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Chemistry

Polyurethanes thermal, hydrolytic and soil degradation: Sytematic literature review

Degradação térmica, hidrolítica e no solo de poliuretanos - uma revisão bibliográfica sistemática

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ABSTRACT

A search in the Scopus and Web of Science databases covering the period from 2016 to 2021 was carried out and used to update the methodology of polyurethanes hydrolytic, thermal and soil degradation assessment. To that effect, three groups of search words were used: (1) polymer degradation AND "hydrolytic degradation" AND polyurethane; (2) polymer degradation AND "thermal degradation" AND polyurethane; and (3) polymer degradation AND "soil degradation" AND polyurethane. It was observed that the studies on the degradation of polymers are disseminated in different research groups and on a continuous basis during the survey period. The main methodologies found to perform the degradation of polymers were: method of immersion of samples in aqueous solution to evaluate hydrolytic degradation, thermogravimetric analysis, differential exploratory calorimetry and accelerated weathering to evaluate thermal degradation and inoculation of the polymer in soils with different characteristics, such as pH, moisture and organic load to assess soil degradation. Polymers can become a biodegradable solution to the environmental issues generated by plastic waste.

Keywords: Degradation; Hydrolytic; Polyurethane; Soil; Thermal

RESUMO

Materiais poliméricos são amplamente utilizados em diversos ramos da indústria e apresentam vantagens como custo baixo, durabilidade, alta resistência mecânica e flexibilidade. O poliuretano (PU) é um polímero de grande interesse para indústria devido a sua versatilidade. O uso desses materiais provoca a geração resíduos de difícil degradação. A degradação de um polímero se dá pela quebra de grandes moléculas em moléculas menores através de reações pela ação de agentes externos, como água, temperatura e a presença de microrganismos, esses agentes estão presentes na degradação hidrolítica, térmica e na degradação no solo. Sendo assim, através da busca nas bases de dados Scopus e Web of Science, foi identificado as principais metodologias para estudar a degradação hidrolítica, térmica e no solo de polímeros. Foi observado que os estudos sobre a degradação dos polímeros são realizados de forma relativamente constante nos últimos 5 anos e ainda que esses estudos são realizados por diferentes grupos de pesquisa, ou seja, é um tema muito disseminado. As principais metodologias encontradas para realizar a degradação de polímeros foram: método de imersão da amostra em solução aquosa para avaliar degradação hidrolítica, a análise termogravimétrica, Calorimetria Exploratória Diferencial e intemperismo acelerado para avaliar degradação térmica e a inoculação do polímero em solos com diferentes características, como pH, umidade e carga orgânica para degradação no solo.

Palavras-chave: Degradação; Poliuretano; Polímero

1 INTRODUCTION

Polymers are materials widely used in different industrial fields such as in the packaging sector, in pharmaceutical applications, in agribusiness and in civil construction works, because polymers are a material that has advantages such as low cost, durability, high mechanical strength and flexibility, in addition to the possibility of incorporating additives into the polymers structure (Ligier, Olejnikzac, Napiórkowski, 2021; Al Hosni, Pittman, Robson 2019).

Polyurethane (PU) is a polymer of great interest to industries; it has a urethane bond in its structure and is obtained by combining a hard segment (generally an isocyanate) and a soft segment (generally a polyol). It is possible to obtain PU with different properties that can be used for different applications, depending on the raw materials used, the reaction characteristics and even the incorporation of additives (Kwiecień *et al.*, 2020; Sahoo *et al.* 2018; Su *et al.* 2017).

Polymers used to be products with a main focus on economic development, but currently environmentally sustainable products are also sought; hence, the focus was shifted to biopolymers as alternatives to synthetic non-degradable materials. When polymeric materials such as polyurethane are not recycled, they can cause environmental problem, such as the generation of waste, which is commonly dumped in landfills, or accumulates and persists in the environment for years. Thus, studies on the degradation of these polymers are of great interest, whether studies to find ways to accelerate degradation or to obtain materials that degrade faster (Al Hosni, Pittman, Robson, 2019; Travinskaya *et al.*, 2017).

The degradation of a polymer occurs through the breaking of large molecules into smaller molecules by the action of external agents. Due to the breakage of these molecules, a change in the chemical structure of the polymers and consequently in their properties ensues, and further the loss of mass of the material can occur. There are several factors/agents that affect the degradation of polymers, such as moisture, temperature, microorganisms and solar radiation. Considering these factors, three types of degradation are important when studying polymeric materials, namely hydrolytic degradation, thermal degradation and soil degradation (Brzeska *et al.*, 2021; Feng *et al*., 2019).

Hydrolytic degradation is caused by the presence of water that causes the hydrolysis of the material and consequently the breakage of the polymer molecules. This type of degradation can be assessed under conditions that mimic the environment (seawater, rivers, etc.) or under physiological conditions (human or animal body), for example, to study drugs release (Xie *et al.*, 2018; Romero-Azogil *et al.*, 2018).

The breakdown of molecules by thermal degradation occurs by the action of temperature/heat on the polymer, which generates a loss of mass. Thermal degradation can be performed dynamically, when the mass loss is evaluated in relation to temperature variation, or isothermal, when the mass loss is evaluated in relation to time and the temperature remains constant. Degradation can occur in an inert or oxygen atmosphere (Zhao *et al.*, 2019; Satti *et al*., 2020).

Soil degradation can occur through two main factors, moisture and microorganisms (MO) present in the soil. In general, the MO produce enzymes that break the polymer chains into smaller molecules that can to be transported into the cell to participate in the metabolism of MO, then the generation of biomass, water, CO^2 or methane can occur. The degradation rate depends on the chemical composition of the polymers and the type of microorganism present; pH and soil moisture can also interfere in the process. Specific enzymes and MO or chemical agents can be added to the soil to accelerate the degradation process (Al Hosni, Plittman, Robson, 2019; Satti *et al.*, 2020; Liu *et al.*, 2017).

