

Discussion on the microscale geometry as the dominant factor for strength anisotropy in material extrusion additive manufacturing

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Abstract

This paper presents a discussion and interpretation of the findings in the review paper “Fused filament fabrication of polymer materials: A review of interlayer bond” by Xia Gao, Shunxin Qi, Xiao Kuang, Yunlan Su, Jing Li, Dujin Wang [Additive Manufacturing (2020): 101658]. This discussion draws different conclusions based on the microscale filament geometry of interlayer bonds as opposed to molecular-scale bonding (diffusion and entanglement of polymer chains), which is predominantly considered in the review. Four complementary arguments on the matter are proposed, demonstrating that microscale geometry rather than incomplete molecular bonding is the predominant cause of strength anisotropy in material extrusion additive manufacturing (MEAM). These arguments consider the evidence from studies that (i) factored microscale geometry into strength calculation; (ii) eliminated the influence of geometry; (iii) improved the geometry to reduce its impact on strength, and (iv) tested the effect of manually reproduced interlayer geometry in bulk material. Overall, this discussion suggests that the underlying cause of anisotropy in MEAM is filament-scale geometric features (grooves and voids between layers), not the deficient bonding as is often theorised. Drawing upon the evidence in the literature, this discussion proposes that specimens attain bulk material strength for a range of printing conditions and materials.

1. Introduction

Material extrusion additive manufacturing (MEAM) is an advanced manufacturing method that produces parts via layer-wise addition of material. A vast number of variable manufacturing parameters enable MEAM to achieve geometrically complex structures, but this inherently introduces significant variability in geometrical and thermal outcomes, as identified by Gao *et al.* in this review [1]. The task of understanding how printing parameters influence mechanical properties is challenging due to their interdependence and the inherent complexity of the micro- and mesoscale geometries found in many printed specimens.

The primary conclusion of the review by Gao *et al.* [1] is that molecular diffusion and entanglement across the bond is the most important factor causing anisotropic mechanical properties. In particular, they concluded that mechanical anisotropy is “*dominated by interdiffusion and/or entanglements across the bond interface*” for amorphous polymers. For semi-crystalline polymers, they conclude that “*crystallization is the hindering factor for the molecular diffusion and entanglement and the partial melting of the strand surface is the driving factor*” for mechanical anisotropy.

However, the review overlooked an important factor that is missing in the conclusions - the effect of contact area between bonded extruded filaments on anisotropy. This discussion

presents the evidence to support a principally different interpretation of the literature, not presented in the review article, namely that:

- the contact area in the bonded region is the dominant factor governing mechanical anisotropy;
- the bond formation is sufficient to provide perfect bond strength in various polymeric materials manufactured with a broad range of printing conditions.

The evidence in the literature is broken down into four complementary arguments (each considered in a dedicated subsection below):

- *2.1 Factoring geometry into strength calculation* - When the influence of geometry was factored into strength calculation, bulk-material strength of interface was observed [2]–[10] (studies [2] and [5] in this discussion are studies [78] and [121], respectively, in the review).
- *2.2 Eliminating influence of geometry* - When the influence of geometry was eliminated, bulk-material strength was found [11]–[14] (studies [46], [71], [70] and [72], respectively, in the review).
- *2.3 Improving geometry* – Mechanical-performance improvements attributed to enhanced intermolecular diffusion could have been a result of changes in interlayer contact area [15]–[18] (studies [132], [164], [165] and [168], respectively, in the review).
- *2.4 Testing the effect of interlayer geometry* - When the interlayer geometry (surface grooves/voids) was manually applied to longitudinally tested specimens, isotropic mechanical properties were achieved [4], [5].

This body of evidence strongly supports a hypothesis of bulk-material strength of interfaces in MEAM rather than poor intermolecular bonding.

2. Evidence of bulk-material strength of interfaces

2.1. Factoring geometry into strength calculation

As the authors state in Section 4.2.1 of their review [1]: “*The contradictories in [the range of findings identified in the literature for the influence of printing speed on bond strength] arise from various characterization methods for characterizing the interfacial bond property, in which the test specimens had different geometries and therefore suffered from differing thermal history during FFF process*”. The authors attributed the differences in bond-strength results to the thermal history caused by variation in the overall size and shape of test specimens rather than the variation in filament-scale geometry. Instead of relating this feature to differences in terms of intermolecular entanglement and diffusion (that could also play their part under some circumstances, as discussed in Section **Error! Reference source not found.**), we believe that the most important interpretation of these contradictions is to justify the need for precise control and characterisation of bond geometry.

