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Sustainable Additive Manufacturing: Regenerated Polymer Materials and Recycling Reuse

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Abstract. With the global shortage of resources and the increasing environmental pressure, there is an urgent need for new sustainable materials to support the development of 3D printing technology. This paper starts from the recycling routes of recycled polymers and the powder characteristics of recycled polymers, analyzes the advantages and limitations of mechanical recycling and chemical recycling, and discusses the influence of particle size distribution and morphology structure of recycled powders on fluidity and forming quality; it focuses on the application and improvement technology of recycled polymers in selective laser sintering and fused deposition modeling, and summarizes the methods to improve mechanical properties and stability such as filler reinforcement and surface modification to promote the smooth progress of the printing process; it introduces the current development status of stabilization technology for recycled polymers and the application of online monitoring technology; it summarizes and analyzes the problems such as low purity of raw materials, poor reusability and difficulty in promoting industrialization, and proposes future research priorities, providing a reference for the development and engineering application of 3D printing materials under the background of circular economy.

Keywords: Regenerated Polymer, Powder flow, Modification, Process adaptation, Industrialization

1. Introduction

Due to the severe global resource shortage and the increasing pressure on environmental protection, solving sustainable manufacturing has become one of the world's major problems. Currently, the recycling rate of traditional thermoplastic polymer materials in the field of additive manufacturing is not high, and the problems of resource waste and environmental pollution caused by the generation of a large amount of waste are still prominent [1]. Using recycled polymer materials to replace traditional raw materials for AM is an effective solution, which can effectively reduce the consumption of primary raw materials and the carbon emissions and solid waste generated during the production process, in line with the development model of "circular economy". The particle size, morphology, and specific surface area of recycled polymer powders have a significant impact on the forming accuracy of SLA [1-5], and the energy consumption, product purity, and economic cost of different recycling processes of recycled polymers (i.e., mechanical recycling and chemical recycling) are also issues that need to be urgently addressed. Regarding material modification, based

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on previous research, filling modification and interface modification can improve the mechanical properties of recycled materials and enhance printing stability. The quality of recycled plastics is being continuously improved to achieve this goal [6-8]. Because recycled polymers have good application prospects, some problems still need to be solved before they can be put into practical use. This paper systematically analyzes the recycling routes, powder characteristics, and process compatibility of recycled polymers, and proposes technical ideas for improving forming consistency and mechanical properties through composite modification and re-stabilization treatment, which can provide theoretical and methodological support for the reliable application of recycled materials in SLS/FDM and other processes. By introducing digital online monitoring and process-material coupling optimization, a feasible path for real-time quality control and closed-loop optimization during the printing process is provided. It is hoped that this work can promote the industrial application of recycled polymers in additive manufacturing and contribute to the realization of the circular economy and carbon reduction goals.

2. Obtaining and characteristics of recycled polymers

2.1. Overview of mechanical and chemical recycling

There are mainly two ways of recycling regenerated polymers: mechanical recycling and chemical recycling. Mechanical recycling refers to the process of directly reusing waste plastics as recycled materials after physical treatments such as crushing, screening, and remelting. This process is relatively simple and has a lower production cost. However, due to issues such as thermal degradation and impurity generation during the recycling process, the performance of some materials may decline [5]. Chemical recycling involves breaking down the polymer chains of waste plastics through chemical reactions such as wet depolymerization, ester exchange, and pyrolysis, and then reassembling them into new chains. This process can restore or optimize the fluidity and molecular weight distribution of polymers and remove various impurities to obtain higher-purity monomers or oligomers. However, it is energy-intensive, complex, and requires the addition of efficient catalysts and control of appropriate reaction conditions [4,9]. Recent studies have found that combined recycling can complement the advantages of both methods. Although there are still differences between virgin polymers and recycled polymers, the approach of first chemically removing impurities and then mechanically crushing and remolding can achieve a balance between economy and usability, providing a new idea for the preparation of powder raw materials for 3D printing.