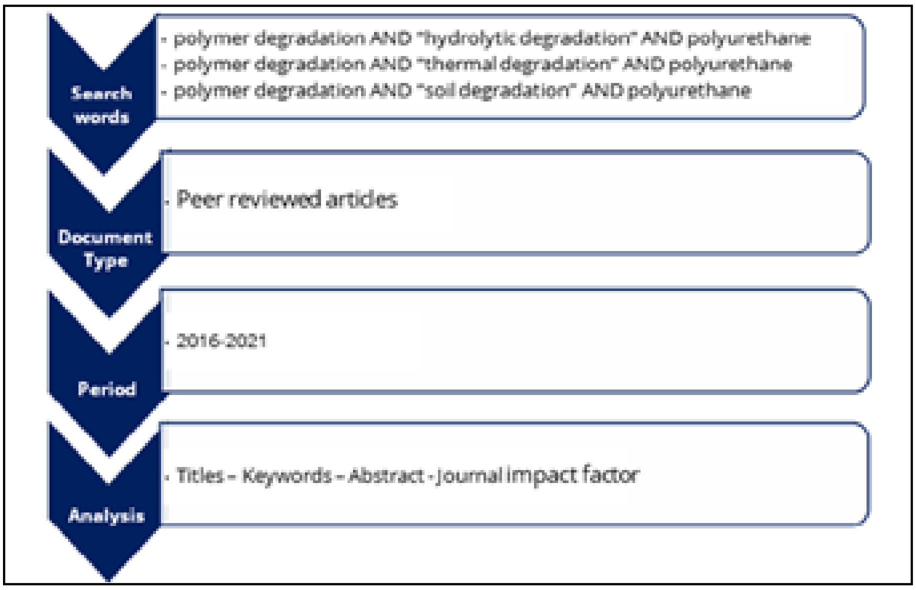
Therefore, the objective of this systematic review was to identify the main methods used for hydrolytic, thermal and soil degradation of polyurethanes, in the period 2017-2021.

2 METHODS

To carry out the systematic literature review, two databases were selected, Web of Science and Scopus, as they are among the largest databases in the world and allow access to a vast number of articles published in high impact and peer-reviewed journals, in different areas of knowledge, such as science, technology, arts and social sciences.

As it is a systematic literature review on polyurethane degradation focusing on the three aspects of thermal degradation, hydrolytic degradation and soil degradation, three groups of search words were chosen (Figure 1). Figure 1 shows the filters applied to search the databases. The search was carried out on 10/03/2021.

Figure 1 – Filters applied to the search in the databases



Source: Authors (2021)

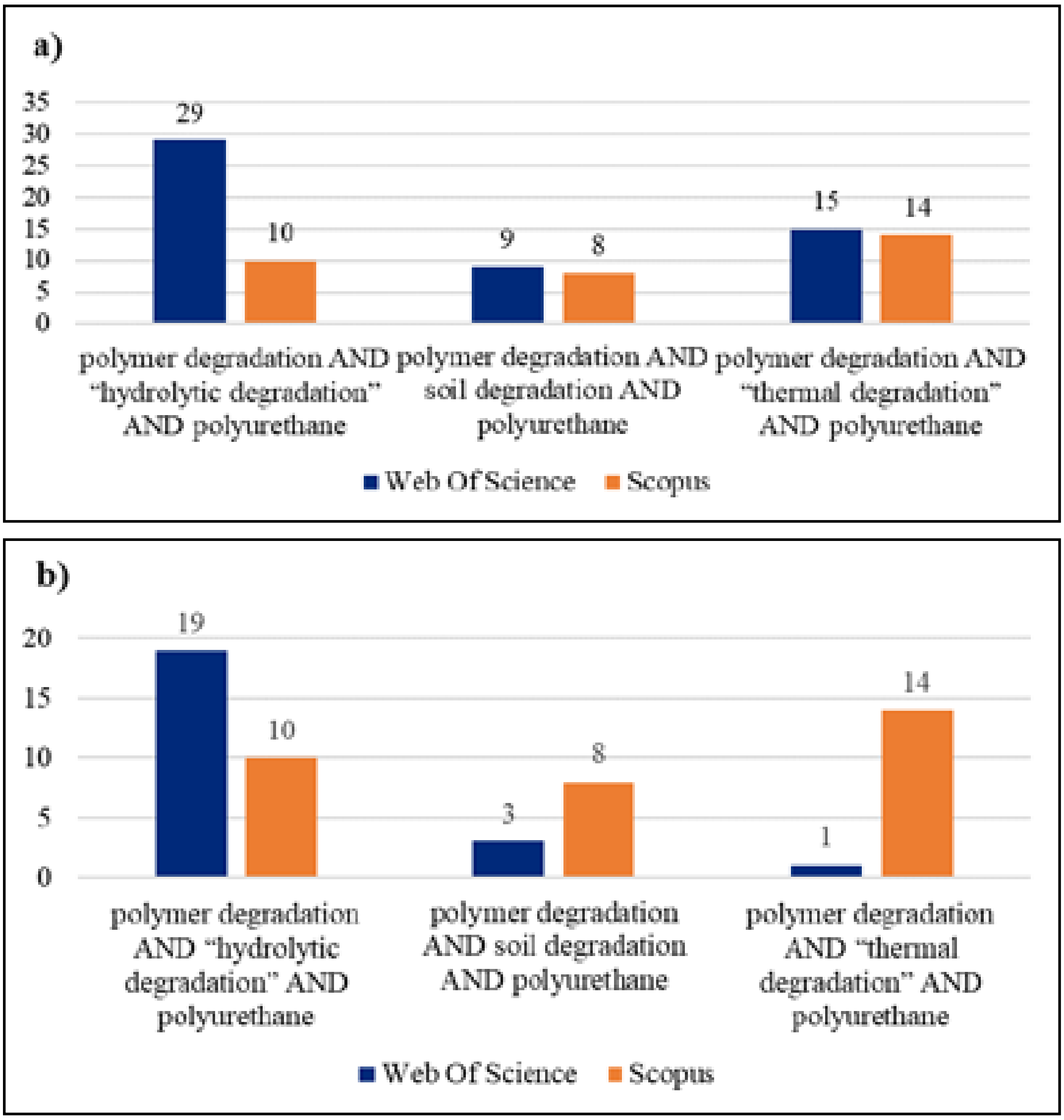
In Figure 1, the filters were used to direct the search in the databases. For the word groups polymer degradation AND “hydrolytic degradation” AND polyurethane, polymer degradation AND soil degradation AND polyurethane the words were searched in the title, abstract and keywords, while for the group of words polymer degradation AND “thermal degradation” AND polyurethane, the words were searched only in the articles abstracts. After the search, the title, keywords and abstract were read. The impact factor of the journals was also reviewed and articles found in journals with an impact factor lower than 1 were excluded.

