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TREND EDITORIAL

Biodegradability of plastics: the issues, recent advances, and future perspectives

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Plastics and microplastics, also known as polymers and polymeric materials, draw large attention today. Biodegradation of plastics has received increasing attention recently due to their recalcitrant nature, their large quantities discharged, and their accumulation in the ecosystems (Amaral-Zettler et al. 2020; Danso et al. 2019; Gu 2020), but little progress and new innovation have been made on this research topic regarding the biochemical processes and mechanisms involved in the available reported literature (Gu 2003, 2017). In this context, we would like to offer our opinions on the specific lack of good science and choice of research methodology to advance the current knowledge and understanding in several selective aspects, which would advance the research, innovation, and development on this highly publicized topic even though many papers are available (Ahmed et al. 2018; Jakubowicz 2003; Orhan and Büyükgüngör 2000; Restrepo-Florez et al. 2014; Shah et al. 2008).

It is clear that there is a large discrepancy in information between our societal needs and scientific advances made in the current situation facing this critical issue on the science and management of plastics from production to microplastics as environmental pollutants through the chain of consumption. Biodegradability of plastics has long been the central research topic for public discussion, and many of the new investigations and published results failed to addressed the fundamental and key scientific issues involved as pointed out earlier (Gu 2003) and also more recently (Amaral-Zettler et al. 2020; Gu 2017; Gu and Gu 2005). The most important research and methodology issues of this topic include (1) the chemistry and composition of the plastics, (2) the methods used for the enrichment and incubation isolation of microorganisms, (3) the metabolism of the microorganisms and the biochemical processes involved, and (4) the proper non-biological controls and reference materials (Gu 2003, 2017; Gu and Gu 2005). With these in mind, we would like to offer a further discussion on each of them to illustrate several critically important issues to facilitate the advancement of the fundamental science on this topic to serve the society better using the information presented in the recently published article as a reference example (Ahmed et al. 2018).

Substrate chemistry and composition

First of all, science is based on its universal occurrence and high reproducibility. One of the most important and fundamental issues, which are probably intentionally ignored by many, is the lack of the basic polymer chemistry of the plastics chosen for any investigations because there is almost no rudimentary characterization data on the purity, molecular weight, and molecular weight distribution of the plastic prior to any (bio)degradation tests conducted (Gu 2003, 2017; Gu and Gu 2005). Without such essential data, the so-called or claimed (bio)degradation results are both highly questionable and non-reliable, as shown in the main stream publications on this topic both previously and also currently (Gu 2017, 2020). Plastics are highly polymerized materials and their composition contains a wide range of similar molecules of the monomer as the basic and repeating unit with different chain lengths of the monomer and reaction intermediates. This property of plastics or polymers, not products, is fundamentally different from the inorganic and organic chemicals of chemistry (excluding polymers). The nature of a mixture must be recognized before any study or claim can be based on degradation. Majority of the published research papers on plastic degradation do not contain a description of such vital needed information, which

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is basic and also fundamental to the evaluation of biodegradability (see also Ahmed et al. 2018).

Plastics do not have a pure chemical composition on the molecular level. Associated with this fundamental issue of plastics, post-consumer plastics and commercial available plastics are often used for convenience in research to report "biodegradation". Consumer products and any processed plastics contain different additives including plasticizers, filler, and colorant, which are required for the final product, but they are potential carbon and energy sources to microorganisms (Gu 2003, 2017). To advance the knowledge on the science of degradation of plastics, high-purity polymer or neat resin of known molecular weight and its distribution and degree of polymerization shall be selected to cast film or mold into the desirable physical forms without involving the industrial processing procedures to avoid any contact with any chemicals to contaminate the plastic materials before the testing is conducted (Gu 2017, 2018; Gu and Gu 2005). This is the first requirement for biologists and environmental scientists to pay a very close attention to the testing materials before conducting research on the choice of materials/plastics.

Commercial films contain a range of impurities that are common in these products, including many biodegradable components, e.g., plasticizers. Because of this, majority of the published results using post-consumption or commercial products assume the products used in their testing are 100% in purity and the molecular weights are uniform completely; both of these assumptions are oversights or shortcomings due to the poor understanding of the polymer chemistry at the beginning. Again, these essential and very important properties of the plastics must be known thoroughly before the research is conducted and, in the same way, such information shall be made available to readers in any publications on this subject as a basic requirement for publication (Gu 2017).

