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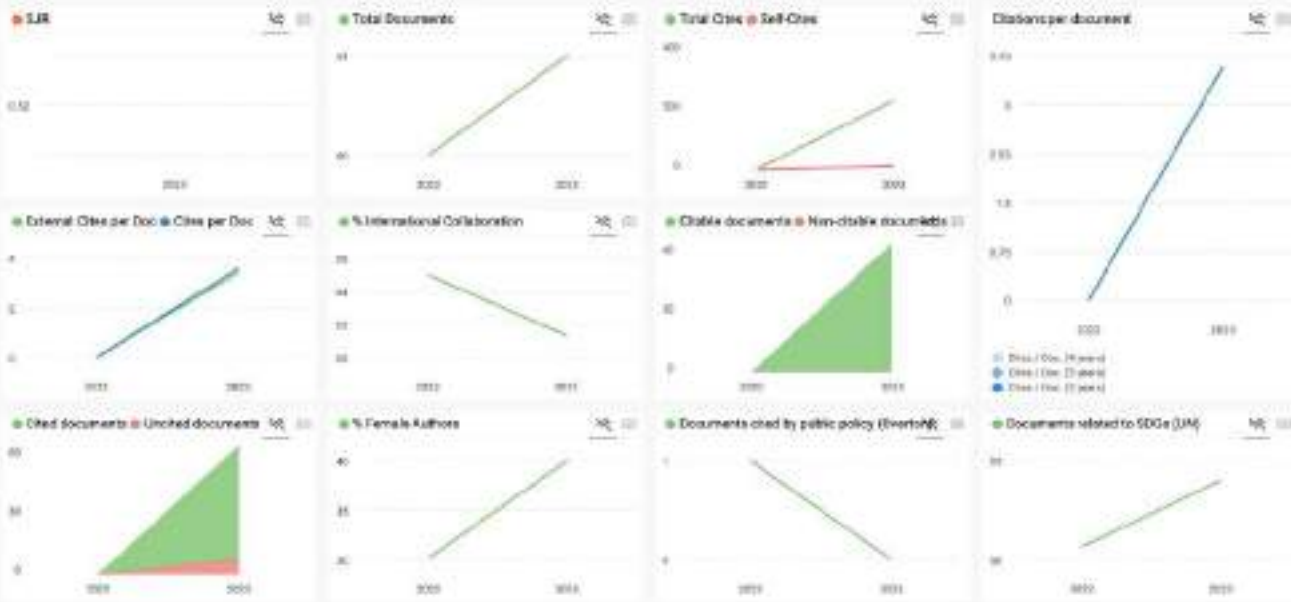
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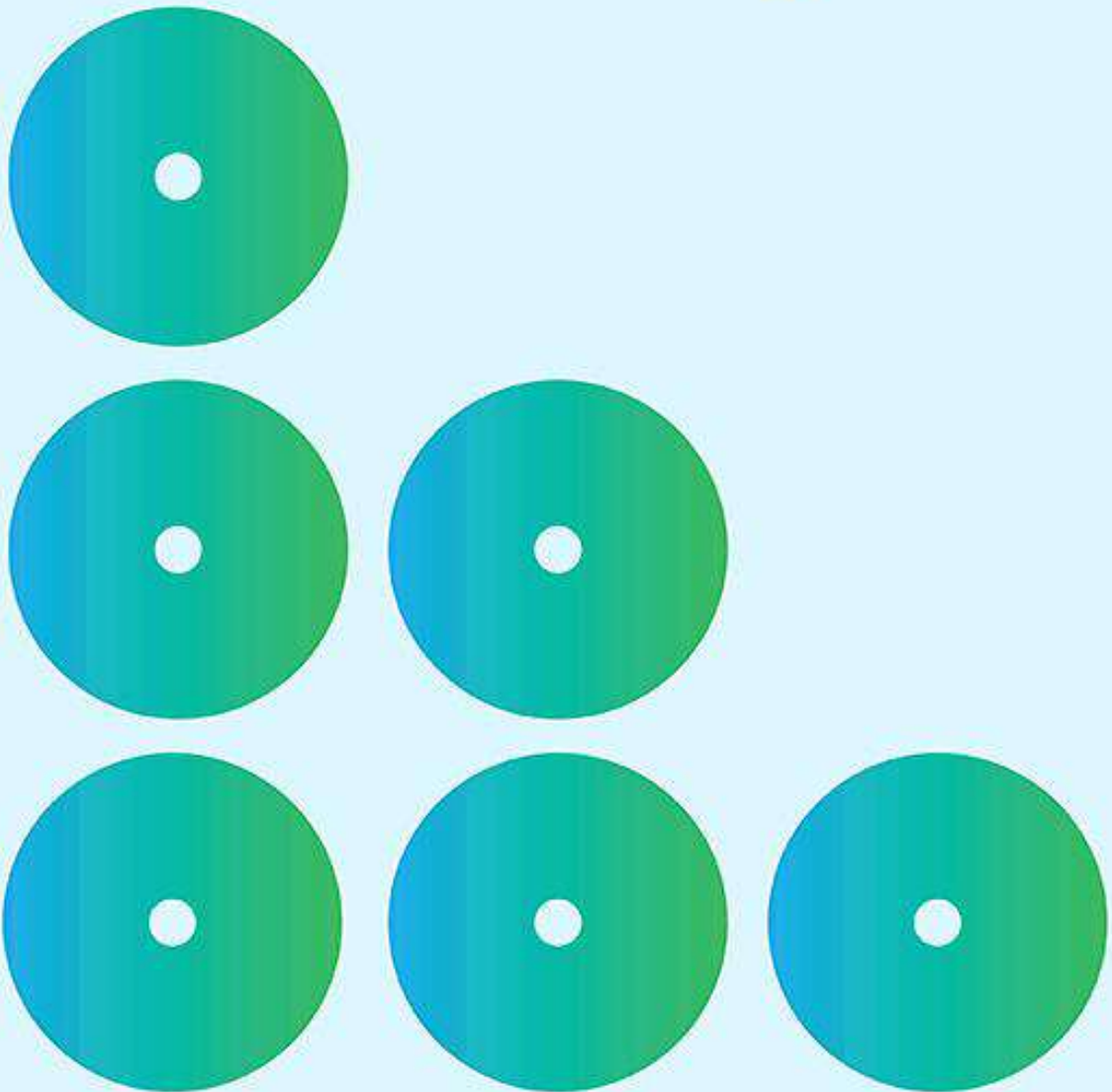
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# Mass transfer kinetics of polyethylene degradation by bacterial-fungal consortium

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## ABSTRACT

Understanding the biodegradation rate of polyethylene (PE) plastic waste mediated by bacterial-fungal consortium (BFC) is important to ensure effective design process of the bioremediation technology. The aims of this study were to scrutinize the behaviors of PE plastic degradation mediated by the BFC colonies numerically simulated the experimental data using the modified mass transfer factor models and to analyze the kinetics and mechanisms of internal, external and global mass transfer. The performance of rectangular reactor (RR) to biologically degrade the PE plastic increased up to 61.5% shows an increased efficiency of 55.9% stimulated by the presence of BFC colonies. Trend in the variation of internal mass transfer is almost the same with that of global mass transfer and is far higher than that of external mass transfer (EMT). The rate-limiting step of PE plastic degradation is dependent on the resistance of EMT. The application of BFC colonies aimed to improve the biodegradation rate of PE plastic waste contributes to advancing the future environmental engineering technologies.

## 1. Introduction

The production of plastics increased during the last few decades has caused an increased global plastic waste of up to around 6300 million tons (Zhang et al., 2020; Zhang et al., 2021). Of 8300 million tons plastics produced in 2017 has caused approximately 60% of the plastic waste accumulated into the environment with 95% of it ended up in the landfills and 5% of it ended up in the oceans and other terrestrial areas (Mohanani et al., 2020). An increased amount of plastic waste with its recalcitrant nature accumulated in the environment could be due to the uncontrolled use and disposal of the plastic materials (Ncube et al., 2021). An increased contamination rate of plastic waste caused by loss of the most recognizable and abundant PE film may threaten the viability of recycling pathway and requires a complete speciation of the recycling stream with a novel level of details (Meert et al., 2021; Seenivasagan et al., 2022). Some of the most common types of thermoplastic produced are polyethylene (PE), polypropylene, polyvinylchloride, polystyrene, polycarbonate, and PE-terephthalate; however, the plastic type of PE is widely used for kitchen utensils,

packaging materials and disposable beverage cups (Rochman et al., 2015). The production of plastic materials increased during the early period of Covid-19 pandemic was due to produce the personal protection equipment of such as gloves, face shields, masks and single-use plastic products contributing to generate more plastic waste (Shilpa et al., 2022). The use of PE mulch as a tool in the agricultural practices could be useful to reach growers production goals resulted from greater nutrient uptake causing roots growing faster to boost earlier ripening and higher yield of fruit (Kasirajan and Ngouajio, 2012). Reasons for the application of PE plastic mulch in the agricultural practices include controlling weed growth, stabilizing soil temperature and soil moisture, improving fertility and health of the soil, and enhancing the visual appeal of the area (Ardisson et al., 2014). Transportation cost of wasted PE plastic mulching reduced by reducing the level of impurity content promotes sustainable management of plastic pollution in the rural areas (Dong et al., 2022).

Long-term exposure of plastics and plastic products to an aquatic environment can lead to a negative effect on the living organisms in an aquatic ecosystem (Rao and Geckeler, 2011). The design and

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optimization of the PE plastic waste analyzed using semi-detailed kinetic model has been proposed to support to thermochemical recycling technologies (Locaspi et al., 2023). The release of PE plastic waste into the environment can lead to leaching of toxic chemicals absorbed by biota tissue, organs, and even cells of the living organisms causing adverse effects on human health (Chen et al., 2022; Yuan et al., 2022). The presence of plastic waste in the environment can be biologically degraded through the mechanisms of hydrolytic degradation, photo-degradation, thermo-oxidative degradation and biodegradation (Dimassi et al., 2022). Scalability of the plastic degradation processes in scientific research needs to be integrated the efforts of state, universities and civil society in the construction of circular ecosystem infrastructures for assessing the various lab-scale prototypes and industrial applications for the management of plastic waste (da Silva et al., 2022). Efforts in making many microorganisms propitious for green chemistry to eliminate harmful plastics from the terrestrial and aquatic ecosystems are suggested by evolving the strategies of using the appropriate organisms to survive for effectively degrading the plastic waste (Dunn and Welden, 2023; Zeenat et al., 2021). The utilization of thermophilic microbial consortia originated from cow dung as the engineering microbes can enhance the biodegradation of low- and high-density PE (Skariyachan et al., 2017). The application of the mesophilic mixed bacteria isolated from the sediment of municipal landfill can biologically utilize the PE particles as a sole carbon source to decompose the PE plastic waste during the test period (Park and Kim, 2019). The performance of microbial strains naturally occurred whether in the soil, activated sludge, farm sludge, or worms' excreta was effective to degrade the PE plastic waste without an inhibition of microbial growth caused by the derived by-products from microbial degradation (Taghavi et al., 2021). The use of fungal laccases and peroxidases could be useful for depolymerization of PE plastic waste (Eldin et al., 2022). The potential of oil seep-contaminated ecosystem for plastic degradation acquired following a long-term adaptation to petroleum compounds has been indicated the diversity of microbial community and the presence of polyethylene terephthalate (PET) degrading bacteria (Babazadeh et al., 2023).

The kinetics and mechanisms of mass transfer for analyzing the movement of degraded-PE particles controlled by either internal diffusion or film mass transfer, which can determine the resistance of mass transfer, could be due to the interdependent biological, chemical and physical processes regulate the speciation, distribution and partitioning of biologically reactive particles (Díaz et al., 2020; Fulazzaky et al., 2013; 2017a). The transport of degraded-PE particles from an internal diffusion to film zone at the surface of PE plastic is influenced by molecular bonding and then from a film zone to cell well of the microorganisms influenced by the environmental conditions (Binda et al., 2021; Lim and Thian, 2022). The internal and external mass transfer of degraded-PE particles released from a PE plastic waste could be affected by the experimental factors of pH, temperature, moisture content, porosity of culture media and biological growth (Fulazzaky, 2011). The porosity of growth media increased when mixing with rice husk could be due to the pore widening by smaller average particle size of rice husks than laterite causing an increased moisture content of the mixed-media affected by an increased surface contact (Oishi and Yagawa, 2020). An increase in the availability of carbon source in the interior channels of growth media causing the utilization of carbon by bacterial-fungal consortium (BFC) to survive under stress conditions can stabilize the pattern of BFC growth affected the rate of internal, external and global mass transfer (Peng et al., 2022; Wang et al., 2019). Even though the kinetics and mechanisms for adsorption, biosorption, precipitation, decolorization of the various pollutants processed using different types of the reactors have been previously suggested to evaluate the behaviors and mechanisms of external and internal mass transfer (Fulazzaky et al., 2013; 2017a; 2017b; Syafuddin and Fulazzaky, 2021), the degradation kinetics and mechanisms of mass transfer for the degraded-PE particles transported from an internal plastic waste to release into the environmental conditions of enriched with the BFC colonies are not fully

understood. The limitations of this study were only focused on the kinetics and mechanisms of mass transfer under a controlled environmental condition (CEC) while a change in the surface characteristics of plastic waste influenced an interpretation of the result findings were not investigated related to the physical structure and chemical composition of PE plastic waste before and after an intervention time of running the experiment for 150 days.

