Journal of the European Ceramic Society 44 (2024) 116780

Contents lists available at ScienceDirect

Journal of the European Ceramic Society

journal homepage: www.elsevier.com/locate/jeurceramsoc

Feature article

Review of the opportunities and limitations for powder-based high-throughput solid-state processing of advanced functional ceramics

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ARTICLE INFO

Keywords:
High-throughput
Functional ceramics
Ceramics processing
Accelerated materials discovery
Solid-state synthesis

ABSTRACT

The production of advanced functional ceramics from dry raw powders through the solid-state reaction method is a highly industrially relevant process used by numerous researchers and industries around the world. Functional ceramics enable many cutting-edge applications and are, as such, a critical material class. Despite this, the development of high-throughput platforms for accelerated ceramics development from dry powders has not been intensively investigated and has lagged behind other combinatorial technologies, such as solution- and vapor-based systems, due to the complexity of automating the various steps in this powder synthesis route while maintaining the phase purity of resulting materials. As such, there are numerous important opportunities for the accelerated experimental synthesis and characterization of functional ceramics in a high-throughput platform that will allow for the production of large, homogeneous datasets required for machine learning, which will be discussed in this review. In addition, limitations and challenges of the various processing steps will also be presented.

1. Introduction

Novel materials have always been fundamental to facilitating significant advancements in established technologies and enabling new functionalities, which is one of the reasons why entire ages of human development were named after the materials that defined them. Design and development of such materials for high performance applications and bringing these materials into use in a shortened time-frame is one of the most challenging tasks in materials science and engineering. Here, improved understanding of the controlling mechanisms is central to engineering materials and developing new material systems, as it allows for extrapolation from existing knowledge to unknown domains. The traditional system of experimental materials development is generally based on recognition of a pattern by the researcher through previous studies that results in the identification of a material of interest, its synthesis, characterization at the length scales of interest, and data analysis. Based on this new information and previous experience, a subsequent decision is made about the next material of interest, thereby acting as a feedback loop. There has been a significant interest in accelerating this traditional process using various technologies, including high-throughput synthesis and characterization techniques [1–11] as well as artificial intelligence and machine learning (ML) methods to assist with pattern recognition and decision making [9,12]. The increased use of automated systems has contributed to the development of many areas of high-throughput synthesis. The ability to evaluate a large number of variables in fast and efficient experiments can be realized both in the processing of starting materials and in the study of final compositions. These capabilities can be extended to other areas of research, such as the design of experiments to optimize the synthesis of new materials with desired properties. The importance of accelerating materials development has been well established by various funding agencies and governments [13,14]. For example, the Materials Genome Initiative that was originally launched in 2011 aimed to reduce the development time of new materials by an order of magnitude through integrating experiments, computation, and theory as well as facilitating access to materials data [14]. In particular, these new

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https://doi.org/10.1016/j.jeurceramsoc.2024.116780
Received 26 June 2024; Accepted 22 July 2024

strategies for accelerated materials development have been successfully used, for example, for photovoltaics [15], electrochemical applications [16], Li-ion batteries [17], thermoelectrics [5], and in other areas of materials science [18].

From the various methods for accelerating materials development and discovery, ML methods have received significant attention. ML uses known data sets to learn and make decisions about future directions [19–22]. In fact, the speed of ML has been shown to be orders of magnitude faster that physics-based simulations while maintaining accuracy [23]. There are numerous examples of the successful implementation of machine learning methods in materials science to develop new materials [12,23–25] and identify material properties, such as grain size [26] and ferroelectric hysteresis loops [27], as well as improved fundamental understanding, including an updated description of the Goldschmidt tolerance factor [28], which was originally proposed to describe the crystal structure of perovskites nearly a hundred years ago and used unchanged since [29]. Design of experiment (DOE) or optimal experimental design (OED) is also a tool used for material development as well as industrial processes and quality control that determines parameter-result correlations from a smaller or even singular set of results and shapes the subsequent experimental boundary conditions accordingly [30,31]. This can offer a more systematic experimental approach, especially if experiments are of long duration or high cost. Applications of DOE combined with ML include phase analysis and optimization of functional properties. For example, Tian *et al.* [32] demonstrated that complex, multicomponent phase diagrams can be predicted using Bayesian Optimization for limiting the experimental space while using a machine learning software in an active feedback loop. Similar results were obtained by Yuan *et al.* [33] for optimization of energy-storage properties of $BaTiO_3$-based ceramics. Here, the property search space could also be drastically reduced by experimental design only focusing on the crossover regions of two phases.

Despite these obvious and important successes, decision making methods, such as ML, require a large volume of correlated datasets for training of the ML algorithms. These datasets can come from high-throughput calculations, data repositories and surveys of previous studies, or experiments. Although physics-based simulations can provide the data volume required for ML methods, the results often vary from experimental results due to simulation constraints, boundary condition requirements, or model assumptions, amongst others. Density functional theory, for example, provides a powerful tool for investigating atomic and electronic properties of materials, although the upper limit in size of the simulated system of hundreds of atoms and restriction to ground state investigations at 0 K often result in differences with experimental data [34]. In contrast, data collected from previous investigations or databases are highly inhomogeneous and suffer from numerous issues in limited data volume, consistency, and repeatability [18,35,36]. In fact, the issue of data reliability appears to be worsening. A statistical analysis of published density values for sodium silicate glasses published between 1889 and 1971 were shown to have smaller residuals than those published between 1972 and 2010, indicating a declining reliability in published data.[37] There have been efforts to automate data mining from text [38], which can potentially improve the data volume issue in some specific instances, but problems with data inhomogeneity persist. It can be argued that the data inhomogeneity issues are particularly serious for ceramics. For the development of ceramics, for example, researchers from around the world synthesize materials with different starting raw components, that may have variations, for example, in purity, crystalline polymorph, particle size distribution and morphology. Additional variability arises from the use of different equipment with variations in calibration, contamination and inconsistencies in processing conditions. As a simple example, humidity is an important factor when processing certain materials, such as compounds with alkali metals [39], especially during weighing of raw materials to ensure stoichiometry, which is not universally controlled to the same levels in every laboratory and not typically reported. Similarly, other materials, such as titania, present issues with electrostatic charge that can be induced through friction during mixing and milling of powder, referred to as tribocharging [40,41], that can also affect analytical weighing. Similar variations are also present during characterization, including issues with equipment calibration, user expertise, and measurement resolution. These variations can lead to significant fluctuations in the observed properties of the resulting materials between different research groups and subsequently highly inhomogeneous datasets. It is also important to note that researchers traditionally tend to publish only successful investigations, despite failed experiments also being a valuable learning tool for ML [42].

In addition to data homogeneity and quality, there is also an issue regarding the quantity of data available from previous studies. There are several materials databases that attempt to collate known materials data [43,44], including crystal structure [45], but often related functional properties are not available or rather general. For example, small signal properties can be provided without critical information about, *e.g.*, frequency or temperature dependence or the effect of grain size. This is largely due to the enormous, highly dimensional datasets and typically highly specific focus of individual studies. As an example, the grain size distribution depends on numerous factors such as sintering temperature and time, heating rate, particle size distribution, packing density, impurities and chemical and phase composition homogeneity, atmosphere, pressure, and even the purity of the reaction vessels [46], making the inclusion of such data in a correlated database a daunting task. And critically, the grain size, as only one important parameter in ceramics, affects numerous other properties, such as the stable structural phase [47] as well as the dielectric [48], mechanical [49], and ferroelectric [50] properties. There are numerous other similar influences on the final properties. For example, an investigation on $(K,Na)NbO_3$, showed that the minor variations in A-site stoichiometry are critical in the stabilization of the crystalline symmetries and, therefore, functional properties [51].

In summary, lack of available large correlated datasets, study-to-study consistency, and missing processing and parameter information is a significant limitation to effectively using ML methods. This is exemplified by an investigation by Balachandran *et al.* that presented the development of high temperature piezoelectrics based on the $xBi\left[Me'_yMe''_{1-y}\right]O_3-(1-x)PbTiO_3$ system, where only two B-site cations were varied [12]. Despite the relative simplicity of this solid solution system, there were over 61,500 possible compositions, of which only 167 had been previously experimentally investigated, representing about 0.28 % of the possible materials space.

Generating correlated data sets between processing conditions, crystal structure and grain size, and properties, with so many processing and compositional variables is a difficult task. Therefore, there is a significant and growing need to improve the speed, reliability, and consistency of experimental synthesis and characterization methods to keep up with rapidly advancing computational and theoretical developments, especially in the area of novel bulk inorganic materials that currently lag in comparison to other material classes [9,10]. In particular, in spite of its industrial importance, the area of powder-based solid-state synthesis of advanced ceramics has been largely overlooked, in part due to the well-known difficulty in automating the numerous processing steps [52,53] and in spite of its industrial importance.

In the following sections, the individual steps of functional ceramics processing using the solid-state reaction method will be discussed with a focus on the opportunities and limitations of incorporating them in high-throughput systems for accelerated materials research. Particular attention is paid to differentiating human interaction time and machine time for various processing steps. This provides important parameters for determining the benefits to parallelization and automation. This review specifically focuses on the development of ceramics from dry starting powders, which is a complex multi-step process. Producing ceramic powders from other methods, such as from precursors in solution-based systems will be briefly mentioned, but is not the main focus.

2. High-throughput solid state processing of polycrystalline ceramics

Processing of dense, bulk polycrystalline ceramics is well-established and industrially relevant. Numerous steps are required, starting with chemical and mechanical powder preparation from raw constituents, forming to bring the ceramic powder into a near final shape, densification through high temperature thermal treatment, sample post-processing, and finally characterization (Fig. 1). Each of these steps can also include multiple individual sub-steps that are also critical to the final product, such as calcination, milling, granulation, and removing of solvents by drying. When possible, water should be used as a liquid medium as it is an eco-friendlier medium. In addition, the powder must be carefully controlled throughout the entire process, as lost powder prior to calcination, *e.g.*, if particles adhere to containers, it can potentially change the final composition and properties. Control of stoichiometry is essential for obtaining reproducible results as even small deviations can cause a change in defect chemistry and, consequently, uncontrolled variations of the functional properties. For example, in the multilayer ceramic capacitors (MLCC) industry the Ba/Ti ratio of $BaTiO_3$ must be controlled within +/- 0.0007 [54]. Similarly, deviations in Bi content of 0.1 % in $(Na_{1/2}Bi_{1/2})TiO_3$–$SrTiO_3$ were found to create a core-shell structure that changed the macroscopic electromechanical properties [55].

Due to the complexity, it is difficult to automate the entire process into one continuous workflow, as has been done in other areas such as photovoltaics [56] and polymers [57]. There have, for example, been numerous studies on high-throughput processing of ceramic thin films by varying composition during deposition [58]. However, the properties of thin films can vary from bulk values due to a variety of reasons, including internal strain fields, surface effects, and differences in microstructure. As a result, thin films are not representative of the bulk materials. In contrast, there have been relatively few comparable studies on high-throughput synthesis of bulk polycrystalline ceramics from dry powders [1,3,4,6–8,53,59]. Nearly all of these studies have focused on a batch process that accelerates individual steps of ceramics processing, allowing for reduced human intervention and potential parallelization of multiple steps simultaneously. These methods involve semi-autonomous solutions [6] as well as parallelizing certain tasks to increase efficiency [1]. The primary focus of most studies was the implementation of a gravimetric dosing unit to accurately control the distribution of fine powders into prepared containers [60]. Following this, most studies performed the remaining processing steps manually [1,4,7,8,59], although some investigators also attempted to accelerate various steps by uniaxially pressing multiple samples simultaneously in a silicone die [1] or uniaxially pressing samples stacked on top of one another [5]. Shuang *et al.* presented a ceramics processing system that automated the ceramics dosing with an accuracy of 5 mg, mixing, and forming processes, resulting in a ceramic green body that can be then densified and characterized [6]. This system has also included a cleaning module consisting of a brush to help avoid cross-contamination. A more recent investigation used ASTRAL (Automated Synthesis Testing and Research Augmentation Lab) that fully automates the sample production through the complete ceramic synthesis workflow, facilitating the synthesis and structure characterization of approximately 1 sample per hour [53]. Another recent study also presented an automated system for solid-state synthesis of inorganic powders that was searching for novel materials with a phase purity above 50 % [61].

