Sintering of 3D printed metal, ceramic and glass multi-material parts

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ABSTRACT

3D printing technologies are well suited to the production of multi-material parts with discrete or continuous material distributions. A range of 3D printing technologies are evaluated to determine promising strategies for the concurrent 3D printing of multiple material classes; specifically metals, ceramics and glasses in a single printed object. A low cost and versatile printing strategy, based on material extrusion and subsequent sintering is identified, and a range of design for manufacture strategies are synthesized. Preliminary examples of multi-material sintered parts including metal-metal, metal-glass and metal-ceramic parts are presented.

KEYWORDS: Sintering; Multiple Material; Material Extrusion; 3D Printing; Metal/Ceramic.

Abbreviations: Additive Manufacturing (AM), Binder Jetting (BJ), Directed Energy Deposition (DED), Fused Filament Fabrication (FFF), Material Extrusion (ME), Material Jetting (MJ), Powder Bed Fusion (PBF), Stereolithography Apparatus (SLA).

1. INTRODUCTION

3D printing technologies are well known for permitting the production of parts with extreme geometric complexity [1-3]. Multi-material 3D printing systems can extend capabilities further, by varying materials within part volumes layer-by-layer, or voxel-by-voxel. Vaezi et al (2013) produced a comprehensive review of academic research into multi-material printing [4]. Common uses of multi-material systems are for the production of parts with varying colours, opacity, elasticity, biological function, mechanical, thermal and electrical properties [5-10]. Applications for multi-material parts will increase as new design tools and manufacturing processes become available [11-14].

Most multi-material systems require the individual materials to have similar processing conditions, e.g. photopolymers with similar viscosity and surface tension. The greater the difference in the processing requirements of the materials, the more difficult it is reconcile the various chemical, thermal, rheological and mechanical characteristics of the materials [15-19]. This problem still needs to be addressed.

The purpose of this paper is to suggest a low-cost but versatile technique for printing multi-class materials in a single print, with strategies for reducing the problems inherent with dissimilar materials, such as large differences in processing temperatures and shrinkage rates.

1.1 Multi-material 3D printing technology evaluation

The challenge of physically printing multi-material parts can be split into two separate issues; placement of the materials in 3D space and consolidation of the materials. Research has shown all seven main AM groups (as defined by ASTM 42) are capable of some form of multi-material printing [4, 20, 21]. Material deposition techniques which can address a single point, or line in space, are the most promising candidates for multi-material parts as they more efficiently allow different materials to be placed within layers. Powder bed fusion and sheet lamination are essentially limited to material changes between layers, while vat polymerisation does not lend itself to the printing of multi-material parts unless it is hybridised with other equipment. Consolidation (i.e. binding or densifying) of the materials may occur as they are deposited, as is the case with FFF and DED, or in a post-process, like sintering or infiltration (e.g. SLA ceramics or binder jetted steel powders infiltrated with bronze).

In this paper the authors have separated polymers, and other low temperature materials, from high temperature materials such as metals, ceramics and glasses. The consolidation temperature is often the most important parameter influencing material compatibility when 3D printing. The importance of other material properties will be addressed in later sections.

Table 1 summarises the seven main AM technologies, whether they are suitable for multiple material printing and whether they process high temperature materials directly or with a secondary step.

Table 1. Evaluation of multi-material printing capabilities for different technologies.

		Material addition		Multi-material		Material groups		
						Polymers	Metal / Ceramic / Glass	
Technologies	Variants	Point	Layer	Within layers	Between layers		Directly	With secondary sinter
VAT Polymerisation (VT)	SLA	□	■	□	□	■	□	■
	DLP	□	■	□	□	■	□	■
Material Extrusion (ME)	FFF	■	□	■	■	■	▨	■
	Paste	■	□	■	■	■	□	■
Material Jetting (MJ)	Polymerisation	■	□	■	■	■	▨	▨
	Nano particle jetting	■	□	▨	▨	▨	▨	▨
Powder Bed Fusion (PBF)	SLS	□	■	□	▨	■	■	■
	SLM	□	■	□	▨	■	■	■
Binder Jetting (BJ)	Adhesive binding	□	■	□	▨	■	□	■
Sheet Lamination (SL)	Ultrasonic consolidation	□	■	□	▨	■	■	▨
	Adhesive bonding	□	■	□	▨	■	■	▨
Directed Energy Deposition (DED)	Melting	■	□	■	■	■	■	▨
	Cold welding	▨	□	▨	▨	▨	▨	▨

■ Commercially available
▨ Possible/Practical
□ Difficult/Unpractical

DED processes can deposit and consolidate high temperature materials directly, with multi-materials usually in the form of functionally graded materials (FGM) [21-24]. Sharp material transitions in directly consolidated FGMs are problematic because the materials experience rapid changes in local temperature and density, causing high thermal stresses and ultimately cracks [25].