The objectives of this study are: (1) to scrutinize the biodegradation kinetics and mass transfer mechanisms of degraded-PE molecules mediated by the BFC colonies using the modified mass transfer factor (MMTF) models, (2) to analyze the variation of the internal with external mass transfer factor allowed to determine the resistance of mass transfer for biodegradation of PE plastic waste, and (3) to predict the performance of rectangular reactor (RR) aimed to assess the capability of BFC colonies as bio-mediator for enzymatic remediation of PE plastic waste in the environment.

## 2. Materials and methods

### 2.1. Cultivation of bacterial-fungal consortium

This study used the growth media of stone mineral salt solution consisting of the macro and micro nutrients of 0.5 g  $\text{CaCO}_3$ , 2.5 g  $\text{NH}_4\text{NO}_3$ , 1 g  $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5 g  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5 g  $\text{KH}_2\text{PO}_4$ , and 0.2 g  $\text{MnCl}_2 \cdot 7\text{H}_2\text{O}$  for the purpose of the cultivation of BFC colonies (Sharpley, 1996). The cultivation of BFC colonies used the mixture of *Clostridium* sp. and *Thiobacillus* sp. bacteria and *Dekkera* sp. yeast (BFC 135 strains) was conducted in an Erlenmeyer flask filled with 80% of growth media and 20% of BFC colonies under the CEC setting of 30 °C. The pour plate method was used for counting the number of colony-forming BFC presented in the petri dish containing of nutrient agar media. The number of BFC colonies was counted using the colony counter of Cole-Parmer (Cole-Parmer® CC-200 Series Stuart Digital Colony Counters, Illinois, USA) after 48 h of incubation and then calculated using the equation:  $N = N_d \times (1/F)$  where  $N$  is the number of BFC per mL,  $N_d$  is the number of BFC per petri dish and  $F$  is the dilution factor.

### 2.2. Laterite

The natural laterites were collected from an area within the campus of Universitas Trisakti in Gunung Putri, Bogor Regency, Indonesia. Approximately 3 kg of the laterites were crushed and then passed through 60 mesh with the sieve aperture size of 0.25 mm yielding the granular laterite size of below than 0.25 mm. Approximately 2 kg of the granular laterites were washed with distilled water and dried at 103 °C for 24 h in an oven and then sterilized at 70 °C for 2 h per day in an oven drying soil of Memmert (Universal oven UN30 - Memmert GmbH, Schwabach, Germany) for 3 days and then sieved to produce a uniform grain-size distribution of the laterites to allow more evenly occurred contact between laterite and BFC colony. Then the sterilization of laterites was performed to kill other microorganisms for allowing the biodegradation of PE plastic only mediated by the BFC colonies.

### 2.3. Rectangular reactor and operating conditions

This study used the RR acrylic glass with its dimension of 14×12×9 cm<sup>3</sup> filled 80% sterile granular laterites, 10% rice husk and 10% BFC colonies, as shown in Fig. 1. The addition of rice husk as a bulking agent aimed to increase the availability of carbon source can improve the porosity of mixed laterite-rice husk media leading to have an optimized growth of BFC colonies for more evenly. The biodegradation of PE plastic with a dimension of 1×1x 0.01 cm<sup>3</sup> was mediated by the BFC colonies under CEC that having a moisture content range of 25 - 28% and pH range of 7.4 - 7.7. The variation of humidity in the RR reaction tank was regularly monitored using the hygrometer of TFA-Haar



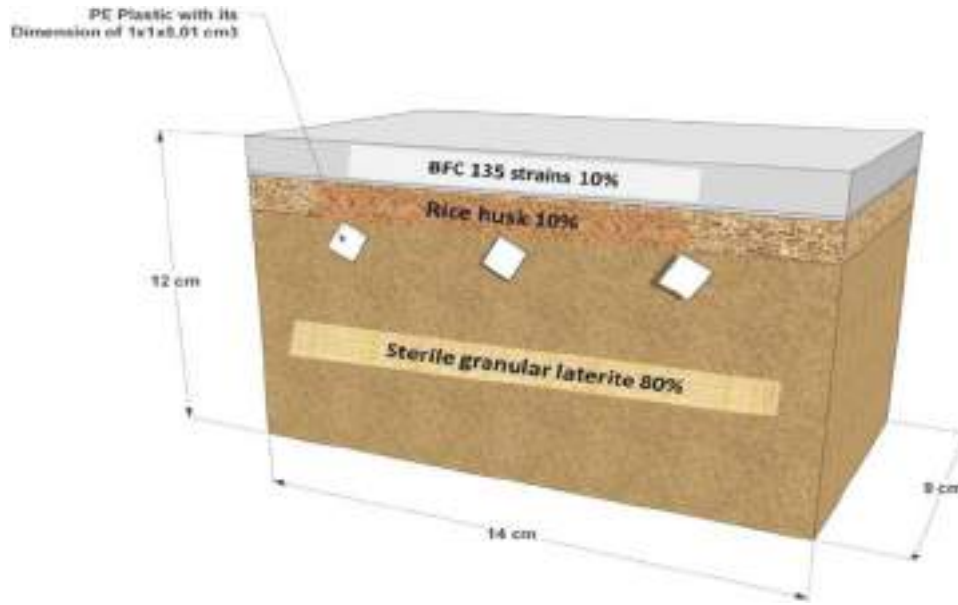


Fig. 1. Design of the rectangular reactor.

Synthetic during the period of the experiment. The variation of pH in the RR reaction tank was monitored using the digital soil pH meter of AMTAST AMT-300 at room temperature. The biodegradation of PE plastic in the RR reaction tank was monitored six times at 0, 30, 60, 90, 120 and 150th day of the experiment.

#### 2.4. Biodegradation kinetic equations and numerical simulation

The performance of RR process for biodegradation of PE plastic mediated by the BFC colonies can be calculated in the percentage of efficiency using the equation of:

$$E = \frac{W_o - W_t}{W_o} \times 100\% \quad (1)$$

where  $E$  is the PE degradation efficiency of the RR process (in %),  $W_o$  is the initial weight of PE in the RR reaction tank (in g),  $W_t$  is the weight of PE in the RR reaction tank at time  $t$  of the experiment (in g).

The accumulated amount of PE degraded by the BFC colonies can be calculated by analogy with the equation (Fulazzaky, 2011) of:

$$q = \int_0^t \frac{(W_o - W_s) dt}{W_o} \quad (2)$$

where,  $q$  is the accumulated amount of PE degraded by the BFC colonies (in g), and  $t$  is the effective time of running the RR experiment to degrade the PE plastic (in d).

This study using the modified mass transfer factor (MMTF) models, named also as the Generalized Fulazzaky (GF) equations, empirically developed using the data of PhD project by Fulazzaky et al., (2013). The forming of GF equations based on the Fulazzaky equations (Wajdi et al., 2023) formerly named as the mass transfer factor models theoretically developed by Fulazzaky (2011) have been used to describe the mechanisms and mass transfer kinetics for adsorption, biosorption, decolorization, precipitation of the pollutants (Fulazzaky et al., 2013; 2017a; 2017b; Syafiuddin and Fulazzaky, 2021). The mass transfer kinetics and mechanisms of PE degradation mediated by the BFC colonies can be investigated using the MMTF equations (Fulazzaky et al., 2013) of:

$$\ln\left(\frac{W_o}{W_t}\right) = [k_L a]_g \times e^{-\beta \times \ln(q)} \times t \quad (3)$$

where  $[k_L a]_g$  is the global mass transfer factor of degraded-PE

molecules released into the environment (in 1/d),  $\beta$  is the parameter of PE molecules affinity (in g d/mg) and  $t$  is the time of running the RR experiment (in d).

By simplifying Eq. (3) into the linear form can be written (Fulazzaky et al., 2013) as:

$$\ln(q) = \frac{1}{\beta} \times \ln(t) + B \quad (4)$$

with

$$B = \frac{\ln\left([k_L a]_g\right) - \ln\left(\ln\left(\frac{W_o}{W_t}\right)\right)}{\beta} \quad (5)$$

where  $B$  is the potential mass transfer index related to driving force of degraded-PE molecules transferred during the degradation of PE plastic mediated by the BFC colonies in the RR reaction tank (in mg/g).

The correlation between external mass transfer (EMT) factor and global mass transfer (GMT) factor has been formulated in the form of mathematical equation (Fulazzaky et al., 2013) of:

$$[k_L a]_f = [k_L a]_g \times e^{-\beta \times \ln(q)} \quad (6)$$

where  $[k_L a]_f$  is the external mass transfer factor of degraded-PE molecules released into the environment (in 1/d).

The variation of  $[k_L a]_g$  accorded to the  $W_o/W_s$  ratio can be calculated using Eq. (5) and that of  $[k_L a]_f$  accorded to an increased value of  $q$  can be calculated using Eq. (6) since the values of  $\beta$  and  $B$  have been verified from the linear graph of plotting  $\ln(q)$  versus  $\ln(t)$  accorded to Eq. (4).

The definition of internal mass transfer (IMT) factor is the GMT factor minus EMT factor can be written in the form (Fulazzaky et al., 2013) of:

$$[k_L a]_d = [k_L a]_g - [k_L a]_f \quad (7)$$

where  $[k_L a]_d$  is the internal mass transfer factor of degraded-PE molecules released into the environment (in 1/d).

The value of  $[k_L a]_d$ ,  $[k_L a]_f$  or  $[k_L a]_g$  can be plotted versus the time  $t$  of running the experiment since the variations of  $[k_L a]_g$ ,  $[k_L a]_f$  and  $[k_L a]_d$  have been obtained from numerical calculations of using Eq. (5), Eq. (6) and Eq. (7), respectively.

### 3. Results and discussion

#### 3.1. Analysis of polyethylene degradation

The biodegradation activity of most enzymes in fungi higher than that in bacteria (Sen and Raut, 2015) requires understanding of PE plastic degraded by the BFC colonies. The degradation rate of PE plastic mediated by the bacterial strains of *Bacillus licheniformis* mixed with *Achromobacter xylosoxidans* and that mediated by the fungal strains of *Aspergillus niger* mixed with *Aspergillus glaucus* isolated from soil of the plastic waste environment have been reported reaching at 32.2% and 40.0%, respectively, for 4 weeks of the incubation (Saeed et al., 2022). The performance of RR process for biodegradation of PE mediated by the BFC colonies can be evaluated from graph of plotting  $E$  versus  $t$  while the change in weight of PE plastic during the experimental period of 150 days can be verified from graph of plotting  $W$  versus  $t$ , as shown in Fig. 2. Empirical evidence (Fig. 2) shows that the degradation rate of PE mediated by the BFC colonies slowly increased by 8.8% from 0 to 8.8% during the first 30 days of the experiment could be higher than the PE degradation of 1.78% mediated by *Bacillus cereus* during the same period of 30 days (Kopecká et al., 2022). Then the degradation rate of PE quickly increases by 43.9% from 8.8 to 52.7% for 60 days of the experiment. Finally, the degradation rate of PE again slowly increases by 8.8% from 52.7 to 61.5% during the last 60 days of the experiment. This is related to the weight of PE decreased by 0.8 mg from 9.1 to 8.3 mg observed from 0 to 30th day and then decreased by 4 mg from 8.3 to 4.3 mg observed from 30th to 90th day and then decreased by 0.8 mg from 4.3 to 3.5 mg observed from 90th to 150th day of the experiment (see Fig. 2). The roughness of PE surface as shown in Fig. 3 gradually changes from a relative smooth of its initial observation at 0 day to very rough surface at 120th day of the experiment. This could be due to the weathering of PE plastic mediated by the BFC colonies is accompanied by a change in the surface morphology from a smooth to the roughness, especially microstructural features in the case of PE plastic. A decrease in the weight of PE caused by the transport of degraded-PE molecules from an internal diffusion of PE plastic migrating to the cell wall of BFC colonies could be due to the microbial enzymes involved in the catabolism of PE material to their constituent parts of small molecules, which form the substrates for metabolic pathways (Zeenat et al., 2021). The modification of molecular structure formed the mass molecular defects of releasing the simple chemical compounds such as carbonyl and hydrogen groups can occur during the biodegradation of PE plastic waste (Tao et al., 2020). The FTIR analysis of the functional groups could be useful to assess the biodegradation of PE mediated by the BFC colonies based on the observation of FTIR spectrum monitored at (a) 0, (b) 30th, (c) 60th, (d) 90th, and (e) 120th day of the experiment (see Fig. 4). Fig. 4a shows that spectrum of PE contains four strong bands located at  $2916\text{ cm}^{-1}$  and  $2848\text{ cm}^{-1}$  attributed to hydrogen group of CH stretch, at  $1472\text{ cm}^{-1}$  attributed to  $\text{CH}_2$  deformation/ $\text{CH}_2$  bend, and at  $718\text{ cm}^{-1}$  attributed to  $\text{CH}_2$  rock (Marinescu et al., 2021) and still

