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Perspective

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Challenges and opportunities in manufacturing highly filled polymers

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Processing highly filled polymers (>50 vol%), whether with particle or short fiber additives, is needed for many applications, such as pharmaceuticals, ceramics, batteries, coatings, etc. Forming these materials into more complex geometries with graded properties is necessary to meet the growing demand for structures with increasingly specialized functions. However, there are many poorly understood processing effects that must be studied to enable the reliable forming of new structures and the qualification of said structures for new applications. Here, we highlight four challenges that impede the processing of highly filled polymers, with a stronger emphasis on additive manufacturing and extrusion processes. They are: (1) understanding process-induced porosity, (2) understanding the solid-liquid interface during the manufacturing process, (3) overcoming equipment limitations, and (4) appropriate use of in-situ monitoring and measurement. We anticipate that by highlighting these research gaps and identifying paths forward, the processing science necessary to form highly filled polymer will advance.

Highly filled polymers are essential across many industries, as they enable maximum volume fraction of functional particles while using a polymer binder to maintain temporary or long-term structural integrity[1]. However, these materials tend to be very difficult to process and shape into products during manufacturing operations due to the high viscosity and tendency for the particles and binder to segregate[1]. Here, we define highly filled polymers as those containing greater than 50 vol% particles or short fibers, though occasionally, we discuss lower volume fractions to highlight specific phenomena that are not yet well-studied at higher packings. With this categorization, applications for highly filled polymers include ceramics[2–5], batteries[6–9], energetic materials[10,11], pharmaceuticals[12–15], dental resins[16–19], concrete[20–23], coatings[24,25], and metals[26,27]. Table 1 provides the typical particle and binder properties and the methods of manufacturing for each. We briefly discuss two of these important applications to provide context for the rest of the perspective: ceramics and pharmaceuticals. These were selected to represent the field with the greatest body of work in this topic (ceramics) and an emerging, but high-value-added field (pharmaceuticals). Specifically, the manufacturing processes, materials, and end applications are extremely different, yet both ceramics and pharmaceuticals would benefit from the same research strategy that we promote in this paper. We then discuss commonly available manufacturing techniques, with an emphasis on additive manufacturing (AM), to form highly filled polymers and the key challenges to overcome.

Additively manufactured ceramics have the potential to deliver custom, cost-effective solutions for high-value applications in healthcare, automotive, and aerospace[2]. Ceramics are presently used due to their excellent high-temperature stability and low thermal and electrical conductivity. AM can significantly decrease tooling and production costs while increasing the flexibility of manufacturing geometrically complex architectures, such as ceramic filters[28]. Additively manufactured ceramics can be prepared using many modalities. While pyrolysis of preceramic polymers is a route to produce ceramics, the most common route is to directly make a ceramic part by burning off the binder material and sintering the solid particles[2]. Therefore, it is desirable to maximize the particle content to minimize shrinkage during sintering. However, the adaptation of high particle content inks to high-resolution AM processes is an open challenge.

Pharmaceutical AM, on the other hand, is not as constrained to very high particle contents. However, by manufacturing on a small scale with the flexibility of AM, pharmaceutical agents can be individually designed to consider the genetic makeup, drug response, and physiology of the patients. This manufacturing approach can also simplify complex drug administration regimens by combining multiple active pharmaceutical ingredients

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Table 1 | Summary of various applications that require highly filled polymers (>50 vol%)