Enrichment and isolation of microorganisms and the mechanisms

Second, any claim on biodegradation shall be supported by an active microorganism and its metabolic capabilities to alter the plastic chemistry, particularly degree of polymerization and molecular weight distribution as strong evidence. It is clear that many of the published papers do not have the key information before the claim on degradation is made very unfortunately. To isolate the potential microorganisms, it is important to conduct the initial incubation of enrichment and transfers with the pure plastic as the sole source of carbon and energy for selection of microorganisms, and series transfers are also necessary to eliminate the non-degrading microorganisms and at the same time to enrich the population of the capable member. This established procedure in pollutant degradation previously has not been adopted into plastic degradation research meaningfully. Based on the fact that a capable microbe has been obtained, the microbiology and biochemical reaction involved can be further elucidated. Currently, many published results on plastic degradation are simply made on culture medium containing a good source of carbon and energy to support the growth of large number of microorganisms, e.g., glucose, yeast extract, and malt extract. With such medium composition, the observed growth of many microorganisms on plastics or in the culture medium is not due to utilization of the plastic carbon, at least not the backbone carbon, as a source of carbon and energy to grow (Gu 2017). Concurrent analyses are also misleading, no matter on the microbial growth in the culture medium or characterization of the retrieved plastics over the time of incubation. The isolated and purified microorganisms have not been enriched selectively for their capability of degrading a selective plastic of high purity and know chemistry, and they therefore do not need the required biochemical enzymes in breaking down the plastics to grow in such culture medium because no pressure for selection is imposed to the microbial population. Unfortunately, a large number of the published papers do not show a good command over enrichment and transferring technique, and the knowledge necessary for the protocols to be used in practice, so claim or conclusion is made boldly and prematurely.

The biochemical mechanisms on degradation of plastics have not been delineated adequately in majority of the reports available except a few (Albertsson 1980; Bonhomme et al. 2003; Yoshida et al. 2016). Using ^{14}C-labeled polyethylene incubated in soil, < 1% of the ^{14}C from the plastic as a potential substrate was recovered in the released $^{14}CO_2$ over a period of more than a year (Albertsson 1980). This is the most convincing result we have so far on the biodegradability of true plastic polyethylene from a scientific research. On the other hand, lipase and laccase are frequently reported for a claimed role in degradation of plastics, but no specific biochemical degradation pathway or mechanisms could be revealed from these studies (Gu 2003; Gu 2017). Biodegradation of any organic chemicals or polymers cannot be established without thorough data on the chemistry, and microbiology and physiology to support jointly. When the chemistry of the polymeric materials and the microbiological culture medium composition are in questions, it is impossible to establish firmly the biochemical reaction mechanisms and the responsible enzymes required as key results. The microorganisms most likely have no role in transformation of plastics. Since the types of non-selective enzymes are not specific to any substrates, especially polymeric materials, natural or synthetic, no biochemical mechanisms can be established for the transformation of the plastic covalent nature—C-C bond, the backbone of the material structure. For example, lipase production can be activated to many chemicals, organic or inorganic, but the enzymatic reaction is not a selective cleavage of the plastic backbone C-C bonds specifically, which contributes then to the substantial breakage of the polymer structure, resulting in a decrease of molecular weights and a shift in the distribution to the lower molecular weights as the dominant fraction in the whole plastic. There are apparently more questions than answers in the available papers on the uncertainties about plastic degradation when the fundamental chemistry is looked into closely. Alkanes are the closest chemical structures to polyethylene and the known degradation reaction for long-chain alkanes is β-oxidation catalyzed by monooxygenase under aerobic condition. In addition, for the so many microorganisms isolated and purified for the "degradation" of plastics, not a single new biochemical reaction type has been discovered so far (Gu 2017) or no breakage of the C-C bond of the plastic can be confirmed (Yoshida et al. 2016).

