

Environmental biodegradation of polyolefins: When, why and how?

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Abstract

The practical usefulness of biodegradable polyolefins is now being realised in commercial products, particularly in agriculture and in packaging. Polyolefins are more ecologically acceptable than traditional materials due to their low energy utilisation (and hence lower cost), lower potential to pollute the air and water during manufacture and high efficiency in protecting perishable goods from contamination during distribution. However, many environmentalists still do not accept the published scientific evidence that polyolefins biodegrade in the environment and particularly in compost and on the soil. Exaggerated claims by industry on the biodegradability of some starch-modified polyolefins in the 1980s resulted in an over-reaction from the standards organisations and has since led to the requirement that the polyolefins should hydro-biodegrade (mineralise) in the same way as pure cellulose. This is unrealistic and counter-productive. Many natural polymers such as rubber, lignin and humus do not biodegrade in this way and consequently much of nature's biological waste cannot satisfy the 90% mineralisation tests which appear to have been chosen because they are more convenient to use than full composting tests. However, such tests are meaningless when applied to hydrocarbon polymers, whether natural or synthetic, which biodegrade by oxo-biodegradation rather than hydro-biodegradation. Presently accepted standards for measuring the biodegradation of polymeric materials are unworkable and recommendations are proposed for a hierarchy of testing procedures to encompass all polymers based on scientific evidence rather than popular preconceptions.

Keywords: Environmentally acceptable polymers; biodegradable polyolefins; testing protocols, composting.

Ecological advantages of the polyolefins

Degradable polyolefins have a long history. During the 1970s a number of rather different products were commercialised based on polyethylene (Table 1). It is relevant to ask why the polyolefins were selected rather than polymers based on natural products such as cellulose acetate which was already used as a plastic in some packaging applications.

Table 1. Commercial degradable polyolefins

- **Photolytic polymers**
Ethylene-carbon monoxide copolymers: E-CO
Ethylene-vinyl ketone copolymers
Ecolyte™ (J.E.Guillet)
- **Oxo-biodegradable polymers**
Antioxidant controlled, transition metal-catalysed
photo- and thermo-oxidisable polymers
Plastor™ (G.Scott-D.Gilead), TDPA™ (EPI)
- **PE-Starch blends**
PE blended with starch (and subsequently with
prooxidants)
Coloroll, St.Lawrence Starch (G.J.L.Griffin)

In the 1970s, the polyolefins had already achieved a central position in the manufacture and distribution of consumer goods because of their combination of flexibility, toughness and excellent barrier properties, which has made them the materials of choice for packaging applications. They were particularly important in thin film technology because of their ease of manufacture and low cost. The present-day efficient distribution of perishable foodstuffs is a direct consequence of the resistance of the polyolefins and other carbon-chain polymers to water and water-borne microorganisms [1] and in agriculture, the new technology of plasticulture based on polyethylene was already making an impact on the growing of soft fruits and vegetables. [2,3]. The low cost of the polyolefins results from the very large scale of production and is associated with a lower energy utilisation during manufacture than is possible in the manufacture of traditional packaging materials [1]. The polyolefins are based on low value oil fractions and the energy utilised in the production of the same volume container is much lower than for metals, glass and paper (Table 2). Furthermore, pollution during the manufacture of polyolefins is less than that in the production of paper Table 3 [4].

Table 2. Energy used in the manufacture of typical packaging [1]

Container	Energy used per container kWh
Aluminium can	3.00
Returnable soft drink bottle	2.40
Returnable glass beer bottle	2.00
Steel can	0.70
Paper milk carton	0.18
Plastic beverage container	0.11

Table 3 Pollution generated during the manufacture of 50,000 carrier bags [4]

Environmental burden	Polyethylene	Paper
Energy (GJ) during manufacture	29	67
Air pollution (kg)		
SO ₂	9.9	28.1
NO _x	6.8	10.8
CH _x	3.8	1.5
CO	1.0	6.4
Dust	0.5	3.8
Water burden (kg)		
COD	0.5	107.8
BOD	0.02	43.1