Application and manufacturing modalities							Binder		Particles used
BJP	DIW	Ex	FFF	IJ	SLS	VPP	Properties	Chemistries	
Ceramics Solids: 40-60 vol% Particulate Size: 0.7–100 µm Solidification: Requires binder burnout or pyrolysis of a green body to obtain a final densified structure (desired > 99% density) Desired feature resolution: >1 mm ref. 2–5							Low viscosity, sacrificial	•Aqueous •Organosilicon •Wax •Thermoplastic •Acrylate	•Silicon carbide (SiC) •Silicon oxycarbide (SiOC) •Silicon nitride (SiN) •Alumina •Zirconia
Y	*Y*	*Y*			*Y*	*Y*			
Batteries Solids: 30–50 vol% (40–60 wt%) Particulate Size: 0.1–30 µm Solidification: Requires binder burnout or pyrolysis of a green body to obtain a final densified structure (desired > 99% density) Desired feature resolution:~10 µm ref. 6–9							Low viscosity, sacrificial	•Polyvinylidene (PVDF) •Aqueous •Solvent	•Carbon •$Li_4Ti_5O_{12}$ (LTO) •$LiFePO_4$ (LFP) •Electrolyte salts
	Y	*Y*	*Y*	*Y*	*Y*	*Y*			
Energetics Solids: 60–75 vol% (70–90 wt%) Particulate Size: 5–500 µm Solidification: Requires binder curing (e.g., thermal) Desired feature resolution: >1 mm ref. 10,11							Elastomeric, high viscosity	•Polyurethane •Polyolefin •Polyether	•Salts •Crystalline solids
	Y	*Y*				*Y*			
Pharmaceutical Solids: vol% typically not reported (90–95 wt%) Particulate Size: <100 µm Solidification: Requires cooling after hot melt extrusion or drying processes Desired feature resolution: ~0.6 mm ref. 12–15							Low and high viscosity	•Starch •PVP •PEG •Cellulose derivatives	•Organic molecules •Salts
Y		*Y*	*Y*		*Y*	*Y*			
Dental resin Solids: 30–70 vol% Particulate size: 10 nm–50 µm Solidification: Requires binder curing (e.g., UV and thermal) Desired feature resolution: <1 mm ref. 16–19							Stiff, high compressive strength, low viscosity	•Methyl acrylate •Comonomers	•Colloidal silica •Quartz •Strontium •Zirconium •Ceramic powder •Glass with barium
	Y	*Y*							
Concrete Solids: 65–70 vol% (>90 wt%) Particulate size: 1–16 mm Solidification: Requires chemical reaction (hydration) Desired feature resolution: 0.6–4 cm ref. 20–23							Stiff, high compressive strength, low viscosity	•Polycarbonate	•Cement and aggregate
	Y	*Y*							
Chemical resistant coatings Solids: 4–50 vol% Particulate Size: ~10 µm Solidification: Requires binder curing (e.g., thermal) Desired feature resolution: <100 µm ref. 24,25							Stiff, elastomers, high viscosity	•Polyurethane •Polyureas •Organosilanes •Polystyrene	•Silica beads •Magnesium •Polymer beads •TiO_2
	Y	*Y*			*Y*	*Y*			
Metals Solids: 60–79 vol% Particulate size: <45 µm Solidification: Requires binder burnout or pyrolysis of a green body to obtain a final densified structure (desired > 99% density) Desired feature resolution: >1 mm ref. 26,27							Low viscosity, sacrificial	•Solvent •Wax •Polyoxymethylene •PEG •Thermoplastic	•SS 316 L •Copper •Ti–6Al–4V •Inconel 718
Y	*Y*	*Y*	*Y*	*Y*	*Y*	*Y*			

General solids loadings, binder properties, binder chemistries, particle sizes, particle chemistries, solidification mechanisms, and desired feature resolutions in the final product are included. The typical manufacturing processes included are: binder jet printing (BJP), direct ink write (DIW), material extrusion into molds (Ex), fused filament fabrication (FFF), ink jet (IJ), selective laser sintering (SLS), and vat photopolymerization (VPP).

(APIs) into one dose[29]. APIs are generally organic molecules and their salts, which are bound into pill form with varying amounts of excipients (non-active ingredients) that improve dissolution and disintegration. A small amount (~5–10 wt%) of binder, which is generally polyvinylpyrrolidone (PVP), polyethylene glycol (PEG), or a cellulose derivative, is used. The primary challenges in AM of highly filled pharmaceutical products are the need for a high degree of process understanding and quality control to ensure patient safety and the ability to prepare tablets with mixtures of different API and excipient particles.

Although the two examples discussed above are for AM, there are many processes that can be used to shape polymer composites, which start from components that come in raw forms such as slurries, powders, liquid polymers, or pellets. However, in the general space of polymers with fillers, there are fewer methods to reliably form highly filled polymers. Here, we focus on processes that have been leveraged significantly in the application areas mentioned above and listed in Table 1, although it is noted that there are new processing techniques that could also be used. Material extrusion (Ex) into molds includes processes such as injection molding, compression molding, and slurry casting. Material extrusion into shapes without mold confinement includes techniques such as fused filament fabrication (FFF), direct ink writing (DIW), and ink jet printing (IJ). Techniques such as binder jet printing (BJP) involve forming structures in a supported powder bed by binding particles together through deposition of the raw polymer droplets. Vat photopolymerization (VPP) techniques involve suspensions of particles in a photopolymer vat, which is solidified via a UV source. Selective laser sintering (SLS) involves direct laser processing of powder materials to fuse them together. SLS is provided for context in Table 1 since it can be used to directly fuse particles, such as ceramics, together without binders. However, it is not often used to process polymer-bound composites, so SLS is not discussed at length in this perspective.

