

INVESTIGATION ON THE BEHAVIOUR OF PLASTICS DURING THE BIOLOGICAL TREATMENT OF ORGANIC WASTE

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Abstract: Microplastics (MP) (plastics <5 mm) are increasingly seen as a burden. Most MP entering the aquatic systems are often disposed on land, but little is known about their possible accumulation in terrestrial ecosystems. Organic fertilizers produced from organic waste might be a potential entry of MP into the soil. This research aim is to understand the behaviour of PE, PBAT, and PLA during the degradation of organic waste to assess their disintegration and to formulate recommendations for fermentation and composting practices. The mechanical, thermal and biological properties of the plastics will be determined in static and dynamic systems with different temperature ranges as well as retention times. For that, laboratory scale reactors for fermentation and composting processes were installed. A composting preliminary experiment was carried out during seven weeks to test the performance of the tumbler and to investigate possible physical changes on low-density polyethylene (LDPE) and high-density polyethylene (HDPE). After seven weeks, microscopic changes were observed on the surface of the HDPE films as scratches. The results from the preliminary experiment provided an insight on the performance of the compost tumbler and the quality of the compost. Preliminary fermentation tests will be carried out in the future and therefore results are not included here.

Key words: Microplastic, composting, fermentation, organic fertilizer, organic waste.

1. Introduction

Plastics play a vital role in the present human society. Its applications extend to the industrial, commercial, medicinal, and municipal sector. Globally approximately 6300 million tons of plastic were generated as of 2015. Of this amount, only 9 % had been recycled, 12 % was incinerated and 79 % was either accumulated in landfills or in the environment [1]. Plastic litter poses a considerable threat to the environment by choking wildlife, distributing potentially harmful organisms, and absorbing toxic chemicals [2]. Plastics can undergo aging processes resulting from physical, biological, and chemical actions and therefore fragmentize, disintegrate, and degrade into microplastics (MP) that may subsequently be ingested [2; 3]. MP are plastics with a size smaller than 5 mm, which are increasingly seen as a burden [4]. Although most MP entering the aquatic systems are produced, used, and often disposed on land, little is known about the sources, pathways, and possible accumulation in terrestrial ecosystems [3; 5].

Polyethylene (PE) is one of the most widely used plastics, but once it is wrongly discarded, it persists in the environment without degrading, which leads to its accumulation in the environment [6]. PE is a linear hydrocarbon polymer that consists of long chains of the ethylene monomer (C₂H₄) and therefore their resistance to degrade under environmental conditions [7].

To overcome the accumulation of polymers either on soil or in water, the production of bioplastics as a substitute to conventional plastics has gained attention. Bioplastics can be either bio-based, which means

synthesized from biomass and renewable resources or they can produced from fossil fuels [8]. They represent about one per cent of the around 335 million tonnes of plastic annually produced [9]. Although some bioplastics offer new ways of recovery and recycling (organically), their suitability is limited to composting plants conditions and not to those in nature [9]. From Fig. 1, it can be noticed that the most used biodegradable plastics are PLA with 10.3 % and PBAT with 7.2 %.

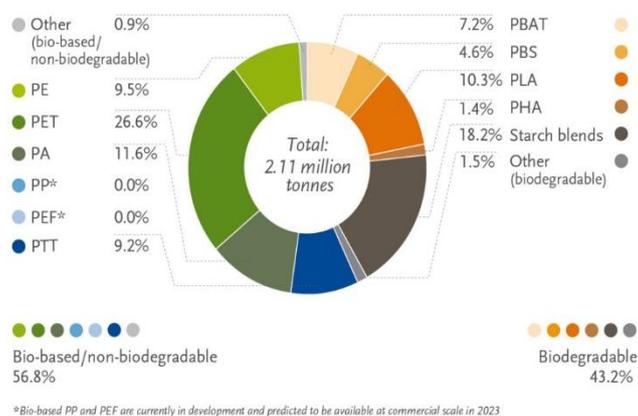


Fig. 1 Global production capacities of bioplastics 2018 (by material type) [10]

Poly(lactic acid) (PLA) is a compostable polymer derived from renewable sources (mainly starch and sugar) [11]. Due to its low toxicity [12], environmentally benign characteristics, and compostability, PLA is slowly becoming an ideal substitute for food packaging [13]. PBAT is 100 % biodegradable and its properties are comparable to those of low-density polyethylene [14]. The biodegradation of PBAT and PLA has been mainly tested in composting under different laboratory conditions [15–19], but to the best of our knowledge not in fermentation. The risk of not meeting the required degradation conditions for PBAT and PLA in the composting or fermentation plants could lead to the accumulation of MP in organic fertilizers.