The popular image of plastics made from petrochemical feed-stocks is very different from this. They are portrayed by the ‘green’ movement as ecologically undesirable because they contain carbon from fossil fuel resources and they are consequently resistant to biodegradation in the environment. It is ironical that the very physical properties (water and microbe resistance) that have made the polyolefins so commercially successful are a disadvantage when the materials appear in the waste stream. This is not unique to the polyolefins. The commercial polyesters and polyamides, although chemically more analogous to the natural polymers, do not biodegrade readily in the environment either and this has led to the popular belief that plastics never biodegrade in the environment. In the 1960s a well known environmentalist, Barry Commoner, suggested that because no biological organisms were known that could attack plastics, the earth would soon be buried in ten feet of plastics waste! Thirty years later, he expressed the same views in rather a different way when he stated in a report for Greenpeace [5]

“Materials made from naturally occurring or biologically produced polymers are the only truly biodegradable ‘plastics’ available. Since living things construct these materials, living things can metabolise them”.

This concept is based on an ideological view of natural processes and has nothing to do with science. Many naturally produced materials also persist in the environment for very long periods. For example certain types of wood, such as the sequoia can survive for 500 years in the biotic environment after being felled [1]. Categorical statements such as the one quoted above have misled politicians and those responsible for environmental legislation into believing that only polymers derived from natural products are eco-efficient whereas polymers derived from fossil sources are unsustainable because the carbon they contain is derived from a non-renewable resource.

The position was further confused by the enthusiastic promotion of starch-polyethylene blends as environmentally acceptable materials by companies with an interest in the commercial exploitation of excess starch. It was subsequently shown that the polyolefins biodegrade by quite a different mechanism from starch. The former undergo oxo-biodegradation and the latter hydro-biodegrade so that, at the

concentrations of starch used in commercial composites, starch does not induce the biodegradation of polyethylene [6]. However, the failure of starch-polyethylene composites to match up to the claims made for them caused a sharp reaction from legislators [7] who insisted that claims of biodegradability should be substantiated. This was a valid reaction at the time but it appeared to reinforce the popular belief that hydrocarbon polymers cannot biodegrade under any circumstances. This view has influenced many biologists who are not aware that abiotic chemistry precedes the attack of microorganisms on synthetic polymers [1] and that abiotic peroxidation plays a particularly crucial role in the bioassimilation of carbon-chain polymers. To the concerned polymer scientist, the important question is not *whether* synthetic polymers biodegrade in the environment but rather *how fast* they biodegrade and whether they will accumulate in any specific environment.

Why ‘natural’ polymers?

As a consequence of the above misunderstandings, many environmentalists have championed the cause of ‘renewable’ bio-based polymers in ‘out-of-the body’ as well as ‘inside the body’ applications. However, for commodity polymers, the concept of ‘renewability’ should take into account not only the fossil resources used in the chemical structure of the polymer, but also the fossil fuels used in the agricultural technology that goes into the production of polymers of biological origin, including fertiliser utilisation transport and the utilisation of land in competition with food crops. Little has so far been published on the eco-efficiency of bio-based raw materials for the polymer industry and there are serious doubts as to whether agriculturally produced polymers can be sustainable in the long-term [1]. The fact that there is current localised overproduction of starch in some developed countries is not a basis from which to plan for the world’s needs for the century which has just commenced. If packaging plastics were all to be manufactured from renewable resources, vast areas of land currently used for food crops would have to be appropriated for polymer crop production [1]. Less than 10% of fossil resources are used in the manufacture of polymers. The remainder goes into energy generation for transport, power stations and industry. It would make ecological sense to conserve fossil carbon resources for synthetic polymer manufacture and to turn waste agricultural biomass into energy.

Nevertheless the renewability concept is superficially persuasive and has inspired many academic scientists with expertise in polymer synthesis to develop polymers derived from natural products in order to utilise nature’s bio-recycling potential. As already seen the ecological arguments are at present unconvincing, as is the underlying assumption of environmentalists that synthetic polymers cannot be returned to the natural carbon cycle. Natural rubber (cis-polyisoprene) is a hydrocarbon polymer which oxo-biodegrades naturally in the form of latex as it comes from the rubber tree. However, it can also be synthesised in a chemical plant and synthetic cis-polyisoprene shows exactly the same behaviour in a bio-active environment [8]. The same is true of other hydrocarbon rubbers (e.g. polybutadiene) not found in nature. Strikingly, however, both natural and synthetic rubbers after formulation with antioxidants, as in automotive tyres, do not detectably biodegrade for many decades in the outdoor environment. This is because the antioxidants added during manufacture inhibit the formation of low molar mass oxidation products which are absorbed and utilised as a source of energy by bacteria and fungi. Thus the non-biodegradability of commercial rubber products thus has nothing to do with whether the rubber itself is natural or synthetic [9,10].

