Biocatalysis and Agricultural Biotechnology 17 (2019) 25–31

Contents lists available at ScienceDirect

Biocatalysis and Agricultural Biotechnology

journal homepage: www.elsevier.com/locate/bab

Biodegradation of polyethylene terephthalate waste using *Streptomyces* species and kinetic modeling of the process

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ARTICLE INFO

Keywords:
Biodegradation
Polyethylene terephthalate
Streptomyces
Environmental pollution
GC-MS
Michaelis-Menten model

ABSTRACT

Polyethylene terephthalate (PET) is one of the most widely used plastics in manufacture of fibers, films, and drinking bottles, etc. It is one of solid wastes which pollutes urban and marine area and gets a lot of sacrifices from creatures. Thus, its removal from the environment is very important for protecting marine life. Different physical, chemical, and biological methods are studied by authors, but because of environmental and economic reasons, biological methods are preferred. These methods are slow and must combined with one or more physical or chemical methods. In this study, biodegradation of PET by *Streptomyces* species was assessed. Drinking bottles as PET wastes were firstly powdered and classified into four particle sizes. Then 50 mg of samples of each particle size were taken and treated with a fixed number of microorganisms in a culture medium for 18 days at 28 °C within an incubator, and degradation values of the samples were calculated on certain days. Also, a PET film was prepared from bottles and was exposed to biodegradation to show and compare differences between degradation of powdered and film samples. Results showed that final biodegradation percentages for PET particles sizes of 500, 420, 300 and 212 μm were 49.2%, 57.4%, 62.4%, and 68.8%, respectively. We showed that particle size and reaction time were the most important parameters on biodegradation. Also, by-products of biodegradation were analyzed by GC-MS to verify biodegradation process. Kinetic modeling of biodegradation showed that Michaelis-Menten activation or inhibition model can predict experimental results, more precisely.

1. Introduction

Heavy polymers with very high molecular weights can be found naturally or be prepared synthetically. While many natural polymers such as cellulose, starch and proteins are known and have widely been used by humans for centuries, but production and utilization of synthetic polymers is relatively new (Koltzenburg et al., 2017). Plastics are mostly man-made synthetic polymers obtained from materials such as coal, natural gas, limestone, oil, salt and water. These materials have very useful characteristics, e.g. they have lower density compared to other solid materials, their strength-to-weight ratio is significantly high, they demonstrate a good stability to mechanical shock, they are resistant to corrosion and chemical decomposition, and their formation is somewhat easy. But like other materials they have some shortcomings and weaknesses (Schwartz and Goodman, 1982). The most fundamental problem facing this industry and civilization as a whole which was revealed for the first time in 1970s, is concern about environment. Although, there is no doubt that the use of plastics helped improvement of life quality, but they have garbage and environmental pollution problems (Brydson, 1999).

Types of plastics currently found in urban waste include PET, high density polyethylene (HDPE), poly vinyl chloride (PVC), low density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), etc. (Tchobanoglous et al., 1993). PET is a well-known plastic and most widely used as soft linear thermoplastic polyester which was entered polymer market since 1977 and quickly found many applications in industry and domestic uses. It can be shaped at normal temperatures and pressures. This polymer is obtained from condensation polymerization reaction of dimethyl terephthalate or terephthalic acid with ethylene glycol as shown in Fig. 1 (Rieckmann and Völker, 2004):

Abbreviations: PET, Polyethylene Terephthalate; GC-MS, Gas Chromatography - Mass Spectrometry; SEM, Scanning Electron Microscopy; HDPE, High Density Polyethylene; PVC, Poly Vinyl Chloride; LDPE, Low Density Polyethylene; PP, Polypropylene; PS, Polystyrene; MSE, Mean Square of Errors

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https://doi.org/10.1016/j.bcab.2018.11.002
Received 2 October 2018; Received in revised form 4 November 2018; Accepted 5 November 2018
Available online 06 November 2018
1878-8181/

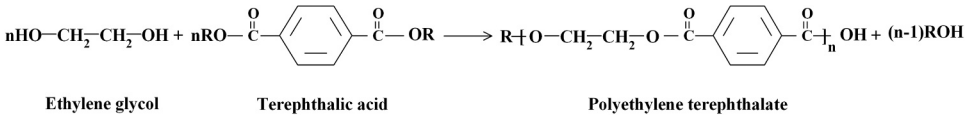

Fig. 1. Reaction of ethylene glycol with terephthalic acid for production of PET.

