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ACTIVE INTERLAYER HEATING FOR SUSTAINABLE SELECTIVE LASER SINTERING WITH RECLAIMED POLYAMIDE 12 POWDERS

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ABSTRACT

Selective laser sintering (SLS) technology produces a substantial amount of un-sintered polyamide 12 powders after the manufacturing process. Failure to recycle and reuse these aged powders not only leads to economic losses but also is environmentally unfriendly. This is particularly problematic for powder particles close to the heat-affected zones that go through severe thermal degradations during the laser sintering processes. Limited procedures exist for systematically reusing such extremely aged powders. This work proposes a systematic method to maximize reusability of aged and extremely aged polyamide 12 powders. Building on a previously untapped interlayer heating, pre-processing, and a systematic mixing of powder materials, we show how reclaimed polyamide 12 powders can be consistently reprinted into functional samples, with mechanical properties even superior to current industrial norms. In particular, the proposed method can yield printed samples with 18.04% higher tensile strength and 55.29% larger elongation at break using as much as 30% of extremely aged powders compared to the benchmark sample.

Keywords: Selective laser sintering; Powder reuse; Powder aging and degradation; Interlayer heating; Sustainability

INTRODUCTION

Selective laser sintering (SLS) is one of the most well established and commonly used additive manufacturing (AM) techniques to rapidly manufacture three-dimensional components [1-4]. Polymeric powders, semicrystalline or amorphous, are the first and still the most widely applied materials in SLS [1,5,6]. Parts printed using amorphous polymer powders are partially consolidated, and consequently can be useful for applications when the strength and durability of parts are not dominant [7]. On the contrary, parts printed using semicrystalline polymer powders are fully consolidated with high mechanical strength and effectively weakened warpage. Among the semicrystalline polymers, polyamide families are most popular for SLS, and polyamide 12 dominates the market because of the capability to generate strong parts for common applications [3,4,8].

Despite the popular applications of the polyamide 12 powders in SLS, the volume ratio of powders that translate to parts is small (5% - 15%). The 85% to 95% residual powders went through deteriorate physical and chemical degradations in the intricate fabricating processes [3,9], but have the potential to be recycled and reused for further applications [3,4,10,11]. However, the deteriorated powders have reduced surface morphologies, larger and more complex molecular chains, decreased flowability, and deteriorated mechanical and thermal properties, which make it challenging to reuse them directly [3,4,10-12]. Also, polyamide 12 powder is relatively expensive, priced around $150/kg in 2019 [9]. Abandonment of the residual polyamide 12 powders can cause not only economic losses but also environmental pollution. Thus, reclaim and reuse are difficult yet necessary for a sustainable SLS AM.

Relevant works on the reuse of the reclaimed polyamide 12 powders have been reported in recent years. L. Feng et al. [9] reclaimed polyamide 12 from SLS and made the powders into filaments for fused deposition modeling (FDM). Clarifying the aging mechanisms on thermal behavior, coalescence behavior and the resulting crystallinity, microstructure and mechanical properties, and investigating systematic aging mechanisms and microstructural evolution, P. Chen et al. [3] and S. Dadbakhsh et al. [4] characterized the aging process of polyamide 12 powders

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in SLS. Dotchev et al. [10], Wegner et al. [13] and Josupeit et al. [14] verified the decreased flowability of the reclaimed polyamide 12 powders compared to new ones through a melt volume rate (MVR) index. The effect of powder reuse on mechanical properties has been studied by R.D. Goodridge et al. [15] and K. Wudy et al. [16], who pointed out the changes of tensile strength as well as elongation at break in parts built from aged polyamide 12 powders. In addition, the relationships between preheating temperature [13], energy density [13,17], combined dwelling time between layers, energy density [18], and part quality were studied.

Although existing efforts have sought to understand the aging mechanisms and reuse of the degraded polyamide 12 powders, the possibility and feasibility of reusing the residual polyamide 12 powders remain not fully exploited. In particular, the reuse of the extremely aged polyamide 12 powders close to the heat-affected zones (HAZs)[2] has not been reported. This paper seeks to bridge the missing link and proposes a new method, hereby referred to as active interlayer heating, to build parts with the reclaimed and extremely aged polyamide 12 powders. We discuss systematic approaches to reuse the polyamide 12 powders of different degradation levels, different mixing percentages, and different combinations with multiple layer printing and superior mechanical properties. The results show that reclaimed polyamide 12 powders can be consistently reprinted into functional samples, with mechanical properties comparable or even superior to current industrial norms.