Polyolefins are hydrophobic hydrocarbon polymers, like the rubbers, and again, like the rubbers they are very resistant to peroxidation and biodegradation as commercial products due to the presence of antioxidants and stabilisers [9-11]. They are also highly resistant to hydrolysis and for this reason they cannot hydro-biodegrade. However, they can be made oxo-biodegradable by the use of prooxidant additives [1,8,9,11,12] leading to hydrophilic surface modification friendly to microorganisms which are thus able to bioassimilate the low molar mass oxidation products.

Fig. 1 summarises the two alternate mechanisms of polymer biodegradation which cut across the biopolymer-petropolymer divide. The first is hydro-biodegradation producing bioassimilable hydrolysis products from cellulose, starch, polyesters, etc. The second is oxo-biodegradation which generates carboxylic acids, alcohols, ketones, etc. that can be utilised by microorganisms as nutrients to produce cell biomass [11,13] similar to the hydrolysis products formed from the aliphatic polyesters [14].

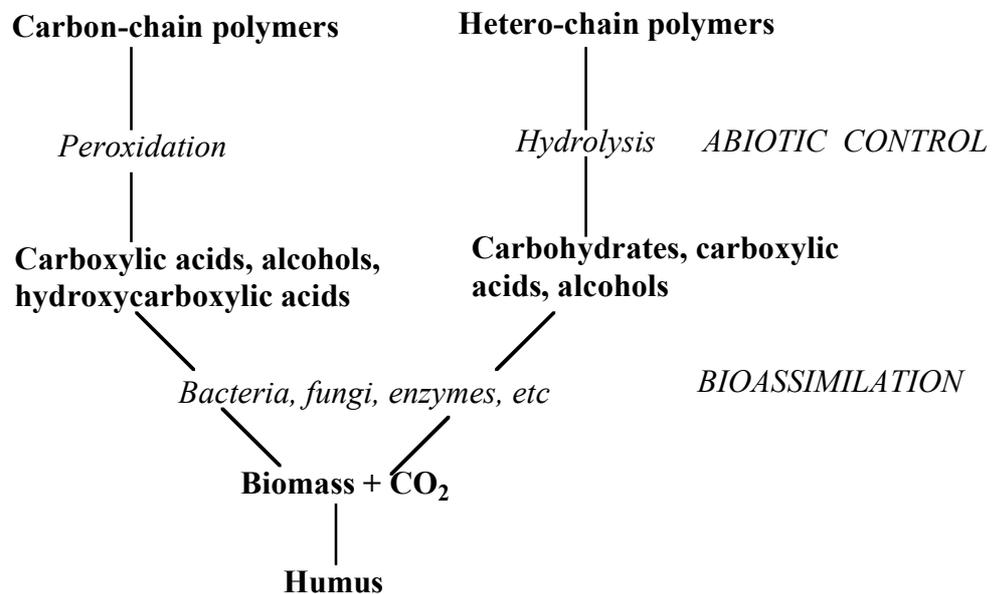


Fig.1 Alternate mechanisms for the biodegradation of the synthetic polymers [1]

Rubbers, if they do not contain substantial concentrations of antioxidants, oxo-biodegrade much more rapidly than the polyolefins. Aliphatic polyesters are more oxidatively stable but because they are hydrophilic, they are hydrolysed and bioassimilated rapidly in an aqueous biotic environment in much the same way as starch and cellulose.

Thus the polyolefins and the polysaccharides stand at the opposite ends of a spectrum of polymer properties (fig. 2). The naturally occurring hydro-biodegradable polymers such as cellulose, starch, etc. are water-wettable or water swellable. Consequently, to be technologically useful, they have to be made less hydrophilic with sacrifice of biodegradation rate. The properties of synthetic hydrocarbon polymers are a mirror image of this [1]. They are resistant to water, which is their main attribute in packaging but, in their conventional commercial versions, they do not oxidise and biodegrade at a practically acceptable rate in the natural environment and they in turn have to be made more oxo-biodegradable by the use of prooxidant additives.