The presence of MP in organic fertilizers coming from organic waste treatment plants have been previously proved [5]. Organic fertilizers can be produced from organic waste either anaerobically digested or composted. Municipal organic waste is often contaminated with plastics and although sieving can significantly reduce its amount, plastic particles remain in the waste. Therefore, organic fertilizers produced from organic waste might be a potential entry of MP into the terrestrial ecosystem. Knowing the form and amount in which plastics enter the organic waste stream will help to determine the contribution that organic fertilizers have on the introduction of MP into the soil.

Therefore, this research aims to understand the aging process of marked plastics during the degradation of organic waste. For that, we installed laboratory scale reactors for fermentation and composting treatment technologies. The influence of the temperature, mechanical stress, and residence time on the fragmentation of plastics to MP is going to be investigated. The assessment of the disintegration of plastics will result in the formulation of recommendations for fermentation and composting practices.

2. Methodology

2.1. Substrate determination

A synthetic organic waste recipe to guarantee a substrate free of plastics was established. In order to simulate the organic waste treated at the treatment plants, samples in two composting and two fermentation facilities were taken during autumn. Most of the treatment plants pre-treat the waste by mechanically removing impurities, mainly metals, glasses, and plastics. The aim of this step is to reduce pollutants from the input material, to optimize its physical properties, and to remove materials that may interfere with the subsequent process steps [20].

Since this step does not guarantee an input material free of impurities, plastics and other inorganics can still be found during the biological process. A way to know the plastic content entering to the biological step is by sampling after the mechanical sorting of impurities in the waste and before the biological treatment, when possible. The information regarding the facilities sampled in means of plant capacity, sample point, and weight of sample taken is depicted in Table 1.

Table 1 Treatment plants sampled and weight of sample taken and treated

Plant Number	Treatment technology	Plant capacity tones organic waste/a	Sample point	Total sample weight (kg)	Treated sample weight (kg)
1	Composting	60000	After sieving < 80 mm	17	3
2	Composting	29000	Receiving bunker	14	4
3	Fermentation with post composting	36000	After sieving < 60 mm	23	3
4	Fermentation with post composting	36000	After sieving < 60 mm	24	3

The German Federal Compost Quality Association establishes the methods to analyse compost samples. For the determination of plastic content, the method establishes to first dry the sample in an oven at 105 °C to later dry sieve the sample and manually sort out the impurities [21]. In order to avoid the breakage of the plastics due to the heat exposure, the plastics were sorted out from the fresh organic waste sample. Afterwards, the plastics found in the sample were wet weighted and placed on graph paper for a total surface area determination.

The sample free of plastics was later on chopped to a size of 40 mm and three subsamples of 150 grams each were taken for water content and volatile solids analysis. The rest of the sample was used to determine the mineral content in means of gravel, sand, and clay. Due to a lack of standardization for the determination of minerals on an organic waste sample at the sampled point, a wet sieving procedure with modifications was followed [22]. The minerals stratification was based on [23–25], where soil particles larger than 2 mm are classified as gravel, from 0,063 to 2 mm as sand, and smaller than 0,063 as silt or clay. Therefore, an analytical sieve shaker type AS300 control from the Retsch Company was used. The plates have a diameter of 315 mm and the sizes from the mesh fabric were 0,063 mm and 2 mm.

Gravel was separated from the mesh with the size 2 mm, whereas sand was found on the 0,063 mm mesh. Finally, clay was assumed to be those minerals that fell under the mesh with a size of 0,063 mm. The followed

method for the minerals determination separates the clay from the water with the use of a continuous centrifuge. However, due to the lack of equipment, it was assumed that clay is the difference from the total minerals content, the gravel and sand content.

2.2. Experimental set-up

Three laboratory-scale treatment plants were set-up as shown in Fig. 2. The design of the plants was based on the operating conditions of the composting and fermentation plants of the region. One plant consists of an acclimatized chamber where nine glass fermenters operate under mesophilic conditions. In a second plant, nine glass fermenters are surrounded by a water bath to provide thermophilic environment. With a propeller, these two plants can also operate dynamically and the effect that the mechanical stress as well as the working temperature have on the fragmentation of plastics can be evaluated.