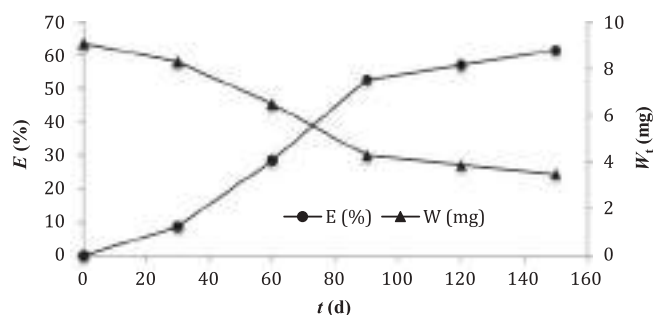


Fig. 2. Degradation rate and decreased weight of PE mediated by the BFC colonies under the experimental conditions of moisture content ranged from 25 to 28% and pH ranged from 7.4 to 7.7.

appear at the same locations in Figs. 4b, 4c, 4d, and 4e after the biodegradation of PE mediated by the BFC colonies during 30, 60, 90, and 120 days of the experiment, respectively. The absorption bands with their medium peaks shown in Fig. 4b appearing at around  $1033$ ,  $1009$ ,  $539$  and  $469\text{ cm}^{-1}$  after 30 days of the experiment could be attributed to Si-O bond vibrations (Ruiz et al., 2023). The appearance of the absorption bands at around  $1033$ ,  $1009$ ,  $912$ ,  $535$  and  $467\text{ cm}^{-1}$  assigned to Si-O-Al bond vibrations with their sharp peaks in Figs. 4c, 4d and 4e after 60, 90 and 120 days of the experiment, respectively, could be attributed to a change in the chemical characteristics of PE due to the biodegradation of PE mediated by the BFC colonies in the presence of laterite and rice husk can result in the breakdown of PE into monomers. The absorption bands with the peaks appearing at about  $1750\text{--}1600\text{ cm}^{-1}$  (see Fig. 4a, b, c, d) could be attributed to CO band stretching of carbonyl compounds (Mobaraki and Hemmateenejad, 2011).

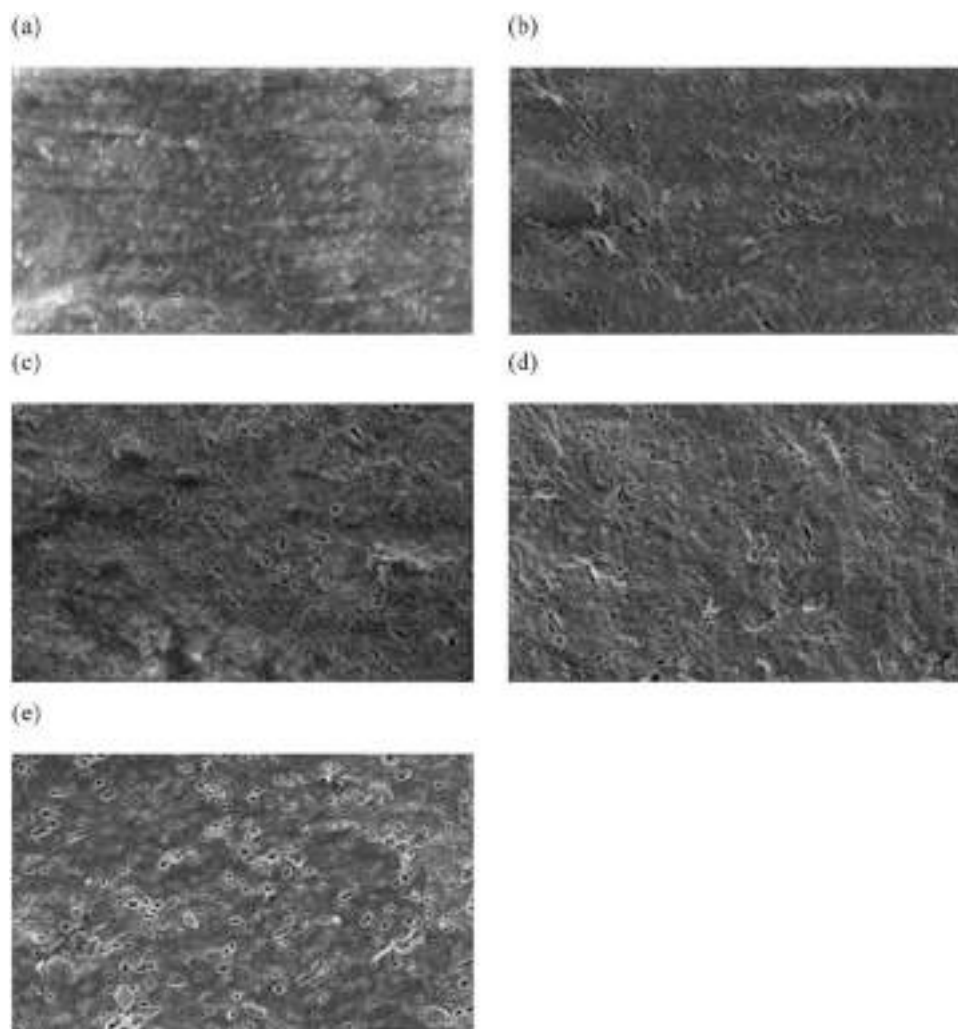
#### 3.2. Straight line graph of the linear function

The plot (Fig. 5) of  $\ln(q)$  versus  $\ln(t)$  accorded to Eq. (4) shows a linear graph yielding the  $1/\beta$  value of  $1.2458\text{ mg/g d}$  obtained from slope and the  $B$  value of  $0.4041\text{ mg/g}$  from the y-intercept. The linear equation of straight line graph showing a good fit to the experimental data with  $R^2 = 0.93754$  means that the use of the parameters  $\beta$  and  $B$  could be useful to predict the mass transfer kinetics of degraded-PE molecules transported from an internal diffusion within the PE plastic to the cell wall of BFC colonies in the RR reaction tank. The application of various methods has been proposed for the calculation of the kinetic parameters used to evaluate the biodegradation of plastic waste (Nisar et al., 2022a). The biodegradation rate of PE mediated by the BFC colonies is dependent on the CEC set of conditioning the PE plastic in the RR reaction tank. The use of rice husk conditioned with a granular laterite for supporting growth of BFC colonies can lead to influence the mechanisms and mass transfer kinetics of degraded-PE molecules, which perfectly control the performance of RR process. The development of BFC colonies effectively degraded the PE plastic could be favorably used the simple substrates of small molecules derived from molecular complexes of PE material when combined with the rice husk as carbon source of BFC metabolisms for the need of cell growth (Xiang et al., 2022).

The variations of the  $[k_L a]_d$ ,  $[k_L a]_f$  and  $[k_L a]_g$  values against the time period of the experiment can be separately traced since the values of  $\beta$  and  $B$  have been verified from the curve of plotting  $\ln(q)$  versus  $\ln(t)$ . Application of the MMTF models is important to predict the IMT mechanism of transporting small derived-PE molecules from an internal diffusion to the surface layer of PE plastic and then to predict the EMT mechanism of released-PE molecules transported from the surface of PE plastic to the cell surface of BFC colonies. An increase in the concentration of degraded-PE molecules accumulated outside of the PE plastic surface affecting driving force influences the behavior of EMT towards the cell wall of BFC colonies. The biodegradation mechanisms of PE mediated by the development of BFC colonies could be associated with the surface functional groups of BFC colonies consisted the various interactions by ion exchange, physical sorption and chemical bond (Gavrilescu, 2004). The cell wall of BFC colonies consisting of lipids, polysaccharides and proteins can offer the abundant functional groups of amino acid, carboxyl, hydroxyl, and phosphate yielding the abundant hydrophobic adsorption sites of aliphatic carbon chains and aromatic rings (Limo et al., 2018; Xiao et al., 2018). The cell surface hydrophobicity and cell surface polysaccharides have an important role in the development of BFC colonies during the biodegradation process of PE plastic in the RR reaction tank.

#### 3.3. Biodegradation kinetics of internal, external and global mass transfer

The biodegradation kinetics and mechanisms of the internal,

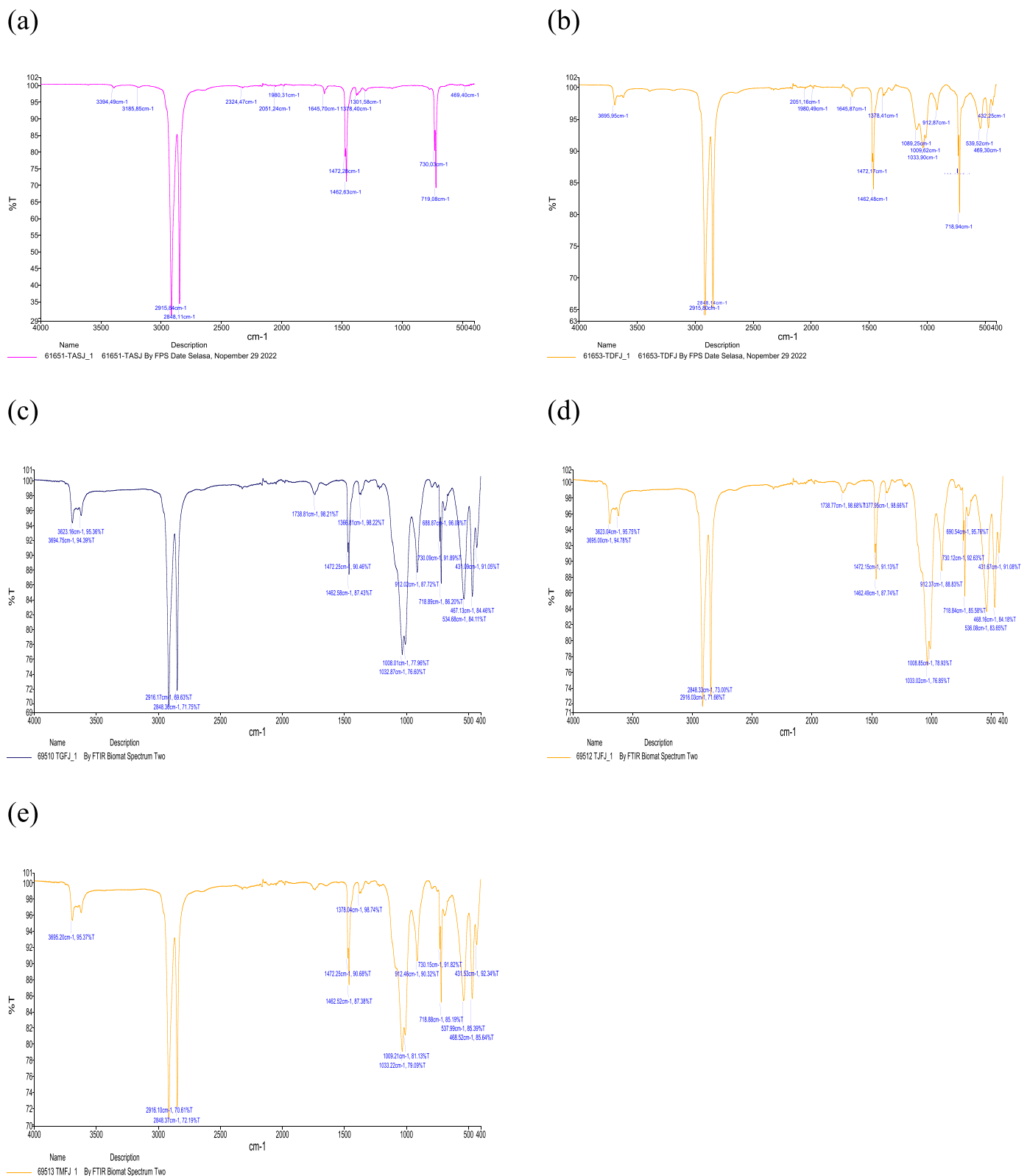


**Fig. 3.** SEM analysis of the PE surface for (a) observed at 0 day, (b) observed at 30th day, (c) observed at 60th day, (d) observed at 90th day, and (e) observed at 120th day of the experiment.

