

TREATMENT OF LANDFILL LEACHATE BY ELECTROCOAGULATION

A Dissertation

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award of the degree of***

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By

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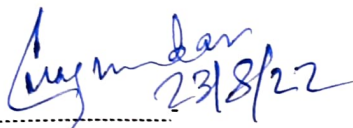


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ABSTRACT

In recent times treatment of landfill leachate by nonconventional methods such as the electrochemical process has been increasing for its treatment efficiency. Among the electrochemical techniques, the process of electrocoagulation (EC) has largely been employed to remove heavy metals from municipal and industrial wastewaters. However, solid waste landfill leachate treatment with the aim of Chemical Oxygen Demand (COD) removal and Ammonia-Nitrogen ($\text{NH}_3\text{-N}$) removal have not been studied so much using different types of electrodes. Thus, in this study, the efficacy of the process of electrocoagulation using iron and aluminium sacrificial electrodes has been evaluated for Chemical Oxygen Demand (COD) removal and Ammonia-Nitrogen ($\text{NH}_3\text{-N}$) removal from landfill leachate. Different operating conditions were studied for the removal of COD and $\text{NH}_3\text{-N}$ which includes inter-electrode distance, current density, pH and operating time. The results of the batch experiments showed a maximum COD removal of 100% using iron electrodes at the optimum operating conditions of current density of 3 mA/cm^2 , electrolysis time of 30 minutes and inter-electrode distance of 2 cm. Whereas the maximum $\text{NH}_3\text{-N}$ removal of 99.83% using aluminium electrodes at the optimum operating conditions of current density of 2 mA/cm^2 , electrolysis time of 45 minutes and inter-electrode distance of 4 cm.

Treatment of landfill leachate wastewater by electrocoagulation process using an aluminium and iron electrode was investigated in a batch electrochemical cell reactor. Response surface methodology based on central composite design was used to optimize the operating parameters for the removal of % COD and % ammoniacal nitrogen ($\text{NH}_3\text{-N}$) together with power consumption from landfill leachate. Effects of three important independent parameters such as current density (X_1), electrocoagulation time (X_2), and solution pH (X_3) of the landfill leachate sample on the % COD and % ammoniacal nitrogen ($\text{NH}_3\text{-N}$) removal with power consumption were investigated. A quadratic model was used to predict the % COD and % ammoniacal nitrogen ($\text{NH}_3\text{-N}$) removal with power consumption in different experimental conditions. The significance of each independent variable was calculated by analysis of variance. In order to achieve the maximum % COD and % ammoniacal nitrogen ($\text{NH}_3\text{-N}$) removal with minimum of power consumption, the optimum conditions were about current density (X_1)—2 mA/cm², electrocoagulation time (X_2)—20.683 minutes and initial solution of effluent pH (X_3)—7, with the yield of COD removal of 72.694%, and ammoniacal nitrogen ($\text{NH}_3\text{-N}$) removal of 86.025% when the inter electrode distance was 4cm and aluminium electrode was used. The electrocoagulation process could be applied to remove pollutants from industrial effluents and wastewater.

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LIST OF ABBREVIATIONS AND NOMENCLATURES

2 FI	Two factor interactions
3 FI	Three factor interaction
A	Ampere
AAS	Atomic Absorption Spectrophotometer
AC	Alternating Current
Aduj. R^2	Adjusted R^2
ANN	Artificial Neural Network
ANOVA	Analysis of Variance
AOP	Advanced Oxidation Process
AP	Adequate Precision
BBD	Box-Behnken Design
BDD	Boron Doped Diamond
BIS	Bureau of Indian Standards
BOD ₅	5-Day Biochemical Oxygen Demand
BP	Back Propagation
CCD	Central Composite Design
CD	Current Density
CETP	Common Effluent Treatment Plant
COD	Chemical Oxygen Demand
CPCB	Central Pollution Control Board, New Delhi
CV	Coefficient of Variance
DC	Direct Current
DOE	Design of Experiments
EC	Electrocoagulation
EDX	Energy Dispersive X-Ray
EF	Electrofloatation
ETP	Effluent Treatment Plant

FD	Factorial Design
FFD	Fractional Factorial Design
GA	Genetic Algorithm
HRT	Hydraulic Retention Time
I	Current
IS	Indian Standards
MCL	Maximum Contaminant Level
MLSS	Mixed Liquor Suspended Solids
MS	Mean Squares
MSE	Mean Square Error
MSW	Municipal Solid Waste
NH ₃ -N	Ammonia-Nitrogen
OFAT	One Factor at a Time
PLOF	Probability of Lack of Fit
Pred. R ²	Predicted R ²
PRESS	Predicted Residual Error Sum Of Squares
p-value	Probability Value
R	Correlation Coefficient
R ²	Coefficient of Determination
RSM	Response Surface Methodology
SBR	Sequencing Batch Reactor
sCOD	Soluble Chemical Oxygen Demand
SEM	Scanning Electrone Microscope
SNR	Signal to noise ratio
SS	Sum of squares
SS-SS	Stainless Steel-Stainless Steel
STP	Sewage Treatment Plant
TDS	Total Dissolved Solids
TKN	Total Kjeldahl Nitrogen
TOC	Total Organic Carbon

TP	Total Phosphorus
TS	Total Solids
TSS	Total Suspended Solids
USEPA	United States Environment Protection Agency
V	Volt
VFA	Volatile Fatty Acids
Y_1 & Y_2	Predicted Results of Removal (COD and Ammonia-nitrogen)
β	The Sum of a Constant
A, B, C	Parameters in Equation
WHO	World Health Organization

CHAPTER – 1

INTRODUCTION

1.1 Water: Global Perspective:

One of the biggest challenges of the 21st century is the availability of safe drinking water. Earth's water supply is only 2.5 percent fresh. Two-thirds of it are frozen in the polar ice caps and glaciers.

While the agricultural sector is the biggest consumer of water, the industrial and domestic sectors are also consuming more water. By 2050, about half of the world's grain production will be at risk due to water stress.

According to the World Bank and the UN, around 40 percent of the world is already affected by water scarcity. The issue is getting worse due to the increasing demand for water. It has been linked to conflicts and human displacement.

Water shortages are expected to become more prevalent in the future. Water shortages can be classified as physical or economic. The former refers to the lack of water resources to meet the demand, while the latter refers to the improper management of these natural resources. Both of these conditions have their own unique characteristics.

Proper water management & treatment techniques can help conserve and improve the quality of our water supply. This process would have a positive effect on society's socio-economic well-being.

1.1.1 Global Water Resources Scenario:

The volume of global water resources is $1.386 \times 10^{18} \text{ m}^3$ i.e., 1386 MKm³ and its distribution in nature is shown below.

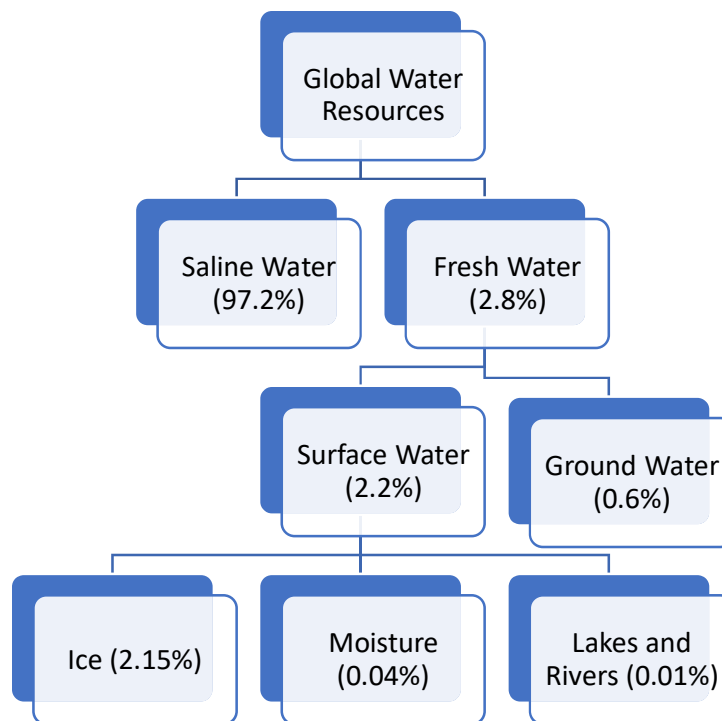


Fig 1: Global Water Resources Scenario

1.2 Indian Perspective:

According to a study conducted in 2007, the discharge of untreated waste water is the leading source of pollution in India. This issue is also linked to the lack of treatment capacity for the wastewater.

Most of the time, the government's sewage treatment plants are not maintained and operated properly. This contributes to the uncontrolled discharge of solid waste into the environment.

In India, the main cause of water pollution is the discharge of untreated sewage from cities, towns, and villages. Currently, the treatment capacity of the urban sewer system is only 11,786 million liters per day.

Many rivers in India are polluted due to the discharge of domestic waste. To prevent this, the government has established a network of water quality monitoring stations.

The monitoring network carries out water quality monitoring in 293 rivers, 94 lakes, 8 ponds, and 23 creeks across India. The samples are routinely analysed for various parameters such as dissolved oxygen, chlorine, and organic matter.

It is also important to minimize the contaminants that enter water bodies. This can be done through the removal of harmful substances and pollutants.

One of the processes by which water gets polluted is from the leachate which comes out from the landfill and that is a major problem in India. The topic is discussed below.

1.3 Landfills and Water Pollution:

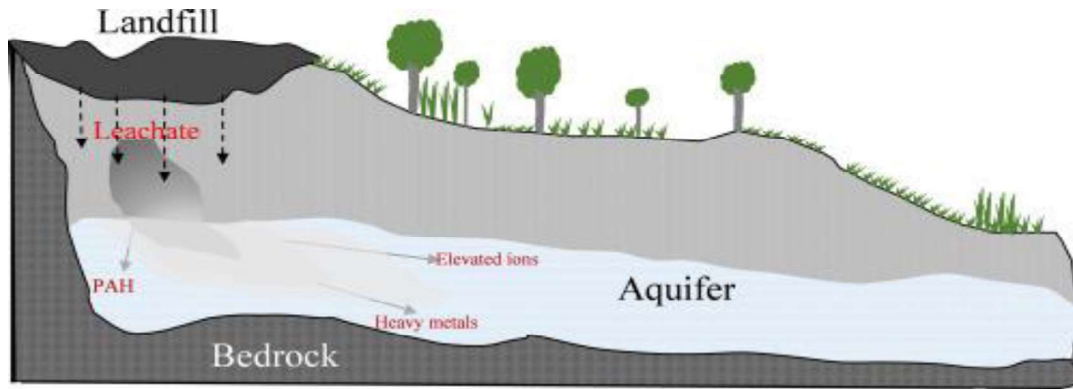


Fig 2: Landfills and Water Pollution scenario

Increased production of industrial and household waste is a result of the industrial and economic growth. In the past ten years, developed nations produced between 300 and 800 kilograms of trash per person annually. Urban solid waste is typically disposed of in sanitary landfills (Abbas et al., 2009). In accordance with different sources, such as municipal waste, biomedical waste, and industrial waste, solid waste can be divided into three categories. Municipal solid waste (MSW), also known as garbage, is the most prevalent type of solid waste and is primarily produced by apartment and business complexes. A landfill is a location where waste is dumped. This is the most traditional and well-known method of waste treatment, and it can also be used for short-term consolidation, storage, and transition. Sanitary landfill methods are still widely accepted and used for the final disposal of solid waste because of their financial advantages. The production of heavily contaminated leachate, which presents significant differences in both chemical composition and volumetric flow, is a significant drawback despite its many advantages. In conclusion, the management of municipal solid waste poses a significant environmental, economic, and social challenge for the world today, primarily due to the fact that waste production is outpacing population growth. About 0.15 million tonnes of municipal solid waste are produced daily in India (Rathod et al., 2013). According to research done by the National Environmental Engineering Research Institute (NEERI) in Indian cities, the amount of MSW generation varies between 0.21 and 0.35 kilogrammes per person per day in urban areas and increases to 0.5 kilogrammes per person

per day in large cities. Authorities have increased the requirements for pollution control every year as a result of the environmental impact of landfill leachate being recognised (Renou et al., 2008). But when there are strict environmental standards for surface and groundwaters are consistently followed, the disposal of landfill leachate becomes a major environmental concern.

Leachate from landfills is an effluent primarily produced by rainfall on the landfill's crest. Rainwater permeates the trash, causing physical mixing and chemical reactions among the waste's constituent parts. High concentrations of organic matter, ammonium, and toxic substances are frequently found in leachate. As a result, it is referred to as high strength wastewater. Surface and groundwater contamination from landfill leachate is the primary potential environmental impact. There are four groups of pollutants found in landfill leachate: dissolved organic compounds, inorganic macro components, heavy metals, and xenobiotic organic compounds. Domestic waste contains little quantity of heavy metals; hence its contribution in leachate is small (Kjeldsen et al., 2002). In tropical weather climate regions leachate generation is comparatively higher due to the greater magnitude of precipitation which penetrates into the landfill cell (Renou et al., 2008).

1.4 Treatment of Landfill Leachate:

Based on the period of the landfilling process, the composition of landfill leachate varies significantly (Altin, 2008). Three types of landfills can be distinguished based on their age: young, medium, and old. Young leachate, for instance, can be identified by its high organic fraction, which includes volatile organic acids, total organic carbon (TOC), high carbon dioxide (COD) levels, BOD₅, and a BOD₅ to COD ratio greater than 0.3 (Umar et al., 2010). Old leachate, in contrast is distinguished by a relatively less biochemical oxygen demand (BOD), slightly basic with pH > 7.5 and BOD₅ to COD ratio < 0.1 (W. Li et al., 2010). At this stage, humic and fulvic acid as well as NH₃-N are also produced as a result of anaerobic decomposition (Bashir et al., 2010). Throughout the stabilisation period, the organic portion will deteriorate. The non-biodegradable portion of COD is left unaltered in this process, which causes the BOD₅/COD ratio to decrease over time (Ahmed & Lan, 2012).

Before final discharge, leachate needs to undergo adequate treatment to reduce the high concentration of contaminants to an acceptable level (Hilles et al., 2016a). Leachate can be treated using a range of techniques, including physical, chemical, and biological ones. Leachate

can be treated physically and chemically to remove recalcitrant or non-biodegradable substances like phenolic compounds, organic halogens, and polychlorinated biphenyls. Biological treatment has been highly efficient in extracting nitrogenous and organic matter (Abbas et al., 2009) from immature leachate when the BOD₅ is high and the BOD₅/COD ratio is greater than 0.5 (Renou et al., 2008). As they have a low initial investment and ongoing operating expenses, biological treatments are still one of the best ways to treat leachate. A lower BOD₅/COD ratio indicates that the leachate is in a stable stage and that further biological decomposition is difficult (Aziz et al., 2010). Biological treatments have low effectivity closer to definitive elements such as halogenated bio-refractory organic compounds (AOX). Consequently, physical-chemical treatments are proposed because of extracting refractory substances beyond matured leachate, then additionally as a polishing quadrant for biologically handled leachate.

Effluents from landfills must be treated on site in order to meet disposal standards before being discharged into the sewer, on-site land, or in surface water. The majority of stabilised or biologically treated leachate is made up of large recalcitrant organic molecules that are difficult for biological treatment to handle. The majority of the time, a single biological or physicochemical process cannot achieve discharge standards (K. Wang et al., 2018); (Torretta et al., 2017); (Gotvajn et al., 2009). To meet the exacting quality requirements for direct leachate disposal into surface water, development of integrated systems for treatment—a combination of chemical, physical, and biological processes—is necessary. For the treatment of landfill leachate, combining oxidation and biological processes will be a wise decision (S. H. Lin & Chang, 2000).

For landfill leachates, new treatment methods under cleaner production technologies have been developed. When conventional biological methods cannot be used to treat complex industrial effluents, advanced oxidation processes (AOPs) are used. To increase the biodegradability of organic components and ensure their mineralization by biological treatment, AOPs are used as a pre-treatment for wastewater. Advanced oxidation processes are appropriate among leachate treatment methods because they chemically expel pollutants rather than transferring toxic chemicals into the leachate (Atmaca, 2009); (Deng & Ezyske, 2011). Conceptually, combining biological treatment with advanced oxidation processes is advantageous because it can result in higher treatment efficiencies when compared to the efficiencies of individual treatment units.

Advanced oxidation enhances oxidation in a treatment process by using radicals, typically hydroxyl radicals (W. Li et al., 2010); (Peralta-Hernández et al., 2009). The Fenton process is a popular AOP for reducing the impact of highly contaminated wastewater (Lopez et al., 2004). The excessive use of Fe^{2+} , which results in the release of a significant amount of iron sludge, is one of the clear drawbacks of the traditional Fenton process (Bui et al., 2019). According to several studies, the Electro-Fenton process (EF) is a promising method for extracting organic matter that is more effective and environmentally friendly than traditional Fenton processes (Barrera diaz et al., 2014; Ignasi Sires et al., 2014) (Sirés et al., 2014). The primary advantages of the EF process include in-situ reagent production, anodic iron electrode dissolution that results in less sludge production, regeneration of Fe^{3+} to Fe^{2+} at the cathode, and a higher and quicker rate of organic pollutant compound mineralization.

When treating landfill leachate or as a final step to meet water quality standards, membrane technologies are frequently used (Kjeldsen et al., 2002). Membrane filtration and traditional biological processes are combined in membrane bioreactors (MBRs). In MBR systems, membrane filtration takes the place of the settling tank, which is used in conventional biological treatment plants to separate solid from liquid. Thus, the advantages of both biological and membrane filtration processes are combined in the MBR process. The MBR treatment plant's footprint can be reduced by up to 50% compared to the traditional activated sludge process, and since the additional tertiary filtration steps are skipped, there are even more footprint reductions (Galleguillos, 2011). Considering the advantages of MBR system, it is ideally suitable for treatment of strong waste water.

The biodegradability of leachate reduces with growing age and conventional biological treatment by itself may be absolutely ineffective to meet the discharge standards. Combined AOP and MBR process can improve the biodegradability index and curtail pollutants at great extent. The total degradation rate of a combined AOP - MBR process is expected to be much higher than MBR process for treating a slowly biodegradable or biologically recalcitrant effluent like matured landfill leachate. This lower kinetics can end up with the requirement of greater capital investment. The integrated process is interesting in terms of lower capital investment and easiness of operation. As a result, the overall treatment series using MBR and the advanced oxidation process tends to be a successful way to treat landfill leachate and meet land disposal standards. The properties of the landfill leachate, technical applicability and constraints, effluent discharge options, cost sufficiency, regulatory requirements, and

ecological effects are crucial factors in determining the most effective treatment method for landfill leachate treatment (Mojiri et al., 2013).

1.5 Electrocoagulation:

The use of electrocoagulation to treat ship bilge water was first documented in early 1900. Since 1970, this technology has advanced significantly, leading to the filing of numerous patents in nations like Japan, Germany, China, France, and the USA. companies with a global presence, like Sanyo Electric Co., Exxon Research and Engineering Co., Germany, Adder Abase Anlagen A-G USA, Hitachi Ltd., Japan, Mitsubishi Electric Corp., and Humber J.M. The development of this electrocoagulation technology is being done by Corp., USA. Electrochemical treatment is a technique in which electricity is passed through water in the presence of sacrificial electrodes (Chen, 2004). Although it has been around for a while, it hasn't really been used much for wastewater treatment. For the treatment of industrial wastewater containing metals, this technology has seen an increase in use in South America and Europe over the past ten years (Mollah et al., 2001). Theory of electrocoagulation involves a conglomeration of many chemical and physical phenomena. The ions are produced *in-situ* and it involves following three stages. (i) Electrolytic oxidation of sacrificial electrodes to form coagulants, (ii) Destabilization of colloidal contaminants including particulate suspension and breaking of emulsion, and (iii) Aggregation of destabilized phases to form flocs (Koren and Syversen, 1995; Mollah et al., 2004) (Syversen, 1995) (Mollah et al., 2004). Aluminum and iron electrodes are common electrodes for *in-situ* generation of coagulating agents. The external power supply provides the potential difference across the electrodes. The anode acts as dissolution of respective ions and called as sacrificial electrode.

Electrocoagulation using iron pair of electrodes has following reaction equations (1.1-1.2):

Anodic reactions:



Cathodic reaction:

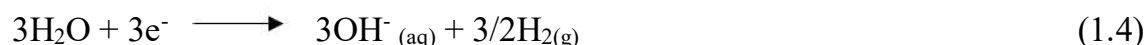


The electrocoagulation using aluminium electrodes produces aluminium cations to form hydroxylated species. The pollutants present in aqueous species are destabilized and then adsorbed on the aluminium hydroxide flocs. The reactions involved are shown in equation (1.3-1.5):

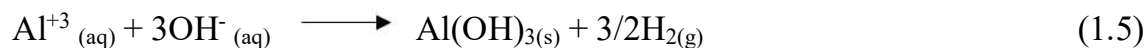
Anodic reaction:



Cathodic reaction:



Overall reaction in bulk solution is:



Metal removal through electrocoagulation has specific advantages in terms of high metal removal efficiency, instant start-up, compact treatment facility, less sludge formation and ease of operation (Chen, 2004; Mollah et al., 2001, 2004). Electrocoagulation has been successfully used for the removal of

metals like arsenic (Kumar et al. 2004) (Gomes et al., 2007) (Hunsom et al., 2005) (Parga et al., 2005) (Vasudevan et al., 2012a), boron (Vasudevan et al., 2010; Yilmaz et al., 2005, 2007b, 2007a, 2008), copper, lead and cadmium (Escobar et al. 2006), iron (Ghosh et al., 2008; Vasudevan et al., 2009), chromium (Aber et al., 2009; Golder et al., 2007; Heidmann & Calmano, 2008b, 2008a, 2010; Ölmez, 2009; Reddithota et al., 2007; Thella et al., 2008; Vasudevan et al., 2011, 2012b), and for the treatment of cutting oil emulsions and metal plating effluents (Bensadok et al., 2008; Kabdaşlı et al., 2009; Tir & Moulai-Mostefa, 2009). In this technique, passing of electrical energy into the wastewater through sacrificial metal electrodes generates electrons at the anode, which causes the reduction of the Cr(VI) into Cr(III) (Chen, 2004). The metal ions thus formed at the anode bring about coagulation and flocculation of the colloidal particles in the wastewater through neutralization of the surface charges, and the metal

hydroxide precipitates thus formed are subsequently removed. According to Heidmann and Calmano (Heidmann & Calmano, 2008a), at higher currents, Cr(VI) is reduced directly at the cathode and precipitated as Cr(OH)_3 . Electrocoagulation produces lesser amounts of hazardous sludge, and can be used under compact treatment facilities (Chen, 2004; Mollah et al., 2001, 2004).

CHAPTER - 2

LITERATURE REVIEW

2.1 Introduction:

In recent decades, urbanization has increased in speed and scale. Today, fifty five percent of the earth's population lives in urban areas, and is predicted to grow to sixty eight percent by 2050. Forecast indicate that urbanization, the incremental change in human population's residence from rural to city areas, combined with the world population growth, could add another two and half billion people to cities by 2050, according to a recent United Nations report set up by the Population Department of the UN Department of Economic and Social Affairs (UNDESA). By 2050, it is expected that India will have additional four hundred and sixteen million urban dwellers.

The Municipal solid waste (MSW) management is a considerable challenge today. Due to several environmental, economic, social and legislative constraints, the recognition of the most suitable disposal method for MSW is a significant issue in almost all developed countries (Adani et al., 2000). The common forms of solid waste handling include landfill, refuse composting, incineration and hydrogenation. According to the Environment Protection Agency (EPA), landfills are engineered area where waste is deposited in the land. Sanitary landfilling has become the most common way of eliminating MSW in most countries. Yet, in low lying areas MSW is disposed of without taking care or operational controls. While landfilling is viewed as the most cost-effective way of the waste disposal process, defective management operations are the main causes of environmental pollution, particularly in developing countries such as India (Wanwari et al., 2020).

The amount of MSW produced depends on different elements such as eating habits, living standards, level of commercial activities and seasons. The percentage constitution of solid waste varies between regions because it is affected by various factors such as community attitudes, location, frequency of collection, population characteristics, legal systems, socioeconomic and lifestyle as mentioned in (Sharholly et al., 2008).

Under Municipal Solid Waste Management & Handling Rule, 2000, Government of India, garbage is classified as municipal solid waste including residential and commercial solid wastes produced in the municipality or in notified regions either in solid or semi-solid state eliminating hazardous wastes from industries but adding treated biomedical wastes. The MSW includes household waste, sanitation residue, demolition and construction debris and waste from streets.

The large number of urban solid waste landfills and the many hazardous materials which they contain can pose serious environmental hazards to soil and groundwater, as landfills can release a wide variety of chemical compounds due to the waste degradation throughout the whole life cycle. Inside the landfill, the biodegradable portion of household waste is biodegraded to create the liquid leachate and landfill gas. Leachate contamination risk depends on the degree to which the pollutant barrier integrity is maintained and on the management efficiency of leachate.

2.1.1 Municipal Solid Waste Generation & Management Practices in India:

India, the world's second highest populated country after China with population of 1.21 billion (census 2011) already containing 17.5% of the world's population, is a land of physical, climatic, geographic, ecological, social, cultural and linguistic diversity. The annual rate of growth of urban population in India is 3.35% (Census of India, 2011). The proportion of population living in urban areas has increased from 17.35% in 1951 to 31.2% in 2011 (Census, 2011). High rate of population growth, declining opportunities in the rural areas and shift from stagnant and low paying agriculture sector to more paying urban occupations, largely contribute to urbanization. It is interesting to note that Currently 1 out of every 3 person is living in urban area and it is projected that as much as 50% of India's population will live in cities in next 10 years (Khurshid & Sethuraman, 2011). No doubt, India has achieved multifaceted socio-economic progress during last 64 years of its independence. However, in spite of heavy expenditure by Civic bodies, Management of Municipal Solid Wastes (MSW) continues to remain one of the most neglected areas of urban development in India. Piles of garbage and wastes of all kinds littered everywhere have become common sight in urban life. For most of urban local bodies in India solid waste is a major concern that has reached alarming proportions requiring management initiatives on a war-footing. Though solid waste management is one of the basic essential services to be provided by municipal authorities in

India, the present scenario provides rather a clumsy picture in terms of service delivery as evidenced by absence of adequate overall waste management mechanism. Present level of service in many urban areas is so low that there is a threat to the public health in particular and environmental quality in general (Supreme Court Committee Report 1999).

2.1.2 Classification of Countries Based on Population:

Details of tier-wise classification of cities based on population are as follows_

(i) Classification of cities (tier-wise):

Table-1: Classification of cities (tier-wise)

Population classification	Population	No. of Cities/Towns (as per Census 2011)
Tier 1	1,00,000 and above	485
Tier 2	50,000 to 99,999	551
Tier 3	20,000 to 49,999	1313
Tier 4	10,000 to 19,999	1087
Tier 5	5,000 to 9,999	473
Tier 6	Less than 5000	132

(ii) Population-group wise classification:

Table-2: Population-group wise classification

Population classification	Population
Rural centre	up to 9,999
Semi-urban centre	10,000 to 99,999
Urban centre	100,000 to 999,999
Metropolitan centre	1,000,000 and above

2.1.3 Present Solid Waste Generation in India for Different Cities:

The generated amount of MSW also depends on living standards, extent, type of commercial activity, eating habits, geographical, and climate conditions. Migrants in search of opportunities worsen the situation. As per the recent data from MNRE Report, India is generating exponentially about 145 million tonnes of waste per year and further expected to reach approximately 260 to 300 million tonnes per day in the year 2047. As per the CPCB, India data, about 117,644 MT collected, and around 49,401 MT is only treated (Central

Pollution Control Board, 2016) (Ahluwalia then Patel, 2018). CPCB together with the assistance of NEERI has conducted land survey over waste management within 59 cities (35 metro cities yet 24 government Capitals). Quantities and waste generation rate in 59 cities is as under_

Table-3: Present Solid Waste Generation in India for Different Cities

S. No	Name of City	Waste Quantity (TPD)	Waste Generation Rate (kg/c/day)
1	Kavaratti	3	0.30
2	Gangtok	13	0.44
3	Itanagar	12	0.34
4	Daman	15	0.42
5	Silvassa	16	0.32
6	Panjim	32	0.54
7	Kohima	13	0.17
8	Port Blair	76	0.76
9	Shillong	45	0.34
10	Shimla	39	0.27
11	Agartala	77	0.40
12	Gandhinagar	44	0.22
13	Dhanbad	77	0.39
14	Pondicherry	130	0.59
15	Imphal	43	0.19
16	Aizawl	57	0.25
17	Jammu	215	0.58
18	Dehradun	131	0.31
19	Asansol	207	0.44
20	Kochi	400	0.67
21	Raipur	184	0.30
22	Bhubaneswar	234	0.36

23	Thiruvananthapuram	171	0.23
24	Chandigarh	326	0.40
25	Guwahati	166	0.20
26	Ranchi	208	0.25
27	Vijayawada	374	0.44
28	Srinagar	428	0.48
29	Madurai	275	0.30
30	Coimbatore	530	0.57
31	Jabalpur	216	0.23
32	Amritsar	438	0.45
33	Rajkot	207	0.21
34	Allahabad	509	0.52
35	Vishakhapatnam	584	0.59
36	Faridabad	448	0.42
37	Meerut	490	0.46
38	Nashik	200	0.19
39	Varanasi	425	0.39
40	Jamshedpur	338	0.31
41	Agra	654	0.51
42	Vadodara	357	0.27
43	Patna	511	0.37
44	Ludhiana	735	0.53
45	Bhopal	574	0.40
46	Indore	557	0.38
47	Nagpur	504	0.25
48	Lucknow	475	0.22
49	Jaipur	904	0.39
50	Surat	1000	0.41
51	Pune	1175	0.46

52	Kanpur	1100	0.43
53	Ahmedabad	1302	0.37
54	Hyderabad	2187	0.57
55	Bangalore	1669	0.39
56	Chennai	3036	0.62
57	Kolkata	2653	0.58
58	Delhi	5922	0.57
59	Greater Mumbai	5320	0.45

2.1.4 Characteristics of Solid Waste for Different Cities:

The composition and the quantity of MSW generated form the basis on which the management system needs to be planned, designed and operated. In India, MSW differs greatly with regard to the composition and hazardous nature, when compared to MSW in the western countries (Gupta et al., 1998; Shannigrahi et al., 1997; Jalan and Srivastava, 1995). The composition of MSW at generation sources and collection points was determined on a wet weight basis and it consists mainly of a large organic fraction (40–60%), ash and fine earth (30 40%), paper (3–6%) and plastic, glass and metals (each less than 1%). The C/N ratio ranges between 20 and 30, and the lower calorific value ranges between 800 and 1000 kcal/kg. The chemical characteristics of MSW in different Indian cities are presented in Table-4 and the physical characteristics of MSW in metrocities are presented in Table-5. It has been noticed that the physical and chemical_characteristics of MSW change with population density, as shown in Table-6 and Table-7 (Garg and Prasad, 2003; CPCB, 2000; Bhide and Shekdar, 1998).