The aerobic plant consists of four stainless steel drums installed in an acclimatized chamber set at 25 °C to avoid disruptions during winter. Windows on the sidewalls and lid of the drum were built to provide aeration of the substrate. The turning is done with the help of a handle placed on the side of the drum (Fig. 2).



Fig. 2 a: fermentation reactors in water bath; b: fermentation reactors in acclimatized chamber; c: aerobic tumbler

Three main variables that could influence the fragmentation or degradation of the plastics under study are temperature range, mechanical stress, and retention time. The most significant environmental factors that should be considered in the biodegradation of organic wastes as well as polymers are pH, temperature, moisture, and oxygen content [18; 26]. Other factors such as C:N ratio, CH₄, and CO₂ formation will be also measured. The arrangement of the experiments is shown in Fig. 3.

For the experimental stage a mixture of PE, PLA, PBAT, and PLA/PBAT films in equally distributed amounts in a size of one cm² will be given to the substrate. These plastics are provided by the Institute of Plastics Technology at the University of Stuttgart to assure that the plastics are additives free and that its properties are known.

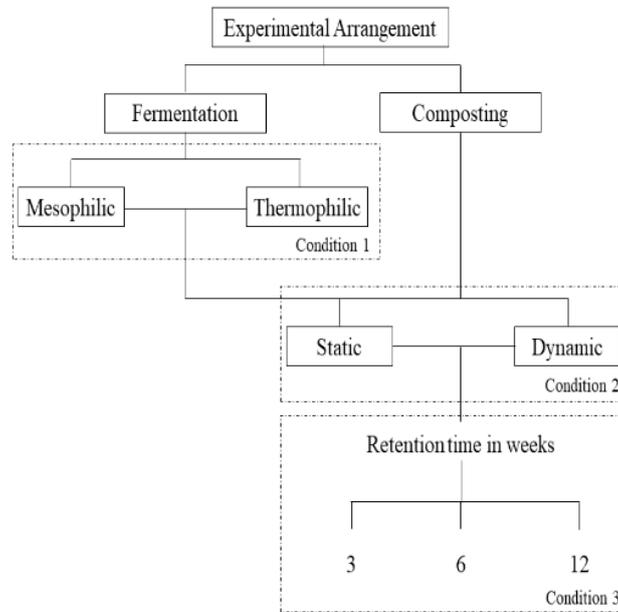


Fig. 3 Scheme of the experimental arrangement

Preliminary experiments were carried out in aerobic treatment during seven weeks. The aim was to test the performance of the compost drum and to investigate possible physical changes on low density and high density polyethylene (LDPE, HDPE) films.

The substrate consisted of organic waste obtained from a local co-fermentation post composting treatment plant. Plastics were manually sorted out from the substrate to avoid interferences with plastics that are not under study. Later on, LDPE and HDPE cut in one cm² size were added to the substrate. The drum was turned once per day to provide material mixing and aeration. The temperature of the pile was recorded daily in three different points of the pile with exception of the weekends. The carbon to nitrogen ratio was measured at the beginning and at the end of the experiment using an elemental analyser. The pH, water content and volatile solids of the sample were analysed once a week following the procedure of the Method Book for the Analysis of Organic Fertilizers, Soil Enhancers and Substrates [21]. The characteristics of the first aerobic preliminary test are shown in Table 2.

Table 2 characteristics of the substrate

Mass [kg]	51 + 26*
Volume [L]	163
Mass of plastics [g]	0,5
WC [%]	66
VS [%]	81
pH [-]	5,7
C/N	29:1

*Mass added on day 14th

After the biological degradation of organic waste, the plastics were taken out from the compost and were washed out with distillate water until all loosely attached impurities were removed. Afterwards, the plastics were left to dry at room temperature. In order to evaluate whether the given boundary conditions have any effects on the physical properties of the plastics, micro and macroscopic aspects of the PE films was carried out.

A first indication of microbial attack on the surface of plastics can be observed on the change in shape, malleability by roughening of surface, formation of holes or any other physical deterioration, which are known as micro and macroscopic aspects [27; 28]. These aspects were investigated before and after the biological treatment, the macro through visual observation and the micro with the use of a Zeiss Upright Axioplan 2 Imaging Microscope, with an objective magnitude of 10x. A microscope camera TUCSON was connected on the top of the microscope to transfer the image to the software of the same company.

