



## New sequential treatment for mature landfill leachate by cationic/anionic and anionic/cationic processes: Optimization and comparative study

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### ABSTRACT

Two new applications for sequence treatment of mature (stabilized) landfill leachate, that is, cationic resin followed by anionic resin (cationic/anionic) and anionic resin followed by cationic resin (anionic/cationic), are employed and documented for the first time in the literature. Response surface methodology (RSM) concerning central composite design (CCD) is used to optimize each treatment process, as well as evaluate the individual and interactive effects of operational cationic resin dosage and anionic resin dosage on the effectiveness of each application in terms of color, chemical oxygen demand (COD), and  $\text{NH}_3\text{-N}$  removal efficiency. A statistically significant model for color, COD, and  $\text{NH}_3\text{-N}$  removal was obtained with high coefficient of determination values ( $R^2 > 0.8$ ). Under optimum operational conditions, the removal efficiency levels for color, COD, and  $\text{NH}_3\text{-N}$  are 96.8%, 87.9%, and 93.8% via cationic/anionic sequence, and 91.6%, 72.3%, and 92.5% via anionic/cationic sequence, respectively. The experimental results and the model predictions agree well with each other.

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### 1. Introduction

Landfilling is an important means of managing the increasing amount of solid wastes generated. However, the habitual disposal of municipal solid wastes in landfills presents the following problems: the contamination of surface and ground water through leachate; soil contamination through direct waste contact or leachate; air pollution through the burning of wastes and the uncontrolled release of methane by anaerobic decomposition of waste; spread of diseases by different vectors, such as birds and insects; and odor [1]. The presence of high concentrations of pollutants in municipal landfill leachate is one of the primary issues usually encountered by landfill operators. Because landfill leachate properties continue to be dangerous and poisonous over long periods, impurity removal (or at least reduction) has become an imperative concern in leachate treatment over recent decades [2–4].

Unfortunately, tropical countries, such as Malaysia, exhibit increased leachate production due to rainfall exceeding the amount that can be evaporated during the rainy season [5]. According to Trankler et al. [6], in hot and humid weathers, leachate production is considerably higher and varies more than in hot and arid regions because of intensive microbial activity.

Typically, the quality and quantity of landfill leachate can be influenced by several factors, including solid waste decomposition, landfill age, hydrology of landfill site, climatic condition, and moisture content [7].

Characteristically, landfills more than 10 years old are normally in the methanogenic phase, and the leachate produced is referred to as mature, “stabilized” leachate. According to Christensen et al. [8], many parameters change dramatically when a landfill becomes mature, in which the stabilized leachate normally contains a large quantity of non-biodegradable organic compounds, such as humic and fulvic substances. Moreover, the leachate contains considerable amounts of inorganic substances, particularly  $\text{NH}_3\text{-N}$  [9]. On account of its complicated characteristics, the application of biological treatment alone for stabilized leachate treatment is not a viable alternative [10]. Therefore, the common focus of research nowadays is on the treatment of stabilized leachate using physico-chemical techniques, such as adsorption, coagulation and flocculation, oxidation, air flotation, membrane filtration, and ion exchange processes [11–18]. Zagorodni [19] reported that ion exchange resins have been broadly employed in water and wastewater treatment for the extraction, separation, and purification of ion and organic substances.

According to the literature, however, the implementation of the ion exchange technique using cation and anion ion exchange resin for stabilized leachate treatment is still rare [4,20]. Recently, the removal efficiency of some ion substances from stabilized leachate using cation exchange resin has been advantageously employed by Bashir et al. [3] and Primo et al. [17]. Furthermore, anion exchange

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**Table 1**  
Characteristics of raw leachate from PBLs.

Parameters	Units	Feb 2008–Jan 2010 (current study)		Sep 2007–Feb 2008 [15]		Standard discharge limit <sup>a</sup>
		Values	Average	Values	Average	
pH	–	8.30–9.17	8.58	7.76	8.2	6.0–9.0
COD	mg/L	1810–2850	2321	2270–2945	2667	400
NH <sub>3</sub> -N	mg/L	1630–2200	1949	983–2170	1760	5
Color	Pt–Co	4250–5760	5094	3860–4248	4059	100
Turbidity	FAU	128–330	211	203–308	248	–
SS	mg/L	114–360	181	177–254	211	50
Conductivity	μS/cm	21,850–26,230	24,340	–	–	–

<sup>a</sup> Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, under the Laws of Malaysia–Malaysia Environmental Quality Act 1974 [24].

resin has been employed for non-biodegradable organic compound [measured as chemical oxygen demand (COD)] removal [18,21].