ME and MJ have favourable material addition methods allowing material changes within layers. ME and MJ are better suited to processing low temperature materials, such as pastes, thermoplastics, photopolymers and ceramic or metallic loaded polymers. ME is a particularly robust method of depositing materials with a wide range of consistencies, including inks, pastes and clays. These materials can then be sintered in a secondary process [17, 26-29]. While the secondary steps require extra time and resources, removing the consolidation step from the printing greatly reduces printer complexity and cost when compared with PBF and DED. For this reason ME and a secondary sintering step is chosen as the most versatile and cost effective method to print multi-class material parts.

2. PRINTER DESIGN

The proposed method of printing is based on the modification of a Makergear M2 printer as shown in Figure 1. In practice any FFF based printer could be modified in this way. The requirements for the extruder design were as follows:

- Forces sufficient to extrude powders, pastes and clays with low binder/water content
- Low cost
- Compatible with conventional 3D printing hardware and software
- Modular so extra materials can be added in parallel
- A limited number of custom parts

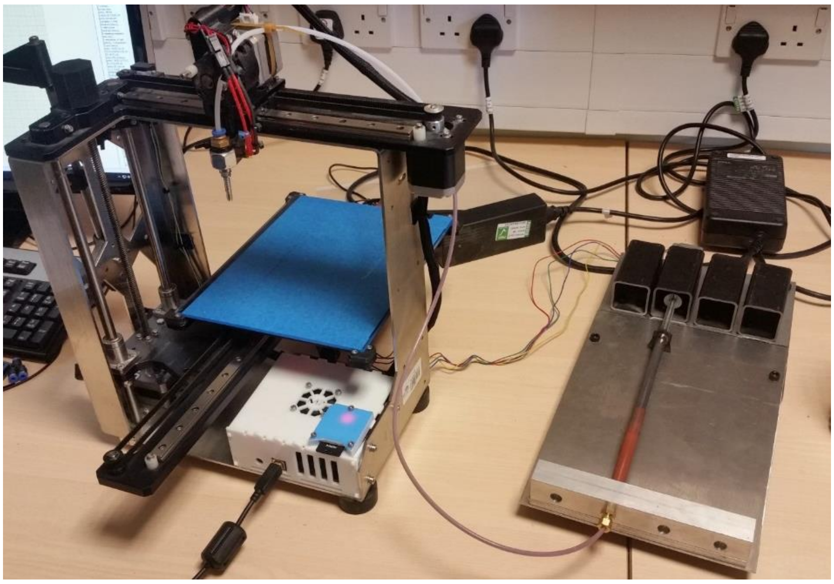

Figure 1. 3D printer modified for the printing of plastic solids.

The final extruder design consists of a geared stepper motor which drives a leadscrew piston. This is a functional and low cost design allowing up to four materials to be printed at once. One downside is that extrusion may be susceptible to lag when printing with compressible materials. All modules were designed to share one nozzle via a double Y pneumatic quick fit joint, so purging of the nozzle during material change over will be necessary. This feature also makes it possible to blend materials, although complete mixing is unlikely.

Materials with low binder content are preferable as this decreases burnout times, shrinkage and porosity, however this comes at the cost of green strength. For this reason support material may be required, even for parts without overhangs.

3. SINTERING STRATEGIES

Sintering of metal and ceramic powders is a well-established and economic process for high volume production of geometrically complex components. Cemented carbide cutting tools and low temperature co-fired electronic components are commercially successful multi-material examples [30, 31]. Sintering is one of the most promising methods to process discrete or FGM and much has been written on the subject in recent years [25, 32-34].

