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3D Printing's Green Revolution: A Review of Usage of Sustainable and Recycled Polymers

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Abstract

The convergence of sustainability concerns and additive manufacturing technologies has sparked significant interest in the use of recycled plastics for 3D printing. This review paper examines the current state of research and practice in this rapidly evolving field. It explores the processes involved in recycling various types of plastics for 3D printing applications, with a focus on commonly used materials such as PLA and ABS. The review finds that while it is feasible to use recycled plastics in 3D printing, particularly in fused deposition modelling (FDM), the quality and consistency of the resulting products can vary significantly. Recycled PLA tends to show some degradation in mechanical properties, while recycled ABS generally maintains its properties better through the recycling process. Interestingly, recycled materials often produce smoother surface finishes in 3D printed objects. The paper also highlights the need for further research in areas such as standardization of recycling processes for 3D printing, long-term performance studies of products made from recycled materials, and life cycle assessments comparing recycled and virgin plastics in 3D printing contexts. Overall, this review underscores the potential of using recycled plastics in 3D printing as a significant step towards more sustainable manufacturing practices.

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Introduction

Sustainability, broadly defined as meeting the needs of the present without compromising the ability of future generations to meet their own needs, has become a critical concern across all sectors of society and industry. In the realm of polymers and plastics, sustainability takes on particular urgency due to the ubiquity of these materials in modern life and their significant environmental impact. The concept of sustainability in polymers encompasses

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not only the sourcing of raw materials but also the entire lifecycle of plastic products, including their production, use, and end-of-life management.

The importance of sustainability in the polymer industry cannot be overstated. Plastics, while invaluable for their versatility and durability, have become a major environmental concern due to their persistence in ecosystems, contribution to greenhouse gas emissions, and dependence on finite fossil fuel resources. The need for sustainable practices in plastic production and use is driven by several factors, including the depletion of natural resources, the accumulation of plastic waste in landfills and oceans, and the growing public awareness of environmental issues.

To address these challenges, two main approaches have emerged: the development of sustainable virgin polymers and the recycling of existing plastics. Sustainable virgin polymers, such as bioplastics derived from renewable resources, offer the potential to reduce dependence on fossil fuels and decrease carbon emissions. On the other hand, recycling existing plastics provides a way to minimize waste and conserve resources by keeping materials in use for longer periods.

Concurrent with the push for sustainability in plastics, the field of 3D printing, also known as additive manufacturing, has undergone rapid evolution and growth. From its origins in the 1980s as a prototyping technology, 3D printing has developed into a versatile manufacturing method capable of producing complex, customized parts across a wide range of industries. The importance of 3D printing continues to grow, with applications expanding in fields such as aerospace, automotive, healthcare, and consumer goods.

As both sustainability and 3D printing have gained prominence, there has been increasing research interest in the use of sustainable plastics, including recycled materials, for 3D printing applications. This intersection of sustainability and additive manufacturing offers the potential to address environmental concerns while leveraging the benefits of 3D printing technology. However, while numerous studies have explored various aspects of using sustainable and recycled plastics in 3D printing, there has been limited effort to collate and synthesize this body of research.

A comprehensive review of the existing research on 3D printing with recycled plastics would be invaluable for several reasons. It would provide a clear overview of the current state of the field, highlight successful approaches and persistent challenges, and identify promising directions for future research. Such a review could serve as a resource for researchers, engineers, and policymakers working to advance sustainable manufacturing practices.

This gap in the literature presents an opportunity for a systematic review that brings together the diverse strands of research on 3D printing with recycled plastics. The present paper aims to address this need by providing a comprehensive overview of the field, synthesizing findings from various studies, and identifying key trends and challenges. It aims to answer the following research questions:

- RQ1: What is the current state of research and practice in the use of sustainable and recycled plastics for 3D printing?
- RQ2: What are the key challenges and opportunities in this field?

Sustainable Polymers

Sustainable polymers (SPs) represent a significant advancement in materials science, offering a promising solution to the environmental challenges posed by conventional plastics. These polymers are designed to minimize environmental impact throughout their lifecycle, being derived from renewable resources, biodegradable, or both1. The concept of sustainability in polymer science encompasses not only the source of raw materials but also environmentally friendly manufacturing processes, maintained or improved performance compared to traditional polymers, and environmentally responsible end-of-life options.

