ORIGINAL ARTICLE



MSW stabilization in an anaerobic bioreactor landfill and evaluation of in-situ leachate treatment potential with the help of quadric model

Anil Nain¹ · Rajesh Kumar Lohchab¹ · Kulbir Singh¹ · Mikhlesh Kumari¹ · Jitender Kumar Saini¹

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Abstract

Recirculation of the leachate using bioreactor technology for in-situ treatment of leachate is an efficient method for reducing the contaminants and cost of ex-situ treatment as well. The current study was performed to evaluate the impact of leachate recirculation in anaerobic bioreactor on leachate treatment as well as municipal solid waste stabilization. The reactor was loaded with properly mixed 23.79 kg of solid waste with compaction of waste and total 20 L leachate was prepared in the bioreactor by adding water. Physico–chemical characteristics of leachate were estimated by using standards methods. The result reveals that the pH, alkalinity and VFA/ alkalinity were varied from 5.56 to 7.58, 7650 to 1875 mg/L and 1.44 to 0.44 which were within the optimal range of anaerobic digestion. Chemical oxygen demand is an indication of organic pollutants and 97% of COD removal was observed after leachate recirculation study. Quadric model was employed to investigate the behavior of operational parameters over time scale. Model indicated the highest phosphate decline (5.3%) with time but highest increase was observed for total kjeldhal nitrogen (4.3%). After completion of leachate treatment study, a 26.09% reduction in volume of MSW was found.

Keywords Municipal solid waste \cdot Leachate recirculation bioreactor \cdot Anaerobic degradation \cdot MSW stabilization \cdot Quadric model

Introduction

Appropriate management of solid waste has become a challenging task particularly in developing countries due to urbanization and industrialization [1]. Foremost, municipal solid waste (MSW) management possibilities involve composting, incineration and landfilling. Landfilling is a dominant approach used for disposal of MSW in developing countries due to its technological and economic aspects [2, 3]. In a traditional anaerobic landfill, adequate measures are required to avoid migration of leachate to groundwater and inflowing of peripheral water to the landfill [4]. The prospect of anaerobic energy generation is more ecofriendly and efficient than another [5–7]. Anaerobic degradation is a biological process that can be employed to treat organic portion of solid wastes. The extent of digestion process *i.e.*

reduction of organic pollution load, biogas production and MSW stabilization is highly dependent on the operational setup, operating parameters, MSW composition, existing microbial population and reactor type [8, 9]. Recent investigations have revealed that the anaerobic decomposition process could be designed for municipal solid waste treatment and biogas production [10–12].

The large fraction of MSW consists of organic matter and when discarded in landfill, it undergoes conversions causing formation of leachate, an extremely contaminating discharge due to the complexity of its constituents, which contains huge quantities of organic pollutants estimated as chemical oxygen demand, suspended solids, nitrogenous compounds, heavy metals and significant amount of inorganic salts [13–17]. Hence, leachate must be treated or managed cautiously to decrease the adverse impact on environment, for instance stubborn organics can combine with organic compounds or heavy metals and transfer contaminants into the environment [18–20]. Based on the principle of anaerobic landfill, the bioreactor technology was developed for the reduction of leachate pollution load and fast solid waste stabilization [21–23]. The main purpose of an anaerobic

Rajesh Kumar Lohchab rajeshlohchab@gmail.com

¹ Department of Environmental Science and Engineering, Guru Jambheshwar University of Science and Technology, Hisar, Haryana, India

bioreactor landfill is to increase microbial activities by leachate recirculation which accelerates the conversion and degradation of organic portion of solid waste [23–27].

Leachate recirculation improves the moisture content which in turn provides conditions for the effective transmission of substrates and nutrients for microbes, redistribution of enzymes and microorganisms throughout the waste mass and dilution of higher concentration of contaminants [28–31]. The rates of leachate recirculation maintain the stability among hydrological and biological processes of solid waste decomposition. In the initial phase of study, the higher rate of leachate recirculation should be implemented to increase dissolution of waste content and establishment of higher population of methanogens [32, 33]. Hussain et al. [34] proposed that initially increased leachate recirculation enhanced hydrolysis, decomposition efficacy and production of volatile fatty acid during acidogenic phase of organic waste. The rate of leachate recirculation is determined on the basis of the volume of MSW bed in a bioreactor. The leachate volume to be recirculated is calculated under different conditions i.e. 2%, 10%, 13% and 30% of MSW volume loaded in the reactor [35, 36]. The intensive rate of recirculation is where a larger volume ($\geq 30\%$ of the waste volume) of leachate recirculated through the waste bed. The intensive recirculation rate formed channels through the waste which increase the leachate flow and decrease the retention time of the leachate in the bioreactor [35]. The moderate rate of recirculation, where sufficient volume (10-20% of the waste volume) of leachate recirculated through MSW filled in bioreactor. The lower recirculation rate is where small volume (2% of the waste volume) of leachate recirculated through the waste. San and Onay [37], studied the impact of the leachate recirculation rate and recirculation approach on the anaerobic decomposition of MSW. The recirculated leachate volume of 2 L and recirculation approach of four times in a week were provided the maximum stabilization of MSW. Chugh et al. [35] stated that the methane generation rate and COD were dropped considerably when the leachate recirculated volume was 30% of the volume of MSW filled the bioreactor. Sponza et al. [36] reported that the increase in leachate recirculation rate from 9 L/day (13% of the reactor volume) to 21 L/day (30% of the reactor volume) influences the production of methane. The three times surge in recirculation rate caused reduction in cumulative methane production and increment in volatile fatty acid and COD concentrations were observed. The findings of this investigation indicated that the recirculation rate of 9 L/day enhances the biodegradation of MSW in anaerobic bioreactors. According to Li et al. [38] moderate leachate recirculation intensities contribute to reducing microbial consortium disruption, which enhances biogas production but intensive leachate recirculation provokes rapid vertical flow of leachate along preferential flow routes, causing insufficient consumption of existing moisture storage. Therefore, the leachate recirculation rates should be carefully chosen and optimized to minimize the disturbance and wash-out of the viable microbial consortium. In addition to this, other operational and design problems are at risk of environmental exposure if leachate is applied to the surface and requirement of liner, leachate and gas collection of management facilities. However, the study on effects of leachate recirculation on anaerobic degradation of MSW requires further investigation, particularly focused on continuous recirculation.