2.2. Powder particle size, morphology and fluidity

The particle size distribution, particle morphology and specific surface area of recycled polymer powder determine the layer thickness selection and uniformity of spreading in the Selective Laser Sintering (SLS) process [2,6]. A narrower particle size distribution (35-100 μm), finer and rounder particles are preferred, as this enables better control of layer thickness and uniform powder spreading, leading to higher quality of the formed parts [6]. At the same time, a smoother surface and a more spherical shape of the powder particles are beneficial for reducing the friction coefficient between the particles, thereby enhancing the powder flowability and bulk density, and achieving better densification and mechanical properties of the formed parts. However, compared to new powder, the recycled powder is more prone to fragmentation and edge burrs due to the high-temperature regeneration and multiple thermal cycles, resulting in uneven packing and unstable

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powder spreading. For such powders, certain methods need to be employed for improvement, such as mechanical ball milling or surface coating. Mechanical ball milling can increase the powder bulk density and interlayer bonding performance by reducing the particle size and increasing the specific surface area and surface roughness. However, high-energy or excessive ball milling may cause polymer chain breakage and thermal oxidation degradation, so the energy input and processing time must be strictly controlled. Surface coating introduces a functional thin layer on the powder surface to adjust the wettability, surface energy and light-heat absorption characteristics, thereby inhibiting agglomeration and improving flowability. The prerequisite is that the selected coating material must be compatible with the base material and have sufficient thermal stability [2]. If the powder has a large specific surface area, it is conducive to laser absorption, melting and bonding, but it may also cause problems such as easy moisture absorption and agglomeration. During powder production and storage, humidity control and the addition of fluidization aids should be noted [6].

3. The application of recycled materials in additive manufacturing

3.1. Direct reuse and compound modification

During the 3D printing process, some studies have reported that for certain polymer powders, simple pretreatment such as vibration screening and appropriate drying can make the powder suitable for SLS technology. For instance, Pezold et al. in their research on PET powder adopted a pretreatment process involving vibration screening (190 μm) and drying at 170 °C for 5 hours. The powder treated in this way showed no significant decrease in melt flow and molecular weight during high-temperature short-time sintering, and the mechanical properties and dimensional stability of the printed parts were within acceptable ranges [10]. However, due to the relatively high incorporation amount, the mechanical properties of the formed parts might slightly decline [2,3,9]. To enhance the printing performance of recycled polymers, composite modification methods are generally adopted by adding inorganic fillers, fibers, and nano-reinforcing phases, etc. [4,5]. Adding rice husk (RH) to rPP recycled powder, the porous structure of rice husk powder mechanically interlocks with the plastic matrix, significantly improving the tensile strength and modulus of the printed parts, and enhancing their thermal conductivity, which is beneficial for improving laser absorption and interlayer bonding strength [3]. Interface modifiers (coupling agents or compatibilizers) can be used to enhance the interfacial bonding strength between recycled polymers and fillers, compensating for the loss of mechanical properties caused by the reduction in molecular weight after degradation [5].

3.2. Application performance in typical processes

In the SLS process, the performance of recycled powder is closely related to powder fluidity and laser absorption characteristics. Some characterization methods can be used to observe the flow properties of polymer powder in selective laser sintering [2]. Generally speaking, in traditional process applications, SLS recycled powder can be recycled while ensuring powder fluidity and laser absorption. The density of the formed powder after recycling can be greater than 95%, and the tensile strength attenuation does not exceed 5%, with no significant difference from the original polymer [6,8]. However, unmodified recycled powder is prone to problems such as many internal pores and poor interlayer bonding in the formed parts, with severe attenuation of mechanical properties [2]. Experiments on the recycling and utilization of recycled powder in the FDM process show that PLA and PETG recycled twice can be used multiple times, and the extrusion temperature and printing speed of the FDM process can be appropriately optimized to compensate for the

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negative impact of material viscosity changes on FDM printing accuracy [3,7]. In summary, for different types of recycled polymers, the corresponding process parameters can be optimized, and composite modification and quality stabilization techniques can be used to ensure their effective use in mainstream additive manufacturing processes. However, for different types of polymers, we still need to match the process, material and performance.

4. Performance and quality control

4.1. Mechanical properties and processing stability

The mechanical properties and processing stability of recycled polymers depend on the molecular weight of the recycled polymer raw materials, the type of fillers, and the printing process parameters. The tensile strength of unmodified recycled powder formed parts is generally 10% - 20% lower than that of virgin materials [6]. After adding reinforcing phases such as carbon nanotubes and glass fibers, the mechanical property decline does not exceed 5% [5,8]. Processing parameters such as processing temperature, powder spreading thickness, and laser power have a significant impact on the forming stability: higher temperatures are prone to thermal degradation and warping, while lower temperatures can lead to poor interlayer bonding and porosity [6]. Therefore, the appropriate process window should be selected based on the specific material properties to balance mechanical properties and printing accuracy.