external, and global mass transfers followed from the graphs of plotting  $[k_L a]_d$ ,  $[k_L a]_f$  and  $[k_L a]_g$ , respectively, against  $t$  allow us to determine whether the resistance of mass transfer controlled by an internal diffusion or film mass transfer (Fulazzaky, 2011; Fulazzaky et al., 2013). The results (Fig. 6) show that the variations of  $[k_L a]_d$  and  $[k_L a]_g$  values are almost similar to each other and are far higher than the variation of  $[k_L a]_f$  value. A variation in the value of  $[k_L a]_d$  ranged 0.124 to 1.314 1/d with an average value of 0.819 1/d is very close to that of  $[k_L a]_g$  ranged 0.127 to 1.322 1/d with an average value of 0.825 1/d (see Fig. 6a) and is very high compared to that of  $[k_L a]_f$  ranged 0.004 to 0.008 1/d with an average value of 0.006 1/d (see Fig. 6a; see also Fig. 6b). Empirical evidence shows that the resistance of mass transfer for the biodegradation of PE plastic mediated by the BFC colonies is dependent on EMT due to the variation of  $[k_L a]_d$  value is far higher than that of  $[k_L a]_f$  value. The rate-limiting step for development of BFC colonies related to an EMT rate could be dependent on the driving force, stagnant film thickness, and microbial activity subjected to the plasma membrane of BFC colonies (Silhavy et al., 2010). The biodegradation of PE caused the molecular decay of PE plastic moved from an internal diffusion to the extracellular accumulation on the cell wall of BFC colonies is controlled by the EMT rate. The driving force of degraded-PE molecules transported from the surface of PE plastic to plasma membrane of BFC colonies increases with increasing of the concentration of molecular decay suspended on the surface of PE plastic (Wajdi et al., 2023). Intracellular diffusion of decaying PE molecules competed with the biochemical

reaction and EMT resistance for the next step of developing the BFC colonies influences the variability of digestion kinetics (Li and Hu, 2020; Syafiuddin and Fulazzaky, 2021). It is suggested that an extracellular precipitation of the decaying PE molecules on the cell wall of BFC colonies is not only supported by the presence of rice husk in the RR tank but also by a moisture content that causing the uniform growth of BFC colonies.

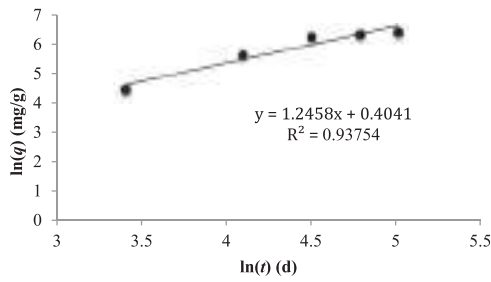
The primary concern of this work is the application of the MMTF models for scrutinizing the biodegradation kinetics of PE mediated by the BFC colonies in the RR tank. This aims to provide an insight into the dynamic behaviors of IMT and EMT for getting a new idea of the response mechanisms and mass transfer kinetics occurred during the biodegradation of PE plastic waste by the BFC colonies. The synergistic interactions, kinetic and thermodynamic analysis of the annual plastic debris could be important to determine the conversion rate of PE degradation (Galiwango and Gabbar, 2022). The purified hydrolases of heating a plastic waste exhibit a good hydrolysis activity for the degradation of high-temperature resistant PET plastic (Zhang et al., 2022). The potential growth of BFC colonies related to biodegradation of PE plastic processed in the RR tank could be supported by the addition of rice husk as carbon source. The effect of rice husk on the biodegradation rate of EMT supported the growth of BFC colonies allows to stimulate the digestion of organic matter transported from growth media to cellular tissue causing an increased EMT degradation rate of PE plastic. Empirical evidence shows that the variation of  $[k_L a]_f$  value increased from



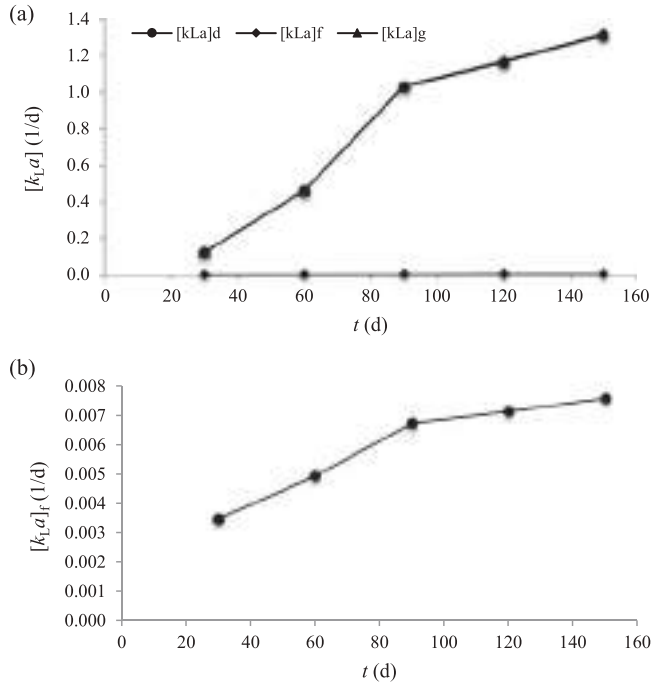
**Fig. 4.** FTIR analysis of functional groups for the biodegradation of PE mediated by the BFC colonies observed (a) at 0 day, (b) at 30th day, (c) at 60th day, (d) at 90th day, and (e) at 120th day of the experiment.

0.0035 1/d at the beginning to 0.0076 1/d after 150 days of the experiment. The biodegradation rate of IMT higher compared to that of EMT could be due to an increase in the carbon source of growth media leading to an increased internal diffusion of decaying PE molecules and then to hinder the movement of molecules passed through a film zone of

towards the plasma membrane of BFC colonies (Camper, 2004). Effect of soil content on the development of BFC colonies can lead to cause a great motive force in the transport of degraded-PE molecules to the cell surface of BFC colonies from the surface of PE plastic and then followed from the cell surface sorption to cellular accumulation of decaying PE



**Fig. 5.** Linear regression analysis of graphing  $\ln(q)$  versus  $\ln(t)$  to verify the values of  $\beta$  and  $B$  used for the calculation of the  $[k_{1a}]_d$ ,  $[k_{1a}]_f$  and  $[k_{1a}]_g$  variations over time of running the experiment.

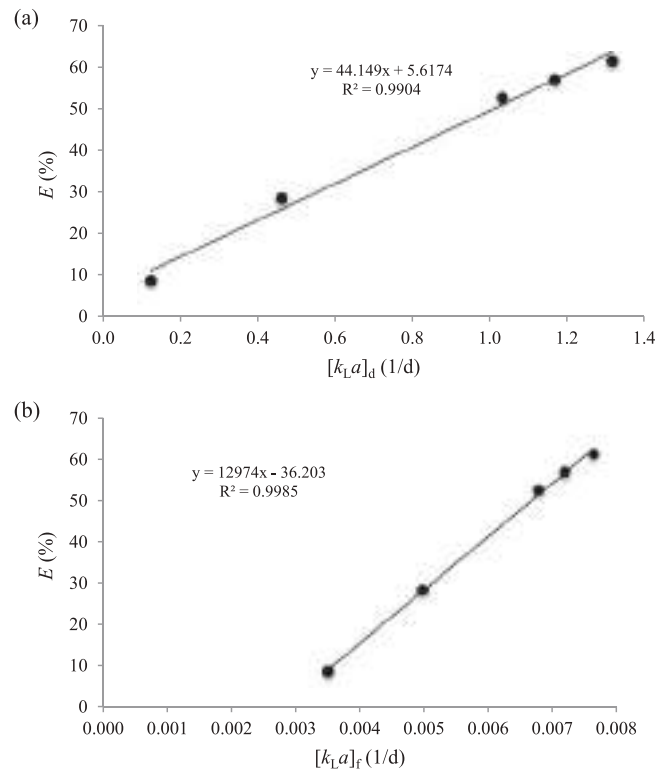


**Fig. 6.** Analysis of the biodegradation kinetics and mechanisms of the internal, external, and global mass transfers based on the variations of the  $[k_{1a}]_d$ ,  $[k_{1a}]_f$ , and  $[k_{1a}]_g$  values, respectively, over time of running the experiment; (a) a plot with the same scaling on y-axis and (b) a plot with detailed scaling on y-axis.

molecules (Fulazzaky et al., 2019). High biotic potential of PE plastic waste degradation used laterite as a living space to deliver food and energy shown by a rapid population growth of BFC colonies could be expected for an environmental condition of the mixed laterite-rice husk media. The role of rice husk enlarged porosity of growth media causing an increased sensitivity to the growth of BFC colonies has a great chance to maintain the appropriate biodegradation rate of PE molecules decay in the RR reaction tank (Yap et al., 2021). The average EMT kinetic rate of 0.006 1/d is far lower than the average IMT kinetic rate of 0.819 1/d for biodegradation of PE plastic and is thus required an improvement of CEC practice aimed to increase the EMT rate of transporting the decaying PE molecules from the surface of PE plastic to plasma membrane of BFC colonies.

### 3.4. Correlation of $E$ to $[k_{1a}]_d$ and $[k_{1a}]_f$

The numerical analysis of graphing  $E$  versus  $[k_{1a}]_d$  and  $E$  versus  $[k_{1a}]_f$  provides the best-fit linear trends of simulated data for scrutinizing the biodegradation of PE plastic mediated by the BFC colonies (see Fig. 7). The RR efficiency of PE degradation under an appropriate



**Fig. 7.** Numerical analysis of PE degradation mediated by the BFC colonies in the RR treatment system under the optimal control of environmental conditions with (a) the efficiency of PE degradation related to the internal diffusion of decaying PE molecules and (b) the efficiency of PE degradation related to the external transport of released PE molecules.

CEC set increased with increasing of the IMT and EMT factors can be expressed in the linear equation of  $E = a [k_{1a}]_{d(or f)} + b$  with  $a$  as slope and  $b$  as the y-intercept. The unit of  $a$  expressed in % d and that of  $b$  expressed in % are defined to have the same definite magnitude of quantity with the unit of  $E$ , which is expressed in %. This linear function equation shown a strong correlation of both  $E$  to  $[k_{1a}]_d$  and  $E$  to  $[k_{1a}]_f$  with  $R^2 > 0.99$  (see Table 1) can be used to predict the performance of RR process in accordance with the value of the IMT and EMT factors. The performance of RR process naturally degraded the PE plastic can be verified at approximately 5.6% of the efficiency (see Fig. 7a). Many types of microorganisms naturally adapted with an environment can grow and act to degrade decaying molecules of PE and this influences the kinetic rates of IMT and EMT in the release of decaying PE molecules supported by other elements and the positive interactions among microbes (Li et al., 2022; Verla et al., 2019). The presence of BFC colonies aimed to boost the performance of RR process can increase the kinetic rate of IMT to transport decaying PE molecules from an internal diffusion to the surface layer of PE plastic. Empirical evidence shows an increase in the value of  $[k_{1a}]_d$  by 1.190 1/d from 0.124 1/d at 30th day to 1.314 1/d at 150th day of the experiment (see Fig. 7a). The acceleration of EMT to transport decaying PE molecules through a film zone closely near the surface of PE plastic to cell surface sorption of the BFC colonies resulted in an increased performance of RR process can be verified from

**Table 1**

Numerical values of the parameters:  $a$ ,  $b$  based on  $E = a [k_{1a}]_d + b$  and  $E = a [k_{1a}]_f + b$ .

