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Editorial

Modelling of Microstructure Formation in Metal Additive Manufacturing: Recent Progress, Research Gaps and Perspectives

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Abstract: Microstructures encountered in the various metal additive manufacturing (AM) processes are unique because these form under rapid solidification conditions not frequently experienced elsewhere. Some of these highly nonequilibrium microstructures are subject to self-tempering or even forced to undergo recrystallisation when extra energy is supplied in the form of heat as adjacent layers are deposited. Further complexity arises from the fact that the same microstructure may be attained via more than one route—since many permutations and combinations available in terms of AM process parameters give rise to multiple phase transformation pathways. There are additional difficulties in obtaining insights into the underlying phenomena. For instance, the unstable, rapid and dynamic nature of the powder-based AM processes and the microscopic scale of the melt pool behaviour make it difficult to gather crucial information through in-situ observations of the process. Therefore, it is unsurprising that many of the mechanisms responsible for the final microstructures—including defects—found in AM parts are yet to be fully understood. Fortunately, however, computational modelling provides a means for recreating these processes in the virtual domain for testing theories—thereby discovering and rationalising the potential influences of various process parameters on microstructure formation mechanisms. In what is expected to be fertile ground for research and development for some time to come, modelling and experimental efforts that go hand in glove are likely to provide the fastest route to uncovering the unique and complex physical phenomena that determine metal AM microstructures. In this short Editorial, we summarise the status quo and identify research opportunities for modelling microstructures in AM. The vital role that will be played by machine learning (ML) models is also discussed.

Keywords: ICME; CALPHAD; cellular automata; phase field model; kinetic Monte Carlo; computational fluid dynamics; finite element method; machine learning

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1. Introduction

The strategy of depositing metallic material layer by layer to make components bestows additive manufacturing (AM) with numerous advantages (e.g., [1–3]) but also creates several challenges (e.g., [4–6]). One of these difficulties is pinpointing the exact nature of the influences determining microstructure formation in AM parts [7]. This is not surprising, considering the complex thermal and fluid flow phenomena preceding the solidification of a highly nonequilibrium nature in a powder bed fusion (PBF) process [8–10] (Figure 1).

Microstructures resulting from solidification are a function of the heat extraction rate, alloy chemistry, and nucleation conditions. They determine the properties (e.g., mechanical, thermophysical) of the AM part and, ultimately, its fitness for use. Since solidification rates are influenced by the position of the fusion zones on the part being built, the AM microstructures are strongly location-specific. Additionally, as the heat source (e.g., laser beam or electron beam) scans the powder bed in directions perpendicular to

the build direction, the asymmetry in heat transfer paths is reflected in the microstructural morphology that results in anisotropy. In addition, microstructures in AM form under rapid solidification conditions as the microscopic melt pools are quenched by instantaneous heat loss to the previously fused layer (or the build plate in the case of the first layer). They are sometimes modified by the heat received from the newly depositing adjacent layers as they remelt (at least partially) and resolidify [11]. Körner et al. [12] pointed out that grains may nucleate on segregated microstructure in the remelt zones since, during remelting, some precipitates or particles that do not fully dissolve may serve as heterogeneous nucleation sites. An example of the distinctive AM microstructures around the microscopic melt pools is given in Figure 2 for the AlSi10Mg alloy solidifying in an L-PBF setting. Thijs et al. [13] explained that the directional solidification in the melt pool caused not only a morphological texture but also a crystallographic texture.

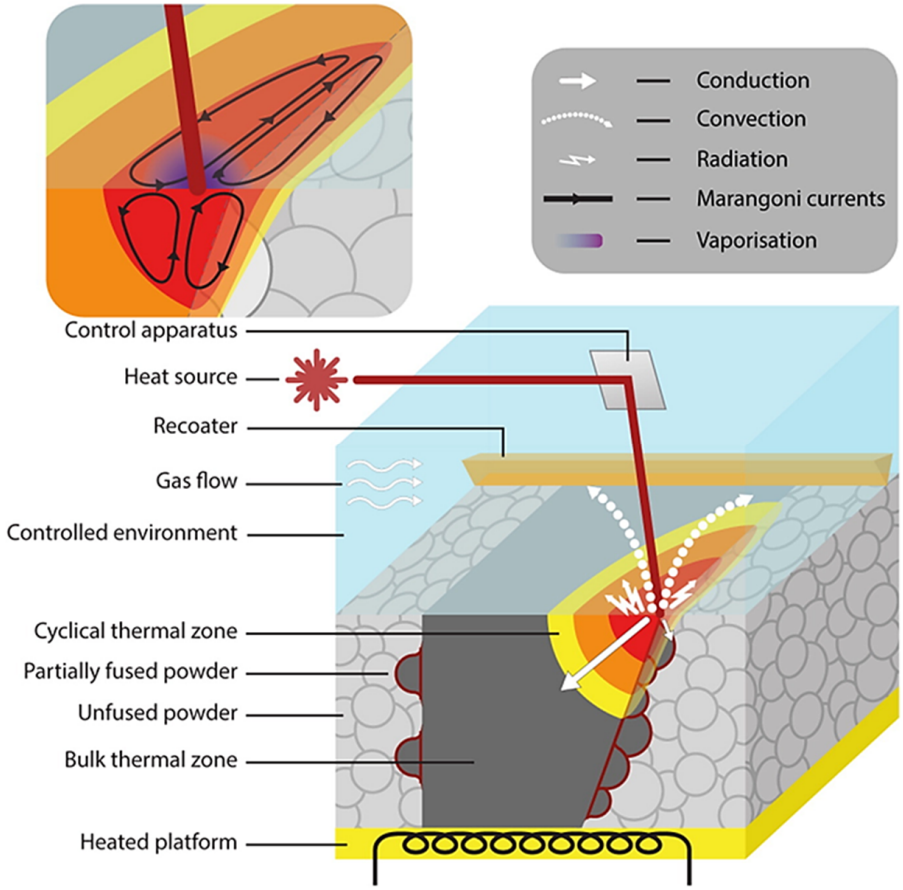

Figure 1. A schematic showing the complex thermal and fluid dynamics phenomena that occur in a powder bed fusion process. Reproduced from [9] with permission from Elsevier. The heat source can be a laser beam or an electron beam. In the latter case, the build platform is usually preheated.

Due to the rapid freezing rates and other factors mentioned above, the AM microstructures significantly differ from those obtained in traditional processes (e.g., casting) in grain morphology distributions, defect populations and crystallographic texture [14,15]—a comparison is provided in Figure 3 for a popular AM alloy, Ti6Al4V. Other alloys are considered elsewhere, e.g., [16].

