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Review

An overview of landfill leachate treatment via activated carbon adsorption process

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ABSTRACT

Water scarcity and pollution rank equal to climate change as the most urgent environmental issue for the 21st century. To date, the percolation landfill leachate into the groundwater tables and aquifer systems which poses a potential risk and potential hazards towards the public health and ecosystems, remains an aesthetic concern and consideration abroad the nations. Arising from the steep enrichment of globalization and metropolitan growth, numerous mitigating approaches and imperative technologies have currently drastically been addressed and confronted. Confirming the assertion, this paper presents a state of art review of leachate treatment technologies, its fundamental background studies, and environmental implications. Moreover, the key advance of activated carbons adsorption, its major challenges together with the future expectation are summarized and discussed. Conclusively, the expanding of activated carbons adsorption represents a potentially viable and powerful tool, leading to the superior improvement of environmental conservation.

Contents

1. Introduction

Concern about environmental protection has increased over the years from a global viewpoint. During the past several decades, the exponential population and social civilization growth, changes in the productivity and consumption habits, increasingly affluent lifestyles and resources use, and continuing development of the industrial and technologies has been accompanied by the rapid generation of municipal and industrial solid wastes, which create the most intransigent paradox around the world [1,2]. In 1994, the global municipal solid waste production rate was recorded at 1.3 billion tonnes per day, or equivalent to an average of two-thirds of a kilogram per capita per day (10 times per capital body weight per year) [3], which in 2008, the figure has risen by 31.1%, designated an generation rate of 1.7 billion tonnes per day [4].

Of major interest, sanitary landfilling is recognized as the most common and desirable integral indispensable solid waste management strategy for sustainable disposal and elimination of residue wastes from separation, recycling and incineration, both in fully industrialized and developing countries [5], in terms of its simplicity, as well as the low exploitation and capital costs, accounting approximately 95% of the total municipal solid waste collected worldwide [6]. By nature, sanitary landfill is a physically, chemically and biologically complex heterogeneous system [7], which underlying the hydrological conditions, refuse composition and compaction, temperature, moisture content along with the seasonal variations as its key functions [8]. In spite of various exploitations, emphasis and researches have been proliferated,

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such implementations are handicapped by the inherent drawbacks of the extensive emissions of highly variable quantity and quality of landfill leachates, which enriched in numerous organic, inorganic, ammonium and toxic constituents [9], resulting in the threatening of surrounding soil, groundwater, and surface water [10].

Simultaneously, the enforcement of environmental rules and regulations concerning the monitoring of contaminants from leachate waste streams by regulatory agencies are becoming more stringent and restrictive, inevitably affect the design, planning, and operation of the sanitary landfills [11]. This has prompted a growing research interest in establishing a leading selective, reliable and durable alternative for the treatment of heavily polluted leachates. Of late, a wide variety of scientific publications covering the collection, storage and appropriate treatment of the highly contaminated landfill leachates or its manifestations have currently been exerted [1]. With the above aforementioned, this bibliographic review attempts to summarize the origin, properties, and environmental impacts of the sanitary landfill leachate. The present work is aimed at providing a concise and up to date picture of the present status of the leachate treatment technologies. The comprehensive literature together with the challenges and future perspectives has been highlighted and outlined, to familiarize the knowledge deficiencies regarding leachate treatment via activated carbon adsorption technology.

2. Landfill leachate

2.1. Definition and background studies

In general, landfill leachate is defined as any contaminated liquid effluent percolating through deposited waste and emitted within a landfill or dump site through external sources [12], of which its route of exposure and toxicity often remains unknown [13]. More precisely, it is a soluble organic and mineral compound formed when water infiltrates into the refuse layers, extracts a series of contaminants and instigates a complex interplay between the hydrological and biogeochemical reactions that acts as a mass transfer mechanisms for producing of moisture content sufficiently high to initiate the liquid flow [14], induced by the gravity force, precipitation, irrigation, surface runoff, rainfall, snowmelt, recirculation, liquid waste co-disposal, refuse decomposition, groundwater intrusion and initial moisture content present within the landfills [4].