The main disadvantage of plastics is their environmental pollution which necessitates their removal from the environment. For this purpose, physical, chemical, and biological methods have been studied and proposed. Biological methods are preferred due to economic and environment reasons. But these methods are slower than physical and chemical methods and must be combined with some pretreatment methods. Various methods have been proposed for the removal of PET wastes from the environment that can be categorized as photo-oxidation, thermal degradation, chemical degradation, and biodegradation (Andrady, 2015).

Currently, industrial methods on removal of PET wastes and other polymers include landfill, incineration and recycling. But with regard to the environment protection, these methods have some disadvantages. Landfill is the simplest and oldest method, but it results many problems such as occupation of land, groundwater pollution, harmful material emission and waste of resources. Similarly, burning of harmful products can lead to serious pollution of the environment. According to the reuse of PET wastes, recycling is an effective and scientific method. However, recycling is limited in practice because of the high costs. In addition, suitable PET wastes for recycling are limited (Foolmaun and Ramjeeawon, 2012; Webb et al., 2013).

Unlike chemical decomposition, biological decomposition has the potential to prevent secondary infection and reduces costs. Many scientists have been engaged in the study of biodegradation of plastics (Harshvardhan and Jha, 2013; Shah et al., 2013; Sangeetha Devi et al., 2015; Farzi et al., 2017), While PET is a polyester, its ester group makes it more resistant to biodegradation compared to other polymers. A few studies on PET biodegradation by enzymatic and microbial methods have been observed which are summarized at below.

Tokiwa and Suzuki (1981) investigated biodegradation and hydrolysis of copolyesters (CPE) of aromatic and aliphatic polyesters such as polycaprolactone and PET by *Rhizopus delemar* lipase. Reaction mixtures containing enzyme, culture medium and powder or film of CPE were incubated on a shaker at 150 rpm at 37 °C for 16 h. But they didn't indicated particle size of CPE powder or dimensions of film they used in culture medium. They investigated the effects of transesterification reaction time and molar ratio of aliphatic to aromatic polyesters on biodegradability of copolyestes and showed that biodegradation increased with increasing reaction time and/or aliphatic mole ratio. Also, they showed that when PET was used as aromatic polyester in copolyester structure, biodegradability was decreased significantly.

Zhang et al. (2004) studied biodegradability of PET fiber and diethylene glycol terephthalate (DTP) by microbes prepared from activated sludge, and lipase enzyme. Their results showed that although degradation ratio of DTP was high, but degradation of PET fiber was very week.

Marqués-Calvo et al. (2006) investigated microbial and enzymatic biodegradation of PET homopolyester and PET copolymers films with 225–275 mm thickness containing nitrated units. Roughness enhancement, swelling and decrease of average molecular weight expressing biodegradation of copolymers were significant only for the case of PET containing 30 mol% of nitroterephthalic units cultured with *Aspergillus Niger*.

Nowak et al. (2011) assessed biodegradation degree of PET films modified with a polyester by filamentous fungi *Penicillium funiculosum* and compared with that of PET. After 84 days of incubation it was found that addition of Bionolle polyester didn't accelerate significantly biodegradation of modified PET films.

Sharon and Sharon (2012) studied biodegradation of PET films using naturally growing microbes on degrading PET such as different microbes from *Actinomycetes* family and some fungi. They cut PET films into 1 cm^2 pieces and inoculated onto the test medium. The results confirmed slow degradation of PET by *Nocardia* with help of esterase enzyme.

Nakkabi et al. (2015) investigated biodegradation of PET films by *Bacillus Subtilis*. They incubated test medium for 30 days at 37 °C. SEM and infrared analysis before and after test showed week biodegradation ratio, but also they showed that the bacteria acted on PET films.

Yoshida et al. (2016) used *Ideonella sakaiensis* bacteria isolated from outside of a bottle-recycling facility for hydrolysis of PET. They incubated PET film at 30 °C for 18 h and found that this strain degrades PET polyester into its monomers with the aid of two enzymes MHETase and PETase. They performed different analyses such as HPLC and SEM to assess performance of bacteria for biodegradation.

In this study, biodegradation of PET polymer obtained from bottles by *Streptomyces* species from *Actinomycetes* family isolated from the soil of East Azerbaijan, Iran, was investigated. For the purpose of determining biodegradation performance, different analyses such as calculation of degradation ratio, GC-MS and SEM were conducted.