After selecting the articles, an analysis of the publications behavior was carried out in relation to the years of publications; the journals where the articles were found and bibliometric analysis with the help of the VOSviwer Software, which allows assessing the relationships between the keywords and the authors of the articles by creating co-occurrence and co-authorship network maps respectively. Since it is a network of words in the abstract and in the title that avoids terms not relevant to the study, the terms that had at least 5 occurrences were selected. A unification of similar terms and acronyms was also carried out. And the main degradation analysis methods were identified within the selected articles.

3 RESULTS AND DISCUSSIONS

Using the search terms, 101 articles were found in the Web of Science database and 71 articles in Scopus. Figure 2 shows the number of articles selected in each database for each group of search words (a), and the number of articles after the exclusion of duplicate articles (b).

Figure 2 – (a) Number of articles selected in each database for each group of search words; (b) and the number of articles after deleting duplicate articles



Source: Authors (2021)

As shown in Figure 2a, after reading the title, abstract and keywords, and excluding articles found in journals with an impact factor lower than 1, a total of 53 articles was selected in the Web of Science database and 32 articles in the Scopus database. The articles covered polymer degradation analysis methods, focusing on thermal, hydrolytic and soil degradation. Duplicate articles were excluded, leaving 55 documents (Figure 2b), 23 in Web of Science and 22 in Scopus.

During the period 2017-2021, 11 articles were obtained for the years 2017, 2018 and 2019, 10 articles published in 2020, and in 2021, 7 articles, but this number may be different because the investigation was carried out before the end the year, in October 2021. With these data, we could observe that the publications on polyurethane degradation are current; however, the impact factor should be taken into account to determine the relevance of these publications. Table 1 shows the number of articles published per journal and the corresponding impact factor.

Table 1– Number of articles (Art.) found in each journal and the impact factor (IF)

Journal	Art.	IF	Journal	Art.	IF
Polymer Degradation and Stability	9	5,03	Polymers	6	4,329
Polymer Testing	2	4,282	Polymer Chemistry	2	5,580
European Polymer Journal	2	4,598	Journal of Polymer Research	2	3,097
Journal of Materials Research	2	3,089	ACS Sustainable Chemistry & Engineering	2	8,198
Advances In Polymer Technology	2	2,389	International Journal of Metal casting	1	1,805
ACS Applied Materials & Interfaces	1	9,229	International Journal of Polymer Science	1	2,642
International Journal of Pharmaceutics	1	5,875	Journal of Materials Chemistry B	1	6,331
Anais da Academia Brasileira de Ciências	1	1,753	ACS Applied BioMaterials	1	2,570
Carbohydrate Polymers	1	9,381	Journal of Cleaner Production	1	9,297
Chinese Journal of Chemical Engineering	1	3,171	Journal of Polymers and the Environment	1	3,667
Coatings	1	2,881	Material and Design	1	7,991
Current Microbiology	1	2,188	Materials	1	3,623
Polymer Bulletin	1	2.870	Materials Science & Engineering C	1	7,328
Journal of Macromolecular Science Part A	1	2,168	Materials Today Communications	1	3,383
RSC Advances	1	3,361	Progress in Organic Coatings	1	5,161
Journal of Coatings Tec. and Research	1	2,382	Polymers for Advanced Technologies	1	3,665
Journal of Inor. and Organomet. Polymers and Materials	1	3,543	Polymer Composites	1	3,171
Waste Management	1	7,145	-		-

Source: Authors (2021)

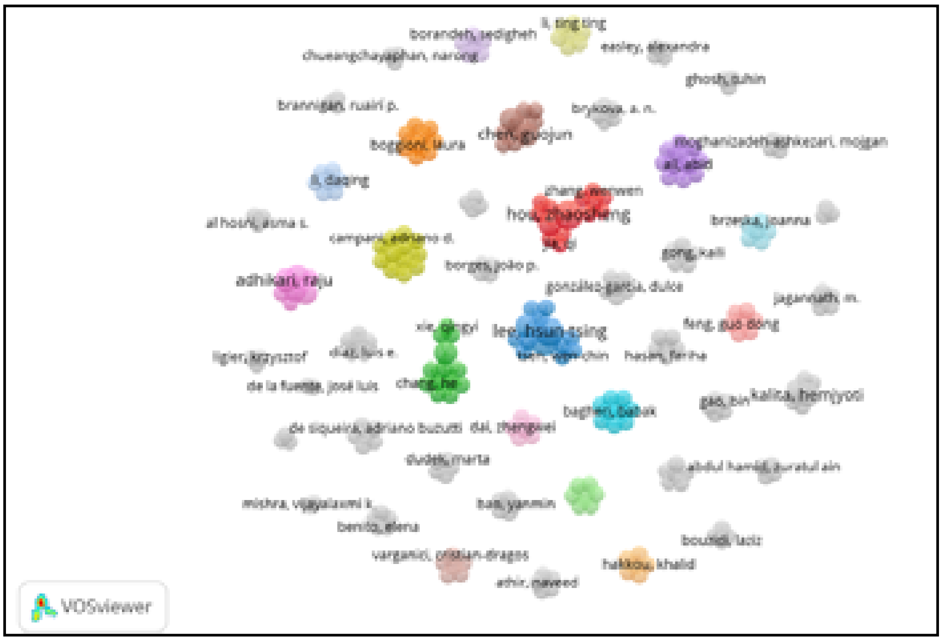
In the review of Table 1, we note that the journals that published the most articles on the subject were Polymer Degradation and Stability (9 articles), Polymers (6 articles), representing 16.36% and 10.90% of the articles, respectively. Seven journals published two articles on the topic, the Polymer Testing, Polymer Chemistry, European Polymer Journal, Journal of Polymer, Research Journal of Materials Research, Advances In Polymer Technology, ACS Sustainable Chemistry & Engineering and the others reported one paper. Thus, the relevance of the theme can be considered, since most papers are published in journals that have an impact factor greater than 2.

3.1 Bibliometrics

With the help of Software VOSwiwer, it was possible to create a network map of co-occurrence of keywords and co-authorship. The software identifies the most frequent terms in the title and abstract of the articles, and links between them and with the authors.