The physical processes involved in sintering are complex and will not be covered in detail here. However, it is important that the sintering temperatures and the shrinkage behaviour of the materials are similar enough to prevent the critical levels of stress which lead to warping, delamination and cracks. The sintering atmospheres and chemical compatibility of the materials also needs to be considered. At first sight this would seem to greatly restrict the number of feasible material combinations that can be co-sintered. Fortunately there are a number of parameters which can be adjusted in order to bring the sintering characteristics of various materials into line.

The first parameters to mention are sintering temperature, shrinkage and powder size. Sintering of a particular material is a function of both time and temperature so increasing the temperature usually reduces the sintering time and vice versa. Additionally changing the materials particle size, composition, or initial apparent density will alter the sintering temperature and shrinkage. This selection of adjustable parameters greatly expands the number of compatible material combinations. Figure 2 shows current common ranges of sintering temperatures for common alloys and ceramics. Drawing a horizontal line through the chart highlights that many materials are compatible on the basis of sintering temperature.

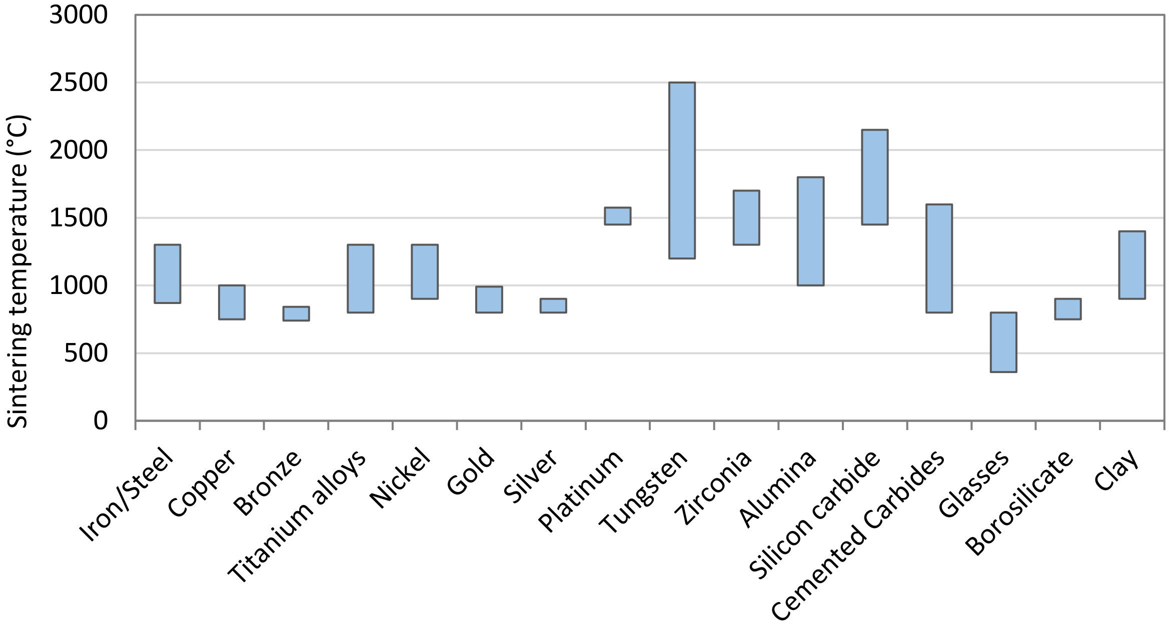

Figure 2. Sintering temperatures for different materials.

Taking a simplistic view, there also appears to be large overlaps for the shrinkage of sintered materials as shown in Figure 3.