3. Results and discussion

3.1. Substrate determination

The average values for the organic, gravel, sand, and clay were 77 %, 0.4 %, 4 %, and 19 %, respectively. The high clay content is attributed to the assumption made where clay is the difference between the total minerals content, the gravel and sand content. The results obtained are comparable to those in [22], where a sample was taken in autumn from the suspensory before the fermentation treatment. The reported values for organic, gravel, sand, and clay content of the sample were 72 %, 2 %, 10 %, and 16 %, respectively [22]. The results referring to the minerals and plastics content determined in the samples from the four treatment plants with their standard deviation are shown in Table 3.

Table 3 Organic, mineral, and plastic content of the samples analysed

Plant Number	Organic Content (%)	Gravel Content (%)	Sand Content (%)	Clay Content (%)	Plastics Content (%)
1	78 ± <0.1	0.1 ± <0.1	4 ± <0.1	18 ± <0.1	0.5
2	82 ± <0.1	0.3 ± 0	4 ± <0.01	14 ± <0.01	0.1
3	76 ± <0.01	1 ± <0.1	3 ± <0.01	20 ± <0.1	0.3
4	70 ± <0.01	0.3 ± <0.1	6 ± 0.02	24 ± 0.02	0.3
Average	77	0.4	4	19	0.3

A broader comparison of results cannot be made since to the knowledge of the author a established method for the determination of mineral content at this specific sampling point does not yet exist. Other studies however, applied other methodologies for the determination of impurities within different size ranges. In [29] impurities were separated from a pre-treated organic waste sample using two methods, combustion with a muffle furnace and elutriation with drying and manual sorting. The same authors tested later on, impurities separation with the use of a hydrocyclone of a pre-treated organic waste sample [30].

3.2. Experimental set-up

Since the experiment was started during winter and the composter was placed in a hall with low external heating, the initial temperature of the substrate was 7 °C. The thermophilic phase lasted five days with a maximum temperature of 53 °C and by the 9th day, the temperature registered was 25 °C. It was assumed that the mass of substrate was too low for the volume of the tumblr and therefore on the 15th day more substrate was added (red line on Fig. 4). This helped to keep the thermophilic phase for other six days followed by a decrease to a mesophilic stage, with temperatures in the range of 30-35°C until the day 33. After this period, the temperature decreased and it remained similar to the ambient temperature until the end of the process.

The pH was slightly acid at the beginning of the process with a value of 5.7. After the first week, the pH became neutral and with the addition of fresh substrate, it decreased again to 6.2. The following weeks were represented by a second increase of pH from neutral to slightly basic conditions and it lasted until the end of experiment where a pH-value of 7.7 was recorded. The behaviour of the temperature on the three measuring points and the pH values recorded during the six weeks are depicted in Fig. 4.

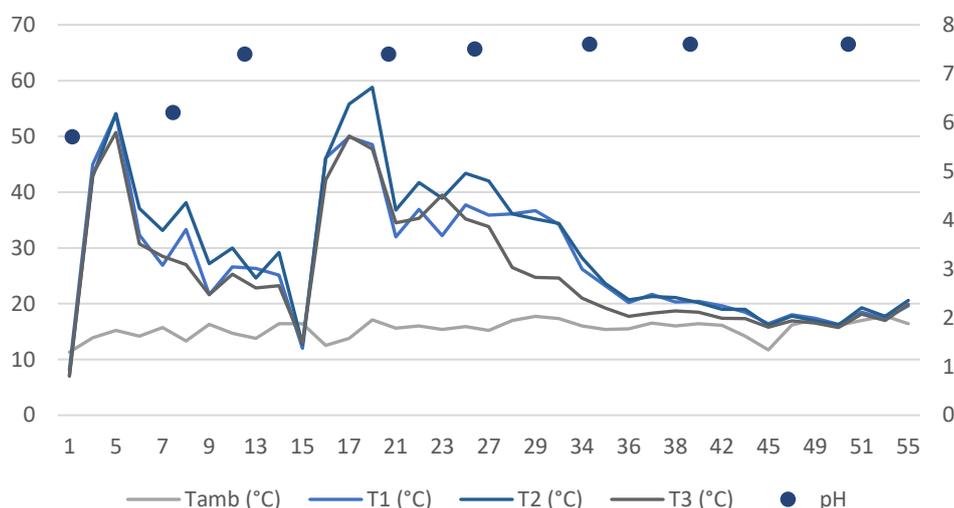


Fig. 4 Temperature and pH values in the composting process during the retention time

The final water content and volatile solids of the compost were 67 % and 71 % respectively. The high value of the volatile solids shows that the organics contained in the substrate did not fully converted into compost. In the literature a finished compost has a volatile solids content of 48 % and fresh compost of 58 % [20]. In the same way, the water content of the compost was higher than those reported in the literature, which are in the ranges of 35 % and 47 % for finished and fresh compost respectively [20]. A high water content can decrease the aeration of the material inside the drum, as water occupies the pores that should be filled with air. This leads to a negative influence on the aerobic performance of microorganisms and consequently in the material degradation [20].

