nature portfolio

Peer Review File

High performance plain carbon steels obtained through 3D-printing

Corresponding Author: Dr Qiyang Tan

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
This paper provides as interesting fashionable perspective, but this is not demonstrated by the presented evidence. The key premise of this paper is that alloys of relatively complex steels can be replaced by simple Fe-C alloys by adopting the layer-by-layer approach of additive manufacturing. However, this is not demonstrated. Components such as gears are subjected to conditions measured by engineering properties different to those shown in the paper. Ashby maps are too simple and to not reflect wear and contact fatigue. These are the real engineering properties to which to compare, rather than toughness, strength or ductility. As such the key premise of the paper is flawed and this reviewer believes is not suitable for this journal.

The paper demonstrates that different microstructures can be tailored and attained through rolling contact fatigue, but the link of microstructure with relevant properties is not there. As such, the presented information is not new.

Reviewer #2

(Remarks to the Author)
Tan et al. discussed the significant advancements in structural alloys over the past century, emphasizing improved mechanical performance but noting the downsides of increasingly complex alloy compositions, such as recycling challenges with higher costs. This research work shows that 3D-printed plain carbon steels can achieve attractive mechanical properties, such as tensile strength and elongation and impact toughness, comparable to, or even better than, more complex ultra-high strength steels and Maraging steels. By adjusting 3D printing parameters, the microstructure of the two carbon steels can be tailored for specific applications, offering new opportunities to reduce alloy complexity without sacrificing performance. This approach highlights the potential of 3D printing to drive the shift towards simpler, more efficient alloy compositions in metal processing.
While the move towards alloy "plainification" via laser powder bed fusion (L-PBF) printing is promising, it raises some concerns. Simplifying alloy compositions might lead to a trade-off in the long-term durability and specific performance characteristics that complex alloys are designed to provide. It is worthy noting that alloying elements are added in engineering alloys not only for enhancing strength and ductility or toughness, but the comprehensive performance including but not limited to processability, corrosion, oxidation, etc. While plain carbon steels may perform well in controlled tests, their real-world application could reveal limitations in environments where advanced alloys typically excel. Also, the reliance on L-PBF printing for achieving desirable microstructures, such as martensite or bainite, could also introduce inconsistencies due to variations in printing conditions (process parameters, printing layout, and scale), potentially compromising material quality.
Though there are some deficiencies in language and lacking universality, this is a quality work presenting plenty of L-PBF printing outcomes and beautiful microstructural characterization. Two carbon steels were additively manufactured using L-PBF with varying process parameters mainly via low to high energy densities. However, it is quite known regarding the correlations between energy density and resultant martensitic or annealed microstructure, leading to strength-ductility trade-off in tensile performance for a wide range of engineering alloys in the community. It sounds that the authors exaggerate the point of “plainification” by making use of established L-PBF printing knowledge. This work should surely be published in Acta Materialia, but may be debatable to publish in Nature series journals.

Reviewer #3

(Remarks to the Author)
The approach of plainification is very interesting and relevant. The experiments carried out are basically suitable to underline the core statements that even with a small number of alloying elements, components with very good properties can be achieved by 3D printing.

However, the structure of the document makes it hard to follow through, and there are also some weaknesses in the discussion.

As the data itself is convincing and mostly sufficient, I recommend a revision of the document to improve readability and representation of the results, and address the flaws listed in the following:

Abstract:
- Inconsistencies between 3D printing / 3D-printing

Main:
- The authors claim (line 50) that there is "currently an active discussion about 'plainification'". This is not supported by the choice of references, being from 2019. An active discussion would yield more recent literature? Thus, calling it a "movement" might be a bit of an exaggeration here.
- 3D-printing is rather a group of technologies than "one" technology (line 74)
- LPBF is an outdated term (see: ISO/ASTM 52900:2021, Annex)
- The range of 10^2 to 10^5 K/s is not the typical one for LPBF (line 88), more recent studies show even 10^6-10^7 K/s
- Any comment on the powder? It seems like there are many satellites.
- Line 138f: how was the sample's build direction? This might be relevant for the discussion on stresses. Isn't the in-situ tempering effect contradictory to the initial thoughts about super high cooling rates being beneficial for martensite? This is dealt with a bit too superficially here.
- Why were the tensile test samples not tested along the build direction which is known to be the weakest, and thus most crucial?
- The comparison of wrought and 3d-printed steel are a bit difficult to understand. Was the wrought material also heat treated, or do you compare different conditions? Wouldn't it make sense to compare also 3d-printed medium or high-alloyed steels? How would you comment on the necessity of heat treating the 3d-printed material after the build in general? What changes are to be expected? Why was the as-built condition only considered in the discussion for the lower alloyed steel? The idea of this approach is not straight forward. The provided Supplementary shows that the complete data does exist. However, the conclusions in the main part refer partially to shown and partially to not shown results. It is claimed that the 3d-printed parts outperform the wrought material, which is a rather daring statement given the fact that it is not clear which data and condition are compared, respectively, and a beneficial test direction was selected.
- The authors might want to consider to swap the microstructure part with the mechanical test part, as the discussion on the mechanical test results and also the reasoning for discussion certain materials seems to require knowledge about the microstructure beforehand. This would make the whole manuscript clearer and easier to follow through. A very clear order could be: processing (complex geometry, density, cracking), microstructure, hardness (correlated with microstructure as well), mechanical properties, comparison with state-of-the-art steels.
- Also, certain parts are repeated throughout the manuscript, e.g., it is referred back and forth and to supplementary quite frequently, and also parts of methods are explained.
Imaging:
- In general: inconsistency in used font and size, also within one Figure (e.g., Figure 1)
- Figure 1b) why does the scale bar (color code) cover a range that is not present in the measurements, making it harder to visually see local differences? Figure 1d is misleading, even though it's just schematically. The overlapping of the melt tracks is no correct, neither in y/x nor z direction, and also does not fit with the described scanning strategy. Fig1i) It's not clear to which parameters the given Energy density refers, especially as some values could be achieved by different parameter combinations (power, speed) given in Table 1.
- Supplementary Figure S2: It is not clear which samples are built with 300 and 600 mm/s, which might be interesting in terms of hardness. Again, the volume energy density does not provide sufficient information, and the scan speed seems also wrong in either a or b.
- Figure 3 and 4: rather low resolution (might be due to PDF version, should be ensured to be sufficiently high in final version)
- Figure 4k: The nanoparticles are really hard to see/identify in contrast to the surrounding material. A larger image might be beneficial.

Methods:
- Precise Version of ImageJ?