The role of filament-scale geometries, i.e., voids naturally occurring at the interlayer bond regions, is not currently accounted for by the geometrical measurement methods employed in many studies, which utilise external caliper measurements, as illustrated in the schematic (Fig. 1). Similarly, most studies referenced in the review [1] did not measure the contact area. This resulted in overestimation of contact (load-bearing) area and underestimation of stresses (due to a larger area used for strength calculation), which could risk being mis-interpreted as

evidence for poor intermolecular bonding. This risk can be overcome by a use of specially designed testing specimens for MEAM [3] and effective geometrical characterisation to delineate contact regions from voids. Experimental measurements of the interlayer contact area used to calculate strength showed that bulk-material strength of interlayer bonding was achieved. [2]–[5] (Fig. 3 (a)) This is also supported by FEA simulations by Tronvoll *et al.* [7], who stated that “for specimens of PLA, much of the anisotropic behaviour could be explained directly by the reduction in residual cross section”.

Also, Fang *et al.* [2], who studied polycarbonate, concluded that: “The strength and modulus change were correlated with geometry changes” and “the strength showed a linear relationship with the bonding area”. Even though extrusion temperature was varied from 230 °C to 290 °C, it had a minimal effect on bond strength (range 45 - 51 MPa; lowest and highest strengths were found at 260 °C and 240 °C, respectively). This suggests that the intermolecular diffusion process was sufficiently completed even at the lowest temperature and highlights the relative importance of geometry compared to temperature. It should be noted that extrusion temperature indirectly affects geometry due to changes in viscosity and extrusion rates [19], complicating the analysis of temperature effects in studies, in which the extrusion rate was not calibrated for each temperature.

Our studies for a different polymer, polylactide [3]–[5], [8]–[10], demonstrated that the interlayer bond had bulk-material strength for a range of layer heights (0.1 mm to 0.3 mm), extrusion widths (0.4 mm to 0.6 mm), and different printing speeds and layer times. It was found that load-bearing capacity was directly related to the respective cross-sectional area (Fig. 2) [3] (study [78] in the review), irrespective of the extruded-filament orientation (transverse or longitudinal), layer height or extrusion width of the specimens. This is evident from the two specimens with similar cross-sectional areas and load-bearing capacity (Fig. 2 (b) and (c)), even though one (Fig. 2 (a)) was testing longitudinally (i.e. along extruded filaments) and the other orthogonal to the interlayer bond (Fig. 2 (b)), This vividly demonstrates that the bond has the same strength as the bulk material, supporting our hypothesis. The layer time and printing speed also did not affect the bond strength [8], even though these parameters, along with layer height and extrusion width, would all have influenced the cooling rate and cyclic heating history. This, again, highlights the apparent insensitivity of bond strength to factors that would be critical for intermolecular diffusion suggesting that it was sufficient to achieve strength similar to that of the bulk material. We also found the evidence

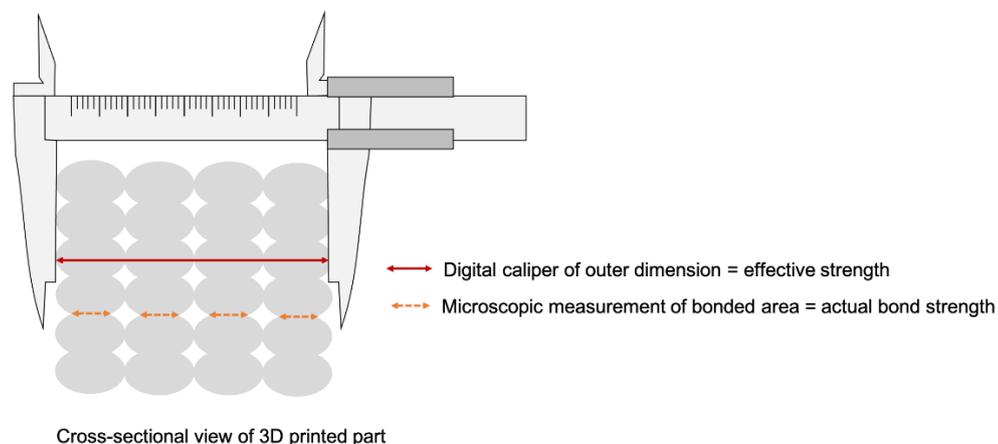


Fig. 1 Schematic of caliper measurement method for a 3D-printed part. This method measures the outer dimension, discounting the narrowing caused by inherent internal and surface void geometries, thus overestimating the load-bearing area, as shown by colour-coded arrows.