Linear equation	Numerical value of parameter		$R^2$
	$a$ (% d)	$b$ (%)	
$E = a [k_{1a}]_d + b$	44	5.6	0.99042
$E = a [k_{1a}]_f + b$	12,974	-36.2	0.99852



the value of  $[k_L a]_f$  slightly increased from 0.006 1/d at 30th day to 0.008 1/d at 150th day of the experiment (see Fig. 7b). Effect of BFC development on the acceleration of GMT (IMT + EMT) can improve the performance of RR process by 52.7% from 8.8% at 30th day to 61.5% of the degradation efficiency at 150th day of the experiment. The performance of RR process to microbiologically degrade the PE plastic waste reaches 61.5% of its maximum efficiency for the release of decaying PE molecules mediated by the BFC colonies obtained at 1.314 1/d of the IMT and 0.008 1/d of the EMT rate. This could be due to the porosity of the mixed laterite-rice husk media has an important role to increase an availability of carbon source speeding the metabolism of BFC colonies in the RR tank and takes place independently from biotic and abiotic environmental factors (Chen et al., 2022).

The RR performance of 100% predicted using the linear equation of  $E = a [k_L a]_d + b$  can be expected at the IMT rate of 2.138 1/d and the EMT rate of 0.011 1/d under an appropriate CEC setting of the experiment. It is suggested that the performance of RR process can be improved by setting an appropriate CEC of conditioning the experiment with the selection and adaptation of the most reliable BFC colonies. The use of GF equations to describe the biodegradation rate of decaying PE molecules mass transfer allows us to get better understanding of the future plastic waste management policies (Lam et al., 2018). The calculation of the kinetic parameters using various kinetic models has been suggested to evaluate the decomposition reaction of PE material in the presence of catalyst (Nisar et al., 2022b). The influence of different CEC factors of setting an experiment for degrading the PE plastic waste contributes to the EMT and IMT degradation rates of PE in an environment (Chamas et al., 2020). The mechanisms of biologically degraded the plastic wastes of PE by the *Plodia interpunctella* strains and polystyrene by the *Tenebrio molitor* strains indicate a variation in the effect of plastic degradation on the gut microbial community of larvae (Navlekar et al., 2023). The physiological response of mealworm larvae to the biodegradation of PE plastic waste has been investigated providing an insight to get better understanding on the physiological response of an invertebrate after plastic ingestion (Peng et al., 2023). An effort to increase the performance of RR process suggested by improving the mobilization of decaying PE molecules transported from the surface of PE plastic to cell wall of BFC colonies could be due to the biodegradation of PE plastic controlled by an EMT rate with its  $[k_L a]_f$  value of 0.008 1/d is below than that supported by an IMT rate with its  $[k_L a]_d$  value of 1.314 1/d observed at 150th day of the experiment.

#### 4. Conclusions

The analysis of linear function used the curve of  $\ln(q)$  versus  $\ln(t)$  showing the best-fit kinetic parameters of  $\beta$  and  $B$  with  $R^2 > 0.93754$  could be able to predict the biodegradation kinetics of decaying PE molecules mass transfer. The biodegradation mechanisms of PE plastic mediated by the BFC colonies analyzed using the values of  $[k_L a]_d$ ,  $[k_L a]_f$  and  $[k_L a]_g$  to predict the kinetic rates of IMT, EMT and GMT could be able to determine the resistance of mass transfer for decaying PE molecules depended on EMT. The combination of laterite with rice husk as the culture media to support the growth of microorganisms has an important role to improve the porosity of growth media and to increase the availability of carbon source for ensuring an effective metabolic acceleration of the BFC colonies. An effort to increase the performance of PE degradation at 100% of the efficiency can be suggested by adjusting the operation of RR at an IMT rate of 2.138 1/d and the EMT rate of 0.011 1/d. The numerical analysis of PE degradation mediated by the BFC colonies limited to the mechanisms and kinetics of mass transfer simulated using the MMTF models contributes to an enrichment of the references for the further researches of bacterial-fungal biodegradation of the plastic waste.

#### Statement

During the preparation of this work the author(s) used Microsoft Excel in order to simulate the experimental data using the modified mass transfer factor models. After using this tool, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

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#### CRediT authorship contribution statement

**Rinanti Astri:** Project administration, Methodology, Formal analysis, Data curation, Conceptualization. **Fulazzaky Mohamad Ali:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Formal analysis, Conceptualization. **Fachrul Melati Ferianita:** Writing – original draft, Resources, Project administration, Methodology, Data curation, Conceptualization. **Sunaryo Thalia:** Project administration, Methodology, Funding acquisition, Formal analysis, Data curation. **Tazkiaturrizki Tazkiaturrizki:** Resources, Project administration, Funding acquisition, Formal analysis, Data curation. **Muda Khalida:** Supervision, Resources, Investigation, Formal analysis.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

The data that has been used is confidential.

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# Paper 3

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## Mass transfer kinetics of polyethylene degradation by bacterial-fungal consortium

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### ABSTRACT

Understanding the biodegradation rate of polyethylene (PE) plastic waste mediated by bacterial-fungal consortium (BFC) is important to ensure effective design process of the bioremediation technology. The aims of this study were to scrutinize the behavior of PE plastic degradation mediated by the BFC colonies numerically simulated the experimental data using the modified mass transfer factor models and to analyze the kinetics and mechanics of internal, external and global mass transfer. The performance of rectangular reactor (RR) to biologically degrade the PE plastic (scraper) up to 61.5% shows an increased efficiency of 55.9% stimulated by the presence of BFC colonies. In the variation of internal mass transfer is almost the same with that of global mass transfer and is far higher than that of external mass transfer (EMT). The rate-limiting step of PE plastic degradation is dependent on the resistance of EMT. The application of BFC colonies aimed to improve the biodegradation rate of PE plastic waste contributes to advancing the future environmental engineering technologies.

### 1. Introduction

The production of plastics increased during the last few decades has caused an increased global plastic waste of up to around 6300 million tons (Zhang et al., 2020; Zhang et al., 2021). Of 8300 million tons plastics produced in 2017 has caused approximately 60% of the plastic waste accumulated into the environment with 95% of it ended up in the landfills and 5% of it ended up in the oceans and other terrestrial areas (Mohanani et al., 2020). An increased amount of plastic waste with its recalcitrant nature accumulated in the environment could be due to the uncontrolled use and disposal of the plastic materials (Neuber et al., 2021). An increased contamination rate of plastic waste caused by loss of the most recognizable and abundant film may threaten the viability of recycling pathway and requires a complete specification of the recycling stream with a novel level of details (Moert et al., 2021; Sreenivasagan et al., 2022). Some of the most common types of thermoplastic produced are polyethylene (PE), polypropylene, polystyrene, polycarbonate, and PE terephthalate; however, the plastic type of PE is widely used for kitchen utensils,

packaging materials and disposable beverage cups (Ruchman et al., 2015). The production of plastic materials increased during the early period of Covid-19 pandemic was due to produce the personal protection equipment of such as gloves, face shields, masks and single-use plastic products contributing to generate more plastic waste (Shilpa et al., 2022). The use of PE mulch as a tool in the agricultural practices could be useful to reach growers production goals resulted from greater nutrient uptake causing its growing faster to boost earlier ripening and higher yield of fruit (Kastirajan and Ngesajin, 2012). Reasons for the application of PE plastic mulch in the agricultural practices include controlling weed growth, stabilizing soil temperature and soil moisture, improving fertility and health of the soil, and enhancing the visual appeal of the area (Andriana et al., 2019). Transportation cost of wasted PE plastic mulching reduced by reducing the level of impurity content promotes sustainable management of plastic pollution in the rural areas (Dang et al., 2022).

Long-term exposure of plastics and plastic products to an aquatic environment can lead to a negative effect on the living organisms in an aquatic ecosystem (Rao and Geckeler, 2011). The design and

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optimization of the PE plastic waste analyzed using semi-detailed kinetic model has been put forward to support thermochemical recycling technologies (Iccampi et al., 2023). The release of PE plastic waste into the environment can lead to leaching of toxic chemicals absorbed by biota tissue, organs, and even cells of living organisms causing adverse effects on human health (Chen et al., 2022; Yuan et al., 2022). The presence of plastic waste in the environment can be biologically degraded through the mechanisms of hydrolytic degradation, photodegradation, thermo-oxidative degradation and biodegradation (Dimassi et al., 2022). Scalability of the plastic degradation processes in scientific research needs to be integrated the efforts of state, universities and civil society in the construction of circular ecosystem infrastructures for addressing the various lab-scale prototypes and industrial applications for the management of plastic waste (da Silva et al., 2022). Efforts in making many microorganisms propitious for green chemistry to eliminate harmful plastics from the terrestrial and aquatic ecosystems are suggested by evolving the strategies of using the appropriate organisms to survive for effectively degrading the plastic waste (Duan and Welden, 2023; Zeng et al., 2021). The utilization of thermophilic microbial consortia originated from cow dung as the engineering microbes can enhance the biodegradation of low- and high-density PE (Skiriyachan et al., 2017). The application of the mesophilic mixed bacteria isolated from the leachate of municipal landfill can biologically utilize the PE particles as a sole carbon source to decompose the PE plastic waste during the test period (Park and Kim, 2019). The performance of microbial strains naturally occurred whether in the soil, activated sludge, farm sludge, or worms' excreta was effective to degrade the plastic waste without an inhibition of microbial growth caused by the derived by-products from microbial degradation (Taghavi et al., 2021). The use of fungal laccases and peroxidases could be useful for depolymerization of PE plastic waste (Eidin et al., 2022). The potential of oil-seep-contaminated ecosystem for plastic degradation acquired following a long-term adaptation to petroleum compounds has been indicated the diversity of microbial community and the presence of polyethylene terephthalate (PET) degrading bacteria (Baharizadeh et al., 2023).

The kinetics and mechanisms of mass transfer for analyzing the movement of degraded-PE particles controlled by either internal diffusion or film mass transfer, which can determine the resistance of mass transfer, could be due to the interdependent biological, chemical and physical processes regulate the species distribution and partitioning of biologically reactive particles (Dhar et al., 2020; Fulazzaky et al., 2013, 2017a). The transport of degraded-PE particles from an internal diffusion to film zone at the surface of PE plastic is influenced by molecular bonding and then from a film zone to cell wall of the microorganisms influenced by the environmental conditions (Rinda et al., 2021; Lim and Thian, 2022). The internal and external mass transfer of degraded-PE particles released from a PE plastic waste could be affected by the experimental factors of pH, temperature, moisture content, porosity of culture media and biological growth (Fulazzaky, 2011). The porosity of growth media increased when mixing with rice husk could be due to the pore widening by smaller average particle size of rice husks than laterite causing an increased moisture content of the mixed-media affected by increased surface contact (Oishi and Yagawa, 2020). An increase in the availability of carbon source in the interior channels of growth media causing the utilization of carbon by bacterial-fungal consortium (BFC) to survive under stress conditions can stabilize the pattern of BFC growth affected the rate of internal, external and global mass transfer (Perez et al., 2022; Wang et al., 2019). Even though the kinetics and mechanisms for adsorption, biosorption, precipitation, decolorization of the various pollutants processed using different types of the reactors have been previously suggested to evaluate the behaviors and mechanisms of external and internal mass transfer (Fulazzaky et al., 2013, 2017a, 2017b; Syaifuldin and Fulazzaky, 2021), the degradation kinetics and mechanisms of mass transfer for the degraded-PE particles transported from an internal plastic waste to release into the environmental conditions of enriched with the BFC colonies are not fully

understood. The limitations of this study were only focused on the kinetics and mechanisms of mass transfer under a controlled environmental condition (CEC) while a change in the surface characteristics of plastic waste influenced an interpretation of the result findings were not investigated related to the physical structure and chemical composition of PE plastic waste before and after an intervention time of running the experiment for 150 days.

The objectives of this study are: (1) to scrutinize the biodegradation kinetics and mass transfer mechanisms of degraded-PE molecules mediated by the BFC colonies using the modified mass transfer factor (MMTF) models, (2) to analyze the variation of the internal with external mass transfer factor allowed to determine the resistance of mass transfer for biodegradation of PE plastic waste, and (3) to predict the performance of rectangular reactor (RR) aimed to assess the capability of BFC colonies as bio-mediator for enzymatic remediation of PE plastic waste in the environment.

## 2. Materials and methods

### 2.1. Cultivation of bacterial-fungal consortium

This study used the growth media of sterilized mineral salt solution consisting of the macro and micro nutrients of 0.5 g  $\text{CaCO}_3$ , 2.5 g  $\text{NH}_4\text{NO}_3$ , 1 g  $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5 g  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5 g  $\text{K}_2\text{H}_2\text{PO}_4$  and 0.2 g  $\text{MnCl}_2 \cdot 7\text{H}_2\text{O}$  for the purpose of the cultivation of BFC colonies (Sharpley, 1996). The cultivation of BFC colonies used the mixture of *Clostridium* sp. and *Thiobacillus* sp. bacteria and *Debkeria* sp. yeast (BFC 135 strains) was conducted in an Erlenmeyer flask filled with 80% of growth media and 20% of BFC colonies under the CEC setting of 30 °C. The pour plate method was used for counting the number of colony-forming BFC presented in the petri dish containing of nutrient agar media. The number of BFC colonies was counted using the colony counter of Cole-Parmer (Cole-Parmer® CC-200 Series Stuart Digital Colony Counter, Illinois, USA) after 48 h of incubation and then calculated using the equation:  $N = N_0 \times (1/F)$  where  $N$  is the number of BFC per mL,  $N_0$  is the number of BFC per petri dish and  $F$  is the dilution factor.

