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PERSPECTIVE ARTICLE

Research, standards, and data needs for industrialization of ceramic direct ink writing

Andrew J. Allen | Igor Levin | Russell A. Maier

Materials Measurement Science Division, National Institute of Standards and Technology, Gaithersburg, Maryland, USA

Correspondence
Andrew J. Allen, Materials Measurement Science Division, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA.
Email: andrew.allen@nist.gov

Abstract

Here, we summarize a panel discussion on the direct ink writing (DIW) of ceramics, organized and moderated by the National Institute of Standards and Technology at the American Ceramic Society's 46th International Conference on Advanced Ceramics and Composites in January 2022. The panel reviewed the current state of the art in DIW, focusing on research directions, standards development, and data needs required to facilitate industrial adoption of these technologies. The panel agreed to form working groups for (1) exploring how the community can work together to develop and curate public databases of feedstock characteristics and properties of end-products and (2) organizing an international round-robin to compare performance properties of DIW-manufactured ceramics, potentially tied to ASTM/ISO standards development.

KEYWORDS

additive manufacturing, ceramics, direct ink writing, extrusion

1 | INTRODUCTION

Ceramic direct ink writing (DIW) refers to a suite of technologies that utilize the extrusion of a ceramic slurry or paste through a nozzle that moves across a building platform to produce a part of a given shape, layer by layer. These techniques technically fall under the materials extrusion definition in the recently revised ASTM/ISO standard: ISO/ASTM 52900:2021(E): additive manufacturing (AM) — general principles — fundamentals and vocabulary. However, the historical context of ceramics manufacturing requires a more nuanced terminology. Low- and high-viscosity ceramic suspensions have long been utilized in traditional ceramics manufacturing using slip-casting and extrusion processes, respectively. The focus here is on the intermediate-viscosity regime between these two extremes. Known initially as robocasting but now more generally accepted as DIW, this regime has opened up one of the most significant and commercially viable approaches to AM of ceramic and multi-material components. The DIW name reflects the near ambient conditions of the direct 3D printing process akin to writing and the fluid properties of the feedstock suspension resembling ink.

The National Institute of Standards and Technology (NIST) organized a discussion panel on DIW of ceramic materials, which took place, on January 26, 2022, during the 6th International Symposium on Additive Manufacturing and 3D Printing Technologies, part of the American Ceramic Society's (virtual) 46th International Conference on Advanced Ceramics and Composites (ICACC 2022). This panel continued a series of NIST-led discussion events[1] on ceramics AM to identify measurement, standards, and data needs hindering the commercialization of ceramics AM and to facilitate collaborative efforts within the ceramics AM community. The international group of panelists included representatives of industrial companies developing these technologies, industrial end-users, government laboratories, ASTM/ISO, America Makes, and academia. The program consisted of brief presentations followed by an extensive discussion of critical aspects of ceramics DIW. These topics included feedstock morphology, process fundamentals, post-processing, defects, and properties of final products, focusing on measurements, data, and standards requirements from both the technology developers and end-user perspectives. In the following, we summarize the main points considered by the panel. The opinions and technical arguments expressed by the panelists reflected their practical experience and perspective of the field, not always traceable to specific references in scientific literature. The purpose of this article is to present such practitioner views of DIW technologies, highlighting the opportunities for research and development, rather than a formal literature review. Recent review articles on DIW can be found.[2,3] A summary of the panelists and discussion panel focus topics is given in the Appendix.