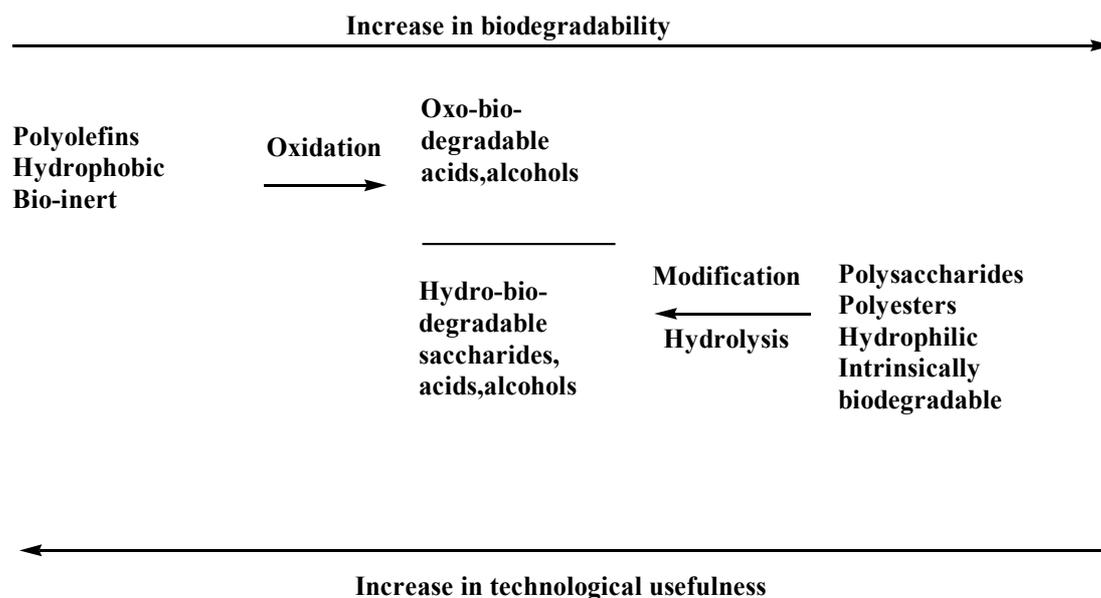


Fig. 2 Environmentally acceptable polymers from polyolefins and natural products

Between these two extremes are the aliphatic polyesters such as polylactic acid and the poly(hydroxyalkanoates). All hydro-biodegradable polymers are on a ‘knife-edge’ balance between the achievement of useful technological performance and end-of-life biodegradability. The polyolefins, by contrast, can be given a programmed lifetime by the use of antioxidants. Sustainability must in practice be a compromise between commercial viability (i.e. cost-performance) and environmental acceptability. This must ultimately mean **adaptability**; namely the potential for adapting polymers (both bio-based and petro-based) to the needs of the environment and to the needs of society. Bio-based polymers have already found important niche applications (for example in medicine) where cost is much less important than function. However, cost with environmental acceptability will continue to dominate the consumer market and it seems very unlikely that biodegradable petro-based polymers will be displaced from their current role in packaging and agriculture.

Programmed life

It is implicit in the above discussion that bioassimilation of plastics residues in the environment is an essential but not the only requirement of technologically useful plastics. In most cases plastics require a controlled lifetime before physical degradation commences. Fig. 3 shows the behaviour of an ideal degradable plastic. There should be no change in the physical and mechanical characteristics of the polymer during the induction period (IP). Moreover the IP must be variable (IP_a, IP_b) and controllable in the end environment if the full potential of the plastics are to be realised. The primary cause of hydrocarbon polymer degradation is peroxidation [11,12] and it is essential to understand and make use of this process if the full potential of biodegradable polyolefins is to be realised. In many applications, and particularly in plasticulture, mulching films and tunnels used to control the microenvironment of the plant [1-3,9], must be timed to disintegrate ($E_b < 10\%$) under slight pressure at the end of their useful life. If the films break too early, there will be a significant loss in crop yield, and if they break down too late the partially degraded plastic will clog the automatic harvester.