Co-authorship analysis consists of identifying the network of collaborations and partnerships between authors of a given subject. The co-authorship network on the subject studied (Figure 3) had 281 authors, divided into 47 clusters, which was identified by a vertex and each cluster by a color.

Figure 3 – Co-authorship network of selected articles on the subject.

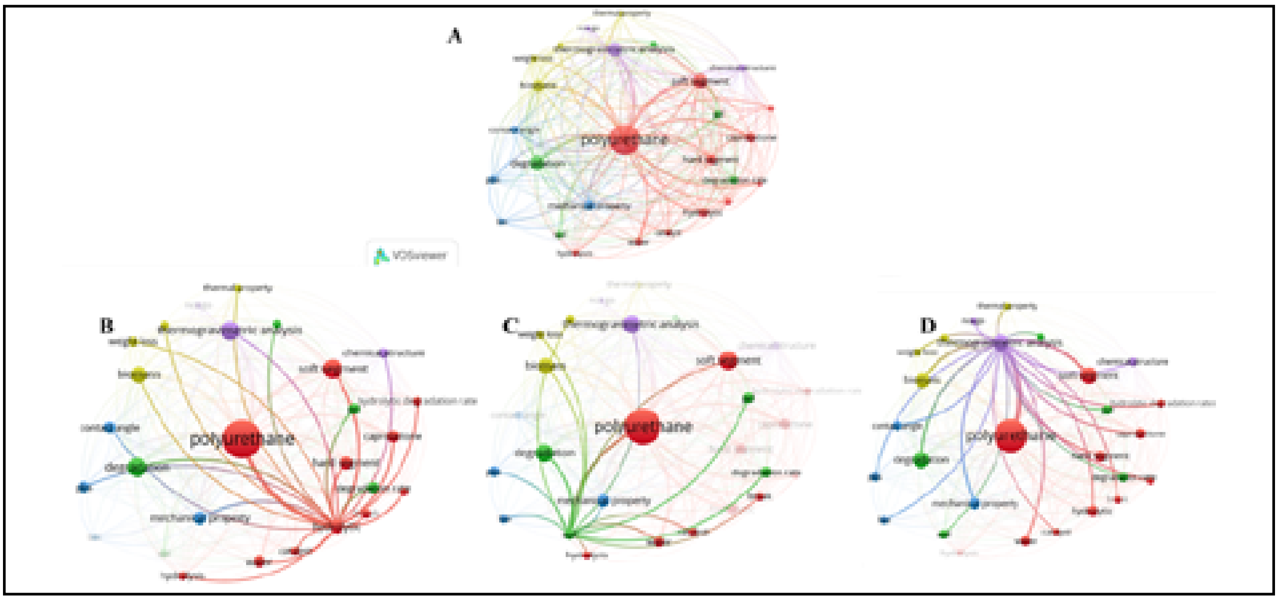


Source: Authors (2021)

In Figure 3, the gray clusters are those with the smallest number of linked authors, with fewer than 6 linked authors. The colored clusters are those with the highest number of linked authors, ranging from 6 to 14. The red colored group included the largest number of researchers, 14 authors. Moreover, it could be observed that most of the authors, the gray groups, did not have so many connections and collaborations. This behavior could be due to the fact that the topic is studied by different research groups in different countries.

The co-occurrence network of the text data was used to examine the most discussed topics in the articles and the connection between them. Figure 4 shows the co-occurrence network of the text data of the 55 articles studied. In Figure 4B, the terms related to hydrolytic degradation were highlighted; Figure 4C shows the association of the term soil related to soil degradation. In Figure 4D, for terms related to thermal degradation, elements associated with thermogravimetric analysis were identified.

Figure 4 – Co-occurrence network of text data from the 55 articles reviewed.



Source: Authors (2021)

The network generated (Figure 4A) was composed of 5 clusters and 27 terms. The size of each intersection (node) varies according to the number of occurrences of the terms. From these terms, we could identify that polyurethane presented the highest occurrence (205) and is the one that has more connections (26) with the other terms; this term was used in the search words of the three groups, so that the articles would bring information about polyurethane degradation. Other terms that stand out in the network in Figure 4A are degradation, hydrolytic, soil and thermogravimetric analysis, which are related to hydrolytic, soil and thermaldegradation, respectively.

Although collaboration between research groups is limited, interdisciplinary partnerships can promote significant advances in the understanding of polymer degradation.

The hydrolytic degradation is prominent in Figure 4A; this can be explained by the increased number (20) of selected articles that presented methodologies and due to the interest in studies on polymers in water bodies (Xie et al., 2018; Ali *et al*., 2021), biomaterials (Hou *et al*., 2021; Shetranjiwall *et al*.; 2017), and also because moisture is a factor that interferes with the degradation of polymers in the soil, and also because it is a simpler methodology to be implemented (Xie *et al*., 2018; Ali *et al*., 2017; Hou *et al*., 2020). The large number of studies conducted in the field of hydrolytic degradation reflects its importance for various technological, medical, and environmental applications. Understanding hydrolytic degradation processes is fundamental to the development of more efficient, safe, and sustainable materials. The study of hydrolytic degradation is important in several areas, including materials engineering, biomaterials, environmental science, and medicine. Understanding the switch and kinetics of hydrolytic degradation is critical for developing materials that are sufficiently durable for their specific application and degrade in a controlled manner when needed, e.g., in medical implants or disposable products.

In Figure 4B, terms related to hydrolytic degradation and polyurethane were highlighted. The term “hydrolytic” is linked to the term “PBS” (buffered saline solution), which was the main aqueous solution used in the studies selected to evaluate the hydrolytic degradation of polyurethanes. The term weight loss appears because the mass loss was monitored during the incubation period of the studied sample.

The terms chemical structure, thermal property and mechanical property are important to note after the incubation period in aqueous solution, as changes in these parameters should occur; those changes are related to polymer degradation, according to Weems *et al.* (2019) and Brzeska *et al.* (2021), which also explains the link with the term thermogravimetric analysis.

The contact angle appears in Figure 4B because it is an analysis a parameter that allows evaluating the hydrophilicity/wettability properties of the polymer, which is related to the ability of this material to absorb water, an important factor in the hydrolytic degradation process.

