NEW COMPOSITE COAGULANT FROM AGRO-WASTE (TAPIOCA PEEL) AND POLYALUMINIUM CHLORIDE FOR PRIMARY LANDFILL LEACHATE TREATMENT

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بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

By Allah's favour and bounty. Allah is all-knowing, All-wise [49:8]

To the world of life; my mom, Hjh. Zamriah Bakri

To my guardian angels; Hj. Rusmin Muriat and Hjh. Pathiah Mohd Salleh

To all my sisters and brothers

&

In memory of Hj. Mohd Salleh Hj. Jefri & Hj. Kamaruzahan Hj. Mohd Salleh

This one’s for you

Should there be a knowledge that others benefitted from this copy may Allah count it as sadaqul jariyah and reward all of you eternally

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ABSTRACT

Coagulation-flocculation is one of the applications that work efficiently for stabilised leachate treatment, alongside the utilisation of chemical coagulants. On the contrary, the use of chemical coagulants has certain drawbacks towards the health of the ecosystem and living organisms. Hence, this leads to the application of natural material (tapioca peel powder (TPP)) originated and recycled from agro-waste in this study. The limited ability of TPP as a single coagulant could be overcome by substituting the material into polyaluminium chloride (PAC) as a composite coagulant (PACTPP) with different weight ratios. PACTPP was tested on stabilised leachate taken from Simpang Renggam's landfill site (SRLS) by investigating the removal percentage of respective parameters. Based on the weight ratio optimisation, TPP/Al = 3.71 or PACTPPg was determined as the optimum composite coagulant. It was characterised that PACTPPg had combined the best benefits from PAC and TPP, with an acidic property of pH 3.45, a low charge density of 3.45 mV, a higher molecular weight of 1.59×10^7 g/mol, and a bigger particle size of 4.528×10^4 d.mn. By using scanning electron microscopy/energy-dispersive X-ray (SEM-EDX), a longer connected and compact structure was observed with a diameter of 142.3 μm, i.e., 70.1% increment from PAC. Through Fourier-transform infrared (FTIR) and X-ray diffraction (XRD) analyses, PACTPPg was identified to comprise new chemical compounds: the functional groups of ketones, aldehydes, and alkanes in a semi-formed crystalline phase. The newly formed compounds could be associated with the bridging function that becomes the predominant mechanism for PACTPPg's coagulation behaviour in removing pollutants. Response surface methodology (RSM) was applied after using the conventional method of one-factor-at-a-time (OFAT) optimisation to find the optimum pH for the leachate sample and dosage of PACTPPg. At the optimum conditions of pH 7.13 and 2,446.18 mg/L (479.49 mg/L of Al content), the responses were 59.4% for chemical oxygen demand (COD), 23.7% for ammonia nitrogen, 91.8% for turbidity, 88.5% for colour, and 96.0% for suspended solids of removal percentages. The

optimisation also revealed that the performance of PACTPPg was at par with that of PAC+TPP, PAC, and TPP coagulants, especially on the removal of physical parameters. The dosages of PACTPPg were reduced by 33.0% and 26.8% compared to the application of PAC and PAC+TPP, respectively. Meanwhile, from the flocs and sludge formation analyses, PACTPPg worked well for the treatment of landfill leachate by inducing adsorption and bridging between particles > charge neutralisation > sweep flocculation mechanisms with denser generated sludge. PAC+TPP, PAC, and TPP also underwent these kinds of mechanisms but with different superiority, which was portrayed by its percentages of parameter removal. Based on the study, it can be concluded that PACTPPg has the potential to be a sustainable coagulant for the primary treatment of stabilised raw leachate in Malaysia.