2. Material and methods

Drinking PET bottles were used as the raw material for biodegradation. Firstly, they were cut into small pieces and liquid nitrogen spilled onto the crushed material and then they ground with a milling machine having a maximum mesh size of 500 μm (Model: ZM 200, Retsch, Germany). To achieve three other sizes, milled material was passed through sieves with mesh numbers of 40, 50 and 70 corresponding to particle sizes of 420, 300 and 212 μm, respectively.

Mineral medium components are presented in Table 1 (Del'Arco and de França, 1999).

The study was conducted in five main stages for biodegradation of PET powdered samples as follows:

- Preparation of pure bacterial strains,
- Treatment of diluted PET powder sample in culture medium with *Streptomyces* species,
- Extraction of residual PET and determination of degradation percentage (Wang et al., 2007),
- Analysis of metabolite by GC-MS equipment in order to identify products of biodegradation,
- SEM analysis on PET film exposed to biodegradation by *Streptomyces* bacteria.

Streptomyces species were prepared from microorganisms' collection of University of Tehran, Iran, isolated from the soil of East Azerbaijan,

Table 1
Mineral medium constituents (Del'Arco and de França, 1999).

Component	Composition (g/L)
NH_4Cl	0.3
NaCl	0.15
KH_2PO_4	0.9
Na_2HPO_4	1.8
$CaCl_2.2H_2O$	0.045
$MgSO_4{\cdot}H_2O$	0.15

Iran. 20 mL of mineral medium was poured into the twisted falcon tubes and the tubes were sterilized by autoclave at 121 °C and 15 psig for 20 min. Four replicate falcon tubes were prepared for each particle size, because sampling must be performed on third, sixth, 12th and 18th day after the start of experiments. 50 mg of powdered and sterilized PET sample were taken and poured into each falcon tube. Also, 0.5 mL of microbial suspension with 0.5 McFarland standard were added to each tube and contents were incubated within a shaker incubator (Model: Pars Azma, Iran) at 28 °C at 120 rpm for 18 days. PET powders were the sole carbon source for bacteria.

Samples including whole falcon tube contents were taken on third, sixth, 12th and 18th days after the start of each experiment in order to determine degradation percentage of PET. For establishing aerobic conditions, doors of falcon tubes were left half-round open (Southam et al., 2001). For extraction of residual PET of each sample, several solvents such as ether, cyclohexane, ethanol, chloroform, dichloromethane, toluene, and xylene, were tested and it was observed that toluene was the best solvent with lowest consumption and highest solubility. 5 mL of toluene was used for PET extraction in a decanter and then the solution was centrifuged (Model: Hettich EBA20, Germany) at 4000 rpm for 5–15 min. Then the organic phase was filtered by Wattman filter paper. Filter paper was weighed exactly by an electric scale (Model: Sartorius, Germany) before and after filtering of the organic phase and drying toluene residue in incubator at 50 °C, to obtain the weight of residual PET. Finally, extent of biodegradation, X, was calculated using Eq. (1).

$$X(t) = \frac{m_0 - m(t)}{m_0} \tag{1}$$

where m_0 and $m(t)$ are initial mass and mass at time t of PET powder sample, respectively.

For assessing metabolites produced by *Streptomyces* sp., filtered liquid sample from previous step which obtained at 18th day of incubation was analyzed by GC-MS. It was injected into a GC-MS apparatus equipped with a gas chromatographic analyzer (Model: Agilent 6890, USA) and a mass spectrometer (Model: Agilent 5973, USA). Extracted components were separated on HP-5 ms capillary column (30 m × 0.25 mm ID × 0.25 μm film, J&W Scientific, Folsom, CA) based on temperature programming while initial temperature of 40 °C was kept constant for 5 min and then it was raised up to 250 °C with constant rate of 5 °C/min. Initial injection was done at 280 °C through an oven (Agilent Technologies, 2012).

Also, a 2 cm × 2 cm film was prepared from PET bottles and exposed to biodegradation with *Streptomyces* bacteria. After 18 days of incubation, the film was analyzed by a scanning electron microscope and its biodegradation was compared with that of powdered samples (Model: VEGA II LMH, TESCAN, Czech Republic).

2.1. Reaction kinetics

Kinetic modeling of biodegradation of powdered PET was also performed to find and propose the best model for prediction of experimental data. As the process was performed in batch mode, dynamic mass balance for the consumption of PET can be written as below:

$$\frac{dC_A}{dt} = -r_A \tag{2}$$

where C_A (g/L) is concentration of PET in sampled metabolite solution at time t (days), and $-r_A$ (g/L.days) is the rate of PET consumption by microorganisms. Three models including power law model, Michaelis-Menten inhibition model, and Michaelis-Menten activation model were tested for predicting PET biodegradation reaction.