The evolution of sustainable polymers has been marked by significant milestones over the past few decades. Initially, the focus was on developing biodegradable polymers such as polylactic acid (PLA) and polyhydroxyalkanoates (PHAs) in the 1980s and 1990s. This was followed by an emphasis on bio-based polymers from renewable resources in the 2000s and 2010s, including bio-polyethylene and bio-PET. The current generation of sustainable polymers, emerging in the 2010s, is characterized by the integration of advanced functionalities and improved performance, such as self-healing and shape-memory properties.[1] Recent discoveries in the field include the development of lignin-based polymers with enhanced mechanical properties, the synthesis of fully recyclable plastics like poly(diketoenamine)s (PDKs), and the creation of bioinspired materials mimicking natural polymers like spider silk.

Among the various sustainable polymers, polylactic acid (PLA) has emerged as one of the most widely used, particularly in 3D printing applications. PLA is a thermoplastic polyester derived from renewable resources such as corn starch, sugarcane, or tapioca roots. It offers a unique combination of biodegradability, biocompatibility, and processability, making it suitable for a wide range of applications. PLA's properties, including a glass transition temperature of 60-65°C, melting temperature of 130-180°C, and tensile modulus of 2.7-16 GPa, contribute to its versatility.[2] The biodegradability of PLA is particularly noteworthy, as it can be composted under industrial conditions, breaking down into carbon dioxide and water. This characteristic addresses the growing concern of plastic pollution, offering a more environmentally friendly alternative to traditional petroleum-based plastics.

Another important class of sustainable polymers is polyhydroxyalkanoates (PHAs). PHAs are aliphatic polyesters produced by bacteria under growth-limiting conditions. These polymers are fully biodegradable and can be produced from various renewable carbon sources, including organic wastes. PHAs can be classified based on their chain length into short-chain-length (SCL), medium-chain-length (MCL), and long-chain-length (LCL) varieties. The most well-known PHAs include poly(3-hydroxybutyrate) (PHB) and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). The physical properties of PHAs can vary widely depending on their composition, with SCL-PHAs typically being more crystalline and rigid, while MCL-PHAs are more elastic and flexible. This versatility allows PHAs to be tailored for various applications, from rigid packaging materials to flexible biomedical devices.[1]

Polybutylene succinate (PBS) is another promising sustainable polymer that has gained attention in recent years. PBS is a semi-crystalline polymer with properties comparable to polyethylene terephthalate (PET). It can be synthesized through the polycondensation of succinic acid and 1,4-butanediol, both of which can be derived from renewable resources. PBS offers a good balance of mechanical properties, including high elongation at break and flexibility. Its biodegradability in various environments, including soil and compost, makes it an attractive option for applications where end-of-life disposal is a concern. Recent research has focused on improving PBS's properties through copolymerization and blending with other biodegradable polymers.[1]

Sustainable polymers offer several advantages over their petroleum-based counterparts, making them superior in many aspects. They contribute to a reduced carbon footprint due to lower greenhouse gas emissions during production and use. By utilizing renewable feedstocks, SPs help conserve fossil resources. Their biodegradability mitigates plastic pollution in the environment, addressing one of the most pressing ecological issues of our time. Moreover, SPs often exhibit reduced toxicity with fewer harmful additives and byproducts. Their improved end-of-life options, including enhanced recyclability or compostability, further contribute to their environmental benefits. Additionally, the unique characteristics derived from natural building blocks offer potential for novel properties, expanding the range of possible applications.[3]

The emerging use of sustainable polymers in 3D printing technologies has opened up new possibilities for eco-friendly manufacturing. This intersection has led to reduced material waste through additive manufacturing processes, which minimize excess material use. The ability to create complex, personalized products with minimal tooling has enhanced customization capabilities. Localized production through 3D printing with SPs has the potential to reduce transportation emissions, contributing to a more sustainable supply chain. In the biomedical field, the development of biocompatible and biodegradable implants using 3D-printed sustainable polymers is particularly promising. Furthermore, the use of SPs in rapid prototyping has accelerated product development while maintaining a focus on sustainability.[4]

In the context of 3D printing, PLA has become the most commonly used material in fused deposition modeling (FDM) due to its ease of processing and good printability. However, the mechanical properties of 3D printed PLA parts can be influenced by various factors, including printing orientation, layer height, raster angle, and infill density. Research has shown that parts printed in X and Y orientations generally yield higher tensile strengths compared to Z orientation, while lower layer heights and higher infill densities typically result in stronger parts.[5] These insights are crucial for optimizing the performance of 3D printed sustainable polymer parts.