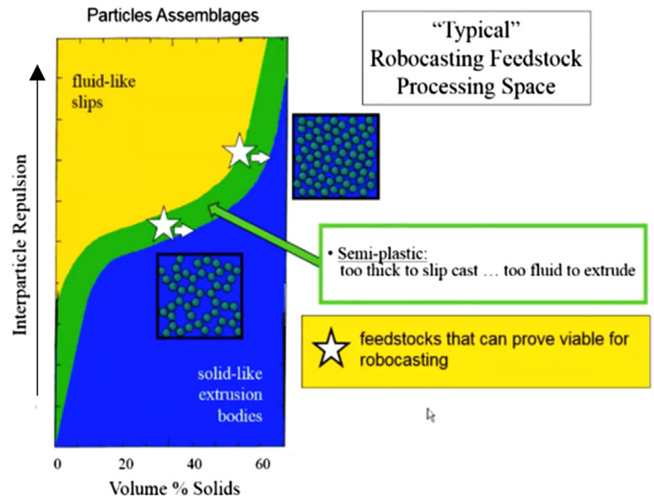

FIGURE 1 A schematic diagram delineating feedstock states suitable for processes like slip casting (top field, colored in yellow), solid-like extrusion (bottom field, blue), and robocasting or direct ink write (narrow border region, green). This diagram illustrates the existence of a wide range of feedstock compositions suitable for robocasting. The axes reflect key controllable parameters that define rheological properties – the solids loading and interparticle forces moderated by pH and additions of dispersants. The stars indicate two acceptable states for direct ink writing (DIW), represented by the cartoons of particle networks, which depend on the degree of repulsion between particles. In reality, there is a continuum of acceptable morphologies along the green band. A database of such feedstock process diagrams could provide a roadmap for the optimization of DIW processes. Courtesy of J. Cesarano

2 | FEEDSTOCK OPTIMIZATION, DATA, AND STANDARDS REQUIREMENTS

Optimizing the ceramic feedstock characteristics is central to process developments by machine vendors and product manufacturers, alike. In DIW methods such as robocasting,[4] the feedstock paste must flow through the nozzle orifice at modest pressures without clogging, should set into a non-flowable mass on dispensing, and, as deposited, must support multiple overlayers without defect formation or artifact inclusion to form a uniform body for the overall build.

For any given system, the paste can behave as a semi-Newtonian fluid for lower particle concentrations (e.g., less than 45% by volume), is pseudo-plastic in the intermediate regime (e.g., 45%–63% by volume), and dilatant for high concentrations (e.g., >63% by volume), with the delineating particle concentrations varying significantly for different feedstock particle morphologies. The optimal behavior for DIW is pseudo-plastic just below its transition to the dilatant. The viscosity rises rapidly with concentration in this regime (e.g., from ≈58% by volume upwards) – so one should aim for the extrusion paste to coincide with the onset of this increase. At the same time, the concentration ranges quoted above can vary markedly with particle surface state and geometry, and also with particle – solvent interactions and solvent fluid (effects reminiscent of the behavior discussed here and captured by the diagram in Figure 1 have been reported for non-ceramic systems).[5] Ideally, on deposition, any drying should move the deposited bead from the pseudo-plastic into the dilatant state, but not too quickly as it needs to support the next layer while also merging with it. As solvent is removed and the pseudoplastic-to-dilatant transition occurs, much larger shear stress is required to register a significant shear strain, enabling the DIW print process to proceed without compromising earlier parts of the build due to the weight of new layers deposited on top.

There appears to be no single optimal particle morphology or size distribution for powders making up feedstock pastes and slurries. For example, the effective repulsion between particles is important: the greater the short-range repulsive forces, the higher the concentration permitted before the slurry becomes dilatant. For robocasting DIW, the need is to stay in the transition zone between fluid-like slip-cast feedstocks and solid-like extrusion bodies. Therefore, there can be a range of acceptable slurries, as indicated in Figure 1. The time dependence of the feedstock rheological response to changing conditions is also essential. For example, close to the center of the nozzle where there are zero shears, the network of local associations between particles forming the slurry is retained. However, near the nozzle walls and at the nozzle orifice, there is high shear stress, and such associations are broken to give more fluid-like conditions. After extrusion, the particle "network" should re-form and the timeframe for this process can be critical to the overall printability of a given component. Finally, there is a transition to a solid state after removing the solvent, with associated shrinkage and possible shrinkage stresses. All these time dependences matter and need to be understood and optimized for a given DIW process. A further important consideration affecting feedstock selection is the target surface roughness of the product (before densification). The dimensional tolerance and roughness are affected by the nozzle diameter and particle morphology within the DIW paste. Another aspect affecting the "granularity" of the build arises due to the shear profile across the nozzle mentioned above, shear being the largest at the walls and least in the middle, where the flow velocity is highest. This spatial modulation in shear can result in intra-bead density variations and micro-inhomogeneity between the layers. Due to these effects, DIW generally gives the greatest strength in fabricated components along the extrusion direction for a length-extruded part.

Thus, printability depends on the feedstock particle size, packing efficiency, surface area, surface chemistry, solvent composition, rate of solvent removal, as well as the size and shape of the nozzle, and the ratio of the syringe-to-nozzle diameter.[6] Moreover, optimal rheology depends on the geometry of a part being printed (e.g., a solid block over an open lattice structure). Even successful printing does not guarantee shape retention after post-processing and densification. Indeed, the exact geometry of a build can be very important. A useful feedstock standard would measure the rate of particle associations relaxing after shear back to the pre-shear arrangement as a way to predict the printability window (at least in time). However, issues with post-processing anisotropy may ultimately turn out to be more important than feedstock standardization.