The present research aimed to observe the effect of leachate recirculation on MSW stabilization in cylindrical shaped anaerobic bioreactor. The physico-chemical characteristics of leachate from bioreactor were also investigated during leachate recirculation. Quadric model was applied to analyze the mode of operating parameters over time scale.

Material and methods

Characteristics of MSW

MSW samples were collected from the main dumping site of Hisar city. 2 kg of this mixed sample was collected in polyethylene bags, brought to the laboratory, and analyzed for moisture content immediately. The remaining samples were stored and analyzed for other important parameters.

The characteristics of MSW samples like moisture content (MC) and volatile contents (VC) were analysis according to ASTM standards [39, 40]. The total organic carbon (TOC) of the sample was analyzed with the help of TOC-V analyzer (Shimadzu) using zero air as a carrier gas. The other parameters i.e. total kjeldahl nitrogen (TKN), sulphur and phosphorus were analyzed in laboratory by using standard methods [41]. All the above physico–chemical characteristics of MSW were also analyzed after completion of leachate recirculation study.

Bioreactor set-up and operation

The Leachate Recirculation Bioreactor setup and dimensions are given in Fig. 1 and the composition of MSW used in bioreactor is shown in Table 1. The properly mixed 23.79 kg of MSW having volume 0.046 m³ was added by compaction to the bioreactor and total 20 L leachate was prepared in the bioreactor by adding fresh water. The bioreactor and leachate collection container were sealed air tight. The bioreactor was kept at steady state conditions for 15 days, afterwards the recirculation of the leachate was started and the bioreactor was operated under anaerobic conditions for 270 days. Leachate recirculation was done at moderate leachate recirculation rates @ 28 mL/min for 12 h daily in such a way that the 20 L collected leachate was completely recirculated because



Fig. 1 Lab-scale leachate recirculation bioreactor

Table 1 Composition of MSW used in bioreactor

S. no	Component	Weight (g)	Fraction (%)
1	Food	13,485	56.70
2	Papers	1538	6.47
3	Cardboard	1225	5.15
4	Plastic/rubber	386	1.62
5	Polythene	880	3.70
6	Textile	1366	5.74
7	Sanitary waste	345	1.45
8	Leather	26	0.11
9	Crockery	247	1.04
10	Yard waste	1265	5.32
11	Wood	184	0.77
12	Aluminium foil	149	0.63
13	Metal	156	0.66
14	Glass	353	1.47
15	Inert	2180	9.17

moderate leachate recirculation intensities contribute to reducing microbial consortium disruption, which enhances biogas production. Therefore, in order to achieve the purpose of continuous recirculation this moderate rate was preferred after reviewing literature. The 50 mL leachate samples were collected on weekly basis for first 5 months and per 2 weeks in last four months from the outlet port and analyzed for various physico-chemical parameters. The leachate volume was made up by adding distilled water equal to the volume of leachate samples collected and accordingly dilution factor was incorporated in the equation used for the calculations. The leachate collected in the vessel was recirculated into the reactor from the top with the help of peristaltic pump.

Analytical procedures for leachate samples

The physico-chemical characteristics of leachate i.e. pH, electrical conductivity (EC), alkalinity, chloride, calcium, magnesium, sodium, potassium, sulphate, phosphate, total Kjeldahl nitrogen (TKN) and chemical oxygen demand were analyzed using "Standard Methods of Analysis of Water and Wastewater" [41].

The volatile fatty acid (VFA) was analyzed with the help of Gas Chromatograph (PerkinElmer 680) using flame ionization detector (FID) and capillary column (Elite-WAX) having length 30 m and inner diameter (ID) 0.32 mm. The pH of samples maintained to 2–3 using ortho-phosphoric acid. The samples were filtered through a membrane having pore size 0.2 μ m and dia. 25 mm using a syringe filter. A small amount (0.5 μ l) of prepared sample was injected through an auto-injector for estimation [42].