### 2.2. Laterite

The natural laterites were collected from an area within the campus of Universitas Triakti In Gading Putri, Bogor Regency, Indonesia. Approximately 3 kg of the laterites were crushed and then passed through 60 mesh with the sieve aperture size of 0.25 mm yielding the granular laterite size of below than 0.25 mm. Approximately 2 kg of the granular laterites were washed with distilled water and dried at 103 °C for 24 h in an oven and then sterilized at 70 °C for 2 h per day in an oven drying soil of Memmert (Univenal oven UN30 - Memmert GmbH, Schwabach, Germany) for 3 days and then sieved to produce an uniform grain-size distribution of the laterites to allow more evenly occurred contact between laterite and BFC colony. Then the sterilization of laterites was performed to kill other microorganisms for allowing the biodegradation of PE plastic only mediated by the BFC colonies.

### 2.3. Rectangular reactor and operating conditions

This study used the RR acrylic glass with its dimension of 14 × 12 × 9 cm<sup>3</sup> filled 80% sterile granular laterites, 10% rice husk and 10% BFC colonies, as shown in Fig. 1. The addition of rice husk as a bulking agent aimed to increase the availability of carbon source can improve the porosity of mixed laterite-rice husk media leading to have an optimized growth of BFC colonies for more evenly. The biodegradation of PE plastic with a dimension of 1 × 1 × 0.01 cm<sup>3</sup> was mediated by the BFC colonies under CEC that having a moisture content range of 25 - 28% and pH range of 7.4 - 7.7. The variation of humidity in the RR reaction tank was regularly monitored using the hygrometer of TFA-Haar

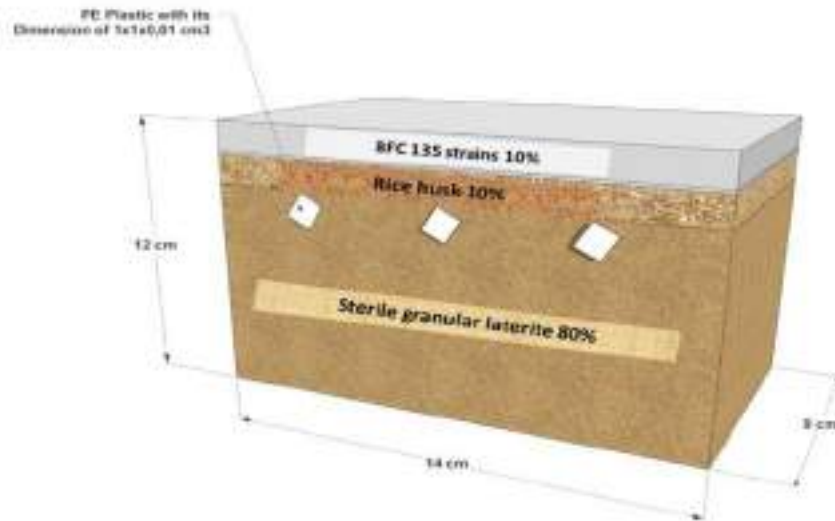


Fig. 1. Design of the rectangular reactor.

Synthetic during the period of the experiment. The variation of pH in the RR reaction tank was monitored using the digital soil pH meter of AMTAST AMT-300 at room temperature. The biodegradation of PE plastic in the RR reaction tank was monitored six times at 0, 30, 60, 90, 120 and 150th day of the experiment.

2.4. Biodegradation kinetic equations and numerical simulation

The performance of RR process for biodegradation of PE plastic mediated by the BFC colonies can be calculated in the percentage of efficiency using the equation of:

$$E = \frac{W_0 - W_t}{W_0} \times 100\% \tag{1}$$

where  $E$  is the PE degradation efficiency of the RR process (in %),  $W_0$  is the initial weight of PE in the RR reaction tank (in g),  $W_t$  is the weight of PE in the RR reaction tank at time  $t$  of the experiment (in g).

The accumulated amount of PE degraded by the BFC colonies can be calculated by analogy with the equation (Fulazzaky, 2011) of:

$$q = \int_0^t \frac{(W_0 - W_t) dt}{W_0} \tag{2}$$

where,  $q$  is the accumulated amount of PE degraded by the BFC colonies (in g), and  $t$  is the effective time of running the RR experiment to degrade the PE plastic (in d).

This study using the modified mass transfer factor (MMTF) models, named also as the Generalized Fulazzaky (GF) equations, empirically developed using the data of PhD project by Fulazzaky et al. (2013). The forming of GF equations based on the Fulazzaky equations (Wajdi et al., 2022) formerly named as the mass transfer factor models theoretically developed by Fulazzaky (2011) have been used to describe the mechanisms and mass transfer kinetics for adsorption, biosorption, decolorization, precipitation of the pollutants (Fulazzaky et al., 2013; 2017a; 2017b; Syaifuldin and Fulazzaky, 2021). The mass transfer kinetics and mechanisms of PE degradation mediated by the BFC colonies can be investigated using the MMTF equations (Fulazzaky et al., 2013) of:

$$\ln\left(\frac{W_0}{W_t}\right) = [k_1\alpha]_g \times e^{-\beta W_0/q} \times t \tag{3}$$

where  $[k_1\alpha]_g$  is the global mass transfer factor of degraded-PE

molecules released into the environment (in 1/d),  $\beta$  is the parameter of PE molecules affinity (in g d/mg) and  $t$  is the time of running the RR experiment (in d).

By simplifying Eq. (3) into the linear form can be written (Fulazzaky et al., 2013) as:

$$\ln(q) = \frac{1}{\beta} \times \ln(t) + B \tag{4}$$

with

$$B = \frac{\ln\left([k_1\alpha]_g\right) - \ln\left(\ln\left(\frac{W_0}{W_t}\right)\right)}{\beta} \tag{5}$$

where  $B$  is the potential mass transfer index related to driving force of degraded-PE molecules transferred during the degradation of PE plastic mediated by the BFC colonies in the RR reaction tank (in mg.g).

The correlation between external mass transfer (EMT) factor and global mass transfer (GMT) factor has been formulated in the form of mathematical equation (Fulazzaky et al., 2013) of:

$$[k_1\alpha]_g = [k_1\alpha]_e \times e^{-\beta W_0/q} \tag{6}$$

where  $[k_1\alpha]_e$  is the external mass transfer factor of degraded-PE molecules released into the environment (in 1/d).

The variation of  $[k_1\alpha]_g$  according to the  $W_0/W_t$  ratio can be calculated using Eq. (5) and that of  $[k_1\alpha]_e$  according to an increased value of  $q$  can be calculated using Eq. (6) since the values of  $\beta$  and  $B$  have been verified from the linear graph of plotting  $\ln(q)$  versus  $\ln(t)$  according to Eq. (4).

The definition of internal mass transfer (IMT) factor is the GMT factor minus EMT factor can be written in the form (Fulazzaky et al., 2013) of:

$$[k_1\alpha]_i = [k_1\alpha]_g - [k_1\alpha]_e \tag{7}$$

where  $[k_1\alpha]_i$  is the internal mass transfer factor of degraded-PE molecules released into the environment (in 1/d).

The value of  $[k_1\alpha]_g$ ,  $[k_1\alpha]_e$ ,  $[k_1\alpha]_i$  can be plotted versus the time  $t$  of running the experiment since the variations of  $[k_1\alpha]_g$ ,  $[k_1\alpha]_e$  and  $[k_1\alpha]_i$  have been obtained from numerical calculations of using Eq. (5), Eq. (6) and Eq. (7), respectively.



### 3. Results and discussion

#### 3.1. Analysis of polyethylene degradation

The biodegradation activity of most enzymes in fungi higher than that in bacteria (Sen and Kasi, 2015) requires understanding of PE plastic degraded by the BFC colonies. The degradation rate of PE plastic mediated by the bacterial strains of *Bacillus licheniformis* mixed with *Achromobacter xylosoxidans* and that mediated by the fungal strains of *Aspergillus niger* mixed with *Aspergillus glaucus* isolated from soil of the plastic waste environment have been reported reaching at 32.2% and 40.0%, respectively, for 4 weeks of the incubation (Saeed et al., 2022). The performance of RR process for biodegradation of PE mediated by BFC colonies can be evaluated from graph of plotting  $E$  versus  $t$  while the change in weight of PE plastic during the experimental period of 150 days can be verified from graph of plotting  $W$  versus  $t$ , as shown in Fig. 2. Empirical evidence (Fig. 2) shows that the degradation rate of PE mediated by the BFC colonies slowly increased by 8.8% from 0 to 8.8% during the first 30 days of the experiment could be higher than the PE degradation of 1.78% mediated by *Bacillus cereus* during the same period of 30 days (Kopecky et al., 2022). Then the degradation rate of PE quickly increases by 43.9% from 8.8 to 52.7% for 60 days of the experiment. Finally, the degradation rate of PE again slowly increases by 8.8% from 52.7 to 61.5% during the last 60 days of the experiment. This is related to the weight of PE decreased by 0.8 mg from 9.1 to 8.3 mg observed from 0 to 30th day and then decreased by 4 mg from 8.3 to 4.3 mg observed from 30th to 90th day and then decreased by 0.8 mg from 4.3 to 3.5 mg observed from 90th to 150th day of the experiment (see Fig. 2). The roughness of PE surface as shown in Fig. 3 gradually changes from a relative smooth of its initial observation at 0 day to very rough surface at 120th day of the experiment. This could be due to the weathering of PE plastic mediated by the BFC colonies is accompanied by a change in the surface morphology from a smooth to the roughness, especially microstructural features in the case of PE plastic. A decrease in the weight of PE caused by the transport of degraded-PE molecules from an internal diffusion of PE plastic migrating to the cell wall of BFC colonies could be due to the microbial enzymes involved in the catabolism of PE material to their constituent parts of small molecules, which form the substrates for metabolic pathways (Zeman et al., 2021). The modification of molecular structure formed the molecular defects of releasing the simple chemical compounds such as carbonyl and hydrogen gas can occur during the biodegradation of PE plastic waste (Jao et al., 2020). The FTIR analysis of the functional groups could be useful to assess the biodegradation of PE mediated by the BFC colonies based on the observation of FTIR spectrum monitored at (a) 0, (b) 30th, (c) 60th, (d) 90th and (e) 120th day of the experiment (see Fig. 4). Fig. 4a shows that spectrum of PE contains four strong bands located at 2919  $\text{cm}^{-1}$  and 2848  $\text{cm}^{-1}$  attributed to hydrogen group of CH stretch, at 1472  $\text{cm}^{-1}$  attributed to  $\text{CH}_2$  deformation/ $\text{CH}_2$  bend, and at 718  $\text{cm}^{-1}$  attributed to  $\text{CH}_2$  rock (Martinez et al., 2021) and still

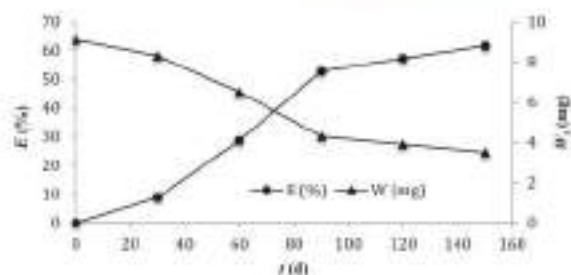


Fig. 2. Degradation rate and decreased weight of PE mediated by the BFC colonies under the experimental conditions of moisture content ranged from 25 to 29% and pH ranged from 7.4 to 7.7.

appear at the same locations in Figs. 4b, 4c, 4d, and 4e after the biodegradation of PE mediated by the BFC colonies during 30, 60, 90, and 120 days of the experiment, respectively. The absorption bands with their medium peaks shown in Fig. 4b appearing at around 1633, 1609, 539 and 469  $\text{cm}^{-1}$  after 30 days of the experiment could be attributed to Si-O bond vibrations (Ruzic et al., 2023). The appearance of the absorption bands at around 1633, 1609, 912, 535 and 467  $\text{cm}^{-1}$  assigned to Si-O-Al bond vibrations with their sharp peaks in Figs. 4c, 4d and 4e after 60, 90 and 120 days of the experiment, respectively, could be attributed to a change in the chemical characteristics of PE due to the biodegradation of PE mediated by the BFC colonies in the presence of laterite and rice husk can result in the breakdown of PE into monomers. The absorption bands with the peaks appearing at about 1750–1600  $\text{cm}^{-1}$  (see Fig. 4a, b, c, d) could be attributed to CO band stretching of carbonyl compounds (Mobaraki and Hermatenejad, 2011).