Further:
- Inconsistencies in references, e.g., line 407
- Grammar (e.g., line 351) : "...of metal 3d-printing technology" -> technologies or "this/the...technology" or (line 533): „ We performed Jominy End Quench Test to evidence the low hardenability…" -> the …test or tests

- Grammar (line 544: „To demonstrate that 3D-printing also enables avoiding of quenching cracking and distortion of plain carbon steels. We prepared…"

Reviewer #4

(Remarks to the Author)
This work systematically investigated microstructure evolution and mechanical performance of 3D-printing plain carbon steels. They found that the sequential nature of the micro-scale melting and solidification made plain carbon steels achieve tensile and impact properties comparable to, or even superior to those of low and medium alloy ultra-high strength steels and some Maraging steels, and demonstrated that 3D printing was a reliable approach to reduce alloy complexity without compromising performance. These findings are interesting and constructive, and it is worth to be published at Nature communications. I suggest authors address the following comments before publication.
(1) Tensile and impact properties of plain carbon steels and other alloying steels are compared in this work. Fracture toughness is also an important index for ultra-high strength steels. Is the fracture toughness of 3D-printing plain carbon steels comparable to the alloying steels?
(2) One important point is that increasing laser energy density can make a martensite-dominant microstructure transformed into bainitic-dominant microstructure, which is attributed to the decreased cooling rate. Increasing laser energy density not only enlarges melting pool size, but also accelerates convection. Both of them could play a vital role in the phase transition path. I suggest that authors could provide a quantitative analysis by finite element simulation or thermodynamic calculation to reveal the relationship between thermal history and phase transition.
(3) L-shape demonstration parts fabricated by 3D-printing and common quenching are compared in Figs. 1(g) and (h). Please list the laser energy density used in 3D-printing. Is the microstructure martensite or bainite? Besides, authors consider that the cyclical thermal profile from the melting of successive layers provides an in-situ tempering effect to reduce the thermal residual stress, leading to the 3D-printing part without cracking. However, the ultrafast heating and cooling always generate high residual stress during AM. I notice that 200 °C preheating is used during AM. Maybe the real reason is the preheating lowers cooling rate, thereby weakening thermal residual stress.
(4) 3D-printing samples always have typical cell structures, i.e., element segregation. Are there cell structures within two kinds of plain carbon steels? What are their effects on mechanical properties? It could be interesting to provide more discussions.

Reviewer #5

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 1:

Reviewer comments:

Reviewer #3

(Remarks to the Author)
The authors have responded convincingly to all my comments. Hence, I recommend acceptance of the manuscript.

Reviewer #4

(Remarks to the Author)
In general, the authors have well addressed the reviewers' comments. I trust the revised manuscript can be accepted for publication now.

Reviewer #5

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response to the Reviewers Comments

We are very grateful to the reviewers for their time and comments on our manuscript. We are sure that by addressing these comments we have significantly improved the article.

In the following, we summarise the comments of each reviewer and directly below each comment we explain how we have addressed the comment. The reviewer's comments are in *black italics*, our responses to the comments are in blue and yellow highlighted text indicates the new text we have modified in the revised manuscript. This modified text is also highlighted in yellow in the revised manuscript for ease of locating.

We begin by addressing the comments of Reviewer #3. This reviewer suggested helpful, but also significant, structural changes to the order of the content presented in the manuscript. We present this first for ease of understanding. We then address the comments of Reviewers 2 and 4&5 (who provided a combined review). We address the comments of reviewer #1 last, since his/her view of the manuscript is very different to the other reviewers.

Reviewer 3

The approach of plainification is very interesting and relevant. The experiments carried out are basically suitable to underline the core statements that even with a small number of alloying elements, components with very good properties can be achieved by 3D printing.

However, the structure of the document makes it hard to follow through, and there are also some weaknesses in the discussion.

As the data itself is convincing and mostly sufficient, I recommend a revision of the document to improve readability and representation of the results, and address the flaws listed in the following:

We appreciate the reviewer's supportive words and constructive suggestion.

1. Abstract:

1.1- Inconsistencies between 3D printing / 3D-printing

This inconsistency has now been corrected throughout the text.

2. Main:

2.1- The authors claim (line 50) that there is "currently an active discussion about 'plainification'". This is not supported by the choice of references, being from 2019. An active discussion would yield more recent literature? Thus, calling it a "movement" might be a bit of an exaggeration here.

We agree that the term 'movement' should be replaced. The sentence on Line 49 on Page 2 was revised in the following manner:

As a result, there has been growing interest in the 'plainification' of engineering alloys – a concept promoting a much smaller number of simpler compositions that can be used across many applications, with consequential benefits for recycling, re-use and security of supply[9,10].

We have also removed the term 'movement' from the Abstract (line 23) and rewritten the relevant sentence as:

The idea of moving away from a large number of complicated alloy compositions towards a smaller number of simpler compositions is known as alloy 'plainification' and is driven by the sustainability pressures of our resources.

2.2 - 3D-printing is rather a group of technologies than "one" technology (line 74).

The sentence on Line 74 on Page 2 was corrected and now reads:

Whilst not a new group of technologies, their extensive uptake by the industry is relatively recent[13].

2.3 - LPBF is an outdated term (see: ISO/ASTM 52900:2021, Annex).

The term 'LPBF' has been corrected to 'PBF' throughout the paper.

2.4 - The range of 10^2 to 10^5 K/s is not the typical one for LPBF (line 88), more recent studies show even 10^6-10^7 K/s.

The cooling rate values for PBF have been updated in the sentence on Line 89 on Page 2:

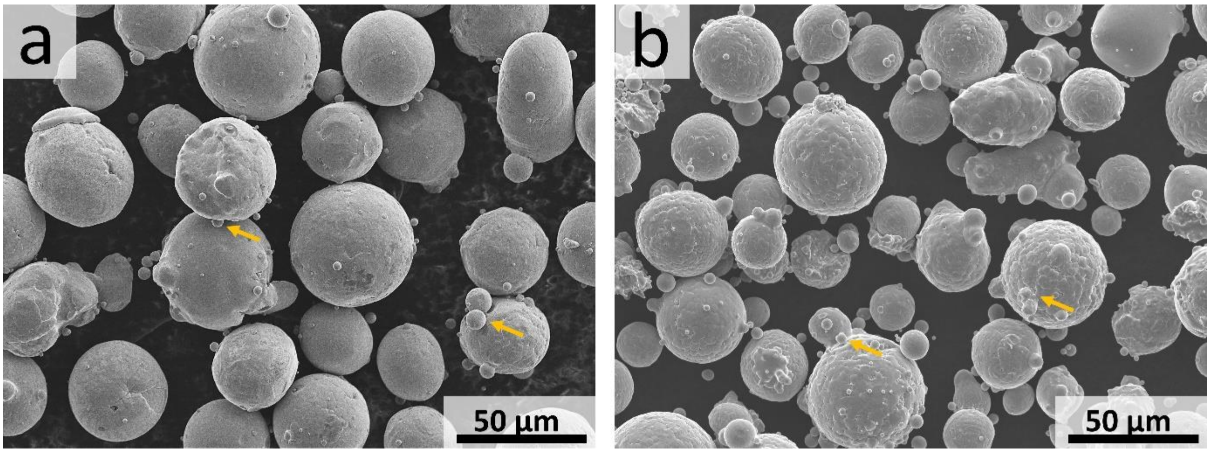
PBF is renowned for its direct fabrication of geometrically complex components and rapid cooling (10^4 to 10^7 K/s) of the melt pools (Fig. 1d)[13,16].