Gas analysis

The composition of gases was analyzed with the help of Gas Chromatograph (PerkinElmer Clarus 680) using

thermal conductivity detector (TCD) and Elite-Plot Q column (30.0 m length \times 80.53 mm ID). Nitrogen was used as a carrier gas at a flow rate of 4.0 mL/min. The fraction of the gases was calculated using standard plots containing a mixture of CH₄ and CO₂ [42].

Quadric model

To capture the behaviour of different variables related to operating parameters (physico–chemical properties of leachate) of the study in relation to time, regression growth model was employed using SPSS 16 software. The data of physico–chemical properties of leachate collected on daily and weekly basis was converted to log scale and used for statistical analysis. Based on the results, quadric model was found best fit to evaluate the change in operational parameters over time scale. The quadric model determines the relationship between dependent variable *Y* and independent variable *X*.

The quadric model equation as below:

$$Y = \alpha + \beta X_i + \Upsilon X_i^2 + U_i,$$

whereas, *Y* is the variable (operating parameter i.e. parameter of physico-chemical properties); *X* is the time; α intercept; β shows the change in *Y* with time (negative value means *Y* decreases with time and positive value means *Y* increases with time). Υ shows the acceleration of change in *Y* with time (negative value means rate of change of *Y* diminishes with time and positive value means rate of change of *Y* increases i.e. accelerates with time); *U* is error term.

Results and discussion

The MSW stabilization through leachate recirculation was studied at lab scale in bioreactor operated under anaerobic condition. The results of change in leachate quality with time are shown in Table 2.

Characteristics of leachate

Initial phase of leachate recirculation determines the modification and acclimatization of MSW decomposition from aerobic to anaerobic phase. After initial adjustment and acclimatization the pH became near to neutral i.e. optimum. Optimum pH ranged from 6.5 to 7.5 for anaerobic degradation [43]. Up to 6th week of leachate recirculation study, the pH of the reactor was decreased from 6.45 to 5.56. It may be due to acidogenesis and formation of volatile fatty acids [44]. After initial decrease, an increase in pH was observed. This increase in pH may be due to onset of methanogenic activity. It results in increased methane production and decreased carbon dioxide, hydrogen and volatile fatty acid production [45, 46]. The pH value varied from 5.56 to 7.92 and ultimately remained at around 7.4. pH of the reactor shows the reactor was working effectively during the study period and anaerobic conditions were maintained properly in the reactor.

The leachate conductivity reflects total ionic concentration of solutes and is an extent of capacity of solution to conduct an electric current [47]. The value of leachate conductivity was 9.52 mS/cm in 1st week of recirculation, after that it increased upto 11.32 mS/cm till 9th week and further then it started to decrease with time. At the end of study, the minimum value of EC had observed and it was 5.35mS/cm. It might be due to formation of metal hydroxides which are insoluble [48].

Chloride is non-degradable in leachate and variation in its concentration is mainly used to estimate the leachate quality [49]. Decline in chloride concentration may be because it was chemically precipitated by their interaction with other substances present inside the bioreactor at suitable conditions of pH and other parameter. Decline in its concentration might be caused by the dilution effect of distilled water used to maintain total makeup of leachate [47]. The initial chloride concentration was observed 2627 mg/L and then it reduced to 1065 mg/L by the end of study period. Erses et al. [47] observed the dilution and washout effect on chloride and they observed 79% removal for chloride during anaerobic degradation of MSW in bioreactor landfill.

Hardness in form of $CaCO_3$ was analyzed in the leachate sample. The hardness decreases with increased leachate recirculation [50]. Ca and Mg contents of the leachate samples showed variability due to more chemically reactive in nature and both are available as significant precipitants. During the leachate treatment period, initially hardness, calcium and magnesium increased and then decreased. This may be due to formation of CaCO₃ and Mg(OH)₂ precipitation and consumption of calcium and magnesium by the microorganisms [47]. Lohchab and Sigh [51] and Snehlata et al. [52] were also observed similar decreasing trend of hardness, Ca and Mg during leachate recirculation study.

The inorganic elements like Na and K act as nutrients for microbes. Potassium is the chief cellular inorganic cation and co-factor for certain enzymes whereas sodium maintains the osmotic regulation within microbial cells [13]. The results showed a decreasing trend of sodium and potassium concentration during the leachate recirculation period in anaerobic bioreactor. The initial sodium concentration in the leachate sample collected from the reactor was 529 mg/L, after that it tends to increase till 675 mg/L up to 5th week and at last it started to decrease with time and reached to 118 mg/L. The potassium in the leachate sample collected from 1050 to 195 mg/L at the end of recirculation study. The results of the present study