#### 3.2. Straight line graph of the linear function

The plot (Fig. 5) of  $\ln(q)$  versus  $\ln(t)$  accorded to Eq. (4) shows a linear graph yielding the  $1/n$  value of 1.2458  $\text{mg/g d}$  obtained from slope and the  $B$  value of 0.4041  $\text{mg/g}$  the y-intercept. The linear equation of straight line graph shows a good fit to the experimental data with  $R^2 = 0.93754$  means that the use of the parameters  $B$  and  $n$  could be useful to predict the mass transfer kinetics of degraded-PE molecules transported from an internal diffusion within the PE plastic to the cell wall of BFC colonies in the RR reaction tank. The application of various methods has been proposed for the calculation of the kinetic parameters used to evaluate the biodegradation of plastic waste (Nisar et al., 2022a). The biodegradation rate of PE mediated by the BFC colonies is dependent on the CEC set of conditioning the PE plastic in the RR reaction tank. The use of rice husk conditioned with a granular laterite for supporting growth of BFC colonies can lead to influence the mechanisms and mass transfer kinetics of degraded-PE molecules, which perfectly control the performance of RR process. The development of BFC colonies effectively degraded the PE plastic could be favourably used the simple substrates of small molecules derived from molecular complexes of PE material when combined with the rice husk as carbon source of BFC metabolites for the need of cell growth (Xiang et al., 2022).

The variations of the  $[k_1]_0$ ,  $[k_1]_t$  and  $[k_1]_e$  values against the time period of the experiment can be separately traced since the values of  $B$  and  $n$  have been verified from the curve of plotting  $\ln(q)$  versus  $\ln(t)$ . Application of the MMTF model is important to predict the IMT mechanism of transporting small derived-PE molecules from an internal diffusion to the surface layer of PE plastic and then to predict the EMT mechanism of released-PE molecules transported from the surface of PE plastic to the cell surface of BFC colonies. An increase in the concentration of degraded-PE molecules accumulated outside of the PE plastic surface affecting driving force influences the behavior of EMT towards the cell wall of BFC colonies. The biodegradation mechanisms of PE mediated by the development of BFC colonies could be associated with the surface functional groups of BFC colonies consisted the various interactions by ion exchange, physical sorption and chemical bond (Gavrilescu, 2004). The cell wall of BFC colonies consisting of lipids, polysaccharides and proteins can offer the abundant functional groups (amino acid, carboxyl, hydroxyl, and phosphate) yielding the abundant hydrophobic adsorption sites of aliphatic carbon chains and aromatic rings (Lima et al., 2011; Jao et al., 2018). The cell surface hydrophobicity and cell surface polysaccharides have an important role in the development of BFC colonies during the biodegradation process of PE plastic in the RR reaction tank.

#### 3.3. Biodegradation kinetics of internal, external and global mass transfer

The biodegradation kinetics and mechanisms of the internal,

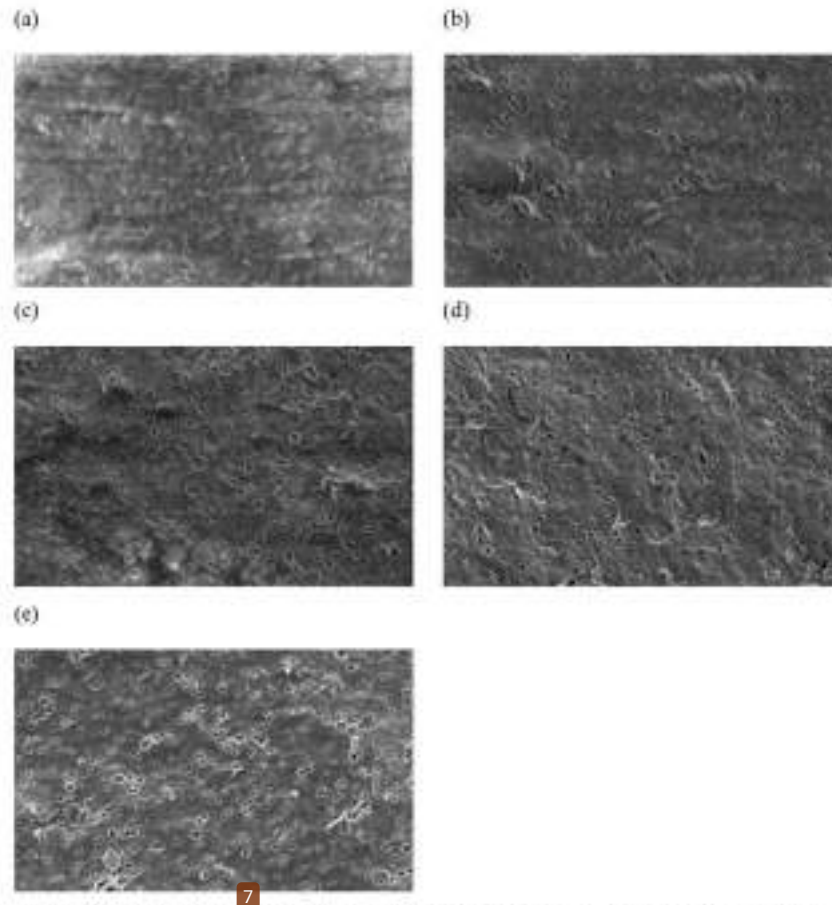


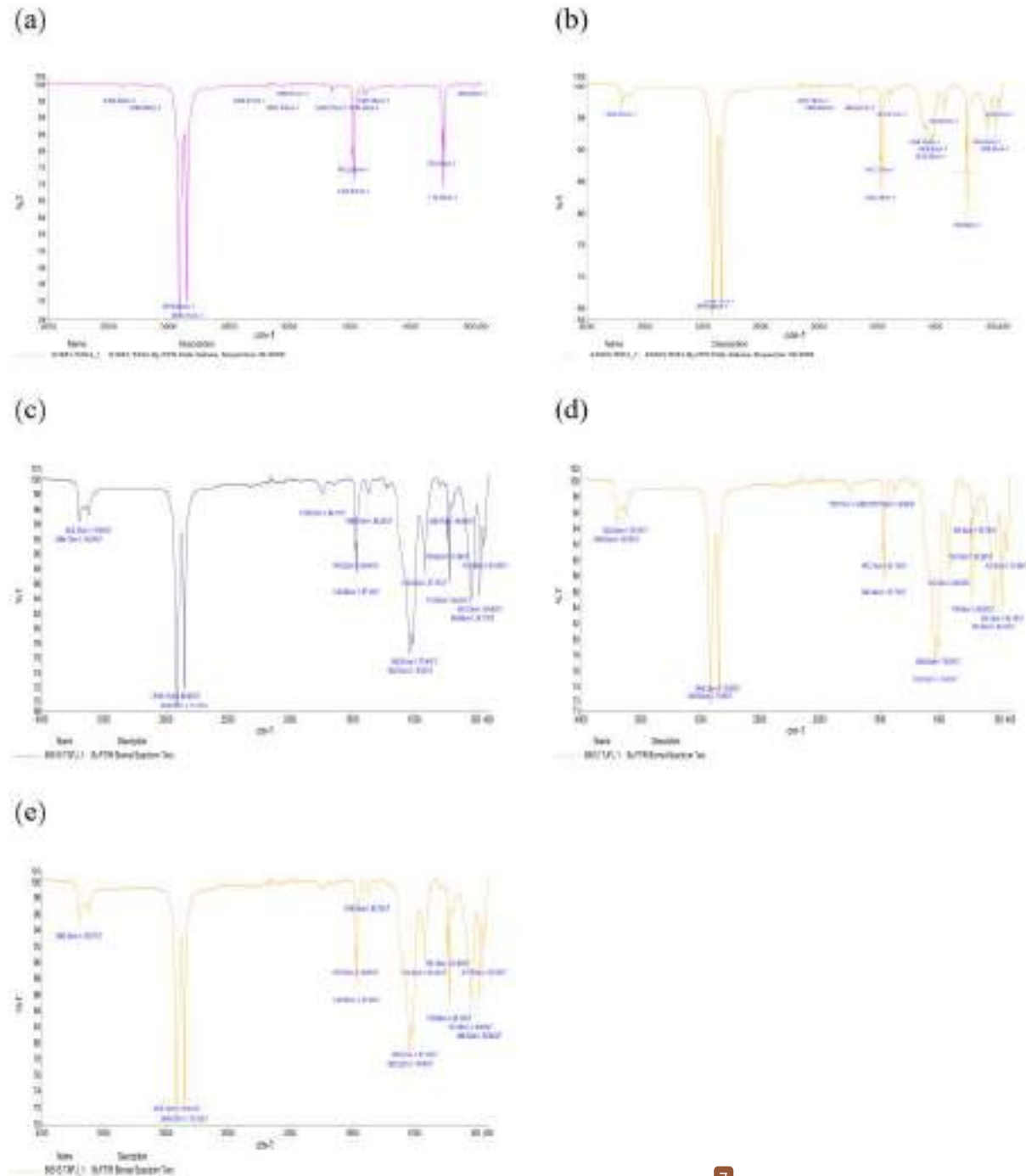
Fig. 3. SEM analysis of the PE surface for (a) observed at 0 day, (b) observed at 20th day, (c) observed at 60th day, (d) observed at 90th day, and (e) observed at 120th day of the experiment.

9 internal, and global mass transfers followed from the graph 3 of plotting  $[k_{1,0}]_d$ ,  $[k_{1,0}]_f$  and  $[k_{1,0}]_g$ , respectively, against  $\tau$  allow us to determine whether the resistance of mass transfer controlled by an internal diffusion or film mass transfer (Pulazzaky, 2011; Pulazzaky et al., 2013). The results (Fig. 5) show that the variation 27  $[k_{1,0}]_d$  and  $[k_{1,0}]_g$  values are almost similar to each other and are far higher than the variation of  $[k_{1,0}]_f$  value. A variation in the value of  $[k_{1,0}]_d$  ranged 0.124 to 1.314 1/d with an average value of 0.819 1/d is very close to that of  $[k_{1,0}]_g$  ranged 0.127 to 1.322 1/d with an average value of 0.825 1/d (see Fig. 5a) and is very high compared to that of  $[k_{1,0}]_f$  ranged 0.004 to 0.008 1/d with an average value 0.006 1/d (see Fig. 5a; see also Fig. 6b). Empirical evidence shows that the resistance of mass transfer for the biodegradation 45 PE plastic mediated by the BFC colonies is independent on EMT due to the variation of  $[k_{1,0}]_d$  value is far higher than that of  $[k_{1,0}]_f$  value. The rate-limiting step for development of BFC colonies related to an EMT rate could be dependent on the driving force, stagnant film thickness, and microbial activity subjected in the plasma membrane of BFC colonies (Silhavy et al., 2010). The biodegradation of PE caused the molecular decay of PE plastic moved from an internal diffusion in the extracellular accumulation on the cell wall of BFC colonies is controlled by the EMT rate. The driving force of degraded-PE molecules transported from the surface of PE plastic to plasma membrane of BFC colonies increases with increasing of the concentration of molecular decay suspended on the surface of PE plastic (Wajidi et al., 2023). Intracellular diffusion of decaying PE molecules competed with the biochemical

reaction and EMT resistance for the next step of developing the BFC 25 colonies influences the variability of digestion kinetics (Li and Hu, 2020; Syahudin and Pulazzaky, 2021). It is suggested that an extracellular precipitation of the decaying PE molecules on the cell wall of BFC colonies is not only supported by the presence of rice husk in the RR tank but also by a moisture content that causing the uniform growth of BFC colonies.