1 Proposed active interlayer heating for reclaimed polyamide 12 powders

The main procedures of the proposed method include powder collection, powder preprocessing, powder mixing, powder characterizations, parameter control, SLS with interlayer heating, and part characterizations, of which the flowchart is presented in Figure 1.

1.1 Materials sample preparation

Materials sample preparation includes powder collection, powder preprocessing and powder mixing. We collected polyamide 12 powders purchased from EOS Corp. and reclaimed from standard SLS processes on an EOS P 390 machine. The powders cover three levels of degradation: (i) new powders without heat treatment, (ii) aged powders located far away from the HAZs during SLS and currently reused in the industry, and (iii) extremely aged powders located close to the HAZs and not being actively reused in SLS.

The extremely aged powders could not be coated smoothly in the SLS chamber because of the existence of the aggregated large particles, as a result of striking drop in flowability. In this work, a sieving process was applied to the extremely aged powders prior to printing. This process was done in a fume hood using a sieve with the mesh size of 200 μm to grind the powders. The extremely aged powders after the sieving process were coated smoothly in the SLS chamber.

[2] Areas close to the laser-material interaction during sintering.

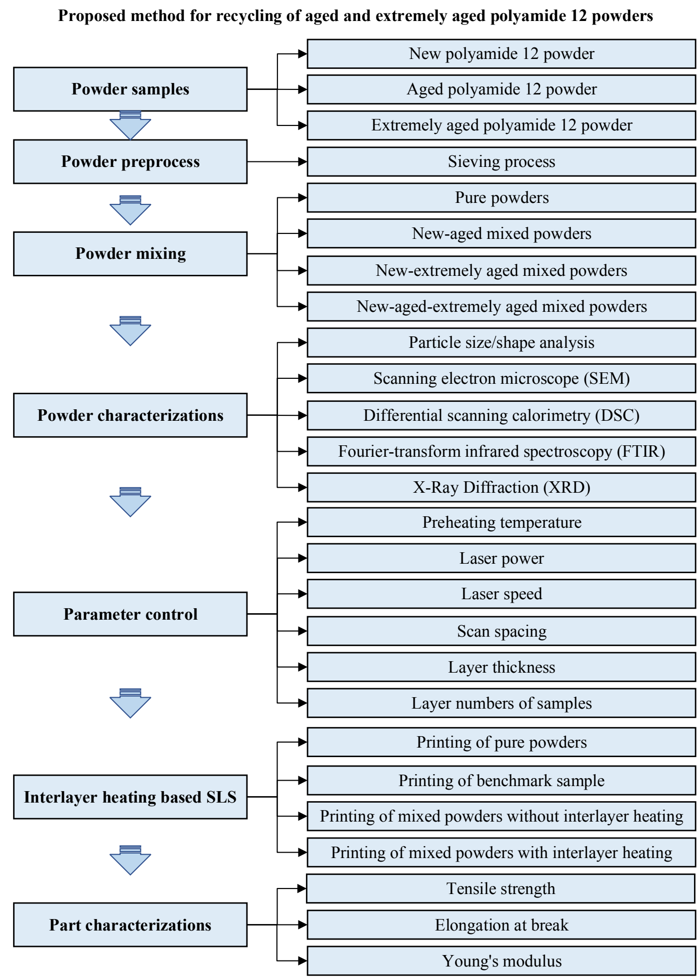


Figure 1: PROPOSED METHOD FOR REUSING AGED AND EXTREMELY AGED POLYAMIDE 12 POWDERS

Four different groups of powders were used in experiment (as shown in Figure 1): pure powders, mix of new and aged powders, mix of new and extremely aged powders, and mix of new, aged and extremely aged powders, to be specific.

