DEFRA Report EV0422: Assessing the Environmental Impacts of Oxo-degradable Plastics Across their Life Cycle

PEER REVIEW on the reply by Loughborough University to the Response by the Oxo-degradable Plastics Industry submitted by the British Plastics Federation.

Key questions:

1. In your opinion, has the available scientific information been adequately reviewed, both in the original report and in the research team's reply to the *Industry Response*? If not, please provide details and explain your reasons.
2. Have all the points raised in the *Industry Response* been adequately addressed by the research team in their reply. If not, please provide details and explain your reasons.
3. Are the conclusions put forward by the researchers logical and reasonable in the context of the evidence considered. If not, please provide details and explain your reasons.
4. Are there any other points that you consider important and that you consider have not been adequately covered and assessed in the original report and reply. Please detail and explain.

Documents provided

1. "Reply to the Response by the Oxo-degradable Industry", a report prepared for the Department for Environment, Food and Rural Affairs by Loughborough University, January 2012.
2. "Industry response to the report", Information consolidated by BPF on behalf of the principal UK industrial stakeholders; EPI Europe, Symphony Environmental and Wells Plastics.
3. EV0422 Assessing the Environmental Impacts of Oxo-degradable Plastics Across Their Life Cycle, Loughborough University.
4. Instructions to reviewers.

REVIEWS

Four reviewers were invited to undertake reviews, two of these were from a list provided by the BPF, one from the UK (A) and one from Sweden (B). Defra independently selected two other reviewers, one from Mauritius (C) and the other from the UK (D). All four reviewers provided confidentiality agreements and declarations that there were no conflicts of interest. The possibility of conflicts of interest (and perceptions thereof) was considered of particular importance in arranging this peer review.

The three reviews that were received are provided below. Reviewer D failed to provide a written report.

REVIEWER A

Professor Norman S. Allen
Professor Emeritus
Manchester Metropolitan University
UK

This peer review report by invitation of DEFRA has been prepared in the hope that it will resolve a number of the issues and perhaps misunderstandings by the authors of the DEFRA Report EV0422 ***Assessing the Environmental Impacts of Oxo-degradable Plastics Across their Life Cycle*** on behalf of DEFRA and the comments made by the Oxo-degradable industry in their response. The issues raised have been addressed under suitable headings, which provides a useful format for the peer review and these will be followed accordingly.

- Mechanism of Oxidation
- Oxidation Products and their Biodegradability
- Timescale of Oxidative Breakdown
- Biodegradation studies
- Toxicity / Toxicological Impact
- Persistence in the Soil / Bio-accumulation
- Recyclability
- Conclusions

Introduction

Over the last 50 years or so with the ever aspiring and increasing usage of thermoplastics like polyethylene in the consumer market there has been much political pressure worldwide on the industry to overcome the problems associated with their end-of-term use and waste problems in the environment. Indeed both industry and academia in its various formats have endeavoured to tackle these issues albeit in some cases partially. The latter wording is used with care as it perhaps reflects the early ignorance of the mechanisms and expectations of the processes involved and the realities and experiences gained. Indeed early attitudes of the general public were simply put “If you cannot see the problem then it was solved”. So, the industry set about developing a range of plastic materials like for example films, which degraded either in light, bacteria/soil, water, thermally to give smaller fragments many of which so called disappeared from sight. Agricultural mulching film was one of the first where organic degradable additives such as starch were incorporated into the polymer. The idea being that the bacteria ate the starch and while they were at it might as well eat the polymer chains also or so they thought. Farmers soon became disillusioned by this process as year after year the residual polyethylene built-up in the soil and became entangled in their machinery. In fact, much of the farmed land began to look like a refuse tip with strips of black/white film sticking out of the ground. It was a good effort but clearly had it been given some afterthought and insight could have been avoided. So, moving on to the modern day plastics waste solution to now, what we call oxy or OXO-degradable plastics (ODBs) the technology has developed quite significantly whereby the plastic is degraded in general to a point no longer visible to the human eye through a number of chemical/bacterial processes. To be brief the science here was to develop plastics with A: built-in functional groups that we knew would assist both thermal and light and bacterial activity i.e. carbonyl groups. If one can enhance the hydrophilic nature of the chains then their degradability can be increased. The second type B: is where the industry could add compounds that were known to enhance/accelerate the mechanisms of oxidation to differing degrees/modes like for example, aromatic carbonyls, transition metal salts and other compounds. Indeed many patents have been filed over the years covering almost every organic and inorganic compound you may find in chemical catalogues. On the biological front of course today we have the newer naturally degradable polymers like Poly(hydroxybutyrates) and poly(lactic acids) to name but a few. The latter are high cost with limited functionality in the commercial place although strenuous efforts are being made to enhance their viability especially now in relation to their thermal stability.

Of course ODBs were developed in each case for a specific fundamental usage. In other words the plastic had to last for its natural useful lifetime and not degrade until its practical use was completed. There are many cases in point here such as bottles and plastic films and bags etc. This meant that the additives were used at a given concentration for controlled use in each functional case.