Based on the behaviour of the process temperature, the water content and volatile solids of the final product, it is concluded that the production of compost was not successful in terms of quality. The temperature behaviour is a parameter that defines the efficiency of the process since it influences the microbial activity. At the beginning of the process the temperature should increase and stay in the thermophilic range for enough days to allow pathogens elimination to then slowly decrease to mesophilic range. This behaviour occurred twice, at the beginning of the process and after the tumbler was fed, which shows a dependency between the substrate mass and the temperature (Fig. 4).

The influence that the retention time and the mechanical stress had on the plastics during the process was observed in the microscopic and macroscopic aspects. A macroscopic degradation was not visible; however, attachment of impurities to the plastic surface was observed in both LDPE and HDPE. Given to the daily turning of the tumbler, most of the plastics removed from the compost were folded or curved. Microscopically, LDPE has an initial surface pattern defined by parallel lines whereas HDPE had a more solid structure in mesh pattern.

Although LDPE has branches at random places having therefore a low packing polymer chain in comparison with HDPE [31]; changes on the surface were only observed in HDPE as scratches. Since both plastics were exposed to the same biological process and boundary conditions, the microscopic aspects results cannot be exclusively attributed to the turning of the tumbler and should be further investigated.

4. Expected outcome

The environment, in which plastics are placed or disposed of, plays an important role on their degradation. Therefore, we expect different behaviours on the fragmentation of plastics during fermentation and composting. The influence of the three experimental conditions previously mentioned will provide an insight on the possible fragmentation of PE during both treatments. The biodegradation of PBAT and PLA has been tested in composting under different conditions [15–19], but to the best of our knowledge not in fermentation. Thus, the behaviour of both plastics during these treatment technologies will contribute to the assessment of their disintegration resulting on the formulation of recommendations for fermentation and composting practices.

5. Acknowledgment

Thanks to the Ministry of the Environment, Climate and Energy Economy of Baden Württemberg for supporting this project.