1.2 Powder characterizations

The particle size/shape distribution of new, aged, and extremely aged polyamide 12 powder samples were examined using a PartAn 3-D particle size and shape analyzer. To get reasonable test results, over 500,000 particles were measured in each experiment. Scanning electron microscope (SEM) was carried out to evaluate the variations in micro surface morphologies of the collected powders. We used a Teneo LVSEM and coated the samples with a thickness of 3 nanometers of Pt/Pd to increase material conductivity. To measure the thermal properties, we conducted differential scanning calorimetry (DSC) on the powder samples of different degradation degrees using a TA Instruments DSC Q20. To measure the differences of molecular components and the chemical microstructures between new and reclaimed materials, we applied Fourier-transform infrared spectroscopy (FTIR) to different powder samples. We also use X-Ray Diffraction (XRD) to check crystalline properties of different polyamide 12 materials.

1.3 Parameter control

As shown in Figure 1, six parameters that have significant influences on part qualities were controlled in this study: preheating temperature, laser power, laser speed, scan spacing, layer thickness, and layer numbers. These parameters were accurately controlled by our in-house developed SLS testbed (Figure 2) in experiment. The selected optimal parameter settings suitable for powders of different degradation levels were 160 °C preheating temperature; 3000 mm/s scan speed; 18 W laser power; 0.3 mm scan spacing; and 150 µm layer thickness. In our testing, we printed 10-layer tensile bar samples for each powder combination.

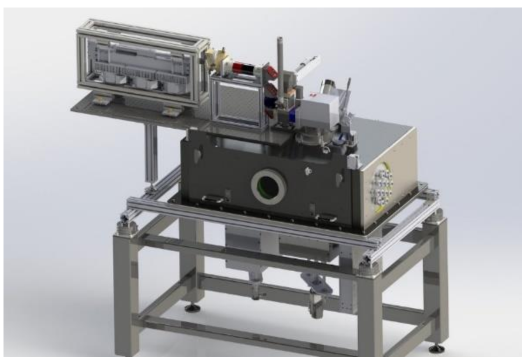


Figure 2: IN-HOUSE BUILT AND CUSTOMIZED SLS TESTBEDS

1.4 SLS with interlayer heating

The proposed SLS with interlayer heating applies carefully controlled thermal energy to the specimens being printed in-between each printed layer. In this process, powder materials were controlled to maintain at a preheating temperature (160 °C for polyamide 12) for a certain heating time. In this work, we tested 0 second (no interlayer heating) and 60 seconds interlayer heating to powders and powder mixtures with different degradation levels to explore the influences of interlayer heating on part mechanical properties. Through the experiment results, it was verified that 60 seconds interlayer heating can provide the 10-layer samples with enough heat and energy.

We carried out a series of continuous experiments to explore the reusability of aged and extremely aged powders and the influences of interlayer heating on part properties, as shown in Figure 3. The experiments were separated into six stages, with a distinct objective for each stage. In stage 1, pure powders were used to verify the feasibility of multi-layer printing with reclaimed powders. In stage 2, the currently available industrial reuse combination (50%-50% new-aged mixed powders) were used to print the benchmark samples of which the mechanical properties were regarded as the baselines in this research. The aim of stage 3 was to check the part mechanical properties when using new-extremely aged mixed powders with the percentages of extremely aged powders increasing. In stage 4, to examine the influences of interlayer heating on part mechanical properties, all the mixed powder combinations in stages 2 and 3 were reapplied and were printed with 60 seconds interlayer heating. By comparing the mechanical properties of samples in stages 2, 3, and 4, influences of interlayer heating on part properties were obtained. In stage 5, different from the previous stages using the combinations of two kinds of powders, parts were printed with mixed powders composed of three types of powders to verify the feasibility and potential benefits of this practice. To ensure the part quality, the combinations of 30% new - 30% aged - 40% extremely aged powders and 30% new - 40% aged - 30% extremely aged powders were selected to print. To examine the influences of interlayer heating, the experiments in stage 5 were repeated with 60 seconds interlayer heating in stage 6.

The SLS with interlayer heating method can provide enough heat and energy to promote the coalescence behaviors of reclaimed powders and improve part densification, and the mixed new-reclaimed powders can improve the diverse grain sizes in reclaimed materials, which are both helpful to improve part tensile strengths when using reclaimed polyamide 12 powders. The method can also explore the influences of interlayer heating and powder qualities on part microstructures, like crystalline ratios and crystal sizes, and part elongation at break.

1.5 Part characterizations

Tensile tests were carried out on all the SLS fabricated samples. The samples were designed based on ASTM standards and were stretched for each tensile measurement using an Instron 5869 Electromechanical testing system with a maximum load frame capacity of 50 kN equipped with Blue Hill control software. A polishing treatment was done on both the top and bottom surfaces of part to remove any skirmish un-sintered particles prior to the tensile testing for accurate measurement.