It is a routine to show the Fourier transform infrared spectroscopy (FTIR) spectra of the plastic specimens after immersion in the culture medium inoculated (bioactive) and without any inoculation (sterile or non-bioactive) as a proof for a conclusion that degradation of the selective plastic occurs by comparing the spectra and the presence of oxygen-containing functional groups, e.g., -OH and -COOH, after exposure (Ghatge et al. 2020; Jakubowicz 2003; Restrepo-Florez et al. 2014; Shah et al. 2008). For this as a common practice by too many, there have been no genuine and good efforts made for a truly scientific comparison between the inoculated and the necessary controls (without any inoculation, and inoculated with sterilized inoculum or only expolymers (EPS) of the microorganisms living in the culture medium) based on the fundamental principle of scientific methods because convincing science evidence must be established on treatments with one single factor as a selective variable to draw conclusion on the cause-and-effect to advance our knowledge with high confidence (Gu 2017). The control without inoculation of microorganisms of the culture medium containing supplementary organic carbon source is a pseudo-control and does not allow yielding a conclusion convincingly with the inoculated ones for a comparison. In addition, the EPS and microbial metabolites as "contaminants" adhere onto plastic specimens to produce interfering signals to the analytical results for a false positive as material changes. EPS of bacteria in culture medium attach quickly onto plastics to yield spectra different from those without any inoculation as a control. Such control information shall be collected for a convincing interpretation of the degradation results using FTIR. Similarly, supplementary organics in culture medium can also yield similar results to mislead the interpretation for degradation. It is a pity that no serious systematic investigations have been made to evaluate this so fundamental and crucially important issue to eliminate the interferences from experimental set up and treatments in biodegradation testing by measuring neat plastics, the ones under abiotic condition, with exposure to microorganisms, and different means of cleaning to document the validity of methods and applications. Progress in science is made through innovative experimental design and the results obtained must be valid and reproducible.

Selection of testing methods

This is also critically important because different plastic products or polymers have variable physical and chemical properties, which lead to their persistent or degradation differently under the selective environmental conditions for testing (Orhan and Büyükgüngör 2000; Otake et al. 1995). Many tests were conducted incorrectly and others used the wrong methods, which show a lack of desired sensitivity to produce useful results. This issue was discussed previously in several publications for matching the plastic physical and degradability characteristics to the testing methods (Gu 2003, 2017; Gu et al. 1996; Gu and Gu 2005). There are a range of methods available to choose from, the highly sensitive electrochemical impedance spectroscopy (EIS) for persistent plastics and gravimetric for easily degradable products, on the two extremes. There are also other methods in between for specific information needs; respirometry can yield mineralization of the plastic materials if properly handled with inoculation of microorganisms and parallel controls in the same set of experiment under identical conditions (Gu and Gu 2005). At least one reference material shall be included when degradability of a new plastic is tested to provide a comparative information. Many published studies did not contain correctly set up experiments to answer the question on degradability due to poor knowledge on plastic chemistry, culture medium composition, and also analytical techniques, e.g., FTIR discussed above.

Biodegradation is defined as the breakdown of organic chemicals of the plastics to innocuous products, including CO_2/CH_4 and H_2O by (micro)organisms, including archaea, bacteria, and fungi (Gu 2003, 2017; Gu and Gu 2005). The biochemical transformation and reaction steps from a selective plastic material of pure resin to mineralization products involve a series of intermediate biochemical reaction steps before achieving the complete degradation. The initial attack on any plastics is the most important reaction and the nature of the enzymes and biochemical reactions shall be the high priority of research focus. Furthermore, degradation testing should be on obtaining mineralization of the polymers as the goal, and a mass balance and stoichiometry analysis of the plastic carbon shall be conducted to assess the degradability on material ground. Such information is hardly available in the currently published papers.

Microbial biofilms observed on materials were regarded as a "proof" of material deterioration or degradation back in the 70s–80s using scanning electron microscopy (SEM), but more advanced techniques are now available to address the key question on biodegradation without any disagreement in science for knowledge and advancement made. With the solid data on depolymerization and biochemical reaction mechanism, SEM observations can be supplementary information to support the biological nature of the transformation, but the reversal of this sequence is not acceptable. Advanced methods shall be used to deal with the scientific questions nowadays.

As such, many, if not all, reported (bio)degradation of plastics in the complex media or open environments (soil and aquatic systems) is fundamental to our understanding on the stability of this class of materials in ecosystems. In the open environments, UV-induced chemical disruption of the C-C backbone in plastics allows depolymerization of the high-molecular weight plastics randomly to low-molecular weights and also degradation intermediate products, including oligomers, dimers, and monomers, which can serve as a very good source of carbon and energy to the indigenous microorganisms. Considering the complex nature in the chemical composition of plastics, a combination of UV of natural light and the available plasticizers will make plastics good niche for a wide range for indigenous microorganisms in the ecosystems, so observation of microorganisms on plastics is expected as a common occurring phenomenon. Because of this, it is not surprising to observe dense microorganisms on surface of plastics recovered from ocean or soils. UV exposure can be used to manipulate experiments for positive results on "biodegradation" of the plastics, which are reported widely (Gu 2017). Under natural condition of aquatic or terrestrial ecosystems, a source of carbon and energy to microorganisms is always available and this will lead to the dense population of biofilm of microorganisms growing on disposed plastics even though the plastic carbon is not utilized by the adhered microorganisms.