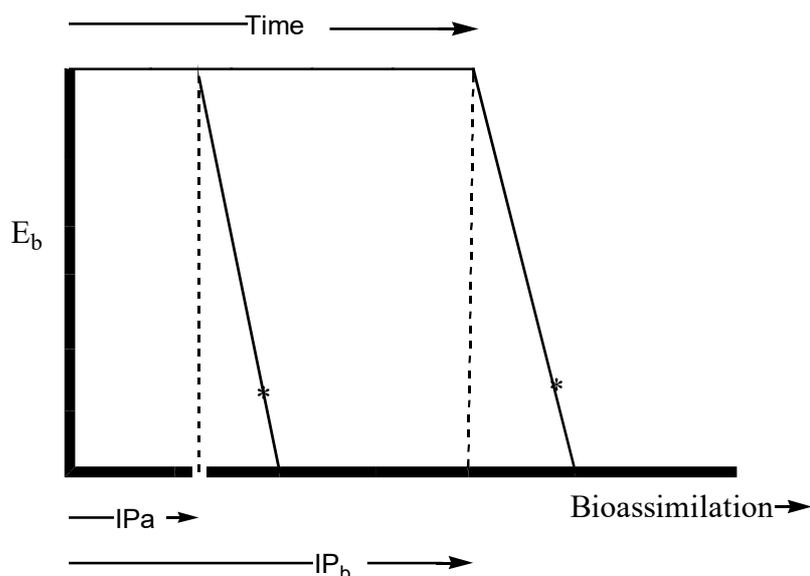


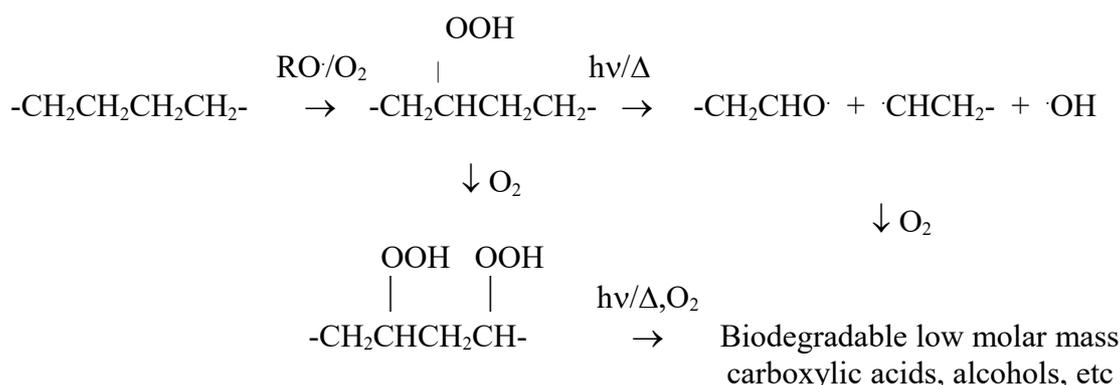
Fig. 3. The ideal behaviour of a degradable plastic in the environment [1]
 E_b = elongation at break, IP_a, IP_b are induction periods during which no change in chemical, physical or mechanical properties should occur, * is the point at which the sample disintegrates (generally $\sim 85-90\%$ loss of E_b)

Many auxiliary plastics products used in agriculture and horticulture also require an extended lifetime before losing mechanical strength. Important examples are polypropylene baler twines, stretch-wrap silage films, bird protective netting and agricultural packaging [1]. The following section summarises our present understanding of how the hydrocarbon polymers degrade in the environment by a combination of peroxidation and bioassimilation.

Oxo-biodegradation: initiation and control

Hydrocarbon polymers degrade by a free radical chain reaction involving oxygen of the atmosphere. The primary products are hydroperoxides (Scheme 1) and the latter either thermolyse (Δ) or photolyse ($h\nu$) with chain scission and the production of biodegradable low molar mass oxidation products such as carboxylic acids, alcohols and ketones. The work of Albertsson, Karlsson and their co-workers has been particularly helpful in illustrating the wide range of oxidation products and low molar mass hydrocarbon waxes that are formed in the abiotic peroxidation of polyethylene [13,15]. The process involves the formation of vicinal hydroperoxides by the 'back-biting' mechanism [15] and it may be inferred from earlier studies of polypropylene [16] that this mechanism will be even more prevalent in the peroxidation of this polymer.

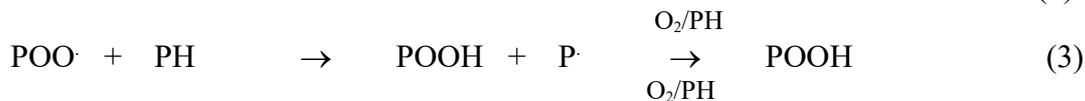
Perhaps the most interesting aspect of this work is that in the presence of microorganisms the concentration of carbonyl compounds is very much reduced [13], indicating that they are assimilated by the microbial cells as they are formed. This is consistent with the findings of Lemaire, Scott et al. [17] that microorganisms can utilise oxidised polyethylene as the sole source of carbon, leading to bioerosion of the polymer surface while leaving M_w essentially unchanged.