Table-4: Characteristics of Solid Waste for Different Cities

S. No	Name of City	Compostable (%)	Recyclables (%)	C/N Ratio	HCV (Kcal/Kg)	Moisture (%)
1	Kavaratti	46.01	27.20	18.04	2242	25
2	Gangtok	46.52	16.48	25.61	1234	44
3	Itanagar	52.02	20.57	17.68	3414	50
4	Daman	29.60	22.02	22.34	2588	53
5	Silvassa	71.67	13.97	35.24	1281	42
6	Panjim	61.75	17.44	23.77	2211	47
7	Kohima	57.48	22.67	30.87	2844	65
8	Port Blair	48.25	27.66	35.88	1474	63
9	Shillong	62.54	17.27	28.86	2736	63
10	Shimla	43.02	36.64	23.76	2572	60

11	Agartala	58.57	13.68	30.02	2427	60
12	Gandhinagar	34.30	13.20	36.05	698	24
13	Dhanbad	46.93	16.16	18.22	591	50
14	Pondicherry	49.96	24.29	36.86	1846	54
15	Imphal	60.00	18.51	22.34	3766	40
16	Aizawl	54.24	20.97	27.45	3766	43
17	Jammu	51.51	21.08	26.79	1782	40
18	Dehradun	51.37	19.58	25.90	2445	60
19	Asansol	50.33	14.21	14.08	1156	54
20	Kochi	57.34	19.36	18.22	591	50
21	Raipur	51.40	16.31	223.50	1273	29
22	Bhubaneswar	49.81	12.69	20.57	742	59
23	Thiruvananthapuram	72.96	14.36	35.19	2378	60
24	Chandigarh	57.18	10.91	20.52	1408	64
25	Guwahati	53.69	23.28	17.71	1519	61
26	Ranchi	51.49	9.86	20.23	1060	49
27	Vijayawada	59.43	17.40	33.90	1910	46
28	Srinagar	61.77	17.76	22.46	1264	61
29	Madurai	55.32	17.25	32.69	1813	46
30	Coimbatore	50.06	15.52	45.83	2381	54
31	Jabalpur	58.07	16.61	28.22	2051	35
32	Amritsar	65.02	13.94	30.69	1836	61
33	Rajkot	41.50	11.20	52.56	687	17
34	Allahabad	35.49	19.22	19.00	1180	18

35	Visakhapatnam	45.96	24.20	41.70	1602	53
36	Faridabad	42.06	23.31	18.58	1319	34
37	Meerut	54.54	10.96	19.24	1089	32
38	Nasik	39.52	25.11	37.20	2762	62
39	Varanasi	45.18	17.23	19.40	804	44
40	Jamshedpur	43.36	15.69	19.69	1009	48
41	Agra	46.38	15.79	21.56	520	28
42	Vadodara	47.43	14.50	40.34	1781	25
43	Patna	51.96	12.57	18.62	819	36
44	Ludhiana	49.80	19.32	52.17	2559	65
45	Bhopal	52.44	22.33	21.58	1421	43
46	Indore	48.97	12.57	29.30	1437	31
47	Nagpur	47.41	15.53	26.37	2632	41
48	Lucknow	47.41	15.53	21.41	1557	60
49	Jaipur	45.50	12.10	43.29	834	21
50	Surat	56.87	11.21	42.16	990	51
51	Pune	62.44	16.66	35.54	2531	63
52	Kanpur	47.52	11.93	27.64	1571	46
53	Ahmedabad	40.81	11.65	29.64	1180	32
54	Hyderabad	54.20	21.60	25.90	1969	46
55	Bangalore	51.84	22.43	35.12	2386	55
56	Chennai	41.34	16.34	29.25	2594	47
57	Kolkata	50.56	11.48	31.81	1201	46
58	Delhi	54.42	15.52	34.87	1802	49
59	Greater Mumbai	62.44	16.66	39.04	1786	54

Table-5: Different types of waste materials in different Indian cities

Name of City	Paper	Textile	Leather	Plastic	Metals	Glass	Ash, fine earth and others	Com-post-able matter
Ahmedabad	6	1		3			50	40
Bangalore	8	5		6	3	6	27	45
Bhopal	10	5	2	2		1	35	45
Mumbai	10	3.6	0.2	2		0.2	44	40
Kolkata	10	3	1	8		3	35	40
Coimbatore	5	9		1			50	35
Delhi	6.6	4	0.6	1.5	2.5	1.2	51.5	31.8
Hyderabad	7	1.7		1.3			50	40
Indore	5	2		1			49	43
Jaipur	6	2		1		2	47	42
Kanpur	5	1	5	1.5			52.5	40
Kochi	4.9			1.1			36	58
Lucknow	4	2		4	1		49	40
Ludhiana	3	5		3			30	40
Madras	10	5	5	3			33	44
Madurai	5	1		3			46	45
Nagpur	4.5	7	1.9	1.25	0.35	1.2	53.4	30
Patna	4	5	2	6	1	2	35	45
Pune	5			5		10	15	55
Surat	4	5		3		3	45	40
Vadodara	4			7			49	40
Varanasi	3	4		10			35	48
Visakhapatnam	3	2		5		5	50	35

In general, the Physical characteristics of MSW in Indian cities population wise are as follows_

All values are in percentage and are calculated on weight basis.

Table-6: Physical characteristics of MSW in Indian cities population wise

Population Range (in million)	Paper	Rubber, Leather & Synthetics	Glass	Metal	Compostable Matter	Inert Material
0.1-0.5	2.91	0.78	0.56	0.33	44.57	43.59
0.5-1.0	2.95	0.73	0.56	0.32	40.04	48.38
1.0-2.0	4.71	0.71	0.46	0.49	38.95	44.73
2.0-5.0	3.18	0.48	0.48	0.59	56.57	49.07
5.0 and above	6.43	0.28	0.94	0.8	30.84	53.90

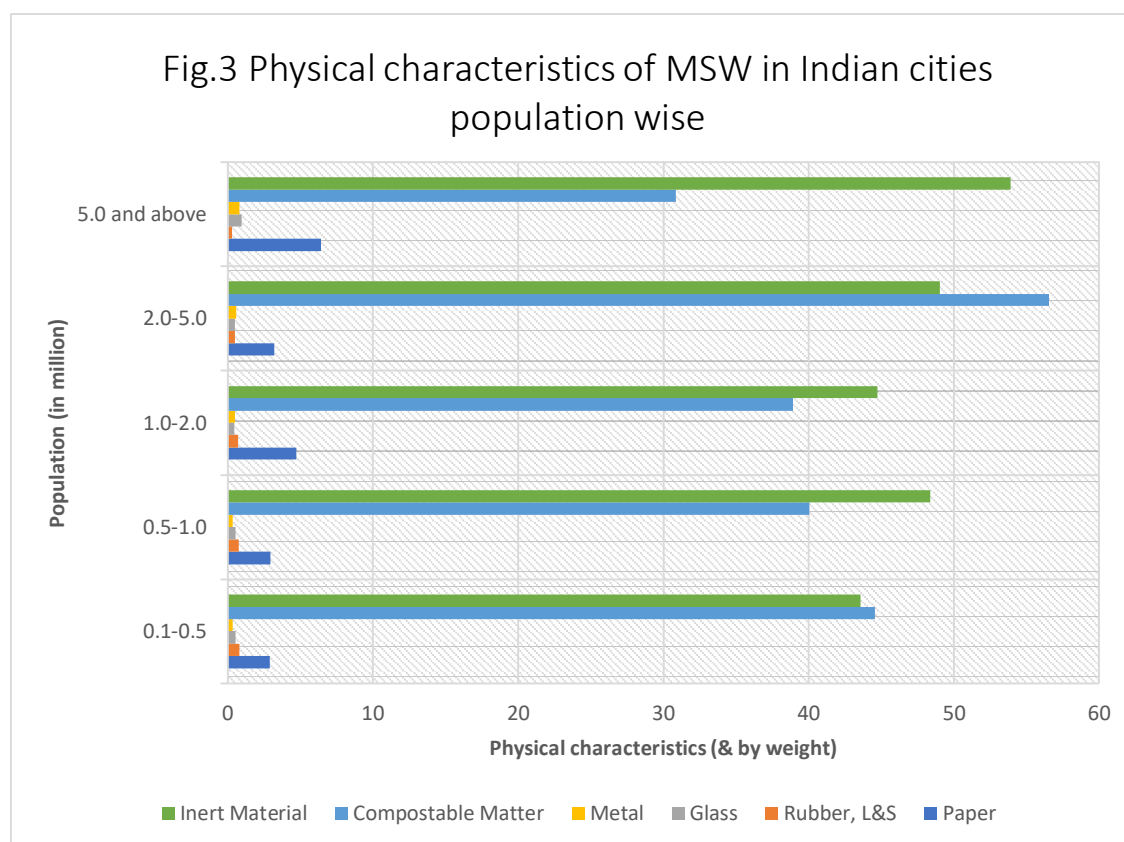


Fig.3 Physical characteristics of MSW in Indian cities population wise

In general, the Chemical characteristics of MSW in Indian cities population wise are as follows_

All values are in percentage and are calculated on wet weight basis.

Table-7: Chemical characteristics of MSW in Indian cities population wise

Population Range (in million)	Moisture (in %)	Organic matter (in %)	Nitrogen as total nitrogen (in %)	Phosphorous as P_2O_5 (in %)	Potassium as K_2O (in %)	C/N ratio	Calorific value (in kcal/kg)
0.1-0.5	25.81	37.09	0.71	0.63	0.83	30.94	1009.89
0.5-1.0	19.52	25.14	0.66	0.56	0.69	21.13	900.61
1.0-2.0	26.98	26.89	0.64	0.82	0.72	23.68	980.05
2.0-5.0	21.03	25.60	0.56	0.69	0.78	22.45	907.18
5.0 and above	38.72	39.07	0.56	0.62	0.52	30.11	800.70

Fig 4: Chemical characteristics of MSW in Indian cities population wise

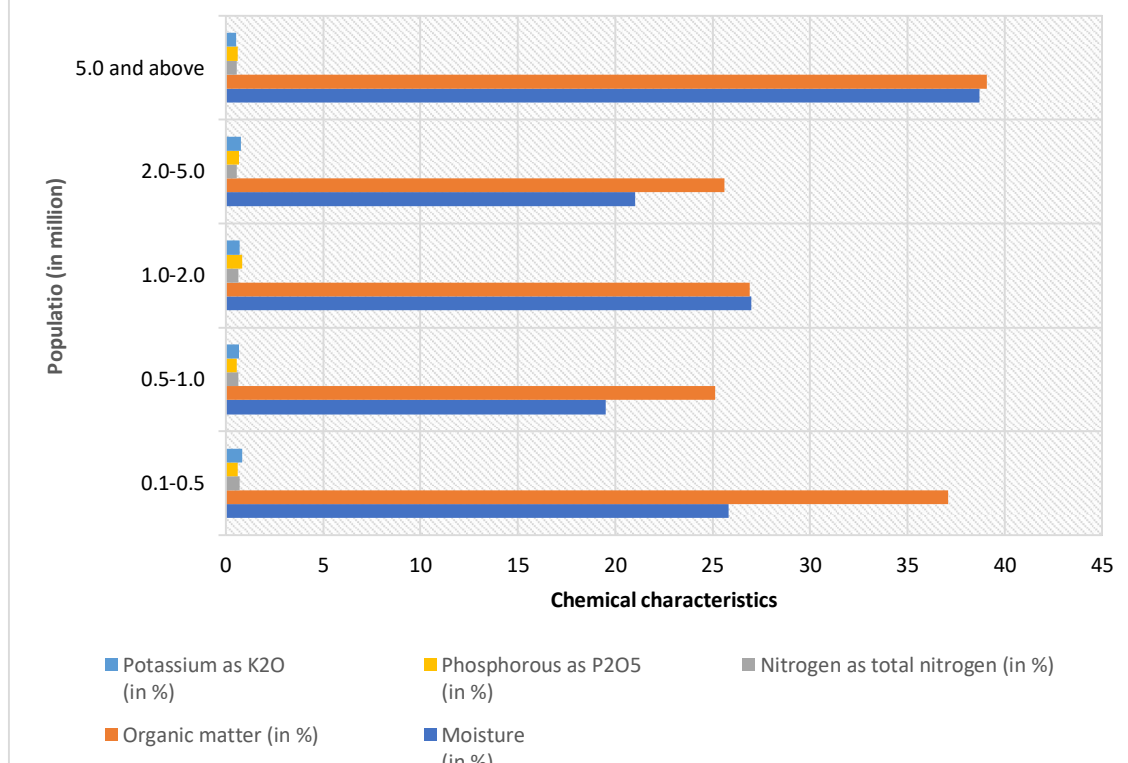


Fig 4: Chemical characteristics of MSW in Indian cities population wise

2.2 Phases of Degradation of Landfill:

The environment in a landfill depends on different physical, chemical and biological processes. Despite large differences in waste composition, water content and management, it is possible to sort out generalizations and identify a group of particular landfill processes. In a landfill with significant amounts of organic matter, biodegradation of organic substance is the controlling process which governs the landfill biogeochemistry. The characteristics of leachate can be described in terms of various chemicals that can be classified as inorganic matters, organic matters and xenobiotic organic compounds. Within landfill many complex reactions occur sequentially which can be categorized as physical, chemical and biological process. The transformation of organic matter moves through a number of different phases that are relatively same among MSW landfills. They are:

I. Aerobic stage

II. Acidic stage

III. Initial methanogenic stage

IV. Stable methanogenic stage

V. Secondary aerobic stage

When deposited, the waste contains oxygen and the landfill is in stage-I, Microorganisms that thrive in oxygen-rich conditions start to degrade the organic matter. The oxygen is depleted relatively quickly, and when new waste is placed on top or the landfill is covered, no additional oxygen can reach the waste. The aerobic stage typically lasts for only a few hours or days and aerobic degradation of organic matter leads to a high rate of CO₂ formation. When the oxygen is depleted anaerobic microorganisms take over. Initially hydrolytic micro organisms are the most active, degrading large organic molecules into mono-saccharides, alcohols, carboxylic acids, etc. This leads to a build-up of volatile fatty acids causing the pH to decrease, sometimes down itowards ipH i5, and the landfill enters stage-II, the acid phase. The chemical oxygen demand (COD) and biological oxygen demand (BOD) in the leachate are high and in between 0.7 and close to 1, the BOD/COD ratio is also high at this stage. The acid leachate enhances dissolution and leads to high concentrations of pollutants, such as heavy metals. Stage-II typically lasts for months to years and ends when microorganisms degrading the intermediate products into CO₂, H₂ and acetate become more active and the pH increases. Methanogenic

bacteria that are sensitive to low pH thrive and produce methane from these molecules. During stage-III, the initial methanogenic phase, most leachate concentrations decrease. Methane production increases until it reaches a relatively stable value.

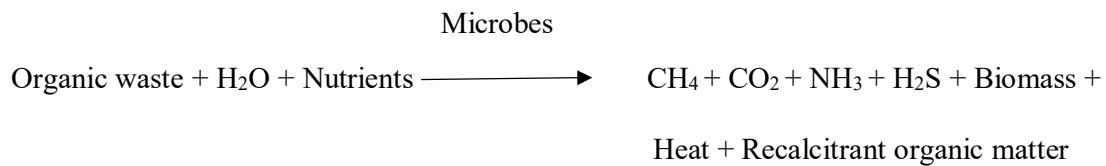
During stage-IV the landfill enters a stable state of methanogenesis with only gradual changes in gas and leachate composition. Landfill gas typically consists of 50–70 % methane and 30–50 % carbon dioxide. The pH during this stage is neutral or slightly alkaline. Ammonium is formed due to the degradation of organic matter and typically accumulates in the leachate, as it has no biodegradation mechanism under methanogenic conditions. The rate of ammonium release is relatively low and thus ammonium is expected to be the most significant long-term pollutant at landfills. The BOD/COD ratio is also low; approaching 0.1, because the most degradable dissolved organic matter is consumed by the microbial population. However, concentrations of more stable organic molecules, such as humic and fulvic acids, increase. After years, or even decades or centuries, when the organic matter that is degradable by anaerobic microorganisms has been depleted, the formation of landfill gas will decrease and eventually, the gas pressure within the landfill will equal the atmospheric pressure. At that point oxygen begins to enter the landfill by diffusion and advection, marking the start of stage-V, the second aerobic phase. This phase has rarely been observed but predicted. Initially, the oxygen is consumed by microbes that oxidize the remaining organic matter and produce stable molecules such as humic substances. At this stage, CO₂ is formed by the oxidation, but as degradation ceases, the gas composition approaches that of the atmosphere. Depending on various factors such as the quality of the cover and the water content of the landfill, the time it takes for a landfill to become completely oxidized varies between hundreds and tens of thousands of years (Kjeldsen et al., 2002).

2.3 The Generation of Landfill Leachate:

Up to 95 percent of the world's solid waste generated is currently disposed of in landfills (Kurniawan et al., 2006; Bohdziewicz and Kwarciak, 2008). Given the many benefits of

landfills, the production of highly contaminated leachates is a major downside, with large variations in both chemical composition and volumetric flows. The leachate from landfills could be generated by two major causes. The first source is external water which goes into the landfill and produces leachate. Due to the biodegradation of organic waste, the second source is leachate generation from the landfill itself.

Organic and inorganic contaminants of landfill leachate are derived from the solid waste due to successive biological, chemical and physical processes. Biodegradable waste can be converted biologically to CH_4 and CO_2 . The pathway of conversion can be explained by the following equation:



Biodegradation in landfill leachate occurs in sequences of five continuous phases with its predominant products in both leachate and landfill gas as shown in Fig. 5

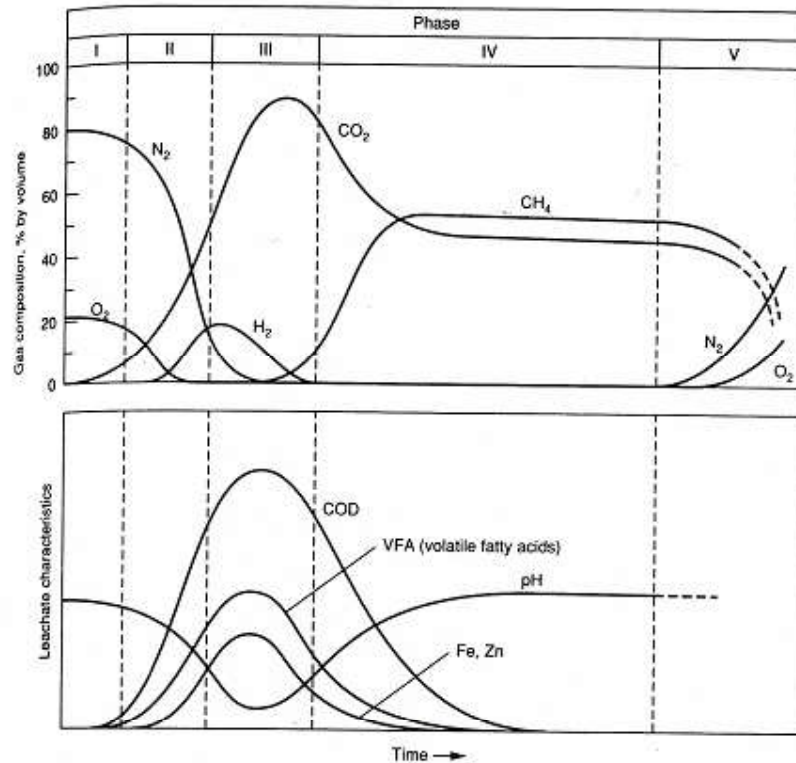


Fig. 5: Schematic diagram of Waste Stabilization in a Landfill (Tchobanoglous et al., 1993)

(a) Phase I- Initial adjustment phase (Lag phase) When the solid waste is buried into the landfill, it takes moisture and air concomitantly with it. Thus, dominant gases are still N_2 and O_2 . Biological decomposition occurs under aerobic condition that results in elevation of CO_2 concentration.

(b) Phase II- Transition phase The transition phase involves the shifting from aerobic to anaerobic environment. Basically, aerobic phase rapidly consumes the confined oxygen and infiltration water by microorganism respiration. Anaerobic conditions result in end products such as volatile fatty acids (VFA) and CO_2 . The pH of the leachate decreases due to the presence of VFA and CO_2 solution. The low pH mobilizes heavy metals from the waste into the leachate.

(c) Phase III- Acid phase Continuous solubilization (hydrolysis) of solid waste, followed by the microbial conversion of biodegradable organic content enhances acetogenic fermentation resulting in high BOD, COD and ammonia-nitrogen concentration. VFA are the main components of the organic matter released (Welander et al., 1997), besides the low pH solubilises heavy metals.

(d) Phase IV- Methane fermentation phase As a landfill matures, the methanogenic phase occurs. Methanogenic microorganisms develop in the waste, and the VFA are converted to biogas (CH_4 , CO_2). In this phase, the leachate composition represents the dynamic equilibrium between the two microbiological mechanisms with lower BOD and COD while the ammonia concentration remains high. Dissolved inorganic materials are continuously released. Occurrence of heavy metals in the leachate tends to decrease because of a high pH value that causes complexation, precipitation and transition to solid phase.

(e) Phase V- Maturation phase With landfill ageing, waste stabilization takes place. Most of the biodegradable compounds have been decomposed. As the VFA content of leachate decreases parallel to the BOD/COD ratio, the organic fraction in the leachate becomes dominated by recalcitrant compounds such as humic substances (Chian and DeWalle, 1976). Thus, gas production drops and leachate stays at a constant level. The atmospheric gases outside the landfill will permeate through the solid waste, resulting to noticeable amounts of N_2 and O_2 once again.

Lema et al. (1988) proposed an anaerobic degradation scheme for the organic material in a sanitary landfill (Fig. 6).

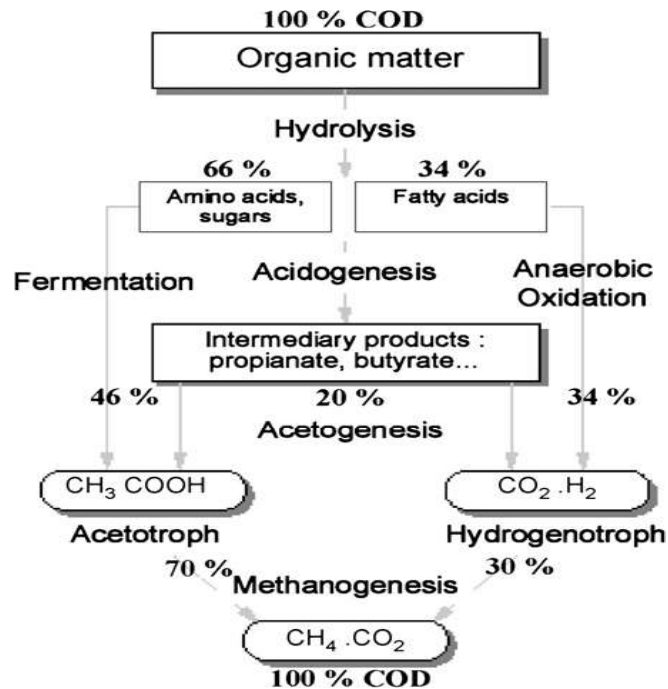


Fig. 6: COD Balance of the Organic Fraction During an Anaerobic Degradation in a Sanitary Landfill (Lema et al., 1988).

Typical data on the composition of leachate are reported in Table 8 for both new and mature landfills

Table 8: Typical Data on the Composition of Leachate from New and Mature Landfills (Tchobanoglous et al., 1993)

Constituent	Value, mg/L ^a		
	New Landfill (< 2 years)		Mature Landfill (> 10 years)
	Range ^b	Typical ^c	
BOD ₅ (5-day biochemical oxygen demand)	2000-30000	10000	100-200
TOC (total organic carbon)	1500-20000	6000	80-160
COD (chemical oxygen demand)	3000-60000	18000	100-500
Total suspended solids	200-2000	500	100-400
Organic nitrogen	10-800	200	80-120
Ammonia-nitrogen	10-800	200	20-40
Nitrate	5-40	25	5-10
Total phosphorus	5-100	30	5-10
Ortho phosphorus	4-80	20	4-8
Alkalinity as CaCO ₃	1000-10000	3000	200-1000
pH	4.5-7.5	6	6.6-7.5
Total hardness as CaCO ₃	300-10000	3500	200-500
Calcium	200-3000	1000	100-400

Magnesium	50-1500	250	50-200
Potassium	200-1000	300	50-400
Sodium	200-2500	500	100-200
Chloride	200-3000	500	100-400
Sulfate	50-1000	300	20-50
Total iron	50-1200	60	20-200

^a Except pH, which has no unit

^b Representative range of values. Higher maximum values have been reported in the literature for some of the constituents

^c Typical values for new landfills will vary with the metabolic state of the landfill

It is to be noted that the chemical composition of leachate will vary greatly depending on the age of landfill and the events preceding the time of sampling. For example, if a leachate sample is collected during the acid phase of decomposition (see Fig. 2.1), the pH value will be low and the concentration of BOD₅, TOC, COD, nutrients and heavy metals will be high. If, on the other hand, a leachate sample is collected during the methane fermentation phase (see Fig. 2.1), the pH will be in the range from 6.5-7.5, and the BOD₅, TOC, COD and nutrients concentration values will be significantly lower. Similarly, the concentrations of heavy metals will be lower because most metals are less soluble at neutral pH values. The pH of the leachate will depend not only on the concentration of the acids that are present but also on the partial pressure of the CO₂ in the landfill gas that is in contact with the leachate. The biodegradability of the leachate will vary with time. Changes in the biodegradability of the leachate can be monitored by checking the BOD₅/COD ratio. Initially, the ratios will be in the range of 0.5 or greater. Ratios in the range of 0.4 to 0.6 are taken as an indication that the organic matter in the leachate is readily biodegradable. In mature landfills, the BOD₅/COD ratio is often in the range of 0.05 to 0.2. The ratio drops because leachate from mature landfills typically contains humic and fulvic acids, which are not readily biodegradable (Tchobanoglous et al., 1993).

2.3.1 External Water:

Majority of the leachate is generated by direct penetration of water i.e. rainwater and snow melt penetration into the waste. Such liquids can penetrate the landfill for several years and will contact with various substances within the landfill during this period. The water is leaching and

dissolving different components until it contains lot of chlorinated organic matter and other substances. The leachate can affect nearby groundwater and surface water. The rainfall intensity, quantity, frequency and duration affect the amount of leachate produced. The humidity in the atmosphere exerts a heavy influence on the production of leachate. The waste is infiltrated by the surface water as well as by the groundwater. The infiltration of surface water is dependent upon the site type. If the landfill is constructed below a sloping field, that has surface water, from the direction of topography it will fall down on the landfill. If the landfill bottom is below the water table, the groundwater can otherwise infiltrate into the waste. The quantity of leachate is dependent on groundwater and waste interface situations.

2.3.2 The Wastewater Generated Within the Landfill:

Water fractions in the waste contribute to the leachate, too. During the waste placement the waste moisture is created by waste. Untreated solid waste in the landfill will generate leachate which is a major source of organic substances within the waste. By anaerobic decay, this becomes a highly contaminated liquid inside the landfill. The damp waste contains excess moisture from the atmosphere or rainwater which includes the adsorbed moisture and its own moisture. The damp waste gets compacted and organically decomposed in the landfill by biological, physical and chemical processes.

The quantity of leachates depends on the infiltration of rainwater through wastes, the inherent water content of wastes, biochemical processes in the cells of waste and its level of compaction of the landfill as mentioned in Abbas et al., (2009). The leachate generation also depends on other factors such as waste content, landfilling techniques, landfill age and humidity, as mentioned in (Bikash Adhikari & Sanjay Nath Khanal, 2015).

2.4 Landfill Leachate Characteristics:

Concentrating on the most usual type of landfill receiving a combination of residential, commercial and blended industrial waste, landfill leachate may be described as a water based solution with four categories of pollutants namely dissolved organic matters, macro inorganic compounds, heavy metals and xenobiotic organic compounds originating from domestic

residue and chemical residues present in low concentrations as mentioned in (Christensen et al., 2001). The leachate has an orange brown or dark brown hue. The smell associated with leachate is foul-smell, mainly due to the presence of organic acids, which arises from the high concentration of decomposed organic substances. Depending on the increasing landfillage, the nature of precipitation and the quantity and quality of the waste, the dark colour and malodour will slowly disappear or decrease as mentioned in Li Rong, (2009).

2.4.1 Dissolved Organic Matters:

The component of dissolved organic matter in leachates such as BOD (Biochemical Oxygen Demand), TOC (Total Organic Carbon) and COD (Chemical Oxygen Demand) is defined using various bulk parameters. Acids, alcohols, aldehydes and other dissolved organic matter commonly quantified as BOD, COD, Dissolved Organic Carbon (DOC), other volatile fatty acid and refractory compounds such as humic-like and fulvic-like compounds. A large number of degradation products of organic matter ranging from small volatile acids to refractory humic and fulvic-like compounds are dissolved in leachate as mentioned in Dewalle and Chian, (1977) (*MANAGEMENT of GAS and LEACHATE i LANDFILLS*, 1977).

Dissolved organic matter may affect the composition of leachate compared to the other constituents by the complex properties of the compounds of high-molecular-weight (HMW) in it. At the most general level, a lower BOD/COD ratio indicates leachate with lesser concentrations of volatile fatty acids and comparatively greater levels of fulvic and humic-like components.