Although each application has its own needs and challenges, we have identified a number of common threads that limit progress. To advance the successful processing of highly filled polymers across multiple fields, we need to improve fundamental understanding in the following crosscutting challenges: (1) process-induced porosity, (2) the solid–liquid interface during the manufacturing process, (3) equipment limitations, and (4) in-situ monitoring and measurement. The cross dependencies of these challenges are shown in Fig. 1. We focus on these areas because they are either critically understudied research areas or new research advances are needed to overcome the stalled progress of existing manufacturing processes. After discussion of those challenges and promising approaches to overcome them, we consider themes of commonality between different application spaces and a path forward.

Challenge 1—Understanding process-induced porosity

Porosity is well-known to degrade mechanical properties in additively manufactured materials[30], promote undesirable convective combustion in energetic materials[31], and indirectly lead to failure in solid-state batteries[32]. These defects develop during use and manufacturing, as illustrated in Fig. 2, and they are particularly important in high solids materials due to the large interfacial area. Manufacturing-induced voids are of interest here. The types and causes of defects depend on specific materials and manufacturing processes. Interfacial voids occur between the particulate and binder phases. Carbon fibers, for example, may become misaligned or damaged[33]. Voids can also form between subsequent tracks for AM methods (e.g., FFF and DIW). Regardless of defect type, the material performance will diminish. As such, a primary challenge in manufacturing highly filled systems is minimizing unplanned void formation. The first step is to identify the causes so that the necessary physics and relevant chemistry can be investigated. The following discussion covers two themes of process-induced porosity that overlap across multiple disciplines and applications: surface chemistry and transport.

Poor chemical compatibility between the binder and particulate phases leads to dewetting and void formation[34]. One approach to resolve this issue is to functionalize particle surfaces where a mismatch in polarity exists between each phase, and this approach has been shown to improve particle dispersion in the composite[35,36]. However, the surface chemistry of some materials is not simply the chemistry of the bulk, nor is it always constant through the entire processing operation. The unknown surface chemistry leads to additional challenges in designing particle surfaces for improved compatibility between the particles and the binder. For example, metal/metalloid particles have passivation layers that have different chemistries than the bulk particle, and these layers can react with polymer functional groups, as is the case with boric acid that coats boron particles[37]. In the case of ammonium perchlorate and cyanoethylated polyamine, which require a manufacturing process with reaction, bonding agents have been developed that covalently bond with the particles to improve compatibility[38]. Overall, tuning the surface chemistry to improve compatibility is a promising approach to prevent void formation during processing, but the design process is difficult due to poorly understood particle surfaces.

Transport processes are also tied to void formation in AM of highly filled polymers. Specifically, processes such as the diffusion/advection from flow and concentration gradients, as well as material delivery to a location, are particularly important for AM. For example, in FFF and DIW, voids are formed between layers if the printer nozzle geometry, tool path, etc., are not chosen properly[39]. At a molecular level, inadequate bonding and/or sintering between layers of extruded AM materials are believed to be due to a decrease in molecular mobility near interfaces, a clear issue for higher solids loading[33]. Finally, gas transport can also be a contributor to void formation during processing, both from air that is trapped between particles and binder[40] and from gaseous byproducts from resin curing reactions[41]. Both experimental and modeling approaches to studying and predicting void