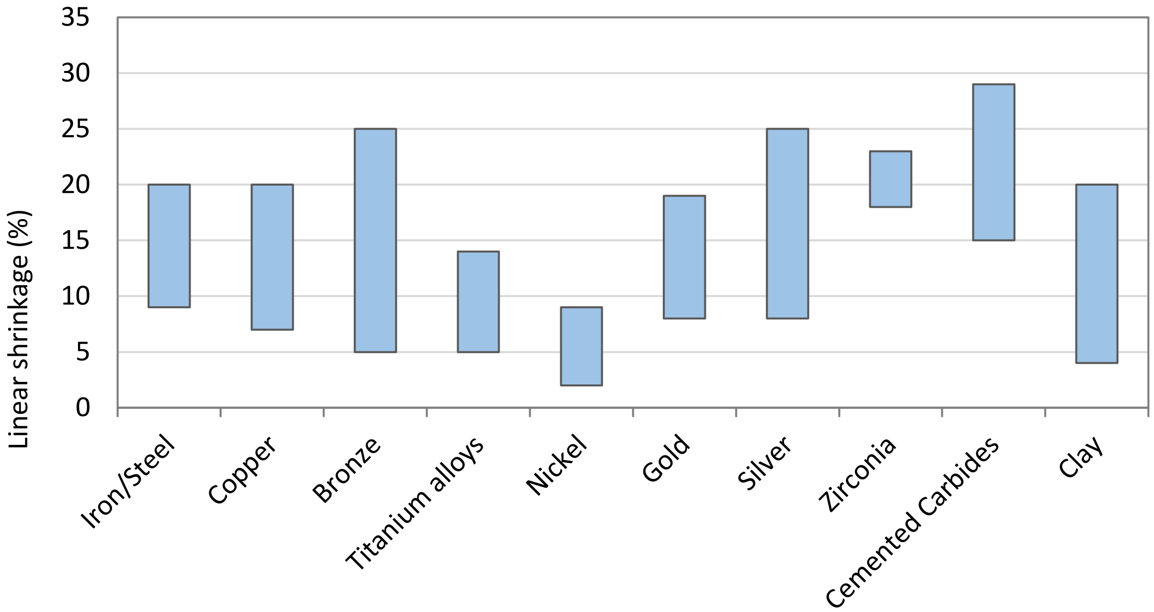

Figure 3. Sintering shrinkage for different materials.

This provides optimism moving forward, however, shrinkage matching is further complicated by the requirement for not only similar overall shrinkages, but similar shrinkage profiles over time. These behaviours can be characterised using dilatometry.

Finally, if materials are so dissimilar that no common sintering conditions can be found, then use of liquid phase sintering may provide a solution. This allows extremely high temperature metals and ceramics to be co-sintered with much lower temperature materials.

4. DESIGN STRATEGIES

Even with materials that share similar sintering temperatures and shrinkage rates there are likely to be complications with high shear stresses or chemical incompatibility at the interfaces. Using copper and bronze as an example of compatible materials with similar shrinkage rates, FEA simulations show that high stresses form at the material interface (Figure 4a). In practice, these stresses will be lower if high levels of diffusion occur between the materials, or if the strain energy is released due to the formation of cracks. One strategy is to accept the creation of cracks along the interface and instead use design strategies to create geometrical interlocks between the two materials as shown in Figure 4b.

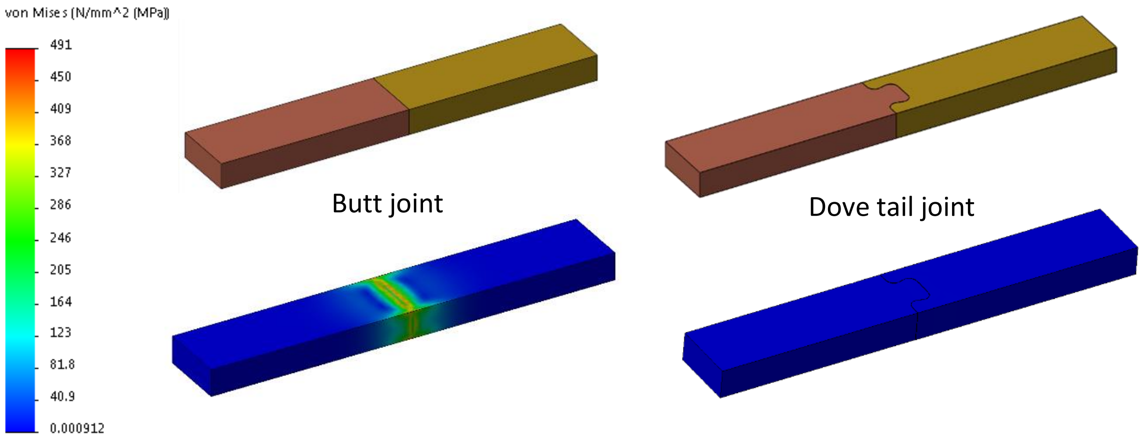

Figure 4. a) Bonded butt joint. b) Un-bonded dove-tail joints for incompatible materials.