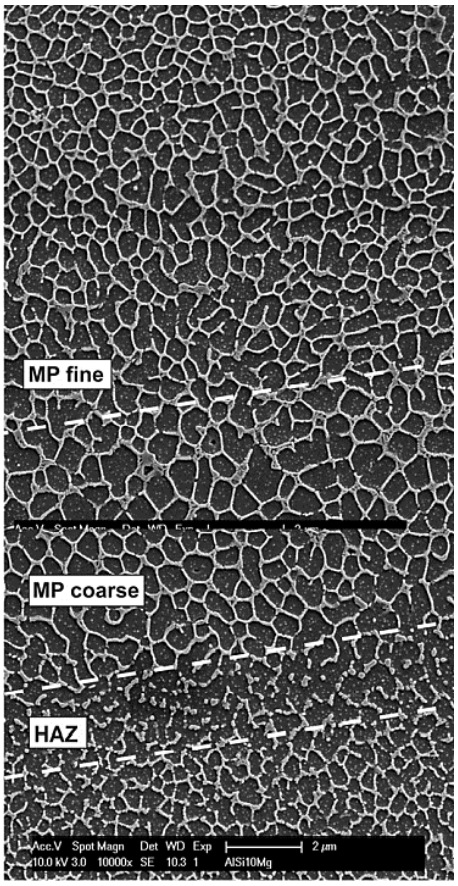

Figure 2. A Scanning Electron Microscope (SEM) image of the melt pool (MP) zone with fine (MP fine) and coarse (MP coarse) microstructures and a heat affected zone (HAZ) in the boundary with the previously fused layer. Reproduced from [13] with permission from Elsevier.

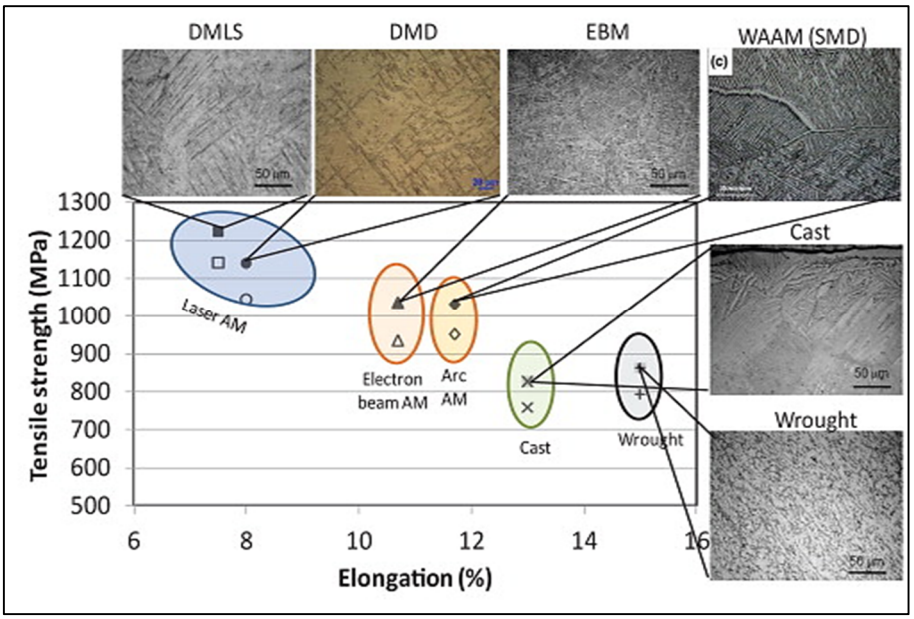

Figure 3. Microstructures and room-temperature tensile strengths of AM Ti6Al4V alloy produced using various AM technologies compared with those of traditional methods. Reproduced from [17] with permission from Elsevier. (DMLS = Direct Metal Laser Sintering, DMD = Direct Metal Deposition, EBM = Electron Beam, WAAM = Wire Arc AM, SMD = Shaped metal Deposition).

Most innovation in materials science and engineering resides in our ability to understand and control the intimate relationship between the structure of materials and their properties [18]. The first step towards achieving such innovation in the sphere of AM involves understanding the relationship between the process parameters and the microstructures they create.

In this article, we first explain why computational modelling can play a major role in advancing the science underpinning microstructure formation in AM. We then provide a brief overview of the status quo in modelling efforts aimed at AM-related microstructure and discuss challenges and research gaps. Since excellent comprehensive reviews of the topic were published recently (e.g., [12,19–21]) and several exceptional texts that address microstructure formation [21] and methods for modelling [22] exist, we restrict ourselves to providing updates on recent progress and complementary insights. For completeness, a summary of the relevant modelling methods is also provided.

2. The Role of Computational Modelling in Advancing the Understanding of Mechanisms

As discussed in the previous section, the AM process comprises complex heat transfer and fluid dynamics phenomena and extreme solidification conditions. Additional complexity is inherited from the fact that the various AM processes offer multiple phase transformation routes by allowing numerous combinations of process parameters (e.g., laser power, scan velocity, scan sequence, hatch spacing) to be employed to build an AM part. Gaining insights from observing the AM process in-situ is made difficult by several factors, including the microscopic nature of the melt pools, the rapid pace of solidification, and the highly dynamic and unpredictable nature of powder-based building processes. The microscopic size of the melt pools and the rapidity of solidification require high-resolution/high-frame-rate sensors to record process signatures such as temperature.

Given the large multidimensional space occupied by AM process parameters, the complex nature of the thermal and fluid dynamics phenomena associated with the PBF process, the asymmetry in heat input and extraction leading to anisotropy, the uniqueness and rapidity of solidification conditions, cyclic reheating, and the difficulties in observing the process in-situ, it is understandable that much of the knowledge concerning AM microstructure formation is still developing. The AM community readily acknowledges the need to create an in-depth understanding of microstructure development to fabricate high-quality products by AM processes—e.g., see [12,21,23–25]. Such exceptional quality products are demanded by industries that use mission-critical AM parts, e.g., space, aerospace, automotive, medical and defence [26].

Against this backdrop, physics-based computational modelling is uniquely positioned to help bridge part of the knowledge gap. For instance, simulations can reconstruct the physical processes by combining known inputs (e.g., rate of heat input, estimated heat loss to the ambient) and predict responses (e.g., temperature evolution) of AM systems by solving the associated governing equations (e.g., Fourier heat conduction). A correctly reconstructed (i.e., experimentally validated) virtual replica of an AM process allows the researchers to 'see' the actual process by interrogating 'virtual probes' for various response histories (e.g., temperature, displacement). Unlike the limited number of high-end physical sensors that may be used in the experiments, there can be a multitude of virtual sensors providing an unparalleled 'view' of the process. For example, any node in an FEA analysis can be used to obtain histories of the field variables. In addition, the analyst can 'freeze' the process at critical junctures for in-depth studies of events leading to an undesirable event.

Importantly, modelling provides a cost-effective scientific method for gaining deep insights into microstructure development in AM. It allows for the testing of various theories on the roles played by different phenomena through the comparison of predictions based on these concepts with laboratory observations (Figure 4). When used in tandem with well-designed validation experiments, modelling can help accelerate the identification of causal mechanisms by recognising the most critical influences from the many potential candidates. Using the well-established 'design of experiments' methods for numerical

simulations as demonstrated elsewhere [27], the number of simulations required to obtain the understanding may be reduced, further quickening the pace of innovation. Based on the knowledge gained through such screening, modelling can further assist with manipulating those key parameters for gain by facilitating 'what-if' studies. In the process of creating models, new theories may also be developed and tested quantitatively by comparing predictions using these with observed microstructures. Ultimately, robust predictive models will be able to help practitioners tailor microstructures to obtain desirable location-dependent properties.