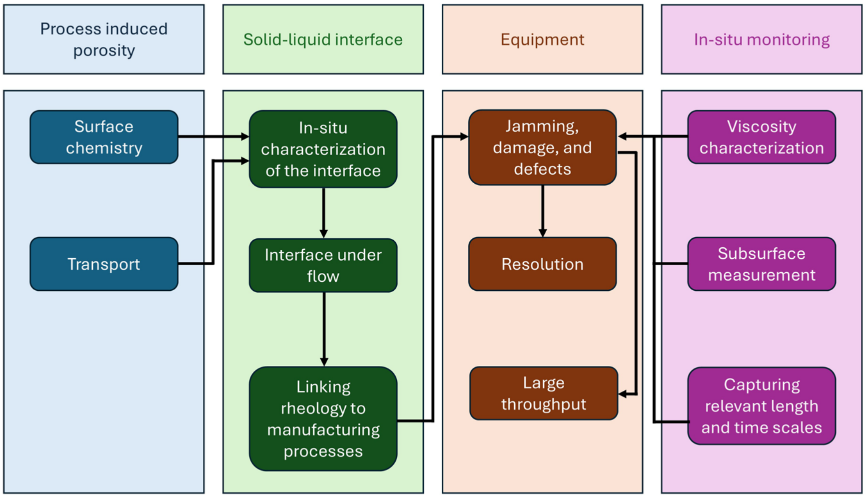


Fig. 1 | Map of the challenges of processing highly filled polymers and their interdependencies.

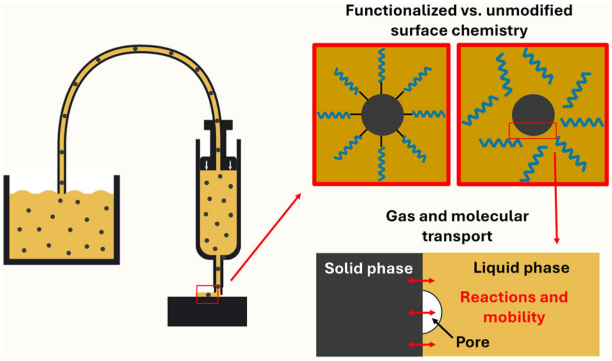


Fig. 2 | Process-induced porosity can be dictated by solid-liquid surface chemistry and gas and molecular transport, which can lead to negative effects such as debonding and pore formation.

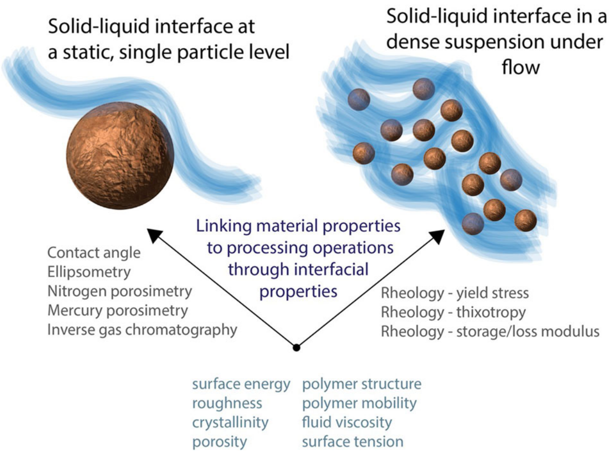


Fig. 3 | Factors influencing the solid-liquid interface under flow that must be understood to control processing of highly filled polymers.

formation due to transport causes are necessary to prevent the formation of undesirable voids during AM. However, approaches with very high particle contents will need to focus more on transport near particle interfaces and the high viscosity during printing rather than binder transport processes that are more crucial in traditional composites with lower particle contents. The breadth of available manufacturing techniques creates many scenarios where different physical and/or chemical processes are critical. Attempts to relate void formation to specific processing conditions should do so with the appropriate dimensionless numbers that relate to the most important competing phenomena and length scales (e.g., interparticle distance vs. layer thickness).

Challenge 2—Understanding the solid–liquid interface during the manufacturing process

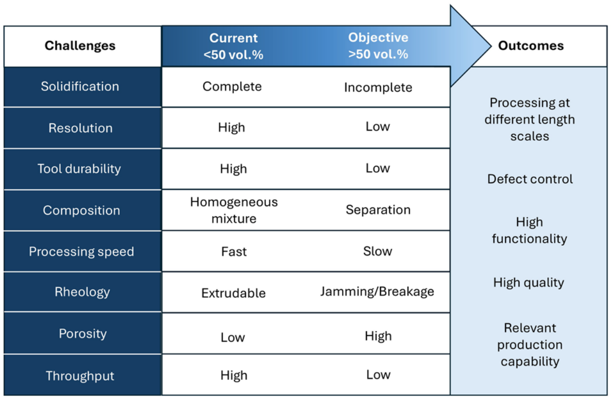
Highly filled polymers have a large amount of interface between the particles and the polymer binder. For example, a composite filled with 60 vol% spherical particles of a given size has 6× more interfacial area than a 10 vol% particle composite with the same size particles. This difference can reach very large magnitudes when small particles, which have high surface area to volume ratios, or particles with abnormal morphologies are used. Thus, the interface is of great importance for highly filled systems. During manufacturing, this interface is typically between a solid particle and a continuous liquid phase, which can consist of melted polymer, polymer in a solvent, or pre-polymers, oligomers and monomers. Since this perspective is focused on manufacturing, we will not discuss the effect of the interface on the composite properties, but the topic has been studied and reviewed elsewhere[42,43]. We will focus on two persistent challenges and opportunities in interfaces during processing as seen in Fig. 3: (1) characterization of the solid–liquid interface and (2) understanding the solid–liquid interface under flow.