In cases where the outer material is known to shrink more than the inner material a simple strategy is to introduce a gap between the two materials at the design stage. The gap can be controlled to produce a wide range of fits, including loose fits or shells with a high level of pretension.

Another situation likely to be encountered is where the inner material is known to shrink more than the outer material. In this case a combination of geometric interlocking, such as dovetails, and the introduction of a small gap between the two materials, will allow the materials to be fixed together with varying degrees of preload as shown in Figure 5.

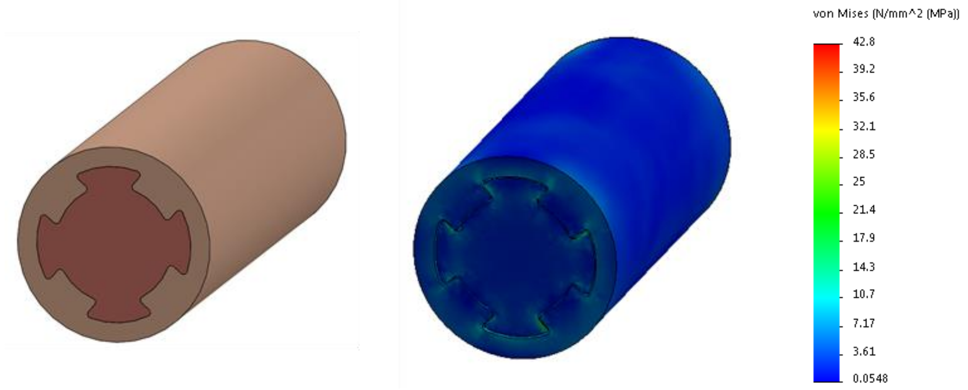

Figure 5. Dovetail joints with small surface offsets for shell and tube structure, where the inner material shrinks more than the outer material.

For large and complicated multi-class material parts automated design tools may be required. It is envisioned that custom software could simulate the shrinkage of the different materials and check for overlapping volumes. Automated software could also provide surface offsets in order to retain coupling but avoid excessive stresses.

4. PRELIMINARY RESULTS

A range of material combinations and geometries were sintered in order to confirm the feasibility of the proposed printing, materials selection and design strategies mentioned in previous sections. Ten materials were trialled including; copper, bronze, aluminium, iron, sterling silver, low-fire clay, soda lime glass, alumina, silicon carbide and boron carbide. All metal, ceramic, and glass powders and clays were commercially available and not modified in any way. The objective of these preliminary tests was to determine if any promising material combinations could be sintered without the need for modifying particle size or material composition. An electric furnace was used for sintering with coconut based activated carbon providing a reducing atmosphere. Figure 6 shows a range of sintered samples made from extrudable metal and ceramic clays.

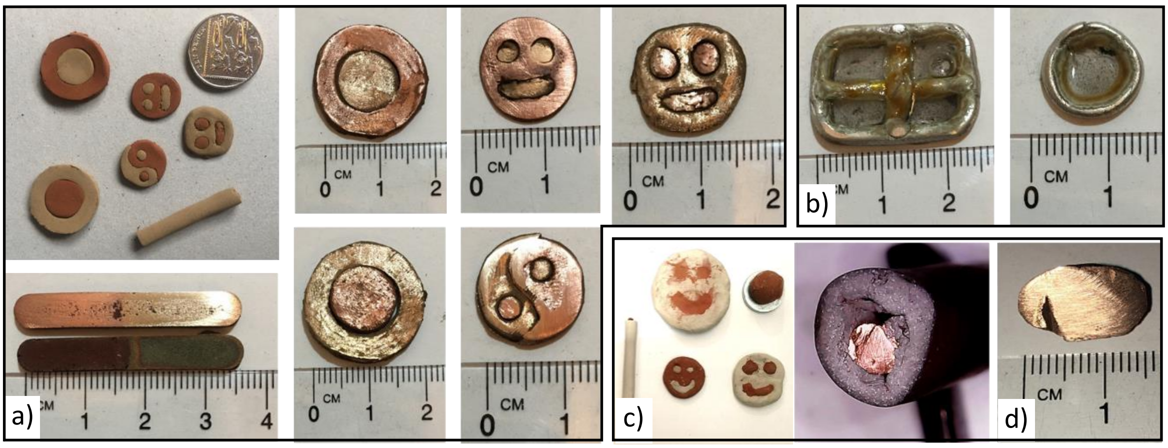

Figure 6. Successful examples of multi-material sintering; a) Copper and bronze. b) Sterling silver and soda lime glass. Low fire clay and copper. d) Liquid phase sintering of copper and bronze.