An updated reference (ref 16) on the cooling rate has also been included:

16. Su, J. et al. Recent innovations in laser additive manufacturing of titanium alloys. *Int. J. Extreme Manuf.* 6, 032001 (2024).

2.5 - Any comment on the powder? It seems like there are many satellites.

An additional figure (Extended Data Fig. 1) has now been included on Page 14 to better illustrate the morphology of the powder feedstocks.



Extended Data Figure 1: *Powder morphology of (a) pure iron and (b) AISI 1080 steel, with arrows indicating small satellites attached to the surface of the spherical powders.*

The following paragraph was added to Line 410 on Page 11.

SEM micrographs (Extended Data Fig. 1) reveal that both feedstock powders have numerous satellites on their surfaces. This satelliting effect is commonly observed in gas-atomized metal powders, resulting from collisions between small particles and incompletely solidified larger droplets[51]. Despite this, the powder showed excellent flowability during the printing process.

2.6 - Line 138f: how was the sample's build direction? This might be relevant for the discussion on stresses. Isn't the in-situ tempering effect contradictory to the initial thoughts about super high cooling rates being beneficial for martensite? This is dealt with a bit too superficially here.

We appreciate the reviewer's comment. The samples that were discussed at Line 138 were built horizontally. We have added the following sentence on Line 467 on Page 13 (Method):

The sample was built horizontally with its wall (i.e., the thickness side) attached to the substrate.

We understand the reviewers' question about cooling rates and self-tempering, and whether these are contradictory.

To form martensite and/or bainite, we need to cool quickly from high temperatures (above ~800C in these alloys) where the material is FCC austenite, to below the martensite or bainite start temperatures, without the formation of allotriomorphic ferrite or pearlite. This is what the high cooling rate of PBF provides, without the need for lots of extra alloying elements. Certainly, once formed, the regions will undergo some self-tempering due to the cyclic thermal profile upon PBF. However, this self-tempering does not get to a high enough temperature to revert the martensite back to austenite [*Yin, Y. et al. Laser additive manufacturing of steels. Int. Mater. Rev. 67, 487-573 (2022)*] – it really does just temper the martensite or bainite and this is actually a desirable effect (it helps relieve any residual or transformation stresses). Therefore, the in-situ tempering effect does not contradict the the need for the high cooling rates to form the martensite in the first place.

To help clarify this point, additional sentences were added on Line 147 on Page 4.

Notably, while high cooling rates in 3D-printing are essential for martensite and/or bainite formation, the subsequent self-tempering from the cyclic thermal profile does not reach temperatures high enough to revert martensite or bainite to austenite. Instead, it tempers the microstructure, relieving transformation and residual stresses without compromising the benefits of rapid cooling[14]. Thus, the in-situ tempering complements the high cooling rates, enhancing the mechanical performance of the 3D-printed parts.

2.7 - Why were the tensile test samples not tested along the build direction which is known to be the weakest, and thus most crucial?

We have also tested samples along the build direction. These materials were quite easy to print with high levels of densification and no lack of fusion or similar defects were observed. In addition, since our martensitic and/or bainite microstructures form by a solid-state phase transformation from the austenite, we observe much less anisotropy between vertical and horizontal builds than perhaps people are used to with PBF metals.

As discussed on Pages 10-11, the martensitic and bainitic transformations in our 3D-printed plain carbon steels after solidification create multiple α'-blocks with different crystallographic orientations within a single prior austenite grain. This effectively counteracts the typical columnar structures and texture that usually forms during directional solidification. Consequently, our 3D-printed plain carbon steels do not exhibit strong anisotropy typically seen in other 3D-printed alloys that do not show extensive solid state phase transformations.

We have added the vertical tensile test data on the 1040 steel to the manuscript, demonstrating uniform and isotropic properties. These results are detailed in an additional section (Supplementary Section 9) with an accompanying figure (Supplementary Fig. S10) and additional Table (Supplementary Table S2) provided in the Supplementary Information on Page 12.

Supplementary Section 10. Evaluation of the anisotropy in mechanical properties

To further assess property anisotropy in the 3D-printed plain carbon steels, additional tensile tests were conducted along the vertical direction (i.e., the build direction) on the 1040 steel. Fig. S10 compares the engineering stress-strain curves of the as-printed 1040 steels in both vertical and horizontal directions. The determined YS, UTS, El, and the corresponding calculated anisotropy ratios are summarized in Table S2. Except for a moderate anisotropy observed in the low-energy-produced samples, the samples produced with medium and high energy inputs exhibited marginal anisotropy in the tensile properties. This result demonstrates that microstructural refinement through martensitic and/or bainitic phase transformations reduced the anisotropy, effectively addressing the common issue of columnar structures in 3D-printed alloys.

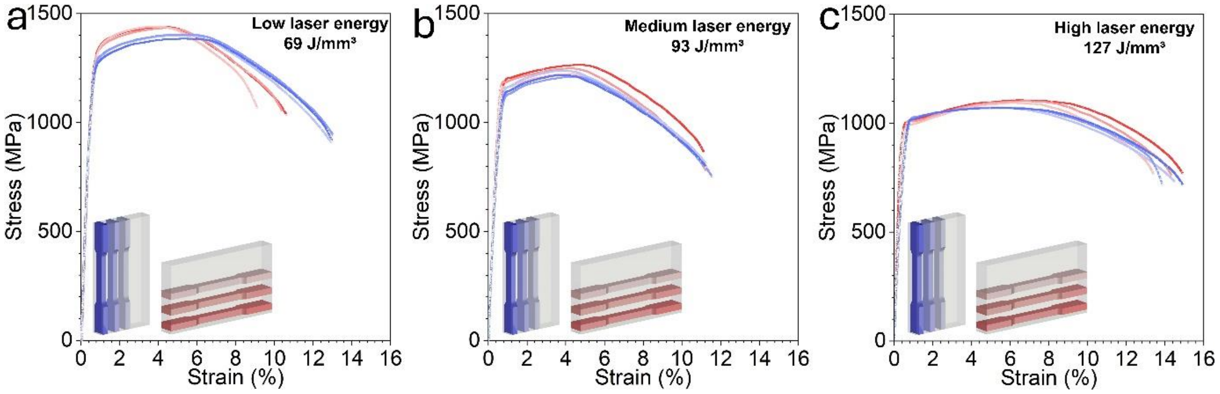


Figure S12: Representative engineering tensile stress-strain curve of the 3D-printed 1040 steel with (a) low laser energy of 69 J/mm^3, (b) medium laser energy of 93 J/mm^3 and high laser energy of 127 J/mm^3 along both horizontal and vertical directions.