Although ODBs degrade and breakdown with severe loss in physical/mechanical properties and in some cases evidence of chemical oxidation this meant the formation of smaller and smaller particles of material of lower molecular weight often down to the macro/microscopic level. The problem now raises its head as to whether or not such material degrades further in the long term before any other type of environmental damage might occur i.e. before aquatic/animal toxicity arises. This question raises an important issue perhaps not realised in either of the reports and that relates to the longevity of the controlled additives used. In small particles of film in water/soil environments extractability effects and consumption will no doubt have a bearing on any further degradability processes. This issue has never been examined scientifically. Any organic/inorganic compounds for example, will have a limited lifespan. Most of the oxidation products in the polymer fragments may have been released as carbon oxides or methane etc. but this still leaves fragments with a lower but still effective molecular weight, albeit less with little or no residual degradents. So where do we go from here as industry has done its job to the best of their ability and yet now in the modern world we have concerns over the eco-system where for example, the oceans are full of planktonic like plastic pieces on the nano-scale that fish tend to eat causing possible harm and death.

Once an ODB has done its job and in many cases been reduced to a fragmentary state there is indeed little one can do any further to alleviate the issue. Very low molecular weight fragmentary plastics are not suitable for recycling especially in the context of mixed waste plastics or virgin material unless the end-use is of a low value state. One of the main issues here is related to the fact that any residual pro-oxidant additives present in the mixed recyclate will transfer to the non-ODB material. If this is the case then the advice would be that recycled ODBs can only be re-used in limited applications i.e. further ODB applicability only. Breakdown products of the

ODB additives have also to be considered and may still reside in the plastics material and with regard to this issue has had little assessment and analysis.

There are indeed many uses of recycled polyethylene/polypropylene for example, railway line sleepers but the use of any ODBs in such material could be devastating in the long term in terms of any failure and liability. Despite all these issues the question remains how to deal with the problems of ODBs in the eco-system-can we reduce the degradation completely to a safe state where particulate toxicity levels are low. This may mean a change in ODB activity and the use of long-term residual active additives. New research and developments perhaps in terms of grafting active components might be one solution for the future.

Mechanism of Oxidation

There is clearly some argument here amongst the groups concerned over the importance of the general mechanistic features of polyolefin oxidation and to be honest is of little relevance to the final outcome of the DEFRA report. The literature is indeed abundant with reports and trials from academia and industrial/government organisations illustrating the viability and efficacy of ODBs in various environments all showing significant reductions in polymer molecular weight and carbonyl group formation. It's not for the Peer review to re-dwell on these outcomes but lest be clear that each report is specific to a particular material and application and degradation condition. Whilst one always wishes for rapid assessments and results there is no alternative to that of natural ageing tests in the long term as pointed out by the Industry. Significant reductions in molecular weight are fine and great step forward but the argument remains with smaller residual particulates and how to overcome their presence in the ecosystem. They may well degrade completely in the very long term but only after the damage has been done.

The issue over the role of metal ions in the degradation route is defined and made clear although the industry response wishes to harden the role they make. It has little relevance other than the fact that metal ions do play a crucial role in accelerating the decomposition of hydroperoxides and enhancing the rate of chain scission-in fact, the higher the ionic valency the faster the rate. So, the mechanisms are not in dispute just the extent to which they play a role in the degree of chain scission and how fast you get there. To conclude both parties have adequately covered the main issues and the response given requires no amendments.

Oxidation Products and their Biodegradability

The role of degradation products such as the alkoxy radicals appears here to be the centre of some clarity in relation to biodegradability of the OBD polymer. The DEFRA report has made its point in fact its known that such radicals are the main point of beta-scission following the breakdown of hydroperoxides on the chain. There is little point in highlighting the matter further. Reference is also made to studies showing that the low molecular weight species produced from accelerated oxidation of oxo-degradable plastics can be dissolved in acetone as if this is an argument for the viability of OBDs in the environment. This is not unexpected, as the response document makes clear. However, we don't live in an acetone environment at least not yet!

Chiellini and co-workers studies have been have been under some discussion and well covered adequately in the DEFRA response and indeed as indicated the work carried out is not definitive evidence for complete biodegradability. The DEFRA response also to the other personal communication by Chiellini is well argued and requires no further amendments. It's again made clear that the studies are not totally relevant showing low levels of mineralisation and this is further illustrated by the work of Eubeler.

One area perhaps not mentioned in the context of oxidation products is in relation to the role and mode of action of the OBD additives or more correctly pro-oxidants. Maybe this needs highlighting in the report as both parties appear to have missed the point mechanistically. Studies on OBD additives for accelerated degradation have clearly shown that they operate as pro-oxidants (term often used to define them). Here the additive post accelerating effect is actually related to its pre-thermal oxidative effect in the polymer during processing giving rise to the formation of carbonyl and hydroperoxide groups. The latter species will subsequently contribute towards the degradation processes.

The industry response makes it clear though that proper standards need to be established indeed this is so but how to establish this is a problem. One case in point is the work undertaken by for example, film archives on the longevity of motion picture film such as cellulose triacetate where the nature of the environment plays a crucial role in film degradability. Here when the environment is inside iron cans the life of the film is 35 years while inside a plastic container can be as high as 90 years. So, the argument here is in laboratory tests on biodegradability of ODBs how the nature of the experimental setup may influence the outcomes-glass containers versus iron or aluminium? This leads us onto the next section of relevance.