	Table 2 Variá	ations i	n leachate qual	lity during	anaerobic treatm	nent in bioreacto	or								
	Time (weeks)	Hq (EC (mS/cm)	Alka- linity (mg/L)	Chloride (mg/L)	TH (mg/L)	Calcium (mg/L)	Mag- nesium (mg/L)	Na (mg/L)	K (mg/L)	Phos- phate (mg/L)	Sul- phate (mg/L)	TKN (mg/L)	VFA (mg/L)	COD (mg/L)
2 6.05 9.88 6.25 2.968 6.052 1740 57.41 57.8 58.9 59.9 3 5.66 1.02 5.33 2.940 6.053 17.3 41.35 41.37 5.06 57.0 57.9 57.0 5	-	6.45	9.52	7650	2627	5625	1620	383	529	1050	33.48	284	645	3400	26,137
3 6.05 10.02 5.32 2.840 6875 1825 561 631 11.53 41.95 61.95 620 520 6 5.56 10.13 51.75 2911 673 123 43.37 420 673 57.07 400 57.07 700 7 5.56 10.88 6.33 27.93 16.30 17.34 37.67 410 900 7740 8 5.54 11.12 6.330 27.93 16.30 17.33 33.4 390 10.05 7740 900 7740 8 5.54 11.12 6.330 27.93 13.35 14.6 54.4 10.35 54.0 7740 900 7740 11 6.58 10.56 58.75 51.50 13.75 38.4 75.7 10.60 7740 78.0 7740 78.0 7740 78.0 7740 78.0 7740 78.0 7740 78.0 75.7 76.0	2	6.26	9.88	6250	2698	6625	1740	552	582	1140	37.41	358	580	3930	23,062
4 5.78 11.13 517 201 6750 1800 546 653 1215 4237 420 670 570 6 556 1123 6137 233 163 673 233 673 730 730 7 566 1058 6575 282 573 153 153 153 153 734 306 733 8 558 1056 653 232 1535 1535 1535 1537 740 733 9 651 1132 6135 2375 1430 1375 441 559 734 700 737 9 651 1132 6135 2375 1430 1375 441 539 234 100 737 703 11 682 054 1375 441 730 1375 441 739 703 703 11 682 055 238 430 <td>3</td> <td>6.05</td> <td>10.02</td> <td>5325</td> <td>2840</td> <td>6875</td> <td>1825</td> <td>561</td> <td>634</td> <td>1155</td> <td>41.96</td> <td>395</td> <td>620</td> <td>5260</td> <td>22,550</td>	3	6.05	10.02	5325	2840	6875	1825	561	634	1155	41.96	395	620	5260	22,550
5 5.60 11.22 5550 2982 6525 1725 537 675 1230 42.96 455 877 700 7 5.65 10.88 6225 3124 6575 2873 1620 498 657 392 7810 7810 8 5.45 10.68 6575 2823 1500 1637 440 555 1060 27.64 290 7300 9 6.31 11.32 6125 2873 1450 475 410 576 790 780 11 655 10.66 5850 2555 1450 1375 414 557 790 790 12 737 1051 5150 1375 446 514 1052 790 700 13 7.55 944 4750 1230 1375 344 535 241 107 557 14 7.56 944 478 921 23	4	5.78	11.13	5175	2911	6750	1800	546	653	1215	43.37	420	670	5700	28,187
6 5.56 10.88 6.22 31.4 6.20 180 498 647 1248 37.67 410 590 7810 7 5.66 10.66 6575 282 5875 1620 443 525 1175 33.4 300 1025 7900 8 5.51 11.5 6530 5765 5555 5555 5555 5555 5555 5555 5555 5555 5556 5130 1375 416 514 205 5560 11 6.52 10.65 5830 2556 5130 1375 416 514 205 557 <t< td=""><td>5</td><td>5.69</td><td>11.22</td><td>5550</td><td>2982</td><td>6525</td><td>1725</td><td>537</td><td>675</td><td>1230</td><td>42.96</td><td>435</td><td>877</td><td>7020</td><td>29,212</td></t<>	5	5.69	11.22	5550	2982	6525	1725	537	675	1230	42.96	435	877	7020	29,212
$ \ \ \ \ \ \ \ \ \ \ \ \ \ $	9	5.56	10.88	6225	3124	6250	1680	498	647	1248	37.67	410	950	7810	33,312
8 5.34 11.15 6.350 2760 56.25 15.25 15.37 14.40 5.59 11.80 31.51 33.4 10.80 7740 10 6.51 11.32 61.52 56.27 53.35 14.40 4.35 55.5 56.7 53.75 14.40 4.35 55.5 56.7 53.75 14.40 4.35 55.5 56.7 53.75 51.50 13.75 51.60 57.64 200 58.6 56.70 57.01 57.00 56.00 56.00 56.00 56.00 57.00 56.00 57.01 57.00 56.00 57.01 57.00 57.00 56.00 57.01 57.00 56.00 57.01 57.00 56.00 57.00 56.00 57.00 57.00 56.00 57.00 56.00 57.00 57.00 56.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00 57.00	7	5.65	10.65	6575	2982	5875	1620	443	625	1175	33.94	390	1025	1960	33,210
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10 6.56 10.66 58.50 2.556 51.50 13.75 416 51.4 10.25 2.4.44 2.53 9.27 6610 11 6.82 10.55 56.55 2485 4875 13.25 380 521 967 23.74 23.7 601 12 7.57 10.51 5100 2414 4750 1170 385 443 865 2.2.64 241 710 5700 13 7.55 9.44 4750 1270 385 433 433 610 23.92 560 570 570 15 7.92 9.42 4170 1080 352 371 733 19 570 570 16 7.75 9.12 3975 1846 3500 850 343 670 1992 217 53 560 17 7.75 9.12 3576 583 732 516 510 510 510 510	6	6.31	11.32	6125	2627	5375	1450	425	525	1069	27.64	290	988	7020	30,545
	10	6.56	10.66	5850	2556	5150	1375	416	514	1025	24.44	253	927	6610	28,187
	11	6.82	10.55	5625	2485	4875	1325	380	521	67	23.74	246	822	6290	23,062
	12	7.37	10.51	5100	2414	4750	1250	394	458	921	23.92	235	780	6170	18,450
	13	7.52	10.45	4175	2272	4525	1175	385	434	865	22.64	241	710	5700	13,837
	14	7.65	9.84	4250	2201	4375	1120	383	408	839	21.06	230	635	5680	12,812
	15	7.92	9.42	4125	2059	4150	1080	352	371	735	19.59	224	610	5290	11,787
	16	7.87	9.36	4050	1917	3850	980	340	343	670	19.92	217	523	5060	12,300
	17	7.75	9.12	3975	1846	3500	850	334	292	635	18.75	219	510	4800	9737
	18	7.68	8.95	3450	1775	3225	825	283	276	590	16.28	207	475	4440	8200
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26 7.43 8.12 2475 1420 1475 400 115 181 305 12.83 192 317 3180 28 7.58 7.75 2325 1491 1225 315 106 173 265 12.57 184 260 2170 30 7.54 6.68 2175 1349 1150 280 109 165 225 11.5 175 238 1970 32 7.43 6.36 2025 1278 1080 255 108 142 212 11.21 172 209 1700 34 7.38 5.45 1950 1207 980 220 104 126 215 10.55 164 202 1250 36 7.35 5.54 1875 1065 850 175 1055 164 205 1050 36 7.35 5.54 1875 1065 850 175 1055 <t< td=""><td>24</td><td>7.49</td><td>8.39</td><td>2550</td><td>1491</td><td>1680</td><td>480</td><td>117</td><td>202</td><td>365</td><td>16.06</td><td>194</td><td>290</td><td>3400</td><td>4612</td></t<>	24	7.49	8.39	2550	1491	1680	480	117	202	365	16.06	194	290	3400	4612
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32 7.43 6.36 2025 1278 1080 255 108 142 212 11.21 172 209 1700 34 7.38 5.45 1950 1207 980 220 104 126 215 10.55 164 202 1250 36 7.35 5.54 1875 1065 850 175 101 117 205 10.55 161 195 1050 38 7.42 5.35 1925 1100 805 155 102 118 195 9.11 162 192 970	30	7.54	6.68	2175	1349	1150	280	109	165	225	11.5	175	238	1970	2562
34 7.38 5.45 1950 1207 980 220 104 126 215 10.55 164 202 1250 36 7.35 5.54 1875 1065 850 175 101 117 205 161 195 1050 38 7.42 5.35 1925 1100 805 155 102 118 195 9.11 162 192 970	32	7.43	6.36	2025	1278	1080	255	108	142	212	11.21	172	209	1700	2050
36 7.35 5.54 1875 1065 850 175 101 117 205 10.25 161 195 1050 38 7.42 5.35 1925 1100 805 155 102 118 195 9.11 162 192 970	34	7.38	5.45	1950	1207	980	220	104	126	215	10.55	164	202	1250	1537
38 7.42 5.35 1925 1100 805 155 102 118 195 9.11 162 192 970	36	7.35	5.54	1875	1065	850	175	101	117	205	10.25	161	195	1050	1282
	38	7.42	5.35	1925	1100	805	155	102	118	195	9.11	162	192	970	1025