Despite the difficulties in full automating the entire processing route, there are significant opportunities and advantages in accelerating and parallelizing various stages of ceramics processing, including drastically improving the data homogeneity, volume, and reproducibility to meet the growing needs of data driven materials development. The following section details the individual processing steps for the preparation of bulk polycrystalline ceramic materials, including the currently available solutions for automation and parallelization as well as the potential opportunities and limitations for various processing steps.

2.1. Powder preparation

The first step in producing ceramic parts is the preparation of the powder with the appropriate chemical and phase composition, including doping concentration and stoichiometry as well as the required particle size distribution and morphology. Realizing this involves several steps, generally including initiating and controlling chemical reactions to produce a desired final single phase composition from known starting constituents as well as mechanical processing through comminution and granulation. This section discusses (i) chemical reaction methods to produce ceramic powder, (ii) comminution methods, and (iii) various other aspects of powder preparation, such as milling, drying, and

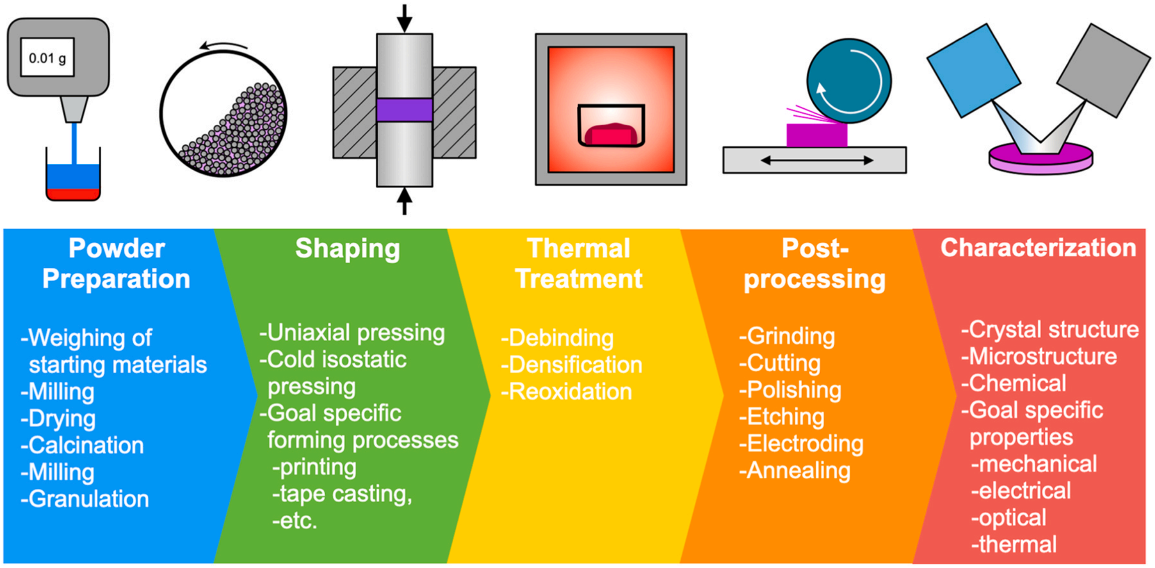

Fig. 1. The general steps of powder-based processing followed by characterization of advanced functional ceramics, including the subsets for each stage.

granulation operations.

2.1.1. Introduction to powder synthesis

There are numerous methods to produce single phase ceramic powder from gas phases, liquid precursors, and solid state reactions [10, 62]. Gas phase reactive methods involve gas-solid [63], gas-liquid [64], and gas-gas [65–68] reactions. Gas-solid and gas-liquid reactions are used extensively for the fabrication of SiC and Si_3N_4 ceramics [69]. In comparison, gas-gas reactions have been previously used to produce various powders, such as TiO_2, SiO_2, Al_2O_3, SiC, and Si_3N_4 [65–68,70]. In fact, gas-phase reactions are an important industrial process for synthesizing large quantities of high purity titania and silica due to the overall simplicity and scalability of the process [66]. Despite this, gas-gas reaction processes are not easily employed for all material systems, such as multi-component oxides, due to the so-called "tyranny of thermodynamics'' [66]. As the need to rapid variations of composition in the range of tens to potentially hundreds of unique compositions per week are required for high-throughput ceramics development, these processes are not considered further here.

2.1.2. Liquid precursor methods for ceramic powder synthesis

Ceramic powder can also be produced using liquid precursor methods, such as sol-gel, spray pyrolysis, and hydrothermal processing, which can be of potential interest in high-throughput ceramics processing systems and are briefly discussed below.

2.1.2.1. Sol-gel synthesis. Sol-gel processing is a well-established method for producing inorganic glasses and ceramics. Different sol-gel methods exist, which are named according to the chemical reagents being used (*e.g.*, alkoxide, acetate, or citrate routes). The term sol-gel originates from the mechanism of the reaction, where, *e.g.*, dissolved metal alkoxides are hydrolyzed, resulting in the formation of a colloidal sol, which on further progress of the process condensate, leading to the formation of a gel. This gel is then heat treated, often leading to homogenous and nano-crystalline powders [71]. For the high-throughput preparation of ceramics, this process is in principle suitable if the different solutions of metal components are compatible with each other. Then, they can be combined with pipetting robots to create libraries. If the solutions are not compatible, and one component's solubility limit is reached prior to the other component (or if one component hydrolyzes faster than the other component), inhomogeneities might arise within the sol-gel-process. These inhomogeneities might influence the screened ceramic properties, leading to false rejection of suitable candidates. For lab scale high-throughput screening, a limiting factor might finally be the size of the individual component of the library, which is often in the sub-gram range [72].

2.1.2.2. Spray pyrolysis. Spray pyrolysis [73,74] is a method for producing ceramic powders, which is also suitable for compounds containing many cations. There are different ways to perform spray pyrolysis experiments, and they can be continuous or non-continuous. These methods have in common that they are based on the aerosolization of a dissolved salt precursor, where the aerosol reacts in a heated compartment / district / zone to form the desired ceramic compound. Different methods exist for the formation of the aerosol, e. g., via a nozzle or an ultrasonic membrane, and how it is reacted to evaporate the solvent and form the product from the dissolved salts, e. g., by spraying it into a flame [75] or by carrying it through a heated reaction zone with "inert" reactor walls [76]. For reaction within a flame, organic solvents can act as a fuel for the flame directly, and alkoxide, acetylacetonate or carboxylate salts of different metals can be dissolved therein. Water can also be used as the solvent for, *e.g.*, hot zone reactors, which is then evaporated leading to the precipitation and final reaction of the salts dissolved therein. The spraying process can be easily performed in a continuous way, which can accelerate the production of ceramic powders through the fact that precipitation and calcination steps are combined in one step. The collection of the particles can occur via electrostatic filtering in a continuous way or via vacuum-assisted filtering on porous materials [77]. The filter needs to be replaced if it gets clogged or whenever a new material is being prepared [78–80]. Numerous advantages exist for this route, such as simplicity, low-cost, and scalability, which make spray pyrolysis attractive for industrial production of several tons of ceramic powder per year. Apart from powder synthesis, it is also possible to directly use this method for the coating of substrates.

However, using spray pyrolysis for high-throughput discovery of ceramics is far from trivial. Reaction conditions like flame temperature, length and temperature profile of the hot zone, type of solvent, and pH for aqueous solutions, can affect the product quality in terms of crystallinity and microstructure. In part, contamination of the reactor tubes or collector units might also have to be taken into account and requires time consuming cleaning steps. This is in principle reflected in studies by Weidenhof *et al.* [75] or Hannemann *et al.* [81], who performed high-throughput screening of various catalyst compositions, with each composition being individually prepared via flame spray pyrolysis. As a result, the screening process was performed at high-throughput, but not the powder production process.

2.1.2.3. Hydrothermal synthesis. Hydrothermal and solvothermal reactions are based on the dissolution of inorganic compounds, which are then placed in an autoclave to react the dissolved metal cations at elevated pressure and temperature to the desired ceramic product. When no external pressure is applied, it is customary to speak of precipitation reactions. Similarly to sol-gel reactions, it is possible to use pipetting robots for the filling of a multitude of autoclave units within a Teflon block [82], leading to a non-continuous batch process for the creation of a library. However, a continuous process has also been demonstrated by Weng *et al.* [83] within a continuous hydrothermal flow synthesis reactor. Tubular reactors are relatively common to scale up precipitation reactions. Similar to other solvent-based routes, the dissolution chemistry of different metal salts and their compatibility must be critically assessed to ensure that multi-metal compositions can react under the same hydrothermal conditions. The hydrothermal synthesis can be quantitatively planned for several systems using thermodynamic modelling to predict the concentration-temperature-pH regions where single-phase products can be obtained [84,85]. Thermodynamic modelling can be coupled with kinetic models based on the population balance approach to understand the crystallization process and develop predictive tools [86,87]. Hydrothermal vessels with temperature and pressure control are commercially available with volumes from about 100 ML to hundreds of liters thus allowing the production of large amounts of powder even at the lab scale.

Despite certain advantages of sol-gel, spray pyrolysis, and hydrothermal processing, integrating liquid phase powder processing routes into a high-throughput synthesis platform is challenging. In particular, precisely controlling the product stoichiometry and dopant concentration are crucial obstacles. Furthermore, the scale-up of hydrothermal processes is often problematic.

2.1.3. Solid state ceramic powder synthesis

Functional ceramics can be produced through the solid-state reaction (SSR) method, which is a relatively simple process involving thermally activated solid-solid interfacial chemical reactions of two or more solid-state reagents to produce a final composition, where chemical reactions can involve decomposition, solid-solid reactions, and reduction/oxidation processes. Although SSR has several advantages, the primary advantage is the ability to produce high purity powder in scalable quantities from a few grams to industrial scale production using relatively low-cost reagents. SSR is broadly used, for example, in the electronic component industry to produce $BaTiO_3$-based MLCCs, piezoelectric ceramics, microwave dielectrics, magnets, and insulating substrates. Despite the process simplicity, however, the chemical reactions can be quite complex and can involve various intermediate reactions [88], depending on the phase diagram and reaction kinetics of the specific system, as well as the formation of liquid phases [89].

Depending on the target composition, raw materials are weighed in stoichiometric proportion, followed by subsequent mixing and mechanical treatment to control the particle size distribution. Finally, the powders are formed into the necessary shape and then heated to an elevated temperature to initiate a chemical reaction that results in a new solid phase as well as potentially gas byproducts, such as CO_2 [88] and volatile components [90]. This process is referred to as calcination. Importantly, calcination works for a wide variety of oxide and nonoxide ceramics and allows for the precise control of composition, dopant levels, and stoichiometry. Starting powders, including simple oxides, carbonates, nitrates, oxalates, sulfates, and acetates, are typically available from multiple industrial suppliers in a wide range of purities, depending on the needs of the particular application [69]. When a large number of samples have to be produced, raw materials from the same lots should be used to avoid uncontrolled variations of the final ceramic properties.