Timescale of Oxidative Breakdown

The response of DEFRA to the oxo-degradable industry response with regard to timescales of breakdown again raises a number of issues. As mentioned above realistic conditions are important here whether or they are accelerated to shorter time scales and this may not be practicable anyway as mentioned. DEFRA have provided a balanced response here again highlighting the fact that many studies are unrealistic. Additive concentrations, humidity, temperature and soil conditions such as pH and metal ion content all reflect on activity and lifetime. Thus, high basicity and acidity and iron contents may be more effective to the lifetime. Arrhenius predictions undertaken by many workers are also irrelevant based on the fact as one moves to more realistic and lower temperatures mechanisms will change. For example, polymers will produce a range of hydroperoxides in their lifetime of differing structures each having a critical thermal activation point such that whilst most peroxides will decompose at the higher temperatures only the less stable ones will continue to operate at the lower temperatures. This needs highlighting if one wishes to argue the point.

The response document also discusses the issue of synergism between thermal and biodegradation. This point is open to many parameters since it may be argued that greater thermal treatment and production of hydroperoxide and carbonyl species will perhaps accelerate biodegradability. Clearly another point requiring clarification and study although difficult. The argument here being that the more you pre-oxidise your polymer the poorer the initial physical properties of the material.

Rates of polymer degradation will also be controlled by the initial amounts of stabilisers added to the material whether they be antioxidants or for example, light stabilisers for differing reasons in providing useful physical characteristics to the polymer during its practical lifetime. In many cases processability is important. These compounds will prolong the life expectancy of the polymer depending upon the conditions required for mineralisation or biodegradability or both. During thermal and light ageing of course the active OBD additives can influence this process during thermal and light ageing. For example, metal ions will catalyse the decomposition of phenolic antioxidants, stripping off their functional alkyl groups from the phenol ring structure. This can be an issue in considering the recycling of waste ODBs when admixed with normal polymers where residual active ODB additives will contaminate and destroy the stabilisers. This is the reason why it’s so important to consider the re-use of waste ODBs for degradable polymer use only.

Biodegradation studies

Here there are two issues in which the industry response and DEFRA are in dispute. The first relating to biodegradability and compostability where there is an apparent misunderstanding and public viewpoint causing problems and the second stating the possibility of producing an effective polymer which can be rapidly mineralised although not yet proven so is based on supposition by the industry. DEFRA have made their point clearly here and require no further clarification other than to make the point perhaps that compostability normally reflects cellulosic or other natural polymers so DEFRA’s response is justified. Frankly, the term compostable is inappropriate for thermoplastics and more applicable to natural polymers. If a polymer is mineralised into mainly carbon oxide gases and water etc. then what is left over is not compost.

Toxicity / Toxicological Impact

I agree totally with the comments made in this section of the reports and have no further comments to make at this time unless standards change and require it.

Persistence in the Soil / Bio-accumulation

Here the the oxo-degradable additive manufacturers criticised the DEFRA report on two grounds the first relating to the hazardous nature of polymer fragments remaining in soils and secondly environmental hazards of fragments of ODB polymer being washed out to sea or rivers and their capability of absorbing pesticides.

Here the arguments presented by DEFRA are strongly supported by the literature and the conclusive evidence as is known anyway in degradation chemistry that polyethylene will crosslink and clearly this will hold off any further degradation taking place resulting in smaller long lasting micro-fragments. Indeed such material would be more difficult to biodegrade.

On the second issue regarding the environmental effects of micro-particulate polymer materials in the air and marine waters this is an area as pointed out by DEFRA for further research. The news media in recent years has highlighted this issue already having found polymer particles in marine environments and inside fish so they must have come from somewhere. This requires no further elaboration other than some concerns for the future direction of ODBs.

Recyclability
On this issue there is a clear conflict of evidence regarding the viability of utilizing recycled ODBs with normal virgin or waste polymer materials. The evidence provided by DEFRA is very clear and conforms with what would be expected that ODBs would have a deleterious effect of the properties of non-ODB polymer materials. Of course the effect may depend very much on the extent to which the ODB polymers have been degraded and how much of the original active additives remain. Since the viable use of an ODB polymer is for it to disintegrate from normal "view" then it would not be from a practical point recoverable. As indicated in the report an often lack of history on the ODB adds further complications in any recycling operation. In any case as pointed out earlier if ODBs were to be recycled then the post consumer usage should only be in the same direction.

Another issue raised by the ODB industry was the fact that stabilisers may be added to the waste polymer to offset any activity of residual additives or pro-oxidant effects. This of course may or may not be possible depending upon the type and nature of the ODB. As pointed out earlier some pro-oxidant additives can destroy the antioxidant and stabilizer action-metal ions being a notable case in point.

Clearly, there are concerns as mentioned in the DEFRA report with recycling ODBs and therefore recommended that collection systems are set up to create separate streams for both of these types of new materials. This is important despite current objections by the ODB industry and the BPF. Similar issues of concern are also mentioned in the report for other polymer waste streams such as PET where for example, discolouration would in many cases prevent its re-use in many applications especially film and bottles.

As indicated in the DEFRA response document in the UK, the British Plastics Federation on their own website raises concerns about recycling of degradable plastics (biodegradable and oxo-degradable). In their response report they now state it's not a problem - a somewhat hypocritical statement.

Conclusions
The original report laid down by DEFRA/Loughborough University is basically sound and concise in their summary and coverage of the mechanistic features associated with the degradation of polyolefins and ODB polymers although not comprehensive as there are many textbooks and reviews on the subjects. This is not the prime objective of the report but to highlight the problems or otherwise associated with the usage and recycling of ODBs in the mainstream industry and its impact on the eco-system with supportive evidence. In this peer review some other perhaps important aspects of the mode of action of ODB polymer additives has been highlighted such as pro-oxidant effects and the antagonistic effects on conventional stabiliser systems.