To date, the application of the ion exchange process in landfill leachate treatment focuses on the use of a particular resin type (cation or anion) to treat a specific pollutant. In line with the above, this study is undertaken to examine, optimize, and compare the effectiveness of using two different sequential treatment systems for stabilized leachate [i.e., treatment by cationic exchanger followed by anionic exchanger (cationic/anionic), and treatment by anionic exchanger followed by cationic exchanger (anionic/cationic)]. The sequential utilization of both media for the simultaneous removal of color, COD, and NH<sub>3</sub>-N from semi-aerobic stabilized landfill leachate is conducted and documented for the first time in the literature. Moreover, the experimental design and optimization for each process is carried out using response surface methodology (RSM) as an ideal statistical method for experimental design and data analysis. According to Montgomery [22], RSM is a technique used for modeling, designing experiments, evaluating the effects of process variables and their interactions, and searching for optimum conditions of variables to predict aim responses.

## 2. Materials and methods

### 2.1. Site characteristics and leachate sampling

Raw leachate was obtained from a leachate aeration pond at Pulau Burung Landfill Site (PBLs). PBLs is a semi-aerobic sanitary landfill situated within the Byram Forest Reserve at 5°24' N Latitude, 100°24' E Longitude in the northwest coast of Peninsular Malaysia (has a tropical climate), approximately 20 km southeast of Penang Island. PBLs has an area of 62.4 ha, of which 33 ha is currently operational and receives about 2200 t of municipal solid wastes daily. Pulau Burung's solid wastes are composed of 40.0% food, 22.0% plastic, 10.5% paper, 2.5% metal, 3.2% glass, 3.5% textile, and 18.2% other wastes. This site was developed in 1991 as a Level II semi-aerobic sanitary landfill by establishing a controlled tipping

technique. In PBLs, leachate is collected through a leachate collection pipe that flows into a detention pond. The site was chosen because it was developed semi-aerobically with leachate recirculation, and is one of only three sites of its kind in Malaysia [2]. These sites were developed consistently with the Fukuoka Landfill Method. PBLs is equipped with leachate collection and gas collection systems, as well as aeration for a leachate pond.

Leachate samples were taken from PBLs in accordance with the Standard Methods for the Examination of Water and Wastewater [23]. The samples were directly transported to the laboratory and stored in a cold room at 4 °C until analysis. The samples were characterized for color, COD, NH<sub>3</sub>-N, SS, and turbidity concentrations and pH as illustrated in Table 1.

### 2.2. Materials

The strong cationic resin, INDION 225 Na, and the strong anionic resin, INDION FFIP MB, supplied by Ion Exchange (INDIA) Ltd. were used in this study. The resins and their physico-chemical properties are presented in Table 2. INDION 225 Na was chosen because of its known characteristics, such as its ability to work as a strong acid cation exchanger when used in the form of hydrogen; its possibility for use in hydrogen form and sodium form; and its application over a wide range of pH levels and temperatures. The same is true for INDION FFIP MB, which acts as a strong base anion exchanger. It can also be used in chloride form and hydroxide form [3,18]. The matrix is polystyrene cross-linked divinylbenzene for both anion and cation which is the most popular matrix [25,26]. Typically, the strong acid cation exchangers have a greater affinity than weak resins for all ionized elements in water [26]. The strong base anion exchangers are principally effective for the removal of many synthetic and natural organic substances that contain weak acids, such as humic and fluvic substances [27–31].

In the current study, anion resin was used in chloride form [18], whereas cation resin was used in hydrogen form [3]. Prior to use, the studied cation and anion resins were rinsed extensively with

**Table 2**  
Physicochemical properties of the studied resins.

Property	INDION FFIP MB	INDION 225 Na
Type	Strongly base anion exchange resin	Strongly acid cation exchange resin
Matrix	Cross-linked polystyrene, isoporous type	Cross-linked polystyrene, gel type
Functional group	Quaternary amine (–N+R <sub>3</sub> )	Sulfonic acid (–SO <sub>3</sub> <sup>–</sup> )
Ionic form (as supplied)	Chloride	Sodium form
Maximum operating temperature	60 °C (OH <sup>–</sup> form) 90 °C (Cl <sup>–</sup> form)	120 °C (H <sup>+</sup> form) 120 °C (Na <sup>+</sup> form)
Operating pH range	0–14	0–14
Particles size range	0.45–0.55 mm	0.3–1.2 mm
Total exchange capacity	1.2 meq/mL	2.0 meq/mL
Bulk density (g/cm <sup>3</sup> )	0.611	0.81
Particle density (g/cm <sup>3</sup> )	1.11	1.328
Porosity (%)	45.04	39.0
Surface area (m <sup>2</sup> /g)	0.5419	0.0996
Appearance	Brown to dark brown	Yellow

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