Under normal conditions, leachate migrates down through the pores within the waste mass, and in modern containment landfills, it drains away in the engineered drainage layer, collected at the lowest point in a sump or storage reservoir [15]. Hereby, the potentiality of formation can be assessed by a water balance which involves the amounts of water entering the landfill, consumed in the biochemical reactions, and quantity leaving as water vapour [16]. Dating back to prehistoric times, the early concept regarding solid waste management has been initiated by the Indian cities with the construction of brick drains during the 3500 B.C. (before century), which in 1900 B.C., the installation of a sewage disposal system (including water closets) in the ancient city of Knossos, and introduction of regulations against littering in the Roman Empire has been witnessed [17].

Meanwhile, in the 1920s, the first household waste collection system was pioneered in an apartment house in Stockholm, with the invention of vehicles equipped specially for waste transportation. Until 1959, the first technical municipal solid waste disposal guidelines have been published [18], of which the controlled disposal of municipal solid waste was recognized as an activity falling within the sphere of the civil engineers in the early of 1930s [19]. In the 1970s, concomitant with the industrialization progress and modernization growth, intensive widespread of a substantial amount of new packing materials (newspapers, glass, plastic and metals) has inspired a drastic rise of the contents and volumes of household wastes [20].

In the early fills, it was a common practice to dispose refuse by uncontrolled tipping or dumping, an operation in which waste is tipped or dumped to fill in a preexisting hole, or in low economic value open dumps on selected pieces of land (inundated swampland, abandoned sand mines and quarries) [18], without taking care of the surrounding environment, nor considering any precautions to compact, cover and prohibit the spreading of leachate to underlying waterways, with the intention of redeveloping the landscape [21]. Today, the application of scientific, engineering, and economic principles has been adopted towards the framework transformation of landfills, of which the monitoring of leachate has routinely performed by the landfill operators and prescribed by the authorities [22].

2.2. Major composition and environmental impacts

The knowledge of the leachate composition is an indication of the types and state of processes occurring within the landfills and relative solubility of the waste matrix, necessary for preliminary implementation of site remediation following barrier breakdown for installation of practicable treatment [23]. Regardless of the concentration changes and vary depending on a complex set of interrelated factors, the complexity of the landfill leachate can be categorized on the basis of four major groups of pollutants: dissolved organic matter, inorganic macro-components, heavy metals and xenobiotic organic compounds [24]. In the perspective, dissolved organic matter is a bulk parameter covering an enormous range of organic species, from methane (CH_4), volatile fatty acids (VFA) to more refractory humic- and fulvic-like compounds, which represents the intermediate degradation organic waste in the landfills [25].Meanwhile, a significant portion of the landfill leachate is contributed by the inorganic constituents, which comprising of the ions calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}), potassium (K^{+}), ammonium (NH_4^{+}), iron (Fe^{2+}), manganese (Mn^{2+}), chloride (Cl^{-}), sulphates (SO_4^{2-}) and bicarbonates (HCO_3^{-}) coupled with heavy metals (arsenic, cadmium, chromium, cobalt, copper, lead, mercury, nickel and zinc), in the microgram per liter to low milligram per liter level, that are readily soluble at fixed concentrations during the degradation processes. Whereas, the presence of a disproportionate amount of xenobiotic organic compounds is originated from the household and industrial chemicals and treatment sludges, with a broad variety of aromatic hydrocarbons, phenols and chlorinated aliphatics [26,27].

Depending upon the nature and partitioning of the waste characteristics (degree of contouring and compacting of solid wastes), moisture content, temperature, pH, oxygen level, microbial activity, groundwater inflow, surface water runoff, movement of entrainment particulate matters, local precipitation patterns, chemical equilibrium solubility, hydro-geological variation, local rainfall regime, the age, maturity, design (size, depth and lining system) and operation of landfill, topography, vegetation and the events proceeding time and separate points of samplings, the spatial distribution, variation and intensification of the landfill leachate is directly proportional to the time spans ranging from decades to centuries [28,29] (Fig. 1).