Loughborough University Polymer department is a renowned centre of excellence worldwide with many areas of expertise and possibly represent the better group of experts able to provide an independent and unbiased overview of the effects of degradation in the recycling industry. There are no serious errors in their assessment.

The fate of the oxo-degradable plastic after it has fragmented to a fine powder of course as mentioned in the report is not clear. If the fine particles are found to persist in the environment for a long period of time then further research is required as mentioned to assess not only the impact this may have on the eco-system in general but also and more importantly to determine whether or not the fine particles emanate from the use of ODBs. One recommendation to the ODB industry here is to perhaps look at more effective ODB additives, which are effective and long lasting and remain in the polymer material to the bitter end. Some of the more viable catalytic metal oxides such as nano-titanias might be worth investigation here.

The other important issue is the uncertainty surrounding the effect of oxo-degradable plastics on the conventional plastics recycling process. A useful way forward here is to either keep ODBs out of the main stream recycling operations or if indeed included then to ensure that the products end up in ODB applications.

As for the use of plastics waste in composting there is clearly some misuse of the term in the industry not for this report to dwell on but basically this is for natural polymers only. How can one obtain “Compost” from polyethylene if in fact it eventually decomposes to carbon oxides and other gases compounded by the fact that small micro-particulates are left which may be undesirable.

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REVIEWER B

Associate Professor Ignacy Jakubowicz, PhD
R&D Manager Polymer Technology
SP Technical Research Institute of Sweden

Mechanism of oxidation
It seems to be an agreement on the general mechanism of oxidation disregarded some specific expressions.

Oxidation products and their biodegradability
The authors responded: "It is questionable to what extent evidence put forward to support biodegradability can be described as *robust* given that **all studies require extensive accelerated aging** of the oxo-degradable plastic films, either by exposure to ultra-violet light or to heat (50ºC - 70ºC)…"

This statement is not correct firstly because the temperature of a material can reach 50 ºC also in Northern Europe during sunny days (cannot be regarded as "extensive accelerated ageing") and secondly the authors disregard the results showing that a similar level of oxidation can be reached at room temperature (23 ºC) in less than 4 months, Jakubowicz (5).

"it is very difficult to tell how such artificial weathering regimes correlate to actual environmental conditions"

The difficulty to correlate the laboratory tests to various environmental conditions is a general problem which is not particularly connected to oxo-biodegradable materials. Nevertheless, the use of accelerated ageing is widely used and accepted.

Time scale of oxidative breakdown
In this part the authors expose their lack of familiarity with the topic. They estimate the mean annual temperature in the UK to 10 ºC and calculate some kind of ageing time which has no significance at all.

In reality only periods at the highest temperatures are important for the rate of degradation. From May to September the air temperature in UK is probably around 20 ºC but during sunny days the true temperature of a thin film can reach 50 ºC or even more. During winter time when the temperature is low the rate of degradation is insignificant.

The authors even recalculated the ageing time in the paper of Jakubowicz (5) to be about 9 years at 10 ºC despite the fact that there is a result in the paper from the simulated ageing at room temperature that shows the degradation time of less than 4 months.

Biodegradation studies
The authors stated in (ii), page 10 that there is a particular paper by Jakubowicz et al (5) which is claimed to show biodegradation… in soil within two years. This sentence is directly followed by: "This study was financed by P-Life Japan Inc, an oxo-degradable additive manufacturer." It is quite natural to get the impression that the

authors are trying to throw suspicion on the scientific integrity of the study that does not fit their private opinion.

In the last sentence on page 10, once again the authors talk about average annual temperature, the value that has no meaning as biodegradation will not happen at low temperatures neither with natural organic materials nor with synthetic ones. They criticize the test being performed at 23 ºC despite the fact that the most widely used international standard in this field viz. ISO 17556 prescribes the test temperature in the range of 20 – 28 ºC and 25 ºC as the preferable temperature.

Once again the authors use their concept of "annual average temperature" which is meaningless and estimate the ageing time at 10 ºC to be 9 years. The authors seem to have some difficulties to understand the science behind accelerated ageing and lifetime prediction. They seem to miss the basic idea of accelerated ageing which is to speed up the normal aging processes. The abiotic part of the testing procedure is supposed to simulate the usage phase of a product and /or a part of disposal.

Persistence in the soil / Bio-accumulation
The authors point out that oxo-degradable fragments may become ingested by living organisms. This is true, but this apply to all plastic fragments and not only to oxo-degradable.

Recyclability
In the last part on page 15 the authors see a problem for the recyclers about knowing how much stabiliser they should add because of a possible presence of oxo-degradables. However, recycling household plastics itself presents a number of challenges related to collection, sorting and upgrading/restabilization. Consumption of all types of biodegradable plastics accounted for less than 1% of the global demand for plastics in 2009, hydro-biodegradable plastics accounting for 90% of the global consumption of biodegradable plastics. In this context, the presence of small amounts of oxo-degradable materials will not have any significance for how much stabilizers that should be add.

Answers to the four review questions:

Question 1. *In your opinion, has the available scientific information been adequately reviewed, both in the original report and in the research team's reply to the Industry Response? If not, please provide details and explain your reasons.*

No. The authors stand by the original conclusions which are partially incorrect and partially speculative.

For instance: In Annex 2 first sentence – the authors declare that the use of prodegradants does not improve the environmental impact, which is a pure speculation not supported by any scientific evidence.