Figure 4C shows the association of the terms soil and polyurethane in the context of soil mining. The term soil occurred 10 times in the articles studied, and 11 articles were found reporting on the analysis of polymer degradation in soil. The term water is related to soil, as soil moisture is a factor affecting polymer degradation; on the other hand, the term bio is related to biodegradation, which is the effect of microorganisms present in the soil. Feng *et al.* (2019) and Liang *et al.* (2021) used soils without the addition of microorganisms, that is, only the effect of the microorganisms naturally present in the soil took place, while Satti *et al.* (2020) worked with the addition of the microorganism *Penicilliumoxalicum* SS2, recently isolated in soil, to evaluate the effect of these microorganisms in the degradation of polymers.

Also, in Figure 4C, it is observed that the term PBS is connected to the term soil, even though it refers to hydrolytic degradation; this occurs because the authors Sahoo *et al.* (2018) and Mishra *et al.* (2020) evaluated the two types of degradations. Due to the difficulty of performing this type of analysis, further studies are required, considering the different factors that interfere in this type of degradation, especially the presence of microorganisms (Satti *et al.*, 2020), moisture (Borrowman *et al.*, 2020), pH (Liang *et al.*, 2021) and the type of soil (Borrowman *et al.*, 2020; Jouyandeh *et al.*, 2020).

Still in Figure 4C, the terms mechanical property and break are related to the fact that the polymers undergo changes in their mechanical properties, such as rupture stress, after the incubation time in the soil, which was observed by Travinskaya et al. (2017). Even though it is a term related to thermal degradation, the term thermogravimetric analysis appears linked to the soil, as it is also a complementary analysis in studies that evaluated soil degradation.

To evaluate the terms related to thermal degradation and polyurethane, elements related to thermogravimetric analysis were identified, as shown in Figure 4D. A total of 15 items were found for the term thermal degradation, but only four occurrences of the term were available, so it was not possible to visualize the term in Figure 4D. However, thermal degradation can be represented by the term thermogravimetry, which is one of the most important techniques for determining the thermal degradation of a polymer, along with other thermal properties (Zhao *et al.*, 2019; Jouyandeh *et al.*, 2020; Reinerte, Kirpluks, Cabulis, 2019; Kalita *et al.*, 2018; Lucio, Fuente, 2017; Mi *et al.*, 2017).

In Figure 4D, it is observed that almost all the terms of the co-occurrence network are linked to thermogravimetric analysis, since the thermogravimetric analysis appears as a complementary analysis to determine thermal properties, as observed in the studies by Ali *et al.* (2021) e Hou *et al.* (2021).

Since thermogravimetric analysis is used to study the thermal degradation of polymers, the term (thermogravimetric analysis) has a strong connection with the term degradation; these studies allow us to determine the durability of the material, which is related to its degradability. The TGA tests can be performed in an inert atmosphere (N_2) or oxidizing atmosphere (O_2), when using the inert atmosphere, it is possible to assess whether there are other degradation methods, while when using an oxygen atmosphere, which simulates the environment, oxygen can modify activation energy and degradation kinetics due to oxidative reactions.

Still in Figures 5, 6 and 7, it was observed that terms such as amphiphilic polyurethane, vegetable oil, azo polymer, nanocomposite, biodegradable polymer, starch, water borne polyurethane, 3D filament, soft tissue engineering, drug release and antibacterial, found in the co-occurrence network, are related to the main types of materials and to the main applications found with the search words used. Medical application (Romero-Azogil *et al.*, 2018), in the field of soft tissue engineering (Moghanizadeh-Ashkezari *et al.*, 2018; Farzan *et al.*, 2020; Abdul Samat *et al.*, 2021) and drug release (Hou *et al.*, 2021; Polo Fonseca, Trinca, Felisbert, 2018; Macocinschi *et al*., 2020) were the terms most found.

3.2 Degradation of Polyurethanes

The degradation of a polymer occurs by breaking large molecules into smaller molecules, through the action of external agents, such as water, temperature and/or microorganisms; these agents are present in hydrolytic degradation, thermal degradation and degradation in the soil.

Degradation in water or hydrolytic degradation of polymers is a parameter used in scientific productions on polymers; using the search words polymer degradation AND “hydrolytic degradation” AND polyurethane, 29 articles were selected. Table 2 shows the title, authors, year and method used for hydrolytic degradation analysis.

Table 2 – Title, authors, year and method used for hydrolytic degradation analysis.

(To be continued)

Year	Authors	Article Title	Method
2017	Brannigan *et al.*	Application of Modified Amino Acid-Derived Diols as Chain Extenders in the Synthesis of Novel Thermoplastic Polyester− Urethane Elastomers	Immersion in 5M NaOH aqueous solution for 100 days
2017	Liu *et al.*	In Situ Polymerization and Characteristics of Biodegradable Waterborne Thermally-Treated Attapulgite Nanorods and Polyurethane Composites	Immersion in 0.1M buffered saline solution (PBS) - pH: 7.4 for 36 days
2017	Mi *et al.*	Post-crosslinkable biodegradable thermoplastic polyurethanes: Synthesis, and thermal, mechanical, and degradation properties	Immersion in buffered saline solution (PBS) for 7 weeks
2017	Mi *et al.*	Biocompatible, degradable thermoplastic polyurethane based on polycaprolactone-block-polytetrahydrofuran-block-polycaprolactone copolymers for soft tissue engineering	Immersion in buffered saline solution (PBS) for 10 weeks

Table 2 – Title, authors, year and method used for hydrolytic degradation analysis (Continuation)