2.1.3.1. Powder weighing. The first step in the calcination process is weighing the starting powders according to the stoichiometry of the desired ceramic composition. Traditionally, this is done manually, which is a tedious and time-consuming process. Here, care must be taken to avoid the influence of external factors that may impact the final weighing accuracy, including, for example, humidity for hygroscopic powders [39] and electrostatic interactions of particles [41]. To avoid such issues, the weighing operation can be performed inside an environmentally controlled chamber, *e.g.*, glove box, with controlled humidity, temperature, and atmosphere, *i.e.*, partial pressure of reactive elements. Among these factors, humidity is a critical consideration for hygroscopic powders, such as alkali oxides [39] and La_2O_3 [91], that can absorb atmospheric moisture, resulting in an artificial increase in apparent powder weight. This is the first primary challenge for any high-throughput ceramics processing system based on dry powder. The powder must be sufficiently dried to remove both absorbed and adsorbed moisture prior to weighing, which may require elevated temperature to remove, *e.g.*, chemically bonded hydroxyl groups [92]. In addition, the powders may contain organic additives used for controlling the crystallization process when powders are produced by precipitation. The temperature required for elimination of volatile impurities can be relatively high (up to 600–700 °C in the case of nanocrystalline powders with large surface area), which may not be desirable since it can lead to changes in the powder size, especially for nano sized powders. To avoid this, a correction for the weight can be applied as an alternative to heat treatment. In this case, the main parameter to be considered is an accurate measurement of the decrease in weight during heat treatment typically done using thermogravimetric analysis. Finally, during the weighing process, a sufficiently low humidity should be maintained to avoid reintroduction of moisture through, for example, the use of vacuum, desiccator media, inert gas, or a dehumidifier unit. Enclosed powder dosing systems can be outfitted with gas ports that can be used to flood the weighing unit with dehydrated inert gas. Importantly, the powder dispersibility [92] and flowability are also affected by moisture content and can, therefore, influence the powder dosing [67].

Several studies have implemented powder dosing units capable of dispensing multiple dry constituent powders with a milligram dosing accuracy [1,4–8,59–61,93], which, depending on the composition and powder amount, have been shown to provide compositional accuracies of approximately ±0.1 mol% for 5 g of powder in perovskite $BiFeO_3$-x-$BaTiO_3$ [93]. Such systems are also used in pharmaceutical research for dosing dry powders [94]. With the integration of a high precision balance, the measurement resolution can be improved into the microgram range, although the actual dosing accuracy may still be in the milligram range. There are several commercially available gravimetric powder dosing systems that are used in pharmaceutical research [5], *e.g.*, from Chemspeed, Mettler Toledo, Labman Automation, and Unchained Labs [95–97], that can also potentially be integrated into automated laboratory workflows for solid-state ceramic powder synthesis. The gravimetric dosing can either be done with a weighing unit integrated into the dispensing head or using an on-deck high precision balance to determine the amount of powder dispensed into the container. In either operation, it is possible that some of the dispensed powder is not transferred to the container due to, *e.g.*, electrostatic interactions with the powder. To reduce this effect, it is possible that dosing systems can be outfitted with an ionizer to neutralize electrostatic particle charging. Fig. 2 shows a Chemspeed dosing platform with a gravimetric measurement head and on-deck balance, where individual dosing units are used to dispense different powders into up to 32 wells. Powder can either be directly dispensed into the containers with the gravimetric weighing unit or moved with a gripper to the on-deck balance for a higher weighing resolution. Importantly, automated powder dispensing systems allow for a rigorous sample tracking system of process steps and attained weighing accuracies without human intervention, increasing the robustness and reliability of the weighing process.

2.1.3.2. Powder flowability. Gravimetric dispensing of dry powders is affected by numerous factors, including the powder flowability [98] and compressibility [99] as well as the dosing system design. Flowability describes the ability of granular powder to flow in a specific system [98]. Despite the simplicity of this description, powder flowability is a complex, multidimensional process dependent on several interrelated properties of the powder, particles, and environment.[100] For example, the bulk density, powder compressibility, cohesive strength, hygroscopicity, electrostatic activity, and surface chemistry, powder particle size distribution and morphology are all important parameters that have been shown to control flowability [101–103]. In addition to these, the temperature and the humidity can also play an important role [102,104] as well as the dispensing system, including the vessel wall friction, electrostatic interactions with the powder, and powder content in the dispensing unit. Moisture can, for example, form liquid bridges at the particle contact points and result in an adhesive force that reduces the flowability [92,105]. The powder compressibility [99], the amount of powder in the dispensing container, and the container design [106], have also been shown to affect the powder flowability. A single measurement to determine the flowability does not exist due to the complexity of powder flowability [98], although there are multiple measurements that can be used to determine powder flowability and compressibility [107–109]. Some of the most commonly used are the angle of repose (AOR) with the fixed funnel method [110,111], the compressibility method (compressibility index, Hausner ratio [112]) [99], and the shear test [113–115], which are shown schematically in Fig. 3. Despite the importance of powder flowability on high-throughput powder dispensing systems presents, there remains little work on the flowability of inorganic ceramic powder necessary for the development of powder-based high-throughput synthesis. This situation is further complicated by the system-to-system variations in the dosing unit design and usage.

To illustrate the effects of powder physical properties on flowability, the AOR was determined using two different commercial Al_2O_3 powders from the same supplier (Almatis): Gilox 63 (99.5 % purity) and CT3000SG (99.8 % purity). Tests were performed based on the ISO 4324:1977 standard [110] (Fig. 4). The Gilox 63 powder had a median particle size (d_{50}) of 17.71 µm (Fig. 4a), whereas CT3000SG had a median particle size of 0.54 µm and a clear secondary peak around 1 µm due to particle agglomeration (Fig. 4d). This can also be observed in SEM images (Fig. 4b,e), which show the larger, irregularly shaped particles in Gilox 63 and smaller, agglomerated particles in CT3000SG. These

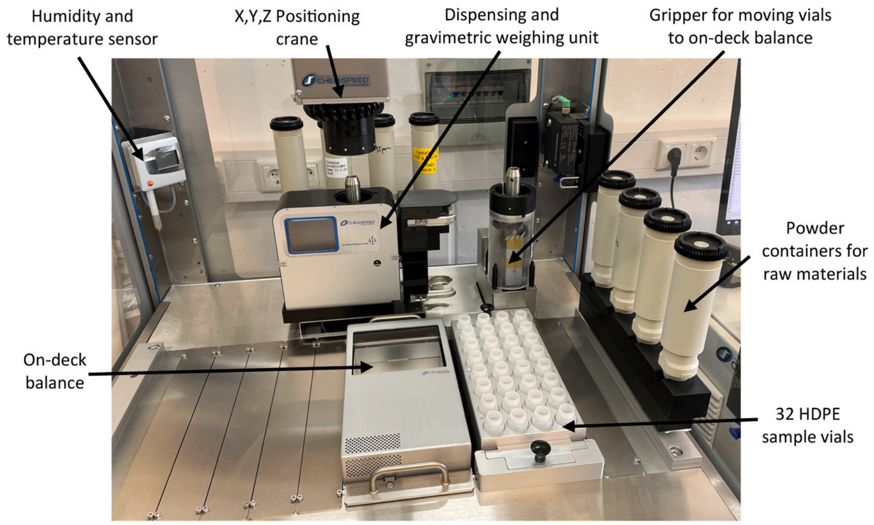

Fig. 2. Chemspeed dosing system with 32 wells and an on-deck balance. Dispensed powder can either be weighed directly from the dispensing unit, which contains an analytical balance with ±1 mg accuracy, or moved with a gripper to an on-deck balance with higher resolution (±10 µg). Powder dispensing containers can be exchanged as needed.

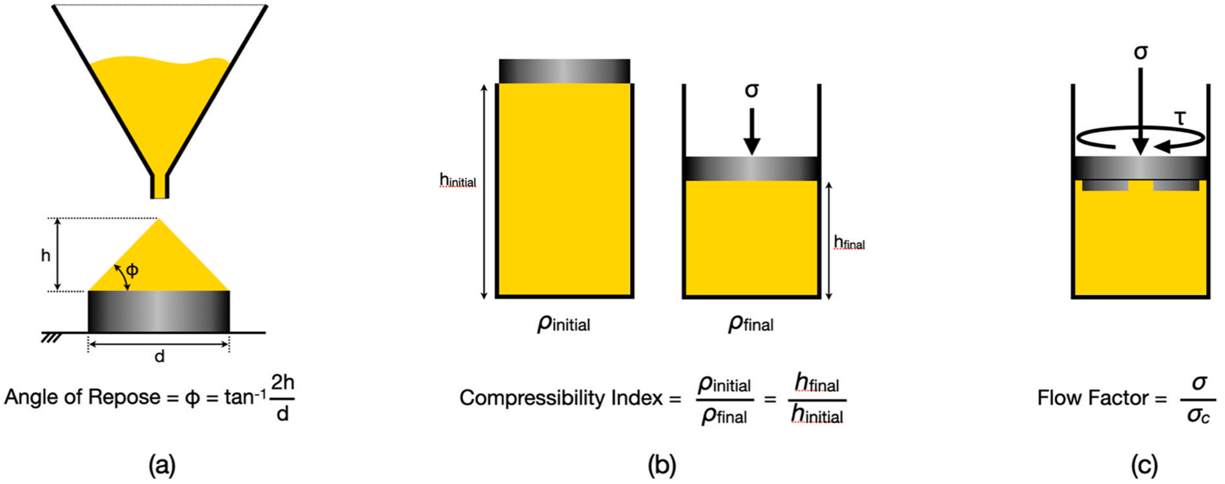

Fig. 3. (a) Flowability as determined by the angle of repose method, (b) the powder compressibility method, and (c) the shear test (Jenike Shear Test). In the shear test, σ is the normal stress compressing the powder and σ_c is the unconfined yield strength under shear.

differences in the particle size and morphology are found to have a significant impact on the flowability (Fig. 4c,f), which was also found to impact the dosing performance. To evaluate this, each powder was dosed 27 times with each dosing having a target value of 3000 mg. Although both powders had a similar average dosing error of approximately 0.3 %, the dosing speed increased from approximately 20 mg/sec for Gilox 63 to 22 mg/sec for CT3000SG. This corresponds well to the observed increase in flowability for CT3000SG, shown by an approximately 10 % increase in the AOR, which is understood to be due to the particle morphology. Additional research is, however, required to better understand the role of flowability on dosing properties. Flowability can be improved by forming spherical granules using a lab-scale spray-drying system. Granulation offers significant advantages in the preparation of green bodies by dry-type forming methods.

2.1.3.3. Powder comminution. During the dosing process, the required components can be directly dispensed into the milling vials in a specific stoichiometric ratio. Following this, the raw materials are mixed and simultaneously milled to produce the necessary particle size distribution for the subsequent calcination step. Although automating the milling process is possible, it is difficult and time-consuming. For that reason, studies to date have focused on extending the capacity of available systems, which reduces but does not eliminate human interaction. Although there are numerous powder comminution methods, milling in a laboratory setting is typically accomplished with a planetary [4], vibratory [5], or rolling ball mill [93]. These methods use milling media, such as stabilized ZrO_2 milling balls and water or organic medium to control the particle size and distribution [69]. The use of a dispersant, *e.g.*, polyacrilic acid, polyethyleneimine, and citric acid, can also help in obtaining a homogeneous mixture. These techniques can also be used for mechanochemically activated calcination processes [116].