Despite the numerous advantages of sustainable polymers, challenges remain in their widespread adoption. These include higher production costs compared to petroleum-based polymers, some performance limitations, and the need for improved recycling infrastructure. For instance, while PLA is compostable under industrial conditions, it requires specific temperature and humidity conditions that are not typically found in home composting systems or natural environments. This highlights the need for appropriate end-of-life management strategies for sustainable polymers.[1]

As research in sustainable polymers continues to advance, their integration with 3D printing technologies promises to revolutionize manufacturing processes, offering a more environmentally friendly approach to product development and production. Future research directions include improving the mechanical properties of sustainable polymers for 3D printing, optimizing printing parameters, developing new sustainable polymer formulations specifically designed for additive manufacturing, and enhancing recyclability and closed-loop processes. The ongoing development of sustainable polymers and their applications in 3D printing represents a significant step towards more sustainable and environmentally responsible manufacturing practices.

3D Printing with Virgin Sustainable Polymers

Three-dimensional (3D) printing with virgin sustainable polymers has emerged as a promising approach to eco-friendly manufacturing. This additive manufacturing technique allows for the creation of complex, customized objects while minimizing material waste. Among the various sustainable polymers used in 3D printing, polylactic acid (PLA), polyhydroxyalkanoates (PHAs), and polybutylene succinate (PBS) have gained significant attention due to their biodegradability and renewable sourcing.

PLA is the most widely used sustainable polymer in 3D printing, particularly in fused deposition modeling (FDM). In FDM, PLA filaments are heated and extruded through a nozzle, depositing layer upon layer to form the desired object. The process parameters, such as nozzle temperature, printing speed, and layer height, significantly influence the final product's properties. For optimal results, PLA is typically printed at temperatures between 180-220°C, with a layer height of 0.1-0.3 mm.[4] The orientation of the printed part also plays a crucial role in determining its mechanical properties. Parts printed in X and Y orientations generally exhibit higher tensile strengths compared to those printed in the Z orientation, due to the alignment of the deposited layers with the direction of applied stress.[5] While PLA offers excellent printability, it faces challenges such as warping and poor layer adhesion, which can affect the final product's quality and strength. To mitigate these issues, researchers have explored various strategies, including optimizing printing parameters and post-processing techniques. For instance, using a heated build plate (around 60°C) can help reduce warping, while careful control of cooling rates can improve layer adhesion. Additionally, post-processing methods such as annealing have been shown to enhance the crystallinity and mechanical properties of 3D printed PLA parts.[2]

PHAs, another class of sustainable polymers, have also been explored for 3D printing applications. However, their use in FDM has been limited due to their high flexibility, which presents challenges during printing. PHAs typically require lower printing temperatures compared to PLA, usually in the range of 160-180°C. The mechanical properties of 3D printed PHA parts are highly dependent on the specific type of PHA used (e.g., short-chain vs. medium-chain) and the printing parameters. One of the main challenges in 3D printing with PHAs is achieving consistent extrusion due to their thermal sensitivity. To address this, researchers have investigated blending PHAs with other polymers or additives to improve their printability and mechanical properties.[1]

PBS, while less commonly used in 3D printing compared to PLA and PHAs, has shown promise due to its good thermal stability and mechanical properties. PBS can be processed using FDM techniques, typically at temperatures between 190-220°C. One of the advantages of PBS in 3D printing is its lower shrinkage compared to PLA, which can lead to better dimensional accuracy of printed parts. However, challenges remain in terms of optimizing printing parameters to achieve the desired balance of mechanical properties and print quality. Research has focused on improving the adhesion between layers and enhancing the crystallization behavior of PBS during the printing process to overcome these challenges.[1]

A common challenge across all these sustainable polymers in 3D printing is achieving mechanical properties comparable to those of conventional, petroleum-based plastics. To address this, various approaches have been explored, including the use of fiber reinforcements and the development of polymer blends. For instance, incorporating carbon fibers into PLA has been shown to significantly enhance the tensile strength and Young's modulus of 3D printed parts.[3] Similarly, blending different types of sustainable polymers or adding plasticizers can help tailor the properties of the printed objects to meet specific application requirements.