of bulk-material-strength bonding in MEAM when testing polylactide specimens in a range of environmental conditions (e.g. submersion in fluid media and at elevated temperatures) [10] and during degradation over 10 months [9]. In the latter case, transverse and longitudinal specimens had a similar evolution of strength (initially increasing before decreasing as degradation proceeded). The results in Fig. 2 (a) indicate that good bond strength can be achieved for a range of bond areas and, therefore, full-strength bonding does not depend on bond geometry even though specimens with lower interlayer contact areas may be expected to have suboptimal conditions for intermolecular diffusion.

Still for a different material - high impact polystyrene, Coogan and Kazmer [6] (study [121] in the review) measured the bond area for strength calculations and established that every specimen printed above the minimum recommended temperature (220 °C) had mean strengths between 89% and 111% of the virgin polymer. They also found these specimens to have complete interlayer diffusion in simulations. This study showed that bulk-material-strength interlayer bonding was present for a four-fold change in layer height, an almost two-fold change in extrusion width, a 50 °C change in extrusion temperature and a four-fold change in printing speed. All these factors would have affected intermolecular diffusion, adding further support to the findings above that the intermolecular diffusion process completes sufficiently for bulk-material interfacial strength under common manufacturing conditions, and that geometric changes to the bond width are the dominant factor affecting strength.

A study by Aliheidari *et al.* [20] (reference [142] in the review) provided several results, which highlight the importance of microscale geometry for fracture resistance of ABS. When the bond area in specimens was factored into calculations, the obtained data indicated that for several nozzle temperatures (220 to 240 °C), bed temperatures (85 to 105 °C) and extruded-filament widths (0.25 to 0.45 mm), the range of fracture resistances of the bond (3390 to 3910 J/m²) was close to the range reported in the study for bulk material (~3500 to 4000 J/m²). There were small variations in bond strength, which may indicate some impact of molecular diffusion, but these variations were minimal by comparison to the effect of microscale