Planetary ball mills use rotating milling containers on a rotating disk to impart large impact energy into the powder, reducing milling times and producing smaller particles compared to rolling ball mills [117]. Despite this, planetary ball mills are considerably more expensive than a roll bank, lead to abrasion of the milling vessel and medium that can contaminate the powder, and are a batch process [117]. Some previous studies on high-throughput ceramic synthesis have modified a planetary ball mill to simultaneously process 16 powders [4], although the milling time was still 3 h and required human interaction to exchange samples

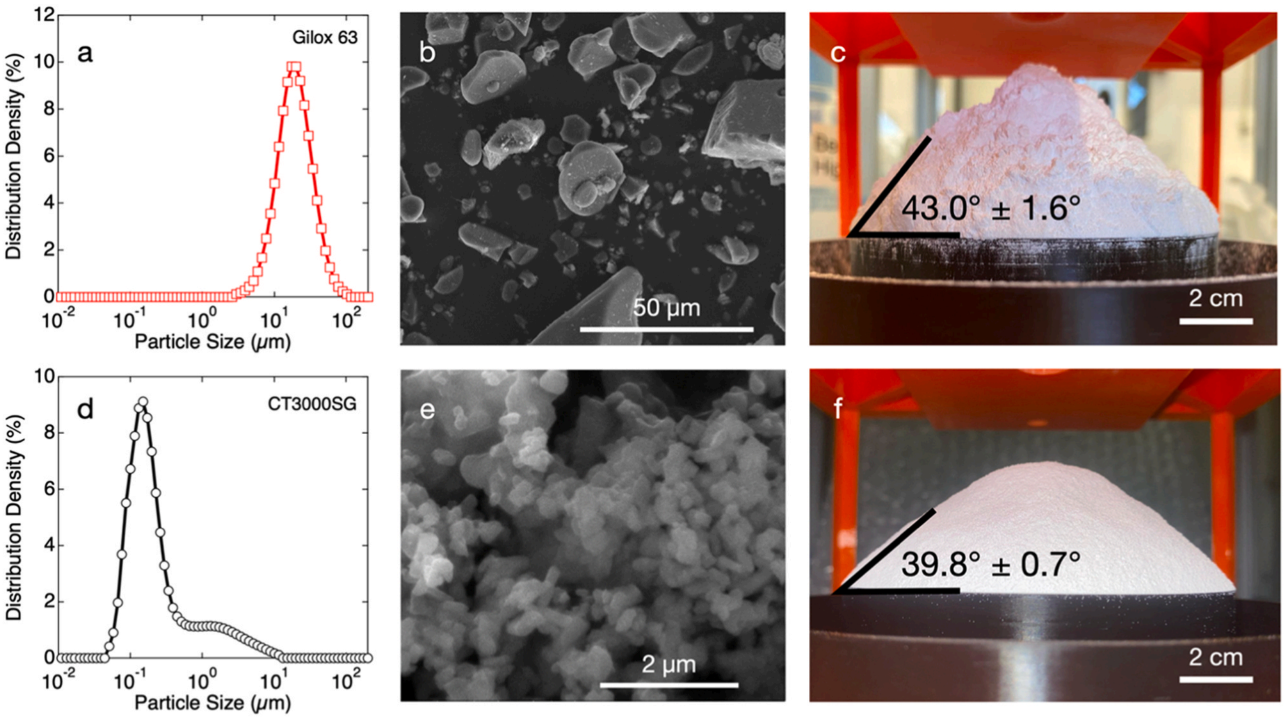

Fig. 4. Powder flowability as determined by the AOR method using commercially available Al_2O_3 powder (Gilox 63 and CT3000SG). Due to the particle morphology in Gilox 63, the flowability was found to decrease. Panels (a) and (d) show the particle size distribution, panels (b) and (e) show SEM images of the particles, and panels (c) and (f) show the resulting AOR values.

for sample volumes larger than the equipment capacity. Stegk *et al.* also modified a conventional planetary mill with a steel fixture that allowed for the simultaneous milling of up to 40 samples [1]. Similarly, a high-throughput ball milling device was developed by Wildcat Discovery Technologies Inc. [118]. Vibratory ball mills, also referred to as high-energy ball mills, agitate the milling container using an eccentric-activated drive mechanism [119], resulting in nano-sized particles. The disadvantages are similar to those for planetary ball milling, including it being a batch process on the time scale of hours that requires relatively expensive equipment. It is possible to extend vibratory mills to include multi-sample adaptors, further accelerating processing. For example, thermoelectric alloys were produced with a dry powder system, whereby the resulting powders were milled with a vibratory ball mill in batches of 8 vials for 1 h [5]. Despite their advantages and potential scalability, both planetary and vibratory ball mills are batch processes with relatively high initial costs. In the case of planetary mills, custom milling vials, from, *e.g.*, ultrahigh molecular weight polyethylene, are required to limit abrasion due to the milling process [1]. Further developments in automation and parallelization are required to accelerate these processes.

Rolling ball mills, in contrast, require approximately 2–20 times longer milling due to the lower imparted mechanical energy, but typically have lower initial costs and can, therefore, be more easily scaled up in production volume and parallelized. This allows for the simultaneous milling of numerous milling vials, significantly reducing the human interaction time, as well as continuous operation (non-batch process). Rolling mills operate through the tumbling action of rotating vials containing powder, milling media, and water or organic medium [69], whereby the impact of the milling media with the powder causes particle crushing. Special milling vials are typically not required and the contamination from abrasive wear of the milling media and vial are relatively lower than in the other processes, reducing cost of consumables. Due to the lower mechanical energy, average particle sizes of 500 nm – 1 μm can be reached, which is considerably larger than from planetary and vibratory ball milling [120]. Nevertheless, these particle sizes are often acceptable for ceramic processing, making the cost, scalability, and low contamination particularly attractive for accelerated material processing. A recent investigation on $BiFeO_3$-$BaTiO_3$, for example, used a rolling mill system (20 ML high-density polyethylene vials with 25 g of stabilized ZrO_2 milling balls ($d = 3$ mm) and 7.5 ML of ethanol) capable of simultaneously milling the entire batch of 72 samples in 24 h, which only required a total of approximately 30 min of human interaction time [93].

2.1.3.4. Drying and granulation of mixed and milled powder. Following the mixing and milling procedures, liquid medium that was used during milling must be removed through a drying process. Special care must be taken during this step to not lose powder or contaminate the raw materials, as it will affect the resulting stoichiometry. This is a time-consuming process that is typically done by manually washing the slurry and milling media out of the milling vial with an organic solvent through a coarse sieve that collects the milling media. Although no automated systems for removing slurry from the milling vials has been developed, automation of this process would lead to a significant decrease in the human interaction time. During the washing process, the slurry passes through the sieve into a container for drying. Subsequently, solvent removal is typically performed either in a rotary evaporator or specially designed drying oven, although there are numerous other drying methods, such as spray dryers, belt dryers, vacuum dryers, infrared dryers, and microwave dryers, that have been developed for drying ceramic powders, slurries, pastes, and tapes. For example, rotary evaporators are designed to be operated with a single container, making the automation or parallelization difficult. Currently, no commercial system for the parallel drying of multiple, independent containers simultaneously with a rotary evaporator is known. In contrast, drying ovens that operate at elevated temperatures can be used to remove solvent from multiple samples simultaneously in a batch process, although care must be taken to avoid loss of powder through solvent evaporation or cross-contamination. Due to the scalability and parallelizability, multiple investigators have used this method [4,59,93], although other studies provide no specific details about the drying step. Importantly, the slurry can be directly washed into the calcination crucible to avoid further powder transfer steps and possible powder loss or contamination.

Spray-drying (SD) is a more effective process allowing for simultaneous rapid drying and granulation, although there are limitations with cross-contamination in the chamber when using different compositions. Therefore, this technique excels for a relatively large number of samples with the same composition. The SD process involves atomizing the concentrated feeding liquid (suspension or solution) into small droplets inside a chamber where hot air circulates leading to evaporation of the water or solvent and the subsequent formation of spherical agglomerates with a size in the micrometer range [121,122]. Typical feed rate for lab scale equipment is 1–2 L/h. The use of SD has also a significant safety advantage avoiding the manual handling of nanopowders. The duration of the SD process is much shorter in comparison to the time needed for oven drying and granulation, resulting in the acceleration of powder processing.

Finally, freeze-drying (lyophilization) is a low temperature dehydration process that involves freezing the suspension and lowering pressure, thereby removing the ice by sublimation [123,124]. It is extensively used in the food industry, pharmaceuticals, and biotechnology. Even with a simple lab-scale lyophilizer, several slurry-containing bottles can be dehydrated in parallel (Fig. 5). There is no need to remove the milling media from the slurry. The absence of a liquid medium results in poorly agglomerated powders after lyophilization due to the absence of capillary forces. The process can be further optimized by atomizing the slurry in liquid nitrogen prior to lyophilization using a spray-freeze-dryer. In a laboratory setting, the productivity is on the order of 1 L of concentrated (40 wt%) suspension per hour. Granulated powders have higher flowability leading to better packing when poured in molds and, consequently, result in a more homogeneous density distribution of the green and ceramic bodies.

2.2. Shaping

With a focus on materials development, the shaping methods of relevance are uniaxial pressing and CIP to produce samples of simple geometries for subsequent characterization steps. Using simple geometries leads to a reduction in complexity and thus to a much simpler sample production. In addition, simple geometries require only simple tooling shapes that are both cost effective and quick to mold. Although additive manufacturing for sample forming is certainly a possibility for unchanging material compositions, it is not a fast manufacturing process when investigating different material compositions or different phase proportions in composite materials.

Fig. 5. Lab-scale manifold freeze dryer that can remove 2 L per 24 h with up to 12 ports. (Image credit: Millrock).

Forming processes are typically performed manually, which can require significant time, although some studies have worked toward accelerating forming through parallelization [1,5] and automation [6]. Ortiz *et al.*, for example, presented a high-throughput approach with custom-built automatic weighing system for producing thermoelectric materials [5]. The as-milled precursor powders were pressed simultaneously in high-density graphite dies, which were compacted during the heat treatment and pressing process. Similarly, parallelizing the forming process, Stegk *et al.* used a custom-built silicone die to simultaneously press 5 samples in a batch process [1]. Shuang *et al.* presented an automated pressing system, where the uniaxial pressing was autonomously done by a pressing rod in up to 10×10 channel molds with a diameter from 5 to 10 mm [6]. Despite these studies, additional work is required in accelerating the pressing operations. In particular, CIP is also often used for further enhancing the green body density of ceramics prior to sintering, which is also a batch process difficult to automate. The dry bag process is a method to form powder filled in a forming rubber mold by transmitting pressure through a pressing rubber mold in a high-pressure vessel. The dry bag CIP process is fully automated and largely used at the industrial scale for high-throughput production of green bodies with simple shape as in the fabrication of alumina spark plugs. Although forming process takes just a few seconds, it is unclear if lab-scale equipment exists. In addition, care must be taken to slowly release the isostatic pressure as to avoid elastic rebounding and the formation of cracks in the green body. Multiple samples can be included in the same CIP cycle to accelerate the process, but further equipment developments and investigations are required.

2.3. High temperature thermal treatment

Processing of polycrystalline ceramic materials with the desired microstructure and properties typically consists of three primary thermal treatments: calcination, binder burnout, and sintering.

2.3.1. Calcination and binder burnout

Calcination is performed during the powder preparation by heating the weighed, milled, dried, and granulated raw constituents to an elevated temperature below the melting temperature inside of a closed crucible [125]. Here, chemical reactions, including oxidation and reduction reactions, can occur through interdiffusion of constituents that result in the formation of a single-phase ceramic powder. Calcination is usually performed in box or tube furnace and is a highly discontinuous process. The need of human intervention and processing time can be reduced by using rotary tube furnaces equipped with automated charge system allowing for continuous powder production, although parallelization through multiple furnaces used simultaneously can also accelerate this step.