To correlate features of the interface to bulk processing and properties, the structure and properties of the interface need to be characterized. Specifically, the characterization of properties such as surface chemistry, surface energy, molecular orientation, crystal structure, surface roughness, porosity, polymer molecular structure, polymer mobility, etc., is needed. The most commonly studied surface properties are the surface energy of the particle or the surface tension between the solid particle and the liquid medium. Contact angle measurements are typically used, but they are highly sensitive to methodology, surface roughness, porosity and compatibility of the surface and liquid used (i.e., reaction and absorption). Hence, the experiments must be very well controlled to ensure proper results[44]. More recent studies of powders with inverse gas chromatography (IGC) have enabled characterization of surface energy heterogeneities[45] and Lewis acid-base interaction numbers[46] of various particles, providing an additional level of detail that can provide enhanced understanding of formulations. Other commonly measured surface properties, including specific surface area and porosity of particles, can be measured by physisorption techniques, such as nitrogen porosimetry with Brunauer–Emmett–Teller theory (BET)[47] and mercury porosimetry[48].

However, all methods mentioned so far are measured for just the particles or particles and binder separately, not for the interfacial properties in situ in the mixtures. Neglecting interfacial measurements is a major weakness in developing a fundamental understanding of how interactions between the particle interface and the continuous phase impact material flow, particle aggregation and final properties, because in mixtures of particles and polymers, the properties at the interface can differ from the bulk, and there can be gradients in properties. For example, work from the Roth group showed that the local glass transition temperature (T_g) of polystyrene (PS) next to silica was 10 K higher when the silica surface had a roughness (R_{rms}) value of 11 nm compared to 0.5 nm[49]. They used a localized fluorescence measurement to determine the local T_g for deposited polymer films, a difficult property to measure locally. The Roth group also used this technique to examine the local T_g across a polymer-polymer interface (PS and poly(n-butyl methacrylate), PnBMA). They showed that the $T_g(z)$ profile, with z indicating distance into the PS layer from the soft material interface, decays continuously and asymmetrically from the higher interface T_g to the lower bulk value over a surprisingly large range of 350–400 nm[50]. In high solids composites, the large distance of T_g differences from the bulk, or the change in T_g based on surface roughness, paired with the high amount of interface, could lead to significantly different behavior than predicted using the bulk T_g. However, further investigation of property gradients from a solid particle to the bulk in high solids materials is still needed.

In processing operations, the solid–liquid mixtures are frequently subjected to flow. Thus, understanding how the interface impacts the flow and rheological properties can aid in manufacturing process design. Correlating interfacial properties with rheology is a challenging task, especially for highly loaded systems where the rheology alone is complex and poorly understood due to multiple factors such as shear history[51], wall slip[52], edge fracture[51,53], shear localization[51], and confinement effects[54]. Rueda et al. provide a more in-depth review on the complexity of rheology measurements of highly filled polymers[1]. Most of the work that studies how surface chemistry impacts the rheology and flow has been focused on concentrated suspensions[55,56], composite melts[57,58], or colloidal particle suspensions[59–61]. To our knowledge, there are none in the highly loaded regime (>50 vol%).

Although few have systematically studied the role of interfacial properties of particles in flow, Johnson et al. focused specifically on the yield stress[55]. They showed that the surface forces acting between colloidal particles, including Van der Waals, electrical double layer, hydration, steric, electrostatic, hydrophobic, bridging, and depletion, significantly influenced the yield stress of suspensions up to 30 vol% particles[55]. For example, when surfactants are added to suspensions of minerals, they result in hydrophobic interactions and lead to an increase in the magnitude of the maximum yield



Challenges	Current <50 vol.%	Objective >50 vol.%	Outcomes
Solidification	Complete	Incomplete	Processing at different length scales Defect control High functionality High quality Relevant production capability
Resolution	High	Low	
Tool durability	High	Low	
Composition	Homogeneous mixture	Separation	
Processing speed	Fast	Slow	
Rheology	Extrudable	Jamming/Breakage	
Porosity	Low	High	
Throughput	High	Low	

