

Microbial upcycling of polyethylene terephthalate wastes into value-added products

Abstract

This mini-review discusses the latest developments in microbial upcycling of PET wastes into high-value products. PET is one of the major types of synthetic plastics that include PE, PP, PS and PVC. PET finds widespread use in making water, soda and beverage bottles and diverse food packaging materials; more than 82 million tons of PET wastes are generated globally per year. PET is highly recalcitrant and massive amounts of PET wastes especially water bottles accumulate and contaminate the environmental sectors including soil, freshwater, and marine ecosystems.

Urgent initiatives are, therefore, warranted for safe disposal, recycling and upcycling of PET wastes. Of late, several reports have demonstrated successful upcycling of PET wastes into value-added products such as adipic acid, gluconic acid, lycopene, paracetamol, and vanillin etc. Microbial bio-upcycling integrates chemical or enzymatic pretreatment of PET wastes with microbial conversion of the building blocks into high-value end-products via natural or engineered microbes e.g. *E. coli*, *Gluconobacter oxydans*, *Ideonellasakaiensis*, *Pseudomonas putida*, and *Rhodococcus jostii* etc.

Further research efforts must now be directed towards scaling up of the production of high-value chemicals at commercial scale from PET wastes, exploring the feasibility of similar upcycling processes for other plastic wastes, and ushering a circular, secure, and sustainable economy of synthetic plastics without further depletion of fossil fuel reserves; continued aggravation of climate change; and further deterioration of Earth's ecosystems.

Keywords: polyethylene terephthalate, upcycling, adipic acid, vanillin, paracetamol, lycopene, *ideonella sakaiensis*, engineered microbes, circular economy

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Introduction

Plastics come in various kinds including polyethylene (PE), polypropylene (PP), Polystyrene (PS), polyvinyl chloride (PVC) and polyethylene terephthalate (PET). PET is the fourth most produced plastic and it constitutes about 10-12% of global plastic production. It finds widespread use in making water, soda, and beverage bottles, and diverse food packaging materials. More than 82 million tons of PET are generated globally per year (National Renewable Energy Laboratory, NREL [now renamed National Laboratory of the Rockies, NLR], 2021). After customer use, most of this PET e.g. plastic water bottles accumulate and pollute the environment. PET is quite recalcitrant, i.e. it has rather low degradability, and its uncontrolled disposal threatens several ecosystems. Several scientific groups have been investigating, over the past few decades, into strategies of how to valorize plastic wastes.

Several recent publications indicate that the several proof-of-concept breakthroughs have been found regarding upcycling of PET wastes into valuable products. Some of these products include valuable products e.g. lycopene, nylon precursors, vanillin, paracetamol, and bioplastic precursors. These value-added products were obtained by integrating chemical or enzymatic pretreatment with microbial bioconversion of PET breakdown products into high-value chemicals via engineered or natural microbes.

Upcycling of PET wastes

To combat the accumulation of post-consumer PET wastes, bio-upcycling, which integrates chemical (or enzymatic) pretreatment with microbial bioconversion has shown promise in recent times. Microbial upcycling involves pretreatment of plastic waste,

depolymerization to generate monomers and oligomers under mild conditions, bio-upcycling in which the resulting small molecules are fed to engineered or natural microbes to use them as "living factories" to convert the feedstocks into high-value compounds leading to product recovery, whereby the valuable products such as PHAs, vanillin, or adipic acid are purified. In this mini-review, let's look at some recent initiatives to transform PET wastes (for example, waste water bottles) into valorized products such as lycopene, vanillin, paracetamol, adipic acid, and gluconic acid.

Lycopene production

Diao et al.,¹ showed that PET can be converted to lycopene in a 2-step process. The first is an alkaline hydrolysis stage that converts PET into terephthalic acid (TPA) and ethylene glycol (EG); alternatively, an enzyme PETase, from the PET degrading bacterium discovered in Japan, *Ideonella sakaiensis*, may be used. The second is a bio-upcycling process. This involves a genetically engineered bacterium-*Rhodococcus jostii* strain RPET (RPET).

RPET has a natural predilection for utilizing PET products as sole carbon and energy sources. But the PET breakdown products can be channeled from the central metabolic pathways e.g. TCA cycle into the methyl erythritol phosphate (MEP) pathway that leads to the formation of lycopene. This research group has genetically engineered strain RPET by adding a gene that optimizes channeling of products into the MEP pathway. They have demonstrated that production of lycopene has been enhanced nearly 500 times by using the GEM strain as compared to the wild-type strain. Lycopene is a valuable product that finds applications as a colorant and as an antioxidant in pharmaceutical and food industries. Diao et al.,² reported that

maximum lycopene yield was about 1,300 mg/L in the fermentation broth.

Similarly, other valuable products such as paracetamol, vanillin, bacterial nanocellulose (BNC), polyhydroxybutyrate (PHB), adipic acid and gluconic acid have been obtained by bio-upcycling of PET wastes.