Table S2. Mechanical properties of 3D-printed 1040 steels in different directions, with calculated anisotropy ratios. Note that anisotropy ratios were not calculated when property variations fell within the error margins.

Dominant phase	**Direction**	**YS (MPa) Anisotropy ratio (%)**	**UTS (MPa) Anisotropy ratio (%)**	**El (%) Anisotropy ratio (%)**
Martensite & bainite (low *E*)	Horizontal	1335 ± 5	1430 ± 5	10.2 ± 0.9
	Vertical	1267 ± 15	1397 ± 10	12.9 ± 0.2
	Anisotropy ratio	-5%	-2%	+26%
Bainitic dominance (medium *E*)	Horizontal	1177 ± 15	1247 ± 25	11.3 ± 0.1
	Vertical	1125 ± 23	1223 ± 16	11.5 ± 0.1
	Anisotropy ratio	-4%	-	-
Complete bainite (high *E*)	Horizontal	1000 ± 7	1100 ± 7	14.3 ± 0.8
	Vertical	992 ± 10	1070 ± 2	14.4 ± 0.6
	Anisotropy ratio	-	-3%	-

Several sentences were also added at Line 381 on Page 11 of the Main text to explain these vertical tests to the reader:

These transformations create multiple α'-blocks with different crystallographic orientations within a single prior austenite grain, effectively counteracting the typical columnar structures and texture formed during directional solidification[14]. The resulting ultrafine, almost texture-free microstructures of our 3D-printed plain carbon steels (Figs. 2 and 3) are testament to this mechanism. This microstructural refinement also contributes to the reduction in property anisotropy, addressing a common issue in 3D-printed alloys with columnar grains, as demonstrated by Supplementary Fig. S10 which compares the tensile responses of horizontally and vertically printed samples.

Several sentences were also added in the Method section on Line 514 on Page 14:

To evaluate the property anisotropy, additional tensile tests were conducted along the vertical direction (i.e., the build direction) on the as-printed 1040 steel. Tensile dog-bone samples with the same dimensions were sectioned vertically from the initially fabricated blocks measuring 25 mm (width) × 12 mm (thickness) × 40 mm (height).

2.8 - The comparison of wrought and 3d-printed steel are a bit difficult to understand. Was the wrought material also heat treated, or do you compare different conditions? Wouldn't it make sense to compare also 3d-printed medium or high-alloyed steels? How would you comment on the necessity of heat treating the 3d-printed material after the build in general? What changes are to be expected? Why was the as-built condition only considered in the discussion for the lower alloyed steel? The idea of this approach is not straight forward. The provided Supplementary shows that the complete data does exist. However, the conclusions in the main part refer partially to shown and partially to not shown results. It is claimed that the 3d-printed parts outperform the wrought material, which is a rather daring statement given the fact that it is not clear which data and condition are compared, respectively, and a beneficial test direction was selected.

We really appreciate these questions. The questions are answered one by one as follows:

Was the wrought material also heat treated, or do you compare different conditions?

The data for the wrought alloy steels were primarily retrieved from the ASTM Handbook, where the commercial steels have undergone their standard heat-treatment procedures to achieve the properties required for practical applications. This comparison data is really from commercial steels in the state used for practical applications.

To clarify this, a sentence was revised on Line 329 on Page 10.

A comparison with conventional wrought alloy steels (Figs. 4e and f) demonstrates that the 3D-printed plain carbon steels exhibit strength-ductility-toughness combinations comparable with many highly alloyed ultra-high strength steels (UHSS) after standard heat treatments, such as 4340, 8640 and 300M low alloy steels, and H11 and H13 medium alloy steels[27], and are even close to maraging steels[27,35].

Wouldn't it make sense to compare also 3d-printed medium or high-alloyed steels?

The comparison between our 3D-printed carbon steel with other 3D-printed alloyed steels is shown in Fig. 4g. This shows that the plain carbon steels perform just as well as the 3D-printed alloy steels, further demonstrating that potential exists for 'plainification' of some alloy grates, for some applications.

How would you comment on the necessity of heat treating the 3d-printed material after the build in general? What changes are to be expected?

Why was the as-built condition only considered in the discussion for the lower alloyed steel? The idea of this approach is not straight forward.

We consider these two comments together because they are in some ways related.

Normally, 3D-printed metals are given, at least, a stress relief heat treatment after printing and before putting into service. We have added a sentence to Line 304 on Page 9 to highlight this.

3D-printed materials typically require post heat-treatments to release the residual stress and homogenize the microstructure, thereby optimize strength-ductility trade-off[13].

While we performed mechanical testing on both the as-printed and as heat-treated 1040 steel (Method), we found that heat treatment had a limited effect on its mechanical performance. The as-built sample already exhibited a good strength-ductility-toughness trade-off, due to the in-situ tempering effect (as discussed on Line 263 in the Supplementary Information). Given that heat treatment is usually required for 3D-printed materials (as per our previous response), this is an additional benefit of the 1040 steel. Therefore, the mechanical properties reported here are based on the as-built condition.

To clarify this, several sentences were revised/added on Line 276 on Page 8.

For the 1040 steel, we found that good strength-ductility-toughness trade-off can be achieved directly after 3D-printing, alleviating the need of heat treatment. This is due to the auto-tempering effect typical of bainitic transformation (Supplementary Section 8). Thus, the mechanical performance of 1040 steel discussed here is focused on the as-printed condition.

The provided Supplementary shows that the complete data does exist. However, the conclusions in the main part refer partially to shown and partially to not shown results.

As mentioned on Line 268 on Page 8, the tempering temperatures used in our study were not optimized but were intended to demonstrate the potential for further improvement in the mechanical performance of 3D-printed plain carbon steels. Hence, we presented only selected data in Fig. 4 to effectively illustrate the range of the achievable properties of our steels whilst provided full data in Supplementary Table S1. This presentation also aims to avoid excessive data overlap, which can make reading the figures difficult. This is the balance we have tried to obtain in writing the paper – having all the data there, but also keeping the figures sufficiently simple that the main message comes across easily to most readers.

To clarify this, one sentence was added in the caption of Fig. 4 on Line 325 on Page 9.

Note that the data of the 3D-printed plain carbon steels in these Ashby plots were selected from Supplementary Table S1 to effectively illustrate the distribution of the achievable properties while avoiding excessive data overlap. The full data for all properties at different processing conditions is listed in Supplementary Table S1.

It is claimed that the 3d-printed parts outperform the wrought material, which is a rather daring statement given the fact that it is not clear which data and condition are compared, respectively, and a beneficial test direction was selected.