Fig. 4 | General equipment considerations that must be accounted for to successfully process highly filled polymers.

stress with surfactant concentration[55]. Further controlled and systematic studies of modified particle systems would enhance our understanding of the role of the interface in rheological properties. However, Johnson et al. focused solely on yield stress as it is the most clearly tied to the interfacial interactions. Moving beyond yield stress requires challenging experimental design to isolate the role of the interface in complex flows, especially for high solids systems, which themselves display complex rheological behavior.

Similarly, few studies have examined the effect of surface roughness[62], polymer binder[63], or other formulation properties on the rheology, especially in high solids suspensions. For example, high surface roughness increases friction between particles and thus hinders flow, and while this has been studied for dry powders[64], it has not been investigated in detail in high solids suspensions. For the polymer binder, one study tied the behavior of the pre-polymer, polyethylene glycol diacrylate (PEGDA), under printing conditions to the rheology and final microstructure of the solidified parts, focusing on the application-specific condition of printing at ultralow temperatures for Lunar environments[65]. Another study showed that a non-adsorbing polymer, polyvinyl pyrrolidone, acts as a lubricant between particles and increases the stress at which shear thickening occurs, which can broaden the window of extrusion-based processing of suspensions[63]. But overall, full studies that link the material and interface properties to the processing operations are scarce, and there is a significant opportunity for further scientific development.

Challenge 3—Overcoming equipment limitations

Several common equipment challenges that impede the production of highly filled polymers and the outcomes of overcoming them are summarized in Fig. 4. When processing highly filled polymers, specialized equipment is often required. For example, high-speed mixers (i.e., resonant acoustic mixer or planetary centrifugal mixer) are required to achieve homogeneous solid-liquid inks for processing at high particle loadings[66–68]. In screw extrusion, the screw must be designed to have the correct pressure profile to carry the material forward, while not damaging the filler[33,69] or entangling the fiber fillers[33,70]. However, despite the high demands on the equipment, there have been advances that enable innovative printing methods. For example, vibration-assisted printing (VAP) is a modified DIW approach that adds vibration to the nozzle tip[71]. This makes it possible to achieve faster deposition speeds and to print more viscous slurries at higher resolution[72,73]. Recently, DIW printers have been equipped with dynamic mixers to enable variation of the composition throughout the print[74,75]. This provides an extended parameter space to tune properties and fully take advantage of the flexibility of AM. Further research is needed to understand how these new techniques affect the solid-liquid interface under flow, but these methods expand the current manufacturing space available for highly filled polymers.

In addition to the high viscosity caused by the high particle loadings, the particles are also abrasive and can wear down the equipment with repeated use. For example, extrusion of carbon fiber-filled polymers requires hardened steel nozzles and regular maintenance. In terms of post-processing manufacturing steps, such as machining, it is well known that specialized tooling is needed to machine carbon fiber reinforced composites, such as diamond-coated tools[76]. Binder jet processes rely on rollers to disperse powders, but they are subject to wear and tear. Furthermore, adding coatings and surface treatments to components is often required to increase resistance to abrasion and corrosion from gas-producing polymers or solvents used as diluents to reduce viscosity[77]. Although these are generally solvable problems, they increase the cost of the equipment and require analysis of the abrasion and corrosion potential of materials used in the manufacturing process, which is not always considered for additive manufacturing process design in a research context. There have been recent feasibility studies on 3D printed polymer molds to enable metal injection molding[78,79]. Although cheaper than conventional machining[78], more research is needed to improve dimensional tolerance, thermal stability, and abrasion resistance. Manufacturing metal molds using laser powder bed fusion would mitigate some of these issues, yet this area has not been thoroughly explored.