While it was encouraging to see a number of the material combinations work with no need for modification, many of the material combinations trialled were unsuccessful. Most failures could be attributed to insufficient binder burnout, high levels of oxygen and insufficient sintering temperature and time. The furnace used in this study had a maximum temperature of 1000 °C which only allowed for liquid phase sintering of the higher temperature materials. Recent developments in office friendly sintering technology, including microwave sintering, mean that many of these problems are unlikely to be long term issues.

5. CONCLUSION

A technical evaluation of current 3D printing technologies revealed that combining material extrusion with subsequent sintering is potentially a versatile and economical strategy for the manufacture of multi-class material parts. A brief survey of common materials shows that while well-matched sintering characteristics are required, there are still likely to be a wide range of materials that are compatible. Furthermore there are a number of material parameters such as particle size and composition which can be varied in order to increase compatibility.

FE simulations show that once compatible materials are found a range of design strategies can be employed to further increase the chances of sintering success. These strategies include the use of geometric coupling (2D and 3D dovetails) and the addition of small gaps to allow for differences in shrinkage.

Finally, the sintering of multi-material samples was successfully demonstrated including metal/metal, metal/ceramic and metal/glass structures. The authors believe that with further developments in extruder technology, customised materials, and design-for-multi-material-3D printing, a wide variety of 3D printed multi-class material parts are possible.

REFERENCES

1. Brooks, H. and K. Brigden, Design of conformal cooling layers with self-supporting lattices for additively manufactured tooling. *Additive Manufacturing*, 2016. **11**: p. 16-22.
2. Gibson, I., D. Rosen, and B. Stucker, *Additive Manufacturing Technologies: 3D Printing, Rapid Prototyping, and Direct Digital Manufacturing*. 2014: Springer New York.
3. Brooks, H. and S. Molony, Design and evaluation of additively manufactured parts with three dimensional continuous fibre reinforcement. *Materials & Design*, 2016. **90**: p. 276-283.
4. Vaezi, M., et al., Multiple material additive manufacturing – Part 1: a review. *Virtual and Physical Prototyping*, 2013. **8**(1): p. 19-50.
5. Guttag, M. and M.C. Boyce, Locally and Dynamically Controllable Surface Topography Through the Use of Particle-Enhanced Soft Composites. *Advanced Functional Materials*, 2015. **25**(24): p. 3641-3647.
6. Willis, K., et al., Printed optics: 3D printing of embedded optical elements for interactive devices, in Proceedings of the 25th annual ACM symposium on User interface software and technology. 2012, ACM: Cambridge, Massachusetts, USA. p. 589-598.
7. Brunton, A., C.A. Arikan, and P. Urban, Pushing the Limits of 3D Color Printing: Error Diffusion with Translucent Materials. *ACM Trans. Graph.*, 2015. **35**(1): p. 1-13.
8. Panesar, A., et al. *Design optimization for multifunctional 3D printed structures with embedded functional systems*. in *11th World Congress on Structural and Multidisciplinary Optimisation*. 2015.
9. Schubert, C., M.C. van Langeveld, and L.A. Donoso, Innovations in 3D printing: a 3D overview from optics to organs. *British Journal of Ophthalmology*, 2014. **98**(2): p. 159-161.
10. Malone, E. and H. Lipson, *Multi-Material Freeform Fabrication of Active Systems.* 2008(48357): p. 345-353.
11. Cheney, N., et al., *Unshackling evolution: evolving soft robots with multiple materials and a powerful generative encoding*, in *Proceedings of the 15th annual conference on Genetic and evolutionary computation*. 2013, ACM: Amsterdam, The Netherlands. p. 167-174.
12. Gaynor, A.T., et al., Multiple-Material Topology Optimization of Compliant Mechanisms Created Via PolyJet Three-Dimensional Printing. *Journal of Manufacturing Science and Engineering*, 2014. **136**(6): p. 061015-061015-10.