Another important consideration in 3D printing with sustainable polymers is the potential for material degradation during the printing process. Repeated heating and cooling cycles can lead to changes in molecular weight and thermal properties, potentially affecting the biodegradability and mechanical performance of the printed parts. To mitigate this, careful control of processing conditions and the use of stabilizers or antioxidants have been investigated.[4]

As research in 3D printing with virgin sustainable polymers continues to advance, efforts are being made to develop new materials and processing techniques that can overcome current limitations. This includes the exploration of novel sustainable polymer formulations specifically designed for additive manufacturing, as well as the development of hybrid printing approaches that combine different materials and printing methods. Additionally, there is a growing focus on understanding the relationship between printing parameters, material properties, and the performance of the final printed objects, with the aim of developing predictive models that can guide process optimization.[5]

Recycled Plastics in 3D Printing

Recycled plastics (RPs) are polymeric materials that have been reprocessed from post-consumer or post-industrial plastic waste. These materials play a crucial role in the circular economy concept, which aims to replace the traditional linear "take-make-dispose" model with a more sustainable approach6. The use of recycled plastics in 3D printing represents a significant step towards more environmentally responsible manufacturing practices, addressing the growing concern of plastic pollution while offering new opportunities for material innovation.

The evolution of recycled plastics in 3D printing has been marked by significant advancements in processing techniques and material properties. Initially, the focus was on simple mechanical recycling of common thermoplastics like PET and ABS. However, recent discoveries have expanded the scope and efficiency of plastic recycling for 3D printing applications.

One of the most recent developments is the improvement in the recycling process for PLA, a popular biodegradable polymer used in 3D printing. It was found that over five recycling cycles, PLA exhibited improved crystallinity, though with a slight reduction in break strain from 1.88% to 1.68%.[7] The molecular weight of PLA decreased by 26.73% after three cycles and 44.91% after five cycles, indicating the need for strategies to maintain material integrity over multiple recycling iterations.[8]

Another significant advancement is the development of recycling techniques for carbon fiber reinforced polymers (CFRPs). A method for recycling and remanufacturing continuous carbon fiber-reinforced thermoplastic composites using 3D printing was proposed.[9] This process achieved a material recovery rate of 100% for carbon fiber and 73% for

PLA matrix, significantly reducing environmental impact while maintaining comparable mechanical properties to virgin composites.[10]

Researchers have also explored innovative additives to enhance the properties of recycled plastics. For instance, the addition of lignin to recycled PLA has been found to improve melting properties, albeit with a slight decrease in tensile strength and Young's modulus11. Similarly, the incorporation of carbon fibers into recycled plastics has shown promise in improving mechanical properties, with some studies reporting a 25% increase in bending strength compared to the original material.[11]

Advantages of Recycled Plastics

Recycled plastics offer several advantages over virgin polymers, making them increasingly attractive for 3D printing applications:

Environmental Benefits

Using recycled plastics can reduce waste, lower carbon emissions, and conserve fossil resources. It's estimated that recycling and remanufacturing plastic can save between 30% and 80% of carbon emissions compared to processing and manufacturing virgin plastics.[8]

Cost-Effectiveness

Material-reuse solutions in 3D printing have shown potential cost reductions ranging from 10% to as high as 70-80% for printed components.[8]

Energy Efficiency

Studies have shown that recycling processes for 3D printing filaments can be significantly more energy-efficient than producing virgin materials. For instance, recycled HDPE filament production resulted in an embodied energy of 38.29 MJ/kg, compared to 79.67 MJ/kg for virgin HDPE feedstock.[8]

Comparable Performance

In many cases, recycled plastics can achieve mechanical properties comparable to virgin materials. For example, PLA specimens obtained from recycled food packaging showed similar thermal stability and only minor differences in mechanical properties compared to virgin PLA.[8]

Versatility

Recycled plastics can be blended with additives or other polymers to tailor their properties for specific applications, offering a high degree of customization.