There exists a need for publicly curated information and data regarding the effects of feedstock characteristics on overall part printability. Examples of such properties are particle size, viscosity, interparticle interactions, including particle surface charge, fluid medium properties, and time dependences characterizing feedstock behavior. Rheological studies of feedstock slurries remain essential, requiring methodical small-amplitude-oscillatory-shear and capillary rheological measurements to determine shear stresses and loss tangents, and the degree of elastic-to-viscous response. Extrapolation of results to zero shear-strain behavior is challenging but needed. For capillary rheological properties, details of the geometry may significantly affect results, and this has a strong bearing on nozzle design.[7,8]

Time-dependent effects, that is, the system's relaxation after undergoing significant shear stress, also require investigation. How do successive layers retain individual structural integrity while binding together sufficiently to maximize overall product strength? Each time the nozzle movement starts and stops, that is, when a turn is executed during the build process, inhomogeneities can be introduced. The rate of print scanning itself is an essential variable with the rheology changing for high or low scan rates – and these effects call for new kinds of shear experiments. This topic could be a particularly fertile area for developing databases and (effectively) feedstock process diagrams that could guide the selection of process parameters. However, good final products may or may not be generated across a broad range of particle morphologies and feedstock viscosities. For example, the inclusion of ultrafine particles may improve green body and final density but potentially at the cost of a significant increase in viscosity. Discrete element modeling of the suspension behavior could provide the necessary insight into such phenomena, facilitating their optimization.

Recycling feedstock powders (excess not incorporated into DIW product, etc.) may not be a significant issue compared to the energy and cost associated with the rest of the process. However, manufacturers may need to recycle to meet increasing certification requirements, especially in Europe.[9] Recycling ceramic feedstocks is likely an energy-intensive process that can negate the recycling advantages. The more stringent European certification requirements may force innovations not currently needed in the North American environment, but which may need to be adopted at a later date.

How feedstock properties propagate into the printed part is a complex issue. For example, the printability window varies with the scale and density of the product required. Optimal feedstock rheology for fabricating highly porous structures is not the same as that for fully dense components. Therefore, testing printed parts are of prime importance to end-users, but, even there, the question is whether to evaluate the as-built "green" or the final densified product. While final-part properties are of most interest to end-users, unintended directional anisotropy of product characteristics can be rooted in the response of feedstock to the nozzle print pattern. The availability of relevant ceramics data is limited; moreover, the community still needs to define the data requirements for ceramics AM via the DIW route.

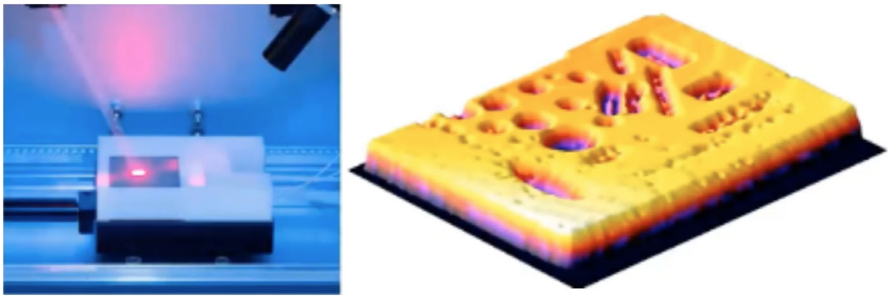

FIGURE 2 (Left) A setup for optical coherence tomography, (right) reconstructed 3D signal for a single printed layer of a ceramic material. Courtesy of A. Michaelis

Another level of complexity exists in the co-printing of several materials which attracts increasing attention with applications including functional gradient materials, hierarchical composite structures, and ceramic electronic packaging. Co-printing can be accomplished either through multiple feedstock streams through the same nozzle, or by multiple single-stream nozzles. Hierarchical issues within the feedstock morphology, and how it is dispensed (nano/micro/macro), are important and can determine defect structures in the final product.[10] Feedstock contamination can also be a serious problem, as is the compatibility of different materials that are printed together. In particular, final-part properties cannot necessarily be determined using the individual material bulk properties, and some of these non-linearities can apply to the feedstock as well.