2017	Panwiriyarat *et al.*	Study on physicochemical properties of poly(ester-urethane) derived from biodegradable poly(ε-caprolactone) and poly (butylene succinate) as soft segments	Immersion in 3% NaOH aqueous solution for 12 days
2017	Shetranjiwalla *et al.*	Effect of hydrothermal ageing on structure and physical properties of one-phase and two-phase entirely lipid-derived thermoplastic poly (ester urethane)	Immersion in water for 30 days
2017	Su *et al.*	Synthesis and properties of novel biodegradable polyurethanes containing fluorinated aliphatic side chains	Immersion in 3% NaOH aqueous solution for 18 days
2018	González-García *et al.*	Synthesis and In Vitro Cytocompatibility of Segmented Poly (Ester-Urethane) s and Poly (Ester-Urea-Urethane) s for Bone Tissue Engineering	Immersion in buffered saline solution (PBS) - pH: 7.4 for 120 days
2018	Lee *et al.*	Preparation and Characterization of Biodegradable Polyurethane Composites Containing Attapulgite Nanorods	Immersion in 3% NaOH aqueous solution
2018	Moghanizadeh-Ashkezari *et al.*	Polyurethanes with separately tunable biodegradation behavior and mechanical properties for tissue engineering	Immersion in buffered saline solution (PBS) - pH: 7.4 for 63 days
2018	Polo Fonseca *et al.*	Amphiphilic polyurethane hydrogels as smart carriers for acidic hydrophobic drugs	Immersion in buffered saline solution (PBS) – pH: 7.4 for 4 weeks and buffer solution of hydrochloric acid and potassium chloride (HCl/KCl) for 3 days
2018	Romero-Azogil *et al.*	Hydrolytic degradation of d-mannitol-based polyurethanes	Immersion in buffer solutions: citrate pH= 2.00, phosphate= pH 7.00, borate pH= 8.00 and carbonate pH= 10.00 for 30 days
2018	Su *et al.*	Synthesis of aromatic-doped polycaprolactone with tunable degradation behavior	Immersion in buffered saline solution (PBS) for 80 days
2018	Vieira *et al.*	Synthesis, electrospinning and in vitro test of a new biodegradable gelatin-based poly (ester urethane urea) for soft tissue engineering	Immersion in buffered saline solution (PBS) - pH: 7.4 for 37 days
2018	Villegas-Villalobos *et al.*	Effect of an organotin catalyst on the physicochemical properties and biocompatibility of castor oil-based polyurethane/cellulose composites	Immersion in buffered saline (PBS)
2018	Xie *et al.*	Poly(ester)-poly (silyl methacrylate) copolymers: synthesis and hydrolytic degradation kinetics	Immersion in artificial seawater (ASW) for 7 weeks
2019	Hakkou *et al.*	Synthesis of novel (bio) degradable linear azo polymers conjugated with olsalazine	Immersion in buffered saline solution (PBS) for 10 days

Table 2 – Title, authors, year and method used for hydrolytic degradation analysis (Continuation)

2019	Li *et al.*	Properties and degradation of castor oil-based fluoridated biopolyurethanes with different lengths of fluorinated segments	3% NaOH solution for 18 days
2019	Shah *et al.*	Thermo-mechanically improved curcumin and zwitterion incorporated polyurethane-urea elastomers	Immersion in buffered saline (PBS) for 8 weeks
2019	Weems *et al.*	Highly Cross-Linked Shape Memory Polymers with Tunable Oxidative and Hydrolytic Degradation Rates and Selected Products Based on Succinic Acid	Immersion in 0.1M NaOH aqueous solution for 140 days
2019	Xiao *et al.*	Degradable Poly(ether-ester-urethane) s Based on Well-Defined Aliphatic Diurethane Diisocyanate with Excellent Shape Recovery Properties at Body Temperature for Biomedical Application	Immersion in buffered saline (PBS) – pH: 7.4 for 12 weeks
2020	Farzan *et al.*	3D scaffolding of fast photocurable polyurethane for soft tissue engineering by stereolithography: Influence of materials and geometry on growth of fibroblast cells	Immersion in buffered saline solution (PBS) - pH: 7.4 for 6 months
2020	Hou *et al.*	Facile preparation of medical segmented poly(ester-urethane) containing uniformly sized hard segments and phosphorylcholine groups for improved hemocompatibility	Immersion in buffered saline solution (PBS) - pH: 7.4
2020	Yang *et al.*	Highly Branched Copolymers with Degradable Bridges for Antifouling Coatings	Immersion in artificial seawater (ASW) for 3 months
2020	Yaoet *et al.*	Degradable polyurethane based on triblock polyols composed of polypropylene glycol and ε-caprolactone for marine antifouling applications	Immersion in artificial seawater (ASW) for 60 days
2021	Abdul Samat *et al.*	Mechanical Properties and In Vitro Evaluation of Thermoplastic Polyurethane and Polylactic Acid Blend for Fabrication of 3D Filaments for Tracheal Tissue Engineering	Immersion in buffered saline (PBS) for 7 months
2021	Ali *et al.*	Synthesis and characterization of caprolactone based polyurethane with degradable and antifouling performance	Immersion in artificial seawater (ASW) – pH: 8.2 for 35 days
2021	Brzeska *et al.*	Degradability of Polyurethanes and Their Blends with Polylactide, Chitosan and Starch	Immersion in buffered saline (PBS) – pH: 7.01 for 36 weeks
2021	Hou *et al.*	Preparation and characterization of highly pH-sensitive biodegradable poly (ether-ester-urethane) and its potential application for drug delivery	Immersion in buffered saline (PBS) – pH: 7.4 for 18 weeks

Source: Authors (2021)

The hydrolytic degradation analysis consists of immersing the material sample in an aqueous solution and weighing the sample periodically; what changes in the methodology are the characteristics of this water and the immersion time; such parameters vary according to the environment to be simulated, which can be a simulation of the human body or the environment conditions. For materials developed for medical application (biopolymers) buffered saline solutions were used tomimic the conditions of the human body, following Shetranjiwalla *et al.* (2017) and Xiao *et al.* (2019), or even solutions with basic or acidic pH, simulating the gastrointestinal system conditions, following Romero-Azogil *et al.* (2018) and Polo Fonseca, Trinca, Felisbert, (2018).

In Table 2, to evaluate the hydrolytic degradation of polymeric materials in the environment (rivers, seas, rain, etc.), Panwiriyarat, Tanrattanakul, Chueangchayaphan (2017), Lee *et al.* (2018) and Su *et al.* (2017), worked with 3% NaOH aqueous solution to evaluate the hydrolytic degradation of polymeric materials. Weems *et al.* (2019) and Brannigan, Walder e Dove (2017) used 0.1M and 5M aqueous NaOH solution, respectively. Brannigan Walder e Dove (2017) used a 5M solution of the same base to evaluate the accelerated degradation of a polyester-urethane thermoplastic at body temperature. The test lasted 100 days and in addition to evaluating the hydrolytic degradation, it also evaluated the water absorption, which was observed in the first days of immersion. Yang *et al.* (2020) and Xie *et al.* (2018) immersed their polymeric materials in artificial sea water (ASW) for 3 and 7 months, respectively, to evaluate polymer degradation in a saline aquatic setting.