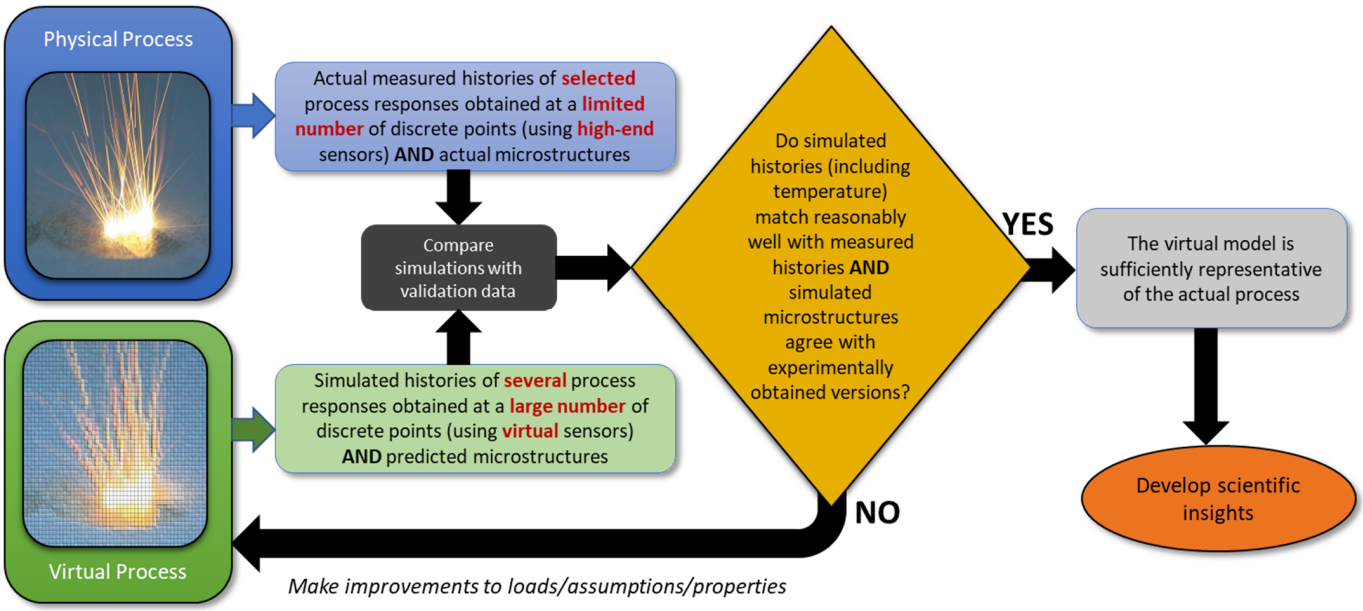

Figure 4. The nexus between experiments and simulations in advancing the knowledge on microstructure formation in AM processes. Temperature is widely accepted as a reliable process signature and its history must be measured and compared with predictions.

3. A Summary of Modelling Methods

Several numerical methods are capable of simulating microstructures in AM at the required level of resolution, i.e., at the mesoscopic scale (that spans from nanometres to micrometres) and at the continuum scale. Some examples of these are front-tracking [28], phase field (PF) [12,19,29–31], level set (LS) [32], lattice Boltzmann (LB) [30,33], Potts kinetic Monte Carlo (kMC) [12,19,30,34], cellular automata [12,19,29,30] and the Johnson–Mehl–Avrami–Kolmogorov (JMAK) theory-based phenomenological model [35,36]. These varied techniques have unique strengths and weaknesses [12,19,30] which means modellers can select the method best suited to their tasks based on these considerations. For example, by deliberating the resolution vs. computational load relationship (Figure 5), a decision may be made on how much accuracy may be sacrificed to obtain a faster solution. Additionally, some methods (e.g., PF) are better suited to considering fundamental physics and may thus be the optimum choice for tailoring microstructures. Each technique also has its own set of unique challenges that modellers need to address—especially when simulating a highly nonequilibrium process such as AM. The hurdles faced by modellers are considered later, in Section 6. While we do not consider atomistic or molecular dynamics simulations here, these may be used to obtain some inputs required for the above methods using first principles.

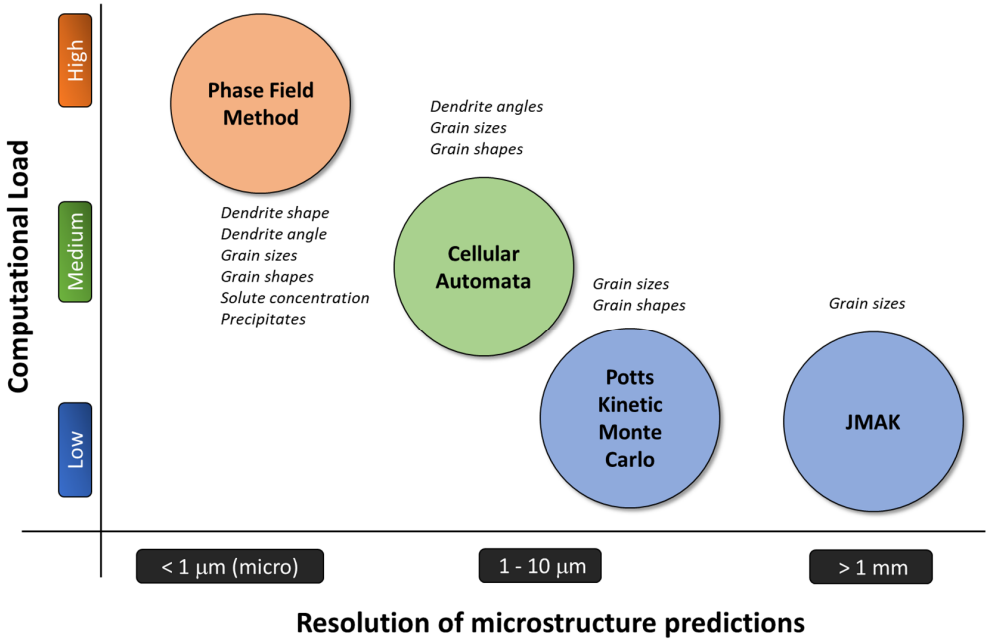

Figure 5. Resolution of microstructures obtained from different techniques vs. computational load involved. Note that the axes are not drawn to scale. (Information for this figure was drawn from [19]).

Temperature histories recorded during AM builds capture the combined influences of heat sources and sinks and provide crucial information on local solidification rates. Specifically, they help quantify the local temperature gradient G and the solid–liquid interface velocity R (i.e., the rate at which the liquidus isotherm moves). The balance between G and R plays an important role in determining the morphology of the microstructures in AM processes such as Direct Energy Deposition (DED) [37], Wire Arc Additive Manufacturing (WAAM) [38], Laser Powder Bed Fusion (L-PBF) [23,39], and Electron Beam Powder Bed [40,41]. One such relationship for an L-PBF process is shown in Figure 6.