The primary concern of this work is the application of the MMDF 20 models for scrutinizing the biodegradation kinetics of PE mediated by the BFC colonies in the RR tank. This aims to provide an insight into the dynamic behavior of IMT and EMT for getting a new idea of the response mechanisms and mass transfer kinetics occurred during the biodegradation of PE plastic waste by the BFC colonies. The synergistic interactions, kinetic and the 56 dynamic analysis of the overall plastic debris could be important to determine the conversion rate of PE degradation (Galivango and Gabbar, 2022). The purified hydrolases of heating a plastic waste exhibit a good hydrolysis activity for the degradation of high-temperature resistant PET plastic (Zhang et al., 2022). The potential growth of BFC colonies related to biodegradation of PE plastic processed in the RR tank could be supported by the addition of rice husk as carbon source. The effect of rice husk on the biodegradation rate of EMT supported the growth of BFC colonies allows to stimulate the digestion of organic matter transported from growth media to cellular tissue causing an increased EMT degradation rate of PE plastic. Empirical evidence shows that the variation of  $[k_{1,0}]_f$  value increased from

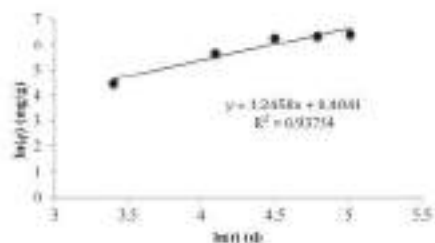




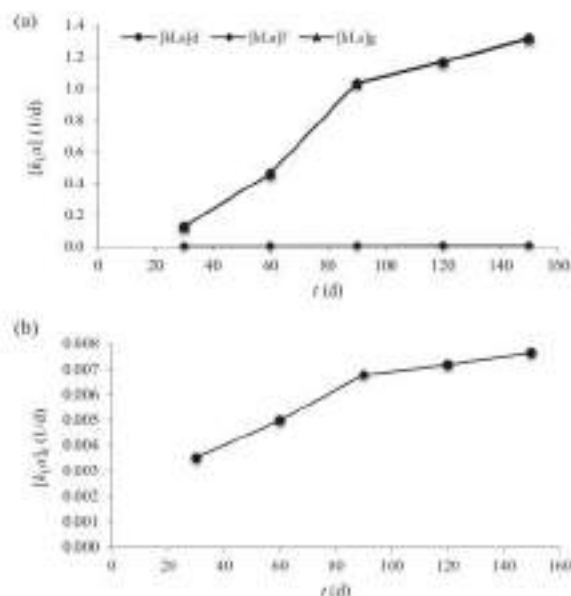
**Fig. 4.** FTIR analysis of functional groups for the biodegradation of PE mediated by the BFC colonies observed (a) at 0 day, (b) at 30th day, (c) at 60th day, (d) at 90th day, and (e) at 120th day of the experiment.

0.0035 1/d at the beginning to 0.0076 1/d after 150 days of the experiment. The biodegradation rate of IMT higher compared to that of EMT could be due to an increase in the carbon source of growth media leading to an increased internal diffusion of decaying PE molecules and then to hinder the movement of molecules passed through a film zone of

towards the plasma membrane of BFC colonies (Comper, 2004). Effect of soil content on the development of BFC colonies can lead to cause a great motive force in the transport of degraded-PE molecules to the cell surface of BFC colonies from the surface of PE plastic and then followed from the cell surface sorption to cellular accumulation of decaying PE



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63 **5.** Linear regression analysis of graphing  $\ln(q)$  versus  $\ln(t)$  to verify the values of  $\beta$  and  $B$  used for the calculation of the  $[k_1\alpha]_D$ ,  $[k_1\alpha]_I$  and  $[k_1\alpha]_E$  variations over time of running the experiment.

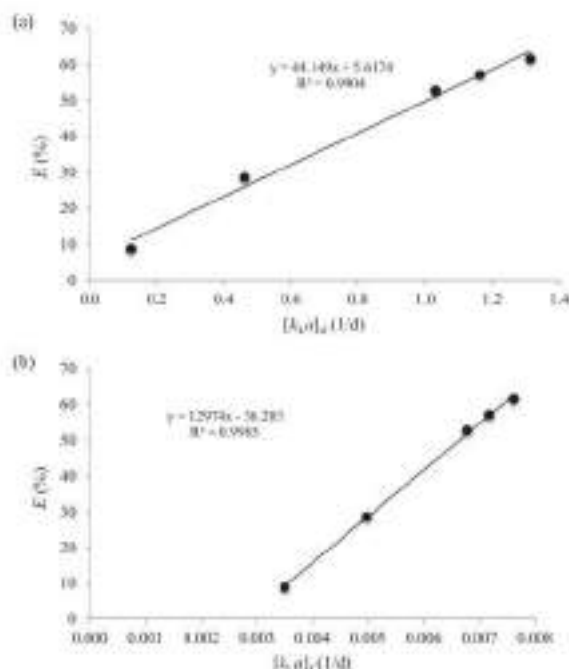


**Fig. 6.** Analysis of the biodegradation kinetics and mechanisms of the internal, external, and global mass transfers based on the variations of the  $[k_1\alpha]_D$ ,  $[k_1\alpha]_I$ , and  $[k_1\alpha]_E$  values, respectively, over time of running the experiment; (a) a plot with the time scaling on y-axis and (b) a plot with detailed scaling on y-axis.

molecules (Pulamazky et al., 2019). High biotic potential of PE plastic waste degradation used laterite as a living space to deliver food and energy shown by a rapid population growth of BFC colonies could be expected for an environmental condition of the mixed laterite-rice husk media. The role of rice husk enlarged porosity of growth media causing an increased sensitivity to the growth of BFC colonies has a great chance to maintain the appropriate biodegradation rate of PE molecules decay in the RR reaction tank (Yap et al., 2021). The average EMT kinetic rate of 0.006 1/d is far lower than the average IMT kinetic rate of 0.819 1/d for biodegradation of PE plastic and is thus required an improvement of CEC practice aimed to increase the EMT rate of transporting the decaying PE molecules from the surface of PE plastic to plasma membrane of BFC colonies.

#### 3.4. Correlation of $E$ to $[k_1\alpha]_D$ and $[k_1\alpha]_I$

The numerical analysis of graphing  $E$  versus  $[k_1\alpha]_D$  and  $E$  versus  $[k_1\alpha]_I$  provides the best-fit linear trends of simulated data for scrutinizing the biodegradation of PE plastic mediated by the BFC colonies (see Fig. 7). The RR efficiency of PE degradation under an appropriate



**Fig. 7.** Numerical analysis of PE degradation mediated by the BFC colonies in the RR treatment system under the optimal control of environmental conditions with (a) the efficiency of PE degradation related to the internal diffusion of decaying PE molecules and (b) the efficiency of PE degradation related to the external transport of released PE molecules.

CEC set increased with increasing of the IMT and EMT factors can be expressed in the linear equation of  $E = a [k_1\alpha]_{D/E} + b$  with  $a$  as slope and  $b$  as the y-intercept. The unit of  $a$  expressed in % d and that of  $b$  expressed in % are defined to have the same definite magnitude of quantity with the unit of  $E$ , which is expressed in %. This linear function equation shows a strong correlation of both  $E$  to  $[k_1\alpha]_D$  and  $E$  to  $[k_1\alpha]_I$  with  $R^2 > 0.99$  (see Table 1) can be used to predict the performance of RR process in accordance with the value of the IMT and EMT factors. The performance of RR process naturally degraded the PE plastic can be verified at approximately 5.6% of the efficiency (see Fig. 7a). Many types of microorganisms naturally adapted with an environment can grow and act to degrade decaying molecules of PE and this influences the kinetic rates of IMT and EMT in the release of decaying PE molecules supported other elements and the positive interactions among microbes (Li et al., 2022; Vesilä et al., 2019). The presence of BFC colonies aimed to boost the performance of RR process can increase the kinetic rate of IMT to transport decaying PE molecules from an internal diffusion to the surface layer of PE plastic. Empirical evidence shows an increase in the value of  $[k_1\alpha]_D$  by 1.190 1/d from 0.124 1/d at 30th day to 1.314 1/d at 150th day of the experiment (see Fig. 7a). The acceleration of EMT to transport decaying PE molecules through a film zone closely near the surface of PE plastic to cell surface sorption of the BFC colonies resulted in an increased performance of RR process can be verified from

**Table 1**

Numerical values of the parameters:  $a$ ,  $b$  based on  $E = a [k_1\alpha]_D + b$  and  $E = a [k_1\alpha]_I + b$ .

Linear equation	Numerical value of parameter		$R^2$
	$a$ (% d)	$b$ (%)	
$E = a [k_1\alpha]_D + b$	41	5.6	0.99042
$E = a [k_1\alpha]_I + b$	12,974	-38.2	0.99852

the value of  $[k_d a]$  slightly increased from 0.006 1/d at 30th day to 0.008 1/d at 150th day of the experiment (see Fig. 7b). Effect of BFC development on the acceleration of GMT (GMT + EMT) can improve the performance of RR process by 52.7% from 8.8% at 30th day to 61.5% of the degradation efficiency at 150th day of the experiment. The performance of RR process to microbiologically degrade the PE plastic waste reaches 61.5% of its maximum efficiency for the release of decaying PE molecules mediated by the BFC colonies obtained at 1.314 1/d of the IMT and 0.008 1/d of the EMT rate. This could be due to the porosity of the mixed laterite-rice husk media has an important role to increase an availability of carbon source speeding the metabolism of BFC colonies in the RR tank and takes place independently from biotic and abiotic environmental factors (Chen et al., 2022).

The RR performance of 100% predicted using the linear equation of  $F = a [k_d a]_d + b$  can be expected at the IMT rate of 2.138 1/d and the EMT rate of 0.011 1/d under an appropriate CEC setting of the experiment. It is suggested that the performance of RR process can be improved by setting an appropriate CEC of conditioning the experiment with the selection and adaptation of the most reliable BFC colonies. The use of GP equations to describe the biodegradation rate of decaying PE molecules mass transfer allows us to get better understanding of the future plastic waste management policies (Lain et al., 2018). The calculation of the kinetic parameters using various kinetic models has been suggested to evaluate the decomposition reaction of PE material in the presence of catalyst (Nisar et al., 2022b). The influence of different CEC factors of setting an experiment for degrading the PE plastic waste contributes to the EMT and IMT degradation rates of PE in an environment (Chomas et al., 2020). The mechanisms of biologically degraded the plastic wastes of PE by the *Plasmodium* spp. strains and polystyrene by the *Tenebrio molitor* strains indicate a variation in the effect of biotic degradation on the gut microbial community of larvae (Novlekas et al., 2023). The physiological response of mealworm larvae to the biodegradation of PE plastic waste has been investigated providing an insight to get better understanding on the physiological response of an invertebrate after plastic ingestion (Peng et al., 2023). An effort to increase the performance of RR process suggested by improving the mobilization of decaying PE molecules transported from the surface of PE plastic to cell wall of BFC colonies could be due to the biodegradation of PE plastic controlled by an EMT rate with its  $[k_d a]$  value of 0.008 1/d is below than that supported by an IMT rate with its  $[k_d a]$  value of 1.314 1/d observed at 150th day of the experiment.

#### 4. Conclusions

The analysis of linear function used the curve of  $\ln(q)$  versus  $\ln(t)$  showing the best-fit kinetic parameters of  $\beta$  and  $B$  with  $R^2 > 0.93754$  could be able to predict the biodegradation kinetics of decaying PE molecules mass transfer. The biodegradation mechanisms of PE biotic mediated by the BFC colonies analyzed using the values of  $[k_d a]$ ,  $[k_d a]$ ,  $[k_d a]$ ,  $[k_d a]$  to predict the kinetic rates of IMT, EMT and GMT could be able to determine the resistance of mass transfer for decaying PE molecules depended on EMT. The combination of laterite with rice husk as the culture media to support the growth of microorganisms has an important role to improve the porosity of growth media and to increase the availability of carbon source for ensuring an effective metabolic acceleration of the BFC colonies. An effort to increase the performance of PE degradation at 100% of the efficiency can be suggested by adjusting the operation of RR at an IMT rate of 2.138 1/d and the EMT rate of 0.011 1/d. The numerical analysis of PE degradation mediated by the BFC colonies limited to the mechanisms and kinetics of mass transfer simulated using the MMTP models contributes to an enrichment of the references for the further researches of bacterial-fungal biodegradation of the plastic waste.

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#### Statement

During the preparation of this work the author(s) used Microsoft Excel in order to simulate the experimental data using the modified mass transfer factor models. After using this tool, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

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#### CRedit authorship contribution statement

Rinanti Astri: Project administration, Methodology, Formal analysis, Data curation, Conceptualization, Fulazzaky Mehamad Ali: Writing – review & editing, Visualization, Validation, Supervision, Methodology, Formal analysis, Conceptualization, Fadhul Metad Ferianita: Writing – original draft, Resources, Project administration, Methodology, Data curation, Conceptualization, Sunaryo Thalia: Project administration, Methodology, Funding acquisition, Formal analysis, Data curation, Tarkiaturrizki Tarkiaturrizki: Resources, Project administration, Funding acquisition, Formal analysis, Data curation, Muda Khalida: Supervision, Resources, Investigation, Formal analysis.

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#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

The data that has been used is confidential.

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