2 Results and discussions

2.1 Powder test results

The particle size distributions show that the percentages of the particles with the diameter greater than 100 µm are 21%, 22%, and 24% for new powder, aged powder, and extremely aged powder, respectively, showing a mild increase after the physical/chemical degradations. The obtained particle shape distributions show that the percentages of particles with good sphericity (0.9~1.0) for new, aged, and extremely aged powders are 21%, 16%, and 14%, respectively. The sphericity of the reclaimed powder decreases mildly, which eventually increases part surface roughness. In other words, the percentage of particles with non-spherical and irregular shapes or rough edges increases.

The SEM results show that with a 500x magnification, few aggregated large size particles were found in new powders, while there were more large size particles found in aged powders and extremely aged powders as aggregation exacerbated. At a 2000x magnification, there were no cracks in new powders, obvious cracks in aged powders, and more cracks in extremely aged powder in the SEM scope. These cracks were attributed to the evaporation of moisture or alcohol during the sintering cycles or repeated expansions/shrinkages during multiple times recycling.

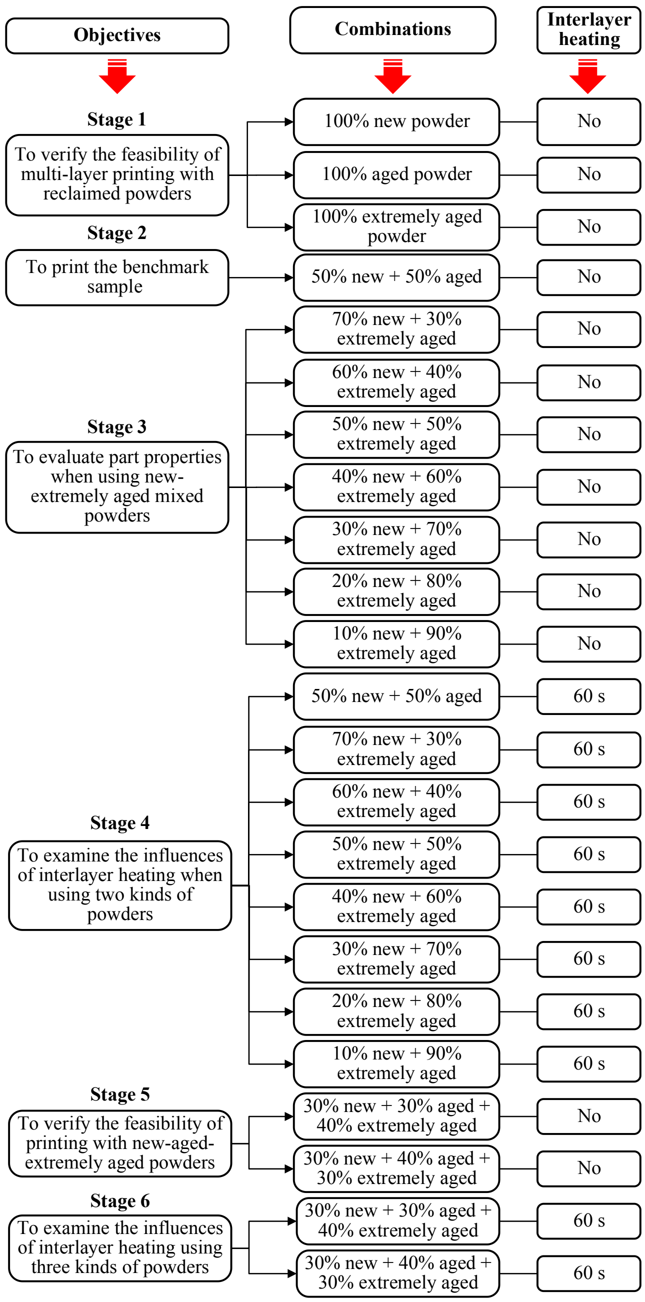


Figure 3: EXPERIMENT PLANS ABOUT REUSABILITY OF AGED AND EXTREMELY AGED POWDERS AND INFLUENCES OF INTERLAYER HEATING (THE MIXED POWDERS ARE IN VOLUME PERCENTAGES)

The DSC test results exhibit that the onset melting temperatures decrease for reclaimed powders (in the heating cycle), which is important knowledge for parameter settings of SLS printing when the aged or extremely aged powders are used. The setting principle of the preheating temperature is that the preheating temperature should be set close to the material onset melting temperature or melting point [12,16,17]. The decrease of onset melting temperature of aged and extremely aged powders gave us significant instructions when printing aged or extremely aged powder because the appropriate setting of preheating temperature is dominant to keep the sintering processes going and to ensure the part quality.