2.4.2 Inorganic Macro Compounds:

These include ammonium (NH_4^+), sulphate (SO_4^{2-}), chloride (Cl^-), calcium (Ca^{2+}), sodium (Na^+), magnesium (Mg^{2+}), potassium (K^+), hydrogen carbonate (HCO_3^-), iron (Fe^{2+}) and manganese (Mn^{2+}). The concentration in the leachate of some inorganic macro compounds depends on the landfill stabilization. Due to a lower dissolved organic matter content and higher pH, the cations like magnesium, calcium, iron and manganese are lesser in methanogenic phase of leachate. In the methanogenic phase, sulphate concentrations are also low due to the

reduction of sulphate to sulphide by microbes. The effects of complex formation, sorption and precipitation are small for some macro compounds such as chloride, sodium and potassium. Diminishing tendency in concentration of contaminants with time could be due to washing out by the process of leaching as stated in Ehrig, (1989). Many research works report the concentration of ammonia nitrogen in leachate ranges from 500 to 2000 mg/l. Ammonia is extracted from the waste primarily by decomposition of proteins. Leaching is the single mechanism by which ammonia concentration can reduce during waste decomposition.

2.4.3 Heavy Metals:

The sum of heavy metals is related to how much industrial waste will be present in landfills. Household waste includes only very small quantity of heavy metals. If there is mixed filling in the landfill, then the industrial refuse is the primary source for the content of heavy metals. This include lead (Pb^{2+}), nickel (Ni^{2+}), copper (Cu^{2+}), cadmium (Cd^{2+}), chromium (Cr^{3+}) and zinc (Zn^{2+}). There is a significant variance in the concentration of heavy metals recorded from various landfills. Average concentrations of metal is, however, very small. Several researchers reported low metal concentrations from full-scale landfills, test cells and laboratory tests. The heavy metals in landfill leachate from MSW are therefore not a prime concern as mentioned in (Kjeldsen et al., 2002) and (Esakku et al., 2005).

2.4.4 Xenobiotic Organic Compounds:

These compounds include hydrocarbon aromatics, chlorinated aliphatics, phenols, pesticides and plasticizers. The mono aromatic hydrocarbons (benzene, ethylbenzene, toluene and xylenes) and halogenated hydrocarbons such as trichloroethylene and tetrachloroethylene are the most frequently occurring Xenobiotic organic compounds (XOCs). Such contaminants are found at the highest levels. In spite of the very complicated structure of landfill leachate, these non-polar organic substances are relatively easy to examine. They have significant negative effects on the aquatic environment. Information on ionic and polar organic contaminants is scarce but many researches are going on as stated by (Kjeldsen et al., 2002).

2.5 Factors Influencing the Leachate Characteristics:

Leachate consists of both organic and inorganic compounds. The chemical constitutions of leachates vary due to various factors like waste composition, landfill age, seasonal weather/climate variability at the deposited site and landfiling process (Raghab et al., 2013).

2.5.1 Composition of Waste:

The waste composition has a great impact on landfill leachate water quality. An element that determines the chemical constitution of the landfill leachate is the kind of waste collected on a landfill. Waste from various sources contains different of these organic and inorganic materials, e.g. municipal, industrial etc. Organic waste products in the waste are mainly waste from kitchen while inorganic components are items such as plastics, metals, glass etc. The leachate from landfill includes COD and BOD that are derived from the organic compounds of domestic waste. The proportion of domestic biological waste, which may be high or low in the waste, may directly affect the COD and BOD concentration as mentioned in Madu Jude, (2008). The residues of dust and soil have the purpose of filtration and adsorption of organic matter present in leachate. Hence the concentration of leachate could also be influenced by the contents of dust and soil within the waste.

2.5.2 Age of Landfill:

Leachate generation in sanitary landfiling is a complex mix of physical, chemical and biological processes whereby waste age has a major impact on the landfill performance that produces leachate. Leachate quality is significantly influenced by the age or length of time after filling waste. Results show that the content of leachate is well associated with the age of waste. This is largely due to the degradation of both organic and inorganic components in the waste by microbes which experience different exposure of acetogenic and methanogenic phases as mentioned in (Lee et al., 2010).

The numerous landfill sites remain with various stages. Leachate can be classified into various forms depending on the landfill age. Leaching from landfills of less than 5 years old is commonly considered as young leachates. It's said that the young leachates are in the acidogenic stage. In this process, landfills carry large quantities of biodegradable organic matter that is usually considered to undergo fermentation anaerobically facilitated by landfill water content resulting in the generation of volatile fatty acids (VFA). As the landfill continues to take place after 5 years, the methanogenic stage begins. Methanogenic microbes grow in the waste, turning the VFAs to CH₄ and CO₂ and the organic portion of the leachate becomes primarily non-biodegradable (refractory) compounds like humic substances. Leachates from landfills between the ages of five and ten are leachates of medium or intermediate age because they may be having both acidogenic and methanogenic characteristics whereas those from landfills more than ten years are considered to be stable or old. Table 9 reports the leachate composition at various age levels.

Table 9: Composition of leachate at different age range

Leachate type	Young	Intermediate	Old
Landfill age (yr)	<5	5-10	>10
pH	<6.5	7	>7.5
BOD/COD	>0.3	0.1- 0.3	<0.1
TOC/COD	0.3	-	0.4
Organic matter	70-90% VFA	20- 30% VFA	High Molecular Wt.
Nitrogen	100-2000 mg/l TKN		
Metals g/l	2	<2	<2

The potential of hydrogen ion (pH) is a measure of a solution's acid or basic content. Table 9 reports that the pH of the young leachate is in acid range while the old one is in basic range.

COD is a measure of the contaminants concentration in leachate that can be oxidized by a chemical oxidizing agent. BOD₅/COD is the relationship among BOD and COD which shows the level of biodegradability of the leachate. As mentioned in (Renou et al., 2008) if BOD/COD ratio is higher than 0.3, it is young leachate. When the ratio is in the range 0.1 - 0.3, it is medium age leachate and if the ratio is lower than 0.1 then it is old leachate. Volatile fatty acids (VFA) are the results of the degradation of organic matter anaerobically. From table 2.2 it is seen that they account for approximately 80 percent of the content of young leachate while old leachates mainly contain organic content of fulvic and humic substances.

2.5.3 Climate /Seasonal Weather Variation:

The variability of climate/seasonal weather affects the leachate production in terms of both quality and quantity. The moisture content of landfills increases during the rainy season. Since the amount of moisture improves the anaerobic degradation of organic matter, in the rainy season biodegradation would be quicker and more compared to the hot season. Thus, the humid and hot climate which aids microbial activities produces more leachate and biodegrades more organic compound than the dry climate. Evaporation also adversely affects moisture content in the dry season which decreases the leachate generation and activities of microbes.

2.5.4 Method of Landfilling:

The landfilling process affects the leachate quantity and quality. The landfill flood control system is useful for aiding the surface water discharge. Additionally, it is better to use the clay layer at the bottom of the landfill to regulate the flowing ground water and surface water into the landfill. Generally, the production is greater when the waste is less compact since compaction reduces the rate of filtration (Bhalla et al., 2012).

2.6 Environmental Issues Pertaining to Landfills:

Conventional landfills contain a combination of various types of waste, including residential, industrial, construction and demolition waste, with diverse organic and inorganic substances

and a wide variety of physical and chemical characteristics. MSW usually contains a significant fraction of biodegradable material, and thus major conventional landfills contain significant quantity of organic matter. Gas and leachate are the two primary emission pathways for pollutants from landfills. Degradation of organic compounds forms landfill gas. Its main components are CO₂ and CH₄, both of which are greenhouse gases, with CH₄ on a one hundred year time scale being over 20 times more potent than CO₂. A combination of physical, chemical and microbial processes carries contaminants from the waste substance to the leachate, making it a complex liquid.

Waste was disposed off in dumps that did not have liner or pollution control methods. Groundwater and surface water contamination is considered as a serious environmental effect of landfills for several years. Organic substances may cause depletion of oxygen in the receiver. The high levels of nutrients and other leachate and wastewater products, such as xenobiotic organic compounds like phenols and pesticides, pose a risk to the environment and human health. Inorganic macro components comprise nutrients that induce eutrophication, and many freshwater organisms are toxic to many inorganic compounds mostly seen in landfill leachate, such as ammonia and chloride (Kjeldsen et al., 2002). Leachate released without sufficient treatment will lead to eutrophication of water bodies causing algal blooms, oxygen depletion, habitat loss and increased turbidity and water toxicity. Leachate needs an efficient treatment before final discharge to reduce the higher contaminants concentration to an acceptable range (Hilles et al., 2016b).

2.7 Leachate Discharge Standards:

The landfill discharge standards should be reviewed in various aspects: the continuous impact on the environment, the leachate quality, economic abilities and the practical situation of the landfill. Standards aim to furnish executive criteria to be followed. The standard differs in different countries. Table 10 shows the limiting concentration of environmental pollutants for discharge in India (Environment Protection Rules, 1986).

Table 10: General standards for discharge of environmental pollutants as per The Environment (Protection) Rules, 1986, Govt. of India

Characteristic	Inland surface	Public sewer	Land disposal
SS mg/L	100	600	200
pH	5.5-9	5.5-9	5.5-9
NH ₃ -N mg/l	50	50	-
BOD mg/L	30	350	100
COD ,mg/L	250	-	-
Chloride mg/L	1000	1000	1000
Sulphate mg/l	1000	1000	1000
Sulphide mg/l	2	-	-
Phosphate mg/l	5	-	-
Bioassay Test	90% survival of fish after 96 hours in 100% effluent	90% survival of fish after 96 hours in 100% effluent	90% survival of fish after 96 hours in 100% Effluent

2.8 Landfill Leachate Treatment Methods:

There are numerous techniques for treating landfill leachates. The choice of apt treatment depends on several factors which correspond to the characteristics of each landfill. There are several things to consider such as quality and quantity of leachate, disposal standards, process residuals, geographical and climatic conditions, economic costs etc. Landfill leachate treatment methods can be generally categorized into physico-chemical and biological. Equalization tanks prior to treatment are sometimes needed due to the changes in the amount of leachate produced.

2.8.1 Physico-Chemical Method:

Physico-chemical methods are non-biological methods used for treating leachate. The method accomplishes treatment by oxidizing pollutants with chemicals after which separation processes are applied physically. This method is also coupled along side the biological process to enhance treatment efficiency. It is applied in the removal of recalcitrant substances (humic substances or undesirable compounds) like heavy metals, polychlorinated biphenyls (PCBs) and absorbable organic halogens (AOXs) or non-biodegradable substances from leachate. Before discharge of effluent, an additional refining step using physical-chemical treatments can be carried out on-site. Air stripping, coagulation/precipitation, Electro coagulation, Activated carbon adsorption, Flotation, Advance oxidation technologies and Electro-chemical oxidation are the physico-chemical methods.

• Air stripping

Air stripping is a physico-chemical technique often used along with other technical solutions for ammonia removal (Yuan et al., 2016). It accelerates the biodegradation of organic compounds and inorganic compounds of nitrogen (mainly ammonia-nitrogen). Significant and sustainable reduction of landfill emission occurs, resulting in minimization of environmental risks caused by polluted leachate. It may speed up the decreasing BOD₅ and TOC concentrations, which is an indicator of organic material degradation; while the decrease in ammonium concentration and the similar intermediate presence of nitrate/nitrite are an indicator of nitrification processes. This process is applied for treating stabilized leachates, reaching high efficiencies with higher retention times and temperatures.

• Coagulation/Precipitation

It is an effective pretreatment before biological or membrane treatment, or an efficient leachate post- treatment. The method supports the reduction of large organic molecular compounds in leachate. Coagulant Ferric chloride at pH values above 9 is more effective than alum in reducing organic constituent of leachate. Lime can remove heavy metals such as Cd, Fe and Cr by up to 90 percent, but increases hardness and pH, offers low COD removal (20-40%), and produces large amount of sludge at high dosages. Due to its low efficiency for the removal of organic content, it is not suitable for complete treatment of leachate. The efficiencies in COD

removal primarily depend on pH, coagulant species, coagulant dose and leachate characteristics, ranging from ten percent to eighty percent.

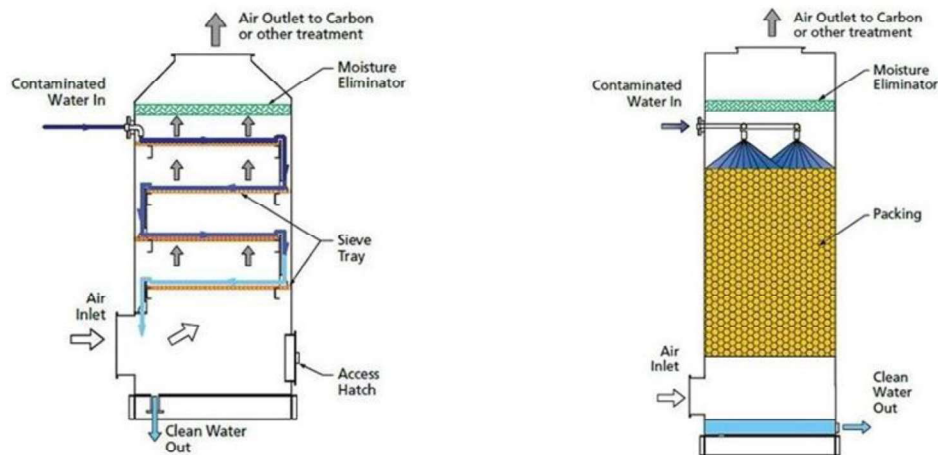


Fig 7: Air Stripping Process

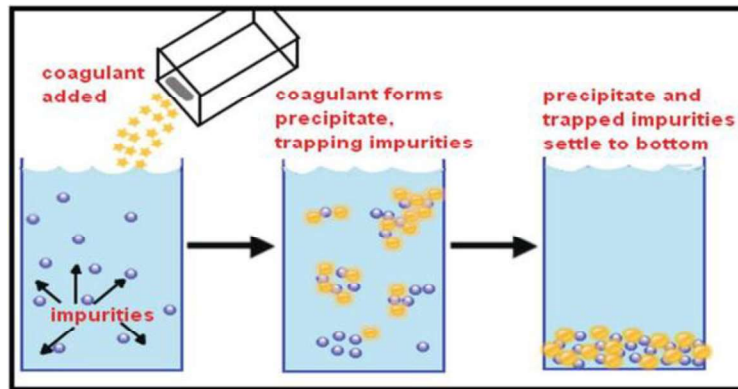


Fig 8: Coagulation/Precipitation Process

• Electro Coagulation

Electro coagulation involves chemical and physical methods, which use sacrificial electrodes for production of the coagulants. The production of metallic cations occurs at the anode, while H_2 production takes place at the cathode. It has better treatment efficiency than chemical coagulation. There are many factors able to influence the Electro-coagulation process, such as the reactor design, electrode material and current leachate density and conductivity (the removal of polluting particles increases with the density). Despite higher removal efficiencies achieved with aluminum, iron is considered the best electrode because it is less toxic, requires less energy, and is less sensitive to inhibitor phenomena (Z. Wang et al., 2016). The disadvantage is that Electro coagulation corresponds to high energy consumption and, therefore, it is necessary to find a compromise between the removal efficiency and consumption of power required (Orkun and Kuleyin, 2012).

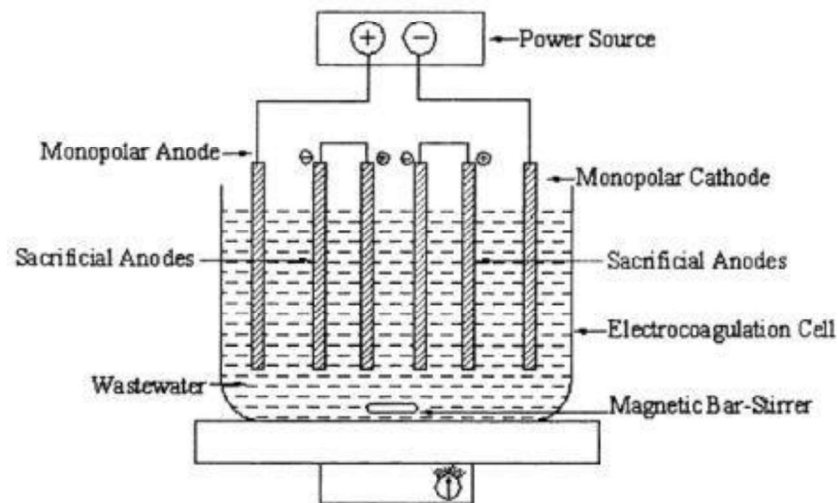


Fig 9: Diagram for Electrocoagulation Process

• Activated Carbon Adsorption

Adsorption is a surface phenomenon through which a multi-component fluid mixture is attracted to the solid adsorbent surface and gets attached via physical or chemical bonding (Foo & Hameed, 2009). Granular or powdered types of activated carbon accomplished a higher

reduction in organic component than that recorded for coagulation or precipitation. It is effective in eliminating non-biodegradable and colour causing organic components that are left after the biological treatment. This method extracts substances that are preferentially low molecular weight. Application is restricted by the need for regular regeneration of carbon particles or, equivalently, high amount of carbon powder.

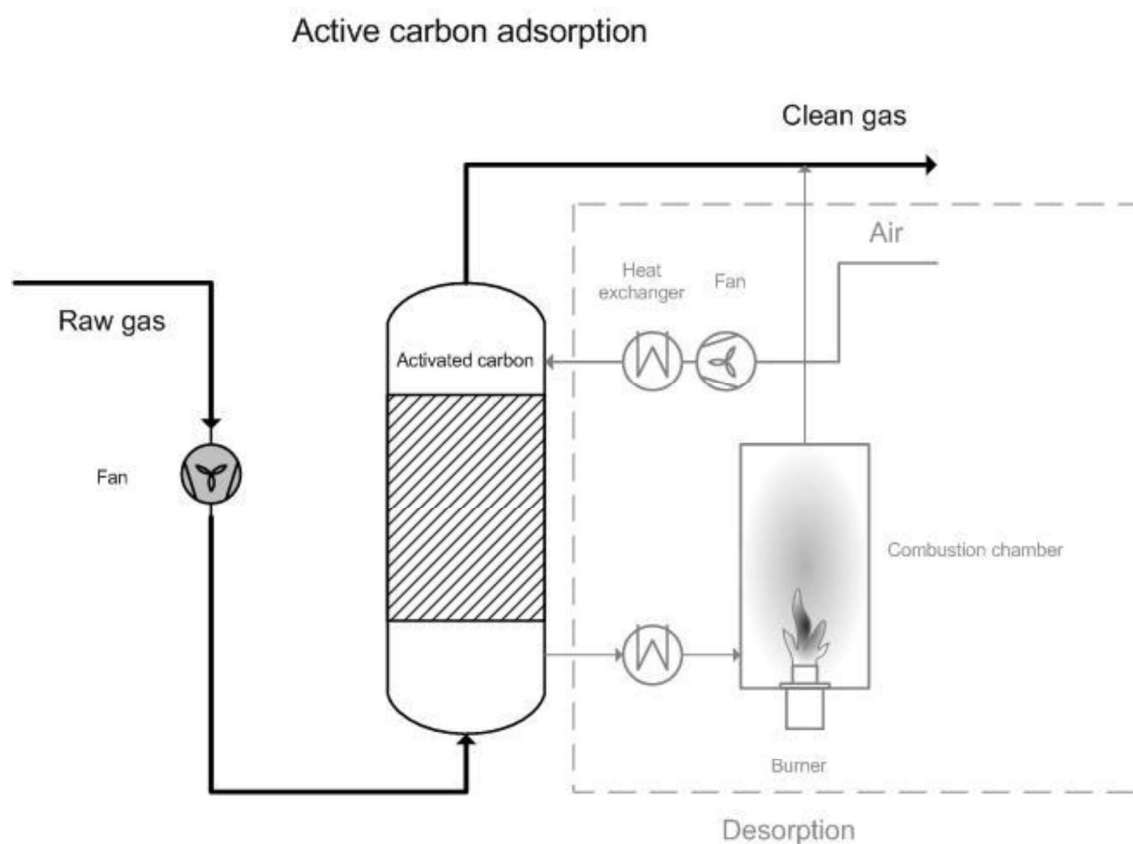


Fig 10: Activated Carbon Adsorption

• Advanced Oxidation Technologies

In recent years, there has been a growing interest towards Advanced oxidation processes (AOP) using the combination of strong oxidizing agents such as O_3 and H_2O_2 , together with ultraviolet radiation or ultrasound. Many processes are based on the direct reaction of the oxidant with the contaminants while AOP is characterized by the generation of OH radicals as reactive species able to oxidize halogenated organics and improve the biodegradability of recalcitrant organic pollutants.

Fenton's reagent ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$) is one of the most efficient method of organic pollutant oxidation among diverse AOPs. The Fenton reaction initiated by Fe^{2+} and H_2O_2 and the Fenton-like reaction initiated by Fe^{3+} and H_2O_2 are used for the treatment of leachates since they can significantly remove recalcitrant and toxic compounds and increase leachate biodegradability low operating cost. O_3 , UV, UV + O_3 , and O_3 + H_2O_2 have particularly high operating costs for leachate treatment. H_2O_2 used alone has a limited COD reduction capacity, although not all organics are oxidized to carbon dioxide and water at a low operating cost.

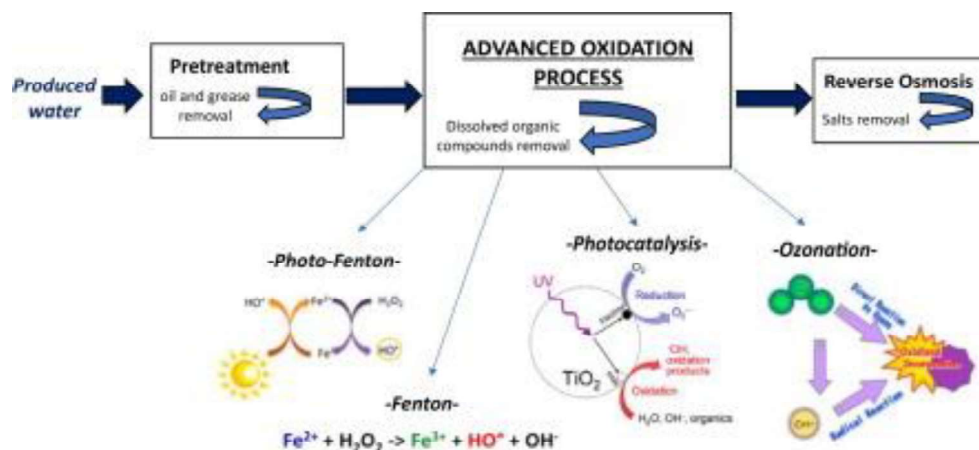


Fig 11: Advanced Oxidation Technologies

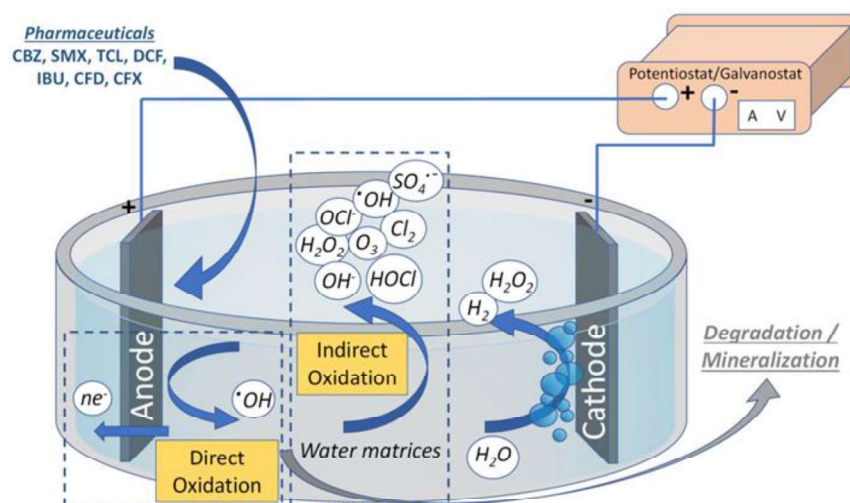


Fig 12: Electrochemical Oxidation

• **Electrochemical Oxidation**

The process is theoretically achieved by two distinct mechanisms: indirect oxidation, in which the oxidation is carried out by a mediator, which is electrochemically generated and direct anodic oxidation, in which pollutants are destroyed at the surface of anode. This method will effectively minimize organic content, ammonia, colour and metals from leachate of landfill. Current density varies extensively from 5-540 mA/cm² for electrochemical oxidation of leachate. However, this technology could be applied to leachates in order to reduce the concentration of refractory organic matter and ammonium. Using this technology alone, it appears that it is not possible to achieve the limits for discharging into sewage, except where local limits for carbonaceous substances and nitrogen are decidedly more permissive. Even though high energy consumption may limit its implementation, Electro Fenton process is an optimistic technology for extracting organic matter more effectively, especially for low iBOD₅/COD or high toxic landfill leachate and environmental friendly compared with conventional Fenton process (Rada et al., 2013).

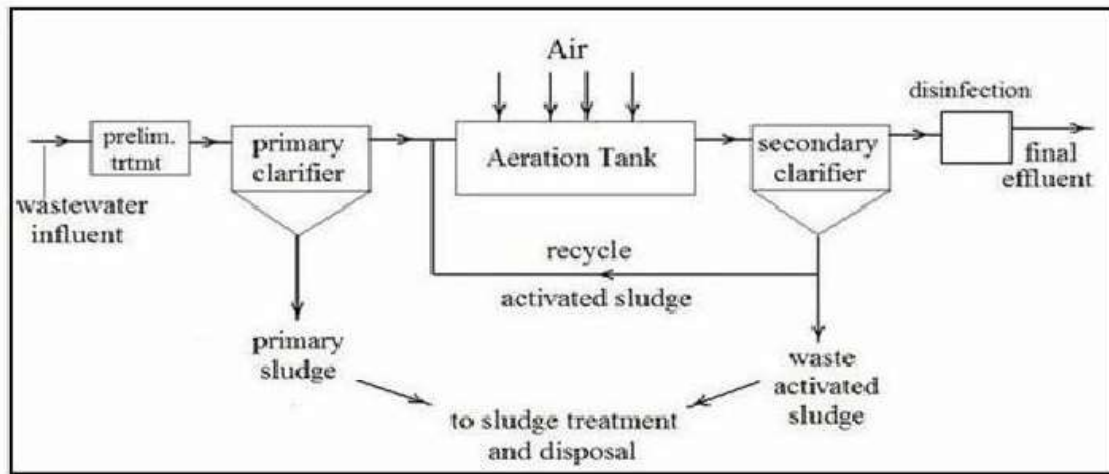
2.8.2 Biological Methods:

Biological processes include the use of microorganisms to remove the organic and nitrogenous matter from young leachates. It involves modifying the microorganism environment for growth which can eliminate the substances. Biological removals of organic compounds are carried out using anaerobic and aerobic decomposition processes. Under anaerobic conditions (absence of oxygen or nitrate) e.g. Digesters, lagoons, anaerobic filters etc, organic compounds are converted into methane and carbon dioxide (biogas) as well as water and a small fraction of fresh biomass (sludge). Non-injection of oxygen in the anaerobic systems, lowers their cost.

Under aerobic conditions (presence of oxygen) organic substances are converted to carbon dioxide, water and biomass. Activated sludge reactors, aerated lagoons and bio rotors are examples of biological processes. Biological processes, however, cannot remove refractory organic compounds. The various biological methods that are used for landfill leachate treatment are Activated sludge process, Rotating biological contactor (RBC), Sequencing batch reactor (SBR), Reed Beds, Biological aerated filters (BAF), Lagoons, Upflow anaerobic sludge blanket (UASB), Anaerobic filters (AF), Moving bed biofilm reactor (MBBR), Membrane bioreactor (MBR).

• Activated Sludge Process (ASP)

Activated sludge is one of the methods used in leachate treatment. The aeration tank is where oxygen is introduced as the leachate flows along the system. The sludge generated at the clarifier (settling tank), settles at the bottom of the tank while the supernatant is runoff as effluent. Part of the settled material (sludge) is returned to the head of the aeration tank for reseeded the incoming leachate with microorganisms. The re-seeding portion of the sludge is called return activated sludge while the excess sludge which accumulates is removed. However, the activated sludge process is not adequate for the leachate treatment due to the significant disadvantages of this treatment like high sludge production, which involves considerable costs for disposal; significant energy demand; the presence of inhibitor microorganisms due to the high concentrations of $\text{NH}_3\text{-N}$ (Renou et al., 2008).



Activated Sludge Wastewater Treatment Flow Diagram

Fig 13: Activated Sludge Wastewater Treatment Flow Diagram

• Rotating Biological Contactor (RBC)

The rotating biological contactor, an attached growth technology is also known as a bio-rotor. It has plastic circular discs mounted on a shaft which partly submerged in a tank containing the leachate. As the shaft rotates gradually, microorganisms adhere to the disc as biological growth, assimilating and treating organic substances from the leached water as they move over the surface of the disc. When the disc rotates out of the leachate, aerobic conditions are maintained and organic compounds get oxygenated. Thus, the disc makes contact between biomass and the leachate, mixing the mixed liquor and aerating the leachate. The performance of RBCs depends upon several design parameters such as rotational speed, organic and hydraulic loading rates, hydraulic retention time (HRT), RBC media, temperature, wastewater and biofilm characteristics, dissolved oxygen level, effluent and solids recirculation, step-feeding and medium submergence. Moreover, RBCs have some operating problems such as difficulty in the maintenance of appropriate biofilm efficiency is generally lower than with that of an activated sludge. thickness under adverse conditions (Cortez et al., 2008).