It is possible that some reactions may not be fully complete during calcination, which will affect subsequent densification processes and final properties. The formation of extra-phases can be a crucial problem in solid-state synthesis. Even though the desired product is thermodynamically stable, other metastable phases can kinetically form and their conversion into the final stable compound can require long times. In these cases, alternative reaction paths can be explored to circumvent the formation of the metastable phase. A typical example is the preparation of the relaxor compound $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) perovskite. The direct fabrication of PMN from $MgO+PbO+Nb_2O_5$ powder mixtures results in the formation of PMN plus the pyrochlore $Pb_2Nb_2O_6$. The transformation of the pyrochlore to perovskite is sluggish and requires repeated calcination and milling steps. The formation of the pyrochlore can be avoided by a two-step process: (i) the reaction between MgO and Nb_2O_5 with formation of $MgNb_2O_6$ (columbite) and (ii) the transformation of columbite in PMN perovskite by reaction with PbO [126]. The presence of impurities can also lead to the formation of unwanted phases. The preparation of $BiFeO_3$ by SSR between Fe_2O_3 and Bi_2O_3 raw materials containing Si impurities can determine the stabilization of the sillenite phase $Bi_{12}SiO_{20}$ [127]. Given the stoichiometry of this compound and the parallel formation of $Bi_2Fe_4O_9$ for mass balance reasons, even small amounts of impurities produce large changes in the equilibrium phase composition.

In addition, for some materials calcination involves the removal of volatile components. Specifically, CO_2 is released in all reactions where one or more precursors is a carbonate, *e.g.*, $BaCO_3+TiO_2 \rightarrow BaTiO_3 + CO_2$. The formation of CO_2, a gas heavier than air, requires a more careful design of the synthesis process. If CO_2 is not efficiently removed, as it occurs when several centimeters of powder are poured in a crucible, CO_2 accumulates at the bottom of the crucible and can result in an incomplete reaction for thermodynamic and/or kinetic reasons as the increase of p_{CO_2} leads to the increase of the decomposition temperature of the carbonate. Therefore, calcination should be performed in open trays using a powder bed of a few millimeters in thickness, resulting in the lowering of the calcination temperature and reduction of the reaction time. As an alternative, a rotary tube furnace can be used. Therefore, a careful characterization of the raw materials, a knowledge-based optimization of the solid-state reaction (*e.g.*, phase diagrams, thermodynamic modelling, diffusion coefficients, *etc.*), and preliminary kinetic studies (thermogravimetry and DTA, quantitative XRD) can contribute to find optimal reaction conditions and improve the throughput of the whole process [128]. In addition, heat treating ceramics with volatile components, such as Pb [129,130] and Bi [131] as well as alkali metals [132,133] might lead to furnace contamination, meaning that it might not be able to be used for other material systems. In order to maintain stoichiometry, this means that samples will need to be located in a powder bed, which is often done in a closed crucible to increase the relevant partial pressure of the volatile element [134]. Nevertheless, care must be taken to eliminate cross contamination of volatile elements.

The process parameters, *e.g.*, calcination temperature, time, and heating/cooling rates, are dependent on the composition and particle properties and, therefore, must be individually determined for each new composition under investigation. Particle size plays a major role, especially as the reaction kinetics is controlled by solid-state diffusion. High-throughput methods are ideally suited to accelerate this step with the use of automated crystal structure analysis, whereby secondary phases, crystal phases, impurities, *etc.* can be rapidly identified [93].

Following calcination, the powder has typically undergone some local densification and coarsening, resulting in agglomerated powder that must then be re-milled to produce the particle size distribution necessary for forming processes and sintering [125]. In addition, sintering kinetics may be enhanced by milling the powder to smaller particle sizes before sintering. [125] The shaped powder (Section 2.2) is then heated to burn out any additives, *e.g.*, binder or plasticizer, used during the forming process and finally densified during sintering [135]. Typically, the binder burnout and sintering steps can be performed during a single heating/cooling cycling with a lower temperature dwell to remove the binder and other additives before subsequently heating to the final sintering temperature [135].

2.3.2. *Sintering*

In solid state pressureless sintering, the shaped powder (called the green body or green compact) is heated to temperatures below the melting temperature, allowing for densification and grain growth by diffusion at the surface, through the lattice, along grain boundaries, or by gas transport [136]. This leads to a fully or partially densified body with controlled microstructure and properties. The starting powder particle size and morphology, *i.e.*, surface to volume ratio, has significant influence on the densification and grain growth [137,138]. It is important to note that sintering is a complex process that depends on many parameters, such as pressure, particle size distribution, green body density, atmosphere, sintering conditions, *etc.*, which are beyond the scope of this work. There are, however, several comprehensive reviews and books available on densification methods [69,125,139,140].

2.3.2.1. Conventional pressureless sintering. The calcination and sintering temperatures are largely determined by the thermally induced mobility of atoms and ions of the material of interest and typically range between 650–1200 °C and 1000–2000 °C, respectively, for ceramics [141]. Conventional pressureless sintering is typically performed in a box or tube furnace, where a controlled atmosphere, *e.g.*, inert, reducing, and oxidizing, can be achieved. There are various possible heating elements available, including metal alloy resistive heaters, SiC [142], $MoSi_2$ [143], and graphite electrodes [144]. The choice of heating elements depends on the sintering requirements, such as maximum temperature and atmosphere. Importantly, as one of the primary goals of high-throughput processing is to produce a large volume of samples with a variation in composition, there is a clear need to calcine and sinter samples at different temperatures, necessitating either different heated zones for parallelization or rapid heating/cooling for automated processing. One of the main drawbacks of conventional furnaces with resistive heating is the time required to heat and cool, generally limited by the power of the heating elements and the heat capacity of the insulation. Although laboratory furnaces can reach heating rates up to 20 K/min, cooling a laboratory furnace with an inner volume of 8 litres from 1500 °C to room-temperature will typically take between 6 and 10 h depending on mass and volume of sintering fixtures used to support the green bodies. Typically, the complete sintering cycle can be 3–24 hours, which can be a significant bottleneck limiting high-throughput ceramics synthesis.

The pressureless sintering step, however, can be accelerated either with parallelization through the simultaneous use of multiple heating zones or the implementation of rapid heating processes [145–151]. Previous investigations, for example, have used multizone ovens capable of separately controlling multiple independent temperature regions [93]. In addition to this, conveyor belt and pusher furnaces are also available, although they are typically large volume, industrial furnaces that are limited in the maximum temperature and the ability to change temperature for each sample.

In summary, conventional pressureless sintering is well-established, repeatable, low complexity and cost, compatible with many ceramic materials, suitable for wide temperature ranges, scalable to industrial production, and can be easily parallelized. However, it requires longer-times and parallelization can be expensive and energy intensive. Importantly, however, the time required is primarily machine time with little to no human interaction, facilitating parallelization with other processing steps.

There are, however, various rapid heating systems that may be suitable to accelerate the heat treatment steps in oxide and non-oxide ceramics [141], which are discussed in the following section with respect to their applicability to a high-throughput workflow and compared to conventional sintering. Other processes, such as cold sintering [149], hot isostatic pressing (HIP) [150], hot pressing [152], and sinter forging [153], are not discussed in detail, as they are batch processes without rapid heating that are technically challenging to integrate into a high-throughput system. In the case of cold-sintering, however, the scale-up of the process has been recently demonstrated with parallel densification of several samples [154]. The key parameters are attainable temperatures, total cycle energy consumption, automation ability, and repeatability. Table 1 presents a brief overview of the heating methods, including the available heating/cooling rates, temperature range, and typical sintering time as well as the advantages and disadvantages.

2.3.2.2. Laser sintering. Selective laser sintering (SLS) is commonly used

Table 1
Comparison of various densification methods for use in high-throughput processing of advanced ceramics.

Method	Heating/Cooling Rate (K/min)	Temperature Range (°C)	Sintering Time (min)	Advantages	Disadvantages
Conventional Pressureless Sintering	20/20	20–3000	360 to ≥1440	Industrially relevant, well-established, applicable to most materials, easy to parallelize, low complexity, inexpensive, good temperature control, any atmosphere can be used, numerous commercial suppliers	Slow, difficult to automate
Laser Sintering	≥1000 inhomogeneous	Bandgap dependent	~5 sample size dependent	Fast, automation possible	Poor penetration depth, limited by bandgap, thermal gradients
Field-Assisted Sintering / Spark Plasma Sintering	2000/400 (with forced cooling)	1000–1700 Preheating required	20–180	Fast densification, dense fine grain ceramics	Batch process, very expensive, intensive human interaction, carbon contamination, requires post-annealing
Ultrafast High-Temperature Sintering	$\leq 10^4$	1100–3000	1–10	Very fast, automatable, parallelizable	Only reducing atmosphere, carbon contamination, requires annealing, not commercially available
Blacklight Sintering	$\leq 10^4$ inhomogeneous	1800 Bandgap dependent	0.5–1	Very fast, automatable, parallelizable	Limited by bandgap, not commercially available
Microwave Sintering	$\leq 10^4$, inhomogeneous	1400–1600	1–16	Fast, automatable	Poor temperature control, complex system requiring expert user

as an additive manufacturing technique producing three dimensional ceramic parts in a layer-by-layer method, allowing for near net shape fabrication of complex geometries and requires no pressing dies or molds as well as no process additives, such as binder and plasticizer [155–158]. SLS has been used to sinter and melt ceramic powders like ZrO_2 [159,160]. For sintering of ceramics, care needs to be given to the interaction of the laser beam with the material as powder particles will scatter the irradiating beam more than a densified structure, while grain boundaries and other material defects will increase laser absorption, leading to an inhomogeneous temperature distribution [161,162]. For an accelerated synthesis approach using multiple compositions that require disk-shape samples with a thickness of approximately 1 mm, SLS is attractive because raw powders can be directly consolidated into dense form with speeds up to 0.89 mm^3/s, as reported for ZrO_2 [155]. For compositional investigations, however, SLS would require individual powder beds for each composition, considering that penetration depths are typically in the 30 μm to 1000 μm range [161,163]. Therefore, direct laser sintering of disk-shaped samples without prior layer-by-layer deposition of powder is impractical. Direct temperature control and comparability to other densification processes is particularly difficult, especially for multi-component powders with different absorbance parameters.

In spite of these challenges, high-throughput laser sintering has been reported to make a large number of samples with a range of sintered microstructures and microstructure sensitive properties [145,164]. It was shown that starting from a tape cast alumina sheet, after drying, a CO_2 laser was used for laser sintering. Under optimized conditions, laser sintering, as opposed to laser melting was realized. The laser was scanned and it was shown that due to the spatial variation of the laser power, there was a corresponding spatial variation in the sintered density and microstructure. In one scan (less than a few minutes), samples with a range of density (from 60 % to 95 %) with corresponding microstructures were produced [145]. In a related study, the tape cast alumina sheet was micro-machined, using picosecond laser, into 100 micro units (450 μm x 450 μm). This array was laser sintered using the approach described above and as a result ten microstructures were produced and for each microstructure there were ten samples. The hardness of these samples was measured and used to confirm that indeed there were ten different sintered samples with different hardness and for each hardness there were ten samples (within experimental error) [164]. As has been shown, capitalizing on the non-uniformity of thermal profile, laser sintering can also provide a high-throughput approach with low total process time and low human interaction time for making samples with a range of microstructures (and hence properties) [145,164].