Typically, the characteristic of the landfill leachate can be best represented by chemical oxygen demand (COD), total organic carbon (TOC), biochemical oxygen demand (BOD), BOD/COD ratio, pH, suspended solids (SS), ammonium nitrogen (NH_3-N), total Kjeldahl nitrogen (TKN), bacterial count, turbidity or heavy metals content [31,32], which provided a prerequisite insight into the prediction of future trends of leachate quality and the design and operation

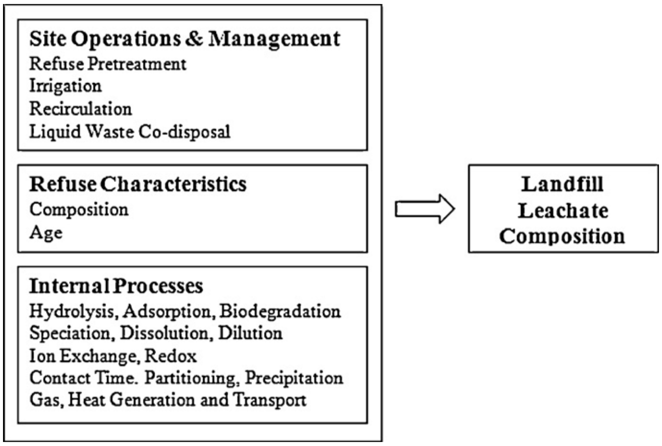

Fig. 1. Factors influencing leachate composition in landfills [30].

of leachate management facilities [10]. Accordingly, four successive stages are involved in the degradation processes: (1) aerobic stage; (2) hydrolysis and fermentation stage; (3) anaerobic acetogenic stage; and (4) anaerobic methanogenic stage [29], of which each stage is dynamic and dependent on the creation of a suitable environment by the preceding stage (the competing ability of the microbiological community to function within a changing chemical environment) [33], leading directly towards the gas and leachate production.

Table 1 summarized the classification of landfill leachate according to the composition changes. In this respect, young acidogenic landfill leachate is commonly characterized by high biochemical oxygen demand (BOD) (4000–13,000 mg/L) and chemical oxygen demand (COD) (30,000–60,000 mg/L) concentrations, moderately high strength of ammonium nitrogen (500–2000 mg/L), high ratio of BOD/COD ranging from 0.4 to 0.7 and a pH value as low as 4 [35,36], with biodegradable volatile fatty acids (VFAs) appear to be its major constituents [37]. During the methanogenic phase, landfill leachate is denoted to the presence of substantial quantities of recalcitrant, difficult-to-treat and hard COD, degradation of volatile fatty acids compounds, reducing of organic strength and pH rises of greater than 7 [38]. Along with the increasing age and domination of anaerobic decomposition over a period of 20–50 years, the stabilized leachate is featured by a high molecular weight (refractory compounds such as humic substances and fulvic-like fractions, which are not easily degradable), high strength of ammonium nitrogen (3000–5000 mg/L), moderately high strength of COD (5000–20,000 mg/L), and a low BOD/COD ratio of less than 0.1 [39].

Despite the evolution of landfill technology, from open uncontrolled dumps to highly engineered facilities designed to eliminate or minimize the potential adverse impact of the waste on the surrounding environment, the generation of contaminated leachate remains an inevitable consequence of the practice of waste disposal in landfills [40]. If poorly managed, a landfill may become a source of hydro-geological contamination due to the risk of leachate infiltrating into the natural environment and groundwater table, thus poses a multiple, synergistic, carcinogenic and acute toxicity and genotoxicity [41]. Relatively, a couple of 100 hazardous compounds have been identified in the heterogeneous landfill leachate (aromatic compounds, halogenated compounds, phenols, pesticides, heavy metals and ammonium) [42], which present an accumulative, threatening and detrimental effect to the survival of aquatic life form, ecology and food chains, by imposing a significant influence on the mobilization and attenuation towards the complexation of organic ligands and colloidal matters [21].