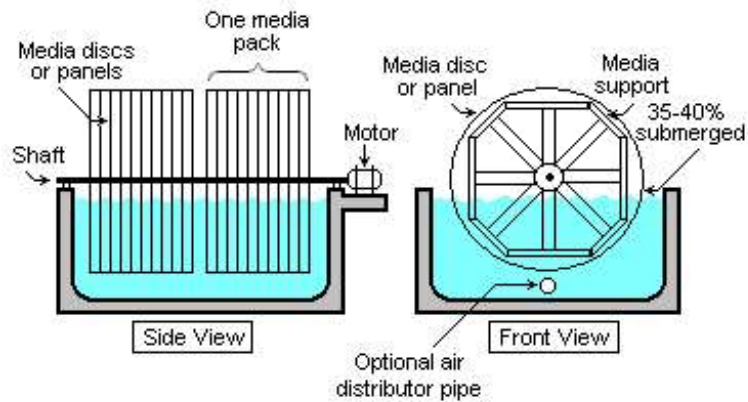


Fig 14: Rotating Biological Contactor (RBC)

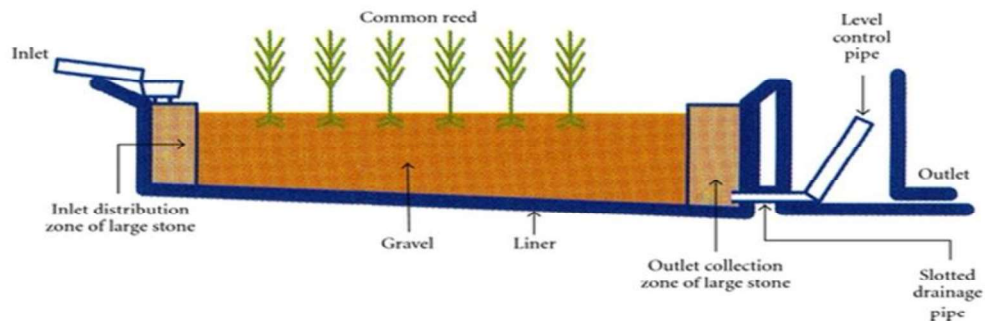


Fig 15: Reed Beds

• Reed Beds

Reed beds usually have gentle sloping beds lined with impermeable barriers and planted with emerging hydrophytes such as reeds (phragmites), bulrush (scirpus), or cattails (typha). Reed beds may have a crushed stone inlet zone to distribute wastewater evenly over the bed and an outlet zone of crushed stone for collecting and discharging effluent. Leachate enters at the inlet and flows gradually through the bed following a horizontal flow direction before leaving. The extensive root system of the reed provides a large surface area for attached microorganisms, increasing the potential for the decomposition of organic matter: Nitrogen and phosphorus are removed through plant uptake while ammonia is removed through volatilization and nitrification / denitrification (Kivaisi, 2001). The gravel or soil where the reeds are planted also serve as a filter medium.

• Biologically Aerated Filter (BAF)

A biological aerated filter is a treatment tank consisting of a submerged aerated fixed film biological filtration system that provides a surface for the biomass and also retains suspended solids which acts both as biological contactor and a filter, eliminating the need for a separate sedimentation step. There are numerous types of filling materials, all of which contribute to the removal of pollutants in different ways which can be reused materials: plastic, wood or rubber from tyres. In the system, both the influent and the process air flow upward from the bottom. The highest biological activity occurs in the lower half of the filter and the treated leachate stays above the media.

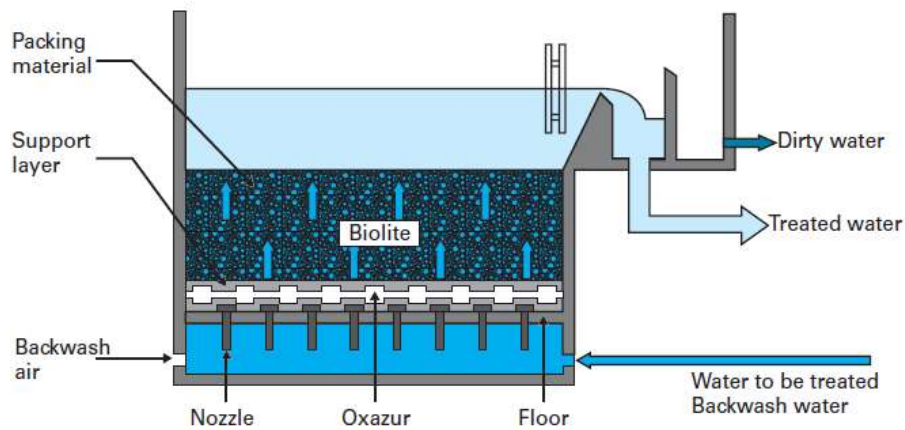


Fig 16: Biologically Aerated Filter (BAF)

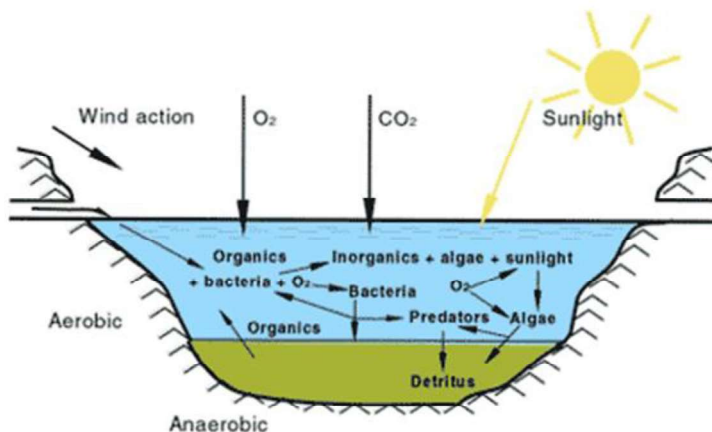


Fig 17: Lagoons

• Lagoons

As in an activated sludge reactor, a lagoon for leachate treatment is normally an artificial pond with microorganisms. Lagoons can be anaerobic or aerobic, artificial or natural. A lagoon is anaerobic because it lacks dissolved oxygen for most of its depth (e.g. lagoons where liquid animal wastes are dumped) whereas an aerobic lagoon is one in which dissolved oxygen exists for most of its depth. Over the years, leachate treatment studies have been performed in anaerobic lagoons as well as in aerated lagoons. If the aerated lagoon method is adopted as a full-scale leachate treatment, hydraulic retention times (HRT) need to be fully evaluated since they can be significant (Mehmood et al., 2009). This treatment is suggested in countries with extensive free areas, with no energy available and low economic funds.

• Up Flow Anaerobic Sludge Blanket (UASB)

UASB technology is a form of anaerobic digester used for treatment of leachate and many other forms of waste water. The process involves an upward passage of leachate through an anaerobic sludge bed inside a tank. When the leachate passes through the sludge, microorganisms degrade organic matter in the leachate producing biogas (methane and carbon dioxide). As the gas moves upwards to escape, hydraulic turbulence takes place in the reactor prompting mixing which result to more degradation as a result of more contact of microorganisms with substrate. The gas is collected at the top of the reactor and the liquid phase is separated from the sludge solid. The effluent is collected after the separation of sludge. Retention of active sludge within the UASB reactor enables good treatment performance at high organic loading rates. Natural turbulence caused by the influent flow and the biogas production provides good wastewater-biomass contact in UASB; hence higher organic loads can be applied in UASB systems. Therefore, less reactor volume and space are required while, at the same time, high-grade energy is produced as a biogas that can be exploited. However, the effluent from UASB reactors usually needs further treatment in order to remove remnant organic matter, nutrients and pathogens (Seghezzo et al., 1998).

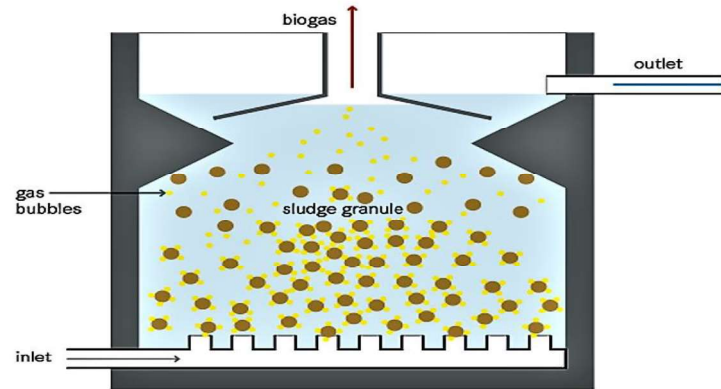


Fig 18: Up Flow Anaerobic Sludge Blanket (UASB)

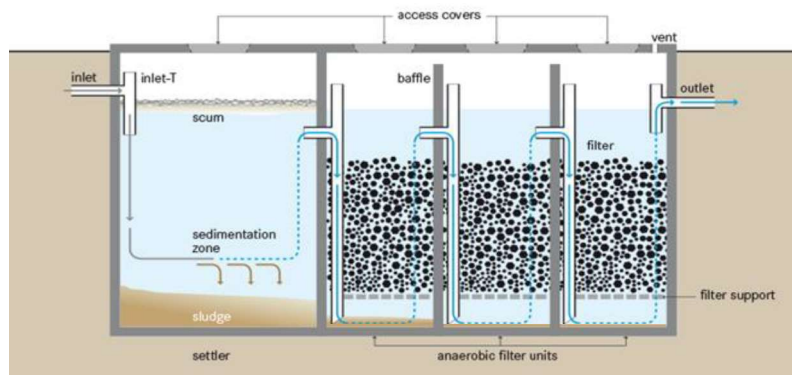


Fig 19: Anaerobic Filter

• Anaerobic Filter (AF)

Anaerobic filter is yet another type of anaerobic digester. The digestion tank contains a filter medium (pieces of polyvinyl chloride plastic, etched glass, baked clay, reticulated polyurethane foam, expanded shale, porous stone) on which communities of anaerobic bacteria can grow. The bacteria remain in the filter, providing a long solid retention time, even though the HRT is much shorter. Soluble organic compounds in the influent wastewater pass in close proximity to the biomass and diffuse into the surfaces of the attached or granulated solids. Here they are converted to intermediates and to end products, specifically methane and carbon dioxide (Gourari & Achkari-Begdouri, 1997). AF has several advantages over aerobic and anaerobic processes. It is more suitable for handling high-strength wastewaters as it presents

high substrate removal efficiencies at short hydraulic retention times and high organic loading rates. It should be joined with other treatment solutions as its removal yields must be improved for aggressive wastewater treatment.

• Moving Bed Biofilm Reactor (MBBR)

The MBBR process is an attached growth biological leachate treatment process. In a reactor with the MBBR process, microorganisms attach themselves and grow on a plastic biofilm carrier that are suspended and are in continuous movement within the reactor which maximizes growth by protecting the biofilm from abrasion on a specified volume resulting in uniform and highly effective treatment. The media carriers may have different shapes and sizes depending on the application. They can occupy different volumes in the reactor, from 30% to 60%, depending on the amount of required biomass (Hemi et al., 1994). There might be a nitrification tank and denitrification tank. Sludge generated at the settling tank, settles at the bottom of the tank while the supernatant is run off as effluent. To re-seed incoming leachate with microorganisms, part of the settled material (sludge) is returned to the head of the aeration tank.

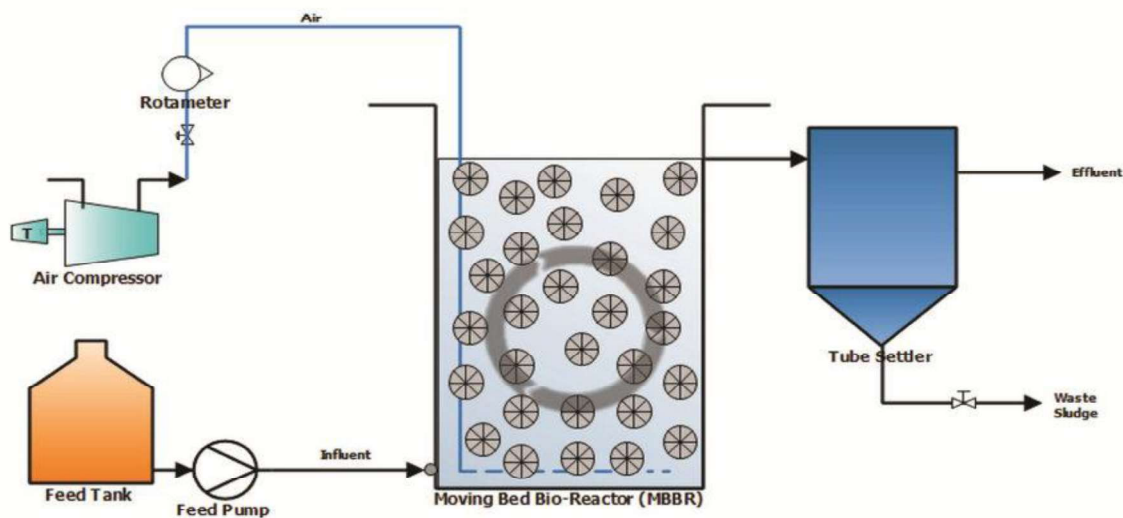


Fig 20: Moving Bed Biofilm Reactor (MBBR)

• Sequencing Batch Reactor (SBR)

SBRs are alteration of the Activated sludge process. They vary from activated-sludge plants as they include all of the treatment measures and process wastewater in a single basin or tank whereas conventional plants depend on multiple basins. The operation systems used in aerobic conditions comprise four steps: (1) feeding (2) aeration (3) settling and (4) discharge. SBR can be attached growth or suspended growth process. Due to the consumption of carbon sources in the aeration stage, wastewater does not have sufficient carbon source in the following anoxic denitrification stage and it leads to low total nitrogen (TN) removal efficiency (Wei et al., 2012).

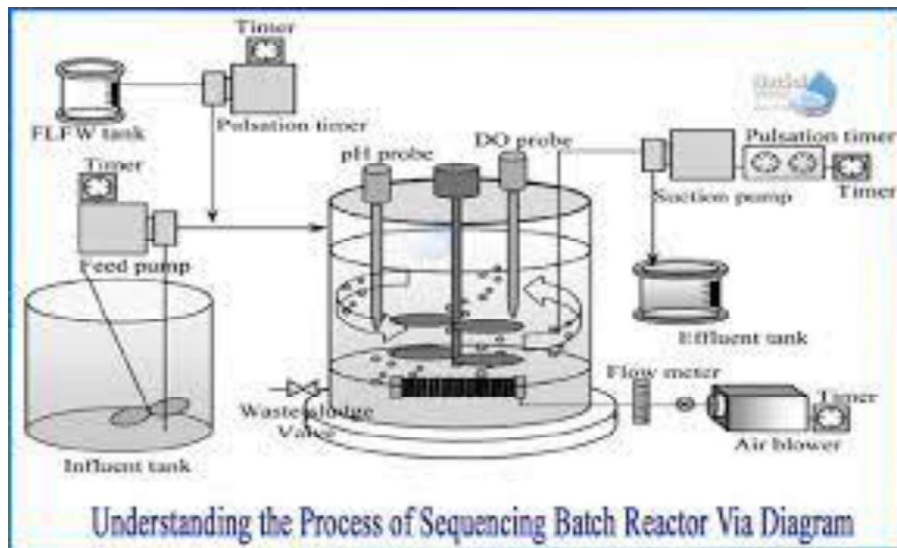


Fig 21: Sequencing Batch Reactor (SBR)

• Membrane Bioreactor (MBR)

Membrane bioreactor technology, which combines the biologically activated sludge method and membrane filtration, has become more common, abundant, and has been approved for the treatment of landfill leachate in recent years. Membrane bioreactor is a biological waste water treatment process which uses membrane to replace the gravitational settling tank for the solid liquid separation of sludge suspension in the conventional activated sludge process. The MBR

produces a clarified and considerably disinfected effluent. In addition, it concentrates the biomass and reduces the necessary tank size, increasing the bio-treatment process efficiency. MBRs thus tend to generate treated waters of higher purity with respect to dissolved constituents such as organic matter and ammonia, both of which are significantly removed by bio-treatment (Santos et al., 2011). MBR's in which biomass is strictly separated by membrane provide many advantages over conventional activated sludge process, including higher concentration of biomass, lower footprint, low output of sludge and improved permeate quality (Naghizadeh et al., 2008).

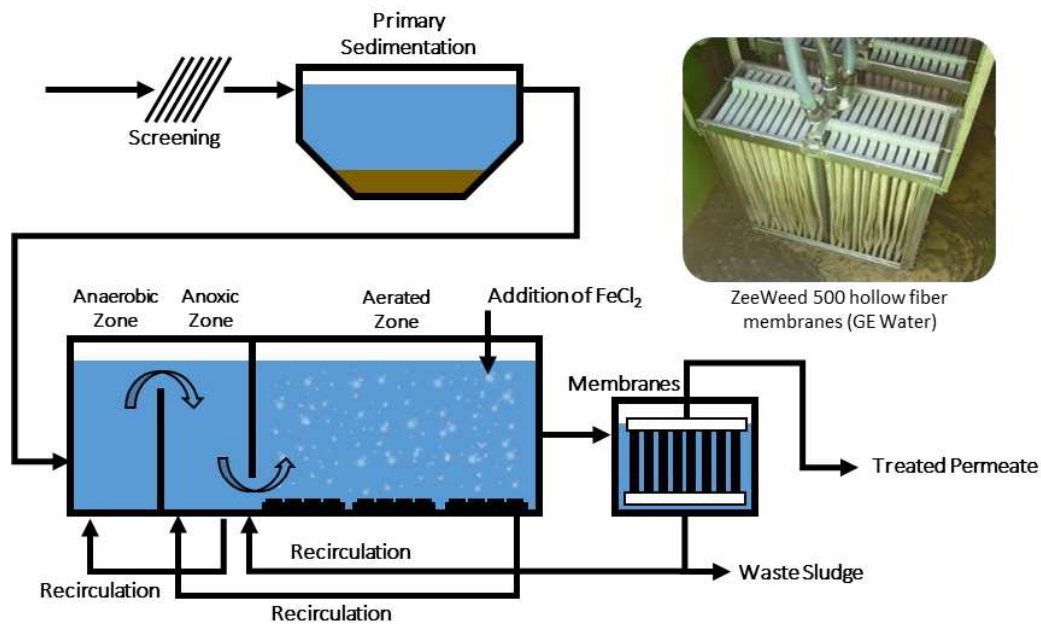


Fig 22: Membrane Bioreactor (MBR)

2.8.3 Coupling Physico Chemical Process and Biological Process:

Taking into account the leachate age, season, climatic conditions, regulation criteria and pollutant concentration, leachate treatment plants are forced to integrate physico-chemical and biological stages which improves the drawbacks and contributes to higher treatment efficiency. Past studies had already highlighted the difficulty of treating landfill leachate by stand-alone conventional chemical/physical or biological treatments due to the high percentage of high-

molecular-weight organic materials and biological inhibition suggesting a combination of technologies in order to achieve high pollutant removal rates. Indeed, biological treatments are useful to decrease organic pollutants, but they are not able to reduce heavy metal concentrations or inorganic chemical compounds usually found within landfill leachate; on the contrary, physical-chemical treatments are not suited to treat young leachates as they are not able to effectively reduce organics fractions. As a result, stand-alone technologies are not useful for leachate pollutant reductions and a combination is suggested in order to achieve regulation limits for the release to water bodies. Many combined treatments (SBR + Electrooxidation, ASP + coagulation + photo fenton, Trickling filter + Electrocoagulation, photo fenton + MBR, Aerobic SBR + Adsorption, constructed wetland + adsorption, MBR + UF + Electro-Oxidation etc.) were studied, which achieved high pollutant removal rates and enhanced treatment efficiencies, thus allowing project developers to implement technological choices suited for specific landfill release management issues. Advanced oxidation technologies are used in situations where the complex industrial effluents are not suitable for conventional biological treatment methods. The role of oxidation depends on the objectives of the treatment and can range from partial remediation to complete mineralization. In the case of partial treatment, oxidation aims at selectively removing the more bio resistant fractions and converting them to readily intermediates which can then be treated biologically. Coupling advanced oxidation with biological post-treatment is conceptually advantageous because it can lead to improved overall treatment efficiency relative to the efficiency of each individual stage. Membrane technology was noted as a viable treatment technology to comply with strict release concentrations, as reported by Abbas et al., (2009). However, fouling issues and high energy requirements are barriers that need to be overcome, specifically in developing countries.

2.9 Difficulties in Treating Landfill Leachate:

At present, the major challenges in handling leachate are as following: (1) Leachate has high organic component and a significant number of toxic and recalcitrant molecules. A single biochemical process or physicochemical process cannot achieve discharge standards; an integration of physicochemical and biochemical processes is required. The first challenge is to select a fair, economical and efficient integrated process (2) Ammonia nitrogen levels are high, and it is difficult to identify an efficient and full nitrogen removal method for leachate. Improving the performance of nitrogen removal is the second main challenge (3) the major variations in water quality and quantity intensify the complexity of finding a reliable discharge

method. Leachate water quality and quantity can be varying which challenges both the identification and the operation of an effective treatment process. The third challenge in leachate treatment is the identification of a suitable integration of available technologies and how to apply them for a stable operation (4) the treatment process is highly complex and the cost of treatment are very high. In order to achieve discharge standards, leachate treatment plants often use nano-filtration and reverse osmosis which makes the treatment costs high. Reducing leachate treatment costs is the fourth main difficulty (K. Wang et al., 2018).

2.10 Summary of Leachate Treatment:

Leachate treatment which can be classified into five major groups – physicochemical treatment, advanced oxidation process treatment, biological treatment, combined physicochemical and biological treatment and combined advanced oxidation process and biological treatment were discussed in the previous section. The strengths and the weaknesses of each of the treatment process were summarized in Table 11.

Table 11: Treatment Process and its Strengths and Weaknesses

Treatment Process	Strength	Weakness	Reference
Coagulation	Applicable for mature landfill leachate (50-60% COD removal)	Not applicable for young landfill leachate (10-25 % COD removal)	Amokrane et al., 2007 (<i>Amokrane.Pdf</i> , n.d.); (Hamidi et al., 2007)
Reverse Osmosis	High removal on COD and $\text{NH}_3\text{-N}$ (96-97%)	Costly and need extensive pretreatment	(Ahn et al., 2002); Tchobanoglous et al., 1993
Ammonium Stripping	94% $\text{NH}_3\text{-N}$ removal was achieved with 11 g/L of lime	Not effective for the removal of recalcitrant organic compounds (COD removal less than 15%)	(Calli et al., 2005); (Kurniawan et al., 2006)
Nanofiltration	Effective in removing heavy metals (88% for the removal of Pb^{2+} , Zn^{2+} and Cd^{2+}) using AFC-30 membrane, and 66% of COD removal	Useful only as a polishing step	(Linde & Jönsson, 1995) (Martinen et al., 2002)

Advanced oxidation process (AOP)	Can achieved 45-85% of COD removal and increase the biodegradability of a mature landfill leachate to 0.3-0.6	Not economically feasible for large-scale effluents	Kim et al., 2001; Roddy and Choi, 1999; (Scott & Ollis, 1995); (Rivas et al., 2003)
Sequencing batch reactor (SBR)	Effective in removing organic and nitrogenous matter (BOD removal 95% and nitrogen removal 50%)	High volume of sludge production which requires handling, treatment and disposal	(Diamadopoulos et al., 1997)
Electrocoagulation	It can lead to better quality effluent, requires a low level of electrical current	Electrodes are impermanent	Li et al., 2011; Huda et al., 2017

2.11 Previous Studies:

Utilizing electrochemistry principles, electrocoagulation is a substitute technology for the removal of metals from wastewater. The in-situ generation of coagulants is carried out using a sacrificial electrode (anode). Similar to coagulation in terms of removal mechanism, but with the added benefit of producing less sludge (Mollah et al., 2001). Even though the electrocoagulation process is 100 years old, the underlying mechanisms and process understanding have not been fully explored. The majority of studies relied purely on empirical model construction and lacked any real insight. This technology has been used more frequently over the past ten years to remove metal in the metal plating and automotive industries. Electrocoagulation involves combination of physical and chemical phenomena in which sacrificial electrodes are used to generate *in situ* ions into the wastewater stream. These ions serve as coagulants and balance the metal ions' net charges. They become closer for agglomerations as a result of the charge neutralisation, which is then settled during the clarification process (Koren and Syversen, 1995);(Syversen, 1995); Mollah et al., 2004).

Ample scientific literature is available for the use of electrocoagulation to treat wastewater carrying metals. Fig. 2.2 summarizes the evolution of published research using electrocoagulation treatment in the last twelve years (data extracted from Scopus). Fig. 2.3 depicts the top fifteen journals publishing articles on electrocoagulation. The use of electrocoagulation has been successful for the removal of different metals, including copper,

cadmium, boron, and arsenic, nickel, lead, iron, and chromium. Removal of arsenic by electrocoagulation with iron and aluminium electrodes was studied in a batch bench scale plant. Results demonstrate that both iron and aluminium electrocoagulation are robust technologies capable of removing arsenic down to $10 \mu\text{g}/\text{dm}^3$ (the level fixed by most environmental and health agencies). Nevertheless, aluminium electrocoagulation is not as efficient as iron. The effect of the current density (from 0.1 to 4.0 mA/cm^2) was studied with both the electrodes. Results show that this parameter influences slightly on the range of pH in which the process works, and also on the efficiency (always over 99.9%), but it is not a limiting parameter. Results can be easily explained taking into account the solubility of iron, aluminium and arsenates species present in the treated water (Lacasa et al., 2012).

Arsenic removal via electrocoagulation from field pilot scale has been studied in Mexico (Parga et al., 2005). According to the findings, the reactor's amorphous iron oxyhydroxide and magnetic particles remove arsenic with an efficiency of more than 99%. Gao et al. (2005) investigated the electrocoagulation-electrofloatation process without a filter for the removal of chromium. The parameters like charge loading, effect of pH, effect of surfactants, effect of Al^{+3} and power consumption were studied. The electrocoagulation process containing 100 mg/L arsenic solution was treated with 98% efficiency at current density of 1.2 A/dm^2 and residence time of 9 min has been investigated by Hansen (Hansen et al., 2006). Escobar (Escobar et al., 2006) has reported using steel electrodes in electrocoagulation to remove copper, lead, and cadmium from natural waters and simulated wastewater electrofloatation of dilute synthetic zinc solution with 96% removal efficiency using sodium dodecyl sulphate as collector, current density of around 8 mA/cm^2 and inlet pH of 7.0 has been demonstrated by Casqueira (Casqueira et al., 2006). Vasudevan et al. (2012) researched the removal of arsenic using direct and alternating current by electrocoagulation.

Several employees have used electrocoagulation to treat wastewater that contains Cu^{+2} , Zn^{+2} , and Cr (IV). To increase efficiency and optimise, variables such as pH, current density, and metal ion concentration have been researched. Electrocoagulation with an aluminium anode and a stainless steel cathode has also been used to remove oil, heavy metals, and organic compounds from oil-in-water emulsions in continuous processes (Saur et al., 1996). Using laboratory studies, the electrocoagulation unit was examined for the purification of soluble oil

waste with a high COD (Rubach and Saur, 1997; Calvo et al., 2003; Gotsi et al., 2005) (Rubach & Saw, n.d.) (Rostan & Lapiquea, n.d.). Studies were done to determine how COD removal was affected by various factors, including current density, sodium chloride concentration, coagulant dose, recirculation rate, electrode gap, etc. Based on a laboratory scale experiment, a pilot scale electrocoagulation unit was designed and constructed (Calvo et al., 2003) (Rostan & Lapiquea, n.d.). Panizza and Cerisola (Panizza & Cerisola, 2006) investigated the anodic oxidation process used to treat wastewater at an olive mill using parallel plate electrodes. The removal of COD, aromatic content, and colour were studied in relation to chloride concentration and applied current.

Heavy metals (copper, chromium, and nickel) from a plating industrial plant in Thailand were treated electrochemically on a laboratory scale with 99% treatment efficiency (Hunsom et al., 2005). (Park & Kim, 2004) have investigated how activated carbon fibres affect the effectiveness of electrochemical oxidation in removing Cr (VI), Cu (II), and Ni (II).

(Adhoum et al., 2004) used electrocoagulation for the treatment of electroplating wastewater with 94% copper removal efficiency at charge loading of 480 A/m² current density, 50 cm² anode surface area in 5 min treatment time. Optimum pH was found to be between 4 and 8, current density of 80–480 A/m² consuming 32 Ah/L energy consumption. (Hunsom et al., 2005) reported that electrochemical treatment of copper containing wastewater showed 99% removal efficiency using 12 h treatment time at 10 A/m² current density. When Cr(VI) was present, the treatment proceeded according to zero order copper removal kinetics, but when Cr(VI) concentration was reduced, the process changed to first order. Escobar has researched the electrocoagulation process' optimization for the removal of copper, lead, and cadmium from natural and simulated wastewaters (Escobar et al., 2006). At pH 5 and 31 A/m² current density, the maximum copper removal of 80% was accomplished in 6 minutes. Additionally, the removal efficiency dropped from 91% to 73% when the initial copper concentration was reduced from 3 mg/L to 1 mg/L. In another study, on copper removal using aluminium electrodes, the optimum pH was found to be between 4 and 6, consuming 43 KWh per Kg of copper removed (El-Ashtoukhy et al., 2010). Akbal and Camci (Akbal & Camciotless, 2011) found 96.7% copper removal efficiency at a pH of 3, 20 min treatment time, 50 A/m² current density, 6 mS/cm conductivity for 45 mg/L of copper concentration.

Textile wastewater has been successfully treated using electrocoagulation (Kobyas et al., 2003) (Bayramoglu et al., 2004) (Bayramoglu et al., 2007) (Can et al., 2006) (Rajkumar et al., 2007) (Zaroual et al., 2006) (Rajkumar & Kim, 2006). Kobyas (Kobyas et al., 2003) has investigated the use of iron and aluminium electrodes in the treatment of textile wastewater. According to the findings, iron is more effective than aluminium at removing COD and uses less energy overall as a sacrificial electrode material. (Bayramoglu et al., 2004) studied electrocoagulation of textile dye wastewater with reference to operating cost using Fe and Al electrodes. Costing has been done on the basis of electrode material dissolution and energy consumption. Operating costs for iron electrodes range between 0.1 and 0.3 US dollars per kilogramme of COD removed. The combined effect of chemical coagulation and electrocoagulation in the treatment of the textile wastewater has been studied by Analysis of COD removal effectiveness and operating costs was conducted. Zaroual (Zaroual et al., 2006) studied industrial textile wastewater in batch reactor. The removal efficiency was 100% colour and 84% COD in 3 min at potential of 600 mV.