FTIR test results present that peaks at the vibrational frequencies of 1369.23, 1159.03, 1062.60 and 948.82 cm^{-1} slightly decreased in aged powders and extremely aged powders compared to new powders, which was a sign of oxidation reactions. Therefore, like the material aging after the sintering process, the aging mechanisms of the recycled powders impacted by the existence of laser and high temperature are thermal oxidations.

The XRD test results of new, aged, and extremely aged polyamide 12 powders presented similar behaviors and are in different colors. The XRD peak at 10.98° was selected as a reference of crystallization. These curves all exhibited α-structures with very high crystallinities, unstable structures and oriented in an anti-parallel manner.

2.2 Part test results

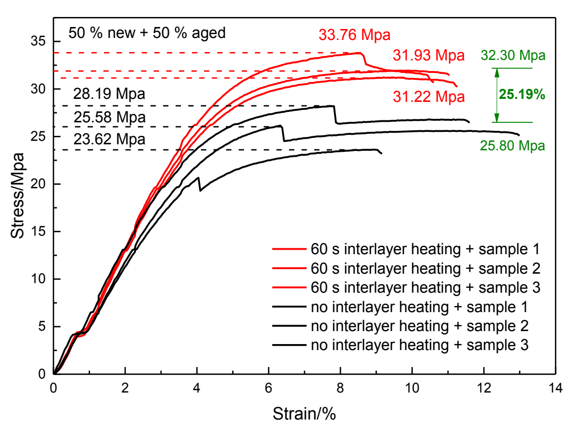
Based on the proposed method, we printed successfully tensile bars with 10 layers of powders. Using differently degraded materials, the same series of optimized parameters were applied to all the designed experiments to ensure the equivalent processing conditions. Tensile test was done, and the results are analyzed in this section.

Known from the stress-strain curves of samples printed using pure powders, the average tensile strengths of samples printed using pure new, aged and extremely aged powders are respectively 22.96 Mpa, 18.12 Mpa, and 11.08 Mpa, and that of samples using aged and extremely aged powders decrease by 21.09% and 51.75% compared to new ones. The average Young's modulus of samples printed using pure new, aged, and extremely aged powders are respectively 503.67 Mpa, 358.50 Mpa, and 177.00 Mpa, and that of samples using aged and extremely aged powders decrease by 28.82% and 64.86% compared to new ones. The average elongations at break of samples using pure new, aged, and extremely aged powders are respectively 5.31%, 12.12%, and 9.56%, and that of samples using aged/extremely aged powders increase by 56.20% and 44.46% compared to new SLS parts. The reason for this is attributed to that the reclaimed powders have smaller crystal size, increased flexibility and decreased brittleness. Inversely, parts using new powders with larger crystal size are easier to break down before the separation of crystals, with decreased flexibility.

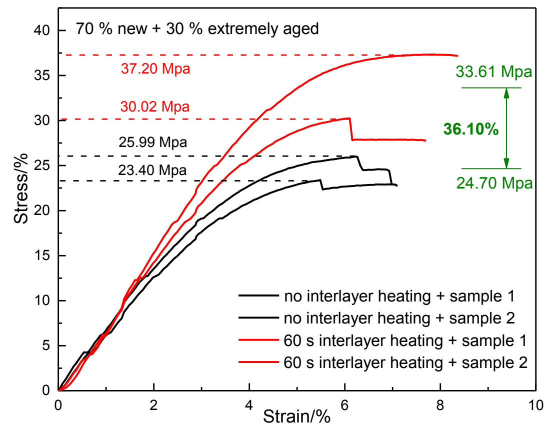
The samples printed using the 50%-50% new-aged mixed powders are taken as the benchmark samples, of which the mechanical properties are baselines in the proposed method. Calculating the average mechanical properties of the benchmark samples, the baselines of tensile strength, Young's modulus and elongation at break are respectively 25.80 Mpa, 568 Mpa, and 11.36%.