4.2. Re-stabilization treatment and online monitoring

Most recycled polymer materials will experience molecular chain breakage and degradation after multiple thermal cycles, leading to performance deterioration [11]. For these materials, implementing "re-stabilization treatment" by adding antioxidants, light stabilizers, or chain transfer agents can effectively reduce molecular weight loss and restore the molecular weight distribution, enabling the density and strength of printed products to recover to over 90% of the original raw materials [8]. Additionally, for the defects generated during printing, methods such as laser scanning thermal imaging and real-time monitoring of powder bed thickness/temperature are adopted to promptly identify and quantify defects such as overburning, incomplete melting, delamination between layers, and uneven powder spreading, thereby providing immediate warnings for abnormal areas. Based on such online information, reverse adjustments can be triggered, significantly reducing the scrap rate and post-processing costs, and helping to suppress residual stress and warpage, thereby enhancing the consistency and mechanical properties of formed parts. Long-term accumulated monitoring data can also be used for process traceability, process-material coupling analysis, and quality prediction based on machine learning, promoting the realization of closed-loop adaptive control strategies and stable large-scale production [1,2,12]. The combination of re-stabilization treatment and online monitoring can better ensure the continuous high-quality and high-efficiency 3D printing.

5. Shortcomings and prospects

5.1. Key bottleneck

At present, the raw material sources of recycled polymer powders are mixed, with different types of waste mixed in, residual additives, and trace impurities that are difficult to remove. This leads to significant differences in particle size distribution, morphology, and chemical composition among

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different batches of powders, resulting in poor uniformity during powder spreading and poor consistency of formed parts. Moreover, after multiple thermal cycles, molecular chain breakage and cross-linking occur, and the more severe the decrease in molecular weight, the more the mechanical properties and thermal stability decline exponentially. Currently, there is no composite modification and re-stabilization method that can fully restore its performance and is cost-controllable. At the same time, research on the coupling optimization of material and additive manufacturing process parameters is still fragmented, and there is a lack of a platform that can achieve digital design and experimental verification to provide a unified solution for the polymer system. Additionally, although many online detection methods based on direct optical measurement, energy spectrum analysis, etc. are still in the concept proof stage, only a small portion can be implemented in industrial sites and achieve real-time full-process tracking through real-time feedback and adaptive control closed-loop, thus making it impossible to achieve large-scale application of recycled powders.

5.2. Future research directions and industrialization prospects

Looking to the future, efforts should be made to develop highly selective and low-energy-consumption purification and separation methods and technologies, to form a traceable raw material supply chain, and to ensure the consistency and controllability of recycled powders. Research should be conducted at the molecular level on precise repair and functionalization, using enzyme catalysis and reversible chemical bond breaking and recombination methods to restore damage to the polymer main chain caused by heating and enhance the strength of the polymer main chain. By means of nano-reinforcement, interface engineering and other methods, an integrated operation of "repair-strengthening" at the molecular level should be achieved. A comprehensive digital simulation experimental verification platform covering powder preparation - 3D printing - post-processing should be established, and based on multi-scale modeling and machine learning-driven parameter optimization schemes, the correlation mapping relationship between multi-scale models and their multi-physical fields should be realized on this platform. On this basis, a multi-source online monitoring system, including thermal imaging, powder bed laser scanning, intelligent sensing and other online monitoring devices, should be developed. Then, based on edge computing and adaptive control strategies, an industrial closed loop that truly achieves the function of "seeing defects - regulating processes" should be realized, providing an important guarantee for the large-scale application of recycled polymer materials in the field of additive manufacturing.

6. Conclusion

After undergoing strict particle size classification, modification and optimization of process parameters, recycled polymers can approach the quality of virgin materials in additive manufacturing processes such as SLS/FDM. The main existing bottlenecks are impurities and uneven components in raw materials, molecular weight degradation, insufficient powder fluidity, and the lack of unified characterization and quality standards. Looking ahead, efforts should be prioritized on promoting efficient purification and molecular repair technologies, composite modification, unified characterization standards, and digital online monitoring and process closed-loop to support the reliable large-scale application of recycled materials.

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