Discolorization and treatment of dyes in wastewater have been investigated by various authors (Xiong et al., 2001); (Roessler & Jin, 2003); (Fernandes et al., 2004); (Daneshtar et al., 2006); (Modirshahla et al., 2007); (Rajkumar & Kim, 2006); (Rajkumar et al., 2007); (Kobyas et al., 2007). Meunier (Meunier et al., 2006) compared the electrocoagulation with chemical treatment for heavy metals. Compared to a chemical process, electrocoagulation was five times cheaper in price. While nickel was easily removed by chemical precipitation, about 99.5% of the lead was removed using electrocoagulation. Yildiz (Yildiz et al., 2007) has demonstrated the treatability of synthetic prepared water containing humic substances. The effects of the supporting electrolytes (Na_2SO_4 , NaCl, and NaNO_3), initial humic concentration, and applied voltage were investigated.

Chen (Chen et al., 2000) studied the effect of electrolysis voltage on electrocoagulation process. The electrolysis voltage depends primarily on inter-electrode distance, conductivity, current density and electrode surface state. Treatment of poultry wastewater by electrocoagulation has also been investigated (Kobyas et al., 2006). On removal efficiency for oil-grease and COD, the effects of process variables including pH, electrode material, current density, and operating time are investigated. Technical and economic analysis of electrocoagulation for the treatment of poultry slaughterhouse wastewater in Turkey was studied by Bayramoglu (Bayramoglu et al., 2006). Iron electrode was superior in removing oil and grease with 98% efficiency, while aluminium electrode performed better in reducing COD

with 93% removal efficiency in 25 minutes of retention time. Koparal and Ogutveren (Koparal & Öütveren, 2002) have reported the removal efficiency of nitrate from water using electroreduction and electrocoagulation.

The combined effect of electrocoagulation and electrofloatation processes to treat restaurant wastewater was studied by (Chen et al., 2000). The process produces sludge with less water and a shorter retention time. Separation of high oil and grease content from restaurant wastewater by electrocoagulation was studied by Chen (Chen et al., 2000). For all of the wastewater tested, the oil and grease removal efficiency was greater than 94%.

High performance electrochemical wastewater system for industrial and municipal wastewater has been documented (Feng et al., 2003). It has been reported that water with *Legionella* bacteria was treated electrochemically for disinfection (Feng et al., 2004). The disinfection was attributed to synergistic effects of the oxide anode, the electric field and the radicals formed during the electrochemical treatment. Vlyssides (Vlyssides et al., 2002) studied the laboratory scale electrochemical treatment of domestic wastewater using Ti/Pt as anode and stainless steel as cathode. At various pH ranges, the effects of variables including COD, volatile suspended solids, ammonia nitrogen, nitrates, phosphorous, chloramines, chloride, anode efficiency, and specific energy consumption were studied. Khemis (Khemis et al., 2006) investigated the treatment of concentrated wastes by electrocoagulation using sacrificial aluminum anode. A straightforward model was put forth to explain the experimental data. (C.-J. Lin et al., 2005) studied the pilot scale electrocoagulation with bipolar aluminum electrodes for on-site domestic grey water reuse. With an area requirement of 8 m², the total unit cost of on-site treatment was US \$0.27 per cubic metre (capacity 28 m³/day), which was less than the local potable water rate.

Operating cost of electrocoagulation treatment including energy consumption was discussed in detail by (Ghosh et al., 2008) using simplified cost equations. However, there hasn't been much work reported on enhancing the performance of electrocoagulation in terms of power consumption (Mollah et al., 2004). Alternating current electrocoagulation for aqueous waste remediation was studied by the United States' Superfund Innovative Technology Evaluation

(SITE) (Barkley et al., 1993). Parameters kind of current, electrode interspace and bearing time had been optimized. Baklan and Kolesnikova (Baklan & Kolesnikova, 1996) studied the influence of electrode material on the electrocoagulation by using iron anode and aluminium cathode.

Alternating pulse current (APC) was used in electrocoagulation removal of hexavalent chromium employing aluminum and iron electrodes. Initial pH, current density, reaction time, initial Cr(VI) concentration, and conductivity were some of the operational parameters whose effects were investigated. At optimal conditions, a chromium removal efficiency of about 99% was attained, and the outcomes were compared to direct current. But, APC produced better water recovery and very low turbidity (Keshmirizadeh et al., 2011). Table 2.4 presents the brief comparison for chromium removal using electrocoagulation technique.

A semi-pilot scale plant for water defluoridation using a 200 L/h electroflocculator has been developed and commercialised in India by the Central Electrochemical Research Institute (CECRI), Karaikudi. At IIT, Bombay, batch experiments were conducted for the electrocoagulation-based removal of arsenic from drinking water (Kumar et al., 2004). According to the study, iron electrodes are more effective at removing arsenic than aluminium and titanium electrodes. Performance of electrochemical reactor and influence of anode material on the electrochemical oxidation for the treatment of tannery wastewater from common effluent treatment plant (CETP) located at Ranipet, Tamil Nadu was studied by (Szpyrkowicz et al., 2001). The treatment of dilute copper cyanide wastewater (synthetically prepared) arising from industrial electroplating has been carried out under hydrodynamic effects by Szpyrkowicz et al. (2005a, 2005b). When the electrocatalytic film of copper oxyhydroxide could form in situ on the anode with minimal turbulence, cyanide and copper could be removed simultaneously (Szpyrkowicz et al., 2005a). South Indian wastewater from the cigarette industry was treated electrochemically using cast iron electrodes (Bejankiwar, 2002). The treatment efficiency was 56 % for COD and 84% for BOD removal with current rating of 3.5 A for 5 hour of electrolysis. Ti/TiO₂-RuO₂-IrO₂ electrodes and an undivided reactor were used to perform the electrochemical treatment of phenolic wastewater from M/s IND-ITAL chemicals, Madavaram, Chennai, at a current density of 5.4 A/dm² (Rajkumar & Palanivelu, 2004). According to the study, highly concentrated bio-refractory organic

pollutants found in industrial wastewater can be effectively treated electrochemically. Cast iron electrodes were used to investigate the electrochemical oxidation of pharmaceutical wastewater from a manufacturing facility in Mysore with a low BOD/COD ratio. After two hours of treatment, the BOD/COD ratio had increased from 0.18 to 0.3, resulting in a 72% removal efficiency for COD. (Deshpande et al., 2005).

Rusdianasari (Rusdianasari et al., 2017) investigated the effectiveness of electrocoagulation in removing Biochemical Oxygen Demand (BOD_5), Chemical Oxygen Demand (COD), NH_3 -N and lead (Pb) of leachate. Optimum conditions and the effectiveness of electrocoagulation for each parameter are in the current density 30 A/m² with 60 minutes of time process. The electrocoagulation method is effectively reduced the value of TSS 46.80%; BOD_5 71.33%; COD 73.77%; Pb 62.5%; NH_3 -N 57.92%; and for the increased of pH from 8.03 to 8.95, respectively.

A combination of biofiltration (BF) and electrocoagulation (EC) processes was used for the treatment of sanitary landfill leachate by Dia Oumar (Oumar et al., 2016). Ammonia nitrogen ($N-NH_4$ removal of 94%), BOD (94% removed), turbidity (95% removed) and phosphorus (more than 98% removed) were removed when BF process was used as secondary treatment. The optimum performances of COD and colour removal from landfill leachate were obtained when a current density of 10 mA/cm² through 30 min of treatment were applied. The COD removal reached 53%, whereas 85% of colour removal was recorded. It has been proved that the alkalinity had a negative effect on COD removal during EC treatment (Dia Oumar et al., 2016) (Oumar et al., 2016).

The factors affecting the efficiency of removing NH_3 -N and COD of leachate, such as electrode material, current density, electrolysis time, Cl^- concentration and pH of solution were studied by Xiangdong Li (X. Li et al., 2011). Treatment was carried out in a discontinuous system and the results show that the electrocoagulation can be applied to leachate pre-treatment. The operating conditions were current density of 4.96 mA/cm², the raw pH, Cl^- concentration of 2319 mg/L, operating time of 90 min with Fe electrode, the highest COD and NH_3 -N removal efficiencies were 49.8% and 38.6%, respectively.

It is stated by Fatih Ilhan (Ilhan et al., 2008) that treatment of leachate by EC can be used as a step of a joint treatment when it is performed in a batch process. Firstly, EC was compared with classical chemical coagulation (CC) process via COD removal. The first comparison results with 348 A/m² current density showed that EC process has higher treatment performance than CC process. Secondly, effects of process variables such as electrode material, current density (from 348 to 631 A/m²), pH, treatment cost, and operating time for EC process are investigated on COD and NH₄-N removal efficiencies. The appropriate electrode type search for EC provided that aluminium supplies more COD removal (56%) than iron electrode (35%) at the end of the 30 min operating time.

During the electrocoagulation (EC) treatment of landfill leachate, the production of chlorine species may result in the formation of harmful disinfection by-products (DBPs) (Xu et al., 2020). With a current density of 100 mA cm⁻², the unregulated HK dominated the weight-based DBP concentration (96% in Leachate A and 44.3% in Leachate B), while the unregulated HAN contributed to >80% of the DBP additive toxicity in both leachates. Applying granular activated carbon as a post-treatment step could successfully reduce the total DBP concentration from 295.33 µg L⁻¹ to 82.04 µg L⁻¹ in Leachate A, leading to a total DBP removal of 72.2% and a toxicity removal of 50%.

Inter-electrode distance, initial pH and electrolyte concentration on colour and COD removals have significant effects on the colour removal and electrolyte concentration was the most significant parameter affecting the COD removal (Huda et al., 2017). Under optimum conditions (initial pH: 7.73, inter-electrode distance: 1.16 cm, and electrolyte concentration (NaCl): 2.00 g/L), the process could remove up to 82.7% colour and 45.1% COD.

A treatment approach resulted in clear, colourless and odourless effluent with the values of all measured parameters significantly lower compared to the upper permissible limit for discharge into the environment was obtained when a combined treatment approach using advanced oxidation, electrochemical methods and microwaves was developed and applied for the treatment of landfill leachate (Orescanin et al., 2012). Due to the complex nature of the effluent

and extremely low bio-degradability (BOD5/COD ratio=0.01) the purification of the leachate started with pre-treatment with ozone followed by simultaneous ozonation and electrocoagulation/electrooxidation using the set of iron and aluminium electrodes, and finally, the degradation of organic residue and ammonia with microwaves. The removal percentages of the parameters: colour, turbidity, suspended solids, ammonia, COD and iron following the combined treatment were 98.43%, 99.48%, 98.96%, 98.80%, 94.17% and 98.56%, respectively.

The removal of chemical oxygen demand (COD) from landfill leachate by electro-coagulation (EC) using iron sacrificial anode has been performed and analyzed in terms of COD removal efficiency and the important cost related parameters such as electrode and energy consumptions, as a function of initial pH, conductivity, current density, and operation time by Merve Oya Orkun et al., 2010 . The optimum COD removal (65.85%) was obtained with typical operating conditions: current density, 30 mA/cm²; operation time, 180 min; conductivity, 16.4 ms/cm; and initial pH, 6.54. At these conditions, the electrode and energy consumptions during the electrolysis were determined as 0.418 kg COD/kg Al and 11.092 kWh/kg COD, respectively. The EC and Fenton processes are applied together referred to as the electro-Fenton process for the removal of COD from landfill leachate and effect of operation time and hydrogen peroxide concentration are found as follows. The process was very fast in the first 15 min, and then it slowed down till it was complete in 60 min. An optimum COD removal efficiency of 74.21% was obtained at 5000 ppm hydrogen peroxide dosage and 60 min.

Electrocoagulation exhibited more efficient BOD removal (84 %–90 %), indicating its excellent capability of removing biodegradable organic material from wastewater. Neanderson Galvão (Galvão et al., 2020), showed the rate of coagulant generation to be high for a high applied current. The best removal efficiencies were obtained by applying J of 128 A m⁻² for 90 min of electrolysis, removing 82 %, 82 %, 40 %, 90 %, and 40 % of colour, turbidity, chemical oxygen demand, biological oxygen demand, and ultraviolet absorbance at a wavelength of 254 (Abs₂₅₄), respectively. The removal rates were slow for the smallest J applied. The sulphates showed complete removal with a J of 96 and 64 A m⁻² and the pH of

the leachate increased with increasing electrolysis time and J owing to electrogenerated hydroxyl (OH^-) ions.

Job Contreras et al., 2008 investigated the effect of four control factors, namely current density, pH, time and fluid conductivity, as well as the interaction among (between) them to get an optimal turbidity removal by a central composite design. The central composite design consisted of 29 experimental points with five replications at the centre point. A second order polynomial model was used for predicting the response. Regression analysis showed that more than 95% of the variation was explained by the model wherein current density with a 60.1% contribution turned out to be the factor with the most significant influence. Analysis of variance showed that time, pH, current density and the interaction time/current density had a significant influence on the turbidity removal. The optimal conditions obtained for the removal of turbidity were time 38.8 min, pH 7.6, current density 109.9 A m^{-2} and NaCl 2.9 g L^{-1} . Experimental results showed that for a 96.9% turbidity removal, similar reduction in Al (97.0%) and Fe (99.5%) concentrations; as well as 66% total Kjeldahl nitrogen removal was obtained. Furthermore, the sludge formed exhibited a good floc size, which separated in less than 10 min by classical sedimentation. The results analysis provided evidence of reduction of chemical pollutants, although the electrocoagulated leachate could not satisfy regulations for the maximum Total Kjeldahl nitrogen leachate discharge level to public wastewater collection systems in the town of Freire, Chile.

The performance of EC process was carried out in batch reactor by Rosie Jotin (Jotin R, Ibrahim S, 2012) to study the effecting parameters such as applied voltage, conductivity and initial pH to achieve higher removal. Samples of 25mL were taken out from the batch at 10, 20, 40, 60, 80 and 100 minutes of contact time. Results obtained show that the most effective removal efficiency could be achieved at 10V of applied voltage. The increase of wastewater conductivity, in the range of 28.00- 31.00 mS/cm, reduced the EC performance on COD removal. EC was found to be very efficient when the range of initial pH is between 4 and 8. The highest COD removal efficiency of 74.08% occurred at a voltage of 10 V, conductivity of 28.00mS/cm, initial pH of 4 and contact time of 100 min.

Ultimately, it is up plant designers to decide which optimization criteria are the most important and what costs they are willing to accept in order to obtain higher COD removal rates (Sedighi et al., 2021). Such decisions are case-dependent (subjective) and will have to be made considering the specific conditions of each leachate treatment plant. The researchers investigated the impacts of initial pH (4.0–8.0), electric current intensity (1.0–5.0 A), initial temperature (25.0–45.0°C), and retention time (15–75 min) on COD removal. The highest COD removal was targeted irrespective of process costs. The best performance was obtained at a pH of 5.4, current intensity of 4 A, and reaction time of 60 min with an initial temperature of 40 °C, leading to COD removal of 71.78%. However, in real-world applications, the use of energy and resources must be limited to viable levels. Hence, under the second optimization scenario, capital and operational expenditures were taken into account. The use of chemicals, electricity and heating was minimized as well as reducing the size of the equipment and tanks. With these restrictions, at a pH of 6, current intensity of 3.4 A, initial temperature of 30 °C, and process duration of 47 min, COD removal of 51% was achieved. Although in the second scenario the COD removal decreased by 20%, the energy usage was halved and the electrode consumption was reduced by more than one-third, in addition to lower capital costs for smaller equipment.

a feasible model containing variables of the electrocoagulation process on COD removal was derived using stepwise regression analysis by Alper Alver (Alver & Altaş, 2017). The leachate was treated by the electrocoagulation (EC) method with aluminium and iron plate electrodes together in a laboratory-scale batch process. The effects of process variables such as current densities, time, and pH were investigated for chemical oxygen demand (COD) removal. Stepwise regression analysis was done for determining which variable, acting independently, best explains or predicts the variation in the removal efficiency of COD.

Electrocoagulation and electro-Fenton processes could be used as a post-treatment method for leachate and an effective method inert COD removal from leachate nanofiltration concentrate. However, both processes could be efficient methods for converting insoluble COD to soluble COD and removal of nonbiodegradable COD (Yazici Guvenc et al., 2019). Removal efficiencies of 60.2% for total organic carbon (TOC), 69.4% for chemical oxygen demand (COD), and 87.6% for colour from the electro-Fenton process at the optimum operating

conditions and 45% for TOC, 57.4% for COD, and 77.1% for colour from the electrocoagulation process at the optimum operating conditions respectively, were obtained by the researchers when electrocoagulation and electro-Fenton processes were carried out using iron electrodes in a batch electrolytic reactor. Additionally, the fraction of biodegradable COD increased from 6.37% to 17.04% and 30.08% after electrocoagulation and electro-Fenton processes respectively. About 39% soluble COD and 82% particulate COD removal efficiencies were obtained by electrocoagulation.

A second stage of treatment based on a biological process could be suitably included in order to abate recalcitrant organic matter and decrease remaining toxicity in a more efficient integrated treatment system. This is stated by Aline Roberta de Paulia (De Pauli et al., 2017), when they investigated organic matter (OM) removal performance along with the assessment of the toxicity of landfill leachate (LL) treated by Electrocoagulation (EC) method. In the context of response surface methodology (RSM), the optimal EC experimental conditions were sought by applying a 33 complete factorial design, regarding pH, current density and electrolysis time as operational parameters. The sum of dissolved organic and inorganic carbon, namely as dissolved total carbon (DTC), was chosen as a global response parameter. Other EC experiments were performed, keeping fixed pH and current density at local optimal values and varying the electrolysis time. Determination of the median lethal concentration (LC50) from bioassays based on *Artemia salina* and *Lactuca sativa* indicators was performed. A second-order polynomial function was statistically validated with good predictions of the DTC data, indicating best removals by setting values of pH, current density, and electrolysis time at 5, 128.57 Am⁻² and 120 min, respectively. Additionally, removals above 90% were achieved for colour, turbidity, iron concentration and dissolved inorganic carbon, whereas reductions on related-to-organic matter parameter values were around 60%. Although the EC treatment reduced the LL effluent toxicity, as verified by toxicity bioassays, 90 min treatment times showed best results on LC50, but higher toxicity was persistent in electrolysis times below 40 min.

the abilities of LL to stimulate oxidative stress in *G. pulex* have been proven. The results revealed that antioxidant parameters are useful biomarkers for determining the treatment efficiency of the electrocoagulation process (Serdar et al., 2018). To evaluate whether

electrocoagulation process is efficient for treatment landfill leachate (LL) or not by using antioxidant biomarkers in *Gammarus pulex*, Glutathione peroxidase (GSH-Px), superoxide dismutase (SOD), and catalase (CAT) activities and malondialdehyde (MDA) and glutathione (GSH) levels in *G. pulex* exposed to untreated, treated, and diluted rates 1/10 and 1/20 in both LL during 24 and 96 h were tested by the researchers. Physiochemical characteristics of leachate (chemical oxygen demand, electrical conductivity, pH, phosphate, turbidity, NH_3 , Cl^- , and colour) were determined pre and post treatment. All physiochemical characteristics of LL decreased after treatment process. GSH-Px and CAT activities and GSH and MDA levels were increased in untreated groups when compared to control ($p < 0.05$). After treatment by electrocoagulation, MDA and GSH levels and CAT activities were returned to control values.

F. Bouhezila (Bouhezila et al., 2011) showed that an increase in current density (125–500 A/m²) enhanced the speed of treatment significantly, the inter-electrode distance was 2.8 cm and the stirring speed was 150 rpm for the studied leachate. The removal efficiencies of COD, total nitrogen, colour and turbidity were respectively 70%, 24%, 56%, and 60% with Al electrodes and 68%, 15%, 28%, and 16% with Fe electrodes. Electrical energy consumption and operating cost with Al electrodes were 0.022 (kWh/L), 0.54 (US\$/m³ leachate treated), respectively, and 0.019 (kWh/L), 0.47 (US\$/m³) with Fe electrodes.

The performance of electrocoagulation (EC) using aluminium electrodes as a pretreatment step for the nanofiltration process was compared against a conventional chemical coagulation (CC) process. It is reported that electrocoagulation is superior over the conventional coagulation process with respect to total organic carbon (TOC) and turbidity removal (Mariam & Nghiem, 2010). At the optimum reaction time, TOC and turbidity

removals by the electrocoagulation process were 67% and 80%, respectively. In comparison, at the optimum dosage of $\text{Al}_2(\text{SO}_4)_3$ obtained by a standard jar testing procedure, TOC and turbidity removals by the chemical coagulation process was only 10% and 65%, respectively. The amount of aluminium released by the electrocoagulation process to the solution was significantly higher than the optimum dosage of the chemical coagulation process. Therefore, better performance of the electrocoagulation process can possibly be explained by the higher coagulation concentration and the formation of polymeric aluminium which is known to be more effective for small organic compounds which are prevalent in landfill leachate. A

remarkable difference between electrocoagulation and chemical coagulation pretreatment was also observed with respect to fouling mitigation in a subsequent nanofiltration process. For the two different nanofiltration membranes (NF 270 and SR2) used in this study, severe membrane fouling was evident when filtering raw landfill leachate or chemical coagulation pretreated landfill leachate. In contrast, fouling was not observed with an electrocoagulation pretreated feed solution. However, the use of electrocoagulation pretreatment did not result in any improvement in treated effluent quality by the hybrid system. Over all, there was no discernible variation between the EC – NF and CC – NF hybrid systems in their TOC and turbidity removals, which were approximately 92% and 99%, respectively. The reported results demonstrate that an EC – NF hybrid system can be a promising candidate for landfill leachate treatment, particularly for small and decentralised landfills where simplicity and robustness are required.

In the removal of chemical oxygen demand (COD), total organic carbons (TOC) and colour from municipal solid waste leachate (L1, L2) by means of chemical- and electro-coagulation (EC) methods were carried out. Using aluminium and iron electrodes simultaneously in the reactor the percent of COD, TOC and colour removal versus the electro-chemically generated Fe^{2+} and Al^{3+} doses were examined by the researchers. The best removal has been achieved in the COD parameter. Thus, in electro-coagulation method, the maximal removal is 87% for the L1 solution, while for L2 it is 90% for Fe^{2+} and 77% for L1 and 88% for L2 for Al^{3+} ion solutions. High colour removal for Fe^{2+} is observed for L1 (86%), while for L2 it is approximately 99%. The removal indices for the leachate treatment by chemical coagulants ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) appeared to be lower than for the electrochemically generated Fe^{2+} and Al^{3+} . The results show that electro-coagulation could be considered as an effective alternative solution for the treatment of leachate (Veli et al., 2008).

To examine the feasibility of electrocoagulation (EC) in treating landfill leachate which has low biodegradability and a high level of inorganic compounds, researchers have carried out a test to cut down the organic matter present in the leachate especially the concentration of chemical oxygen demand (COD) which must be less than 120 mg O_2/L . the effects of the operating parameters of EC, such as the current density j (23–95 A/m^2), electrode gap (2–4 cm), and cathode natures were evaluated via COD, turbidity, absorbance at 254 nm, colour,

NH_4^+ , NO_3^- , and electrical energy consumption. The COD objective of the French landfill has been obtained after 135 min of electrocoagulation with the following optimum conditions: $j = 95 \text{ A/m}^2$, 2 cm gap between aluminium electrodes. In this experimental condition, residual turbidity was around 5 NTU and 80% discoloration ratio was acquired. The decrease absorbance at 254 nm showed a removal of humic substances. Nitrate removal was 40% but EC treatment was not appropriate for removing NH_4^+ (Ricordel & Djelal, 2014).

Simultaneous removal of carbonaceous and nitrogenous pollutants under optimal conditions were achieved by Jing Ding (Ding et al., 2018). Ammonia removal was due to the free chlorine generation of EO while organic matter degradation was achieved by both EO and EC processes. Nitrate removal was attributed to both ER and EC processes, with the higher removal achieved by ER process. In a scaled-up reactor, the EO/EC/ER process was able to remove 50–60% organic matter and 100% ammonia at charge of 1.5 Ah/L with energy consumption of 15 kW h/m³. Considering energy cost, the process is more efficient to meet the requirement of organic removal efficiency less than 70%. These results show the feasibility and potential of the EO/EC/ER process as an alternative tertiary treatment to achieve the simultaneous removal of organic matter, ammonia, nitrate, and colour of leachate.

COD and turbidity removal was strongly influenced by the initial pH (C.B. Shivayogimath et al., 2013) (., 2013). The effects of process variables such as pH, applied cell voltage, and operating time were investigated on COD and turbidity removal. The EC process was carried out in a batch reactor of 1 litre capacity and samples of 750 ml were taken out for batch at 05, 10, 15, 20, 25, 30, 35 and 40 minutes of operation. The highest COD and turbidity removal efficiency of 95.8% and 96.6% were obtained at an applied cell voltage of for 9V, 40 minutes of operation.

Different operating conditions were studied by Sushmita De et al., 2019 (Mukherjee, 2020) for the removal of Hg which includes pH, inter-electrode distance, current density and operating time. The results of the batch experiments showed a maximum removal of 82.72% of Hg at the optimum operating conditions of current density of 4.93 mA/cm², electrolysis time of 60 min, pH 7 and inter-electrode distance of 2.5 cm.

2.11 Response Surface Methodology:

Response surface methodology (RSM) is a group of mathematical and statistical methods for empirical model construction. The aim of RSM is to optimize an output variable (response) which is affected by various input variables (independent variables) by carefully designing the experiments. An experiment is a sequence of tests, known as runs, in which the input parameters are changed in order to identify the causes for changes in the output variable. The aim of the RSM is to optimize, find the best set of factor levels to achieve a target. An important feature of RSM is the experiment design, which is usually abbreviated as DoE. The aim of DoE is to select the points where the response should be assessed. Such designs are able to fit a second order equation for predicting the response. Within these equations the quadratic terms model the curvature in the true response function. When within the factor region a maximum or minimum exists, it can be estimated by RSM.

The two most common response surface designs are the central composite design and the Box Behnken design. The central composite design incorporates a fractional factorial of two level and two other kinds of points.

- Points at the centre, where all factor values are at zero or midrange value.
- Axial points, where except one all other factors are set at mid-range or zero and that one factor is set at outer axial values.

The substitute to central composite designs is the Box Behnken design. One distinctive aspect of the Box Behnken design is that each component has only three levels. Each independent variable or component is set at one of three values that are equally spaced, normally coded as -1, 0, +1. The design should be capable to suit a quadratic model, including squared terms and products of two variables.

The response can be presented graphically in the 3-dimensional space (Response surface) or in 2-dimensional contour plots that aid in visualizing the response surface form. The graph is useful for showing the shape of a response surface; hills, valleys and ridges. Contours are

constant response curves drawn in the x_i, x_j plane fixing all other variables. Each contour represents a particular value of the response surface. Such graphs help to analyze the interactive relationships between the input variables and output responses.

Variance analysis (ANOVA) is carried out to assess the interactions between the variables and to calibrate the models according to experimental findings (Mahmoud Zaeri et al., 2012). ANOVA uses conventional standardized terminologies. The sample variance is shown using Eq (2.9).

$$S^2 = (1/n-1) \sum (y_i - \bar{y})^2 \quad \text{Eq (2.9)}$$

Where the divisor is called the degrees of freedom (DF), the summation is known as the sum of squares (SS), the squared terms are deviations from the sample mean and the result is the mean square.

ANOVA confirms the model appropriateness. ANOVA splits the variability among every value into one component that is due to variability among group means (due to the treatment) and another component which is due to variability within the groups (also known as residual variation). Variability within groups is quantified by the sum of squares of the differences between each value and its group mean. It is the residual sum of squares. Variation among groups (due to treatment) is quantified as the sum of the squares of the differences between the group means and the mean of all values in all groups. Adjusted to the size of each group, this becomes the treatment sum of squares. Each sum of squares is correlated with a certain number of degrees of freedom (DF) derived from number of subjects and number of groups, and the mean squares (MS) is determined by dividing the sum of squares by the appropriate number of degrees of freedom. These can be thought of as variances. The square root of the mean square residual can be considered as the standard deviation.

How well the estimated model fits the data can be calculated by the value of R^2 . R^2 is the fraction of the total variance attributable to the group mean differences. It matches the variability between group means and the variability within the groups. The R^2 value is determined from the ANOVA and is equal to the group sum of squares divided by the entire sum of squares. It is a descriptive statistic which quantifies the intensity of the relation between group members and the variable calculated. The R^2 lies in the $[0, 1]$ interval. If R^2 is closer to the 1, the better is the regression equation estimate fitting the sample data (Lenth, 2009).

CHAPTER- 3

RESEARCH GAPS

In the recent years treatment of landfill leachate by nonconventional methods such as electrochemical process has been increasing for its treatment efficiency. Among the electrochemical techniques, the process of electrocoagulation (EC) has largely been employed to eradicate COD, ammonia, heavy metals etc. from municipal and industrial wastewaters. Many researchers have reported that to meet strict quality standards for direct discharge of leachate, an integrated treatment strategy has to be developed. However, solid waste landfill leachate treatment with the aim of eradicating COD and Nitrogen(ammonia) removal has not been studied so much for the Dhapa landfill site in Kolkata.

Hence in this study the efficacy of the process of electrocoagulation using iron and aluminium sacrificial electrodes will be evaluated for COD and Nitrogen(ammonia) removal from Dhapa landfill leachate.

CHAPTER- 4

OBJECTIVES AND SCOPE OF WORK

4.1 Objectives:

Keeping in view the behaviour of leachate, the present study has been formulated with the following main objectives_

- i) Removal efficiency of COD and Nitrogen(ammonia) from leachate by electrocoagulation with cost analysis.
- ii) To study the effect of process variables viz. current density, treatment time and spacing between electrodes.

4.2 Scope of Work:

The scope of the study is fixed after keeping the aspects of the literature review.

- Collected leachate was analyzed with respect to. pertinent chemical properties.
- Laboratory scale reactor was designed.
- Electrocoagulation was performed with varying current density, electrodes, operating time, inter electrode distance and pH.
- Removal efficiency was measured for each case.
- The optimum operational conditions for leachate treatment were determined.

CHAPTER- 5

MATERIALS AND METHODS

5.1 Introduction:

Enhancement of biodegradability of landfill leachate by electrocoagulation has been investigated in this study. The technical feasibility of an integrated process for treating landfill leachate to meet the disposal standards has been studied here.

5.2 Landfill Leachate Sample Collection:

In the present study, leachate from the Dhapa uncontrolled landfill in Kolkata has been used. The Dhapa landfill site was commissioned in the year 1981 and absorbs around 3000 tons of MSW per day. The characteristics of leachate has been determined and has been compared with the Indian leachate discharge standards which are specified in Table 12. All the analyses were processed according to internationally accepted standard methods.