Samples using new-extremely aged mixed powders (from 70%-30% new-extremely aged mixed to 10%-90% new-extremely aged mixed) without interlayer heating were printed in stage 3. Known from the stress-strain curves of these samples that with the percentages of extremely aged powders increasing from 30% to 90%, the average tensile strengths of samples are 24.69 Mpa, 29.18 Mpa, 25.32 Mpa, 24.44 Mpa, 20.93 Mpa, 22.65 Mpa and 29.97 Mpa, and the average elongations at break are 7.10%, 7.76%, 8.60%, 8.43%, 12.33%, 13.38% and 15.36%. From an overall perspective, the average tensile strength of parts from the combination of 60%-40% new-extremely aged mixed powders to the combination of 30%-70% new-extremely aged mixed powders show decreases. But this didn't apply to the remaining mixing percentages, which can be ascribed to that the mechanical properties of samples are not only related to the properties of powders but also relevant to the thermal or laser conditions in the sintering chamber. However, the elongations at break increase with the increasing of extremely aged powders because the microstructure changes.

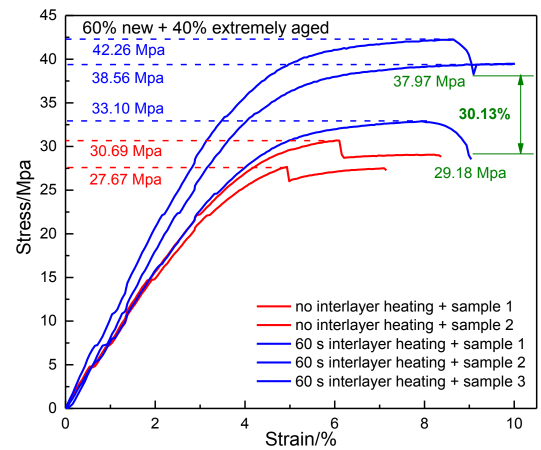
The comparisons of stress-strain curves of benchmark samples and samples using new-extremely aged mixed powders with and without interlayer heating are shown in Figure 4. As shown, the tensile strengths of some samples remain no significant changes after interlayer heating, while that of the other samples increase. In Figure 4 (a), (b), (c), (e), (f), the tensile strengths of samples with 60 seconds interlayer heating increased by 25.19%, 36.10%, 30.13%, 5.46% and 22.51%. Thus, the tensile strengths of samples using mixed powders can be improved to some extent after interlayer heating because of the better melting and coalescence behaviors of particles on each layer.