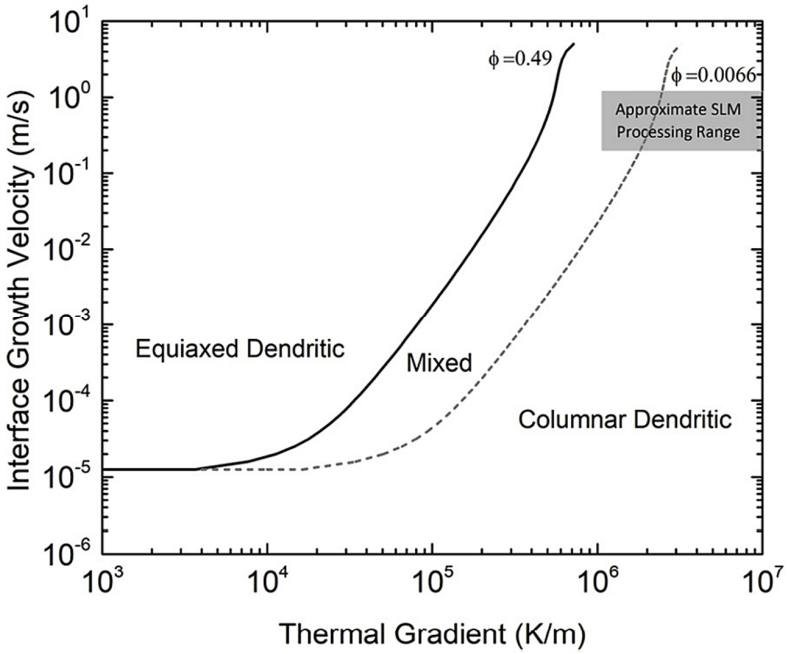

Figure 6. The G vs. R relationship that determines the microstructural morphology for the Al-10wt.%Si-0.5wt.%Mg alloy processed via the Selective Laser Melting (SLM) (i.e., L-PBF) route Reproduced from [39] with permission from Elsevier.

Therefore, it comes as no surprise that all of the microstructure prediction models need location-specific temperature histories as inputs for their predictions. These are either measured during experiments or estimated at the continuum scale using computational fluid dynamics (CFD) or the finite element method (FEM). Recently, the use of lattice Boltzmann method-based hydrodynamics tools for the purpose was demonstrated [33,42]. These can take into account the random distributions of powder particles by size in a layer, and the propagation of the laser (or electron beam) that includes multiple reflections, phase transitions, thermal conductivity, and detailed liquid dynamics of the molten metal. The influences taken into consideration include evaporation of the metal and recoil pressure. In the current Special Issue, a similar hydrodynamics-based approach is presented in an article by Cummins et al. [35] where temperature histories were estimated using Smoothed Particle Hydrodynamics (SPH) simulations that consider melt pool dynamics (Figure 7). SPH is a meshless or Lagrangian method in which virtual particles move with the local material velocity and store all field variables (such as temperature, velocity and microstructure phases). This means that the heat transfer, phase change and melt flow initiated by the laser passing over the powder bed are naturally modelled with SPH in a coupled scheme. While CFD methods consider the detailed fluid flow characteristics of the melt pool when calculating temperature histories, FEM is frequently used to obtain approximate estimates within much shorter timeframes—allowing the simulation of large builds. Particle-based hydrodynamics-based tools are the least productive in terms of computational efficiency, although they are otherwise ideal for simulating powder-based AM systems.

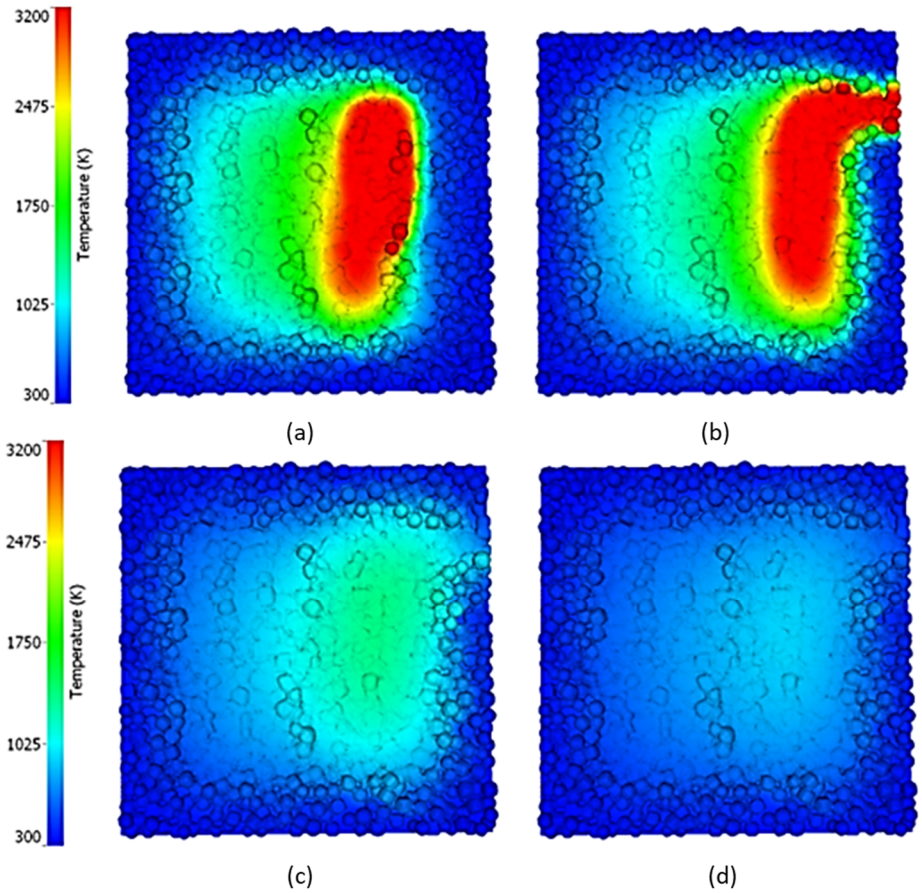

Figure 7. Temperature of the powder bed calculated by SPH following a laser scan over Ti-6Al-4V powder grains for selected times from the start of scan at time = 0 s [35]. Laser power = 114 W and scan speed = 0.6 m/s. Time steps (ms) extracted here are for: (**a**) 12, (**b**) 14, (**c**) 18 and (**d**) 22.

A critical challenge common to all the above methods is the treatment of nucleation of grains [12]. It is a hurdle that is far from being resolved due to the difficulties surrounding observing the mechanism and the potentially coupled nature of the numerous causal factors. To further complicate matters, it has been shown that the several free parameters that are part of the existing models of nucleation have an overwhelming influence on final microstructure predictions [25].

4. Modelling of Nonequilibrium Microstructures Encountered in AM: Recent Progress

It is well recognised that understanding microstructural development in AM under highly nonequilibrium cooling conditions and the consequent effects on mechanical properties of the final component is critical for accelerating industrial adoption of the metal AM process [39]. The distinctive mechanisms that influence microstructure formation under extremely high solidification rates force a rethink of traditional theories or the development of new ones. In this section, we highlight some recent progress made on this front.