(Dhapa is a locality on the fringes of East Kolkata, India. The area consists of landfill sites where the solid wastes of the city of Kolkata are dumped. "Garbage farming" is encouraged in the landfill sites. More than 40 percent of the green vegetables in the Kolkata markets come from these lands. There are four sectors in Dhapa for dumping garbage that are filled with tonnes of waste per day.)

Table 12 Characteristics of leachate from Dhapa uncontrolled landfill

Parameter	Landfill leachate	Indian leachate discharge standards
pH	-	5.5–9
Conductivity ($\mu\text{S}/\text{cm}$)	-	-
COD (mg/L)	-	250
BOD ₅ (mg/L)	-	30
BOD ₅ /COD	-	-
NH ₃ -N (mg/L)	-	50
Hg (mg/L)	-	0.01
Pb (mg/L)	-	0.10

5.3 Description of Dhapa Dumping Ground:

The Dhapa dumping ground/landfill site is maintained by the Kolkata Municipal Corporation and closely lays in western part of ward No. 57 of KMC administrative boundaries, on the eastern part of Kolkata. The disposal site has been functioning as an open dumping ground and serving for the city of Kolkata since 1987. It has a long history in siting and selecting as a dumping ground. From 1960 to 1980, the dumping ground in Kolkata was shifted four times from Dhapa garage to Kadapara, then Bantala, then P.C Connector (Science city) and finally shifted to Dhapa because of available space to hold increasing quantity of waste every day. But now, the present dumping ground is in a condition that it become unable to hold more waste after 3 to 4 years as the horizontal extension is already filled and vertical height is already reached up to 30m.

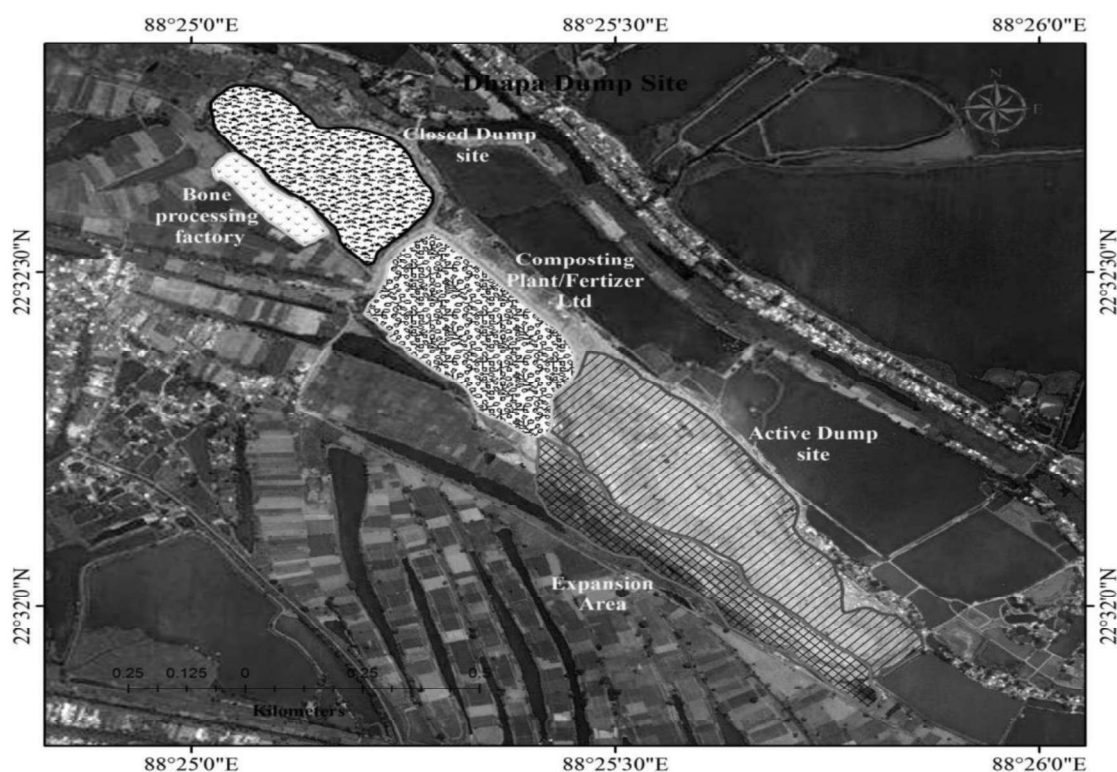


Figure 23: Structural Pattern of Dhapa Dumping Ground

Currently, the disposal site is spread over an area of about 35 hectares. Geometrically, the dumping area is divided into two uneven portions, one is closed dump and another is active dump which is separated by a private composting plant. Out of total 35 ha areas, 12.14 ha area

consists of closed dump which had begun operation during 1987 and was closed in 2009 and the rest about 23 ha areas consists of active dumps which had also commenced operations during the same time and is still now used for disposing waste (Figure 23). 10 ha more areas were added beside active dumping ground in 2010 to cover more waste from the city.

5.4 Location of Dhapa Dumping Ground:

Dhapa dumping ground is located beside the Eastern Metropolitan Bypass (EM bypass), on the eastern side of the city near the “Parama Island,” a small plot in East Kolkata Wetland (EKW). The EM bypass lays on the east, Belegghata/Sealdah on the west, China Town on the south and the Metropolitan Cooperative Housing Society on the north. It is just 4 km from science city. It is roughly located between $22^{\circ} 32' 30.75''$ N to $22^{\circ} 32' 46.56''$ N and $88^{\circ} 25' 1.53''$ E to $88^{\circ} 25' 15.92''$ E. By locational extent, two dumpsites (closed and active) are not contiguous and spaced about 0.5 km separated by composting plant (M/S Eastern Organic Fertilizer) and roads (Figure 24).



Fig. 24: Location of active and closed dumping ground at Dhapa uncontrolled landfill in Kolkata, India

(Source: Google Earth)

5.5 Waste and Leachates: Quantities and Properties:

According to latest census data, KMC has a population of 4.5 million with covering a total area of 200.45 km². Recently KMC has 144 wards which are divided in to 16 boroughs. KMC produces 4500 MT of solid waste per day which is estimated as 0.667 kg/capita. It is observed that Municipal Solid Waste generated from various sources including household, street sweeping, institutional waste, commercial and market waste in the city (Chattopadhyay et al., 2009). The whole generated wastes are collected from primary and secondary storage places; KMC has provided 664 storage places. These whole collected wastes are transported for final disposal. In KMC, there are total 2 disposal sites in KMC at Dhapa and Garden Reach of which Dhapa is the main one, where 95 percent generated wastes are disposed (Hazra & Goel, 2009). In Dhapa dumpsite, mechanized compost was installed in April 2000 with a 700 t/d capacity. MSW carried by trucks is received at the compost plant first; larger sized and non-biodegradable materials are manually separated then prepare for compost and remaining wastes are directly thrown for disposal in Dhapa dumping ground (Chattopadhyay et al., 2009).

Because of high population density and high pressure of floating population, large quantities of waste are generated in the city. On an average, nearly 4200-4500 mt waste are generated daily (D/SWM/KMC). It was 3600 mt 10 years ago, meaning during a 10-year interval about 600-900 mt/d has increased. The data on MSW shows that out of total 4500 mt/d generated waste, household sector or residential area shared highest percentage about 60 percent of total waste generation. The second most waste generating source is commercial refuse by contributing about 16 percent. Along with street sweeping, constructional site, institutional refuse and industrial area also share 11 percent, 5 percent, 4 percent and 4 percent respectively in generating waste. The generated MSW vary with sessional changes. From the variation of collected MSW data, it was found that highest and lowest waste generation session is rainy and winter respectively. During rainy session waste generation rate is higher due to wet weight of waste and the reverse case occurred during winter.

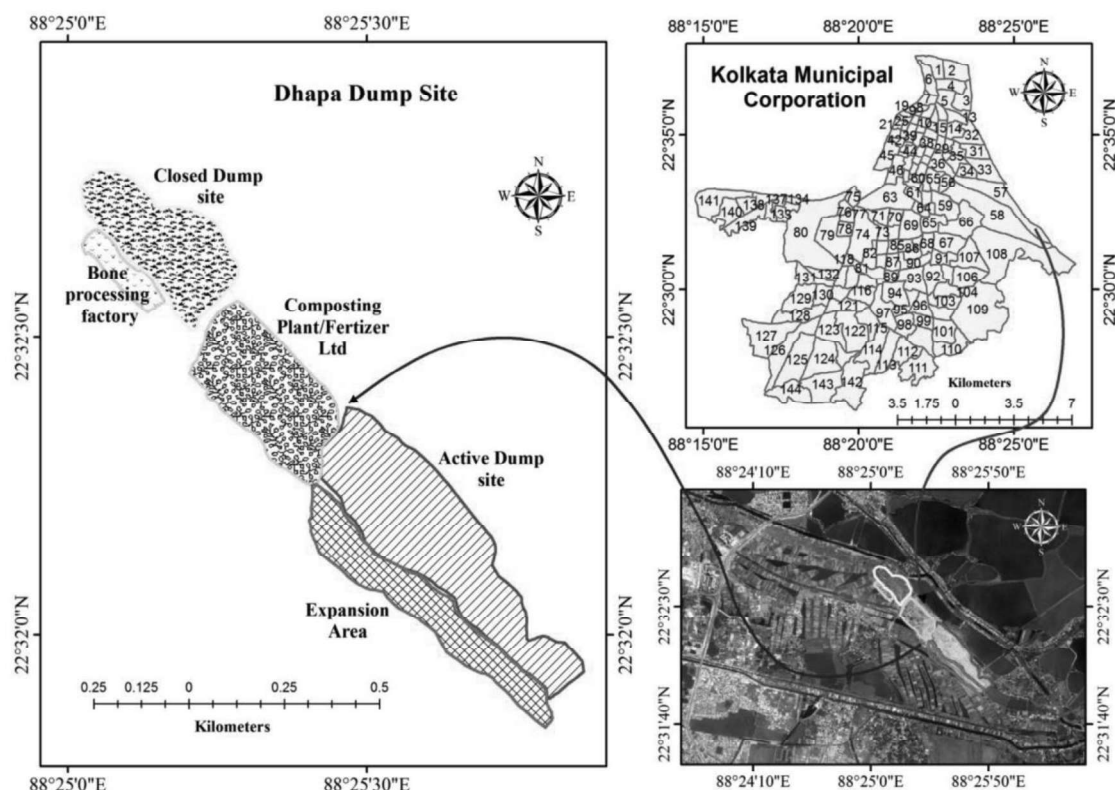


Figure 25: Geographical Location of Dhapa Dumping Site, Kolkata

The waste composition data on MSW was collected from official records at Dhapa. The records revealed that out of total waste disposed here are mixed waste, and out of which maximum come from domestic and residential areas followed by plastic from street and market areas (Figure 26).

The variation in chemical characteristics are found in solid waste is given in Table 13. The quality of normal leachates, tested from the present solid waste disposal site at Dhapa, indicates that concentrations of solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), and chloride are much higher than those permissible for discharge into inland surface water (Mandal, 2007). Similar result was found from Kolkata Environmental Improvement project shown that the concentrations of total solids (TS), BOD, COD, and chromium (Cr) are higher than permissible limit in the canal at the Dhapa area (KEIP, 2005). But the concentration of toxic elements such as arsenic (As), mercury (Hg), lead (Pb), cadmium (Cd), chromium (Cr), copper (Cu), zinc (Zn), nickel (Ni), and fluoride are reported to be slightly lower than the permissible limit to discharge on land (Chattopadhyay et al., 2009).

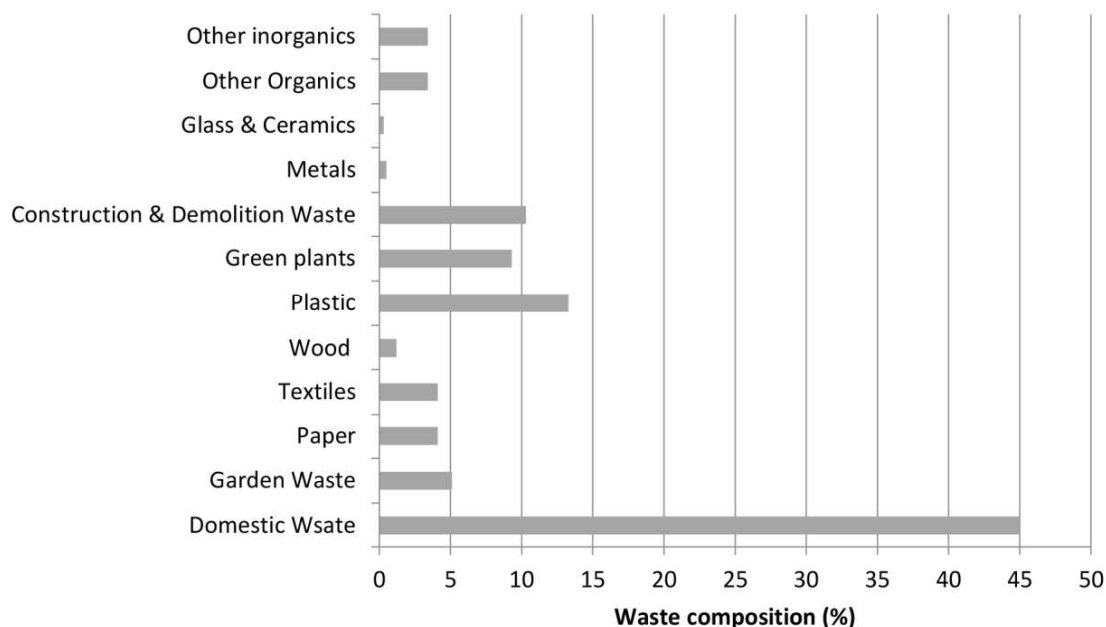


Fig 26: Percentage of Waste Composition in Dhapa Disposal Site

Source: Assessment Report, Dhapa Disposal Site, KMC.

. Waste Composition – Dhapa Disposal Site

Variation of Chemical Characteristics of MSW in Kolkata

Parameters	Measure
Moisture	46.00
pH	8.07
Loss on ignition	38.53
Carbon	22.35
Nitrogen as N	0.76
Phosphorous as P_2O_5	0.77
Potassium as K_2O	0.52
Carbon/Nitrogen (C/N) ratio	31.81

Note: All values are in percent by dry weight basis except pH and C/N ratio.
Source: National Environmental Engineering Research Institute (NEERI), Kolkata

Table 13: Variation of Chemical characteristics of MSW in Kolkata

5.6 Electrode Material:

After going through different kinds of literature, it is found that the best electrode materials till now in terms of removal efficiency for COD & Nitrogen(ammonia) are aluminium and iron. Hence, aluminium and iron electrode were used for the experiment.

5.7 Experimental Set Up:

The electrocoagulation unit has been run in a batch process within a reactor of 10L capacity which has the occupancy of holding a maximum number of 12 electrodes at a time. In this case, the inter-electrode distance would be 2 cm. In the second case, 6 numbers of electrodes were used making the inter-electrode distance 4 cm. In every batch process, only one type of electrode i.e. either iron or aluminium were used.

The dimensions of the electrodes were 21.6 cm (height), 11.7 cm (width) and 1 mm (thickness). The electrocoagulation unit has been run in a batch process with 500ml of raw leachate diluting with 5L of distilled water making a total of 5.5L of sample per run. So, the effective area of the electrodes was (11.7 cm * 11.8 cm) 138.06 cm². Keeping this effective area in mind, all the required values of electric current (DC current) were calculated so that a specific set of current density (viz. 2 mA/cm², 3 mA/cm² etc.) can be send to the reactor. For each type of current density send to the reactor; six numbers of samples were taken from the reactor at the regular time interval of 15 minutes i.e. the total operating time for each type of current density was 90 minutes. This process was repeated for each type of electrodes (for iron and aluminium electrodes) and also for each type of inter-electrode distance (IED) i.e. for 2 cm and 4 cm IED.

The required values of electric current were again calculated in the case of 4 cm IED because of the change in the effective area. (the effective area was changed because the number of electrodes got reduced to 6 from 12)

The optimum conditions for COD and Nitrogen (ammonia) removal have been determined by changing the current density, operating time, and inter-electrode distance by one variable at a

time (OVAT) method. In this method, each single parameter will be once varied while other parameters will be kept constant to optimize each and every parameter.

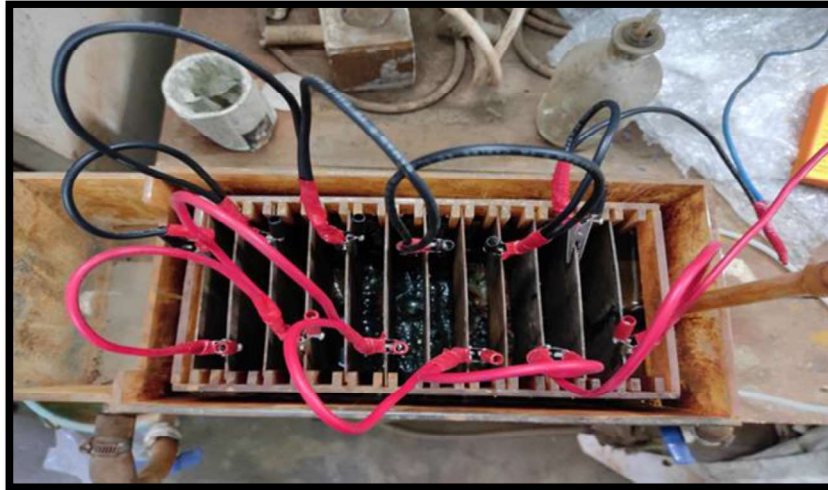


Fig 27: Experimental Set Up (1)



Fig 28: AC to DC Converter Machine

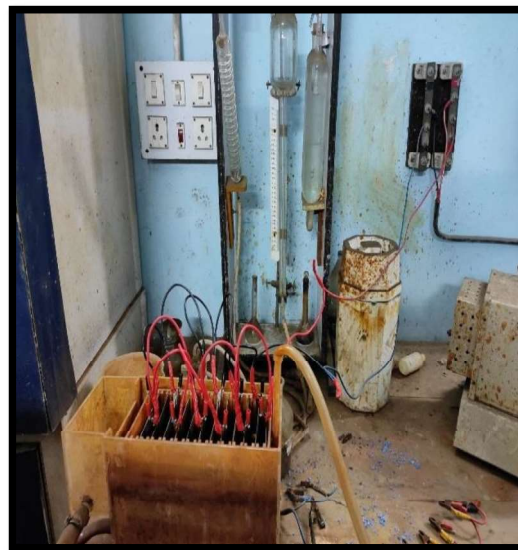


Fig 29: Experimental Set Up (2)

5.8 Analysis of Parameters:

The different parameters (COD, Nitrogen ammonia) have been analysed as per standard procedure (APHA, 2005). The various analytical methods used for analysing the parameters are recorded in table 14.

Table 14: Methods and instruments used for analysing parameters

Sl No.	Parameter	Method	Equipment used
1	COD	Potassium Dichromate Closed Reflux Method	COD apparatus
2	Nitrogen Ammonia	Nessler's Method	Spectrophotometer

The COD was measured by closed reflux, colorimetric method, and NH₃-N was tested by the Nessler's reagent spectrophotometry.

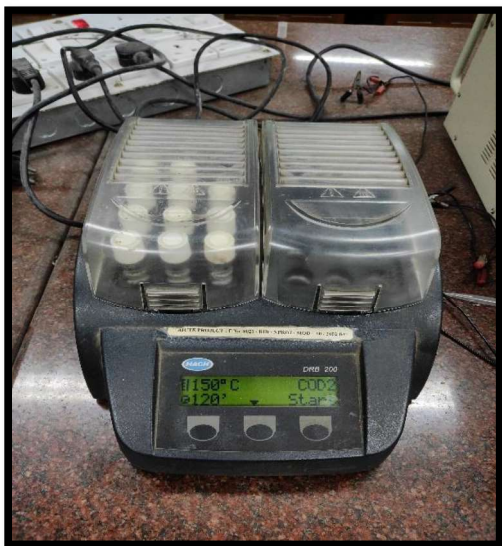


Fig 30: COD Closed Reflux Digestor



Fig 31: Spectrophotometer



Fig 32: pH meter

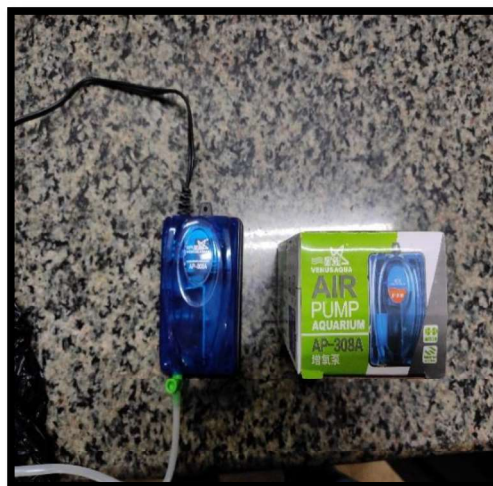


Fig 33: Air Pump

Determination of Chemical Oxygen Demand by Closed Reflux, Colorimetric Method:

General Discussion:

a. Principle: Most types of organic matter are oxidized by a boiling mixture of chromic and sulfuric acids. A sample was refluxed in strongly acid solution with a known excess of potassium dichromate ($\text{K}_2\text{Cr}_2\text{O}_7$). After digestion, the remaining unreduced $\text{K}_2\text{Cr}_2\text{O}_7$ was titrated with ferrous ammonium sulfate to determine the amount of $\text{K}_2\text{Cr}_2\text{O}_7$ consumed and the oxidizable matter is calculated in terms of oxygen equivalent. Keep ratios of reagent weights, volumes, and strengths constant when sample volumes other than 50 mL were used. The standard 2-h reflux time might be reduced if it had been shown that a shorter period yields the same results. Some samples with very low COD or with highly heterogeneous solids content might need to be analyzed in replicate to yield the most reliable data. Results were further enhanced by reacting a maximum quantity of dichromate, provided that some residual dichromate remained. When a sample is digested, the dichromate ion oxidizes the COD material in the sample. This results in the change of chromium from the hexavalent (VI) state to the trivalent (III) state. Both of these chromium species were colored and absorbed in the visible region of the spectrum. The dichromate ion ($\text{Cr}_2\text{O}_7^{2-}$) absorbs strongly in the 400-nm region, where the chromic ion (Cr^{3+}) absorption is much less. The chromic ion absorbs strongly in the 600-nm region, where the dichromate has nearly zero absorption. In 9M sulfuric acid solution, the approximate molar extinction coefficients for these chromium species are as follows: Cr^{3+} - 50 L/mole cm at 604 nm; $\text{Cr}_2\text{O}_7^{2-}$ - 380 L/mole cm at 444 nm; Cr^{3+} - 25 L/mole cm at 426 nm. The Cr^{3+} ion has a minimum in the region of 400 nm. Thus, a working absorption maximum was at 420 nm. For COD values between 100 and 900 mg/Lan, increase in Cr^{3+} in the 600-nm region is determined. Higher values can be obtained by sample dilution. COD values of 90 mg/L or less can be determined by following the decrease in $\text{Cr}_2\text{O}_7^{2-}$ at 420 nm. The corresponding generation of Cr^{3+} gave a small absorption increase at 420 nm, but this was compensated for in the calibration procedure.

b. Interferences and Limitations:

Oxidation of most organic compounds was 95 to 100% of the theoretical value. Pyridine and related compounds resist oxidation and volatile organic compounds reacted in proportion to

their contact with the oxidant. Straight-chain aliphatic compounds were oxidized more effectively in the presence of a silver sulfate catalyst. The most common interferent was the chloride ion. Chloride reacted with silver ion to precipitate silver chloride, and thus inhibited the catalytic activity of silver. Bromide, iodide, and any other reagent that inactivated the silver ion could interfere similarly. Such interferences were negative in that they tend to restrict the oxidizing action of the dichromate ion itself. However, under the rigorous digestion procedures for COD analyses, chloride, bromide, or iodide could react with dichromate to produce the elemental form of the halogen and the chromic ion. Results then were in error on the high side. The difficulties caused by the presence of the chloride could be overcome largely, though not completely, by complexing with mercuric sulfate (HgSO_4) before the refluxing procedure. Although 1 g HgSO_4 was specified for 50 mL sample, a lesser amount might be used where sample chloride concentration was known to be less than 2000 mg/L, as long as a 10:1 weight ratio of $\text{HgSO}_4:\text{Cl}^-$ was maintained. For samples containing more than 2000 mg Cl^-/L were not to be tested. Techniques designed to measure COD in saline waters are available.^{1,2} Halide interferences might be removed by precipitation with silver ion and filtration before digestion. This approach might introduce substantial errors due to the occlusion and carry down of COD matter from heterogeneous samples. Ammonia and its derivatives, in the waste or generated from nitrogen-containing organic matter, were not oxidized. However, elemental chlorine reacted with these compounds. Hence, corrections for chloride interferences were difficult. Nitrite (NO_2^-) exerts a COD of 1.1 mg $\text{O}_2/\text{mg NO}_2^- \text{-N}$. Because concentrations of NO_2^- in waters rarely exceed 1 or 2 mg $\text{NO}_2^- \text{-N/L}$, the interference is considered insignificant and usually was ignored. To eliminate a significant interference due to NO_2^- , add 10 mg sulfamic acid for each mg $\text{NO}_2^- \text{-N}$ present in the sample volume used; add the same amount of sulfamic acid to the reflux vessel containing the distilled water blank. Reduced inorganic species such as ferrous iron, sulfide, manganous manganese, etc., were oxidized quantitatively under the test conditions. For samples containing significant levels of these species, stoichiometric oxidation could be assumed from known initial concentration of the interfering species and corrections can be made to the COD value obtained. The silver, hexavalent chromium, and mercury salts used in the COD determinations create hazardous wastes. The greatest problem is in the use of mercury. If the chloride contribution to COD is negligible, HgSO_4 can be omitted. Smaller sample sizes (see Table 5220:I) reduce the waste. Recovery of the waste material might be feasible if allowed by regulatory authority.³ Volatile organic compounds are more completely oxidized in the closed system because of longer contact with the oxidant. Before each use inspect culture-tube caps for breaks in the TFE liner. Select culture-tube size according to block

heater capacity and degree of sensitivity desired. The 25- _ 150-mm tube were used for samples with low COD content because a larger volume sample could be treated. This procedure was applicable to COD values between 40 and 400 mg/L. Obtain higher values by dilution. Alternatively, use higher concentrations of dichromate digestion solution to determine greater COD values. COD values of 100 mg/L or less could be obtained by using a more dilute dichromate digestion solution or a more dilute FAS titrant. Overall accuracy could be improved by using an FAS titrant which was less than the 0.10M solution specified below. Higher dichromate concentrations or reduced FAS concentrations probably require titrations was to be done in a separate vessel, rather than in the digestion vessel, because of the volumes of titrant required.

c. *Quality control (QC)*: The QC practices considered to be an integral part of each method are summarized in Table 15.

TABLE 5220-I. SAMPLE AND REAGENT QUANTITIES FOR VARIOUS DIGESTION VESSELS

Digestion Vessel	Sample mL	Digestion Solution mL	Sulfuric Acid Reagent mL	Total Final Volume mL
Culture tubes:				
16 × 100 mm	2.50	1.50	3.5	7.5
20 × 150 mm	5.00	3.00	7.0	15.0
25 × 150 mm	10.00	6.00	14.0	30.0
Standard 10-mL ampules	2.50	1.50	3.5	7.5

Table 15: Samples and reagent quantities for various digestion vessels

2. Apparatus:

a. Digestion vessels: Preferably borosilicate culture tubes were used, 16- * 100-mm, 20- * 150-mm, or 25- * 150-mm, with TFE-lined screw caps. Alternatively, borosilicate ampules, 10-mL capacity, 19- to 20-mm diameter were used. Digestion vessels with premixed reagents and other accessories were available from commercial suppliers.

b. Block heater or similar device to operate at 150 _ 2°C, with holes to accommodate digestion vessels. Use of culture tubes probably required the caps to be outside the vessel to protect caps from heat. **CAUTION: The possibility of leaking samples could generate a corrosive and possibly explosive atmosphere, so an oven was not used. Also, culture tube caps might not withstand the 150°C temperature in an oven.**

c. Microburet.

d. Ampule sealer: Only a mechanical sealer was used to ensure strong, consistent seals.

Optical quality of reaction vessels was ensured. Other types of absorption cells with varying path lengths might be used. The extinction coefficients of the ions of interest were used for this approach.

e. Spectrophotometer, for use at 600 nm and/or 420 nm with access opening adapter for ampule or 16-, 20-, or 25-mm tubes. The instrument operates in the region of 420 nm and 600 nm were verified. Values slightly different from these might be found, depending on the spectral bandpass of the instrument.

3. Reagents

a. Digestion solution, high range: 500 mL distilled water was added to 10.216 g $K_2Cr_2O_7$, primary standard grade, previously dried at 150°C for 2 h, 167 mL conc H_2SO_4 , and 33.3 g $HgSO_4$. Then it was dissolved to cool to room temperature, and was diluted to 1000 mL.

b. Digestion solution, low range: was prepared as in ¶ a above, but was used only 1.022 g potassium dichromate.

c. Sulfuric acid reagent: Ag_2SO_4 was added, reagent or technical grade, crystals or powder, to conc. H_2SO_4 at the rate of 5.5 g Ag_2SO_4 /kg H_2SO_4 . It was standed for 1 to 2 d to dissolve. Mix.

d. Sulfamic acid is required only if the interference of nitrites was to be eliminated.

e. Potassium hydrogen phthalate (KHP) standard: $HOOC-C_6H_4-COOK$: was lightly crushed and then was dried to constant weight at 110°C. 425 mg was dissolved in distilled water and was diluted to 1000 mL. KHP had a theoretical COD of 1.176 mg O_2 /mg and this solution had a theoretical COD of 500 μ g O_2 / mL. This solution was stable when refrigerated, but not indefinitely. One had to be alerted for the development of visible biological growth. Solution was prepared and transferred under sterile conditions. Weekly preparation usually was satisfactory.