13. Meisel, N. and C. Williams, An Investigation of Key Design for Additive Manufacturing Constraints in Multimaterial Three-Dimensional Printing. *Journal of Mechanical Design*, 2015. **137**(11): p. 111406-111406-9.

14. Zegard, T. and G.H. Paulino, Bridging topology optimization and additive manufacturing. *Structural and Multidisciplinary Optimization*, 2016. **53**(1): p. 175-192.

15. Perelaer, J., B.J. de Gans, and U.S. Schubert, Ink-jet Printing and Microwave Sintering of Conductive Silver Tracks. *Advanced Materials*, 2006. **18**(16): p. 2101-2104.

16. Liu, Z.H., et al., Interfacial characterization of SLM parts in multi-material processing: Metallurgical diffusion between 316L stainless steel and C18400 copper alloy. *Materials Characterization*, 2014. **94**: p. 116-125.

17. Scheithauer, U., et al., *Processing of Thermoplastic Suspensions for Additive Manufacturing of Ceramic-and Metal-Ceramic-Composites by Thermoplastic 3D-Printing (T3DP)*, in Additive Manufacturing and Strategic Technologies in Advanced Ceramics. 2016, John Wiley & Sons, Inc. p. 19-30.

18. Mott, M. and J.R.G. Evans, Zirconia/alumina functionally graded material made by ceramic ink jet printing. *Materials Science and Engineering*: A, 1999. **271**(1–2): p. 344-352.

19. Kumar, P., et al., Direct-write deposition of fine powders through miniature hopper-nozzles for multi-material solid freeform fabrication. *Rapid Prototyping Journal*, 2004. **10**(1): p. 14-23.

20. Gibson, I., D. Rosen, and B. Stucker, *Additive Manufactruing Technologies*. 2010: Springer US.

21. Jones, J.B., et al., Gateways Toward Dissimilar Multi-material Parts. 2012.

22. Yakovlev, A., et al., Laser-assisted direct manufacturing of functionally graded 3D objects. *Surface and Coatings Technology*, 2005. **190**(1): p. 15-24.

23. Jiang, W., R. Nair, and P. Molian, Functionally graded mold inserts by laser-based flexible fabrication: processing modeling, structural analysis, and performance evaluation. *Journal of Materials Processing Technology*, 2005. **166**(2): p. 286-293.

24. Carroll, B.E., et al., Functionally graded material of 304L stainless steel and inconel 625 fabricated by directed energy deposition: Characterization and thermodynamic modeling. *Acta Materialia*, 2016. **108**: p. 46-54.

25. Kieback, B., A. Neubrand, and H. Riedel, Processing techniques for functionally graded materials. *Materials Science and Engineering*: A, 2003. **362**(1–2): p. 81-106.

26. Ghazanfari, A., et al., A novel freeform extrusion fabrication process for producing solid ceramic components with uniform layered radiation drying. *Additive Manufacturing.*

27. Grida, I. and J.R.G. Evans, Extrusion freeforming of ceramics through fine nozzles. *Journal of the European Ceramic Society*, 2003. **23**(5): p. 629-635.

28. Onagoruwa, S., S. Bose, and A. Bandyopadhyay, Fused deposition of ceramics (FDC) and composites. *Pro SFF, Texas*, 2001: p. 224-31.

29. Wu, G., et al., Solid freeform fabrication of metal components using fused deposition of metals. *Materials & Design*, 2002. **23**(1): p. 97-105.

30. Imanaka, Y., Multilayered low temperature cofired ceramics (LTCC) technology. 2005: *Springer Science & Business Media*.
31. Upadhyaya, G.S., Materials science of cemented carbides — an overview. *Materials & Design*, 2001. **22**(6): p. 483-489.
32. Birman, V. and L.W. Byrd, Modeling and Analysis of Functionally Graded Materials and Structures. *Applied Mechanics Reviews*, 2007. **60**(5): p. 195-216.
33. Miyamoto, Y., et al., *Functionally Graded Materials: Design, Processing and Applications*. 2013: Springer US.
34. Mehrali, M., et al., Dental implants from functionally graded materials. *Journal of Biomedical Materials Research Part A*, 2013. **101**(10): p. 3046-3057.