The first conclusion under "Degradation and biodegradation" is incorrect. The length of time to degradation can be predicted quite accurately. Moreover, using suitable types and amounts of stabilizers and prodegradants the lifetime of a material can be customized.

Another example is the statement “Biodegradation of oxo-degradable plastics, if it occurs at all…” which questions the evidence for biodegradability presented in the comprehensive scientific literature.

Question 2. *Have all the points raised in the Industry Response been adequately addressed by the research team in their reply. If not, please provide details and explain your reasons.*

No. For instance, the authors question the evidence put forward to support biodegradability because “all studies require extensive accelerated ageing…”. Firstly, the authors disregard the results of naturally aged materials (Ojeda, Jakubowicz), secondly the authors do not seem to understand that accelerated ageing is the same as natural ageing achieved in a shorter time.

Another example is from the pages 6-7 where the authors try to show that it will take very long time for oxo-degradable materials to degrade. Unfortunately, they make a mistake assuming “the average outdoor temperature” in their calculations.

The third example is from page 8 where the authors found some lack of information in the scientific papers claiming synergistic effect between oxidation and biodegradation. For this reason the authors consider themselves to be in position to question the conclusion of the scientists that performed the work.

Question 3. *Are the conclusions put forward by the researchers logical and reasonable in the context of the evidence considered. If not, please provide details and explain your reasons.*

Some conclusions are reasonable but some are not and should be changed. Especially when contradictory conclusions are drawn. For instance on page 7 it is concluded that the oxo materials will last for many years while on page 25 under “Re-use” it is stated that bags “are unsuitable for storing items for an extended length of time.”

Question 4. *Are there any other points that you consider important and that you consider have not been adequately covered and assessed in the original report and reply. Please detail and explain.*

Biodegradable plastics have an expanding range of potential applications, and driven by the growing use of plastics and the perception of being environmentally friendly, their use is predicted to increase. However, issues are also emerging regarding the use of all kinds of biodegradable plastics and their potential impacts on the environment and effects on established recycling systems and technologies.

There is a need of an objective and scientific review to identify issues of relevance including various technologies, applications, disposal options, standards, recycling, labeling, education of consumers etc. However, this issues are not only related to oxo-biodegradable materials but to all kinds of biodegradable materials. An objective report should provide information about advantages and limitations of various materials and also identify the areas where more research is needed.

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REVIEWER C

Professor Dr Romeela Mohee, PhD, FRSC
National Research Chair
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Question 1. *In your opinion, has the available scientific information been adequately reviewed, both in the original report and in the research team's reply to the Industry Response? If not, please provide details and explain your reasons.*

1. The research team's reply to the Industry Response to the concern of adequate cross referencing of previous available scientific literature on this topic of research has been addressed. A number of relevant and recent (2008 to 2011) research work published in high-impact factor and specific journals on biodegradation have been revisited by discussing the merits and demerits and other aspects related to the degradation of oxodegradable plastics. The research team has also specified that the procedures of the standards have been followed systematically and that experimental conditions for carrying the analyses on the degradation of the oxo-degradable plastics using standards had been followed. The research team also comments on findings from studies like Jakubowicz (5), Fontanella et al (7), Jakubowicz paper (5), Husarova's paper (8), Chiellini's paper (9) and Corti et al. (10) to either support or compare the results of this study. It is fair to say that the available scientific information has been adequately reviewed by the research team.

2. Whilst in principle, the results of the present work are in fair agreement with the published data, the conditions imposed in these work are far from being encountered in nature, except for some very scarce situations. A thorough reading showed that the actual conditions of testing did not realistically encompass a long time scale which could have yielded a more comprehensive set of degradation and biodegradation behaviour of the OBD-plastics. It is also felt that it would be of interest to assess the preliminary effects of variable heat/temperature conditions on the degradation rates and any enhanced susceptibility to (bio)degradation. At this stage, it is probable that additional strains of bacterial or other microorganisms flora would have to be selected and studied. In this same line of thinking, would there be any biodegradation occurring, it would also be of interest to explore and trace down the fate and mobility of any metabolites of biodegradation into the environment.

3. While considering biodegradability in the scientific literature, the scope could be enlarged by looking at the real meaning or definitions of biodegradability. "Biodegradable" product has the ability to break down, safely, reliably, and relatively quickly, by biological means, into raw materials of nature and

disappear into nature.". This definition is also shared by Patel et al. (2011)[1]. Several different definitions have been published by national and international standardization bodies and organizations[2]. The ISO definition of biodegradable plastics states only a chemical change of the material (e.g., oxidation) by microorganisms; the CEN and DIN standard define biodegradation as the conversion of plastics into microbial metabolic products.

4. I believe that a broader definition of biodegradability is more appropriate to consider for oxodegradable plastics which would give a wider scope in analysing relevant scientific studies and reports.

Question 2. *Have all the points raised in the Industry Response been adequately addressed by the research team in their reply. If not, please provide details and explain your reasons.*

The points raised are grouped as follows:

Mechanism of oxidation

There is no dispute between Defra response and the Industry report. The research team agrees with the industry's explanation of how the oxodegradable plastics degrade but not totally with the fact that the oxidation catalysts used in OBD technology accelerate the normal degradation processes. Furthermore, in the industry report, the various studies cited do not support this point.