ABSTRAK

Pengumpalan-pemberbukuan adalah salah satu aplikasi yang berfungsi dengan cekap dalam rawatan larut resapan, bersama penggunaan koagulan kimia. Sebaliknya, penggunaan koagulan kimia mempunyai kelemahan tertentu terhadap kesihatan ekosistem dan organisma hidup. Oleh itu, ini membawa kepada penggunaan bahan semula jadi (serbuk kulit tapioka (TPP)) berasal dan di kitar semula daripada sisa agro dalam kajian ini. Kemampuan terhad TPP sebagai koagulan tunggal diatasi dengan menjadikannya sebagai pengganti polyaluminium klorida (PAC) di dalam koagulan komposit (PACTPP) dengan nisbah berat yang berbeza. PACTPP diuji pada larutan resapan yang diambil dari tapak pelupusan Simpang Renggam (SRLS) dengan menyiasat peratusan penyingkiran parameter masing-masing. Berdasarkan pengoptimuman nisbah berat, TPP/Al = 3.71 atau PACTPPg ditetapkan sebagai koagulan komposit optimum. Ia dicirikan bahawa PACTPPg telah menyatukan manfaat terbaik daripada PAC dan TPP, dengan mempunyai sifat asid pada pH 3.45, ketumpatan cas rendah pada 3.45 mV, berat molekul yang lebih tinggi pada 1.59×10^7 g/mol, dan saiz zarah yang lebih besar pada 4.528×10^4 d.mn. Dengan menggunakan pengimbas mikroskop elektron/sinar-X penyebaran tenaga (SEM-EDX), struktur sambungan yang lebih panjang dan padat dapat diperhatikan dengan 142.3 μm diameter, yakni 70.1% pertambahan daripada PAC. Melalui analisis inframerah transformasi Fourier (FTIR) dan belauan sinar-X (XRD), PACTPPg dikenal pasti terdiri daripada sebatian kimia baharu; kumpulan berfungsi keton, aldehid, dan alkana dalam fasa kristal separa terbentuk. Komponen yang baru terbentuk dapat dikaitkan dengan fungsi penghubung yang menjadi mekanisme utama untuk perilaku pembekuan PACTPPg dalam membuang bahan pencemar. Metodologi permukaan tindak balas (RSM) telah digunakan selepas menggunakan kaedah konvensional pengoptimuman satu-faktor-pada-satu-masa (OFAT) untuk mencari pH optimum pada sampel larut resapan dan dos PACTPPg. Pada keadaan optimum pH 7.13 dan 2,446.18 mg/L (479.49 mg/L kandungan Al), respons adalah 59.4% untuk permintaan oksigen

kimia (COD), 23.7% untuk nitrogen ammonia, 91.8% untuk kekeruhan, 88.5% untuk warna, dan 96.0% untuk pepejal terampai bagi peratusan penyingkiran. Pengoptimuman juga mendedahkan bahawa prestasi PACTPPg adalah setanding dengan PAC+TPP, PAC, dan TPP koagulan terutamanya pada penyingkiran parameter fizikal. Dos PACTPPg adalah 33.0% dan 26.8% dikurangkan berbanding penggunaan dos PAC dan PAC+TPP. Sementara itu, melalui analisis pembentukan lumpur dan enap cemar, PACTPPg berfungsi dengan baik untuk rawatan pelupusan larut dalam tanah dengan mendorong penjerapan dan penyambungan di antara zarah-zarah > peneutralan caj > mekanisme pemberbukuan sapuan dengan enap cemar yang padat. PAC+TPP, PAC, dan TPP turut menggunakan mekanisme seperti ini tetapi dengan keunggulan yang berbeza, yang digambarkan pada peratus penyingkiran parameter. Berdasarkan kajian ini, dapat disimpulkan bahawa PACTPPg berpotensi menjadi koagulan mampan untuk rawatan awal larut resapan stabil di Malaysia.

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LIST OF SYMBOLS AND ABBREVIATION

AAS	Atomic absorption spectroscopy
Al	Aluminium
ANOVA	Analysis of variance
AP	Adequate precision
APHA	American Public Health Association
ASTM	American Society for Testing and Materials
BAKAJ	Johor Water Regulatory Authority
BOD	Biochemical oxygen demand
C	Carbon
Ca	Calcium
CCD	Central composite design
CDSS	Cross-linked *Durio zibethinus* seed starch
CEP	Complex Event Processing
Cl	Chlorine
COD	Chemical oxygen demand
CPS	Cassava peel starch
d.nm	Diameter values in nanometres
DO	Dissolved oxygen
DOE	Department of Environment
DSS	Durian seed starch