According to an official toxicity study conducted in 56 conventional municipal waste landfills, a disproportionate amount of 133 different toxic chemicals (32 cause cancers, 10 cause birth defects and 21 cause genetic damages) has been evidenced compared to 72 toxic chemicals in the industrial waste landfills [4]. Nonetheless, ammonia nitrogen, resulting from the decomposition process was demonstrated as the major long term toxicant (as confirmed by toxicity analyses carried out using bioassay methods and test organisms: *Daphnia magna* [43], *Salmo gairdnieri* and *Onchorhynchus nerka* [44], freshwater fish (*Sarotherodon mossambicus*) [45], luminescent bacteria [46] and zebrafish (*Danio rerio*) [47],) which stimulate the algal growth, inhibit the degradation process and deplete dissolved oxygen through eutrophication, that differ upon the oxidation, wind-drift, dilution, pH and salinity alterations [48].

3. Landfill leachate treatment technologies

Throughout recent decades, the wastewater treatment industry has identified the emission of organic, inorganic and heavy metals compounds due to leachate seepage into the waterways as a risk to the natural environments [49]. The adverse impacts of overloading in the sensitive ecosystems are becoming increasingly noticeable with several substances with confirmed carcinogenic or co-carcinogenic potential were indicated in the landfill leachate while others were expected to be persistent and highly bioaccumulative [50]. In view of the above matter, a wide range of new tertiary treatment processes has been abounded. Extensive of work has focused on the enhanced coagulation–flocculation, clarification and biological processes (aerated lagoons, activated sludge, anaerobic filters, stabilization ponds, upflow anaerobic sludge blanket, sequence biological reactor, rotating biological contactors, and nitrification or denitrification processes) as plausible circumstance for leachate treatment, mainly hinges of its reliability, simplicity, high cost-effectiveness, reduction of stabilization time and acceleration of biogas production [51,52].

Regardless of the biological reactions and quality of waste involved, the manipulation of residence time (sludge age), food–microorganism ratio (F/M), hydraulic retention time (HRT) and sludge volume index (SVI) is usually adapted for insuring

Table 1
Classification of landfill leachate according to the composition changes [32,34].

Type of leachate	Young	Intermediate	Stabilized
Age (years)	<5	5–10	>10
pH	<6.5	6.5–7.5	>7.5
COD (mg/L)	>10,000	4,000–10,000	<4000
BOD_5/COD	0.5–1.0	0.1–0.5	<0.1
Organic compounds	80% volatile fatty acids (VFA)	5–30% VFA+ humic and fulvic acids	Humic and fulvic acids
Ammonia nitrogen (mg/L)	<400	N.A	>400
TOC/COD	<0.3	0.3–0.5	>0.5
Kjeldahl nitrogen (g/L)	0.1–0.2	N.A	N.A
Heavy metals (mg/L)	Low to medium	Low	Low
Biodegradability	Important	Medium	Low

Table 2
Lists of researches for the landfill leachate treatment via activated carbon adsorption process during the last 15 years.

Activated carbon type/precursor	Adsorbate	Leachate type	Maximum adsorption capacity (mg/g)	Percentage removal (%)	Reference
Desotec	Adsorbable organic Halogens (AOXs)	Stabilized	0.59	–	[9]
	COD		268		[9]
Norit SA 4	COD	Intermediate	–	38	[10]
Commercial PAC	COD	Synthetic	–	87	[28]
	Ammonia			16	[28]
DARCO	COD	Stabilized	–	38	[31]
	Dissolved organic carbon (DOC)			40	[31]
Commercial PAC	Hydrophobic organic chemicals (HOCs)	Intermediate	–	89.2	[49]
	COD			24.6	[49]
Commercial GAC	HOC			73.4	[49]
	COD			19.1	[49]
GAC (type PHO 8/35 LBD)	COD	Stabilized	165.46	60	[51]
	Ammonia		53.58	95	[51]
Commercial PAC	COD	Intermediate	–	75	[52]
	Ammonia			44	[52]
	Phosphate			44	[52]
Oil Palm shell	COD	Stabilized	1460	50	[64]
Norit 0.8	COD	Intermediate	0.253	68	[65]
Chemviron AQ40			0.258	55	[65]
Picacarb 1240			0.148	48	[65]
Commercial GAC	Benzene	Synthetic	0.23	–	[66]
	Trichloroethylene		0.54		[66]
	1,2-dicholoroethane		0.48		[66]
Carbotech	COD	Intermediate	0.250	75	[67]
GAC-40	COD	Stabilized	38.12	–	[68]
Commercial PAC	COD	Intermediate	–	49	[69]
	Ammonia			16	[69]
Commercial PAC	COD	Stabilized	4300	38	[70]
Commercial PAC	DOC	Stabilized	50.00	–	[71]
Calgon Filtrasorb 400	COD	Stabilized	564	70	[73]
Commercial PAC	COD	Young	–	49	[76]
	Ammonia			78	[76]
	Colour			50	[76]
Rice husk	COD	Young	–	70	[78]
	Colour			60	[78]
Norit 0.8	COD	Stabilized	88.80	90	[80]
Commercial PAC	COD	Stabilized	6.5	-	[82]

