofilms, which favor denitrification. In addition, the filtration also allows for almost total nitrification or the reduction of 99% of the ammonium ions. After coupling with the filtration system, there is a very important reduction of the COD, the BOD_5, the SS, the nitrogenous forms, the orthophosphate, the sulphates, and up to a 100% reduction of the SS through the filtration column with concentrations much lower than those of the Moroccan standard of rejection. The following points can explain this strong elimination:

- The small particle size of the fly ash does not exceed 20 µm, which allows an increase of the adsorption surface resulted from the decrease of the dimensions of the adsorbent grains.
- The high content of SiO2 (silico-aluminous structure) of the fly ash [38]. It is an adsorbent with a high electrical polarity, rich in silica, aluminum, and iron, and positively charged.

The latter contributes to the neutralization of the negative charges of nitrites, nitrates, orthophosphate, and sulfates, which traps them by chemical bonds. We can also explain this increase in abatement rates by the formation of biofilms inside the column. The pollutants of the effluent can either adsorb on the cell membrane of the organisms forming the biofilms or be assimilated by the biofilms [39]. Table 3 presents the results of the microbiological analyses of the leachate before and after treatment with SBR coupled with filtration. Table 3 shows that the indicators of fecal contamination (Fecal Coliform and Fecal Streptococci) are eliminated with a 100% abatement rate and a total absence of pathogenic germs (Staphylococci); also, there is a complete elimination of yeasts.

The results reported in Table 4 reflect a reduction of metal ions in the leachate filtrate, with concentrations below Moroccan standards for direct discharges and high abatement rates.

Table 2. Leachate analysis results after SBR treatment coupled with the filtration column.

	Average values raw leachates	Average values of leachates treated with SBR	Average values of leachate treated by filtration	% abatement	Limit values of discharges into surface and underground waters
Temperature (°C)	12	12	20	-	-
pH	8,21	7,61	8,24	-	5,5-9,5
Electrical conductivity (ms/cm)	45	35,7	18,99	-	-
NH_4 (mg/l)	2877	366	1,2	99	-
NO_3^- (mg/l)	5,35	208,05	3,79	99	-
NO_2^- (mg/l)	8,74	2,485	0,95	61	-
SO_4 (mg/l)	304,32	203,54	54	73	600
PO_4 (mg/l)	2,44	4,96	0,13	97	2
COD (mg/l)	12619,29	1742,28	136,36	92	500
BOD_5 (mg/l)	10250	80	25	68	100
SS (g/l)	5,72	4,41	0	100	100 (mg/l)
TKN (g/l)	2,6	0,5	0,1	80	-

Table 3. Results of microbiological analyzes of raw leachates treated with RSD coupled with filtration.

	Raw leachates	Leachate treated by the SBR	Leachate treated by filtration	% Abatement
Total coliforms(UFC/ml)	$83\ 10^4$	$70\ 10^4$	0	100
Fecal coliforms (UFC/ml)	0	0	0	100
Fecal streptococci (UFC/ml)	8700	0	0	100
Staphylocoques (UFC/ml)	$67\ 10^4$	$56\ 10^4$	0	100

Table 4. Heavy metal results from pilot-processed leachate effluent.

	Raw leachate	leachate treated by SBR	leachate treated by filtration	% Abatement	Limit values of discharges into surface and underground
Aluminum (mg/l)	7,03	15,397	0,622	95	10
Cadmium (mg/l)	<0,01	<0,01	<0,01	-	0,25
Chromium (mg/l)	14,66	11,71	0,411	97,1	0,5
Iron (mg/l)	3,432	<0,01	<0,01	-	5
Fer (mg/l)	25,08	15,55	0,093	99,6	2
Zn (mg/l)	9,46	5,58	0,0923	98	5

Fig 9. Discoloration of leachate treated with SBR and filtration A: Raw leachate, B: leachate treated by SBR, C&D: leachate treated by filtration after coupling (tank 1 and tank 2).

From Figure 9, we observed a total discoloration of the leachate effluent treated by filtration. And this is explained by the retention of organic and mineral matter by fly ash. According to Tables 3 and 4, the proposed treatment system has successfully eliminated all fecal contamination germs and pathogens. And this could be explained by the fact that the bacteria are retained in the support (fly ash) by adsorption. As well, there is a reduced rate of Fe, Mn, and Ni up to 95%, 97%, and 98%, respectively. This can be justified by the high content of SiO_2 (silico-aluminous structure) of these ashes [40]. It was reported by N. Koshy and D. Singh (2016) that silico-aluminous structure could retain heavy metals by trapping them in their pores. It is an adsorbent with a strong electrical polarity in addition to its richness in silica, aluminum, and iron, which are positively charged [41]. Besides, according to F. Adams et al. (2019), silicate is one of the major functional groups that exist in the adsorbents [42]. In comparison with other leachate treatment studies, the proposed system proved its effectiveness without any malfunction during treatment compared to other systems. For example, the evaporation treatment consists of a chemical treatment to eliminate carbonate scaling problems, which requires cleaning with sulfamic acid and high-pressure rinsing water [43]. On the other hand, the reverse osmosis treatment uses modular open-channel systems and requires cleaning with great efficiency regarding scaling, fouling, and especially biological fouling [44]. Furthermore, the one-way ANOVA test confirmed that the combination of SBR and filtration using fly ash affected significantly the removal of all physicochemical parameters, pathogenic microorganisms, and heavy metals (Tables 2, 3, and 4)

($P = 0.001$) ($\alpha = 0.05$).

The proposed treatment system has allowed for effective removal of the pollutant load, proving the reliability and power of the technique. In addition, it offers a satisfactory solution at a reasonable cost, as well as requiring limited manpower and a low technical skilled staff, the possibility of reusing the treated leachate in irrigation, and the absence of sludge recirculation, particularly in the SBR. This reduces the operating cost.

4. Conclusions

The proposed treatment system, consisting of a sequential batch reactor operating under aerobic conditions coupled with a physical system using a two-stage packed column filtration with fly ash, has shown high leachate removal capacity. It allowed for a 99% reduction of BOD_5, 98% of COD, 78% of NO_2^-, 99% of NH_4, and 100% of SS. This treatment system eliminated all fecal contamination germs and pathogenic germs, so the leachate treated with the system could be used in irrigation since it meets Moroccan standards of rejection. The obtained results show significant biodegradation by microorganisms in the SBR system. Besides, the high performance of the treatment system, especially the filtration column, is due to the efficiency of the fly ash, which has a silica-alumina matrix that exceeds 94% in terms of silica, aluminium, and iron. These contribute to the neutralization of the negative charges of the chemical parameters and adsorb organic matter and microbial germs. In conclusion, the proposed system has the capacity to be used for the treatment of all the leachates in the local landfill, as well as in controlled landfills in other Moroccan cities. And it can be used efficiently as an international model.

Acknowledgements

Our acknowledgement goes to the Moroccan Ministry of Environment for financing and monitoring this project during the study period.

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