3 | BUILD AND POSTPROCESSING: IN SITU CONTROL AND DEFECTS

Accurate control of the build process calls for more precise feedstock delivery mechanisms[11] (e.g., plunger-driven volumetric feed-scalable syringe units, multi-material in-line mixing interchangeable nozzles, micro-dispensing systems, etc.) and instrumenting DIW with sensors to provide feedback to the overall feedstock entry pressure and other parameters. Innovative designs exist that employ sensor feedback at every step of the DIW process with automated real-time adjustments of printing parameters. The results can be compared continually to model data obtained by computer simulations of the DIW process for the feedstock of interest. Laser line scanning provides a means to identify and mitigate defects as they occur (by modifying the printing parameters). Defect correction can be performed in-process by milling the surface above the layer height to preserve adhesion with the next layer, assuming that the deposited bead has transitioned into the dilatant state before the milling. Machine learning and artificial intelligence methods provide promising means for improved and automated defect detection.[12–16] Working examples exist of efficient detection of small defects during DIW of ceramic parts using optical coherence tomography (Figure 2) combined with artificial intelligence algorithms.[16] Other nondestructive methods suitable for in situ monitoring, like electrical resistive tomography, also appear to be worth exploring. Suitable validation of in-line process-monitoring feedback mechanisms, together with validation of associated model simulations predicting process behavior, will likely prove essential for realizing the potential of these methods.

The goal of such "smart manufacturing" is dual: print qualification and in situ identification and repair of printing defects. The print qualification enables the flagging of parts with flaws that may lead to catastrophic failure. In-process repair has the potential for software to make decisions related to characterizing a flaw and implementing a routine to add additional material to the local area for correcting this defect. Print qualification and defect-repair methods rely on "point cloud" data.[17] A "point cloud" is a 3D set of points extracted from an optical or acoustic image and converted into Cartesian coordinates representing the geometry of a printed part. This point cloud can be compared to a CAD dataset to determine the differences between a physical printed object and the 3D model of the desired component. The engineering methods used to acquire point clouds and the software for analyzing such data can drastically reduce defect densities of 3D-printed parts and are under active development.

Like most other ceramic AM methods, DIW provides a porous "green" part that remains to be densified via binder burnout followed by sintering – the same steps that are involved in conventional ceramic processing. Compared to traditional manufacturing, ceramic AM introduces additional types of defects (e.g., porosity specific to a certain printing process, extrusion-related defects, and air-trapped pores).[18,19] Defects tend to occur on a hierarchical basis, associated with the DIW process: inter-filament defects associated with the DIW tool path (where a filament is a single continuous stream of DIW-extruded material between changes in extrusion direction), intra-filament defects caused by binder removal; and grain size effects possibly due to particle size segregation in the feedstock, and there are also other defect modalities such as filament stacking. For example, because of the inverse relationship between print speed and voxel resolution, parts produced quickly and economically will necessarily contain larger extrusion-related defects and flaws compared to slower high-resolution processes. Considerations such as surface finish, line resolution, and pore size must be carefully weighed against print methods by ceramic AM end-users to make decisions related to the balance of cost, quality, and reliability. In particular, as with conventional ceramic manufacturing, the surface finish can have a major effect on the mechanical properties, such as those found in mechanical bend tests, and so forth.

4 | CURRENT STATE OF STANDARDS DEVELOPMENT

Recently, both ASTM and ISO initiated activities toward developing standards specific to ceramic AM. The ASTM Additive Manufacturing Committee (F42) formed a Ceramic Materials Task Group (F42.05.05) while ISO Additive Manufacturing Technical Committee (TC261) formed a joint Ceramic Materials Task Group: TC261/WG2/JG82. It is envisaged that these new initiatives, both involving ASTM and ISO, and separately organizations such as ASME, will need become significantly more focused on ceramics AM issues than AM standards work to date. Standards development is a consensus-based process, making coordination and collaboration critical for success. In this connection, a recent survey[20] conducted as a part of these ASTM/ISO efforts indicated that materials of most interest are alumina, zirconia, and non-oxides, while AM methods that would benefit most from standards development are DIW aspects of extrusion (as discussed here), vat polymerization, and binder jetting. Finally, the feedstock attributes that appeared to be of most concern are particle size, particle morphology, and processing ease.

For ceramics AM, there is strong motivation to leverage existing standards from metal or polymer AM and those from conventional ceramics where this is possible. General standards for characterizing powder particle sizes and the rheology of concentrated particle slurries are also relevant. With these points in mind, there are currently two main standards initiatives. One targets standard test specimens for ceramic parts built by the most significant AM methods: DIW, binder jetting, vat polymerization, and material jetting. The other addresses feedstock materials, mainly for vat photopolymerization, but some aspects are also common to DIW: powder characterization, crystallinity, chemical composition, rheology, and refractive index. The main ceramic AM technical focus areas for supporting standards development will likely be modeling & simulation with appropriate experimental validation, feedstock thermodynamics, material property databases, round-robin studies, and post-build processing.