Finally, manufacturing at the extremes of size, be it high-resolution fine features or large-format additive manufacturing (LfAM), leads to distinctive demands on the equipment. For fine features through extrusion-based AM, the particle size may begin to approach the size of the nozzle, leading to high demands for extrusion pressure to prevent clogging. For VPP printing methods, high particle content can lead to increased light scattering and require tunable UV wavelengths to minimize scattering to produce finer features[80]. At large scales, it is easier to extrude higher solids loading of particles or fibers since the particles are significantly smaller than the nozzle diameter and screw extrusion processes are used[33]. However, there are still viscosity limits to overcome, and LfAM processes require additional tooling, such as a tamper, to flatten out the bead to produce parts with high geometric fidelity[70]. Finally, LfAM systems for unfilled thermoplastics can produce high throughput rates due to processing pellets, but making large batches of highly loaded polymers, such as pellets[70] or suspensions, as is done for concrete[81], is challenging and limits the throughput rate. Post-processing highly filled polymers that contain sacrificial binders that must be burned out at both small and large scales is challenging. Although higher filler content generally improves dimensional stability, thickness plays a huge role in the burnout process[82]. Furthermore, cracking, porosity, and defects at fine and large scales are a challenge with ceramic AM in general[2].

The promise of AM designs with high solids materials for applications such as those in Table 1 leads to interest in these extreme length scales and thus is pushing forward research to design improved equipment. At least in terms of material extrusion, simple unifying viscoelastic models that consider the rheological and thermo-physical properties of candidate mixtures have been developed to vet various polymer mixtures across length scales for FFF, DIW, and LfAM processes[83]. However, there is still significant work to be done to produce controllable processes across extreme length scales with flexible material feedstocks. Linking understanding of process-induced porosity and the solid–liquid interface under flow will also be crucial for driving new innovations in equipment.

Challenge 4—Appropriate use of in-situ monitoring and measurement

The successful processing of highly filled polymers presents significant challenges. One of the most critical aspects is the ability to perform in-situ monitoring and measurement to ensure the quality, reliability, and efficiency of the process. As filler content in polymers increases, traditional measurement techniques often become inadequate, necessitating the development of advanced sensors and methodologies tailored for these complex systems.

Not all sensors used in polymer manufacturing are viable in the manufacturing of highly filled systems. For instance, optical sensors have proven useful in polymer manufacturing[84–89], but the opacity of highly filled systems reduces their effectiveness. The same is true for embedded ceramic sensors due to more prevalent particle-particle interactions[90]. Machine vision systems, which use cameras and image processing algorithms to monitor manufacturing processes, have been applied to identify anomalies such as porosity, layer misalignment, and surface defects in real time[91]. Spectroscopic sensors, such as those utilizing FTIR, ultraviolet-visible, and Raman spectroscopy, have been used to analyze material composition and detect defects in polymer composites during additive manufacturing[92]. Mechanophores, which are stimuli-responsive molecules that change their chemical structure in response to mechanical stress, offer a novel approach to monitoring the mechanical behavior of highly filled polymers. When incorporated into the polymer matrix, mechanophores provide real-time feedback on stress distribution, enabling precise control over the manufacturing process[93,94]. However, with machine vision, spectroscopy, and mechanophores, the defect or color change must be geometrically accessible, which means that these approaches are not adequate to visualize complex geometries or internal voids.

Viscosity is a key parameter in determining the flow behavior of highly filled polymers. However, as filler content increases, the material often exhibits non-Newtonian behavior, wall slip, and shear banding, complicating the use of conventional rheometers. Advanced rheological sensors, such as those designed for in-process monitoring during extrusion, have been developed to address this challenge. These sensors provide real-time data on material behavior, allowing for adjustments in processing parameters to ensure consistent quality[95,96]. However, with highly filled polymers, the mixture microstructure will vary with external forces, which makes it challenging to define global recommendations for viscosity monitoring due to thixotropic evolution.

X-ray measurements provide a powerful, non-destructive method for characterizing the internal structure of highly filled polymers. These measurements can reveal the distribution of fillers within the polymer matrix and identify any defects that may arise during processing. Techniques such as X-ray computed tomography (CT) allow for detailed internal analysis, making them ideal for quality control in manufacturing environments[97]. Integrating X-ray measurements with other in-situ monitoring techniques like rheological measurements can comprehensively be used to understand the material behavior during processing, improving product reliability and performance[98,99]. The challenge is in ensuring that the collection time scales are commensurate with the manufacturing and material relaxation timescales. While increasing the collection rate is possible to some extent, resolution and field of view are the cost. Synchrotron X-ray measurements have shown great promise in monitoring pore evolution in polymers[97,98], metals[99], and composites[71]. The key considerations here are using a frame rate that is comparable to manufacturing time scales and a pixel size and field that can capture length scales of interest and representative regions. Also, the constituents must have a measurably different density from air and one another to observe defect formation. Finally, while synchrotron experiments show promise for cases when the material is exposed, this technique may not be suitable for methods like injection molding, especially when the mold density far exceeds the density of the paste. It can also be prohibitively expensive to use a synchrotron as a monitoring technique, especially since beamtime is limited in most facilities. The size of the manufacturing platform may further limit the use of this technique.