4. Procedure

a. Treatment of samples: Suitable volume of sample and reagents into tube or ampule as indicated in Table 5220:I were measured. Samples, blank, and one or more standards as

directed in 5220C.4 were prepared, digested and cooled. **The safety precautions were noted.** It was critical that the volume of each component be known and that the total volume be the same for each reaction vessel. If volumetric control was difficult, digested sample were transferred, diluted to a known volume, and read. Premixed reagents in digestion tubes were available commercially.

b. Measurement of Dichromate Reduction: Samples were cooled to room temperature slowly to avoid precipitate formation. Once samples were cooled, vented, if necessary, to relieve any pressure generated during digestion. Contents of reaction vessels were mixed to combine condensed water and dislodge insoluble matter. Suspended matter was let to be settled and was ensured that optical path is clear. Absorption of each sample blank and standard at selected wavelength (420 nm) were measured. A digested blank to confirm good analytical reagents and to determine the blank COD was analysed; blank COD from sample COD was subtracted. Alternately, digested blank as the reference solution was used once it was established that the blank had a low COD.

At 420 nm, reagent water was used as a reference solution. All samples, blanks, and standards against this solution were measured. The absorption measurement of an undigested blank containing dichromate, with reagent water replacing sample, gave initial dichromate absorption. Any digested sample, blank, or standard that had a COD value gave lower absorbance because of the decrease in dichromate ion. A digested blank with reagent water replacing sample was analysed to ensure reagent quality and to determine the reagents' contribution to the decrease in absorbance during a given digestion. The difference between absorbances of a given digested sample and the digested blank was a measure of the sample COD. When standards were run, differences of digested blank absorbance and digested standard absorbance versus COD values for each standard were plotted.

c. Preparation Of Calibration Curve: At least five standards from potassium hydrogen phthalate solution with COD equivalents were prepared to cover each concentration range. Reagent water were made up to volume; same reagent volumes were used, tube, or ampule size, and digestion procedure as for samples. Prepare calibration curve for each new lot of tubes or ampules or when standards prepared in ¶ a above differ by _5% from calibration curve. Curves should be linear. However, some nonlinearity may occur, depending on instrument used and overall accuracy needed.

5. Calculation:

If samples, standards, and blanks are run under same conditions of volume and optical path length, calculate COD as follows:

$$\text{COD as mg O}_2\text{/L} = \frac{\text{mg O}_2 \text{ in final volume} \times 1000}{\text{mL sample}}$$

Preferably analyze samples in duplicate because of small sample size. Samples that are inhomogeneous may require multiple determinations for accurate analysis. These should not differ from their average by more than _5% for the high-level COD test unless the condition of the sample dictates otherwise. In the low-level procedure, results below 25 mg/L may tend to be qualitative rather than quantitative.

energy consumptions were determined using following equations (3.1-3.3):

$$\text{COD reduction efficiency (\%)} = (\text{Ci}-\text{Cf}) / \text{Ci} \times 100 \quad (3.1)$$

$$\text{Energy consumption (Wh)} = \text{V} \times \text{I} \times \text{h} \quad (3.2)$$

$$\text{Energy consumption (KWh/L)} = \text{Wh} / (1000 \times \text{Vol.}) \quad (3.3)$$

where Ci and Cf represent the initial and final concentrations of COD in mg/L.

V (voltage), I (amperage) and h (electrolysis time in hours) and Vol. (volume of solution in litres).

6. Precision and Bias:

Forty-eight synthetic samples containing potassium hydrogen phthalate and NaCl were tested by five laboratories. At an average COD of 193 mg O₂/L in the absence of chloride, the standard deviation was _17 mg O₂/L (coefficient of variation 8.7%). At an average COD of 212 mg O₂/L and 100 mg Cl₂/L, the standard deviation was _20 mg O₂/L (coefficient of variation, 9.6%). Additional QA/QC data for both high- and low-level procedures may be found elsewhere.

CHAPTER- 6

RESULTS & DISCUSSION

The removal efficiency (in %) of COD of the leachate samples with respect to a specific set of current density (mA/cm²), time (minute) and different types of electrodes with different electrode distance are produced below:

Case 1: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=2 cm

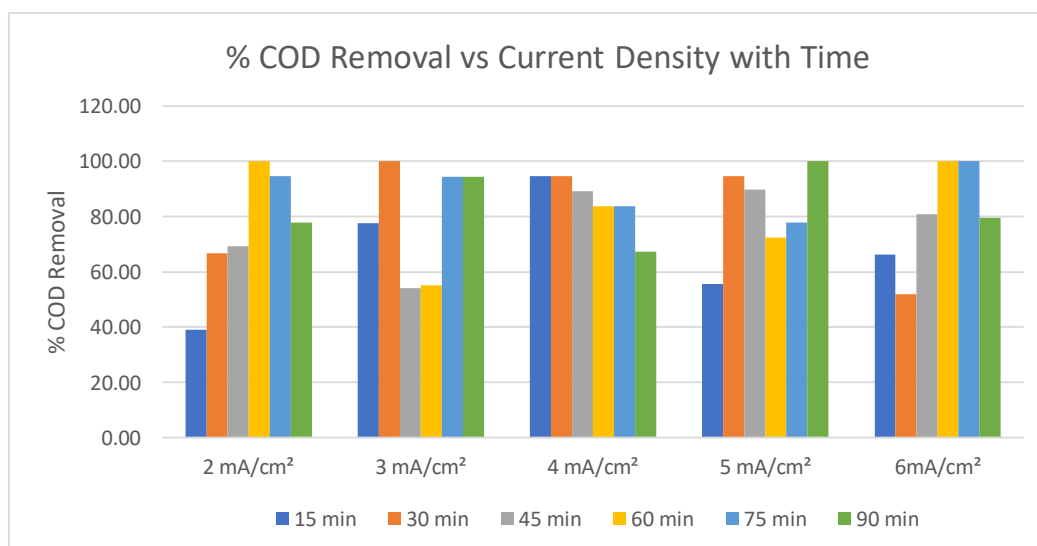
iii) Initial COD= 3104.64 mg/L

Table 16: Experimental data for Iron electrode when IED is 2 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	38.92	77.55	94.56	55.56	66.27
30 min	66.67	100.00	94.56	94.44	51.81
45 min	69.39	54.08	89.12	89.80	80.73
60 min	100.00	55.10	83.67	72.22	100.00
75 min	94.44	94.39	83.67	77.88	100.00
90 min	77.78	94.39	67.35	100.00	79.59

The graphical representation of the above data is plotted below:

Fig 34: Graphical representation of experimental data, Iron electrode, IED=2cm



Case 2: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial COD= 2902.24 mg/L

Table 35: Experimental data for aluminium electrode when IED is 2 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	0.00	70.89	58.82	76.71	100.00
30 min	23.52	88.36	82.35	53.43	94.12
45 min	45.42	65.07	78.17	45.42	45.42
60 min	58.82	76.71	64.71	53.43	94.12
75 min	70.59	65.07	11.76	59.25	94.12
90 min	11.76	88.36	79.89	76.71	94.12

The graphical representation of the above data is plotted below:

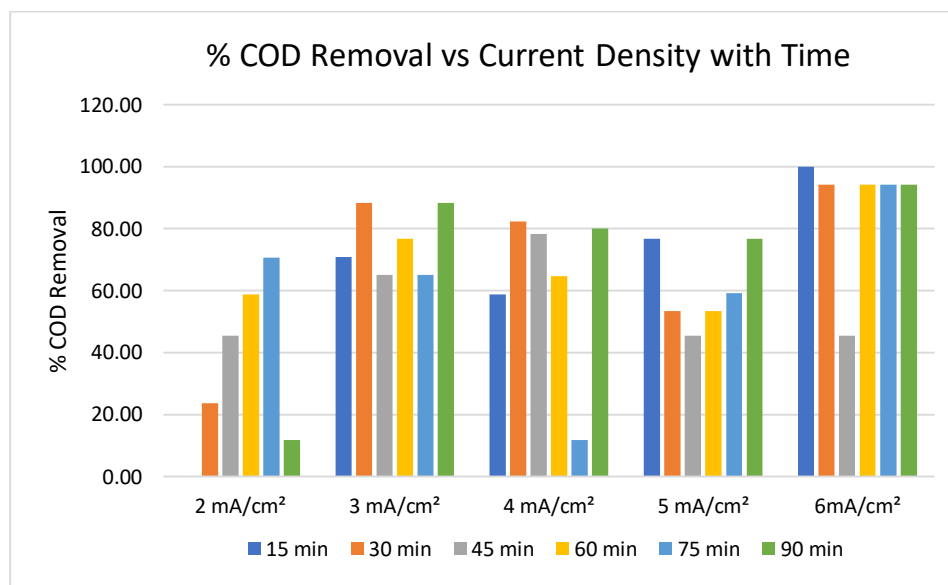


Fig 35: Graphical representation of experimental data, Aluminium electrode, IED=2cm

In the above two cases both the iron and aluminium electrodes were used when the inter electrode distance was 2 cm. In both the cases the COD removal efficiency gradually increases

with the increase in current density and operating time. Then after reaching an optimum point, the COD removal efficiency starts to decrease gradually with the increase of current density and operating time. It can be seen from the figures that on average the iron electrode gives better COD removal performance than the aluminium electrode. The results of the batch experiments showed a maximum COD removal of 100% using iron electrodes at the optimum operating conditions of current density of 3 mA/cm², electrolysis time of 30 minutes while the aluminium electrode also reached 100% removal efficiency but at the cost of 6 mA/cm² of current density with an operating time of 15 min. With the passage of time more and more amount of Fe and Al hydroxides are formed by the oxidation of the anode metal and subsequently which participate in flocculation to remove COD from leachate. When the electrolysis time increases, the concentration of iron ions and their hydroxide flocs increase, also the rate of bubble-generation increases. The pollutants in leachate were removed by the effect of coagulation and flotation. The settleability of particle formed by Fe(OH)₃ is better than that formed by Al(OH)₃. In this study, the iron electrode is regarded as the optimum choice.

Case 3: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial COD= 2902.24 mg/L

Table 18: Experimental data for Iron electrode when IED is 4 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	60.10	69.68	65.80	69.68	60.52
30 min	60.10	69.68	60.10	75.74	49.24
45 min	60.10	57.55	54.40	63.61	4.12
60 min	60.10	51.49	60.10	75.74	43.60
75 min	43.00	39.36	60.10	51.49	37.96
90 min	57.53	63.61	60.10	69.68	60.52

The graphical representation of the above data is plotted below:

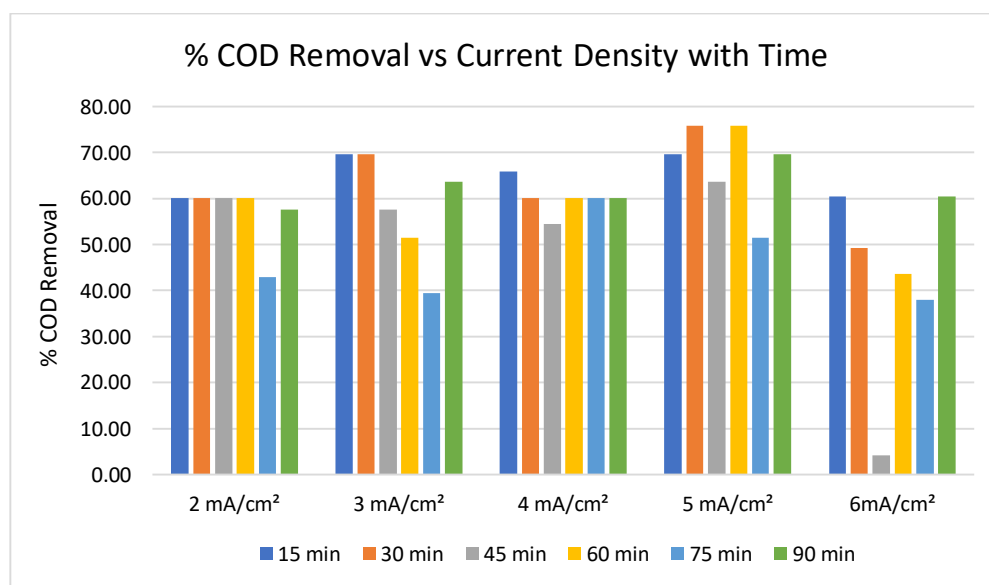


Fig 36: Graphical representation of experimental data, Iron electrode, IED= 4cm

Case 4: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial COD= 2902.24 mg/L

Table 19: Experimental data for Aluminium electrode when IED is 4 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	76.71	65.80	59.67	31.59	59.69
30 min	76.71	88.60	65.43	54.40	53.91
45 min	65.07	54.40	53.91	48.70	65.43
60 min	65.07	71.50	59.67	54.40	65.43
75 min	65.07	54.40	53.91	37.30	71.19
90 min	70.89	48.58	48.15	43.00	65.43

The graphical representation of the above data is plotted below:

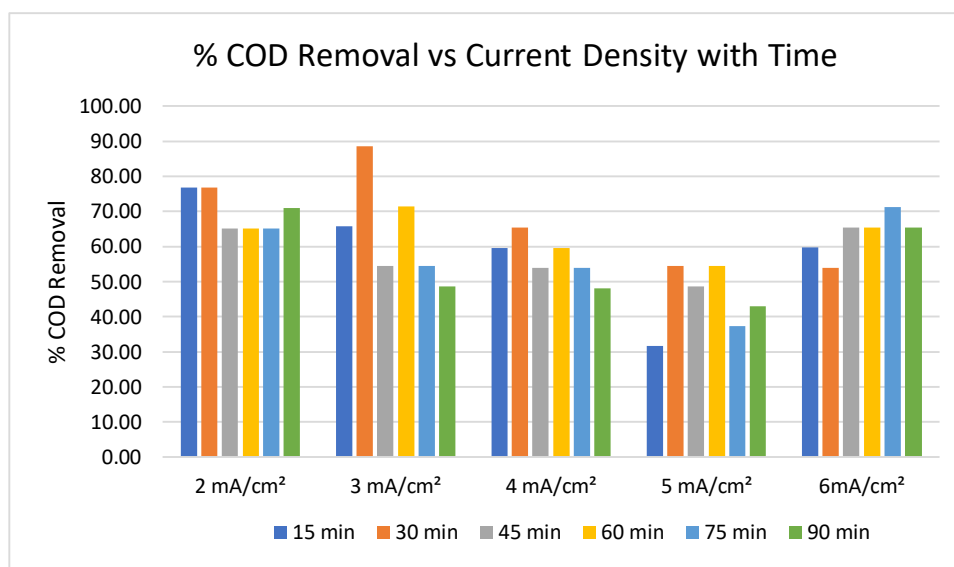


Fig 37: Graphical representation of experimental data, Aluminium electrode, IED= 4cm

In the above two cases (i.e., in Case 3 and Case 4) both the iron and aluminium electrodes were used when the inter electrode distance was 4 cm. It can be clearly seen from the figures that

the COD removal efficiency has fallen down when the inter electrode distance was increased from 2 cm to 4 cm while the other parameters like current density and operating time were same for both types of electrodes. The maximum COD removal for iron electrode was 75.74% when the current density was 5 mA/cm² and operating time was 30 min and maximum COD removal for aluminium electrode was 88.60% when the current density was 3 mA/cm² and operating time was 30 min. So, it can be said that effect of electrostatic field become less intense or effective to remove COD when the inter electrode distance is increased.

The removal efficiency (in %) of $\text{NH}_3\text{-N}$ of the leachate samples with respect to specific set of current density (mA/cm^2), time (minute) and different types of electrodes with different electrode distance are produced below:

Case 1: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial $\text{NH}_3\text{-N}$ = 1517.24 mg/L

Table 20: Experimental data for iron electrode when IED is 2 cm

Electrocoagulation Time	Current Density				
	2 mA/cm^2	3 mA/cm^2	4 mA/cm^2	5 mA/cm^2	6 mA/cm^2
15 min	85.30	96.32	95.22	99.03	40.59
30 min	89.31	48.71	68.66	39.59	93.31
45 min	66.55	43.60	26.66	36.18	48.71
60 min	70.26	29.27	24.56	20.75	83.69
75 min	57.93	35.28	93.52	26.16	17.44
90 min	68.36	28.47	71.97	13.23	5.21

The graphical representation of the above data is plotted below:

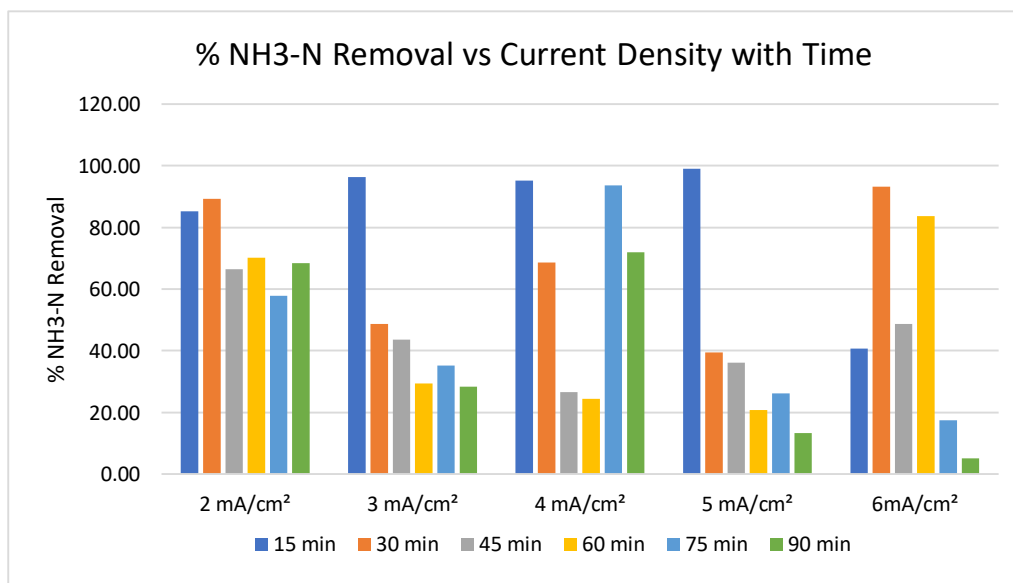


Fig 38: Graphical representation of experimental data, Iron electrode, IED=2cm

In the above case iron electrode was used when the inter electrode distance was 2 cm. It can be clearly seen from the graph that the NH₃-N removal efficiency gradually decreases with the increase of current density and operating time. The maximum NH₃-N removal was found to be 99.03% when the current density was 5 mA/cm² and operating time was 15 min.

Case 2: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial NH₃-N= 1517.24 mg/L

Table 21: Experimental data for aluminium electrode when IED is 2 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	95.72	21.45	42.20	4.11	6.62
30 min	18.34	93.41	16.24	1.80	38.49
45 min	10.42	76.58	50.22	19.14	11.23
60 min	29.37	80.89	41.60	14.23	23.65
75 min	71.36	89.41	6.62	28.67	8.32
90 min	38.79	47.51	0.40	0.80	37.09

The graphical representation of the above data is plotted below:

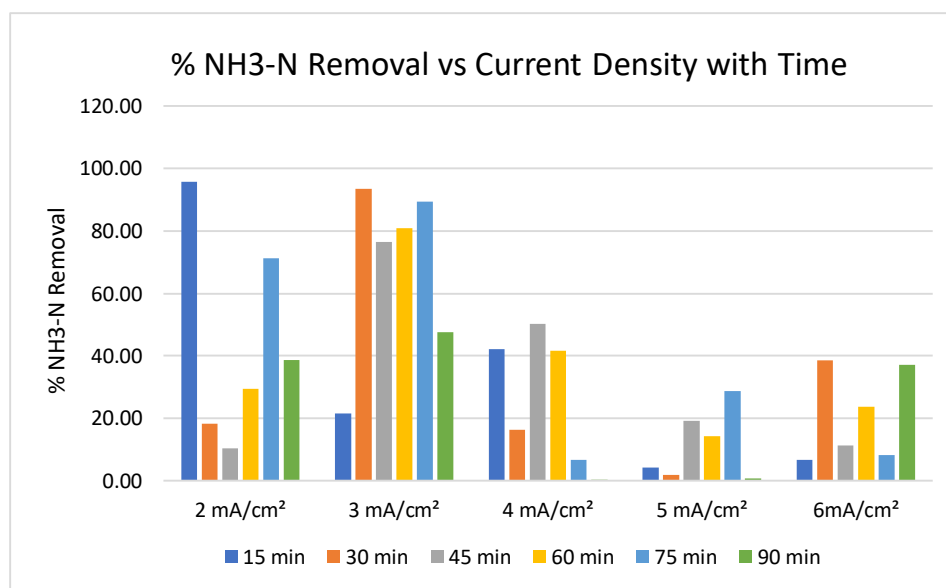


Fig 39: Graphical representation of experimental data, Aluminium electrode, IED=2cm

In the above case aluminium electrode is used when the inter electrode distance was 2 cm. It is hard to identify any special pattern of removal efficiency in relation with different operating parameters. The maximum $\text{NH}_3\text{-N}$ removal was 99.03% when the current density was 5 mA/cm^2 and operating time was 15 min.

Case 3: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial NH₃-N= 1517.24 mg/L

Table 22: Experimental data for iron electrode when IED is 4 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	26.76	98.93	55.23	48.31	54.43
30 min	41.29	99.43	61.44	95.22	70.96
45 min	89.91	49.11	48.61	98.13	62.94
60 min	96.52	48.21	45.60	80.59	98.03
75 min	67.25	99.03	54.73	53.12	58.23
90 min	86.90	44.30	32.27	68.86	62.24

The graphical representation of the above data is plotted below:

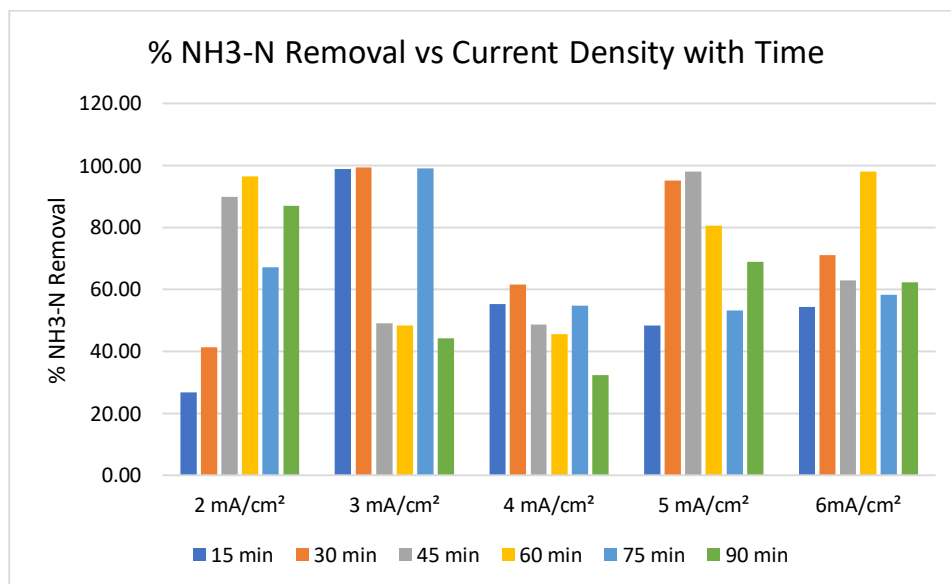


Fig 40: Graphical representation of experimental data, Iron electrode, IED= 4cm

Case 4: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial NH₃-N= 1517.24 mg/L

Table 23: Experimental data for aluminium electrode when IED is 4 cm

Electrocoagulation Time	Current Density				
	2 mA/cm ²	3mA/cm ²	4 mA/cm ²	5 mA/cm ²	6mA/cm ²
15 min	98.63	99.73	99.03	99.53	99.43
30 min	98.93	98.83	36.88	97.12	99.23
45 min	99.83	97.52	99.33	48.71	97.22
60 min	99.43	96.72	59.64	95.82	33.58
75 min	93.31	97.93	24.76	96.62	40.49
90 min	98.83	94.82	98.83	67.05	18.14

The graphical representation of the above data is plotted below:

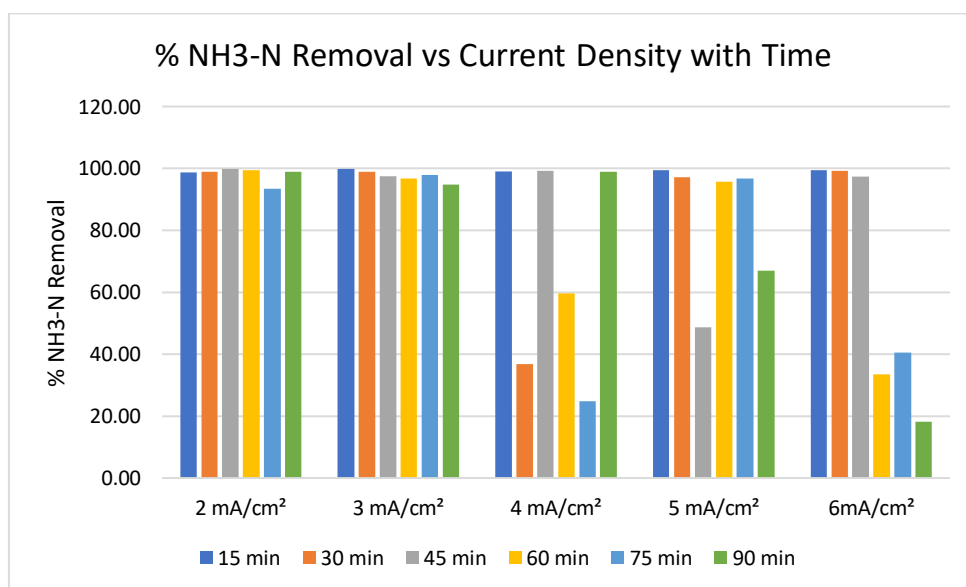


Fig 41: Graphical representation of experimental data, Aluminium electrode, IED= 4cm

In the above two cases both the iron and aluminium electrodes were used when the inter electrode distance was 4 cm. In both the cases the $\text{NH}_3\text{-N}$ removal efficiency is higher than the cases where inter electrode distance was 2 cm. It can be seen from the figures that on average the aluminium electrode gives better $\text{NH}_3\text{-N}$ removal performance than the iron electrode. The results of the batch experiments showed a maximum $\text{NH}_3\text{-N}$ removal of 99.43% using iron electrodes at the optimum operating conditions of current density of 3 mA/cm^2 , electrolysis time of 30 minutes while the aluminium electrode reached 99.83% removal efficiency when the current density was 2 mA/cm^2 with an operating time of 45 min. In this study, the aluminium electrode is regarded as the optimum choice.

At a certain point the removal of $\text{NH}_3\text{-N}$ was hampered by the formation of a metal oxide film on the electrode surfaces. This is because when the operating time was prolonged, the solution was rich in OH^- concentration, leading to a stronger capability of oxygen which reacted on the surface of the electrodes to produce the metal oxide film. The formation of this metal oxide film, commonly known as a passive oxide film, is the drawback when using iron or aluminium as an electrode. Therefore, when the gas bubbles were produced in massive numbers, especially when the plates vibrated, this triggered a thicker metal oxide film to form on the surface of the electrode plates, hence the removal of $\text{NH}_3\text{-N}$ decreased.

Effect of Operating Variables on The Performance of Electrocoagulation Process and Optimizing the Operating Variables Using Response Surface Methodology:

Case 1: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=2 cm

Central composite design: A 3-factor and 3-level CCD was used to optimize the operating parameters of an electrocoagulation process on the responses such as the % COD Removal and % NH₃-N removal efficiency. The total number of experiment combinations was 20, with 6 replications at the design central to determine the pure error. The total number of runs, experimental conditions, response of % COD removal and % NH₃-N removal together with the predicted values are shown in Table 24.

Table 24: CCD Data

Std	Run	Current Density (mA/cm ²) X1	Operating Time (min) X2	Initial pH X3	% COD Removal	% NH ₃ -N Removal
15	1	4	52.5	8	86.4	25.61
6	2	6	15	9	64.27	45.59
19	3	4	52.5	8	86.4	25.61
8	4	6	90	9	77.59	10.21
17	5	4	52.5	8	86.4	25.61
12	6	4	115.567	8	67.35	71.97
13	7	4	52.5	6.31821	91.4	27.61
7	8	2	90	9	75.78	73.36
9	9	0.636414	52.5	8	50	50
3	10	2	90	7	82.78	70.36
1	11	2	15	7	43.92	87.3
4	12	6	90	7	84.59	7.21
11	13	4	-10.5672	8	50	50
2	14	6	15	7	71.27	42.59
14	15	4	52.5	9.68179	84.4	30.61
5	16	2	15	9	36.92	43.92
20	17	4	52.5	8	86.4	25.61
10	18	7.36359	52.5	8	90.36	66.2
16	19	4	52.5	8	86.4	25.61
18	20	4	52.5	8	86.4	25.61

Table 3 Experimental design matrix and response based on the experimental runs on the COD removal (%) and NH₃-N removal (%) proposed by the CCD

Evaluation of experimental results with design of experiments

The % COD removal (Y_1) and % NH₃-N removal (Y_2) are the function of operating parameters such as current density (X_1), operating time (X_2) and initial pH (X_3) at constant. The quadratic model regression equations were obtained from Design Expert Software as shown in following equations_

Final Equation in Terms of Actual Factors

$$\begin{aligned} \text{COD Removal} = & -11.21726 + 22.35421 * (\text{Current Density}) + 1.40021 * (\text{Electrocoagulation} \\ & \text{Time}) + 2.96365 * (\text{Initial pH}) - 0.085133 * (\text{Current Density} * \text{Electrocoagulation Time}) \\ & + 7.18589 * 10^{-15} (\text{Current Density} * \text{Initial pH}) - 3.34977 * 10^{-16} (\text{Electrocoagulation Time} * \\ & \text{Initial pH}) - 1.65805 * (\text{Current Density})^2 - 0.367245 * (\text{Initial pH})^2 \end{aligned}$$

Final Equation in Terms of Actual Factors

$$\begin{aligned} \text{NH}_3\text{-N Removal} = & +295.76942 - 42.69926 * (\text{Current Density}) - 1.60561 * (\text{Electrocoagulation} \\ & \text{Time}) - 30.60473 * (\text{Initial pH}) - 0.138767 * (\text{Current Density} * \text{Electrocoagulation Time}) \\ & + 2.89875 * (\text{Current Density} * \text{Initial pH}) + 0.154600 * (\text{Electrocoagulation Time} * \text{Initial pH}) \\ & + 2.69902 * (\text{Current Density})^2 + 0.008403 * (\text{Electrocoagulation Time})^2 + 0.546578 * (\text{Initial} \\ & \text{pH})^2 \end{aligned}$$

The equation in terms of actual factors can be used to make predictions about the response for given levels of each factor. Here, the levels should be specified in the original units for each factor. This equation should not be used to determine the relative impact of each factor because the coefficients are scaled to accommodate the units of each factor and the intercept is not at the center of the design space.