5 | END-USER PERSPECTIVE AND FURTHER ACTIONS

From the industrial end-user standpoint, the adoption of ceramic AM will only happen in applications where it can provide a cost-effective solution unachievable by other technologies. Therefore, proven success and educational effort are required to inform project managers of ceramic AM's viability and inherent limitations (e.g., dimensional tolerances and surface finishes). End-users are most interested in knowing the properties of final printed parts rather than the often-proprietary details of the build process itself. Indeed, engineers need these data (e.g., thermal, mechanical, and dielectric) for their design models. Standardized test structures and measurement methods for providing properties representative of AM-built ceramic parts remain to be developed. In such testing, factors like as-printed surface finish versus that required by existing standards for conventional ceramics, as well as the inherent directional anisotropy of AM parts have to be considered. Likewise, design engineers would benefit from a database of AM ceramic materials and their properties after printing, as always, the integrity of the data curation process is an important factor for such databases to be useful. The pre-competitive information should be stored using a taxonomy that is accessible and meaningful to stakeholders in the field.

The Discussion Panel concluded with some agreement on needed follow-up actions to advance both data and standards development for DIW: (1) Explore how to work together to develop and curate public databases/process diagrams for feedstocks; (2) organize an international round-robin to compare properties of DIW-manufactured ceramics, which could potentially be tied to ASTM/ISO standards development. NIST will facilitate forming working groups that will coordinate the proposed actions, and plans are underway for a pilot round robin to establish synthesis and measurement reliability with regard to mechanical properties.

ACKNOWLEDGMENTS

This article summarizes presentations and discussions by the panelists: Joseph Cesarano (Robocasting Enterprises, LLC), Dale Cillessen (Sandia National Laboratories), Brandon Cox (Honeywell), Paul Deffenbaugh (nScrypt/Sciperio), Brian Derby (University of Manchester, UK), Nicholas Ku (Army Research Laboratory),

Alexander Michaelis (Fraunhofer Institute of Ceramic Technologies and Systems, Germany), Brandon Ribic (America Makes), Paula Vilarinho (University of Aveiro, Portugal), and Jim Weigner (Lockheed Martin). Also, see the Appendix.

ORCID

Andrew J. Allen https://orcid.org/0000-0002-6496-8411
Igor Levin https://orcid.org/0000-0002-7218-3526
Russell A. Maier https://orcid.org/0000-0003-4024-589X