2.1.1. Power law model

The general model of power law was tested for this process whose rate equation is as follows:

$$-r_A = kC_A^n \tag{7}$$

where exponent n can be obtained by nonlinear regression. Applying the above equation into Eq. (2) and integrating results:

$$C_A = [-(1-n)kt + C_{A0}^{1-n}]^{\frac{1}{1-n}} \tag{8}$$

2.1.2. Michaelis-Menten inhibition model

There are different models of Michaelis-Menten that stand on the mechanism of absorption of reactant(s) on microorganisms or enzymes surface, reaction on them, and desorption from their surface. Because, the models obtained by this technique have theoretical basis, they usually predict biological reactions more precisely than above-mentioned models. Different variants of Michaelis-Menten model include simple model, inhibition model, activation model, and combined inhibition and activation model. As explained by Nauman (2002) inhibition model is used when increasing of reactant(s) concentration decreases reaction rate, while in activation model it increases reaction rate. In this study, these two models were examined. Michaelis-Menten inhibition model can be written as below:

$$-r_A = \frac{kC_A}{1 + k_1C_A + k_2C_A^2} \tag{9}$$

where k, k_1, and k_2 are constants. The details of derivation of the model can be found in Nauman (2002). The result of integration of Eq. (2) when using the above reaction rate is as follows:

$$\log(1-X) + (k_1C_{A0} + k_2C_{A0}^2)X - \frac{k_2}{2}C_{A0}^2 = kt \tag{10}$$

Constants of the above equation can be found by linear regression.

2.1.3. Michaelis-Menten activation model

The following equation represents the model:

$$-r_A = \frac{kC_A^2}{1 + k_1C_A + k_2C_A^2} \tag{11}$$

Integration of Eq. (2) when using this model results:

$$\frac{1}{C_{A0}(1-X)} - k_1\log(1-X) + k_2C_{A0}X = kt - 1 \tag{12}$$

Constants of this model can also be calculated by linear regression.

3. Results and discussion

3.1. Results of biodegradation of powdered PET samples

Fig. 2 shows the results of biodegradation of powdered PET samples by *Streptomyces* species for different particle sizes calculated at different days of incubation using weighing method as explained above, and Eq. (1).

As shown above, till third day there was a small degradation for all particle sizes. After that and till sixth day of incubation, particle size of 212 μm had the most degradation ratio while particle sizes of 300, 420, and 500 μm had almost the same level of degradation. Degradation was continued approximately with the same rate till 12th day where particle sizes of 500 and 420 μm had lower degradation than size 300 μm. At last day of incubation, degradation rates of all particle sizes approximately became very small which can be because of saturation of the solution by byproducts, reduction of carbon source supplied by PET particles, and shortage of other minerals required for microorganisms.

Nowak et al. (2011) obtained 0.08% of biodegradation of PET film samples with *Penicillium funiculosum* fungi which is much lower than the results of this work and can be due to powdering of PET samples before biodegradation in this work.

Also, by comparing the above results with the results of biodegradation of HDPE with *Streptomyces* from the work of Farzi et al.

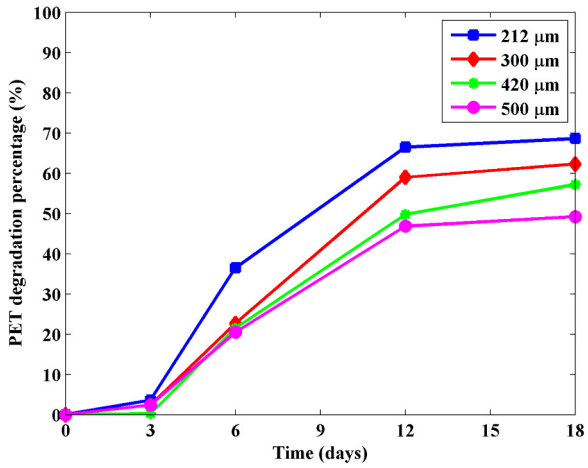

Fig. 2. Degradation percentages of powdered PET samples for different particle sizes with respect to contact time with *Streptomyces* species.

(2017) it can be seen that biodegradation of PET is much higher than HDPE at the same conditions which is because of higher molecular weights of HDPE plastics and their more compact structure compared to PET plastics.