Oxidation products and biodegradability

Both Defra reponse and Industry report agree that intermediates are produced but Defra questions the "robust evidence" statement. The Defra response goes on with evidence and examples to show how the results are highly dependent on some controlled conditions, such as heat, light, aging or bacterial inoculation, pre-aging requirements and these cannot be replicated in actual environmental conditions.

My personal view is that studies cited in the industry report are not totally representative of what may or will happen in the real world as there has been no study until now showing that the intermediates after aging degrade completely. It is recommended that the time scale of the research on an uncatalysed and unassisted degradation be widened and deviate from an artificial environment to more naturally plausible ones.

Timescale of oxidative breakdown

The views of the research team are not in agreement with the reasoning suggested by the industry report about the timescales of oxidation. The industry response assumes that since the oxodegradable materials have degraded to a certain extent, then the fragments will disappear, which is not forcibly true. Also, the industry response assumes linear degradation across a whole temperature range which is disputed by the Defra response. The latter analyses the effect of temperature used

[1] Biodegradable Polymers: an Ecofriendly Approach in Newer Millenium : Parth N. Patel, Khushboo G. Parmar, Alpesh N Nakum, Mitul N Patel, Palak R Patel, Vanita R Patel, Dhrubo Jyoti Sen. Asian Journal of Biomedical and Pharmaceutical Sciences 1 (3) 2011, 01-17

[2] http://www.wiley-vch.de/books/biopoly/pdf_v10/vol10_19.pdf

to assess degradation and the use of the Arrhenius equation to predict levels of degradation at different temperatures.

In my personal view, the changes in degradation rates are not linear across the whole scale of temperature. There are thresholds of activity at 25°C, 37°C, 45°C and 60°C and although the Arrhenius equations are valid at these temperature, they cannot be used outside the range of temperatures of 25°C to 60°C and definitely not in the UK environment where there are seasons where temperatures are as low as zero to 10°C and degradation by bacteria can be very limited. For example in composting, literature has shown very low degradation rates at very low temperatures for example less than 20°c and at very high temperatures, my own personal work has shown that composting can be shut down as these bacteria cannot work beyond 70°C.

Furthermore, there is no evidence nor any studies which clearly show that the oxo materials can be degraded to the point of total embrittlement at ambient temperatures in periods of less than one year, as indicated in the industry response. It is true that the timescale will depend on the amount of antioxidant, but the latter is a control parameter of the industry, which should bear the responsibility of providing to the scientific community, data/studies showing the correlation between amount of oxidant used and biodegradation rates.

Biodegradation studies
The industry response states that Defra has been wrong in saying that biodegradability and compostability are the same thing. Although Defra has not actually said that, it has implied so by using standards pertaining to composting environments to assess the degradability of the oxodegradable materials, which is a bit unfair.

However, the point of Defra is that oxodegradable plastics can be perceived and mistaken to be compostable materials and while entering the composting chain, they might have a residual effect on the final compost quality. Further studies have to be done to assess the effect of OBD composting and the quality of the final product.
Defra has also responded to the study of mineralisation of OBD plastics in soil 90% after two years by stating that this study was done at 23°C and the industry has not given rates for typical annual temperatures encountered in the UK and this point is very valid.

Compostable and Oxodegradable materials cannot be compared as the mechanisms of degradation are not similar and even the end results are not similar. Composting would refer to the production of a material that should be applied to land and be beneficial to crops etc whereas for oxodegradable materials, the challenge is for the materials to degrade under natural conditions without having any significant impact. My research has shown that certain types of degradable plastic remain brittle in the composting environment for several months and can have a negative impact if the compost if applied to seedling applications.

I believe that the real objective is to ensure that the oxodegradable materials do not have any harmful effect on the environment either as pieces or fragments but they should not be assessed as per same standards as for compostable materials. The latter require the analysis of the final product especially when the compost is used in agricultural applications or food crops. The OBD biodegradability standards should not be as stringent as compost standards.

Pathways other than composting should be used to assess the impact of the OBD and appropriate standards should be developed to assess the material, taking into account the sequential mechanism of oxidation followed by degradation.
However, it is agreed that if the oxodegradable material were to degrade in a composting environment, then it would be an added plus as it could then together with the other compostable plastics, enter the composting stream especially if these composting facilities exist in the country.

Also, biodegradation does not involve only aerobic conditions, it is wrong to assume that a material that cannot be composted cannot biodegrade. My own research has shown degradability of plastics in a composting environment and in an anaerobic environment as well.
Again the Defra response that the results presented by industry are lab scale controlled results where the samples have undergone pre-aging, which may not necessarily reflect actual onsite conditions, is valid.

Toxicity
There is no dispute between industry and research team about the biodegradability of the additives and there is no evidence on ectotoxicity in the studies available until now.

Persistence in soils
The industry response states that there is no evidence of oxodegradable materials persistent in the soils, however there is no counterevidence as well that they will not be present in the environment and for how long they might stay there, This has been indicated in one of the studies cited by the research team, the Feuillofey study where fragments have been seen in soil where oxodegraddable plastics mulch had been applied, indicating the possibility of cross linking.

Plastics in general when they degrade become brittle and get fragmented to small pieces that can indeed enter watercourses, pollute marine environment and get ingested by animals. Unless oxodegradable plastics can show that they disappear completely under natural environmental conditions, it is difficult to assess the disposal impact and the risks of them staying around in the environment. Furthermore, the issue of crosslinkage must be further researched upon and addressed as these crosslinked intermediates might not appear in laboratory experiments or in air ovens, etc but still be present.