corresponded with the finding of Reddy et al. [53]. They observed 66% reduction for sodium and 81% reduction for potassium.

During study period, value of phosphate in leachate sample was 33.48 mg/L in 1st week, after that it increased to 43.37 mg/L till 4th week and then, it started to decrease with time. At the end of study, phosphate concentration reduced to 9.11 mg/L. The phosphate concentration may be decreases due to its assimilation by microorganisms in the bioreactor. The similar investigation of leachate recirculation in anaerobic bioreactor showed similar decreasing trend of phosphate concentration [51]. Sulphate is used as micro nutrient for microbes but its higher concentration is inhibitor for the anaerobic digestion [13]. The initial value sulphate content in leachate sample was 284 mg/L which increased up to 435 mg/L and then, sulphate concentration started to decrease and reduced to 162 mg/L at the end of leachate recirculation study. In addition to it, most of the sulfur compounds were converted into the H₂S gas, which cause decrease in the sulphate content of the reactor [54].

Most of nitrogen in the bioreactor is ammonia produced during degradation of protein [55]. Initial concentration of TKN in leachate sample was 645 mg/L, which increased to maximum (1050 mg/L) on 8th week of recirculation. After that there was a gradual decrease to minimum level of 192 mg/L at the end of study. Initially increase in nitrogen may be due to liberation of organically bound nitrogen during decomposition of waste. Nain and Lohchab [56] observed the similar decreasing trend of TKN in anaerobic bioreactor study.