It is known that a certain amount of undercooling (curvature, thermal, constitutional, pressure or kinetic) is necessary for solidification to occur since there is no driving force under equilibrium conditions [30]. As the degree of undercooling grows, the velocity of the solid–liquid interface (which is related to the rate of solidification) increases. This departure from equilibrium gives rise to a well-defined 'equilibrium hierarchy' (Figure 8).

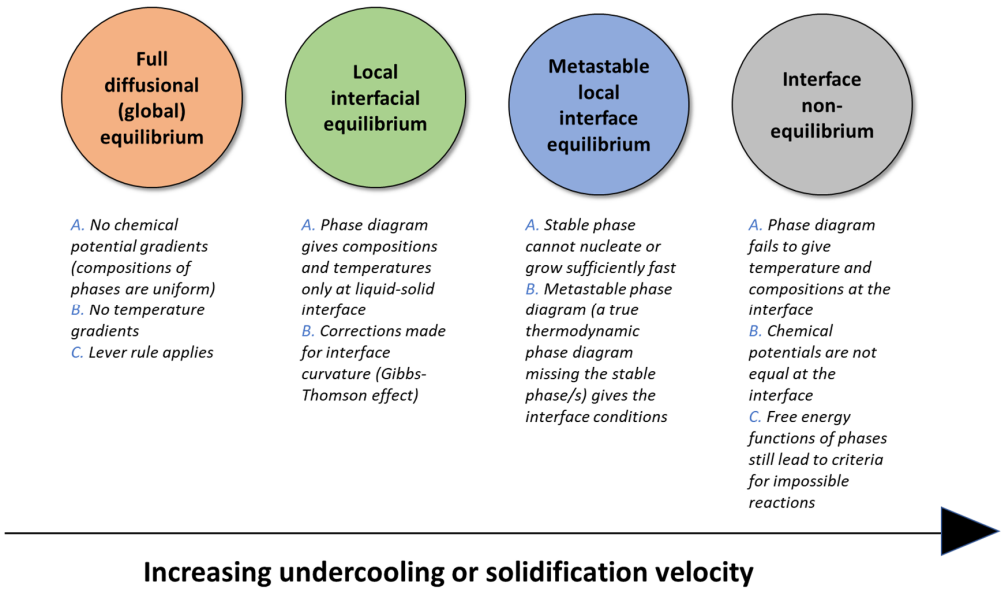

Figure 8. Hierarchy of equilibrium. (Information for this figure was drawn from [43]).

AM processes operate under the 'interface nonequilibrium' conditions where the equilibrium phase diagram fails, and chemical potentials on either side of the liquid–solid interface are no longer equal. In these instances, when the solidification velocity exceeds a certain critical velocity for the multi-component alloy system, the solute is trapped in the rapidly freezing primary solid phase. This is because the solute atoms are unable to diffuse ahead of the fast-moving liquid–solid interface, and the concentration exceeds the solubility limit. Several models have been advanced to describe this experimentally observed phenomenon [31]. Recently, purely quantitative phase field models have been developed from fundamental considerations for predicting solute trapping at velocities relevant to AM, e.g., [31] for the Si-9at.%As system. Similar numerical models are required for other alloys used in AM to simulate microstructure formation accurately. In many cases, the experimental data are not yet available—partially due to the considerable effort involved in collecting all the relevant information, e.g., the extensive range of AM process parameters that generate alternative phase transformation routes. Therefore, the numerical

models can help limit the number of physical experiments needed to determine the solute trapping behaviour.

For predictions involving multi-component alloy systems, phase field models use thermodynamic data obtained from the phenomenological CALPHAD (calculation of phase diagrams) method. The information is used to evaluate alloy phase constitution, the solidification path, basic alloy properties such as partition coefficients (for calculating solute trapping), slopes of liquidus, and solidus phase boundaries. Since the CALPHAD data are typically developed for equilibrium scenarios, they are modified for simulating nonequilibrium processes by using empirically derived models (e.g., Scheil–Gulliver). However, the approaches used so far have limitations [44] and can be improved to support more realistic predictions of microstructures in an AM process simulated at the continuum (or engineering) scale. Some recent efforts (e.g., [44,45]) have improved the quality of microstructure predictions for AM builds. For instance, in an article that is part of the current Special Issue, Sargent et al. [44] have combined continuum scale thermal simulations of the L-PBF process of stainless steel SS316L with a phenomenological microstructure model. Solute redistribution (or microsegregation) was calculated using DICTRA [46], a module that works with the commercial CALPHAD software Thermo-Calc [47]. (DICTRA and Thermo-Calc are registered trademarks of Thermo-Calc Software AB, Solna, Sweden).DICTRA is used to add kinetics that are responsible for the deviations from equilibrium behaviour into the predictions; this makes it possible to incorporate those phenomena (e.g., diffusion-controlled microsegregation) that influence microstructure formation in AM. As shown by the results of Sargent et al. [44], calculations that integrate kinetics can fine-tune predictions using the various location-specific cooling rates in a solidifying alloy under AM conditions. This may be contrasted with a single result obtained for all cooling rates corresponding to maximum microsegregation from the highly idealised Scheil–Gulliver model due to its assumptions of negligible diffusion in the solid phase and perfect mixing in the liquid. (It must be noted here that results obtained from the Scheil–Gulliver model become increasingly valid at extremely high cooling rates [48], e.g., during the rapid quenching of the first AM layer deposited on a build plate, due to its assumptions holding in the physical domain). In a separate recent work [45] that simulated the solidification of AlSi10Mg alloy under AM conditions, a similar fine-tuning was demonstrated by comparing predictions from the original Scheil model and its modified forms. The amendments added solute trapping based on a collection of previously published empirical models (Figure 9) that took into consideration the solidification front velocity and composition.

O'Toole et al. [20] confirmed the power of using diffusion kinetics combined with CALPHAD for predicting microstructures of the AlSi10Mg alloy system freezing under AM conditions. The PF model used by O'Toole and co-workers incorporated the finite interface dissipation (FID) model [49] to account for diffusion across phase interfaces based on kinetics in place of the equilibrium partitioning assumption. (The FID treatment does away with the assumption of equilibrium at the solid–liquid interface and is thus highly suited for the extreme 'interface nonequilibrium' category found on the extreme right in the 'hierarchy of equilibrium' shown in Figure 8). In that study, temperature histories were transferred from a continuum scale CFD model to a PF microstructure model. This multiscale model was then used to quantify the discrepancy in the CFD predictions of the position of the liquid–solid interface between the equilibrium and highly nonequilibrium treatments. O'Toole et al. also introduced a novel volume mapping method to pass the temperature field from the continuum scale grid down to the mesoscopic scale mesh. The FID model [49] was also used by Nomoto et al. [50] for simulating two-dimensional dendritic growth in the Ni (Bal.)-Al-Co-Cr-Mo-Ta-Ti-W-C alloy solidifying under L-PBF conditions and is reported in an article that is part of the current Special Issue. Their nonequilibrium multi-phase field model (MPFM) coupled with the CALPHAD database predicted cell sizes that were in agreement with experimental data. The approaches mentioned above are generally limited to simple binary and ternary systems, as increasing the number of components rapidly escalates the computational complexity [51]. Therefore,

in a recent study, Liu et al. [51] introduced a time-discrete semi-analytical inversion of the thermodynamic relations applicable to general forms of the Gibbs free energy. Linking it with a CALPHAD database enabled the authors to study the grain boundary precipitation and microchemistry evolution in a quaternary multi-component Al-Zn-Mg-Cu system, at length and time scales relevant to typical industrial heat treatment processes. Another recent PF model that merits mention is that of Yang et al. [21], where the solidification of SS316L under L-PBF conditions was simulated. Although solute trapping was neglected entirely as their liquid–solid interface was controlled only by the temperature field variable, they incorporated a noteworthy nucleation model based on classical nucleation theory and the initial grain structures of powder particles and substrate.