Dielectric sensors are another candidate for monitoring highly filled polymers during processing. These sensors can track viscosity, the degree of cure, and the glass transition temperature of polymers by applying an alternating electrical field and measuring impedance[100]. This is particularly useful in highly filled systems where other sensor types may struggle due to the opacity of the material or high interfacial area[101]. For instance, dielectric sensors have been successfully used to monitor the curing process in composite materials, which is crucial in preventing defects such as voids or incomplete curing[102]. Overall, selecting measurement techniques that capture relevant time and length scales with reasonable cost and accessibility are important considerations to make to advance fundamental understanding of processing highly filled polymers. A summary of the discussed measurement techniques and their advantages and challenges is shown in Fig. 5.

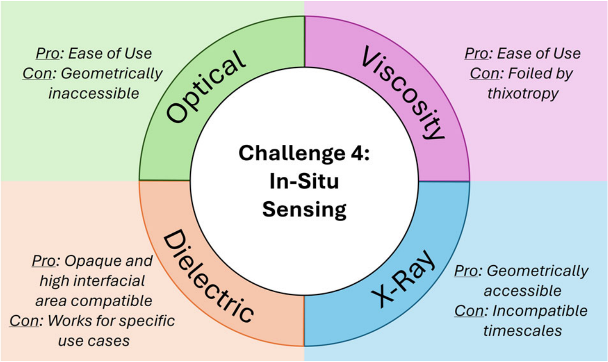


Fig. 5 | Common measurement techniques that can be leveraged for quality measurement and control of highly filled polymers.

Commonality across applications and recommendations for future research

A major barrier to manufacturing highly filled polymers into functional, high-quality parts is linking fundamental phenomena all the way to the processing method. While there is general knowledge about how pores form in manufacturing processes, a deeper understanding of the role of surface functionalization and gasification from chemical reactions is needed. Fundamentally, there are knowledge gaps in linking static measurements of filler or polymer properties to what happens to the solid-liquid interface under flow. Even when that connection is made, the knowledge still needs to be linked to manufacturing processes, which have fundamentally different flow characteristics than typical shear rheology measurements. When considering 3D printing alone, even the different printing techniques tie best to different rheological measurements. For example, rheology related to DIW of high solids materials was reviewed by Marnot et al.[103], covering different techniques than those for stereolithography (SLA) of particle-containing solutions employed by Song et al.[104]. This leads to a broad experimental design space that needs to be explored to fully link materials to manufacturing through rheology.

Limited understanding of process-induced porosity and the solid-liquid interface during the manufacturing process is a barrier to consistent manufacturing. Increased understanding in these areas can be used to overcome common equipment limitations that prevent the production of high-resolution or large-throughput parts. Additionally, in-situ measurement techniques are needed to assess the material state in the manufacturing process or to determine the final internal quality of the manufactured part, yet there are limitations to the type of information that can be obtained. This process is further complicated by the fact that the processes from the solid-liquid interface to the bulk flow in the manufacturing process occur at vastly different length and time scales. Efficient linking of measurements from small to large scales and fast to slow processes is needed, as well as judicious selection of in situ measurement techniques to collect relevant data at the appropriate resolutions.

We generally recommend focusing attention on building knowledge at the intersections between static, dynamic, and processing conditions with the aid of novel measurement techniques or approaches to improve manufacturing control of highly filled polymers. To more efficiently solve these problems, it is important to share knowledge that is transferable across applications. Void evolution during the particle-binder flow process and during processing are phenomena that occur across fields and require deeper exploration. To better translate results, leveraging parameter-based criteria such as normalized maximum packing fraction to describe printability[105,106] will be useful. Furthermore, more standardized viscosity measurements will allow a better understanding of the mixtures. Finally, the understanding gained in some applications will be specific to the material system. For example, surface chemistry and binder solidification techniques will vary. However, developing more standardized measurement frameworks via in-situ diagnostics will expedite the rate at which researchers can use such tools to understand how to control the flow properties of a highly filled polymer for their specific application.

Data availability

No datasets were generated or analysed during the current study.

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Competing interests

The authors declare no competing interests.

Additional information

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