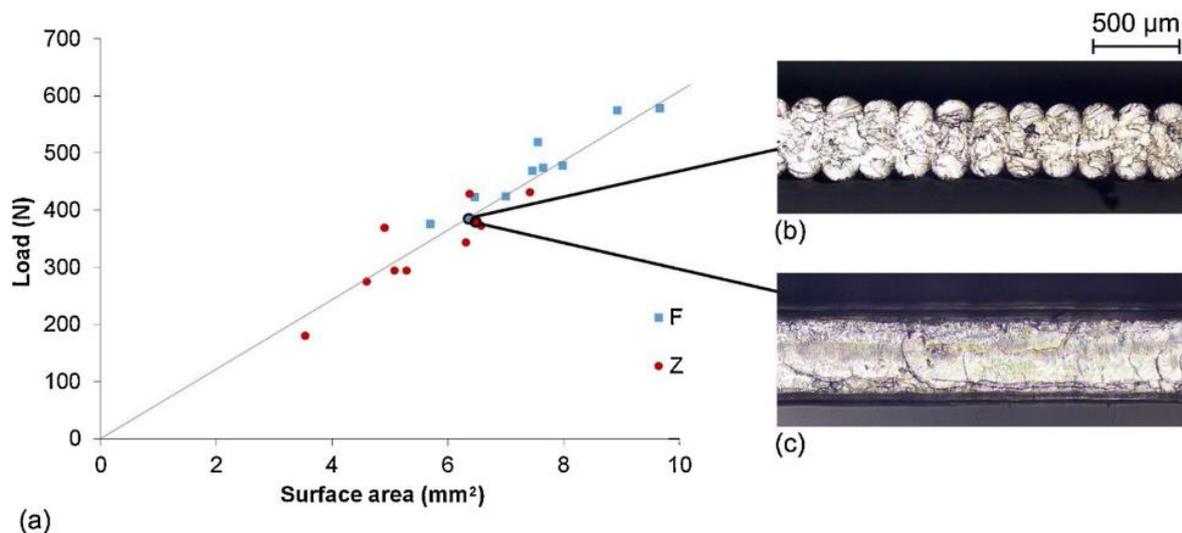


Fig. 2 Mean load-bearing capacity plotted against mean surface area for each specimen type in the study, including F (longitudinal orientation) and Z (transverse orientation). The inset micrographs show specimens with similar fracture surface areas and equivalent load-bearing capacity even though they were tested in different orientations and had different geometric designs [3]. Figures reproduced with permission (Elsevier, 2020).

geometry: when the bond area was considered in calculations, fracture toughness was obtained to be on average ~70% greater than that in calculations that did not consider the bond area. An important example is when the layer height was reduced from 0.3 to 0.2 mm, fracture resistance increased by approximately 33% when bond area was considered in calculations but increased by almost 200% when the bond area was not considered. This clearly demonstrates that it is critical to account for the real bond area in analysis of the bond's mechanical properties.

Other studies demonstrated that geometrical modifications, which increased the interlayer contact area, improved structural strength [3], [21] (studies [78] and [136], respectively, in the review). For ABS specimens, Coogan and Kazmer [21] showed that reduction of the layer height from 0.3 mm to 0.15 mm resulted in the contact-area increase from 60% to 88% and strength increasing from 23 MPa to 29 MPa. The filament strength in this study was measured at 36.3 MPa, meaning that the specimens with layer heights of 0.3 mm and 0.15 mm both had bond strengths comparable with that of the bulk material, when accounting for their relative contact area: 60% of 36.3 MPa = 21.8 MPa and 88% of 36.3MPa = 31.9 MPa, close to the recorded values of 23 MPa and 29 MPa, respectively). This evidence suggests that geometry influences the structural strength whereas bonding has bulk-material strength, irrespective of the geometry.

2.2. Eliminating influence of geometry

The review [1] found some studies [11]–[14] ([46], [71], [70] and [72], respectively, in the review) to have low mechanical anisotropy (<10%) for polyamide, polyethylene and polypropylene. This was attributed to better intermolecular diffusion than for other polymers. However, a common feature of these studies is that there was very low level of interlayer voids, as shown in Fig. 3 (b). This provides a geometrical explanation for the near-isotropic behaviour of these specimens. In cases of such low mechanical anisotropy (10%) and apparent structural homogeneity, digital caliper measurements were appropriate for geometrical characterisation. This also explains why these results indicated that the interlayer bond had perfect strength, a point not communicated in the review article. This evidence additionally supports the message presented in Section 2.1: specimens with voids required the bond area to be measured, whereas the lack of voids justifies the caliper measurements of the external geometry for adequate characterisation of the bond area. Regardless of whether voids are present or not, correct assessment of the real bonded area (in contrast to the supposed one) in the cross-sectional region of fracture (normal to the loading direction) resulted in the identification of bulk-material strength of interlayer bonding. It is not necessary to eliminate voids to achieve bulk-material-strength interlayer bonding.

2.3. Improving geometry

A number of studies identified by the review [1] demonstrated that interlayer mechanical performance was improved by chemical or post-process modifications [15]–[18] (studies [132], [164], [165] and [168], respectively, in the review). Whilst this improvement was hypothesised to be linked to intermolecular diffusion or crystallinity, it can be concluded that the improved mechanical performance was associated with an increasing contact area. Hence, such geometrical improvement may have been the true cause of interlayer's enhanced mechanical performance. One of the studies [18] (study [168] in the review) used a Diels-Alder reaction to improve the mechanical performance of PLA. The authors stated directly that the method was able to *“reduce the impact of mechanical defects introduced by the printing process”*,

acknowledging that the improvements observed were related to the enhanced geometrical outcome. This is supported by the microscopic observations and schematics (Fig. 3 (c)), demonstrating that the void volume was significantly reduced by the chemical treatment. Another study annealed ABS specimens [16] (study [164] in the review) and established that this procedure improved the interlayer toughness significantly. Inspection of the internal void geometry showed that annealing significantly altered the volume and geometrical characteristics of voids (Fig. 3 (d)): the voids were no longer aligned within an interlayer plane, removing material's anisotropy. The alignment of pores is also a critical issue for interlayer