2.3.2.3. Field-assisted sintering / spark plasma sintering. Field-assisted sintering (FAST) or spark plasma sintering (SPS) exposes a sample to an external applied AC or DC electric field [146], which rapidly heats the sample. In SPS, the sample is heated by a low-voltage, high-intensity current flowing directly through a graphite die containing the powder, which also acts as a heating element. This design allows one to obtain very high heating rates, due to the optimal thermal transfer existing between the heating element (the graphite die) and the sample, and the low thermal inertia of the graphite die itself. Heating rates as high as 1000 K/min have been reported, with typical values being between 100 and 300 K/min [165]. In some FAST implementations, the sample itself could serve the function of the resistive heater. Samples with low conductivity can either be exposed to voltages not exceeding the dielectric breakdown strength of the sample or be preheated to increase conductivity [166–168]. Experiments with ZrO_2 showed that rapid Joule heating is the primary mechanism for densification during SPS [169]. Sintering times of 5 s – 30 s have been reported to densify metal powders [170], and total cycle times of 20 min are reported for carbide ceramics [171]. The primary appeal is that heating and cooling rates of 1000 K/min and 400 K/min, respectively, are feasible, in addition the applied uniaxial stress can further enhance densification [166,172], especially when the fabrication of dense nanoceramics is required.

Despite the high heating and cooling rate, SPS is a batch process containing a limited number of samples and requires intensive human intervention throughout. The possibility of parallelization with SPS is also exceptionally low due to the very high equipment and tooling costs. Similarly to other rapid sintering techniques, temperature distribution within the sample is not homogeneous [173]. Carbon contamination from the pressing die may occur, requiring an additional reoxidation step after sintering, although physical separation with a buffer material such as ZrO_2 could be employed [168,174]. In the case of functional oxides, a reoxidation step is needed in most cases because a high cooling rate quenches the defect chemistry, in particular the oxygen non-stoichiometry, that forms at high temperature. Non-reoxidized ceramics often show functional properties quite different from those fabricated by conventional sintering, usually displaying increased conductivity, high dielectric losses, and lower density [175,176]. Sometimes the core of the ceramic is darker (grey or bluish) than the surface and this is a clue that the material has a nonhomogeneous defect distribution with a reoxidized skin and a conductive core. Furthermore, exceedingly high cooling rate can lead to the formation of cracks.

2.3.2.4. Ultrafast high-temperature sintering. Ultrafast high-temperature sintering (UHS) is a recently developed method of sintering ceramics at heating rates up to 10^4 K/min at up to 3000 °C [147]. It uses Joule heating of a graphite felt surrounding the sample in an inert atmosphere that prevents the oxidation of the carbon-based electrode. In contrast to SPS, the sample conductivity is not a determining factor. Although temperature gradients are inevitable, thermal insulation, stepping the heating current, or using a graphite powder bed can increase the temperature homogeneity [147,177]. The technique has been used to sinter a wide range of ceramics, including alumina [178,179], zirconia [180], silicon nitride [181], tricalcium phosphate [182], strontium titanate [183], and barium titanate [184] as well as inorganic solid electrolyte $Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ for battery applications [185]. Studies on high entropy oxides with complex stoichiometries such as $(La_{0.2}Nd_{0.2}Sm_{0.2}Eu_{0.2}Gd_{0.2})_2Hf_2O_7$ for application in thermal barrier coatings [186] and $Ce_{0.2}Zr_{0.2}Y_{0.2}Gd_{0.2}La_{0.2}O_{2-\delta}$ [187] have also been reported. UHS shows promise for parallelization and automation, which when coupled with the rapid heating rate, makes it especially attractive for high-throughput synthesis. Nevertheless, the major area of concern for compositional engineering is the inevitable contamination of the specimen with carbon, which could be eliminated using a tungsten-based heater adapted from a SPS system [188]. Further, due to the requirement of reducing atmosphere to prevent electrode oxidation, leads to similar issues as FAST for some oxides.

2.3.2.5. Blacklight sintering. Blacklight sintering uses the temperature-dependent absorption of electromagnetic waves in the visible and UV-light frequency spectrum directly heating the sample, eliminating energy transfer losses compared to conventional sintering [148]. The photon energy of the irradiating light source needs to match or exceed the bandgap energy of the to-be sintered material. Blacklight sintering has been successfully applied to various advanced ceramics [189]. Technical realization includes Xenon-flash lamps with densification times under 1 min and densities between 94 % and 98 % [190,191], which could also be parallelized and automated more easily than most other densification methods. The major drawbacks are the difficult temperature control due to thermal variation in bandgap of the material during heating and the limited penetration depth of approximately 2 mm. The issue with the material bandgap matching the photon energy of the light source remains fundamental, especially when considering a composition engineering high-throughput approach. Despite this, blacklight sintering has considerable potential for high-throughput sintering.

2.3.2.6. Microwave sintering. Microwave sintering is a rapid and efficient technique to directly heat the specimen by converting electromagnetic energy into heat avoiding heat transfer losses encountered during conventional sintering [151,192]. Densities of up to 98 % are attainable for Al_2O_3 with total cycle times below 2 h. The feasibility of sintering commercial 0603 MLCCs with nickel electrodes in microwave field has also been demonstrated. The microwave sintered samples showed a dense, fine, and uniform microstructure and the properties were comparable to the conventionally sintered samples. The microwave processing was found to have enhanced sintering kinetics of the MLCCs, lowering sintering temperature by about 100 °C and also the processing time by about 90 % [193]. While this may be the ideal compromise between capital cost, cycle time, and density achieved, the control and observation of the database relevant parameter temperature is difficult, preventing a clear correlation between process parameters and material properties [194,195]. Despite this, the microwave sintering method can potentially be automated to densify ceramic samples [196], although the complexity and lack of commercial availability of such a system is prohibitive.

2.3.2.7. Comparison of densification methods. These densification methods have a range of human interaction times and total processing times, which are mainly driven by the complexity of the process, including the required sample preparation steps, loading sample into heated zone, the heating and cooling rate and required densification time. The total process time and the human interaction time as a fraction of the process time are schematically shown in Fig. 6. The total processing time, even for rapid heating methods such as UHC and blacklight sintering, is largely controlled by sample forming processes to produce a green body, such as uniaxial pressing and CIP, which are common for nearly all the discussed methods. Without automation, the human interaction times as a fraction of the total process time of these methods are significantly higher than other methods, such as conventional sintering, where the total process time is primarily defined by the long heating/cooling cycle required for sintering. In addition, even for rapid processes that can densify a sample in seconds, the total human interaction time can still be significantly higher than in longer processes, such as conventional sintering, without automation.

As an example, assuming that a researcher must manually load and unload a green body sample into the system, which requires 2 min, and the densification process takes 1 min. For 100 samples, the researcher will require 300 min of human interaction time, which is substantially longer than that required to load the same 100 samples into a sintering furnace, despite the considerably longer densification time. As such, rapid processes with higher human interaction times will greatly profit from automation and slower processes with lower human interaction times will benefit significantly from parallelization to allow multiple devices to operate simultaneously. In addition, the acceleration of shaping processes through either automation or parallelization would be a major advancement in high-throughput synthesis of advanced ceramics, which has been previously recognized [5,10]. There have been some efforts to parallelize the uniaxial pressing step [1,5,6], but additional work is required.

In addition to the overall time, one critical consideration is whether the process can also be used for calcination and binder burnout. If the sintering method cannot be readily used for calcination and binder burnout, additional furnaces and steps will be required for this task, which will significantly increase costs and human interaction time and reduce the attractiveness. For example, FAST/SPS, laser sintering, and UHC have not been demonstrated for calcination and binder burnout. In

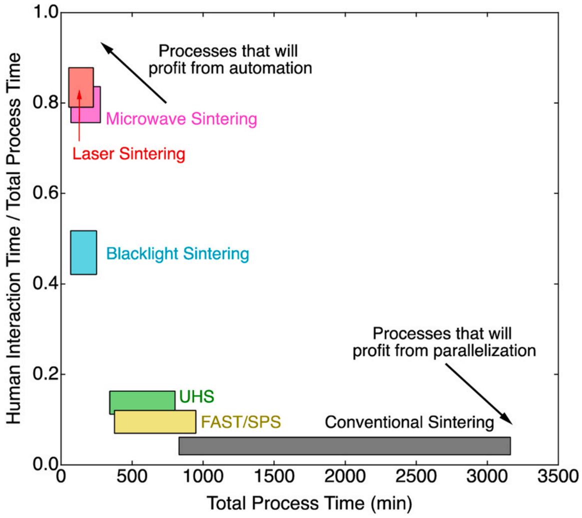

Fig. 6. Comparison of the amount of human intervention time required for each sintering method to the total sintering process time. Processes with low human interaction time and longer processing time benefit from parallelization, whereas processes with longer human interaction times and shorter processing times would profit from automation.

addition, due to the difficult temperature control, calcination using microwave sintering would be a technical challenge, especially considering the sensitivity of many advanced ceramics to the calcination temperature [197].

2.3.2.8. Issues with consumables. One major issue that does not receive sufficient attention is the dramatically increased consumption of consumables during high-throughput processing. For processes at or near room temperature, such as dosing, milling, and drying, consumables can be typically cleaned and reused, *e.g.*, milling balls and dosing units. For custom parts, such as specialized milling vials for planetary or high-energy ball mills for production of nano-sized particles or mechanochemical synthesis [1], the reuse is required due to the prohibitive cost, which should influence the selection of such processes. In addition to this, the use of nanoparticles in dry powder dispensing systems can be complicated by the higher tendency for agglomeration in nano-sized powders [198]. With very fine powders, necks form by elastic deformation, causing agglomeration to increase. This degree of neck formation by simple elastic deformation and adhesion is related to surface adsorption of powders and their cleanliness, which will affect all processing. Some of these technologies, such as rolling ball mills, should also be considered due to the significantly lower acquisition and operational costs. Here, for example, commercially available, low-cost HDPE milling vials can be used in a rolling ball mill, which are significantly less expensive, although overall milling time will increase.

For high temperature processes, such as calcination and sintering, consumables typically cannot be readily reused or must be thoroughly cleaned, *e.g.*, with acid, to avoid contamination. The crucible material must be carefully chosen as chemical reactions between the crucible and sample can occur, which may alter the stoichiometry of subsequent samples. For example, the purity of the alumina crucibles used were found to affect grain growth in ultrapure alumina (> 99.999 %) [46], whereas even the use of platinum influenced the sintering behavior of functional ceramics [199]. In addition, acid cleaning of crucible can only be performed a finite number of times, after which replacement is necessary. In general, the use the same crucible with different materials is discouraged as cleaning and acidic washing are not always effective to avoid cross-contamination.

One alternative is to mass produce crucibles and high temperature reaction vessels in-house, which has the distinct advantage of considerably lower cost per unit as well as the ability to control the crucible material composition, purity, and geometry. There are several 3D printing methods for ceramics that can be used to produce crucibles and other ceramic consumables at the required production volume [200–202]. Some conventional approaches, such as lithography-based ceramic manufacturing [203] and material extrusion (MEX) [204–206], can be time consuming and expensive. In addition, a major disadvantage of the MEX is the rough surface, which leads to the adhesion and retention of components in the crucible during powder synthesis, thus potentially altering the concentration ratios and making cleaning difficult or even impossible. Ceramic injection molding [207] avoids the disadvantages of additive manufacturing, but requires the creation of a mold for the injection molding machine. In addition to very high initial investment costs, it is not possible to change the part geometry without redesigning or modifying the tool.