Literature review showed that none of authors used powdered PET samples for biodegradation except Tokiwa and Suzuki (1981), but they have not indicated powder sizes and also they worked on copolyesters and not PET homopolyester.

3.2. Analysis of metabolites by GC-MS

As mentioned before, a sample with highest degradation value at 18th day of incubation of powdered samples was extracted by toluene and then filtered for analysis by GC-MS apparatus. Fig. 3 shows retention times and abundances of components existing in metabolite detected by GC equipment. As it can be seen, many peaks have been detected and by calculating area percent of each peak, composition of components can be obtained.

To identify each component, outlets from GC column were sent into a mass spectrometer (MS) attached to it. Fig. 4 shows an example of MS analysis at retention time of 2.328 min detected as o-Xylene. Table 2 shows all constituents of the metabolite sample detected by GC-MS. As it is evident, maximum carbon number is 15 and thus *Streptomyces* bacteria were able to degrade PET powder to very smaller molecules. Also, toxicity of the components reveals that none of degradation products are so harmful for human life, albeit some of them such as ethyl benzene or o-xylene must be separated before releasing metabolite to the environment.

As can be seen from the above table, the main constituents of metabolite are ethyl benzene and hexamethyl cyclotrisiloxane. The first one is produced by breaking molecular chain of PET containing aromatic groups. The second one contains silicon element as one of its constituents, because PET polymer is filled by class-E borosilicate glass-fibers before injection molding to increase its glass-transition temperature (Scheirs, 2003; Alqaflah et al., 2017). Other elements such as S and N were detected in other degradation products which may be due to digestion of available ingredients in mineral medium. Also, all detected components have aromatic groups within their structure which is due to the existence of terephthalate group in PET structure.

3.3. Results of biodegradation of PET film

As mentioned in previous section, biodegradation of the surface of a PET film by *Streptomyces* species was also performed for 18 days. At the

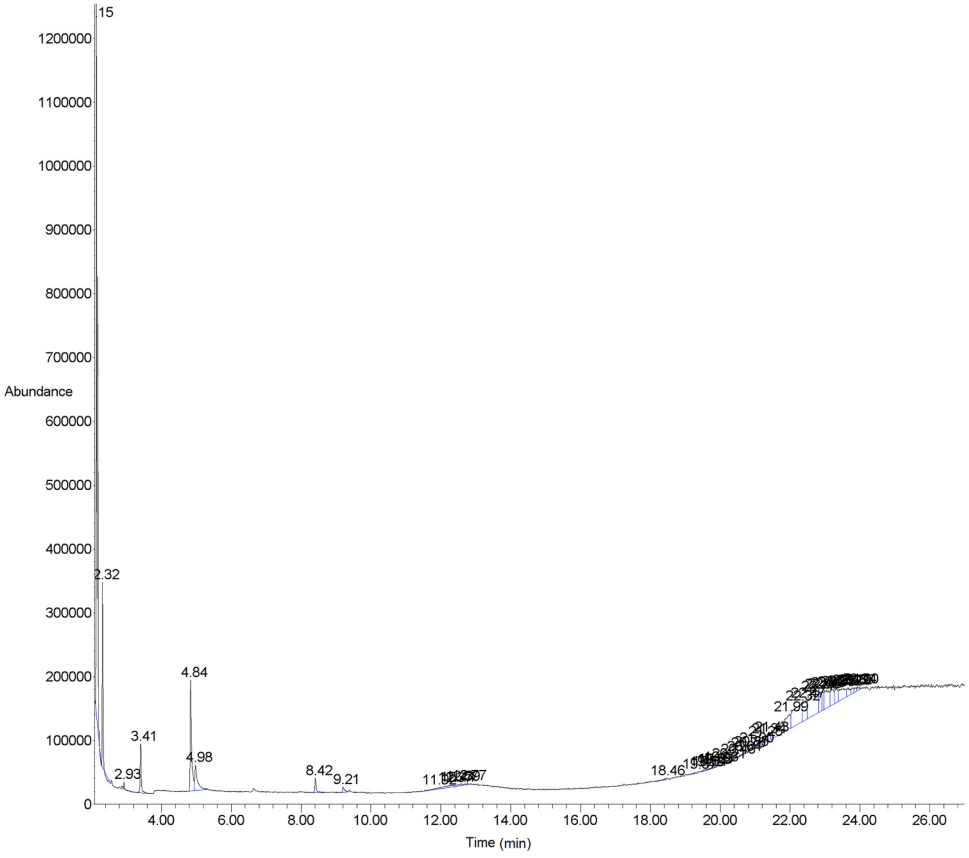

Fig. 3. Results of analysis of metabolite sample by GC equipment.