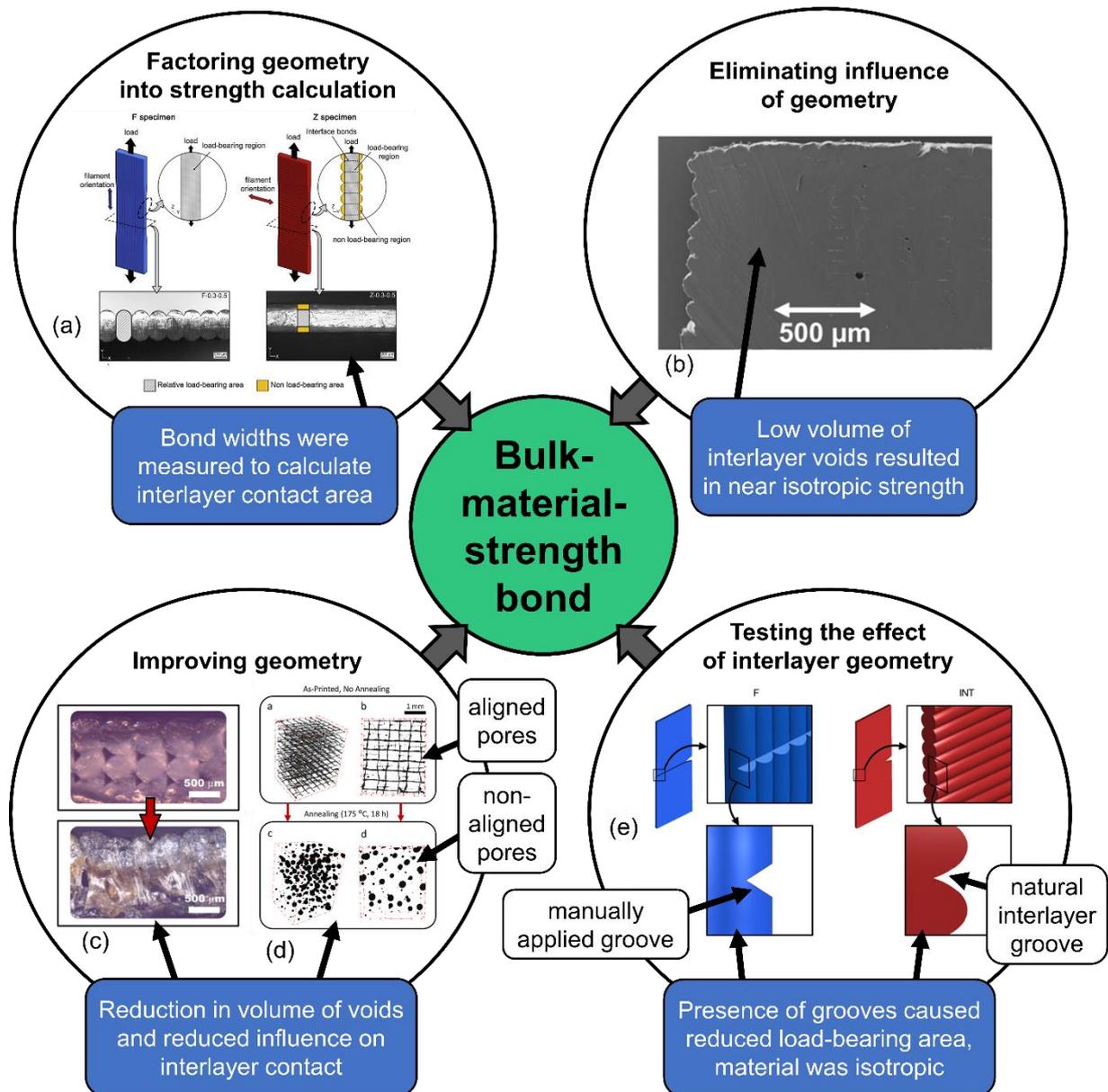


Fig. 3 Studies supporting our hypothesis of perfect bond strength: (a) a study factoring the actual geometry into strength calculation [3] (study [78] in the review); (b) a study eliminating the influence of geometry [12] (study [71] in the review); (c) and (d) studies with improved geometry [18], [16] (studies [168] and [164], respectively, in the review); (e) a study, which tested the effect of interlayer geometry by manually applying grooves to longitudinal specimens (left) to replicated interlayer geometry in transverse specimens (right) [4]. Figures modified and reproduced with permission (ACS publications, 2016, Elsevier, 2017, Elsevier, 2018, Elsevier, 2019, Elsevier, 2020.)