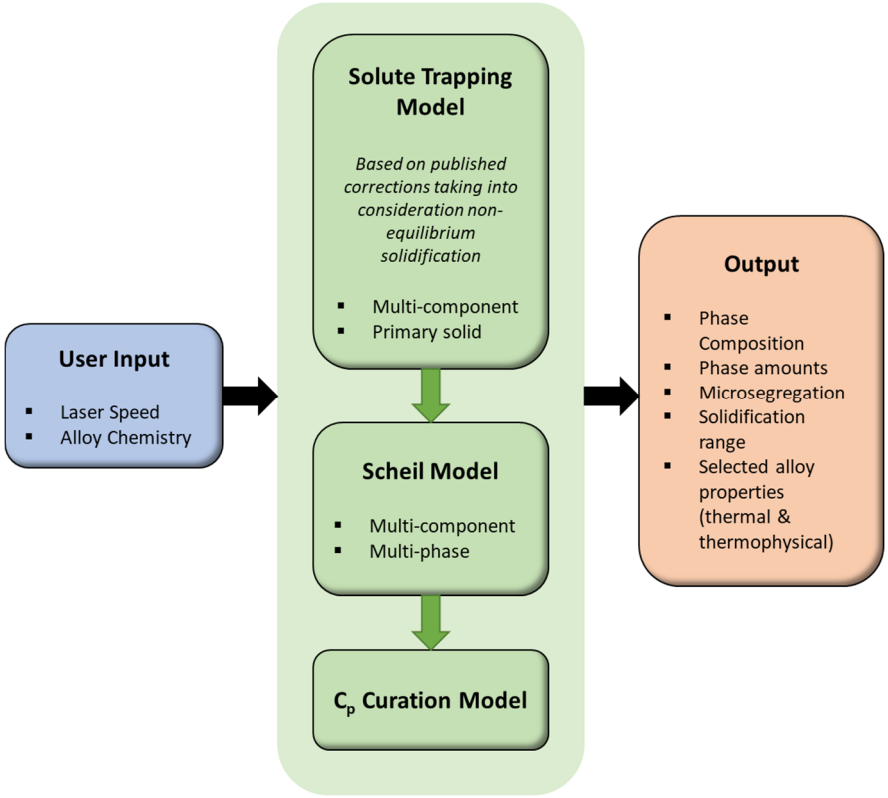

Figure 9. A computational framework [45] for predicting microsegregation in AM. It uses previously published modifications made to the Scheil model that account for solute trapping. The 'Cp curation model' refers to the calculation of specific heat as a function of chemistry and temperature using the Thermo-Calc software. The figure is adapted from that work.

In an article that is part of the Special Issue, Mohebbi et al. [52] used a CA approach to simulate primary particle development and its impact on microstructural evolution of Sc-modified aluminium alloys during AM. They developed a precipitation model that took into account the significant influence exerted by solid intermetallic particles in the undercooled liquid. They also proposed an initiation criterion based on the precipitation kinetics of primary particles, to address solute trapping under high solidification rates. Finally, they used the Avrami equation to track the progress of precipitation. Their experimentally validated approach can predict the distinct fine- (FG) and coarse-grained (CG) zones at the fusion boundary and the melt pool core, respectively. The model was also shown to be able to address the FG zone under lower scanning speed and higher platform temperatures.

5. Machine Learning Models

The use of ML models has increased tremendously in recent times, thanks to increasingly powerful hardware capabilities and contemporary algorithms. These models need to

be trained using a sufficiently large and representative data set to be accurate and reliable. The 'big data' could be obtained from experiments or physics-based models [53].

Machine learning (ML) models are efficient alternatives to physics-rich models (deterministic or statistics-based) as they typically solve in seconds or minutes rather than in hours or days. ML models are particularly suited to tackling higher-dimensional problems such as microstructure formation in metal AM due to their greater competence in obtaining complex correlations than humans and without human bias.

The ML models also play another critical role [53]: they help overcome the difficulties of connecting the various models of sub-processes that comprise the complete AM process. The disparate mathematics and software codes used to solve the vastly different governing equations associated with the sub-processes (e.g., powder flow, heat transfer, melt dynamics, solidification, microstructure formation, residual stress development, distortion, etc.) make it hard to introduce a 'mathematical handshake' between the sub-models (see [54] for details). However, if each sub-process can be distilled into separate ML models, it becomes easier to instead connect these ML models to obtain a complete model. For example, it would be easier to tie an ML model that predicts mesoscale microstructures to an ML model that predicts related continuum scale residual stresses than directly linking the two physics models (e.g., PF with FEA). However, it must be noted that accounting for one-way or two-way interactions between sub-processes would be hard to achieve under this strategy.

In an article [55] that is part of the current Special Issue, an efficient workflow for an ML model was presented where microstructures were correlated to the thermal stress response of a material. Such stresses play a significant part in the usability of AM parts because the relaxation of residual stresses (i.e., when they are removed from their build substrate) are responsible for the distortion of these components.

In closing, it must be recognised that ML models cannot be developed for outcomes that are unusual [56], i.e., statistical outliers. That is because the ML algorithms are typically designed to ignore such events as spurious. Additionally, for the best outcomes, the correlations obtained using ML models must be checked using domain expertise to ensure these are physically valid.

6. Research Gaps and Opportunities

The subject of solidification emerged as an engineering science back in the twentieth century. Gulliver (1913) and Scheil (1942) derived material balance equations at the solid–liquid interface of a solidifying material. They were followed by Chalmers (1956) who developed the constitutional undercooling concept and codified correlations between alloy compositions and cooling conditions that led to observably stable or unstable solid–liquid interfaces (or crystal-melt fronts) [57,58]. While the early treatments applied to equilibrium solidification where sufficient time was available for diffusion within the phases, corrections (e.g., by Aziz [59]) were introduced in the latter half of the twentieth century to consider the nonequilibrium conditions that dominate industrial processes. The arrival of digital computers around this time marked a leap forward in the modelling and simulation of microstructure formation. In the past couple of decades, the rise of AM as an industrial process has added a new dimension to the field with its unique rapid solidification characteristics.