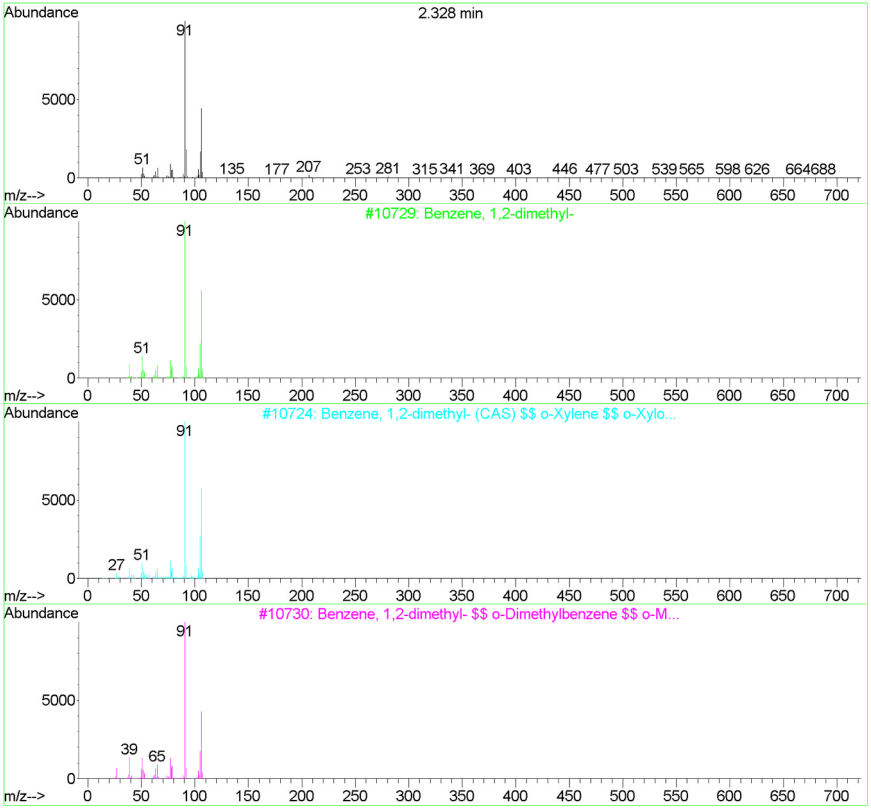

Fig. 4. Mass spectrometry analysis of a component at retention time of 2.328 min detected as o-Xylene.

Table 2
Components existing in metabolite sample and their compositions detected by GC-MS.

Component	Chemical formula	Composition (wt%)	Toxicity	Reference
Ethyl benzene	C_8H_{10}	33.13	LD50 > 3.5 g/kg rats	ScienceLab Chemicals & Laboratory Equipment (2005a)
o-Xylene	C_8H_{10}	5.18	May cause skin and eye irritation	ScienceLab Chemicals & Laboratory Equipment (2005b)
1,2- β Pinene	$C_{10}H_{16}$	0.16	LD50 > 4.7 g/kg rats	Cayman Chemical (2017)
1,8- Cineole	$C_{10}H_{18}O$	2.49	LD50 > 2.48 g/kg rats	ScienceLab Chemicals and Laboratory Equipment (2012)
I-Menthone	$C_{10}H_{18}O$	6.85	May cause skin and eye irritation	Sigma Aldrich (2014)
p-Menthan − 3-one	$C_{10}H_{18}O$	2.98	LD50 > 0.5 g/kg rats	Lewis (1996)
2-(4′-Nitro − 2′-Thienyl)-Pyrimidine	$C_8H_5N_3O_2S$	0.87	Not available	
Germacrene D	$C_{15}H_{24}$	0.49	Non-toxic (allergic)	Essence (2018)
Hexamethyl Cyclotrisiloxane	$C_6H_{18}Si_3O_3$	45.02	LD50 > 50 g/kg rats	Moretto et al. (2005)
6-Methyl − 2-Phenylindole	$C_{15}H_{13}N$	0.95	May cause skin and eye irritation	JandK Scientific LTD (2010)
N-Methyl − 1-Adamantaneacetamide	$C_{13}H_{21}NO$	0.09	Not available	
Cyclobarbital	$C_{12}H_{16}N_2O_3$	0.31	Non-toxic (drug)	Breimer and Winten (1976)