A possible solution for the affordable, high-volume production of crucibles is ceramic transfer molding [208,209] shown in Fig. 7. This combines the advantages of additive manufacturing (complex shapes and high precision) with those of ceramic injection molding (material independence, cost efficiency, and improved surface quality) or cold isostatic pressing (CIP). In this process, a CAD model of the crucible is produced by VAT photopolymerization of polymer resins or, for lower surface requirements, by MEX, with a small nozzle (0.4 mm) and a lower layer thickness than in direct ceramic extrusion, resulting in a smoother surface. This positive model is used with a special silicone compound to create the negative mold, which can then be filled with a ceramic mold mass, with no restrictions on the crucible material (Fig. 7). After successful casting, the green crucibles are demolded, debinded in a powder bed, and sintered. The silicone molds can be reused until they are damaged. Design changes can be implemented very quickly with a new silicone mold. With this approach, large number of crucibles with good surface finish can be produced at low cost.

2.4. Post-processing and sample preparation for characterization

After densification, ceramic samples typically require post-processing for subsequent characterization. As such, the sample preparation is dependent on the planned characterization methods, but typically includes shaping. This is particularly important for microstructural investigations, which is a crucial aspect of ceramics development. Sample preparation for microstructural examination involves multiple steps, including cutting, grinding, lapping, polishing, and etching. These processes can be time consuming and are typically batch processes, necessitating automation of various steps. For example, Song *et al.* [8] modified a high-throughput tissue grinder with an adapter for ceramic disk samples, allowing for simultaneous surface grinding of 20 samples per batch and potential for automatic, robotized sample removal. Similarly, Jauregui *et al.* [10] demonstrated a translational sample holder for both electron backscatter diffraction preparation and characterization by allowing for a precision in the 10–100 μm range while polishing 32 samples in a row that are directly inserted in the SEM on a translatory stage.

Following preparation of the desired sample shape and surface finish, additional steps are often required, including thermal or chemical etching, application of electrical contacts, or annealing. The required steps depend on the goals of the individual study and the material systems under investigation and, therefore, cannot be discussed comprehensively in this work. Nevertheless, the determination of the grain size

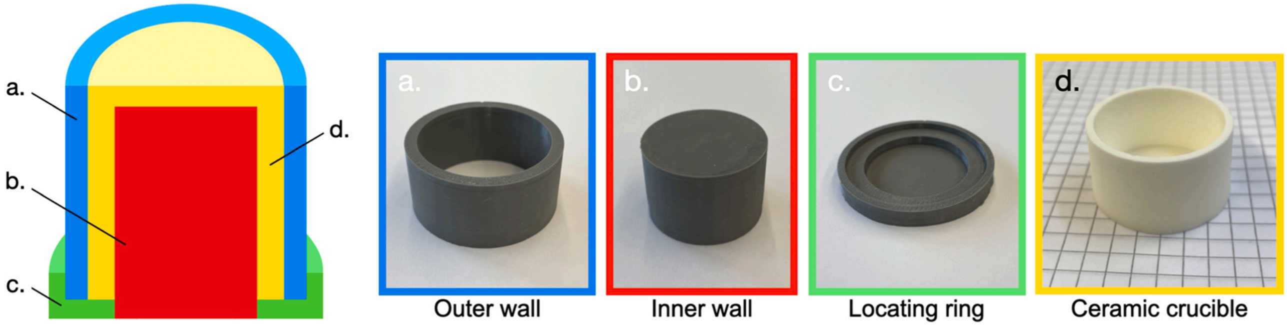

Fig. 7. Production of crucibles using the ceramic transfer molding process with a multi-part negative silicone mold. The slurry (d, yellow) can be poured into the mold, and subsequently the crucible parts (a, b, and c) can be disassembled to remove the ceramic crucible for firing. Using this method, the crucible geometry can be rapidly changed depending on requirements.

is central to all studies on bulk polycrystalline ceramics as it is an important controlling factor in many properties. Established metallographic methods can be adapted to the needs of ceramography, thereby adopting automated metallography operations. Common preparation methods for microstructure characterization involve etching of the polished surface to reveal grain boundaries and the domain structure. The mechanism relies on selective depletion of matter at interfaces either chemically or thermally [1,14]. Thermal etching occurs below sintering temperature to prevent etching induced changes in microstructure [15]. Chemical etching prevents thermally activated grain size alterations but, for ceramics, requires highly toxic and corrosive etching media. As an example, an etchant with HF, NH_4F and H_2O at a molar ratio of 2:1.2:3 is commonly used for ceramics [16]. Recent progress on high-power lasers enables surface etching with beam widths in the cm range through surface ablation in hydrogen atmosphere to chemically modify and subsequently evaporate selective parts of the sample surface [17]. Plasma etching uses high-frequency, high-voltage discharge in a reactive gas species to achieve similar results, but may not be applicable to all materials due to conductivity of the specimen itself [18].

2.5. *Characterization*

One of the major challenges to effectively implement high-throughput methods for developing advanced ceramics is the required characterization methods. Here, accelerated characterization methods or automation of existing techniques are both interesting options, which can be separated in two different general categories: (i) basic characterization that is typically necessary for all ceramic materials and (ii) advanced characterization methods that are specific to the particular material system under investigation.

For a bulk sintered ceramic, the basic characterization requires: (a) determination of the sintered sample dimensions; (b) bulk density; (c) solid density; (d) the nature of the crystalline phases and their relative amounts; (e) and the microstructure, specifically, the grain size, the volume fraction of porosity and pore size, and the distribution of phases in a multi-phase material. These characteristics of the processed material are critically important for establishing the effect of the processing parameters on the microstructure and hence the properties.

Currently, most of the techniques for characterizing these features are manual batch processes that involve significant time and human engagement. As an example, for a disc shaped sample, the thickness and diameter need to be measured. Assuming it takes one minute to measure the thickness and diameter accurately and to input the data into a table, a person would spend more than 1.5 hours for 100 samples. Automation of such a step would be a significant acceleration. Automated sample size measurement systems based on optical 3D metrology and robotic placement are currently in use for, *e.g.*, pick-and-placers-based additive manufacturing [210,211] and should be considered for high-throughput ceramics development. Fast and accurate 3D sample geometry and dimension measurement, before and after densification treatment can be used to obtain quantitative information about dimension change during densification, and together with weight measurement, for green and sintered bulk density. Helium pycnometry on finely ground sintered samples, can be used for solid density and together with bulk density can be used to obtain quantitative volume fraction of porosity in the green and sintered samples.

2.5.1. *Crystal structure analysis*

Determining the crystal structure is also central to the development of ceramic materials, as the crystal phase, lattice parameters, and defects strongly influence material properties. Here, there are commercially available automated sample changing systems for powder X-ray diffractometers, in addition to crystal structure analysis programs with batch-analysis options. Such systems have already been used in high-throughput powder based ceramics development [212–216]. For example, Yanase *et al.* [217,218] reported a powder-based approach using powder dispensing robot in conjunction with a powder XRD sampler, consisting of a moveable plate with 16 recesses for powder that was pressed *in-situ* before diffraction measurement. Importantly, the measurement time can be crucial, depending on the sample characteristics that are to be determined. An example of an automated sample changer in a room-temperature XRD is shown in Fig. 8. This system can measure up to 90 samples without human interaction.

2.5.2. *Microstructural analysis*

Microstructural analysis is needed to determine the grain size, the volume fraction of porosity and pore size, and the distribution of phases in a multi-phase material. Other features may also be important for specific applications and materials including, for example, anisotropy in grain or pore size or ferroelectric domain structure. This analysis can be complex, especially with the presence of multiple phases, chemical heterogeneity, anisotropic grains, and porosity, amongst others. For example, Frömling *et al.* [219] and Koruza *et al.* [220] demonstrated that small variations in A-site stoichiometry as well as sintering time can result in the formation of core-shell structures through chemical gradients that have a significant impact on the macroscopic properties. Such effects should be ideally included in any microstructural investigation. Microstructural characterization involves both the data acquisition and the image processing of appropriately sectioned, polished, and etched polycrystalline samples. There are, however, numerous features that can complicate a microstructural analysis, including domains, porosity, secondary phases, anisotropic grains, and textured microstructure, amongst others. Some examples of ceramic microstructures are shown in Fig. 9.

Following sample preparation, most modern scanning electron microscopes are capable of automated image acquisition and chemical analysis, including positioning and focusing, allowing for the accelerated imaging of several samples and several images of one sample in one operation. Here, the number of possible samples depends on the sample stage and chamber sizes. This type of automated image acquisition has the added benefit of avoiding implicitly biased analysis, as microstructural areas are pre-programmed and not subject to user pre-selection bias. Following this, image analysis presents a significant challenge requiring acceleration through automated methods, as manually analyzing microstructural images is time-consuming. Here, there have been numerous investigations [221–223]. For instance, Lee *et al.* [221] reported an online focusing system that uses deep learning software

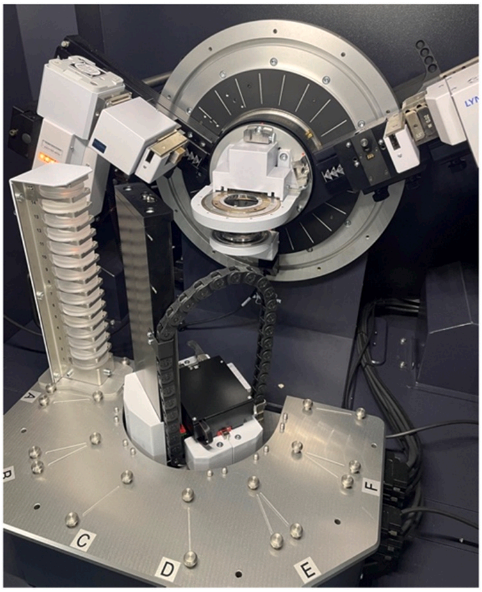

Fig. 8. Automated sample changer for room temperature XRD with a 90 sample capacity.

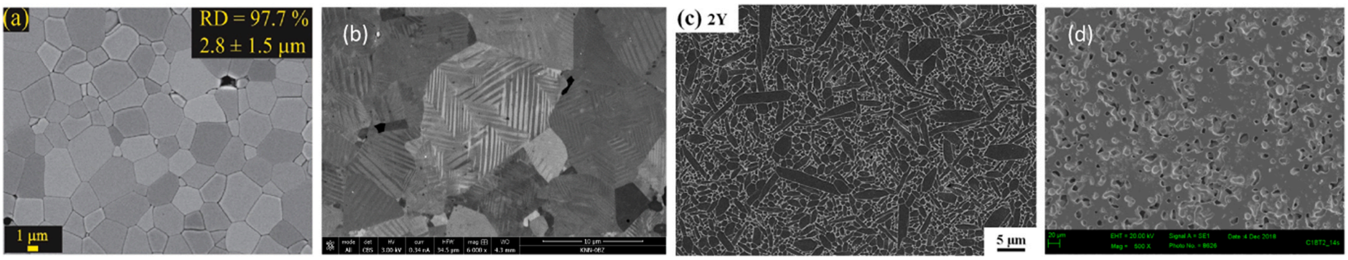

Fig. 9. SEM images of (a) $0.9Pb(Mg_{1/3}Nb_{2/3})O_3–0.1PbTiO_3$ (Copyright 2016, Elsevier) [227], (b) $0.92(Na_{1/2}K_{1/2})NbO_3–0.06Bi_{1/2}Li_{1/2}TiO_3–0.02BaZrO_3$, (c) $0.02Y_2O_3$-doped Si_3N_4 ceramic (Copyright 2018, Elsevier) [228], and (d) porous $BaTiO_3$, showing various common features observed in advanced ceramics, such as domains, anisotropic grain sizes, and porosity.

trained by a skilled SEM operator to identify structures of interest. Commercially available pattern recognition software such as MIPAR has been validated to correctly identify grain boundaries [224] but can also be implemented in a user-customized MATLAB program [225], in addition to opensource programs like Fiji with plugins for automatic segmentation like DenoiSeg [226]. Overall, microstructural analysis times can be reduced to a few seconds. However, the validity of automated image analysis approaches needs to be critically evaluated for the range of microstructures and microstructural features. The field will benefit greatly from access to validated high-throughput image analysis approaches for ceramics.