Scheme 1. Formation of biodegradable carboxylic acids by peroxidation of polyethylene

As mentioned earlier, antioxidants inhibit the chain reaction that leads to hydroperoxides and consequently the subsequent biodegradation of hydrocarbon polymers [9,10,12]. Thus, chain-breaking (hindered phenol) antioxidants inhibit the oxo-biodegradation of the polyolefins under thermooxidative conditions during service [1,10,12]. However, it is not a viable technological solution to the environmental persistence of hydrocarbon polymers to simply omit antioxidants and stabilisers during conversion to commercial products since they are added to protect the polymer against mechanooxidation during the processing operation and to provide the required shelf life [18]. Furthermore, commercial processing stabilisers such as the hindered phenols or phosphite esters considerably extend the life of the polyolefins in the outdoor environment [19] even though they are not normally considered to be light stabilisers.

It has been known since natural rubber became a technological product that many transition metal ions are effective accelerators for the peroxidation and hence biodegradation of the hydrocarbon polymers (Scheme 2) and that this can be retarded by certain types of antioxidant [11,20]. Hindered phenols control the metal catalysed melt degradation of polyolefins during processing but are relatively ineffective in controlling photodegradation [21]. Photodegradable polyolefins based on transition metal compounds and processing stabilisers are thus very effective in short-term applications such as garden waste bags that are intended to biodegrade in aerobic compost but not in programmed-life products such as mulching films, silage film or baler twines where an outdoor service life of up to 12 months may be required. Some of the naturally occurring oxygenases (e.g. Cytochrome P450) act in a similar way to initiate an abiotic peroxidation process and this has been shown to occur when polyethylene contains no protective antioxidant [10]. The evidence suggests that the combination of abiotic and biotic initiation of peroxidation causes a co-operative bioassimilation of the polymer [9].



Scheme 2. Iron catalysed peroxidation of hydrocarbon polymers. PH = hydrocarbon polymer, POOH = macromolecular hydroperoxide.

Since the sequence of reactions shown in Scheme 2 leads to the rapid build-up of hydroperoxides in the polymer, peroxidation cannot be controlled by chain-breaking antioxidants and the latter are rapidly destroyed by hydroxyl and peroxy radicals. By contrast, some peroxidolytic antioxidants (PDs) such as the metal dithiocarbamates, $(\text{R}_2\text{NCSS})_2\text{M}$, are able to catalytically destroy hydroperoxides as they are formed in the polymer in a process not involving the formation of radicals [11]. Consequently, peroxydolytic antioxidants based on transition metal ions inhibit peroxidation until the antioxidant has been depleted by the action of light or heat, liberating the prooxidant metal ion in a controlled manner.

$(\text{R}_2\text{NCSS})_2\text{M}$ Dialkyldithiocarbamates

M = Fe, thermo-antioxidant, photo-prooxidant

M = Zn, thermo-antioxidant, weak photo-antioxidant

M = Ni, thermo-antioxidant, photo-antioxidant.

This process has been discussed in a number of reviews of antioxidant-controlled biodegradation of carbon-chain polymers [1,8-12] and will not be discussed further here.

The biodegradation environment

The popular folklore surrounding the fate of plastics in the environment discussed above has in recent years dominated the thinking of the standards organisations. There is no question that the development of standard tests to measure the rate of biodegradation of polymers is necessary in order to ensure that residues from plastics packaging do not create a long-term pollution problem in the environment. Unfortunately the standardisation of polymer biodegradation has now reached the point where many natural products do not pass the tests laid down by ASTM in the USA and CEN in Europe. Quite unscientific explanations are put forward to explain why synthetic polymers, but not natural polymers, must be shown to be mineralised within a very short time, for example in compost. It is argued that there is no need to demonstrate that natural materials such as leaves and twigs (which contain the relatively slowly oxo-biodegrading lignin) do not need to mineralise during composting but hydrocarbon polymers are ‘different’. The following rationale is given by CEN to explain why this distinction can be made [22].