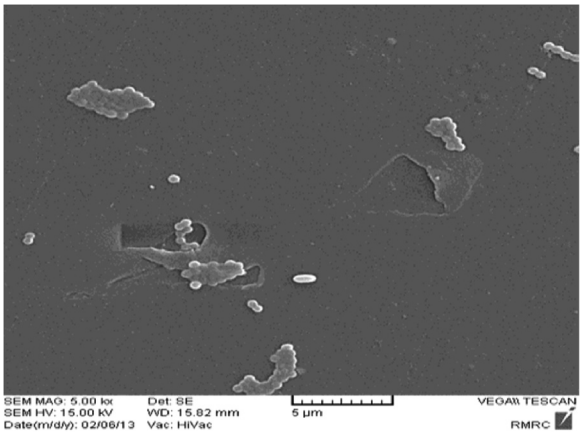

Fig. 5. SEM photo from the surface of PET film sample after biodegradation by *Streptomyces* species.

end of experiment, an SEM photo was taken which is shown in Fig. 5.

As can be seen, microorganisms degraded the surface of the film, but it is not significant. Comparison of biodegradation of powdered samples with film sample, proves that powdering as a physical pretreatment method is highly effective, and high degradation efficiencies could be obtained.

3.4. Kinetic modeling of PET biodegradation by Streptomyces species

Results of kinetic modeling of the process based on experimental data using different models presented before are shown in Table 3. Two parameters, namely coefficient of determination (R^2) and mean square of errors (MSE) were used for validation of the results of models whose values are also shown in Table 3. These data represent that R^2 values for Michaelis-Menten inhibition and activation models are very close to 1.0 with very low MSE values for all particle sizes.

Fig. 6 illustrates results of kinetic modeling of biodegradation of powdered samples with different sizes using different kinetic models. As

Table 3
Estimated values of reaction rate constants for different proposed kinetic models for prediction of biodegradation of powdered PET samples with different particle sizes.

Particle size (μm)	Kinetic model	R^2	MSE × 10^3	$k \times 10^3$	n or k_1	k_2
212	Power law model	0.9239	39.2512	79.8065	0.7497	–
	Inhibition model	0.9998	0.0840	2.4370	− 1.3725	0.4404
	Activation model	0.9999	0.0638	2.1815	− 1.4350	0.4813
300	Power law model	0.9256	31.8616	76.0253	0.4244	–
	Inhibition model	0.9983	0.7208	2.0521	− 1.2273	0.3632
	Activation model	0.9931	2.9496	2.0320	− 1.2703	0.3926
420	Power law model	0.9349	22.4729	70.2717	0.3109	–
	Inhibition model	0.9828	5.9251	3.7931	− 1.1252	0.3250
	Activation model	0.9901	3.4149	3.5170	− 1.1423	0.3438
500	Power law model	0.9290	18.7046	46.7856	0.7925	–
	Inhibition model	1.0000	0.0062	0.5994	− 1.1004	0.2964
	Activation model	0.9996	0.1031	0.4299	− 1.1175	0.3061