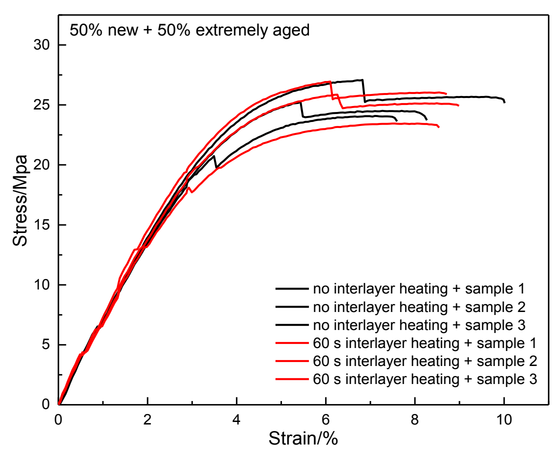
(a) 50% new + 50% aged



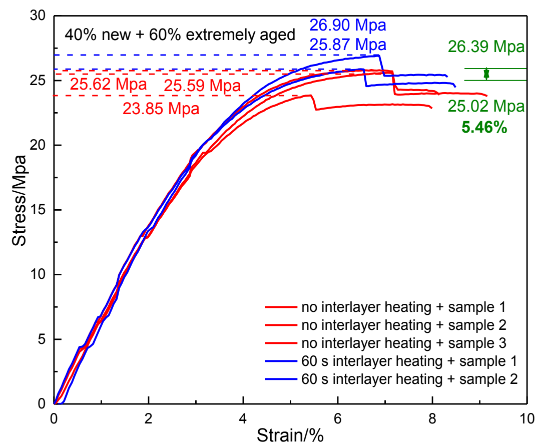
(b) 70% new + 30% extremely aged



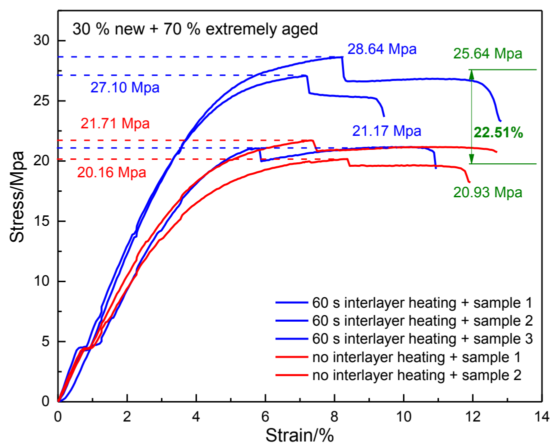
(c) 60% new + 40% extremely aged



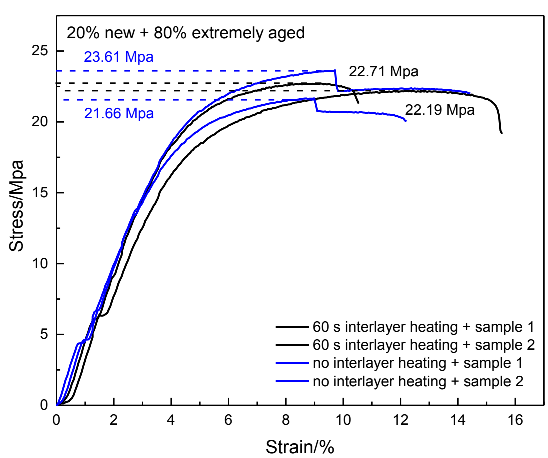
(d) 50% new + 50% extremely aged



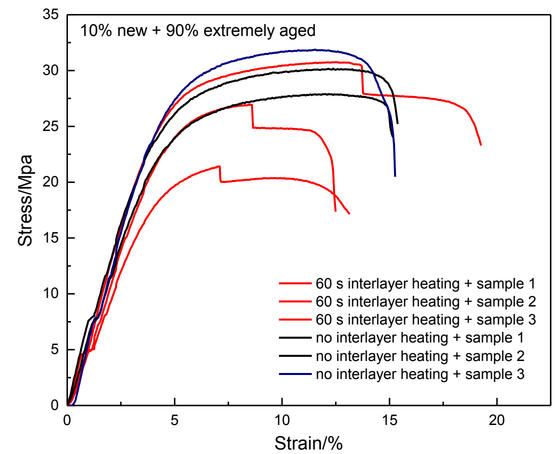
(e) 40% new + 60% extremely aged



(f) 30% new + 70% extremely aged



(g) 20% new + 80% extremely aged



(h) 10% new + 90% extremely aged

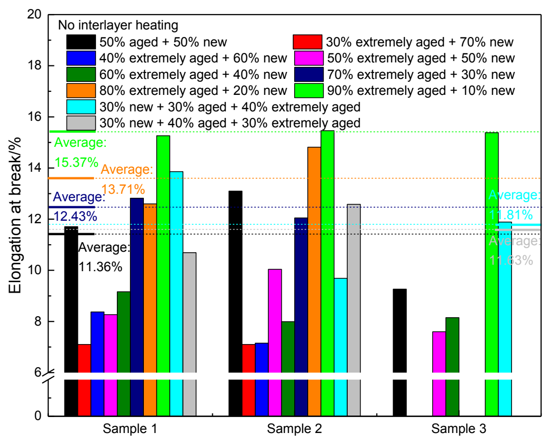
Figure 4: COMPARISONS OF STRESS-STRAIN CURVES OF BENCHMARK SAMPLES AND SAMPLES USING NEW-EXTREMELY AGED MIXED POWDERS WITH AND WITHOUT INTERLAYER HEATING

Comparisons of stress-strain curves of samples using new-aged-extremely aged mixed powders with and without interlayer heating show that after 60 seconds interlayer heating, the tensile strengths of samples using 30%-30%-40% new-aged-extremely aged mixed powders increase by 1.19%, and that of samples using 30%-40%-30% new-aged-extremely aged mixed powders increase by 18.04%. It is concluded that the tensile strengths of samples using new-aged-extremely aged mixed powders can be improved after interlayer heating.