As noted in our response above, the data for the comparison wrought alloy steels were after their standard heat treatment procedures. Additionally, our 3D-printed plain carbon steels exhibit near-isotropic properties, indicating that there is no advantageous test direction. We appreciate that adding the vertical test data to the manuscript was really worthwhile to help address this question that other readers may also share. We feel that the claim that the 3D-printed plain carbon steels are comparable or even superior to some wrought alloy steels is a reasonable claim given the data presented. We have sourced comparison wrought data in the most beneficial commercial and practical states to maximise the validity of the comparison.

2.9 - The authors might want to consider to swap the microstructure part with the mechanical test part, as the discussion on the mechanical test results and also the reasoning for discussion certain materials seems to require knowledge about the microstructure beforehand. This would make the whole manuscript clearer and easier to follow through. A very *clear order could be: processing (complex geometry, density, cracking), microstructure, hardness (correlated with microstructure as well), mechanical properties, comparison with state-of-the-art steels.*

We very much appreciate the reviewer's suggestion. We have now reorganized the order of the paragraphs to follow a Processing-Microstructure-Properties sequence, which we believe makes the manuscript clearer and easier for readers to follow. For brevity, these changes are not detailed in the Response Letter; please refer to the revised manuscript for the updates.

2.10 - Also, certain parts are repeated throughout the manuscript, e.g., it is referred back and forth and to supplementary quite frequently, and also parts of methods are explained.

The repetition and frequent references back and forth were largely due to the original structure of the paper. We have now revised the manuscript to follow a clearer structure (as suggested by Reviewer #3) and conducted a thorough review to eliminate redundancy. We are very grateful to Reviewer #3 for this really helpful suggestion.

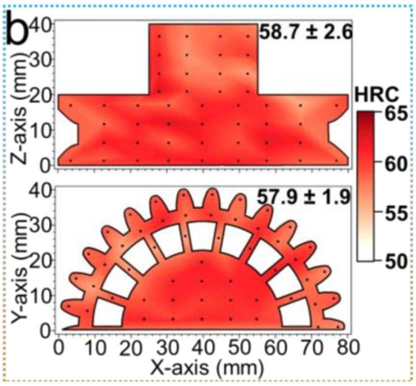
3. Imaging:

3.1 - In general: inconsistency in used font and size, also within one Figure (e.g., Figure 1).

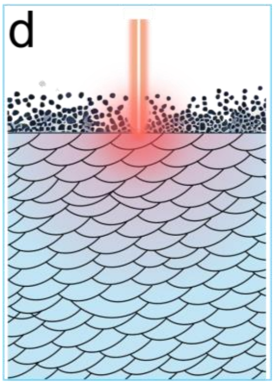
We have revised the figures to achieve consistency throughout. However, achieving uniform font size across all sub-figures is challenging due to the varying dimensions of each sub-figure, such as the scale tick labels in Figs. 1b and 1f. Nevertheless, we have ensured that the font size in all figures remains readable and clear. For brevity, these changes are not detailed in this Response Letter; please refer to the revised manuscript for the updates.

3.2 - Figure 1b) why does the scale bar (color code) cover a range that is not present in the measurements, making it harder to visually see local differences? Figure 1d is misleading, even though it's just schematically. The overlapping of the melt tracks is no correct, neither in y/x nor z direction, and also does not fit with the described scanning strategy. Fig1i) It's not clear to which parameters the given Energy density refers, especially as some values could be achieved by different parameter combinations (power, speed) given in Table 1.

We appreciate these questions. Regarding Fig. 1b, we have narrowed the colour scale bar to 50-65 HRC to more effectively highlight the local variations in hardness.



Regarding Fig. 1d, we have revised the schematic melt pools based on our previous experimental observations and modelling to avoid the misleading.



Regarding Fig. 1i, we agree with the reviewer that the specific processing parameters are important for readers to understand the context of the energy density values. However, including all this information in a single figure could obscure its clarity. Therefore, we have created an additional table (Extended Data Table 2) on Page 15 that lists the energy density alongside its corresponding processing parameter set.

Extended Data Table 2. The laser energy density corresponding to each processing parameter set. Note that the layer thickness (t) and the hatch space (h) were fixed at 0.03 mm and 0.12 mm, respectively.

Process parameter set	**Laser energy density, *E***
P = 100 W, v = 600 mm/s	46 J/mm^3
P = 125 W, v = 600 mm/s	58 J/mm^3
P = 150 W, v = 600 mm/s	69 J/mm^3
P = 175 W, v = 600 mm/s	81 J/mm^3
P = 200 W, v = 600 mm/s	93 J/mm^3
P = 225 W, v = 600 mm/s	104 J/mm^3
P = 250 W, v = 600 mm/s	116 J/mm^3
P = 275 W, v = 600 mm/s	127 J/mm^3
P = 300 W, v = 600 mm/s	139 J/mm^3
P = 325 W, v = 600 mm/s	150 J/mm^3
P = 350 W, v = 600 mm/s	162 J/mm^3
P = 375 W, v = 600 mm/s	173 J/mm^3
P = 350 W, v = 400 mm/s	243 J/mm^3
P = 375 W, v = 400 mm/s	260 J/mm^3