Applications of Recycled Plastics in 3D Printing

The use of recycled plastics in 3D printing is rapidly expanding, driven by both environmental concerns and technological advancements:

Filament Production

Companies and projects like Reflow, Nefilatek, and Filabot are developing technologies to transform various types of plastic waste, including PET bottles, into high-quality 3D printing filaments.[11]

On-Site Recycling

The concept of distributed recycling, where consumers recycle their own waste into 3D printing filament using devices like "recyclebots," is gaining traction. This approach reduces transportation emissions and promotes local, sustainable manufacturing.[11]

Composite Materials

Recycled plastics are being combined with reinforcing materials like carbon fibers to create high-performance composites for 3D printing. These materials find applications in industries such as aerospace, automotive, and defense.[10]

Novel Applications

Researchers are exploring the use of unconventional waste plastics for 3D printing, such as nylon from fishing nets and thermoplastic urethane from recycled ski boots.[11]

Improved Processing Techniques

Advancements in extrusion technology and post-processing methods are enabling the production of higher quality recycled filaments. For instance, techniques like coating recycled PLA filaments with polydopamine have been developed to improve their thermal stability and mechanical properties.[11]

3D Printing with Recycled Plastics

The integration of recycled plastics into 3D printing processes represents a significant step towards sustainable manufacturing and circular economy practices. This section explores the methods, challenges, and mitigation strategies associated with 3D printing using recycled plastics.

The process of 3D printing with recycled plastics typically involves several key steps:

1. Collection and Sorting: Plastic waste is collected and sorted according to type.
2. Cleaning: The sorted plastic is cleaned to remove contaminants.
3. Shredding: The cleaned plastic is shredded into small flakes or pellets.
4. Extrusion: The shredded plastic is melted and extruded into filaments suitable for 3D printing.
5. Printing: The recycled filament is used in a 3D printer, typically using Fused Deposition Modeling (FDM) technology.

For different types of recycled polymers, the process may vary slightly:

Polyethylene Terephthalate (PET)
Commonly sourced from water bottles, PET is often recycled into filament through a process of grinding, washing, drying, and extrusion. The resulting filament can be used in standard FDM printers.[12]

High-Density Polyethylene (HDPE)
HDPE, often from milk jugs or detergent bottles, can be processed similarly to PET. However, it may require higher extrusion temperatures and careful cooling to maintain dimensional stability.

Polylactic Acid (PLA)
While not traditionally "recycled" in the same sense as petroleum-based plastics, PLA from failed prints or support structures can be reground and reextruded into new filament.[13] It was studied that PLA recycled from food packages and found that the mechanical recycling process induced some chain scission, leading to a decrease in mechanical properties.[14]

Acrylonitrile Butadiene Styrene (ABS)
ABS can be recycled through similar grinding and extrusion processes. It was studied that ABS recycled from car dashboards and found that, unlike PLA, recycled ABS did not show significant changes in mechanical properties compared to virgin ABS.[14]

Carbon Fiber Reinforced Polymers (CFRP)
It was found that the researchers developed a novel method for recovering and remanufacturing composite materials using 100% recycled continuous carbon fiber (CCF) and polylactic acid (PLA) plastic. The recovery rates achieved were 100% for CCF and 73% for PLA.[9]

Challenges
Despite the potential benefits, 3D printing with recycled plastics presents several challenges:

Inconsistent Material Properties
Recycled plastics often exhibit variability in mechanical properties due to contamination, degradation during previous use, and mixed polymer types. This can lead to unpredictable print quality and structural integrity. It was found that recycled PLA showed a 33% decrease in both tensile stress and flexural strength compared to virgin PLA.[14] To mitigate this, researchers have explored blending recycled plastics with virgin materials to achieve more consistent properties.[13]

Thermal Degradation
Repeated heating and cooling cycles during the recycling and printing process can lead to thermal degradation of the polymers, affecting their performance. It was observed that the thermal stability of recycled PLA and ABS was not significantly affected by the recycling process, as evidenced by thermogravimetric analysis (TGA).[14]