the optimum growth of the complex interrelated and mixed microbiological populations [27]. Nevertheless, such efforts are always hampered by the presence of bio-refractory organics (humic substance or surfactants) and limited suitability in treating the stabilized (less biodegradable) leachate due to the recalcitrant characteristics of its organic carbon [53]. On the other hand, the recirculation and recycling of landfill leachate has shown to be one of the least expensive available options which increases the moisture content in a controlled reactor system, provides the distribution of nutrients and enzymes between the methanogens and liquids [54], and shortening of the stabilization time from several decades to 2–3 years [55]. In contrast, high volume of recirculation is constrained by the occurrence of saturation, ponding and inhibition of the methanogenesis [56].

Lately, the implementation of advanced oxidation processes (AOPs) which complies a combination of strong oxidants (ozone, chlorine, permanganate, calcium hydrochloride and hydrogen peroxide), assisted by the irradiation of ultraviolet (UV), ultrasound (US), electron beam (EB) or photo-catalysis in enhancing the degradation and biodegradability of pollutants [57], has gaining popularity worldwide. However, its treatability is often deteriorated by the potentiality of chlorine oxidation, resulting in the formation of chlorine or hypochlorite, and the major drawback of poor economic acceptability for large-scaling processes [58]. More recently, the employment of membrane filtration technologies (ultrafiltration, nanofiltration, microfiltration and reverse osmosis) has emerged to be a justifiable and viable tool in pre-treatment or in partnership with chemical treatments for elimination of colloids and suspended matters, and fractionation in evaluation of the preponderant molecular mass of organic pollutants in a given leachate, achieving a COD and heavy metals rejection coefficient value of 98 and 99%, respectively [59,60]. On the contrary, the pressure-driven processes are subjected to the fouling effect by a wide spectrum of constituents (which requires extensive pre-treatment or chemical cleaning of the membranes, resulting in short lifetime of the membranes and decreases of process productivity) and a huge generation volume of concentrate (which is unusable and need further treatments) [1].

4. Landfill leachate treatment via activated carbon adsorption process

Over the last few years, adsorption process, a surface phenomenon by which a multi-components fluid (gas or liquid) mixture is attracted to the surface of a solid adsorbent and form attachments via physical or chemical bonds, is recognized as the most efficient and promising fundamental approach in the wastewater treatment processes [61]. A notable trend in the development of activated carbon, an adsorbent with its large porous surface area, controllable pore structure, thermostability and low acid/base reactivity has been promulgated [62], owning to its superior ability for removal of a wide variety of organic and inorganic pollutants dissolved in aqueous media, even from gaseous environment [63]. Table 2 presents lists of researches for the landfill leachate treatment via activated carbon adsorption process during the last 15 years. In most cases, activated carbon adsorption (Fig. 2) has revealed the prominence in removal an essential amount of organic compounds and ammonium nitrogen from the leachate samples.

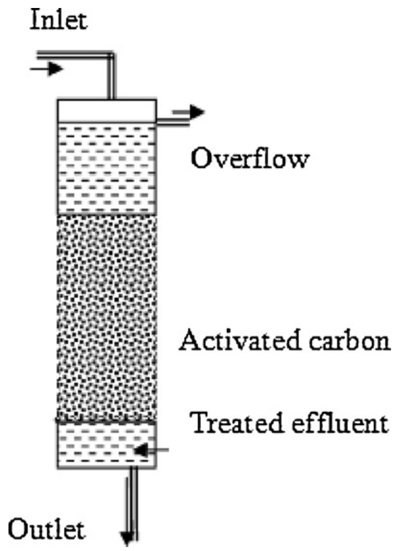

Fig. 2. Experimental set-up of activated carbon column of the leachate treatment [72].