REFERENCES

1. Allen AJ, Levin I, Witt SE. Materials research & measurement needs for ceramics additive manufacturing. J Am Ceram Soc. 2020;103:6055–69. https://doi.org/10.1111/jace.17369
2. Rocha VG, Saiz E, Tirichenko IS, Garcia-Tunon E. Direct ink writing advances in multi-material structures for a sustainable future. J Mater Chem A. 2020;31:15646–57. https://doi.org/10.1039/D0TA04181E
3. Balani SB, Ghaffar SH, Ghougan M, Pei E. Processes and materials used for direct ink writing technologies: a review. Results Eng. 2021;11:100257.
4. Lewis JA, Smay JE, Stuecker J, Cesarano J. Direct ink writing of three-dimensional ceramic structures. J. Am. Ceram. Soc. 2006;89(12) 3599–609. https://doi.org/10.1111/j.1551-2916.2006.01382.x
5. Campbell I, Marnot A, Ketcham M, Travis C, Brettmann B. Direct ink write 3D printing of high solids loading bimodal distributions of particles. AlChE J. 2021;67(12):e17412. https://doi.org/10.1002/aic.17412
6. Cesarano J, Stuecker J, Calvert P. 3D printing of ceramics: processes and constraints. Adv Mater Proc. 2019;177:28–31.
7. Ji H, Zhao J, Chen J, Shimai S, Zhang J, Liu Y, et al. A novel experimental approach to quantitatively evaluate the printability of inks in 3D printing using two criteria. Addit Manuf. 2022;55:102846. https://doi.org/10.1016/j.addma.2022.102846
8. Lewis JA, Ober TJ. Microfluidic active mixing nozzle for three-dimensional printing of viscoelastic inks. US Patent US2018/0133670 A1. 2018
9. WindEurope–Cefic–EuCIA. Accelerating wind turbine blade circularity. 2020.
10. Weingarten S, Scheithauer U, Johne R, Abel J, Schwarzer E, Moritz T, et al. Multi-material ceramic-based components – additive manufacturing of black-and-white zirconia components by thermoplastic 3D-printing (CerAM-T3DP). JoVE J Eng. 2019. https://doi.org/10.3791/57538
11. Pelz J, Ku N. Additive manufacturing utilizing a novel in-line mixing system for design of functionally graded ceramic composites. Army Research Laboratory. 2019. https://doi.org/10.13140/RG.2.2.18758.42560
12. Lin W, Shen H, Fu J, Wu S. Online quality monitoring in material extrusion additive manufacturing processes based on laser scanning technology. Prec Eng. 2019;60:76. https://doi.org/10.1016/j.precisioneng.2019.06.004
13. Kim H, Cha M, Kim BC, Lee I, Mun D. Maintenance framework for repairing partially damaged parts using *3D* printing. Int J Prec Eng Manuf. 2019;20:1451.
14. Madrigal CA, Branch JW, Restrepo A, Mery D. A method for automatic surface inspection using a model-based *3D* descriptor. Sensors. 2017;17:1. https://doi.org/10.3390/s17102262
15. Li R, Jin M, Paquit VC. Geometrical defect detection for additive manufacturing with machine learning models. Mater Design. 2021;206:109726. https://doi.org/10.1016/j.matdes.2021.109726
16. Alarousu E, Alsaggaf A, Jabbour GE. Online monitoring of printed electronics by spectral-domain optical coherence tomography. Sci Rep. 2013;3:3. https://doi.org/10.1038/srep01562
17. Ye Z, Liu C, Tian W, Kan C. A deep learning approach for the identification of small process shifts in additive manufacturing using 3D point clouds. Proc Manuf. 2020;48:770–5.
18. Smith P, Derby B, Reis N, Wallwork A, Ainsley C. Measured anisotropy of alumina components produced by direct ink-jet printing. Key Eng Mater. 2004;264:693–6. https://doi.org/10.4028/www.scientific.net/KEM.264-268.693
19. Celko L, Gutierrez-Cano V, Casas-Luna M, Matula J, Oliver-Urrutia C, Remesova M, et al. Characterization of porosity and hollow defects in ceramic objects built by extrusion additive manufacturing. Additive Manuf. 2021;47:102272. https://doi.org/10.1016/j.addma.2021.102272
20. Cox B. Committee Chair: ASTM F42.05.05: Proposed standard for technical specification to characterize ceramic feedstock materials for digital light processing (DLP). ASTM/ISO. 2022.

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APPENDIX

NIST Virtual Discussion Panel on Ceramic Direct Ink Writing (DIW), January 26, 2022

(Held as part of the 6th International Symposium on Additive Manufacturing and 3-D Printing Technologies at the 46th International Conference on Advanced Ceramics and Composites, ICACC 2022, Daytona Beach, FL)

NOTE: Panelists and discussion focus titles are listed below but the discussion ranged both more broadly and in greater depth, than the titles suggest, as evident in the main text.

Dale Cillessen (Sandia National Lab., Albuquerque, NM, USA)

Developing and characterizing ceramic feedstock for DLP (digital light processing – lithography) additive manufacturing

Joseph Cesarano (Robocasting Enterprises LLC, Albuquerque, NM, USA)

Are standards for ceramic feedstocks for additive manufacturing feasible?

Nicholas Ku (DEVCOM, Army Research Lab., Adelphi, MD, USA)

Ceramics at the DEVCOM Army Research Lab

Paul Deffenbaugh (Sciperio Inc. & NScrypt, Palm Bay, FL, USA)

High-performance requirements on ceramic-based printed circuit structures

Paula Vilarinho (University of Alveiro, Alveiro, Portugal)

Direct writing (robocasting) of ceramics towards industrialization

Brandon Cox (Honeywell Inc., Kansas City, MO, USA, & ASTM/ISO)

NIST panel discussion: a standards perspective

James Weigner (Lockheed Martin, Clay, NY, USA)

Ceramic additive manufacturing perspectives from an end-user

Alex Michaelis (Fraunhofer IKTS, Germany)

Ceramic multi-materials additive manufacturing (AM)

Brandon Ribic (NCDMM/America Makes, Youngstown, OH, USA)

Ceramic DIW AM technology maturation requirements and standardization

Brian Derby (University of Manchester, Manchester, UK)

Defect characterization and length scale in ceramics additive manufacturing