Further, Table 2 shows that the buffered saline solution (PBS) was used to evaluate the hydrolytic degradation; this solution simulates the conditions of the human body and within the selected articles PBS was frequently used to study polymeric materials applied in the area of tissue engineering, as in the trials of Abdul Samat *et al.* (2021), Vieira *et al.* (2018) and Farzan *et al*. (2020).

In 2018, Romero-Azogil *et al.* used 4 buffer solutions as an aqueous medium: citrate buffer pH 2.00, phosphate buffer pH 7.00, borate buffer pH 8.00 and carbonate buffer pH 10.00 for 30 days. To evaluate the hydrolytic degradation of D-mannitol-based polyurethane with controlled drug release. The pH variation was used to simulate the gastrointestinal tract, obtaining better results with pH 7.0 and 10.00.

Thus, to evaluate the degradation and/or durability of polymers in the environment, such as rivers and seas, basic solutions or artificial seawater solutions were used, following Li *et al.* (2019) and Ali *et al.* (2021).

The absolute majority of these polyurethanes is not soluble in aqueous solutions and undergo surface or mass erosion, so polyurethanes erosion profiles are defined not only by the hydrolytic sensitivity of the chemical bonds, but also by the physical characteristics of the material, such as hydrophobicity and morphology, which have an effect on the diffusion of water in the matrix. Water-soluble hydrolytically degradable systems are less common and of particular interest, as the impact of hydrophobicity and morphology on these systems' degradation is limited and polymer breakdown is mainly controlled at the molecular level by chemical reactions (Decollibus, Marin e Andrianov, 2010).

Table 3 – Title, authors, year and method used for soil degradation analysis.

(To be continued)

Year	Authors	Article Title	Method
2017	Liu et al.	Environmentally Friendly Slow-Release Urea Fertilizers Based on Waste Frying Oil for Sustained Nutrient Release	Incubation in garden soil for 210 days
2017	Travinskaya et al.	(Bio) degradable Ionomeric Polyurethanes Based on Xanthan: Synthesis, Properties, and Structure	Incubation in soil of medium microbiological activity (pH=6.82; relative humidity 60%) for 4 months
2018	Sahoo et al.	Degradation Study of Biobased Polyester–Polyurethane and its Nanocomposite Under Natural Soil Burial, UV Radiation and Hydrolytic-Salt Water Circumstances	Burial in soil with pH=6 for 40 days
2019	Al Hosni et al.	Microbial degradation of four biodegradable polymers in soil and compost demonstrating polycaprolactone as an ideal compostable plastic	Burial in commercial soil with pH=7.0 and moisture=26% for 10 months
2019	Feng et al.	Polyurethane-coated urea using fully vegetable oil-based polyols: Design, nutrient release and degradation	Burial in soil with 30% moisture for 120 days
2020	Borrowman et al.	Environmental degradation and efficacy of a sprayable, biodegradable polymeric mulch	Incubation in different types of soil for 3500 hours
2020	Borrowman et al.	LC-MS analysis of the degradation products of a sprayable, biodegradable poly(ester-urethane-urea)	Mixing/backfilling in Low Organic Matter (LOM) and High Organic Matter (HOM) Soil for 57 days.

Table 3 – Title, authors, year and method used for soil degradation analysis. (Continuation)

2020	Satti et al.	Biodegradation of Poly(3-hydroxybutyrate) and Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) by Newly Isolated Penicillium oxalicum SS2 in Soil Microcosms and Partial Characterization of Extracellular Depolymerase	Burial in garden soil for 2 weeks
2020	Mishra; Patel.	Synthesis and characterization of flame retardant polyurethane: Effect of castor oil polyurethane on its properties	Burial in soil with 60-70% moisture for 160 days
2021	Liang et al.	Preparation and characterization of an eco-friendly dust suppression and sand-fixation liquid mulching film	Burial in the ground for 90 days
2021	Ligier et al.	Wear of polyethylene and polyurethane elastomers used for components working in natural abrasive environments	Abrasive natural soil in rotating drum

Source: Authors (2021)

Soil degradation is a very important parameter in terms of polymer biodegradation. Using the search words polymerdegradation AND soildegradation AND polyurethane, 12 articles were selected. Table 3 shows the title, authors, year and method used for soil degradation analysis.

The soil degradation methodology consists of burying, mixing or incubating the sample in a quantity of soil for a period of time that varies from days to months. According to Table 3, different types of soil, which have different characteristics (pH, moisture and organic load), can be used to carry out the trials. Borrowman et al. (2020) used two types of soil, low organic matter (LOM) and high organic matter (HOM) and incubation time of 57 days.

Travinskaya *et al.* (2017) worked with soil of medium microbiological activity with pH 6.82 and relative humidity of the soil 60% and with an incubation period of 4 months. In addition to assessing soil degradation, Travinskaya *et al.* (2017) identified that the presence of microorganisms such as *Rhizopus*, *Aspergillus* and *Penicillium* affected the physical and mechanical properties of degradable xanthan-based ionomeric polyurethanes.

Liu *et al.* (2017) and Satti *et al.* (2020) worked with garden soil. Satti *et al.* (2020) evaluated the action of *Penicillium oxalicum* SS2, isolated from the soil of a garbage dump in Pakistan, in the degradation of polymers in the soil and observed that the presence of this microorganism accelerated the degradation of the materials assessed; within six days about 99% of the film samples of aliphatic polyester was degraded; on the other hand, in the soil without the presence of the microorganism, there was no loss of mass.

Liang *et al.* (2021) and Sahoo *et al.* (2018) and Mishra & Patel (2020) did not identify the type of soil used, only the incubation time, 90 days, 40 days and 160 days respectively. Sahoo *et al.* (2018) identified only the pH of the soil and Mishra & Patel (2020) described the soil moisture during the trial.

Table 4 – Title, authors, year and method used for thermal degradation analysis in the the selected articles.