integrity, since it was found that a 12% void fraction can result in a 68% reduction in the interlayer's bond area [20] (study [142] in the review). A study using a bimodal blend of PLA [15] (study [132] in the review) concluded that it promoted "*stronger interfaces*" compared to a unimodal one. However, analysis of the microscopic geometries of the bimodal specimens compared to the 'neat' specimens demonstrated that the change in the PLA blend resulted in a significant reduction of voids, resulting in a near-homogenous internal structure of the specimen and, hence, improved mechanical performance. Similar geometrical changes are noted in a study utilising heat treatment to improve mechanical performance of polyphenylene sulphide [17] (study [165] in the review). In this work, based on microscopic analysis, the authors stated that "*increasing the print temperature decreased the appearance of roads in elevated temperature heat-treated samples*". This is further evidence that variation in manufacturing parameters affected in many cases geometry, bringing about the increase in strength.

2.4. Testing the effect of interlayer geometry

Another approach to probe the hypothesis about the effect of filament-scale geometrical features on interfacial strength was developed in [3] and [4]. Utilising specially designed specimens of polylactide, manual surface grooves (with dimensions similar to the ones naturally occurring in MEAM) were introduced in longitudinally loaded specimens to physically simulate the interlayer geometry in material without interlayer interfaces (i.e., regions with manufacturing-induced molecular diffusion) normal to the loading direction. These specimens performed similarly to transversally loaded specimens, i.e., with natural interlayer geometry (grooves between layers). This proved directly that the presence of grooves - either naturally occurring or manually generated - had a critical effect on mechanical performance rather than the physical entanglement and intermolecular diffusion of polymer chains across the interlayer bond. Specimens with similar contact areas had close load-bearing capacities and strengths, irrespective of the orientation of their extruded filaments. This clearly demonstrates that the bond strength was equivalent to that of the bulk filament (Fig. 3 (e)), indicating the critical role of microscale geometry.

2.5. Applicability of findings

This paper argues that there is a body of evidence indicating that real bond strength in MEAM is similar to bulk-material strength for a range of materials, printing conditions and microscale geometries. However, the authors acknowledge that there could be cases when deposited filaments does not have bulk-strength bonding. The causes of mechanical anisotropy in MEAM of polymers are complicated, and further research must explicitly consider bond geometry to build broad understanding about dominant factors in mechanical anisotropy and the conditions, under which these factors are most critical. The following non-exhaustive list highlights some factors that require further investigation, with explicit consideration of bond area, to determine whether they influence the attainment of bulk-strength intermolecular bonding:

- Material formulation (including composites);
- Size of the additive manufacturing hardware, extrusions and overall part;
- Broad thermal factors and other printing settings;
- Residual stresses;
- Printing defects and use of suboptimal printing parameters;
- Post-processing techniques.

3. Conclusions

The review article by Gao *et al.* [1] concluded that anisotropy in MEAM was caused by deficiencies of the intermolecular diffusion and entanglement, which resulted in poor bond strength. In contrast, this discussion presents the direct evidence that supports an alternative theory that the interlayer bond strength is similar to that of the bulk material and anisotropic strength is predominantly caused by geometric features - the interlayer grooves and voids between layers - reducing the contact (load-bearing) area (Fig. 3). It also suggests that studies, which did not identify the perfect bond strength, may have inadvertently overlooked the microscale grooves/voids in geometrical characterisation. The current typical measurement methods – assessing the outer geometry with digital calipers - cannot distinguish the bond and void regions, thus resulting in overestimation of the load-bearing area. This discussion proposes that future studies must characterise a bond area (e.g., with microscopy) for calculation of real bond strength. The overall (effective) structural strength of MEAM parts (affected by their internal structure with voids etc.) should be considered as a separate concept. It can be valuable for practical applicability but is not suitable for conclusions on the bond strength. Thus, the most important factor to investigate, control and improve mechanical performance in MEAM is the microscale geometry of the extruded material.

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