The variation in concentration of volatile fatty acid with time seems to be a significant parameter for estimation of MSW decomposition [57]. Initially, the concentration of VFA was increased from 3400 to 7960 mg/L upto 7th week of recirculation. It may be due to accumulation of organic acids by hydrolysis and acidogenesis. Acidogens are most effective at pH 5.5-6.5. Their concentration grows quickly by conversion of complex organic compounds into VFAs [58]. The result was consistent with other studies that in a conventional landfill, VFA contents reached to its highest level before decreasing due to the establishment of methanogenic phase [20, 59]. After that VFA shows the decreasing trend and reached minimum level of 970 mg/L at the end of study. The VFA concentration reduced as the daily biogas production increased, this may be due to utilization of VFA by methanogenic bacteria as a substrate to produce biogas and new cells.

Alkalinity is essential to maintain optimum pH for maximum microbial activity [60]. The initial value of alkalinity in leachate sample was 7650 mg/L and at the end of study it gets reduced to 1925 mg/L. The value of alkalinity in leachate sample collected from reactor shows the decreasing trends during treatment. Alkalinity greater than 2000 mg/L in the reactor indicated adequate alkalinity to maintain optimum condition for methanogenesis [61]. After 32th weeks of recirculation, the reactor alkalinity was less than 2000 mg/L. This may be due to dissolution and precipitation of metals carbonates [62].

The VFA/Alkalinity is a measure of buffering capacity which reflects stability of bioreactor. The lower ratio usually imitates higher efficiency of anaerobic degradation [63]. Initially, VFA/Alkalinity was increased from 0.44 to 1.40 due to increase in VFA concentration during acidogenic phase, then it was gradually decrease and reached a minimum to 0.50 (Fig. 2). The ratio was observed within the limits of operational condition during the methanogenic phase i.e. less than 0.8 [64]. Result clearly shows that performance of the reactor was excellent as VFA/ Alkalinity was in optimum range with maximum biogas yield during the methanogenic phase.

After slight decline, the COD in the bioreactor increased significantly at the beginning i.e. reached 33,312 mg/L till 6th week. This is due to hydrolysis and leaching of soluble organic and inorganic compound from solid waste into leachate. After initial increase it started to decrease with minimum value 1025 mg/L at the end of leachate recirculation. This decrease in COD may be due to fast decomposition of solid wastes under anaerobic condition resulting in conversion of VFA into CH_4 , CO_2 , and H_2S etc. [36]. These results are similar to the outcomes of Han et al. [23], they reported 93.01%, 96.85% and 95.74% of COD removal. The similar trends of reduction were also observed in the other investigations [52, 57, 66–68].



Fig. 2 VFA/alkalinity ratio during recirculation of leachate

Quadric model discussion

To capture the change in operating parameters over time, by using regression analysis, the quadric regression model was identified/ selected on the basis goodness of fit criteria. The correlation coefficient (R^2) value of 0.696 or more and p value (significance F) close to 0 of the quadric model showed that model was best fit. The observed values and predicted values were compared for each case to check the model suitability and Fig. 3 shows that the variance patterns of the observed values and the predicted values are similar for every case. The estimated results of the model are presented in Table 3. The term 'Y' is a variable which denotes different operating parameters i.e. parameters of physico-chemical properties. '\beta' shows the change in operating parameters with time. The negative value of β means the operating parameter (Y) decreases with time and positive value means parameter increases with time. 'Y' denotes the rate of change of operating parameters. The negative value of Υ means rate of change of parameter (Y) decreases with time and positive value of Υ means rate of change of Υ increases with time i.e. accelerates with time.

The positive value of β (0.029) in result shows that pH was increased with time. The rate of increase of pH with time was 2.9%. The value of Υ (0.00) denotes that the rate of pH increment does not increase or decrease with time, though the change of acceleration was zero. The alkalinity has negative value of β (- 0.015) which shows decline in alkalinity was 1.5% with per unit time. The negative value of Υ (-0.001) shows that the rate of reduction in alkalinity content decreases with time with a rate of 0.1%. The negative values of β for chloride (-0.010) and sulphate (-0.046) represent the decreasing trends in their contents with per unit time, but acceleration of change are zero because the value of Υ is 0.00. The amount of decline per unit of time was 1% for chloride and 4.6% for sulphate, respectively. The rate of reduction of magnesium and sodium with time was 1.5% and 0.9%. The negative value of Υ exhibits that the rates of decline in magnesium and sodium quantity was decreased with time at a rate 0.2%. The result also reveals that the phosphate was decreased with time at a rate 5.3% and rate of reduction of phosphate has been decreased with time at a rate 0.008%. The phosphate has shown highest amount (5.3%) of decline with time but decrease in acceleration was lowest. This decrease in phosphate may be due to its uptake by microbes for their growth and multiplication. The quadric model showed that with time electric conductive increase was 2.8% with diminishing rate 0.2%, total hardness increase was 0.9% with diminishing rate 0.3%, total kjeldahl nitrogen increase was 4.3% with diminishing rate 0.3%, potassium increase was 0.7% with diminishing rate 0.3%, volatile fatty acid increase was 1.3% with diminishing rate 0.6% and chemical oxygen demand increase was 3.4%

with diminishing rate 0.5%. The amount of increase was highest in case of total kjeldahl nitrogen 4.3% with highest diminishing rate of 0.3%. These trends of quadric modelling are similar with trends of observed values.