While solidification studies that centred around thermodynamic equilibrium (or near-equilibrium) were sufficient in the past to trigger innovations in manufacturing, this no longer holds for AM. Therefore, the characterisation, manipulation and, ultimately, control of material properties far from equilibrium offers almost completely untapped possibilities for uncovering novel states and phases of materials [18].

Modelling of microstructure formation in AM processes is still in its early stages partly due to the relative novelty of the process itself and partly due to the new paradigms and computational strategies that must be introduced to support AM's distinctive complexities, including extreme nonequilibrium solidification. Another contributing factor is the presently available computing power, which, despite recent advances, is as yet inadequate

to accommodate the simulation of part scale volumes—especially in methods such as PF (and lower scale atomistic or molecular dynamics simulations) that use fundamental physics. Clearly, the more accurate the simulations become, the higher the chances are that these can reliably detect the different phase transformation pathways available for AM alloys under the numerous permutations and combinations of process variables.

Below, we list some of the research gaps that researchers may pursue as opportunities.

6.1. Supporting Multiscale Simulations Associated with the Temperature-Microstructure Linkage

Temperature histories required for the mesoscopic scale microstructure models are typically obtained using simulations conducted at the continuum scale. The challenges here are:

1. **Multiple length scales:** The ability to accommodate the microscopic melt pool caused by the AM heat source in the continuum simulations while concurrently allowing for the build of a complete part to be simulated is a major hurdle in this category. In CFD and FEA simulations, which are usually faster than particle-based hydrodynamics versions, using mesh sizes that cater for the microscopic melts can result in an excessive computational load at the part level. While it is possible to resort to adaptive meshing where mesh sizes coarsen as the heat source moves away, there is often a significant computational overhead associated with remeshing. As an alternative, it may be possible to simulate the effect of the heat source at selected strategic locations of the part in finer detail, predict microstructures at those locations, and interpolate between those locations.
2. **Efficacy and accuracy in passing information between scales:** An important part of the multiscale setup is the ability to pass the temperature data, efficiently and accurately, from the larger grid used for its predictions to the smaller meshes used for microstructure formation. A viable computational strategy for this purpose was recently demonstrated [20].

6.2. Accounting for Two-Way Coupling between Temperature and Microstructure

In reality, there is two-way coupling between temperature and microstructure. That is, the developing microstructure can—in turn—influence the temperature of its surroundings. This is partly because of the release of the latent heat (source term) and partly because of the changing thermophysical properties in the range as the liquid phase changes to solid (more details in Section 6.3 below). Therefore, there is scope for developing a strategy for concurrent two-way coupling in this area. However, early simulations are likely to be restricted to small volumes due to the limitations imposed by computational capabilities.

6.3. Impact of Melt Flow at the Mesoscopic Level

Modelling and simulation of melt flow in AM processes, or welding in general, is a considerable challenge of its own. Numerous studies at the mesoscopic level consider the influences of the beam pressure and capillary and Marangoni forces on the melt pool. For an overview, see, e.g., [60]. These phenomena need to be incorporated to model the solidification problem at the mesoscopic level accurately. At this scale, columnar solidification will impede the melt flow whilst equiaxed dendrites are transported to and deposited at locations different from their nucleation site. Additionally, the transport of solute and heat is affected significantly. Recently a simplified treatment, utilising the Rappaz–Thevoz model of equiaxed solidification [61] and treating extra-dendritic melt well mixed by forced convection, was applied to model the columnar to equiaxed transition in a slab caster [62]. Similar treatments can be applied to an AM solidification scenario. Direct simulations using, e.g., CA or PF models, are not yet available to the best of our knowledge.

6.4. Strategies/Experiments to Obtain Accurate Boundary Conditions

Heat transfer simulations are highly sensitive to some boundary conditions used, e.g., heat conduction away from the substrate (build plate), convection to the ambient (e.g.,

powder bed, cover gas), and radiation. In addition, phenomena such as metal vapour formation, Marangoni flow, etc., also exert influence over temperature predictions. Therefore, for the temperature predictions to be accurate, models with accurate quantification of these phenomena are required. These may be obtained from controlled and well-instrumented laboratory experiments. Another area that must be addressed carefully concerns the quantification of heat transferred from the heat source. This is especially true for laser-based systems since a fraction of the heat is reflected away by the material being printed—and thus must be accounted for accurately to obtain reliable temperature histories in the part being built. If there is any uncertainty associated with predictions, sensitivity studies must be carried out in order to gauge their influence on final temperature estimates and quantify the potential errors.

6.5. Improved Models for Grain Nucleation

A major hurdle to microstructure predictions at the moment is the uncertainty surrounding how to account for the appearance (in terms of time and space) of grains [12]. This subject is particularly relevant to AM because the initial columnar growth attached to the substrate is strongly affected by the microstructure on its surface. While the nucleation model of Yang et al. [21] discussed earlier may be considered a step in the right direction, a deeper understanding is required for the reliable replication of this phenomenon in models.

6.6. Improved Strategies to Account for Multi-Component Diffusion

PF models use nonequilibrium diffusion kinetics linked to equilibrium-based CALPHAD databases to describe solute trapping. A convenient and, therefore, popular, way of achieving this is using a commercial module such as DICTRA [46] that adds on to the thermodynamic database supplied by Thermo-Calc [47], or Pandat [63]. (Pandat is a registered trademark of CompuTherm LLC, Middleton, WI, USA). Assumptions made in developing these modules must hold or be sufficient, e.g., one-dimensional diffusion is assumed by DICTRA. Such unidimensional treatment cannot support diffusion in dendrite tips [48]. In addition, Sargent et al. [44] have reported that convergence issues associated with DICTRA calculations affected half of the temperature histories investigated by them. There is thus scope to make improvements in this area and develop strategies for multi-dimensional diffusion calculations.

6.7. The Need for Reliable Thermodynamic and Mobility Databases of Alloys Relevant to AM

The reliability of PF calculations is predicated strongly on the accuracy of the information contained in the thermodynamic and mobility databases. Sargent et al. [44] and Korner et al. [12] have highlighted the need to have data generated for alloy systems widely used in AM to support related modelling activity. Some of this data may be generated using lower scale simulations at the atomic or molecular dynamics levels, e.g., the quantum-mechanics based Density Functional Theory (DFT) [64].

6.8. The Need for Reliable Material Data for AM Materials

The non-availability of reliable material properties, especially at elevated temperatures, is an issue that has been plaguing simulations for decades and applies to alloys used in AM. It has been the subject of extensive discussion elsewhere [53]. In AM, as suggested in Section 6.4, temperature-dependent laser absorptivity is another property that is critical since the amount of energy transferred from the heat source to the system depends on this value (that changes with the surface condition) and thus can have a strong influence. For PF models, the situation is presently worse. The need to input relatively esoteric quantities such as interfacial energy [12] makes these simulations particularly vulnerable. Thus, there is an opportunity for carrying out targeted work towards filling these gaps.