Vanillin production

Scientists at the University of Edinburgh³ achieved conversion of poly (ethylene terephthalate) into a value-added product, vanillin. This product finds numerous applications as additives in foods, cosmetics and perfumes. First, PET is broken down into TPA and EG by using the enzyme called LCC (leaf-branch compost cutinase) or polyethylene terephthalate hydrolase (PETase) obtained from *Ideonella sakaiensis*. Then the TPA is bio-upcycled using a genetically engineered strain of *E. coli*. The engineered organism performs a series of reactions such as oxidation, methylation, and reduction to bio transform TPA into vanillin.

The most attractive aspect of such bio-upcycling of plastic wastes such as PET is that it achieves a circular economy. Usually, the plastic production process is a linear economy in which fossil fuels are used to make plastics which consumers use in the form of a variety of plastic products. For example, PET is used on a large scale as water (and beverage) bottles and packaging materials. But post-consumer use, the PET products end up in landfills, oceans, or other water bodies. Bio-upcycling enables a circular economy in which the fossil fuels are used to make plastics e.g. PET, consumer products are generated e.g. water bottles, and the wastes are now valorized into useful products e.g. vanillin.

Vanillin production is done at mild conditions, i.e. moderate temperature, near neutral pH, low pressure, free of harsh reagents and is almost carbon-neutral. This ensures sustainable circular economy of plastic bottles. This allows reduction in the use of fossil fuels as the end products may be upcycled to produce useful products without consuming more fossil fuels. Also, as the upcycling process is either carbon-neutral or has a low carbon-footprint, it enhances mitigation of the global climate change.

Paracetamol production

Recently, the research group of Stephen Wallace at the University of Edinburgh⁴ converted PET into paracetamol (also known as acetaminophen) by using a genetically engineered *E. coli*. The process underwent several stages. First, PET is degraded into its building blocks, TPA and EG. This was achieved through alkaline hydrolysis or via enzymes such as PETase from *Ideonella sakaiensis*. Next, they used the reprogrammed *E. coli* as “living factories” to generate acetaminophen. The bacteria were engineered to block the synthesis of para-amino benzoic acid (PABA), necessary for DNA synthesis, compelling them to use the plastic-derived products e.g. TPA.

This process also takes place under mild conditions (room temperature and neutral pH). The Lossen rearrangement needed to convert TPA into PABA (a precursor of paracetamol) usually takes place in the test tube. But these Edinburgh scientists for the first time demonstrated that this rearrangement can also take place inside the living cells. Genes from mushrooms and other bacteria had been added to the genetically modified *E. coli* to enable them to biotransform PABA into paracetamol. The yield was also quite high, about 92%, i.e. 92% of TPA was converted into paracetamol. The group is now working with AstraZeneca to scale up and optimize the process so that production of paracetamol may be achieved at the commercial level.⁵

Paracetamol production is currently dependent on dwindling fossil fuels. But the Edinburgh group has created an engineered *E. coli* that convert the building blocks obtained from breakdown of plastic bottles into a painkiller in less than a day. On a rough calculation, it was found that one 1-litre PET bottle could roughly produce about nine 500 mg paracetamol tablets. As the process needs no extreme heat or pressure, it produces only a small fraction of the carbon footprint as compared to the traditional process involving fossil fuels. Therefore, microbial upcycling is either carbon-neutral or leaves a much lower carbon footprint.

Adipic acid production

The Wallace group also earlier achieved the bio-upcycling of PET into adipic acid (a nylon precursor).⁶ This value-added product would otherwise have been manufactured from diminishing fossil fuel reserves. They used a genetically engineered *E. coli* strain. First, PET is transformed enzymatically into TPA and EG. The TPA is then converted by engineered *E. coli* into adipic acid. Adipic acid can then react with other chemicals like diamines to produce nylon 6, 6, which can then be used to make many valuable products such as nylon fibers, textiles, and packaging materials.

Werner et al.,⁷ produced beta-keto adipic acid (bKA) from bis (2-hydroxyethyl) terephthalate (BHET), a breakdown product of PET, by using an engineered *Pseudomonas putida* strain KT2440.

Gluconic acid production

Kim et al.,⁵ reported the bioconversion of PET-derived ethylene glycol (EG) into glycolic acid by using *Gluconobacter oxydans*.

Attempts are currently underway to bio-upcycle PET into other value-added products such as polyhydroxyalkanoates, gallic acid, muconic acid, catechol and hydrocarbons (for generating biofuels).

Conclusion

A multiplicity of chemicals may be produced by bio-upcycling of waste PET.² These include flavor compounds such as vanillin, textile components e.g., nylon, food additives such as lycopene, pharmaceuticals e.g. paracetamol, and cosmetic ingredients such as gluconic acid. PET also may be upcycled to PHAs (bioplastic components), gallic acid, muconic acid, and catechol (pharmaceutical intermediates), and hydrocarbons (for biofuel generation).

Already, high yield production of lycopene, paracetamol, and vanillin have been shown at the laboratory scale. The next challenge is to scale up these processes so that these high-value products may be generated at the commercial scale. There are now prospects for microbial upcycling of PET and other plastics into value-added chemicals to usher a cyclic, sustainable, and secure economy without further depletion of fossil fuel reserves and without further aggravating the worsening global climate change scenario.

The recent reports open a window onto a bright future where the boundary between ‘waste’ and ‘resource’ gets blurred and where healing the human body would still be compatible with healing the planet.

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None

Conflicts of interest

The author declares that there are no conflicts of interest.

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