In particular, stabilized leachate from the Goslar landfill, Germany was firstly evaluated using a granular activated carbon (GAC) column in 1995, illustrating a COD removal of 91% with an initial concentration of 940 mg/L. Accordingly, film diffusion and internal surface diffusion were demonstrated to play a key role in determining its kinetic rate [73]. In Greece (Thessaloniki landfill), similar study has been conducted with powder activated carbons (PAC) of varying dosage (from 0.2 to 10.0 g/L), suggesting the applicability of Freundlich isotherm with a COD removal of 95% (initial concentration of 5690 mg/L) [74]. Lately, Wasay et al. [75] performed a separate investigation utilizing granular activated carbon, granular activated alumina and ferric chloride for the treatment of heavy metals [Cd(II), Cu(II), Cr(III), Mn(II), Pb(II) and Zn(II)], indicating granular activated carbon to be the most competent adsorbent with the removal of 80–96%, at a pH range of 6–7.7 (initial concentration of 184 mg/L). In Malaysia, a comparative study for the removal of ammonium nitrogen has been undertaken by Aziz et al. [14] using granular activated carbons and limestones in the Burung Island landfill. Approximately 40% of ammonium nitrogen with an initial concentration of 1909 mg/L was eliminated with 42 g/L of GAC while 19% removal was achieved using 56 g/L of limestone under the same concentration.

Coinciding in coping the temporal fluctuations in varying strength and composition of landfill leachate, and ameliorating the single step adsorption process, recently, the development of collaborated multistage treatments, which combine adsorption process with numerous complementary approaches have received stern attention and various encourages.

A substantial amount of simultaneous adsorption and biological treatment investigations have been practiced, offering a number of advantages, including the enhancement of nitrification efficiency (activity of nitrifiers), improvement of sludge dewaterability (filterability), and removal of refractory organic compounds [10,76]. Under the co-treatment processes, the existence of activated carbons is believed may contribute a synergy effect for providing an attachment surface for bio-regeneration (microorganisms) and serving as a nucleus for the occurrence of floc formation [77]. Moreover, it has always been linked to the biodegradation beneficial as supporting medium in the biofilm reactors and dampening effects of leachate in the combined domestic wastewater and landfill leachate systems [78].

In the latter case, two combinative set processes of coagulation–flocculation-activated carbon and Fenton's oxidation-activated carbon has been examined by Ramírez et al. [79], claiming the improvement of adsorption process due to the generation of smaller and adsorbable molecules. In recent years, activated carbon adsorption processes has prevailed as one of the most satisfactory and feasible options for the treatment of contaminated landfill leachate in junction with ozonation oxidation [80] (Fig. 3). Upon the decomposition, ozone was reported capable of oxidizing organic substances to their highest stable oxidation states, producing water and carbon dioxide, while activated carbon can remarkably accelerate the kinetic rate through the formation of •OH radicals [81].

5. Major challenges and future prospects

The world is currently facing the worst environmental crisis in its entire history. Within the last few decades, the enthusiasm of huge waste production and environmental preservation has been one of the most challenging topics which have focused greatest public concern and critical considerations towards the recovery of contamination resources. In line with the growing anxiety of