Experimental data were analyzed by sequential model sum of squares and model summary statistics to obtain the most suitable models among various models such as linear, interactive, quadratic and cubic. The results are tabulated in Tables 4, 5 and 6 for the % color removal, % TOC removal and power consumption, respectively. From Tables 4, 5 and 6, it can be seen that

quadratic model gives the highest R^2 , adjusted R^2 and predicted R^2 values when compared to the other models after excluding the cubic model. The cubic model cannot be used for further modeling of experimental data because it was found to be aliased. An aliased model was a result of insufficient experiments run to independently estimate all the terms of the model. Thus, not all parameters can be estimated and it is unwise for further studying an aliased model. The highest order polynomial from the sequential model sum of squares, quadratic model, was selected for modelling the treatment of landfill leachate using electrocoagulation process where the additional terms are significant and the model is not aliased.

Sequential Model Sum of Squares [Type I]

Response 1: COD Removal

Table 25: SMSS Response1: COD Removal

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Mean vs Total	1.109E+05	1	1.109E+05			
Linear vs Mean	2587.73	3	862.58	4.94	0.0129	
2FI vs Linear	326.15	3	108.72	0.5730	0.6427	
Quadratic vs 2FI	2122.50	3	707.50	20.56	0.0001	Suggested
Cubic vs Quadratic	286.27	4	71.57	7.43	0.0166	Aliased
Residual	57.78	6	9.63			
Total	1.162E+05	20	5812.05			

Fit Summary

Response 1: COD Removal

Table 26: Fit Summary of COD Removal

Source	Sequential p-value	Lack of Fit p-value	Adjusted R^2	Predicted R^2	
Linear	0.0129		0.3836	0.1614	
2FI	0.6427		0.3300	-0.3387	

Quadratic	0.0001		0.8785	0.5148	Suggested
Cubic	0.0166		0.9660	-1.3672	Aliased

Sequential Model Sum of Squares [Type I]

Response 2: NH₃-N Removal

Table 27: SMSS Response1: NH₃-N Removal

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Mean vs Total	34493.99	1	34493.99			
Linear vs Mean	1574.72	3	524.91	1.04	0.4008	
2FI vs Linear	1404.30	3	468.10	0.9144	0.4609	
Quadratic vs 2FI	3392.70	3	1130.90	3.47	0.0588	Suggested
Cubic vs Quadratic	3227.76	4	806.94	141.44	< 0.0001	Aliased
Residual	34.23	6	5.71			
Total	44127.70	20	2206.38			

Fit Summary

Response 2: NH₃-N Removal


Table 28: Fit Summary of NH₃-N Removal

Source	Sequential p-value	Lack of Fit p-value	Adjusted R ²	Predicted R ²	
Linear	0.4008		0.0066	-0.4255	
2FI	0.4609		-0.0096	-1.2730	
Quadratic	0.0588		0.3567	-1.6160	Suggested
Cubic	< 0.0001		0.9887	0.2168	Aliased

Experimental versus predicted:

COD Removal

Color points by value of
COD Removal:

36.92  91.44

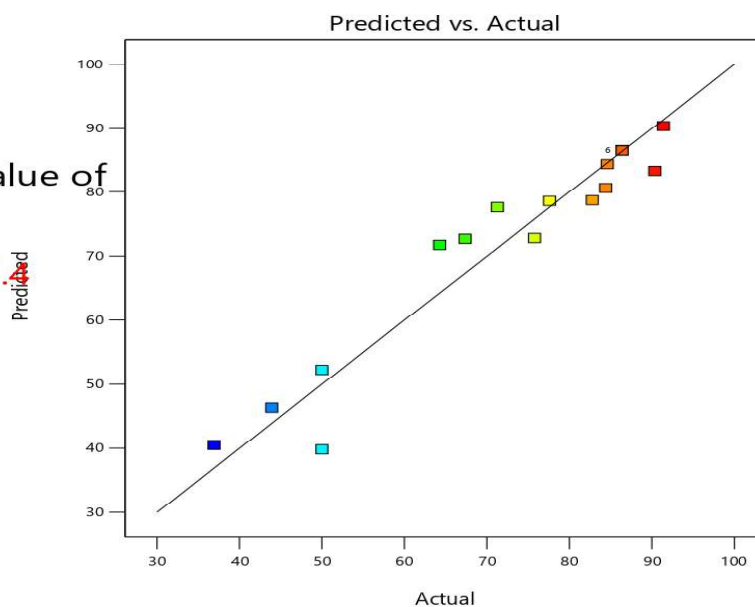



Fig 42: Predicted vs Actual COD Removal graph in Design Expert Software

NH3-N Removal

Color points by value of
NH3-N Removal:

7.21  87.3

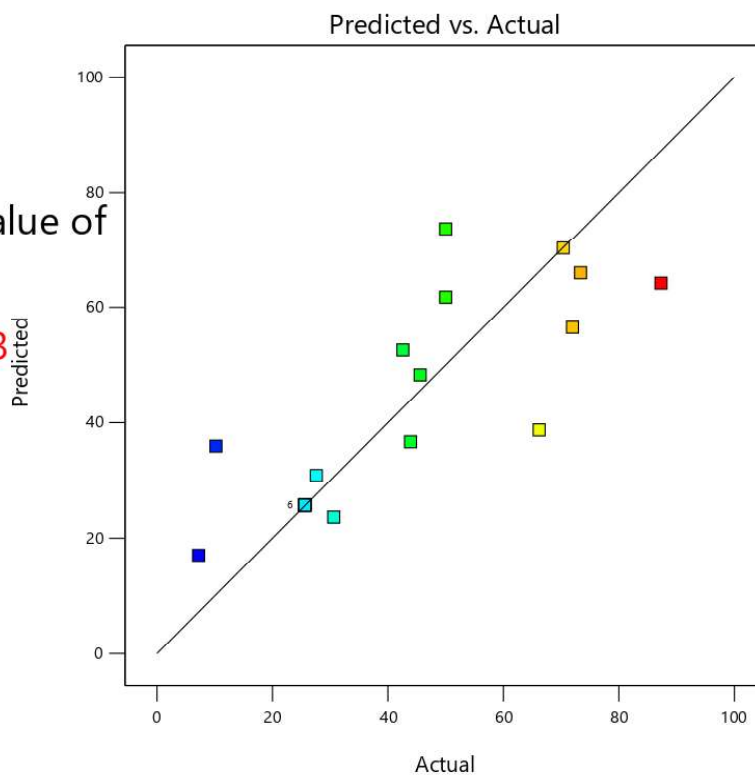


Fig 43: Predicted vs Actual NH3-N Removal graph in Design Expert Software

Optimization:

The main objective of this study is to determine the optimal operating parameters for the maximum % COD and % NH₃-N removal with the minimum of power consumption from landfill leachate wastewater using the electrocoagulation process. The results were optimized using the regression equation of RSM based on CCD. While optimizing, all the input variables such as current density (X_1), electrocoagulation/operating time (X_2) and initial pH (X_3) were selected as within the range while the output variables such as % COD removal and % NH₃-N removal were maximized.

The optimized operating parameters are as following:

When the inter electrode distance is 2 cm and the electrode material is iron.

current density (X_1)—2 mA/cm², electrocoagulation/operating time (X_2)—90 min and initial pH (X_3)—7.00 with the result of COD removal to be 78.673%, NH₃-N removal of 70.274%.

C

Factor Coding: Actual

COD Removal (%)

Design Points:

● Above Surface

○ Below Surface

36.92 91.4

X1 = A

X2 = B

Actual Factor

C = 7

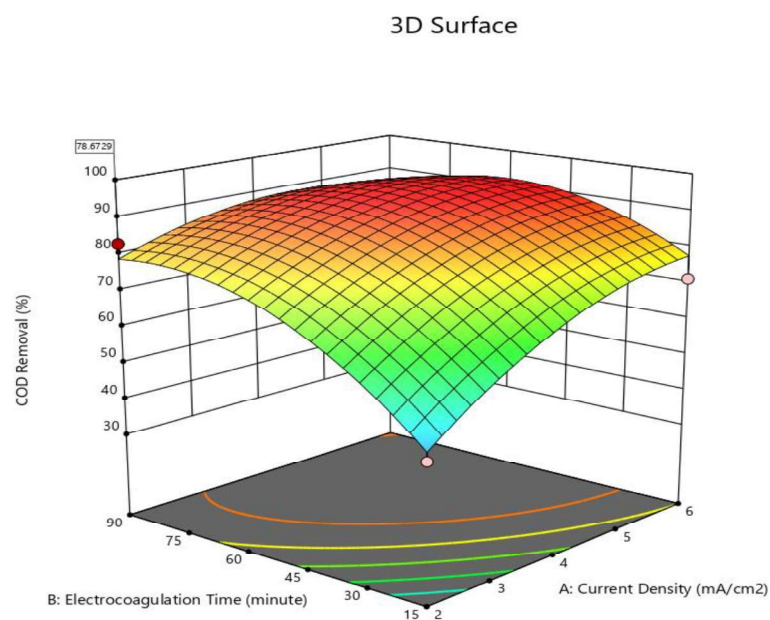


Fig 44: Combined effect of electrocoagulation time and current density on COD removal

Factor Coding: Actual

NH₃-N Removal (%)

Design Points:

● Above Surface

○ Below Surface

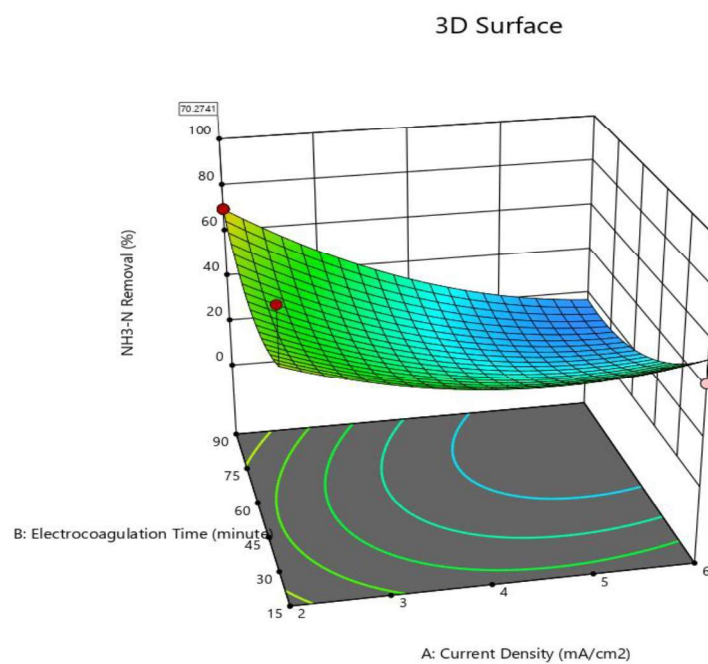
7.21 87.3

X1 = A

X2 = B

Actual Factor

C = 7

Fig 45: Combined effect of electrocoagulation time and current density on NH₃-N removal

The optimized operating parameters are as following:

When the inter electrode distance is 2 cm and the electrode material is aluminium.

current density (X_1)—6 mA/cm², electrocoagulation/operating time (X_2)—90 min and initial pH (X_3)—9.00 with the result of COD removal to be 91.484%, NH₃-N removal of 44.302%.

Factor Coding: Actual

COD Removal (%)

Design Points:

● Above Surface

○ Below Surface

3.1 98

$X_1 = A$

$X_2 = B$

Actual Factor

$C = 9$

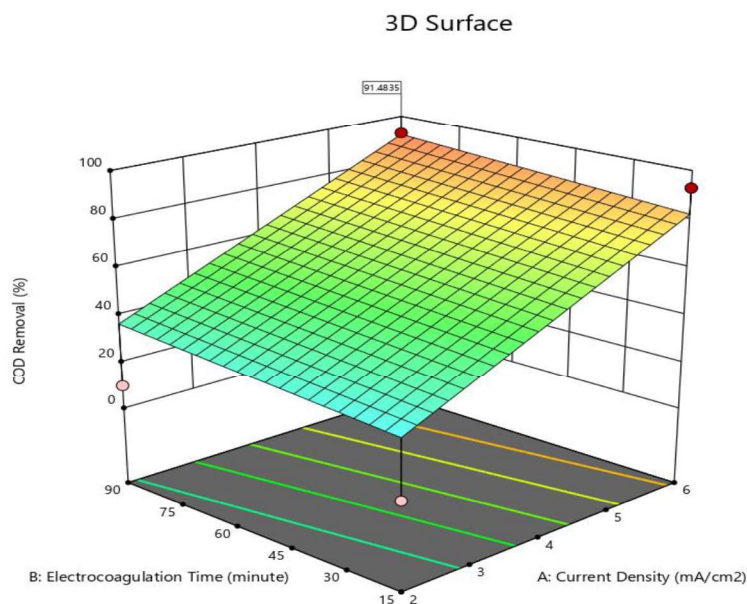


Fig 46: Combined effect of electrocoagulation time and current density on COD removal

Factor Coding: Actual

NH₃-N Removal (%)

Design Points:

● Above Surface

○ Below Surface

5.4 98.72

$X_1 = A$

$X_2 = B$

Actual Factor

$C = 9$

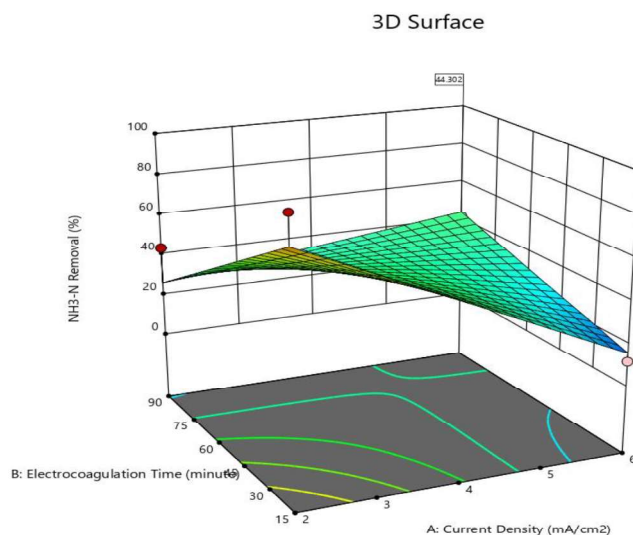


Fig 47: Combined effect of electrocoagulation time and current density on NH₃-N removal

The optimized operating parameters are as following:

When the inter electrode distance is 4 cm and the electrode material is iron.

current density (X_1)—2 mA/cm², electrocoagulation/operating time (X_2)—90 min and initial pH (X_3)—7.00 with the result of COD removal to be 65.163%, NH₃-N removal of 81.551%.

Factor Coding: Actual

COD Removal (%)

Design Points:

● Above Surface

○ Below Surface

23.86 65.52

$X_1 = A$

$X_2 = B$

Actual Factor

$C = 7$

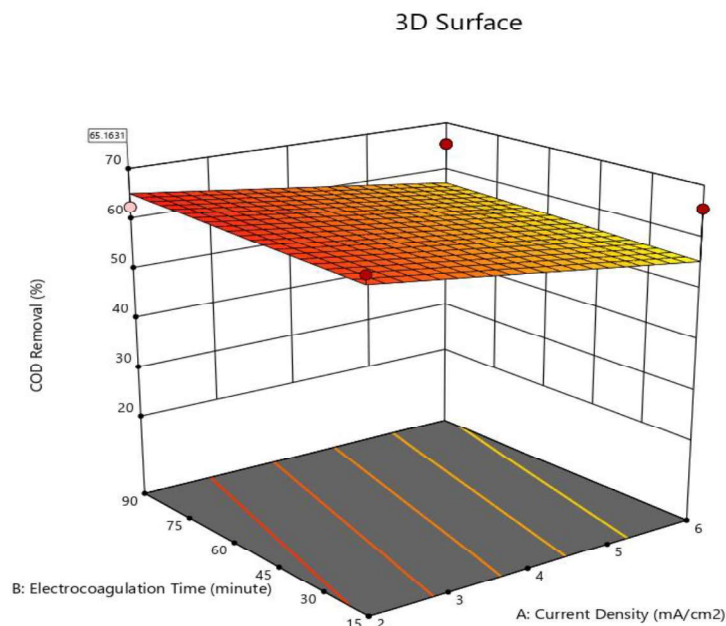


Fig 48: Combined effect of electrocoagulation time and current density on COD removal

Factor Coding: Actual

NH₃-N Removal (%)

Design Points:

● Above Surface

○ Below Surface

28.76 91.9

$X_1 = A$

$X_2 = B$

Actual Factor

$C = 7$

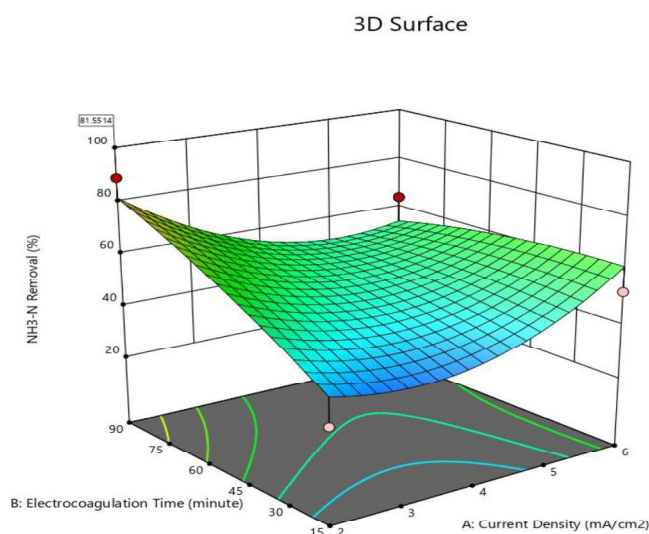


Fig 49: Combined effect of electrocoagulation time and current density on NH₃-N removal

The optimized operating parameters are as following:

When the inter electrode distance is 4 cm and the electrode material is aluminium.

current density (X_1)—2 mA/cm², electrocoagulation/operating time (X_2)—20.683 min and initial pH (X_3)—7.00 with the result of COD removal to be 72.694%, NH₃-N removal of 86.025%.

Factor Coding: Actual

COD Removal (%)
 ● Design Points
 48.15 81.71

$X_1 = A$

$X_2 = B$

Actual Factor
 $C = 7$

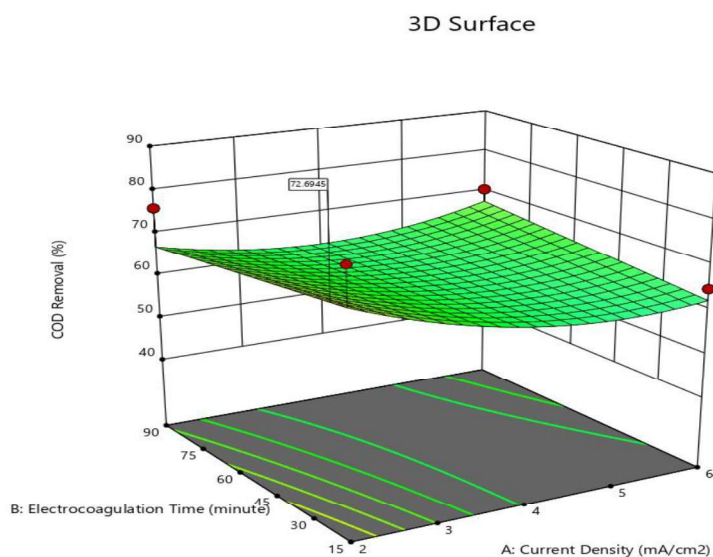


Fig 50: Combined effect of electrocoagulation time and current density on COD removal

Factor Coding: Actual

NH₃-N Removal (%)
 Design Points:
 ● Above Surface
 ○ Below Surface
 20.14 99.43

$X_1 = A$

$X_2 = B$

Actual Factor
 $C = 7$

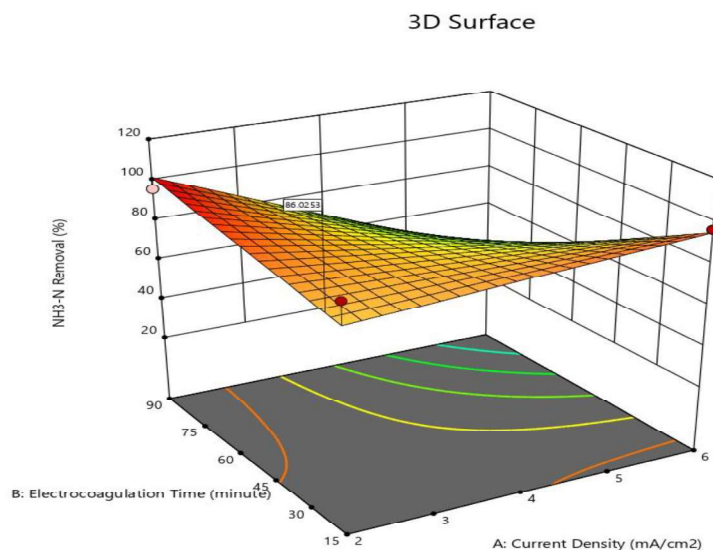


Fig 51: Combined effect of electrocoagulation time and current density on NH₃-N removal

The combined effect of current density (X_1) and inter-electrode distance on % COD and % $\text{NH}_3\text{-N}$ removal with power consumption was tested by varying X_1 from 2 to 6 mA/cm^2 and IED from 2 to 4 cm, it can be observed that the % COD removal and % $\text{NH}_3\text{-N}$ removal were increased as the current density increased, but after the optimum value, further increase in current density does not help in improving the removal of % COD and % $\text{NH}_3\text{-N}$. The increase in current density resulted in the production of large amount of Fe^{2+} and Al^{3+} ions via anodic metal dissolution, more H_2 bubbles was formed at the cathode, which are profitable for the separation or flotation process (Ozyonar and Karagozoglu 2015). An increase in current density caused an increase in cell voltage, which had a direct impact on the power consumption of the electrochemical process. Since a proportional relationship was established between the current density and power consumption, it is necessary to identify the optimum value of current density to reduce the power consumption and operating cost (Heidmann and Calmano 2008).

Inter-electrode distance was varied from 2 to 4 cm in order to study its effect on the % COD removal, % $\text{NH}_3\text{-N}$ removal. It was observed that the % COD removal was decreased as the inter-electrode distance increased from 2 to 4 cm at any value of current density in the range of 2-6 mA/cm^2 for iron and aluminium electrode but the % $\text{NH}_3\text{-N}$ removal was increased as the inter-electrode distance increased from 2 to 4 cm at any value of current density in the range of 2-6 mA/cm^2 . This is because there is an increased in ohmic voltage drop as the distance between the anode and cathode was increased (Khandegar and Saroha 2013). Besides, Faraday's law also stated that the amount of oxidized metal decreased as the gap between the electrodes was increased. However, the power consumption increased as the inter-electrode distance increased. This was due to the fact that there is more resistance offered when the electrodes gap increase and power consumption is directly proportional to the cell voltage (Ricordel and Djelal 2014).

Cost Analysis:

Energy consumptions were determined using following equations (3.1-3.3):

$$\text{COD reduction efficiency (\%)} = (C_i - C_f) / C_i \times 100 \quad (3.1)$$

$$\text{Energy consumption (Wh)} = V \times I \times h \quad (3.2)$$

$$\text{Energy consumption (KWh/L)} = \text{Wh} / (1000 \times \text{Vol.}) \quad (3.3)$$

where C_i and C_f represent the initial and final concentrations of COD in mg/L.

V (voltage), I (amperage) and h (electrolysis time in hours) and Vol. (volume of solution in litres).

For COD removal:

Case 1: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial COD= 3104.64 mg/L

The maximum removal of COD in this case was found to be 100% when the current density was 3mA/cm² and electrocoagulation time was 30 minutes.

So, the energy consumption in this removal process is = $(3.1 \times 9 \times .5) / (1000 \times 5.5)$ KWh/L
 $= 2.54 \times 10^{-3}$ KWh/L

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.5 h

Energy consumed = $(3 \times .5) / (1000 \times 5.5)$ KWh/L = 2.73×10^{-4} KWh/L

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 2: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial COD= 2902.24 mg/L

The maximum removal of COD in this case was found to be 100% when the current density was 6mA/cm² and electrocoagulation time was 15 minutes.

So, the energy consumption in this removal process is = $(4.7*18*.25)/(1000*5.5)$ KWh/L
 $= 3.84*10^{-3}$ KWh/L

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 15 min = 0.25 h

Energy consumed = $(3*.25)/(1000*5.5)$ KWh/L = $1.36*10^{-4}$ KWh/L

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 3: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial COD= 2902.24 mg/L

The maximum removal of COD in this case was found to be 75.74% when the current density was 5mA/cm² and electrocoagulation time was 30 minutes.

So, the energy consumption in this removal process is = $(2.96*6.8*.5)/(1000*5.5)$ KWh/L
 $= 1.83*10^{-3}$ KWh/L

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.5 h

Energy consumed = $(3*.5)/(1000*5.5)$ KWh/L = $2.73*10^{-4}$ KWh/L

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 4: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial COD= 2902.24 mg/L

The maximum removal of COD in this case was found to be 88.6% when the current density was 3mA/cm² and electrocoagulation time was 30 minutes.

So, the energy consumption in this removal process is = $(2.35*4.1*.5)/(1000*5.5)$ KWh/L
 $= 8.76*10^{-4}$ KWh/L

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.5 h

Energy consumed = $(3 \times 0.5) / (1000 \times 5.5)$ KWh/L = 2.73×10^{-4} KWh/L

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

For NH₃-N removal:

Case 1: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial NH₃-N= 1517.24 mg/L

The maximum removal of NH₃-N in this case was found to be 99.03% when the current density was 5mA/cm² and electrocoagulation time was 15 minutes.

So, the energy consumption in this removal process is = $(4.6 \times 15 \times 0.25) / (1000 \times 5.5)$ KWh/L
 $= 3.14 \times 10^{-3}$ KWh/L

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 15 min = 0.25 h

Energy consumed = $(3 \times 0.25) / (1000 \times 5.5)$ KWh/L = 1.36×10^{-4} KWh/L

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 2: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=2 cm

iii) Initial NH₃-N= 1517.24 mg/L

The maximum removal of NH₃-N in this case was found to be 95.72% when the current density was 2mA/cm² and electrocoagulation time was 15 minutes.

So, the energy consumption in this removal process is = $(2.46 \times 15 \times 0.25) / (1000 \times 5.5)$ KWh/L

$$= 6.71 \times 10^{-4} \text{ KWh/L}$$

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.25 h

$$\text{Energy consumed} = (3 \times 0.25) / (1000 \times 5.5) \text{ KWh/L} = 1.36 \times 10^{-4} \text{ KWh/L}$$

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 3: i) Electrode type: IRON

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial $\text{NH}_3\text{-N}$ = 1517.24 mg/L

The maximum removal of $\text{NH}_3\text{-N}$ in this case was found to be 99.43% when the current density was 3mA/cm^2 and electrocoagulation time was 30 minutes.

So, the energy consumption in this removal process is = $(2.13 \times 4.1 \times 0.5) / (1000 \times 5.5) \text{ KWh/L}$

$$= 7.94 \times 10^{-4} \text{ KWh/L}$$

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.5 h

$$\text{Energy consumed} = (3 \times 0.5) / (1000 \times 5.5) \text{ KWh/L} = 2.73 \times 10^{-4} \text{ KWh/L}$$

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

Case 4: i) Electrode type: ALUMINIUM

ii) Inter Electrode Distance (IED)=4 cm

iii) Initial $\text{NH}_3\text{-N}$ = 1517.24 mg/L

The maximum removal of $\text{NH}_3\text{-N}$ in this case was found to be 99.83% when the current density was $2\text{mA}/\text{cm}^2$ and electrocoagulation time was 45 minutes.

So, the energy consumption in this removal process is $= (2.00 \times 2.7 \times .75) / (1000 \times 5.5) \text{ KWh/L}$

$$= 7.36 \times 10^{-4} \text{ KWh/L}$$

Additional cost of air pump: i) Power of air pump = 3W

ii) Run time = 30 min = 0.75 h

$$\text{Energy consumed} = (3 \times .75) / (1000 \times 5.5) \text{ KWh/L} = 4.09 \times 10^{-4} \text{ KWh/L}$$

Now if we multiply the above two values with the per unit rate of electricity then we will get the actual cost of the electrocoagulation process.

CHAPTER- 7

CONCLUSION

Electrocoagulation can conclusively be applied to treat COD and $\text{NH}_3\text{-N}$ of landfill leachate. This study clearly indicates that COD removal can be observed at the optimal conditions of the different experimental parameters of 3 mA/cm² of current density, 30 min of electrocoagulation time and 2 cm of inter-electrode distance. The maximum removal percentage of COD was observed to be 100% using iron electrodes. Whereas in the case of $\text{NH}_3\text{-N}$ removal, it was observed that the optimal conditions of the different experimental parameters are 2 mA/cm² of current density, 45 min of electrocoagulation time and 4 cm of inter-electrode distance. The maximum removal percentage of $\text{NH}_3\text{-N}$ was observed to be 99.83% using aluminium electrodes. The results distinctly demonstrate that COD and $\text{NH}_3\text{-N}$ removal were satisfactory and electrocoagulation can successfully be applied in combination with other physicochemical methods to bring down the concentration of COD and $\text{NH}_3\text{-N}$ within the stipulated leachate discharge standard of India.

CHAPTER- 8

FUTURE SCOPE OF WORK

1. A proper collection system for the landfill leachate generated at the MSW disposal site can be designed.
2. Impact assessment of the landfill leachate on the surrounding environment and people can be investigated.
3. Experiments may be conducted by mixing landfill leachate with highly biodegradable wastewater like Municipal sewage, Dairy wastewater, etc. and treatment efficiency and cost estimation can be studied.
4. Efficiency on the removal of different heavy metals from the leachate by the electrocoagulation method can be examined.
5. An integrated combination of different leachate treatment methods can be designed and further the removal efficiency to be checked with cost estimation.

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