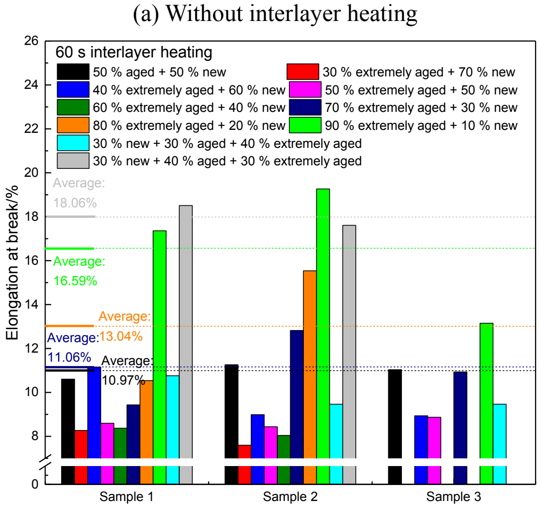
2.3 Discussions

We compare here sample tensile strengths without interlayer heating and with 60 seconds interlayer heating. The baseline tensile strength comes from the samples printed from the 50%-50% new-aged blend without interlayer heating, i.e., 25.80 Mpa. For samples without interlayer heating, the blends of which the tensile strengths are larger than the baseline are 40%-60% new-extremely aged, 30%-30%-40% new-aged-extremely aged, and 10%-90% new-extremely aged. When the percentages of extremely aged powders increased from 50% to 80%, the averaged tensile strengths decrease. The reasons are that it is more difficult to move dislocations through and between the grains when there are more extremely aged powders. For samples with interlayer heating, it is concluded that there are three blends with 60 seconds interlayer heating of which the tensile strengths are larger than the baseline (25. 80 Mpa), 50%-50% new-aged, 40%-60% new-extremely aged and 30%-30%-40% new-aged-extremely aged (56.78%, 25.19% and 24.69% better than baseline). From an overall perspective, samples using more new powders have larger tensile strengths.

The comparisons of sample elongations at break are shown in Figure 5, (a) without interlayer heating, (b) with 60 seconds interlayer heating. The baseline of elongation at break is also from the samples printed using 50%-50% new-aged blend without interlayer heating, 11.36%. Known from Figure 5 (a) and (b), there are several blends that have similar or larger values of elongations at break compared to the baseline. For the samples without interlayer heating, the samples using more extremely aged powders have larger elongation. The largest elongation is from the 10% new - 90% extremely aged blend (35.30% better than baseline), for increased extremely aged powders yields the smaller crystal size, and flexibility increases. The other mixing percentages of samples without interlayer heating of which the elongations at break are better than baselines are 30%-30%-40% new-aged-extremely aged and 30%-40%-30% new-aged-extremely aged mixed blends. For the samples with interlayer heating, the largest elongation is from the 30%-40%-30% new-aged-extremely aged mixed blend (64.63% better than baseline), of which the elongation at break increases by 55.29% after interlayer heating (from 11.63% to 18.06%) for interlayer heating enhances bonding and microstructures.



(a) Without interlayer heating



(b) With 60 seconds interlayer heating

Figure 5: COMPARISONS OF SAMPLE ELONGATIONS AT BREAK

In addition, parts with high percentages of extremely aged powders have lower Young's modulus. Also, the Young's modulus of samples can be controlled consistently at the same level as the standard benchmark samples (50%-50% new-aged mixed blend) in the proposed method.

3. Conclusions

A comprehensive method was proposed in this paper to explore the possibility and feasibility of reusing the differently degraded polyamide 12 powders in different combinations, especially the extremely aged polyamide 12 powders close to the heat-affected zones. The proposed method was successfully applied to the practices of reusing reclaimed polyamide 12 powders into functional samples, with mechanical properties even superior to current industrial norms.

Tensile test results show that parts with higher tensile strength (e.g. 56.78% better than the baseline) and larger elongation (e.g. 35.30% better than the baseline) are obtained. In particular, the proposed method yields printed samples with 18.04% higher tensile strength and 55.29% larger elongation at break using as much as 30% of extremely aged powders compared to the benchmark sample. Besides, Young's modulus of samples is controlled consistently at the same level as the standard benchmark samples.

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