Recyclability
The Defra response comments on the same study cited by the industry response, with a different conclusion that oxodegradable materials are capable of causing deterioration in the production of recycled materials.

Defra response is right in saying that the safest solution is to keep oxodegradable materials out of the mainstream plastics recycling process. To my opinion, the issue of whether the oxo-degradable plastics can enter the normal well-established recycled chain and have a recycled product which is comparable to the current recycled products is very important. A UK study by Grenier et al. (2007) [3] which evaluated two brands of oxo-degradable and hydro-degradable bags, indicated that neither type of bag are perfectly compatible with the traditional plastic bag recycling stream. The oxo bags proved more compatible with the recycling process “from the viewpoint of the preparation of mixtures as well as the extrusion of profiles and films”. One of the recycled content bags made from recycled oxo bags performed fairly well in accelerated aging tests, but the[4] second brand did not.

It has taken many years and efforts for the recycling of wastes materials, including plastics to be established and to be successful, moreso to be accepted by the consumer. It would be disastrous at this point in time for a recycled material to fail in quality or strength due to the addition of the oxo additive and destroy the reputation of the industry. The risks of allowing OBD materials to enter the current recycling streams far outweigh the benefits.

As it is not known yet up to what extent, an oxo degradable plastic can weaken the recycled product and jeopardize its use, especially when it is to be reused as other plastics such as PET, a completely separate industry should be developed for the OBD material if ever it is to be recycled. Oxo should not appear in the chain as the sorting mechanism of the current recycling industry might not be able to handle it effectively.

Finally, the whole objective of the OBD plastics is to render it safe for disposal so the issue of it getting into the recycling industry should not arise or be entertained.

Question 3. *Are the conclusions put forward by the researchers logical and reasonable in the context of the evidence considered. If not, please provide details and explain your reasons.*

Yes, the conclusions put forward by the researchers seem to be logical and reasonable for most of the points except for the assessment of biodegradability using composting standards or biodegradability under specific conditions.

Composting is an engineered aerobic process employed presently as sustainable technology to stabilize a wide variety of organic wastes/substrates. To claim a certain “compostability” of OBD-plastics would be only possible if the OBD-plastics are “artificially” treated as one feedstock in a specific proportion in a larger mix of other organic and far more easily biodegradable matter. Having said so, the research team seemingly missed a key step in analysing the degradation behaviour OBD-plastics in true composting environments. This lack of data on the degradation behaviour of OBD-plastics in a composting environment has induced hasty inferences on their overall biodegradability. O’Brine et al. (2010) investigated the breakdown of two oxo-biodegradable plastics, compostable plastic and standard polyethylene in the marine environment. Tensile strength of all materials decreased during exposure, but at different rates. Compostable plastic disappeared from a test

[3] Grenier D. and Cote L. 2007. Evaluation of the impact of Biodegradable bags on the recycling of traditional plastic bags. California state.

O’Brine, T., Thompson, R.C. 2010. Degradation of plastic carrier bags in the marine environment. Marine Pollution Bulletin, 60, 2279–2283.

rig between 16 and 24 weeks whereas approximately 98% of the other plastics remained after 40 weeks.

Also, when relating to biodegradation, it cannot be only restricted to an aerobic set of environmental conditions. Composting being an inherently aerobic process, to put forward a persistent argument of biodegradability is inadequate. The study should be more clearly defined in terms of its scenarios of environmental conditions and testing. It would be more complete, in my opinion, to at least divide the tests under the following three sets: aerobic degradability conditions, anaerobic degradation conditions and compostability. It may be uncommon to the understanding and knowledge of some but plastics may also be wastes piling up in semi-aerobic and/or completely anaerobic conditions. The degradation behaviour under these conditions cannot be ignored and left unassessed. Hence, biodegradation of OBD-plastics should also be measured by anaerobic microbes.

It is also important to clearly understand the mechanism of biodegradation of OBD-plastics, in order to select the most appropriate standards that will accurately determine the biodegradability and impacts of these plastics on the environment.
As explained in previous sections, using ASTM 6400 and EN13432 standards to assess the biodegradability would limit the scope of the study as it relates to specific composting parameters to enhance degradation.

The ASTM Standards and Methods employed by EPI[5] to test degradable plastics (2007) give a brief description of oxo-biodegradable plastics degradation mechanism. Initially, there is an abiotic/chemical stage that begins with the cleavage of carbon chain backbone in PE, PP or PS into shorter chain molecules and the incorporation of oxygen into these molecules as organic functional groups. The hydrocarbon polymers change their behaviour from hydrophobic to hydrophilic thereby allowing the fragmented polymer to absorb water. The biodegradation process then occurs through a microbiological stage in which biodegradation of the fragmented polymer takes place. During this process, the molecular weight of the polymer which has been reduced during the first stage to lower molecular weight, will allow bacteria, fungi and algae to consume the carbon backbone fragments into their tropic process. The end products are CO_2, H_2O and a small quantity of minerals and biomass with no toxic of harmful residues to soil, plants and macro-organisms.
The American Society for Testing and Materials (ASTM standards) recommends the oxo-biodegradable technology in ASTM D 6954-04-"*Exposing & Testing Plastics that Degrade in the Environment by a Combination of Oxidation & Biodegradation*", which has been specifically developed for plastics that degrade initially by an oxidative process and finally biodegrade. ASTM D 6954-04 references ASTM D5510-Standard practice for heat ageing of oxidatively degradable plastics. This method defines the exposure conditions of plastics at various temperatures when exposed solely to heat conditions. After heat exposure, samples will be tested based on this method which replicates the conditions in landfills and composting. It also references ASTM D5208- Standard practice for operating fluorescent ultraviolet (UV) and condensation apparatus for exposure of photodegradable plastics, which is used to determine the usable life based on UV degradation. These two test procedure in combination make up the 1-tier of the testing process for verifying whether OXO

[5] http://www.epi-global.com/files/brochures/1258559621ASTM01091118EPI.pdf

plastic is degradable[6]. These standards would indeed be much more appropriate to use in the case of OBD.