EQA	Environmental Quality Act
FAO	Food and Agricultural Organization of the United Nations
Fe	Iron
FKAAB	Faculty of Civil Engineering and Built Environment
FTIR	Fourier-transform infrared
g/mol	Gram per mol
HCl	Hydrochloric acid
HCN	Hydrogen cyanide
IC	Ion chromatography
ICP-MS	Inductively coupled plasma-mass spectrometer
IEP	Isoelectric point
IR	Infrared
JSS	Jackfruit seed starch
K	Potassium
KATS	Ministry of Water, Land, and Natural Resources
kDA	Kilodalton
KPKT	Ministry of Housing and Local Government
MBP	Bioprocessing Laboratory
MPS	Multiprobe system
Mg	Magnesium
MKAP	Environmental Engineering Analysis Laboratory
MKAS	Wastewater Engineering Laboratory
MKP	Environmental Engineering Laboratory
MPRC	Micropollutant Research Centre

mg/L	Milligram per liter
mS/cm	Millisiemens per centimetre
mV	Millivolt
Na	Sodium
NaOH	Sodium hydroxide
NOM	Natural organic matter
O	Oxygen
OFAT	One-factor-at-a-time
PAC	Polyaluminium chloride
PACTPP	Polyaluminium chloride-tapioca peel powder
PACTS	Polyaluminium chloride-tapioca starch
PAFC-Starch-g-p (AM-DMDAAC)	Polyaluminium ferric chloride-starch graft copolymer with acrylamide and dimethyl diallyl ammonium chloride
PAFSiC	Polyaluminium ferric silicate chloride
PDM	Polydimethyldiallylammonium chloride
PFASC	Polymeric ferric aluminium sulphate chloride
PFC-PDM	Polyferric chloride-polydimethyldiallylammonium chloride
PFTS	Polymeric ferric titanium sulphate
PFZSSB	Polysilicate zinc ferric sulphate with boric acid radical
PHITF	Prehydrolysed iron-tapioca flour
POME	Palm oil mill effluent
PTSS	Polytitanium silicate sulphate
rpm	Revolutions per minute
RSM	Response surface methodology
S	Sulphur

SEM-EDX	Scanning electron microscopy/energy-dispersive X-ray
SI	Système Internationale/International system
Si	Silica
SRLS	Simpang Renggam's landfill site
SSR	Sludge settling rate
SVI	Sludge volume index
TDS	Total dissolved solids
TFC	*Tympanotonos fuscatus*
TPP	Tapioca peel powder
TS	Tapioca starch
XRD	X-ray diffraction
XRF	X-ray fluorescence
YSI	Yellow Springs Instrument

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CHAPTER 1

INTRODUCTION

1.1 Research background

The population of Malaysia is increasing, so does the waste generation. The skyrocketed economic and consumption activities typically in the capital city contribute as one of the factors of waste generation problem (Budhiarta *et al.*, 2012). High waste generation leads to high disposal volume rates, which leads to insufficient utilities available in Malaysia to manage this problem; thus, waste ends up in dumpsites without any proper management and close monitoring (Tarmudi *et al.*, 2009). According to Bories *et al*. (2009), almost 1,000 million tonnes of agro-waste are discarded annually on earth, and about 1.2 million tonnes are found in landfills in Malaysia. The statistics showed that the country recorded a terrifying daily waste production with an increasing more than double, from 19,000 tonnes in 2005 to 38,142 tonnes in 2018 (Muzamir, 2020). The management of solid waste is reaching the unfavourable phase due to the difficulty in providing the anticipated facilities for public services (Bhalla *et al.*, 2014). Malaysia comprises 13 states and three federal territories, and the country is categorised as middle-income countries alongside Myanmar, Thailand, and India, with the rate between 0.78 and 1.16 kg/capita/day (Ismail & Manaf, 2013). Therefore, it can be stated that the growing countries' development and activities will increase solid waste generation.

The expansion of technology and further research has led to the introduction of sanitary landfills in Malaysia. The implementation of sanitary landfills is not yet widely practised as only a few of them are operating and available due to high cost and financial constraints (Jalil, 2016). The rest of the landfills are just non-sanitary and

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