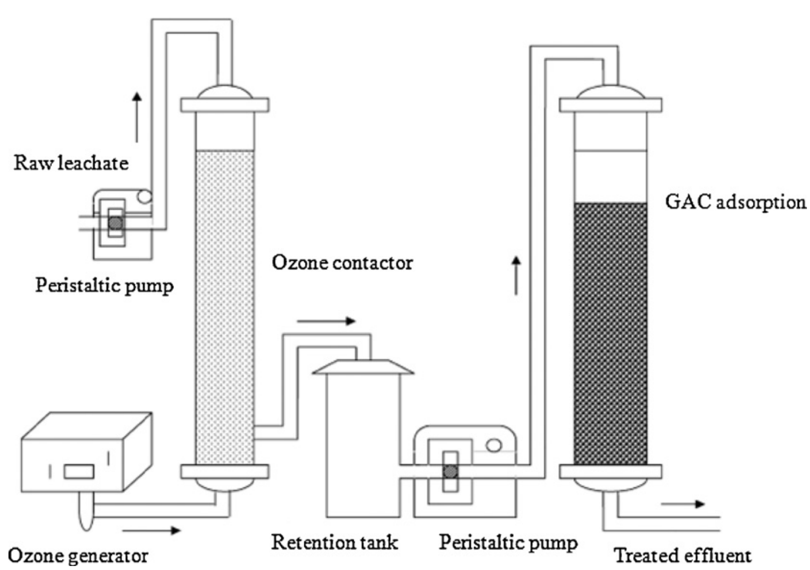

Fig. 3. Schematic diagram of the integrated ozone-GAC adsorption system [51].

the environment-friendly technologies and achieving the status of green environmental policy, various research and development efforts have been advocated to utilize activated carbons contemplated mainly for landfill leachate treatment, in accruing worldwide environmental benefit and shaping the national economy [83]. Although there have been some successful industrial-scale applications and implications, generally the industry is still facing various challenges, the availability of economically viable technology, sophisticated and sustainable natural resources management (cost-prohibitive adsorbent and difficulties associated with regeneration) [84], and proper market strategies under competitive markets.

Amidst these shortcomings, the developing exploration in evaluating the adequacy and suitability of natural, renewable and low cost materials (palm shell, pall fiber, palm stone, bamboo dust, peat, chitosan, lignite, fungi, moss, bark husk, chitin, coir pith, maize cob, pinewood sawdust, rice husk, sugar cane bagasse, tea leaves, and sago waste) as alternative precursors has currently been executed [85,86]. Explicitly, a wide range of integrative approaches which encompassing physical, chemical and biological technologies (membrane filtration-biological [87], ozonation-biological [88], chemical precipitation-biological oxidation [89], activated carbon-biological, and anaerobic–aerobic-rotating biological contactor systems [90]) are attracting extensive momentum and high priorities. Varying upon the alterations of time, place and context, environmental effectiveness, technological feasibility, social acceptability and economical affordability (chemicals, energy consumption, treatment facilities, labor, transportation, collection and maintenance) are usually the key drivers deciding its flexibility, reliability and sustainable manner. Parallel to the central principles of waste management hierarchy, the paradigm shift of individual and groups recycling, recovering, reuse and reduction (quantity, weight, volume and toxicity) throughout the waste chain has seen a panacea and new menu to the waste minimization strategy [91,2].

Accordingly, the urgency of conceiving and administering of strategic, corrective and transparent polices, mandates and standards which governing the collection, transportation, disposal prevention, recycling, reuse, monitoring, designing and supervision of solid waste management ought to be pointed out and well-planned. Increasingly, the sound professional knowledge of creating environmental awareness for adequate financial provisions, engineering and operating standards, responsibilities sharing, product stewardship, staff capacities upgrading, public participation, formal procedures redressing, regular opinion survey, site rehabilitation and aftercare maintenance need to be properly assigned and counteracted [92,82]. Ultimately, full co-operation and joint venture between different parties (nations, states, local government, private sector and communities) from upstream till the bottom line with compatible technologies is a directive motivation for the race to the end line.

6. Conclusion

Over the years, the world's giant factories and processing industries are gradually expanding, driving towards the overwhelming solid waste generation. Predictions for the next 20 years indicate an upward trend in waste production and, subsequently in leachate infiltration. Today, the growing discrepancy and limited success of remediation in field applications has raised apprehensions over the use of activated carbon (or its integrated technologies) as a measure to the environmental pollution control. The evolution has turned from an interesting alternative approach into a powerful standard technique by offering a numbers of advantages. Despite various drawbacks and challenges has been identified and clarified, a widespread and great progress of in this area can be expected in the future.

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