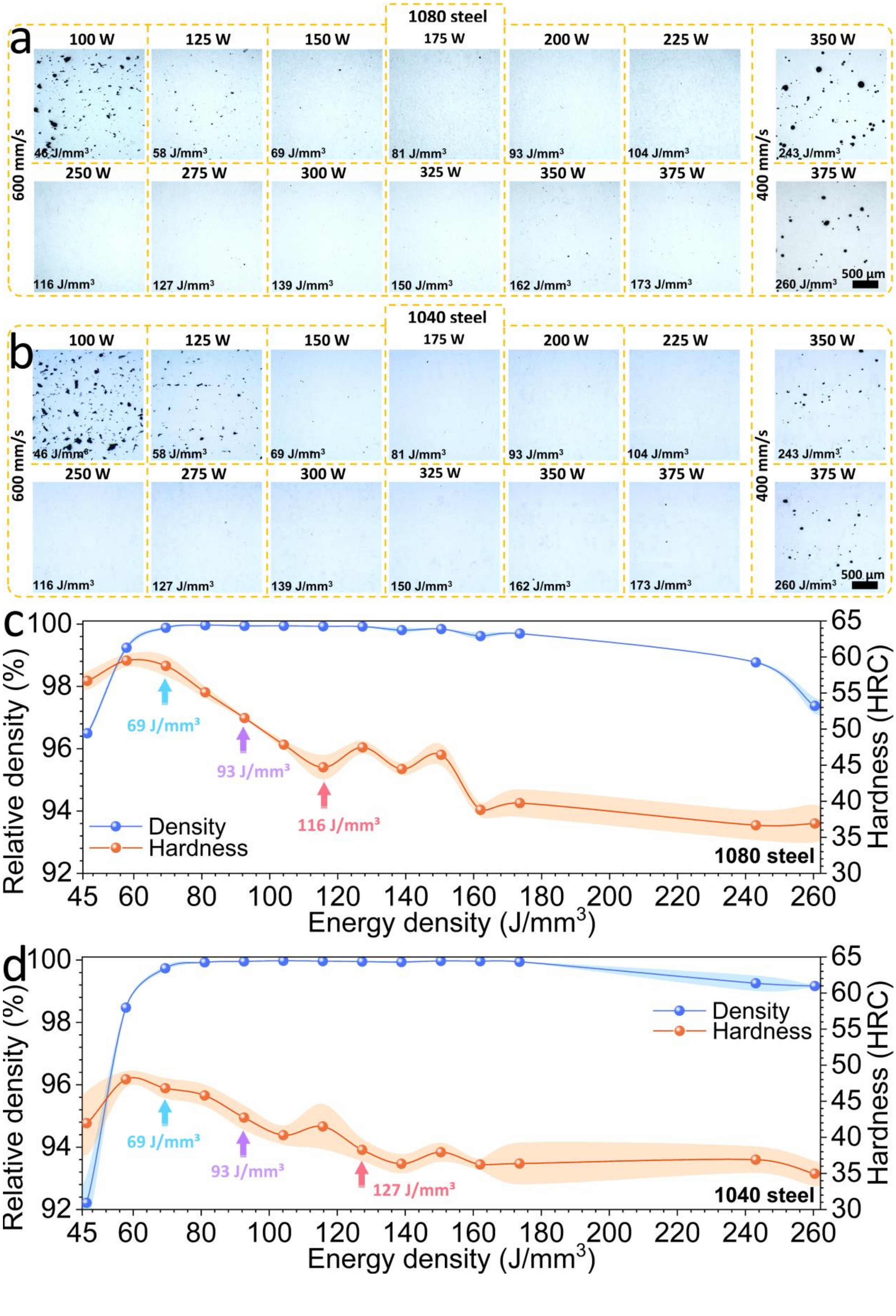
Accordingly, one sentence was added on Line 421 on Page 12.

These variables were integrated into the volumetric energy density (E, J/mm^3), with $E = \frac{P}{vht}$ for easier comparison, as listed in Extended Data Table 2.

3.3 - Supplementary Figure S2: It is not clear which samples are built with 300 and 600 mm/s, which might be interesting in terms of hardness. Again, the volume energy density does not provide sufficient information, and the scan speed seems also wrong in either a or b.

As mentioned in the above response (3.2), we have updated the laser energy densities corresponding to the processing parameter sets in Extended Data Table 2. Additionally, to enhance readability, we have updated the sample information and energy density details in Figs. S2a and S2b, making it easier for readers to connect this information with Figs. S2c and S2d.

We have also double checked the scan speeds in Figs. S2a and S2b to ensure they are accurately presented.

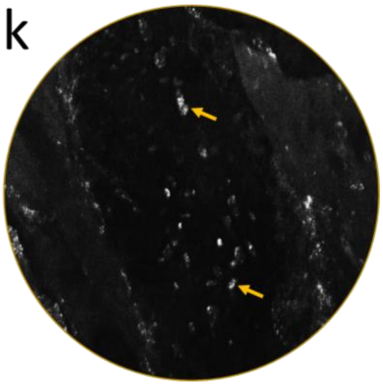


3.4 - Figure 3 and 4: rather low resolution (might be due to PDF version, should be ensured to be sufficiently high in final version)

The low resolution of the figures is due to the image compression of the online PDF conversion. We will ensure high-resolution images are uploaded and published (if the paper is ultimately accepted).

3.5 - Figure 4k: The nanoparticles are really hard to see/identify in contrast to the surrounding material. A larger image might be beneficial.

We have enlarged this figure (Fig. 3k in the revised paper) to ensure a higher readability as shown below.



4. Methods:

4.1 - Precise Version of ImageJ?

The ImageJ version has been provided on Line 428 on Page 12.

…using the ImageJ software (Version 1.52a)…

5. Further:

5.1 - Inconsistencies in references, e.g., line 407

The reference list has been thoroughly checked and corrected to ensure consistency, following the standard Nature Communications referencing style.

5.2 - Grammar (e.g., line 351) : "...of metal 3d-printing technology" -> technologies or "this/the...technology" or (line 533): „ We performed Jominy End Quench Test to evidence the low hardenability…" -> the …test or tests

- Grammar (line 544: „To demonstrate that 3D-printing also enables avoiding of quenching cracking and distortion of plain carbon steels. We prepared…"

These have been corrected as follows:

Line 376: "…of metal 3D-printing technology" was revised to "…of metal 3D-printing technologies".

Line 439: "We performed Jominy End Quench Test…" was revised to "We performed the Jominy End Quench Test…"

Line 461: "To demonstrate that 3D-printing also enables avoiding of quenching cracking and distortion of plain carbon steels. We prepared…" was revised to "To demonstrate that 3D-printing also enables avoiding of quenching cracking and distortion of plain carbon steels, we prepared…"

The manuscript receives an extra proof reading, and a few typos and grammar issues were corrected. For example:

Line 237: "The colour scales represents…" was revised to "The colour scales represent…"

Line 471: "The phase analysis of the 3D-printed steels were…" was revised to "The phase analysis of the 3D-printed steels was…"

Reviewer 2

1. "Tan et al. discussed the significant advancements in structural alloys over the past century, emphasizing improved mechanical performance but noting the downsides of increasingly complex alloy compositions, such as recycling challenges with higher costs. This research work shows that 3D-printed plain carbon steels can achieve attractive mechanical properties, such as tensile strength and elongation and impact toughness, comparable to, or even better than, more complex ultra-high strength steels and Maraging steels. By adjusting 3D printing parameters, the microstructure of the two carbon steels can be tailored for specific applications, offering new opportunities to reduce alloy complexity without sacrificing performance. This approach highlights the potential of 3D printing to drive the shift towards simpler, more efficient alloy compositions in metal processing."

Thank you

2. "While the move towards alloy "plainification" via laser powder bed fusion (L-PBF) printing is promising, it raises some concerns. Simplifying alloy compositions might lead to a trade-off in the long-term durability and specific performance characteristics that complex alloys are designed to provide. It is worthy noting that alloying elements are added in engineering alloys not only for enhancing strength and ductility or toughness, but the comprehensive performance including but not limited to processability, corrosion, oxidation, etc. While plain carbon steels may perform well in controlled tests, their real-world application could reveal limitations in environments where advanced alloys typically excel."

We fully agree with the reviewer. Of course, if the alloy needs corrosion resistance, there is no alternative to adding Cr. This is required. Similarly, for high temperature oxidation, Al may be needed in some applications. We have not intended to give the impression that a 3D-printed plain carbon steel can be used for every imaginable application, in every type of environment. We do not feel that the manuscript as it is written gives such an impression.