2.5.3. *Characterization of properties*

As functional ceramics represent a diverse group of materials, it is not possible to discuss in detail all the measurement techniques here. Broadly speaking for bulk ceramics, the properties of interest include electrical, electromechanical, electrochemical, magnetic, thermal, mechanical, and optical properties. Further for each class of properties, several individual properties are of interest and in many applications, these properties are needed under operating conditions, *e.g.*, temperature, time, cycle numbers. As such, it is not within the scope of this work to discuss all of the possible measurement options, both while the number of techniques is large and the measurement of interest depends on the goal of the investigation.

As an example, elastic modulus is an important mechanical property. There are several measurement methods to consider, including dynamic mechanical analysis, resonant ultrasound spectroscopy, impulse excitation method (resonant frequency and damping analysis), contact resonance atomic force microscopy, quasistatic stress-strain measurements, indentation, and Brillouin spectroscopy. Available techniques and conditions under which properties are needed represent a significant challenge for high-throughput studies, as the high-throughput development is only as good as the characterization and subsequent data analysis. This means that investigators working on high-throughput methods have a strong incentive to focus on a particular application that requires the same measurement techniques of functional properties beyond basic characterization. Similar to the processing steps, measurements that require longer times, such as temperature dependent characterization, will significantly benefit from parallelization to allow multiple samples to be simultaneously characterized, while other measurement techniques with shorter characterization times, like elastic modulus, will profit from automation. An additional complication is that some critical properties of ceramics are not dependent on average features of the sample. An important example is the fracture strength of ceramics that is controlled by the largest critical flaw, not the average flaw size or most prominent flaw, meaning that the observed strength requires a statistical analysis with multiple measurements of samples produced under identical conditions. This is an excellent example of a property measurement that will benefit from automation including sample preparation and loading in a test frame.

3. Outlook and opportunities

The acceleration of the processing and characterization of functional ceramics presents a significant opportunity in the development of new materials for specific technologies and in the understanding of physical phenomena responsible for the observed properties. Cutting-edge ML methods demand large volumes of reproducible, homogeneous, correlated datasets, which are not currently available. Although data can be obtained from previously published results and data repositories, it is often incomplete and suffers from inhomogeneity. As such, there is considerable opportunity for the development of accelerated processing and characterization methods for functional ceramics to address this issue, which will drastically improve the data volume, reproducibility, and homogeneity. This has been highlighted by previous studies on development of such systems, which have demonstrated a substantial and largely untapped potential. It is important to note, that accelerating materials research is not only possible but likely a future requirement.

For the development of a high-throughput processing system, several factors need to be considered, such as process scalability, transferability, cost, and required user experience. Processing approaches that can scale easily to larger volumes should be preferred, if possible, as they will facilitate easier transfer to industrial operations. Similarly, the transferability of the processing method is also an important consideration. Here, more common methods that are widely employed by other researchers should be preferred, as this will allow other research groups to quickly reproduce results, further accelerating development. Highly specialized methods pose a particular barrier to entry for many, which will limit participation of other scientists. Cost is a critical issue and systems with high up-front costs will limit their usefulness. The equipment cost presents a significant limiting factor to getting started in accelerated functional ceramics processing and characterization. Finally, well-established techniques that require less initial user experience will also help other researchers contribute, whereas methods that require specialized knowledge will have limitations.

In addition to these considerations, the reproducibility, parallelizability, and automation potential of various processes needs to be considered. Not all processes will benefit equally from parallelization or automation. For example, longer processes with little human interaction time will primarily benefit from parallelization, whereas faster processes that have a higher proportion of human interaction time will benefit from automation. The human interaction times for the various processing steps required in the solid-state processing of ceramics are schematically shown in Fig. 10. Currently, all processing steps (shown on the left side of Fig. 10) are done manually, resulting in approximately 5 hr of human interaction time for each sample (time for process steps, such as furnace run time, where the researcher is not actively involved, are not included). Using currently available acceleration technologies, such as powder dosing robots, milling in the dosing vials, and drying directly in the calcination crucibles in a heated vacuum chamber, the human interaction time can be reduced by approximately 40 % (shown on the right side of Fig. 10). In general, the overall processing time for each sample is approximately the same, however, through accelerated

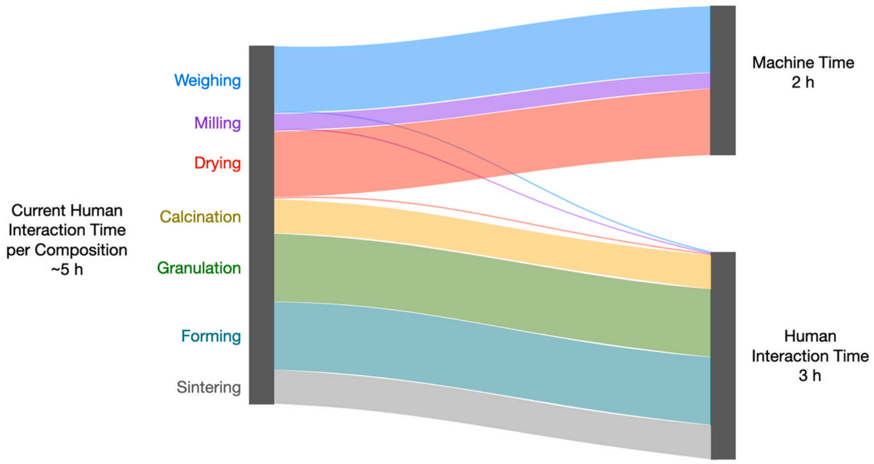

Fig. 10. Current reduction in time for synthesis of one composition through the use of currently available technologies. Some processes remain manually performed and represent areas requiring further development. The machine time is based on experiences with the ChemSpeed automatic powder dispensing system, where 32 compositions can be dosed in one step. The proportional time for a single sample was taken here.

methods, the proportion of human interaction time is reduced, allowing the researcher to focus on other tasks. Additional work is required to further reduce human involvement. Of course, with up-scaling the number of compositions and running various processes in parallel, the human interaction time can be further reduced. There are, however, areas that require additional attention, in particular granulation and forming processes that still account for substantial human interaction times. As previously noted, there have been some developments in accelerating forming processes [1,6], but additional work is required and would represent a significant advancement. The role of data reproducibility should also not be underestimated. A particular potential benefit from such a system is the fidelity in the resulting data that may not be possible with human-to-human and lab-to-lab variations. It is also important to mention, although a detailed discussion is outside of the scope of this work, that a data ontology is also required to successfully store and make the resulting data widely available. Others have also noted the need to integrate high-throughput systems through standardized data management that facilitates cross-platform and cross-user integration [94].

Characterization, of both microstructure and properties, is a major issue that is not yet fully addressed and a central area that requires focus to accelerate materials development. Automated diffraction, spectroscopy, microscopy data collection systems are commercially available, including automated or semi-automated data analysis using various commercial and free software packages. These techniques are important in the basic characterization of ceramics, including the microstructure and crystal structure, although several manual steps are still required, such as grinding, polishing, and etching. Property characterization methods are specific to the particular application of interest. There are limited high-throughout systems for these tasks. As such, there is a pressing need to develop such characterization techniques and many researchers are developing their own high-throughput methods and instruments. Ideally, techniques will be developed that accelerate characterization through, for example, measuring multiple properties simultaneously or in a shorter time-frame through accelerated lifetime testing. Due to the specific nature of these systems, this topic, although very important, has not been discussed in this review.

One of the fascinating things about ceramics is the diversity of materials and functionalities that can be found, in addition to the multilength scale interactions that make the observed performance possible. Through the acceleration of the processing and characterization of functional ceramics, new and exciting discoveries are expected. As examples, a few areas of special interest are mentioned here, although there are many others. Firstly, high entropy oxides have been developed [229], and are being actively investigated as energy storage [230], thermal barrier coatings [186], and electrocatalysts [231]. These materials have a large possible compositional space that would significantly benefit from high-throughput methods to better understand the physical phenomena responsible for the properties. Similarly, phase boundaries are critical to numerous properties in functional ceramics [232], but the influence of numerous factors, such as stoichiometry, composition, and processing conditions, is not well understood. Accelerated techniques would help to address these issues. And finally, sustainable functional ceramics development is an increasingly important area that encompasses numerous issues. For example, the development of new materials would allow materials engineers to focus on non-toxic and conflict-free elements, while maintaining or improving functionality. In addition, accelerated materials development methods could be used to simplifying increasingly complex and, therefore, less well-understood compositions, without a loss of performance. This is particularly important when end-of-life issues, such as recycling, are considered, where complex compositions are difficult to recycle. Finally, more sustainable processes could be envisaged, whereby materials with lower energy consumption or lower CO_2 emissions during processing could be developed.

4. Conclusions

High-throughput processing and characterization of advanced functional ceramics is highly relevant for accelerating materials development but has been to-date not widely implemented or investigated. This is largely due to the complexity and cost of automating and parallelizing ceramics processing steps. However, there are numerous opportunities in this research space that are detailed in this review. There are currently commercial systems available that automate some tasks, such as powder dispensing, as well as several methods for rapid thermal processing. Despite this, there remain several bottlenecks, particularly in the preparation of the powder and shaping operations that require significant human interaction time. Property characterization is a major bottleneck for high-throughput materials development and is specific to the individual goals of the research. However, there have been significant advances in automated and semi-automated methods in diffraction, spectroscopy, and microscopy for data collection and analysis, which are accelerating materials development.

Despite the technical difficulties, there is a significant need and potential for further development of high-throughput methods for functional ceramics. It is suggested that the best practice is to, therefore, not automate at all costs but where necessary and practical. Adaptive task sharing between humans and machines is an established technique in industrial manufacturing with a lot of knowledge and practices available to be transferred into automated materials research. In the field of advanced functional ceramics, automation can increase data integrity and reduce process times while still relying on human interaction where automation would have little impact on total system performance. Based on the research opportunities and financial considerations, scientists should, therefore, identify weaknesses and bottlenecks in their workflows and focus on them.

CRediT authorship contribution statement

Kyle Webber: Conceptualization, Funding acquisition, Project administration, Supervision, Visualization, Writing – original draft, Writing – review & editing. **Oliver Clemens:** Writing – original draft, Writing – review & editing. **Vincenzo Buscaglia:** Writing – original draft, Writing – review & editing. **Barbara Malič**: Writing – original draft, Writing – review & editing. **Rajendra K. Bordia**: Writing – original draft, Writing – review & editing. **Tobias Fey:** Writing – original draft, Writing – review & editing. **Udo Eckstein**: Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

KGW, TF, and UE gratefully acknowledge the financial support of the Deutsche Forschungsgemeinschaft under GRK2495 D/F/H/S. Also, KGW thanks Prof. Jürgen Rödel for helpful discussions. OC thanks Prof. Klaus Stöwe. BM acknowledges the financial support of the Slovenian Research Agency (core funding P2-0105). RKB acknowledges partial support from the US National Science Foundation Grant No. 2119833.

Summary novel conclusion

- Detailed discussion of opportunities and limitations for accelerating ceramics processing steps of advanced functional ceramics
- Discussion of advantages to parallelization versus automation in reducing human interaction time
- Discussion of various issues related to acceleration, including consumables, characterization, and heating method

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