Above trends of quadric modelling as well as observed values of most parameters of leachate during their recirculation indicate that the organic and inorganic contaminants levels of leachate were increased initially during hydrolysis and solubilisation of MSW but as MSW degradation/stabilization progressed with time they had decreased.

Biogas production

Biogas production rate and cumulative biogas production during leachate recirculation study are presented in Fig. 4. The changes in composition of methane and carbon dioxide in biogas during study period are shown in Fig. 5. The biogas generation in bioreactor was started from 2nd week of recirculation and it was exponentially increase from 6th week. The highest biogas production rate of 18 L/day was observed on the 18th week of leachate recirculation (Fig. 4). The methane percentage of biogas was also highest during this week and it was about 67.8% (Fig. 5). The methane peak percentage was noted 69.50% in anaerobic bioreactor [68]. Xu et al. [21] also noticed methane content about 68% during the anaerobic phase. The major gases formed during anaerobic degradation of MSW were methane and CO2. Initially methane content was lower up to 6th week and after that a gradual increase in CH₄ contents was observed with progress of MSW degradation. The average composition of biogas in bioreactor during the study period was 50% CH₄ and 30.8% CO₂. During methanogenic phase, methane (40-70%) and carbon dioxide (30-60%) are two major constituents of landfill gas [69]. The maximum methane production rate of 12 L/day was observed in 18th week. The result is consistent with the outcomes of San and Onay [37] and Erses et al. [47]. The cumulative biogas and methane productions were 1876 L and 938 L during the leachate recirculation study period. Erses et al. [47] determined maximum yield of methane as 158 L/kg dry refuse. Other studies have reported methane generation in the range of 20-170 L CH₄/ kg-dry waste [21, 65, 66, 68-71].

Stabilization of MSW

The characteristics of MSW before and after anaerobic treatment in bioreactor are shown in Table 4. The most crucial parameter enhancing waste degradation is the moisture content of the waste. Leachate recirculation provides necessary moisture content in the reactor which enhances the treatment of MSW as well as leachate recirculating through bioreactor. After leachate recirculation study, volatile contents were reduced to 45%. The total organic carbon and TKN were



Fig. 3 Comparison between variance patterns of the observed values and the predicted values of quadric model

reduced to 40% and 29% which may be discharged into leachate or gaseous phase. Similar study showed 30% reduction in carbon and 10% reduction in nitrogen [72]. Moreover, 43% and 27% reduction in phosphorus and sulphur contents were observed. The depth of MSW used in bioreactor was reduced from 65.5 cm to 25.8 at the end of study. Initially,



Fig. 3 (continued)



Fig. 3 (continued)



Fig. 3 (continued)

Tal	bl	e 3	3 (Qua	dric	model	s	howing	c	hange i	n	varial	ble	s wi	th	time
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Variable (Y)	α	В	Υ	R ²	F	Р
pH	1.686 (42.274)	0.029 (4.768)	0.000 (-3.161)	0.696	29.779	0.000
Electric conductive (EC)	2.265 (85.654)	0.028 (6.878)	-0.002 (-12.797)	0.964	344.756	0.000
Alkalinity (Alkal)	8.825 (148.729)	-0.015 (-1.645)	-0.001 (-3.963)	0.953	266.061	0.000
Chloride (Cl ⁻)	8.011 (226.785)	-0.010 (-1.754)	0.000 (-5.375)	0.971	430.510	0.000
Total hardness (TH)	8.781 (164.479)	0.009 (1.045)	-0.003 (-11.252)	0.986	898.376	0.000
Calcium (Ca)	7.412 (238.276)	0.024 (5.014)	-0.004 (-23.898)	0.996	3.115E ³	0.000
Magnesium (Mg)	6.332 (48.306)	-0.015 (-0.744)	-0.002 (-2.944)	0.899	115.341	0.000
Sodium (Na)	6.492 (135.663)	-0.009 (-1.235)	-0.002 (-8.044)	0.981	732.409	0.000
Potassium (K)	7.098 (128.680)	0.007 (0.852)	-0.003 (-9.824)	0.982	693.713	0.000
Phosphate (PO_4^{3-})	3.826 (65.015)	-0.053 (-5.811)	$-8.394E^{-5}(-0.287)$	0.961	317.589	0.000
Sulphate (SO_4^{2-})	6.065 (84.674)	-0.046 (-4.208)	0.000 (1.133)	0.865	83.391	0.000
Total kjeldhal nitrogen (TKN)	6.545 (66.126)	0.043 (2.842)	-0.003 (-6.835)	0.918	146.399	0.000
Volatile fatty acid (VFA)	8.253 (114.006)	0.113 (10.129)	-0.006 (-15.559)	0.962	332.530	0.000
Chemical oxygen demand (COD)	10.200 (115.596)	0.034 (2.543)	-0.005 (-11.784)	0.983	746.528	0.000



 $\ensuremath{\mbox{Fig.4}}$ Biogas production from anaerobic bioreactor during leachate recirculation

the stabilization of solid waste in bioreactor was observed 5–8% that may be due to the self-weight of MSW. The solid waste will also undergo fast and composite settlements as the result of self-weight and biological degradation when leachate recirculation is implemented along with a bioreactor technology [27, 73, 74]. At the end of leachate recirculation study, 25.8% settlement was observed in anaerobic bioreactor (Fig. 6). The MSW settlement provides more space for solid waste reached at the dumping site which increases the closure period of the site. The similar observations were observed in the investigations performed by [66, 67, 75].