Currently, for wrought high-strength steels, martensite or bainite are mostly used. To obtain such microstructures in commercial wrought steels, lots of alloying additions need to be added to provide the hardenability for wrought processing. We show in this contribution that this is not necessary in 3D-printing via PBF, such microstructures can be obtained in plain carbon steels, with comparable or even better basic mechanical properties. If the 3D-printed steel also needs corrosion resistance, then Cr will need to be added.

The concept of "plainification" does not suggest that simplified compositions can replace every highly alloyed material, especially those specifically designed for functional or environmental considerations. Instead, "plainification" emphasizes the use of simpler compositions that can still achieve characteristics similar to those of more complex alloys, with consequential benefits on cost, recyclability, and security of supply of materials [*Raabe et al., Nature 575 (2019) 64-74; Li et al., Science 364 (2019) 733-734*]. Following this concept, the claim of our work is not to replace advanced alloys in every application but rather to demonstrate the feasibility of simplifying alloy compositions through 3D-printing while maintaining high mechanical performance like alloy steels. Therefore, we focused on evaluating and comparing the basic mechanical properties, such as toughness, strength, and ductility, as they provide an indication of the material's potential as a structural material.

To avoid any misunderstanding, several sentences were revised/added on Line 392 on Page 11 to clarify the future work required.

The promising properties achieved in the 3D-printed plain carbon steels underscore the imperative need for future work in this domain, particularly in other properties such as fatigue resistance, fracture toughness, and stress corrosion cracking. Naturally, improving properties like corrosion resistance will require the addition of elements such as chromium, while other strategic alloying elements may be necessary for specific properties such as oxidation resistance. The principle remains to add complexity to the composition only when absolutely necessary. Such developments align closely with the ongoing focus on material sustainability and plainification, positioning metal 3D-printing as versatile and forward-looking choices in the evolving landscape of materials technology.

Throughout the manuscript text, whenever talking about performance of the plain carbon 3D-printed steels, we have also modified the text so it is clear we are only talking about 'mechanical performance', to try and avoid any misunderstanding regarding corrosion or oxidation resistance. These changes are highlighted in yellow through the manuscript text.

3. "Also, the reliance on L-PBF printing for achieving desirable microstructures, such as martensite or bainite, could also introduce inconsistencies due to variations in printing conditions (process parameters, printing layout, and scale), potentially compromising material quality."

The reviewer is right that variations in printing conditions can lead to different microstructure and then different properties, which is very common in PBF printing of many materials. For this reason, we did investigate how the processing parameters that defined the energy density influence the microstructure and properties. We observe excellent consistency in terms of microstructure and properties, regardless of sample geometry and scale (Fig. 1).

The plain carbon steels investigated in this work have high 3D-printability evidenced through their very wide processing windows as shown in Fig. 1i and Supplementary Fig. S2d. Both 1080 and 1040 steels achieved nearly full densification across a wide processing window. Hence, plain carbon steels are very suitable for metal 3D-printing. Furthermore, the variation of the microstructure and properties with 3D-printing processing parameters provides room for us to tailor the steel properties for different requirements. This can be regarded as an advantage of 3D-printing.

A sentence was added on Line 155 on Page 4 to emphasize the benefits of the excellent 3D-printability of plain carbon steels.

This not only ensures reliable production quality but also offers the scalability in tailoring their properties using 3D-printing.

4. "Though there are some deficiencies in language and lacking universality, this is a quality work presenting plenty of L-PBF printing outcomes and beautiful microstructural characterization."

We appreciate the reviewer for recognizing the quality of our outcomes. To address the reviewer's concerns regarding language and universality, we have carefully revised the manuscript to enhance clarity and readability, and to polish the written English. Additionally, following **Reviewer 3**'s suggestion, we have reorganized the order of the paragraphs to follow a Processing-Microstructure-Properties sequence, making it easier for readers to understand.

5. "Two carbon steels were additively manufactured using L-PBF with varying process parameters mainly via low to high energy densities. However, it is quite known regarding the correlations between energy density and resultant martensitic or annealed microstructure, leading to strength-ductility trade-off in tensile performance for a wide range of engineering alloys in the community. It sounds that the authors exaggerate the point of "plainification" by making use of established L-PBF printing knowledge. This work should surely be published in Acta Materialia, but may be debatable to publish in Nature series journals.

While we appreciate the reviewer's comment, we would like to further clarify the impact and novelty of our work.

The key novelty of our work is that martensitic and/or bainitic microstructure can be obtained uniformly on large cross sections in plain carbon steels using metal 3D-printing, which cannot be achieved through conventional manufacturing processing unless highly alloy steels are used. Given that plain carbon steels are among the most widely used metallic materials globally and that material sustainability and simplification are increasingly important, our work can lead to significant impact to wide ranges of engineering communities. We demonstrate a concept that has quite broad applications and implications. Hence, we submitted our work to *Nature Communications*. In contrast, papers in Acta Materialia focus more on a particular topic with specific advancements.

Reviewer 4

This work systematically investigated microstructure evolution and mechanical performance of 3D-printing plain carbon steels. They found that the sequential nature of the micro-scale melting and solidification made plain carbon steels achieve tensile and impact properties comparable to, or even superior to those of low and medium alloy ultra-high strength steels and some Maraging steels, and demonstrated that 3D printing was a reliable approach to reduce alloy complexity without compromising performance. These findings are interesting and constructive, and it is worth to be published at Nature communications. I suggest authors address the following comments before publication.

We appreciate the reviewer's supportive words and constructive suggestion.

1. Tensile and impact properties of plain carbon steels and other alloying steels are compared in this work. Fracture toughness is also an important index for ultra-high strength steels. Is the fracture toughness of 3D-printing plain carbon steels comparable to the alloying steels?

The reviewer is absolutely right that fracture toughness is an important property for high strength steels. The challenge is that it is difficult to measure accurately because of sample size requirements compared with the expected plastic zone size. This difficulty is not unique to 3D-printed materials but a challenge for all ductile materials. We have, like most other authors, used the standard Charpy test to provide a measure of the fracture resistance, and compared this with wrought materials.

There is just starting to be some fracture toughness data becoming available in the literature on PBF metals such as steels. They show excellent fracture toughness, and we anticipate that our plain carbon steels, based on their impact toughness values and the comparison with wrought steels, will also exhibit excellent fracture toughness values.

e.g. MJ Paul, H Li, E Brodie, JJ Kruzic, CR Hutchinson, B Gludovatz, The effect of micro- and mesoscale heterogeneity on the fracture of laser powder bed fusion processed duplex stainless steels, *Scripta Materialia*, 255, art#116334, 2025.

e.g. MJ Paul, JJ Kruzic, U Ramamurty, B Gludovatz, The importance of fracture toughness evaluation for additively manufactured metals, *Acta Materialia*, art#120061, 2024

Given the potential of 3D-printing in producing high-performance plain carbon steels, we believe that this is an important area for future research to further validate the process in terms of other properties such as fracture toughness, fatigue resistance and stress corrosion cracking. Accordingly, we have added a sentence on Line 392 on Page 11 to indicate this direction.