Plasticizers and other additives

In the degradation studies, the chemical composition of the plastic has to be known and documented as a basic requirement, and this rule shall apply to plastics in a more strict way because of the high variabilities associated with plastics in their molecular weight and its distribution. It is rare to see such information of molecular weight and its distribution, and degree of polymerization in a research paper on plastic biodegradation, but plastics are high polymer products and both the monomer type and molecular weight are basic information for the description of a material type. Polyethylene has a basic chemical structure of $-(CH_2-CH_2)_n-$ as monomer; its molecular weight will determine the degradability or non-degradability and degradation rate if degradable. Furthermore, the purity of the materials is another unavoidable issue, but many investigations used commercially available products than pure resin to cast or mold their specimens for research because finish products contain a range of chemicals, including plasticizer, filler, and colorant (Gu 2003). On this aspect, processing of polymers into products requires the facilitation of plasticizer and molding chemicals so that the expected products can be made successfully. Without them, pure polymer resin cannot be converted into any quality products, so this process introduces chemicals into the finish products in the routine manufacturing processes. Few researchers have the basic knowledge about this and pay attention to this issue, which affect the degradability claim for their utilization as the sole source of carbon and energy by microorganisms (Gu 2018; Li and Gu 2007).

Plastics consist of not only the neat resin but also a range of additives to make the products in manufacturing process. The additives are potential source of carbon and energy to support the growth of environmental microorganisms, resulting in growth observed, enzyme activities, and also failure of the materials during services (Gu 2003). Commonly used plasticizers of the phthalate ester family are known to be degradable by a wide range of microorganisms, aerobic and anaerobic, from wastewater sewage sludge to sediment in the deep ocean (Gu 2003; Li and Gu 2017). Their biochemical degradation pathways are also elucidated (Li and Gu 2007). Packaging, disposable, and household plastic products are different from engineering plastics used in electronic as insulation materials (Gu et al. 1996) and carbon/glass fiber–reinforced composites in aviation and space industries due to their high durability (Gu 2003). In such case, their degradation has to be conducted with new methods of much high sensitivity than the conventional ones by gravimetric weight change or respiration (Gu 2003; Gu and Gu 2005).

Future perspectives

Science is a rigorous process involving a good hypothesis coupling with reliable methods to advance and enrich the knowledge. Because of the above, it is clear that the fundamentals on (bio)degradation of plastics have not been addressed effectively to advance the fundamental science on this subject matter of biodegradability, including published papers available (Gu 2017). Among them, the first basic information, probably the most important one, is the polymer chemistry as discussed above at the beginning, which shall be provided with not simply the monomer name, but molecular weight, degree of polymerization (number of monomers in a polymer chain) and the distribution of molecular weights as a good and basic description of the materials used similar to purity of other (inorganic and organic) chemicals used in research. In addition, the microbiology shall be enhanced for a mechanistic understanding of enrichment, series transferring, and medium composition than isolation and description of isolates from the non-meaningful media used without any selection pressure, and the verification on degradation of plastics shall be made with the most relevant methods for sensitive detection and confirmation than one method for all plastics.

Furthermore, in order to control the high quality of the results, plastic shall be experimentally made from pure resin, not from industrial machinery to avoid contamination with other chemicals. At the same time, proper control reference materials shall be implemented to produce high-quality results of the research.

Lastly, it would not make a good finish without touching on the next-generation sequencing (NGS) as a technology, which has been enjoyed by many environmental scientists and engineers greatly to provide the microbial community information of their samples/treatments for a statement. One great pitfall with the adopting of this technology in research is that a clearly defined scientific question has to be formulated to justify the meaning and value of the technology as data. Currently, many studies incorporate this technology largely due to its convenience and large data set available quickly, but there is little meaningful conclusion from the application because the abundance ranking from different samples/treatments is virtually identical. Since genomic DNA-based NGS does not provide information on the active members in the population, this apparent weakness cannot justify the active members and their contribution to any biochemical processes in the system. It may serve as cosmetics to the papers to a very limited extent and for a very short period of time. But bioinformatics of NGS data and analysis have contributed significantly to new discovery and new knowledge on ammonia-oxidation by archaea and the non-methanogenic archaea as examples for its usefulness.

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Compliance with ethical standards

The author declares that he has no conflict of interest.

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