References

- [1] Geyer, R., Jambeck, J.R., and Law, K.L., "Production, use, and fate of all plastics ever made", Vol. 3, e1700782, 2017.
- [2] Barnes, D.K., Galgani, F., Thompson, R.C., and Barlaz, M., "Accumulation and fragmentation of plastic debris in global environments", Vol. 364, 1985–1998, 2009.
- [3] Souza Machado, A.A. de, Kloas, W., Zarfl, C., Hempel, S., and Rillig, M.C., "Microplastics as an emerging threat to terrestrial ecosystems", Vol. 24, 1405–1416, 2018.
- [4] National Oceanic and Atmospheric Administration, What are microplastics?, <https://oceanservice.noaa.gov/facts/microplastics.html#transcript>. 16.08.2018.
- [5] Weithmann, N., Möller, J.N., Löder, M.G., Piehl, S., Laforsch, C., and Freitag, R., "Organic fertilizer as a vehicle for the entry of microplastic into the environment", Vol. 4, eaap8060, 2018.
- [6] A.K. Mohanty, M. Misra, G. Hinrichsen, "Biofibres, biodegradable polymers and biocomposites: An overview", Macromolecular Material and Engineering, 2000.
- [7] Lee, B., Pometto, A., Fratzke, A., and Bailey, T., "Biodegradation of Degradable plastic Polyethylene by *Phanerochaete* and *Streptomyces* Species", 1990.
- [8] Tokiwa, Y., Calabia, B.P., Ugwu, C.U., and Aiba, S., "Biodegradability of plastics", Vol. 10, 3722–3742, 2009.
- [9] European Bioplastics, Biodegradable Plastics, <https://www.european-bioplastics.org/bioplastics/materials/>. 16.04.2019.
- [10] European Bioplastics, nova-Institute, Global production capacities of bioplastics 2018, <https://www.european-bioplastics.org/market/>. 4/16/2019.
- [11] Lim, L.-T., Auras, R., and Rubino, M., "Processing technologies for poly(lactic acid)", Vol. 33, 820–852, 2008.
- [12] Conn, R.E., Kolstad, J.J., Borzelleca, J.F., Dixler, D.S., Filer, L.J., Ladu, B.N., and Pariza, M.W., "Safety assessment of polylactide (PLA) for use as a food-contact polymer", Vol. 33, 273–283, 1995.
- [13] Weber, C.J., Haugaard, V., Festersen, R., and Bertelsen, G., "Production and applications of biobased packaging materials for the food industry", 19 Suppl, 172–177, 2002.
- [14] Ferreira, F.V., Cividanes, L.S., Gouveia, R.F., and Lona, L.M.F., "An overview on properties and applications of poly(butylene adipate-co-terephthalate)-PBAT based composites", Vol. 63, 2223, 2017.
- [15] Weng, Y.-X., Jin, Y.-J., Meng, Q.-Y., Wang, L., Zhang, M., and Wang, Y.-Z., "Biodegradation behavior of poly(butylene adipate-co-terephthalate) (PBAT), poly(lactic acid) (PLA), and their blend under soil conditions", Vol. 32, 918–926, 2013.
- [16] Leejarkpai, T., Suwanmanee, U., Rudeekit, Y., and Mungcharoen, T., "Biodegradable kinetics of plastics under controlled composting conditions", Vol. 31, 1153–1161, 2011.
- [17] Mihai, M., Legros, N., and Alemdar, A., "Formulation-properties versatility of wood fiber biocomposites based on polylactide and polylactide/thermoplastic starch blends", Vol. 54, 1325–1340, 2014.
- [18] Kale, G., Kijchavengkul, T., Auras, R., Rubino, M., Selke, S.E., and Singh, S.P., "Compostability of bioplastic packaging materials: an overview", Vol. 7, 255–277, 2007.
- [19] Arrieta, M.P., López, J., Rayón, E., and Jiménez, A., "Disintegrability under composting conditions of plasticized PLA-PHB blends", Vol. 108, 307–318, 2014.
- [20] Kranert, M., "Einführung in die Kreislaufwirtschaft", Springer Fachmedien Wiesbaden, 2017.
- [21] Kehres, B., ed., *Methodenbuch zur Analyse organischer Düngemittel, Bodenverbesserungsmittel und Substrate*, 5th ed., Selbstverlag, Köln, 2006.
- [22] Sandgehaltsbestimmung in Bioabfällen und Restmüll, Institut für Abfalltechnik und Umweltüberwachung der Fachhochschule Braunschweig/Wolfenbüttel, 2002.
- [23] DIN EN ISO 14688-1.

- [24] Reddi, L.N., Jain, A.K., and Yun, H.-B., "6 - Soil materials for earth construction: properties, classification and suitability testing", in: Hall, M.R., Lindsay, R., and Krayenhoff, M., eds., *Modern earth buildings*, Woodhead Pub Ltd, Oxford, pp. 155–171, 2012.
- [25] Su, Y.Z., Zhao, H.L., Zhao, W.Z., and Zhang, T.H., "Fractal features of soil particle size distribution and the implication for indicating desertification", Vol. 122, 43–49, 2004.
- [26] Massardier-Nageotte, V., Pestre, C., Cruard-Pradet, T., and Bayard, R., "Aerobic and anaerobic biodegradability of polymer films and physico-chemical characterization", Vol. 91, 620–627, 2006.
- [27] Mahalakshmi, V., Siddiq, A., and Andrew, S.N., "ANALYSIS OF POLYETHYLENE DEGRADING POTENTIALS OF MICROORGANISMS ISOLATED FROM COMPOST SOIL", 2012.
- [28] Shah, A.A., Hasan, F., Hameed, A., and Ahmed, S., "Biological degradation of plastics: a comprehensive review", Vol. 26, 246–265, 2008.
- [29] Jank, A., Müller, W., Waldhuber, S., Gerke, F., Ebner, C., and Bockreis, A., "Impurities in pretreated biowaste for co-digestion: A determination approach", Vol. 52, 96–103, 2016.
- [30] Jank, A., Müller, W., Waldhuber, S., Gerke, F., Ebner, C., and Bockreis, A., "Hydrocyclones for the separation of impurities in pretreated biowaste", Vol. 64, 12–19, 2017.
- [31] Arutchelvi, J. Sudhakar, M., Arkatka, A., "Biodegradation of PE and PP", 2006.