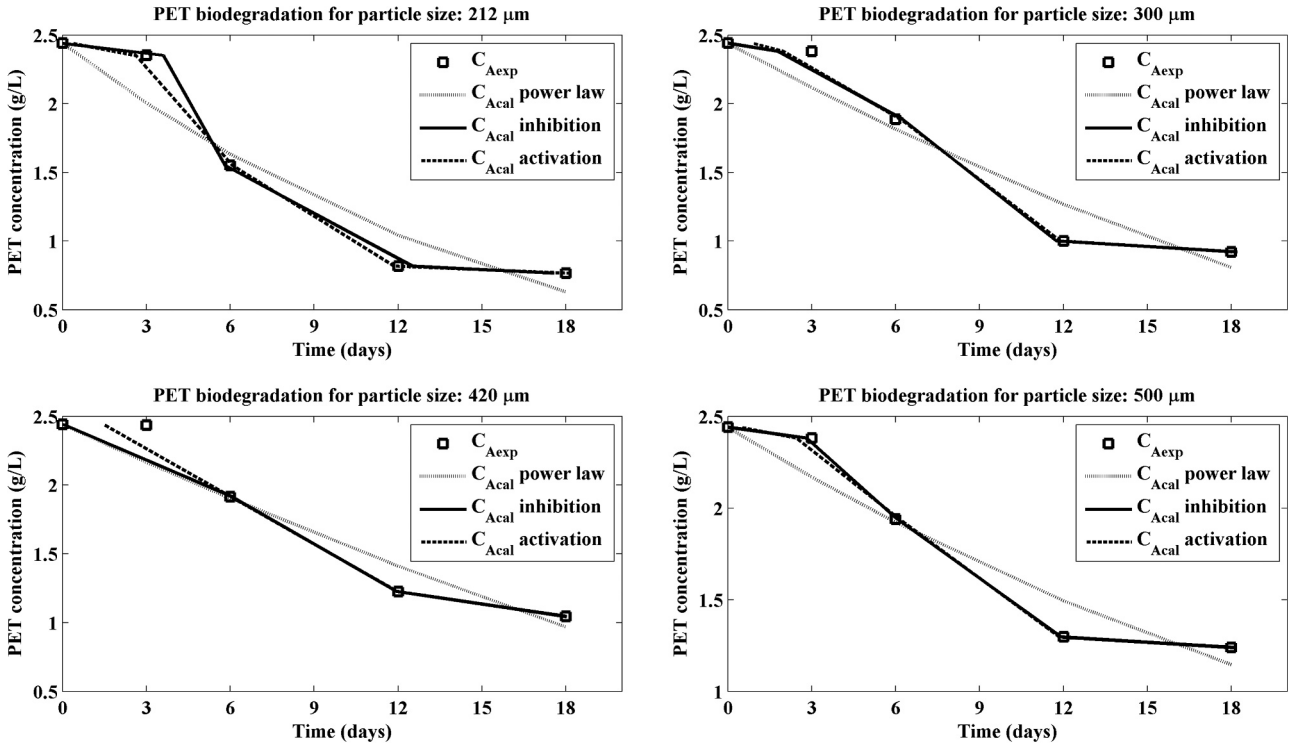

Fig. 6. Results of kinetic modeling of powdered PET biodegradation, □ experimental data, … power law model, — Michaelis-Menten inhibition model, - - - Michaelis-Menten activation model.

can be seen, power law model has medium accuracy, but it cannot predict Z shape of experimental data because of substrate activation or inhibition behavior. Both Michaelis-Menten inhibition and activation models have precise prediction of experimental data and can be used as the kinetic model of the process. Although these models are usually used for enzymatic processes (Nauman, 2002), but it was shown here that they can be used for biological processes containing microorganisms, because microorganisms also perform substrate digestion by the aid of their enzymes.

4. Conclusions

In this study, biodegradation of PET waste by *Streptomyces* species was investigated in laboratory scale. As discussed in introduction section, different methods of waste removal such as landfill, incineration, and recycling which are currently used for PET waste, have some disadvantages such as environmental pollution and high cost of operation. Thus, PET wastes must be completely degraded to smaller molecules and then generated by-products can be utilized for some applications or can be removed more safely. Among different degradation methods, biological methods are preferred because of their low cost of operation, being environmental friendly, and operation at moderate conditions. But, because they are so slow compared to physicochemical methods, they must be combined with one or more of physical or chemical ones to increase degradation rate. Different authors studied biodegradation of PET polymer films and they also showed that PET biological removal is very slow process. For this reason, in this work we used grinding as a physical method together with biological degradation to investigate and show effectiveness of combined methods on removal of PET waste.

PET samples were cut, grinded and classified into four different particle sizes. In each experiment, 50 mg of PET powder were incubated in a culture medium containing 20 mL of a mineral medium and 0.5 mL suspension of *Streptomyces* species at 28 °C at 120 rpm for 18 days. The highest biodegradation value was obtained as 68.8% for particle size of 212 μm on 18th day. Also GC-MS analysis of the sample having highest degradation ratio, showed that bacteria degraded PET powders into less harmful components with low carbon numbers. Finally, kinetic modeling of biodegradation was performed and three kinetic models were

examined.

It was shown that combination of a biological method with a physical or chemical pretreatment method such as grinding and powdering increased degradation efficiency. Also, it was found that finer particles can be degraded more rapidly. SEM analysis of PET film showed very low biodegradation of the surface by microorganisms which is because of low surface area available for bacterial degradation. Michaelis-Menten inhibition and activation models are proposed as best kinetic models for predicting biodegradation of powdered PET samples.

Declarations

Authors of the paper declare that no ethical issues are applicable to the results of this paper and accept publication of the results. All data for the figures of this papers are available and can be presented upon request. None of the authors have competing interest on the results of this paper and no funding support has been received for this work.

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