In addition, while testing biodegradability under anaerobic conditions, the most accurate standard test method for anaerobic environments is the ASTM D5511 - 02 Standard Test Method for Determining Anaerobic Biodegradation of Plastic Materials under High-Solids Anaerobic-Digestion Conditions. Another standard test method for testing in anaerobic environments is the ASTM D5526 - 94(2002) Standard Test Method for Determining Anaerobic Biodegradation of Plastic Materials under Accelerated Landfill Conditions, although this test has proven extremely difficult to perform. Both of these tests are used for the ISO DIS 15985 on determining anaerobic biodegradation of plastic materials.

Question 3. *Are there any other points that you consider important and that you consider have not been adequately covered and assessed in the original report and reply. Please detail and explain.*

To assess the environmental impacts of the oxo-biodegradable plastics, a cradle-to-grave analysis of the OBP-plastics need to be carried out from raw material extraction to final disposal. The environmental impacts should include climate change, resources, ecosystem quality and human health. The inventory analysis should quantity all inputs, for instance, materials/resources used, energy consumption, and all outputs in terms of air and water emissions, solid wastes, by-products, involved in the manufacture and final disposal or treatment of the OBP-plastics. The environmental impacts could be determined for different end-use scenarios, for instance, landfilling, composting, incineration and recycling.

For example, a study carried out by Harding et al. (2007)[7] showed that the dominant contributions to the environmental burden in the production of PHB are the large requirement for energy, in particular steam, as well as the high water requirement (65 dm^3 per kg polymer). The use of fertiliser (from agricultural processes), acids and a significant number of salts, adds to the toxicity levels of wastewater and the eutrophication potential. Despite this, the production of polyhydroxy--butyrate is more beneficial in a full cradle-to-gate life cycle assessment study than polypropylene (PP) production.

Similar studies can be conducted for OBD plastics to identify the real environmental impacts of the oxodegradable materials across the whole chain. Looking only at the environmental impacts for disposal of the product is not enough. Although oxo degradable plastics can be regarded as more sustainable than ordinary plastics for not staying many years in the environment, the manufacturing of the oxoplastics, furthermore the additive still require petroleum, energy and other resources.

Therefore, the degradation and/or biodegradation of OBD-plastics needs a thorough life cycle analysis/life cycle assessment (LCA) spanning at least 2 to 3 years of

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[7] K.G. Harding, J.S. Dennis, H. von Blottnitz, S.T.L. Harrison. 2007. Environmental analysis of plastic production processes: Comparing petroleum-based polypropylene and polyethylene with biologically-based poly--hydroxybutyric acid using life cycle analysis. Journal of Biotechnology 130, 57–66.

detailed monitoring and tracing down the interactions of the most prevalent environmental conditions. It is also suggested to take on board such LCA work a number of most plausible/actually prevalent environmental scenarios.

Question 4. *A short summary of your overall assessment of the reply and your conclusions and comments back to Defra.*

My analysis of the reports has shown that despite a comprehensive set of degradation and biodegradation studies were verified and used to compare the results in this specific work, it is still not correct to advocate that these results are complete. This is because the present work has not addressed the crux of the degradability behaviour of OBD-plastics in a true environmental framework. It is totally wrong to have assumed mimicry of lab-based results which are promising to be a more or less, if not almost similar, to expected prevailing natural conditions to which the OBD-plastics would be exposed. Also, I consent on that oxidation catalysts used in OBD technology have not yet proven to accelerate the normal degradation processes. It is hence *recommended that the time scale of the research on an uncatalysed and unassisted degradation be widened and deviated from an artificial environment to more naturally plausible ones.*

To this end, it is recommended that extensive field studies be carried out to exhaust the most plausible environmental conditions to which the OBD-plastics may be exposed. All the more, it is important to clearly understand the mechanism of biodegradation of OBD-plastics, in order to select the most appropriate standards that will accurately determine the biodegradability and impact of these plastics on the environment. The choice of one or more best suited standards to be used to assess the several types of biodegradability of the OBD-plastics seemed not to be clear in the original report. Finally, it is strongly felt that the degradation and/or biodegradation of OBD-plastics needs a thorough life cycle analysis/life cycle assessment (LCA) of detailed monitoring and tracing down the interactions of the most prevalent environmental conditions.

Also, if the OBD were to enter a landfill which is one of the commonest means of disposal of wastes, the merits of the plastic will not be fully met as in the deep layers of the landfill, the OBD plastics behave as ordinary plastics as they are totally inert in the absence of oxygen or light which further warrant the need for further research into the several biodegradability pathways.

Finally, if the above studies show full compliance with the standards and no negative impact on important environmental categories, as per typical LCA studies, Oxo-biodegradable plastics can prove to be promising and will encourage the use of plastics in its numerous forms in several applications without worrying about polluting the environment OBD could be a better solution instead of banning plastic.

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