The promising properties achieved in the 3D-printed plain carbon steels underscore the imperative need for future work in this domain, particularly in other properties such as fatigue resistance, fracture toughness, and stress corrosion cracking.

2 One important point is that increasing laser energy density can make a martensite-dominant microstructure transformed into bainitic-dominant microstructure, which is attributed to the decreased cooling rate. Increasing laser energy density not only enlarges melting pool size, but also accelerates convection. Both of them could play a vital role in the phase transition path. I suggest that authors could provide a quantitative analysis by finite element simulation or thermodynamic calculation to reveal the relationship between thermal history and phase transition.

We appreciate the reviewer for this thoughtful comment.

We have performed simulations of solidification in melt pools in other alloy system using Computational Fluid Dynamics-Discrete Element Method (CFD-DEM), [*Q. Tan et al., J. Mater. Sci. Technol. 175 (2024) 153-169*], however, liquid convection is expected to have a limited impact on the martensite and bainite formations as they are solid-solid phase transformations occurring at temperatures far below the solidification point. In this system, the liquid solidifies as δ-ferrite at temperature of ~1500 °C which then transforms into γ-austenite (FCC) during cooling and the martensite and bainite transformations occur from this γ-austenite at temperatures below ~500 °C. Since the martensite and bainite form as a solid-solid phase transformation, they are less affected by the solidification conditions, unless there are significant solute segregations or similar factors (discussed further below, point #4).

3. L-shape demonstration parts fabricated by 3D-printing and common quenching are compared in Figs. 1(g) and (h). Please list the laser energy density used in 3D-printing. Is the microstructure martensite or bainite? Besides, authors consider that the cyclical thermal profile from the melting of successive layers provides an in-situ tempering effect to reduce the thermal residual stress, leading to the 3D-printing part without cracking. However, the ultrafast heating and cooling always generate high residual stress during AM. I notice that 200 °C preheating is used during AM. Maybe the real reason is the preheating lowers cooling rate, thereby weakening thermal residual stress.

The laser energy density and the expected microstructure of the 3D-printed L-shaped part has been added to Method section on Line 465 on Page 12:

…the other was directly 3D-printed using 1080 steel powder with the laser power of 150 W and scanning speed of 600 mm/s (i.e. the low-energy-input of 69 J/mm^3 for martensitic microstructure).

Regarding the thermal stress, we acknowledge that the generation and relief of stresses in 3D-printed parts is a complex process. The less distortion and cracking tendency observed in the 3D-printed L-shaped part indicates lower thermal stress level during 3D-printing compared to conventional water quenching processes. As substrate preheating is a very common process involved in many PBF processes, it facilitates the weakening of the thermal residual stress as indicated by the reviewer.

Accordingly, one sentence was revised on Line 145 on Page 4:

…whilst the substrate preheating (Method) and the cyclical thermal profile from the melting of successive layers provides an in-situ tempering effect, reducing the thermal residual stress[13,14].

4. 3D-printing samples always have typical cell structures, i.e., element segregation. Are there cell structures within two kinds of plain carbon steels? What are their effects on mechanical properties? It could be interesting to provide more discussions.

The reviewer is absolutely correct, many systems show cell structures and elemental segregation. In our case, because we only have the fast-diffusing carbon as the major alloying element, we do not have any segregation effects. This is an advantage. Also, because after solidification, the alloy passes through several solid state phase transformations, any cell structure that might have been present immediately after solidification is removes by the structural rearrangement associated with the solid-solid phase transformations (δ to γ, and γ to martensite or bainite).

We have added a paragraph on Line 339 on Page 10 discussing this point that other readers may also be interested in.

Notably, in some alloy steels, the high level of alloying element additions can lead to pronounced solute segregation and the formation of cellular structures under rapid solidification conditions during 3D-prinitng[13]. These structures can strengthen the steel by providing effective grain boundaries that impede dislocation movement. However, significant solute segregation may also result in the formation of brittle phases at the cell boundaries, decreasing ductility and toughness. A typical example is the formation of mechanically unstable retained austenite networks in the 3D-printed H13 steels[37]. In contrast, the plainified composition of carbon steels circumvents this issue while capitalizing on rapid cooling to facilitate steel hardening.

Reviewer 5

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

We appreciate the collaborative effort in the review process and the constructive feedback provided by both reviewers, who have helped us improve the quality of our manuscript.

Reviewer 1

1. "This paper provides as interesting fashionable perspective, but this is not demonstrated by the presented evidence. The key premise of this paper is that alloys of relatively complex steels can be replaced by simple Fe-C alloys by adopting the layer-by-layer approach of additive manufacturing. However, this is not demonstrated. Components such as gears are subjected to conditions measured by engineering properties different to those shown in the paper. Ashby maps are too simple and to not reflect wear and contact fatigue. These are the real engineering properties to which to compare, rather than toughness, strength or ductility. As such the key premise of the paper is flawed and this reviewer believes is not suitable for this journal."

We are very sorry if there has been some misunderstanding. Our paper does not make claims about qualifying a new material to be used as a gear. This is not what our manuscript is about.

The gear-like component shown in Fig. 1 was intended to demonstrate how 3D-printing can address challenges typically encountered with conventional water quenching of components with complex geometries. Of course, if a new material is going to be used for an application such as a gear, all the relevant properties need to be measured to ensure suitable performance.

This paper is not about qualifying a material to be used as a gear and we do not think our manuscript gives that impression. The properties we show are the basic mechanical properties that form the basis of any consideration of mechanical performance. Figs. 1 and 4 demonstrate that our 3D-printed plain carbon steels (1080 and 1040) are comparable with and even better than alloy structural steels processed with conventional processes in terms of hardenability, mechanical tensile properties, and Charpy impact toughness. Of course, qualifying a new material requires more, but this paper is about a new concept and approach, not about qualification of materials. If it was about qualification, a scientific journal would not be appropriate.

To help clarify this, a sentence was added on Line 135 on Page 4:

It should be emphasised that this gear-like component was not intended for specific practical applications as a gear but rather to demonstrate how 3D-printing can address challenges typically encountered with conventional water quenching of engineering structural parts with complex geometries.

2. "The paper demonstrates that different microstructures can be tailored and attained through rolling contact fatigue, but the link of microstructure with relevant properties is not there. As such, the presented information is not new."

We are very sorry, but we do not understand this comment.

Our paper has not looked at rolling-contact fatigue in any way. The main message of our paper has nothing to do with rolling-contact fatigue or the microstructures that result from that purpose.

The key message of our paper is to show that metal 3D-printing has potential to reduce alloy compositional complexity without compromising mechanical performance, and therefore can play an important role in driving alloy plainification.