Solid waste management system is very poor in India and requires research in this field according to local conditions. Large part of MSW in Hisar and other cities of India remain



Fig. 5 Gas composition of anaerobic bioreactor during leachate recirculation

unattended due to lack of information of the waste generation and their management technologies. This research provides the base for proper utilization of MSW according to local need and resources in eco-friendly manner with generation biogas as an energy source. Leachate recirculation bio-landfill can reduce the long-term risk in addition to several other advantages. The biogas and leachate analysis is performed on the weekly leachate recirculation basis at lab scale; though data of precise estimation of this process with field study is unavailable and impacts of individual operating parameters are to be investigated separately. The anaerobic bioreactor at different scales shows similar performances

 Table 4
 Characteristics of MSW before and after anaerobic treatment in bioreactor

Parameters	Before treatment	After treatment
Waste depth in bioreactor	65.5 cm	48.6 cm
Mass of waste	23.79 kg	11.54 kg
Volume of waste	0.046 m^3	0.034 m ³
Moisture content (%)	28.76	46.82
Volatile content (%)	45.38	25.17
Total organic carbon (%)	26.32	15.91
Total nitrogen (%)	1.27	0.9
Phosphate (%)	0.84	0.48
Sulphate (%)	0.49	0.36



Fig. 6 MSW settlement in anaerobic bioreactor during study period

and demonstrates better efficiency in eliminating leachate

contamination in terms COD, alkalinity, VFA, TKN etc. Leachate recirculation bioreactor landfill is one of the most significant and cost-effective technique for enhanced biodegradation and MSW stabilization (Table 5). Therefore, this cost effective and environmentally sound technology can be adopted after suspicious research and precautions.

Conclusion

After initial increase, a decreasing trend was observed in most of the physico-chemical parameters during the leachate treatment period. Removal of chloride, calcium, magnesium, sodium, potassium, phosphate, sulphate and TKN were 66%, 92%, 82%, 83%, 84%, 79%, 63% and 82%, respectively. The cations and anions present in leachate were used as a source of nutrients by the microbes, therefore their concentration were reduced due to their consumption by the microbes during anaerobic digestion of leachate in bioreactor.

The quadric model indicated that phosphate had highest amount of decline with time but sodium has lowest. Whereas, TKN has shown highest amount of increase with time but potassium has lowest. Subsequently, quadric modelling represented that the organic and inorganic pollutants of leachate were decreased and pH was increased with time.

The pH, alkalinity and VFA are important parameters which govern the working of bioreactors under anaerobic conditions and all these parameters were within optimum range of anaerobic treatment. The COD is the best indicator of pollution load of the leachate and the maximum COD reduction was 97%, whereas the reduction in the VFA was 88%. High reduction of COD and VFA indicate that the reactor working efficiency was very high. Therefore, the bioreactor landfill is another suitable option for disposal of the solid waste. One of the most significant and cost-effective technique for enhancing biodegradation in a bioreactor landfill is the leachate recirculation. Table 5 Comparison of different bioreactor technologies used for MSW leachate treatment

Treatment process	Leachate recirculation rate	Opera- tion time (days)	Parameters	Removal (%)	MSW stabilization	References
Anaerobic bioreactor	28 mL/min	270	COD	97	Settlement: 26%	This Study
			Alkalinity	75	VC reduction: 45%	
			VFA	88	TOC reduction: 40%	
			TKN	82		
Anaerobic bioreactor	Without Recirculation	222	COD	55	Settlement: 16%	[47]
			VFA	50	TOC reduction: 14%	
Anaerobic bioreactor	9 L/day	225	COD	51	Settlement: 27%	
			VFA	27	TOC reduction: 28%	
Anaerobic bioreactor	21 L/day	215	COD	42	Settlement: 26%	
			VFA	33	TOC reduction: 26%	
Anaerobic bioreactor	1 L of collected leachate was recir-	630	COD	98	Settlement: 5%	[44]
	culated once per week		Alkalinity	49	VC reduction: 43%	
			TKN	47		
Aerobic bioreactor	1 L of collected leachate was recir-	374	COD	96	Settlement: 37%	
	culated once per week		Alkalinity	85	VC reduction: 55%	
			TKN	93		
Hybrid bioreactor	1-3 times/day, 250 mL/time	300	COD	91	Settlement: 15%	[21]
			VFA	90	VC reduction: 52%	
Pilot-scale anaerobic bioreactor	0.8 m ³ leachate was recirculated	400	COD	97	Settlement: 19%	[20]
	over 1 h twice a week		Alkalinity	56		
			VFA	74		
Semi-aerobic bioreactor	0.8 m ³ leachate was recirculated	400	COD	95	Settlement: 26%	
	over 1 h twice a week		Alkalinity	60		
			VFA	83		

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