Moisture Absorption
Many recycled plastics, especially PET and PLA, are hygroscopic and can absorb moisture from the environment, leading to poor print quality and reduced mechanical properties. Proper drying of the filament before printing and storage in moisture-free conditions can help mitigate this issue.[12]

Surface Quality
It was found that both recycled PLA and ABS produced smoother surfaces compared to their virgin counterparts, with a decrease in mean surface

roughness between 55% and 65%. This could be seen as an advantage, potentially eliminating the need for post-processing in some applications.[14]

Chemical Composition Changes

FTIR analysis showed that the chemical composition of recycled PLA remained largely unchanged compared to virgin PLA.[14] However, recycled ABS showed some differences in carbonyl group presence, possibly due to oxidation during the recycling process.

Crystallinity Changes

It was observed changes in the crystallinity of recycled PLA compared to virgin PLA, which could affect mechanical properties and processing behavior.[14]

Mitigation Strategies

To address these challenges, researchers and practitioners have explored various strategies:

Material Blending

Mixing recycled plastics with virgin materials or different types of recycled plastics can help achieve a balance between sustainability and performance.[13]

Process Optimization

Careful tuning of processing parameters, including extrusion temperature, print speed, and cooling rates, can significantly improve the quality of prints made with recycled materials.

Advanced Recycling Techniques

Developing more sophisticated recycling processes, such as chemical recycling for certain polymers, can help produce higher quality recycled materials suitable for 3D printing.[12]

Quality Control

Implementing strict quality control measures during the recycling process can help ensure more consistent material properties in the recycled filament.

Design for Recycling

Considering the entire lifecycle of 3D printed products, including their eventual recycling, can help create more sustainable closed-loop systems for plastic use in additive manufacturing.[7]

Thus, while 3D printing with recycled plastics presents numerous challenges, ongoing research and development are continually improving the viability of this approach. As techniques for processing and printing recycled materials advance, the potential for creating a more circular economy in additive manufacturing grows.

Conclusion

This comprehensive review has examined the current state of research and practice in 3D printing with recycled plastics. The findings indicate that while it is indeed possible to use recycled plastics in 3D printing processes, particularly fused deposition modeling (FDM), the quality and consistency of the resulting products can vary significantly depending on the type of plastic and the recycling process used. Recycled PLA and ABS have emerged as the most commonly studied materials, with studies showing that they can be successfully used for 3D printing, with some changes in material properties compared to virgin plastics.

The review has highlighted that the mechanical recycling process can lead to some degradation in material properties, particularly in semi-crystalline polymers like PLA, while amorphous polymers like ABS tend to maintain their properties better through the recycling process. Some studies have found that recycled materials can produce smoother surface finishes in 3D printed objects, possible eliminating the need for post-processing in certain applications. The practical implications of this research are significant. The ability to use recycled plastics in 3D printing opens up new possibilities for sustainable manufacturing, helping to reduce the environmental impact of both plastic waste and 3D printing processes. It suggests that with proper material selection and process optimization, recycled plastics could be viable alternatives to virgin materials in many 3D printing applications, from prototyping to small-scale production. This could lead to reduced costs for raw materials and contribute to the development of more circular economic models in manufacturing. However, the variability in material properties observed in recycled plastics also implies that careful quality control and new design approaches may be necessary when working with these materials. Industries may need to develop new standards and testing protocols specifically for 3D printed products made from recycled plastics.

This review has also revealed several areas where further exploration is needed. Future research should focus on developing more efficient and standardized recycling processes specifically tailored for 3D printing applications. There is a need for more comprehensive studies on the long-term performance and durability of 3D printed objects made from recycled plastics, particularly in demanding applications. Additionally, there is scope for expanding the range of recycled plastics suitable for 3D printing, including mixed plastic waste streams. Life cycle assessments comparing the environmental impact of products made from recycled versus virgin plastics in 3D printing contexts would provide valuable insights for decision-making in sustainable manufacturing.

In conclusion, the field of 3D printing with recycled plastics is currently at an exciting point, with the potential to significantly contribute to more sustainable manufacturing practices.

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The sole author was responsible for the conceptualization, methodology, data collection, analysis, writing, and final approval of the manuscript.

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