“Natural products (leaves, wood, small stones are....generally known to be non-toxic. They are universally recognised as biodegradable. On the

other hand, residues of synthetic polymers would be perceived by the general public as being contamination of the compost”

The basis for this statement is that;

“The accumulation of lignin in the environment is a natural event which is beneficial for the fertility of the soil. On the other hand, the accumulation of other foreign materials cannot be encouraged because, while it is well known that lignin is ultimately degradable and helps environment and soil structure, this cannot be claimed for synthetic products whose behaviour in the environment is not known”

From this it is reasoned that

“the CEN scheme considers lignin and the natural non-chemically modified materials as biodegradable *by definition*”

This ignores the extensive chemical studies that have been carried out on the degradation of polyolefins over the past 40 years. Nor does it recognise that lignin, a major constituent of natural products is also an oxo-biodegradable polymer similar in structure to the synthetic phenol-formaldehyde resins and like the latter, it is very stable due to the presence of the polyphenolic antioxidant functions in the polymer chain. Consequently, polyethylene, PF resins, lignin, humic acid and tannic acid oxo-biodegrade relatively slowly but are all ultimately converted to carbon dioxide and water. Humus is a complex mixture of polyphenolic compounds and the slower materialisation occurs, the more beneficial is this organic matter to the fertility of the soil.

The time-scale for bioassimilation into the environment

Biodegradable plastics may end up in sewage systems, in compost or on the soil as litter (Fig. 4).

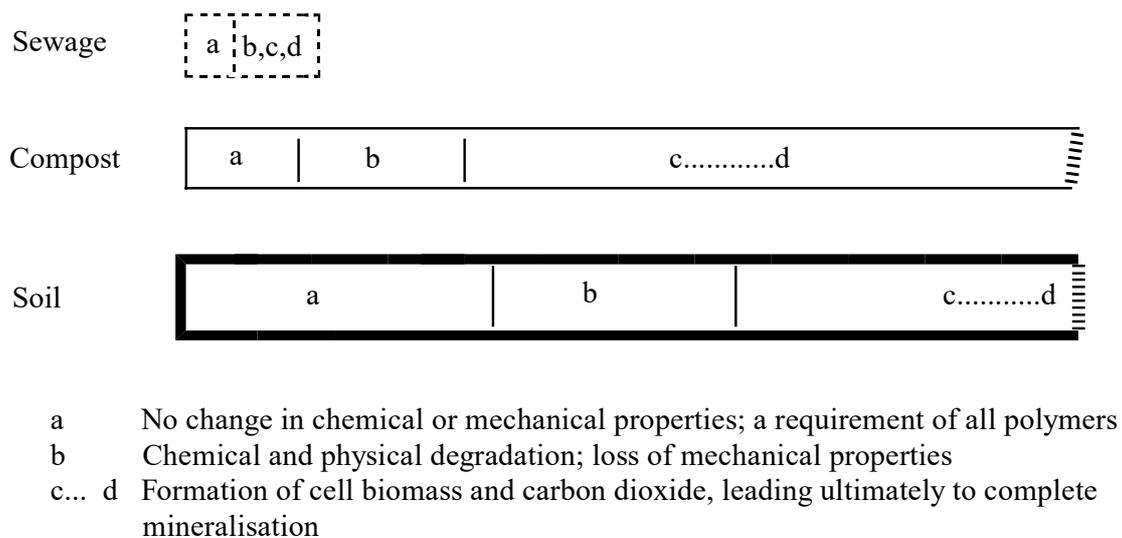


Fig. 4. Biodegradation time-scale for polymers in different environments [1]

The CEN standard for synthetic polymers in compost stipulates that they must be substantially (>90%) converted to carbon dioxide and biomass in an aqueous biotic environment within six months [23] (see below). This is to simulate the behaviour of pure cellulose which is rarely found as such in nature. The presence of lignin in natural products such as leaves and twigs slows down this process in an ambient biotic environment for the reasons already discussed. A good deal of packaging based on cellulose in combination with lignin and/or other bonding agents does not satisfy this requirement, raising serious doubts about its validity as a general standard even for bio-based materials.