(To be continued)

Year	Authors	Article Title	Method
2017	Manikandan *et al.*	Formation of functional nanofibrous electrospun polyurethane and murivenna oil with improved haemocompatibility for wound healing	Thermogravimetry (TGA) in a nitrogen atmosphere
2017	Lucio; La Fuente.	Structural and thermal degradation properties of novel metallocene-polyurethanes	Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) in an argon atmosphere
2018	Kalita *et al.*	Mechanical, thermal and accelerated weathering studies of bio-based polyurethane/clay nanocomposites coatings	Thermogravimetry (TGA) in a nitrogen atmosphere and study of accelerated weathering (QUV camera)
2018	Moghanizadeh-Ashkezarial.	Polyurethanes with separately tunable biodegradation behavior and mechanical properties for tissue engineering	Thermogravimetry (TGA) in a nitrogen atmosphere
2019	Monteiro *et al.*	Weathering Resistance of Waterborne Polyurethane Coatings Reinforced with Silica from Rice Husk Ash	Differential Exploratory Calorimetry (DSC), Thermogravimetry (TGA) in an inert atmosphere and study of accelerated weathering (QUV camera)
2019	Reinerte et al.	Thermal degradation of highly crosslinked rigid PU-PIR foams based on high functionality tall oil polyol	Thermogravimetry (TGA) in a nitrogen atmosphere
2019	Zhao et al.	Thermal Decomposition Studies of EPS Foam, Polyurethane Foam, and Epoxy Resin (SLA) as Patterns for Investment Casting; Analysis of Hydrogen Cyanide (HCN) from Thermal Degradation of Polyurethane Foam	Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) in nitrogen atmosphere and air (O_2)

2019	Zheng et al.	Functionalization of graphene oxide with different diisocyanates and their use as a reinforcement in waterborne polyurethane composites	Thermogravimetry (TGA) in a nitrogen atmosphere
2020	Bossa et al.	Upgrading Sustainable Polyurethane Foam Based on Greener Polyols: Succinic-Based Polyol and Mannich-Based Polyol	Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) in an inert atmosphere
2020	Ghosh et al.	Polystyrene/thermoplastic polyurethane interpenetrating network-based nanocomposite with high-speed, thermo-responsive shape memory behavior	Thermogravimetry (TGA) in an air atmosphere
2020	Jouyandeh et al.	Thermal-Resistant Polyurethane/Nanoclay Powder Coatings: Degradation Kinetics Study	Thermogravimetry (TGA) in an air atmosphere
2020	Kwiecień et al.	Durability of PS-Polyurethane Dedicated for Composite Strengthening Applications in Masonry and Concrete Structures	Differential Exploratory Calorimetry (DSC), Thermogravimetry (TGA) in an air atmosphere and accelerated weathering study (QUV camera)
2020	Pelufo et al.	Kinetic study of the thermal decomposition of castor oil-based polyurethane	Thermogravimetry (TGA) in an air atmosphere
2020	Zhou et al.	Estimating the feasibility of using industrial solid wastes as raw material for polyurethane composites with low fire hazards	Thermogravimetry (TGA) in an atmosphere of air and nitrogen
2021	Li et al.	Preparation of flexible, highly conductive polymer composite films based on double percolation structures and synergistic dispersion effect	Differential Scanning Calorimetry (DSC) and Thermogravimetry (TGA) in an air atmosphere

Source: Authors (2021)

Table 4 presents the title, authors, year and method used for thermal degradation analysis of the 15 articles found using the search words polymer degradation AND “thermal degradation” AND polyurethane.

As shown in Table 4, the technique most used to determine the thermal degradation of polymers was the thermogravimetric analysis (TGA), which allows determining the weight loss of a sample in relation to temperature and time and also in relation to inert atmosphere N2 and oxidizing atmosphere O2 and air.

In Table 4, it was observed that Kalita et al. (2018), Monteiro et al. (2019) and Kwiecień *et al.* (2020) used an accelerated weathering chamber (QUV) as a study method for polymer degradation and thermogravimetric (TGA) and/or differential scanning calorimetry (DSC) analyses were performed to verify the thermal stability of the material, as well as the thermal degradation stages. The accelerated weathering technique consists of exposing the material to an environment that mimics in a combined way the environment conditions, such as sunlight, UV, temperature and humidity. Combining three techniques, Kwiecień *et al.* (2020) managed to study the durability of PS-Polyurethane used in civil works, as well as the kinetics of thermal degradation of the material. The other authors used TGA to evaluate the thermal stability of a new material.

Thermogravimetry can work with parameters such as moisture, mass loss, volatile components and ash. Pelufu *et al.* (2020), Zhao et al. (2019), Zhou *et al.* (2020) and Jouyandeh *et al.* (2020) were able to identify, through thermogravimetric analysis, the stages of thermal degradation of polyurethane copolymers. Another methodology used was the DSC, which also allows assessing the thermal properties of different materials. Bossa *et al.* (2020) and Li *et al.* (2021), who worked with polymeric film and greener polio-based polyurethane foam, respectively, used TGA and DSC to evaluate the thermal stability of those materials. The DSC analysis complemented with information on glass transition, melting and crystallization and the thermal properties of a new material.

The controlled release technology of an active ingredient has been studied for drug delivery in humans and animals, pest control in agriculture and vectors that put public health at risk. Biodegradable polymers have been used in the controlled release of active ingredients as a means of prolonging the action of the agent in the body, in a water body, in the soil and in plants, without the need to remove the device after the desired effect. Polymer degradation studies are potential biodegradable solution to the environmental problems generated by plastic waste. These represent one of the science frontier areas, which involves a multidisciplinary scientific approach, contributing to human well-being.

4 CONCLUSIONS

A systematic literature review was carried out and identified the main methodologies to evaluate the hydrolytic, thermal and soil degradation of polymeric materials.

Polymers are widely used materials, which generate a large amount of waste in nature. Polyurethane is a polymer with several applications and of great interest to different sectors of the industry, both in the rigid segment and in the flexible segment; thus, studies on the degradation of polymers, such as polyurethane, are of great importance for society.

The literature review shows that there are many studies on polymer degradation and the number of publications in the last five years has some stability. Hydrolytic and thermal degradation are the most studied, as a greater collection of articles with these methodologies was found.

The co-authorship analysis shows that studies are spread across different research groups and that the number of collaborations among authors from different groups is low. The network of co-occurrence of keywords shows that studies aimed at application in health have a lot of strength.

The main polymer degradation analysis methodologies included thermogravimetric analysis (TGA), Differential Scanning Calorimetry (DSC), accelerated weathering study to evaluate thermal degradation. Also degradation in the soil through incubation of the sample for a set period, with the addition of microorganisms to accelerate the degradation. And hydrolytic degradation through immersion of the sample in an aqueous medium, which can be NaOH, saline water and buffer solution with different pH.

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