EN 13432:2000 Requirements for packaging recoverable through composting and biodegradation - Test scheme and evaluation criteria for the final acceptance of packaging

- **Mineralization**
90% to CO₂ and biomass by gas evolution in 6 months
(Need not apply to natural products*)
- **Ecotoxicity**
Metal analysis
No adverse effects on compost
- **Background scientific studies**
Not considered relevant; public perception definitive*

***The CEN TC 249 WG 9 position: N18**

There are thus important ecological implications of the CEN standard since polymers that mineralise rapidly bring no benefit to compost or to the soil and by releasing CO₂ rapidly to the environment, they increase the 'greenhouse' effect. They are not therefore 'recoverable in the sense required by the European Directive since carbon dioxide is not a useful product. Rapid mineralisation is thus a waste of resources and contributes to degradation of the environment. To make it a requirement for synthetic polymers entering the compost or soil environment is perverse and counterproductive in the bio-recycling of man-made materials. The proposed standards are causing a great deal of concern to the polymer using industries because of their arbitrary nature. No oxo-biodegradable polymers including those found in nature can possibly pass the mineralisation test demanded by EN 13432. The effect of this standard is to discriminate against hydrocarbon polymers and to favour less ecologically efficient bio-based polymers, thus offering an unfair competitive advantage to the companies who manufacture them.

A number of polymer scientists, specialists in polymer degradation, have pointed out that laboratory biometric tests are no substitutes for full scale composting tests as a basis for environmental testing (see Position Statement, below). Compost evaluation permits the assessment of ecotoxicity on the final product in the field under real conditions. Equally importantly laboratory measurement of abiotic peroxidation and the rate of subsequent bioassimilation of low molar mass oxidation products provides complementary information that can be used to assess the time scale for ultimate absorption of the polyolefins into the environment.

Position Statement concerning biodegradable plastics

A-C.Albertsson, N.Billingham, E.Chiellini, G.George, J.Lemaire, B.Raninger, G.Scott, G.Swift, D.M.Wiles.

“Biodegradability tests that have been developed largely reflect the behaviour of hydro-biodegradable polymers (e.g. aliphatic polyester, modified starch). These materials are ideal for rapid biodegradation in sewage sludge where a maximum rate and extent of mineralisation is required. The fundamental characteristic and most positive value of compost or mulches is the presence of biomass. Without biomass, there simply would be no product.

Rapid mineralisation is not ideal for polymers in compost where the carbon in the original plastic should be converted over a longer period of time to biomass and only slowly to carbon dioxide. The oxo-biodegradable polymers (e.g. the polyolefins) are ideal for this purpose since controlled peroxidation is the rate-determining step in the overall process. Furthermore they cannot give toxic or otherwise objectionable by-products during bio-assimilation”

Similar concerns have been expressed by the European Commission Directorate for Industry and the Environment [24]

European Commission Directorate E - Industry and the Environment. Comment on prEN 13432; Organic recovery

- **“ISO 14851 (Oxygen consumption) and ISO 148 (Sturm test) do not simulate composting conditions**
- **What is really needed is to know what is the fate of materials under composting conditions and what happens once it is released to the soil**
- **If the packaging material does not completely biodegrade during the composting process, it should be demonstrated that it eventually degrades in the soil”**

It has been proposed that there should be an immediate revision of EN 13432 to take into account scientific studies on the biodegradation of polyolefins carried out during the past 8 years. This should be based upon the following hierarchy of tests and the scientific basis for them.

Composting of polyolefins: test hierarchy

- **Full scale composting trials**
 - Particle size reduction**
 - Visual impact**
- **Ecotoxicity measurements**
 - Plant germination and growth rate**
 - Accumulation of transition metals in stems, leaves and fruit**
 - Effect on macroorganisms (worms, daphnia, etc.)**
- **Background scientific studies**
 - Rate of abiotic peroxidation at composting temperatures**
 - Rate of biomass formation and polymer weight loss**
 - Biometric measurements on peroxidised polymer**
 - Correlation of bioerosion with extent of peroxidation**

A number of basic scientific studies have already been published which are relevant to the oxo-biodegradation of polyethylene both *in vivo* [25,26] and *in vitro* [17,27]. For example it has been shown that some soil bacteria and fungi can utilize peroxidised polyethylene in the absence of any other source of carbon leading to bioerosion of the polymer surface [17]. Biometric tests (oxygen absorption) have confirmed that the thermal peroxidation products formed on the surface of the polymer are rapidly mineralised [27]. Studies are currently in progress to correlate the extent of peroxidation of polyolefins with polymer weight loss both during abiotic and biotic assimilation processes.

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