6.9. Improved Strategies to Account for Solid-State Precipitation

In solid phases, precipitation occurs if the concentration of one solid is above the solubility limit in the host solid, due to, e.g., rapid quenching, and the temperature is high enough that diffusion can lead to segregation into precipitates [65]. In light alloys, which are useful for aerospace applications, controlled solid-state precipitation is engineered to obtain desirable properties such as high mechanical strength [66]. Unfortunately, the nucleation of these usually metastable precipitates is often difficult to predict and the atomic-scale mechanisms are poorly understood, thus hampering efforts towards rational materials design [67]. These precipitations can occur in alloys after partial or complete solidification or after secondary precipitation (regardless of whether eutectic or peritectic). These phenomena are affected by the high heat extraction rates and consequently the short time available for attaining phase equilibrium. In addition, the occurrence and distribution of metastable phases must be considered. Additionally, these phases may dissolve again during self-tempering from reheating caused by the repeated heating cycles inherent in AM processes. Wu et al. [45] have sounded a need to improve data in TC-PRISMA [68], the precipitation module that links to Thermo-Calc [47]. (TC-PRISMA is a registered trademark of Thermo-Calc Software AB, Solna, Sweden).

6.10. Predictive Modelling of Hot Tearing

One of the most prominent defects in AM microstructures is hot tearing. This phenomenon appears to be influenced by the alloy composition coupled with the pronounced segregation in liquid channels due to the delay of secondary precipitation [66,67]. This topic has not been resolved conclusively even in conventional casting and thus remains a challenge in AM. It would be highly beneficial for an AM microstructure model to have the predictive capabilities for hot tearing. Any model for quantifying the actual tendency for hot tearing must, however, be coupled to a simulation of local stress and strain subject to (i) macroscopic distortion of the specimen during processing, (ii) microscopic stresses due to non-local thermal expansion, and (iii) transformation strain due to the precipitation of secondary and tertiary phases.

6.11. Accounting for Novel AM Mechanisms at the Microscopic Level

Researchers are increasingly paying attention to the distinctive microstructural features in AM that result from the unique heat transfer profiles. For instance, side-branching in the dendrites as they form in the solidifying melt was the subject of recent studies, e.g., [23]. It was shown that perturbations on the sides of cells (or dendrites) facilitated crystals to change growth direction by side-branching along orthogonal directions in response to changes in local heat flux in L-PBF. Accurate modelling of such phenomena is not possible with one-dimensional diffusion calculations.

6.12. Strategies for Parallelisation and Preparing for Petascale and Exascale Computing

While many of the modelling methods used for microstructure modelling are highly parallelisable [12,33], they still fall short of providing the speeds required to simulate the formation of microstructures in good detail for a reasonable volume of a part. Thus, there is scope to improve on computational efficiency by taking advantage of new hardware paradigms such as petascale and exascale computing that are currently on the horizon (e.g., see [53]). This might involve rewriting some codes to take advantage of the architecture of the new hardware. Although the cost of these rewrites is likely to be prohibitive in the near term, they may offer excellent value in the medium to long term.

6.13. Heat Treatment of AM Microstructures

The repeated thermal cycles mentioned previously may cause in-situ heat treatment-like effects to various degrees, depending on the alloy being printed and process parameters being used. Parts can also be heat treated in the traditional manner, i.e., after printing, to relieve the large residual stresses that result from high thermal gradients characteristic of

AM builds. These treatments trigger changes to the original structures as additional energy and time are provided for the previously curtailed diffusion processes to take place to different extents. For more details, see works such as [69,70]. Incorporating these changes in models will provide researchers the capacity to predict final part microstructures and optimise heat treatment parameters to obtain the desired results.

6.14. High Entropy Alloys

Unlike conventional alloys, which usually feature one main metal mixed with small quantities of others, high-entropy alloys (HEAs) typically use five metals mixed in roughly equal proportions [71]. The resulting crystal structures can endow the alloy with a combination of useful properties, such as strength, toughness and resistance to corrosion. HEAs were used in AM only very recently, and the impact of rapid solidification on phase selection in these alloys was highlighted by Gorsse [16]. They showed that significant grain refinement and nonequilibrium solute-trapping effects were obtained, which in turn avoided the segregation effects and relieved the solubility limitations in printed HEAs compared with their cast counterparts. Since HEAs represent a modern family of alloys that may find use in demanding applications (e.g., additively manufactured jet engine parts), they are likely to be investigated in detail in the coming decades. Thus, it is useful to consider applying computational modelling techniques to assist with deepening the understanding around these materials.

6.15. Extracting Additional Scientific Value from ML Models Using a 'Grey Box Big Data' Approach

Despite the several advantages of data-driven ML models and their sharply increasing popularity, they are primarily designed to be 'black box' models. As a result, the science that underpins physical mechanisms is hidden from the researchers. Therefore, researchers can assess the possibility of developing ML models that use a hybrid approach when physics-based mechanistic models are available. This would enable physics to be embedded into the predictions of ML models. When using this 'physics-informed ML (or PIML) technique, ML models are constructed from 'grey box' big data (i.e., a combination of 'black box' data from experiments or the field and 'white box' data from physics-based models)—see [53] for a detailed explanation on the subject. In developing a hybrid PIML model, the physics-based model can be used to provide the domain-expertise based checks and balances by penalising any physically inconsistent outputs. Additional discussions on the subject are available elsewhere, e.g., [72–74].

7. Future Perspectives

The metal AM market is expected to grow by USD 4.42 billion during 2020–2024, expanding at a compound annual growth rate (CAGR) of over 14% (Figure 10), according to a market research report [75] released in April 2021. Significantly, this report takes into consideration the effect of the current global pandemic. The demand for metal AM products from the industries that manufacture mission-critical products is likely to increase over the coming years. This, in turn, will have implications for product quality and associated requirements for part qualification (i.e., testing to ensure parts are within specifications) and certification.

The need to uncover the science underlying microstructure formation in AM is expected to remain an ongoing requirement for the foreseeable future as the quality demands of end-users increase with time. This, in turn, is likely to fuel demand for aids such as computational modelling and keep the area fertile for research and development for some time to come. While several efforts currently focus on experimental studies, only a few of them are linked with modelling the same process to obtain a digital replica. By working hand in glove with modellers, experimentalists will be able to generate a deeper understanding of the mechanisms involved. This can only accelerate the pace at which several of these phenomena are better understood—and potentially tuned to obtain desirable tailored microstructures. Investigations of materials far from equilibrium require

the development of new techniques capable of following dynamic processes in materials with extreme spatiotemporal resolution [18]. It is encouraging that new breakthroughs are being reported in the in-situ sensor technologies that can assist the AM community in deepening its knowledge of the nonequilibrium phenomena. For example, recently Siwick et al. [18] explained that ultrafast electron-based methods have become a major new frontier in materials science due to the capability of following dynamics on time scales as short as femtoseconds with the high spatial resolution and sensitivity afforded by electrons. Such equipment is likely to be capable of providing information for modellers and experimentalists on phenomena (e.g., phase transitions, metastable states) that hold the key to accurately recreating the solidification processes in AM. Advances in sensors could eventually provide clues that shed light on the currently obscure nucleation mechanisms.

Figure 10. Technavio's latest market research report titled 'Metal Additive Manufacturing Market by Application and Geography-Forecast and Analysis 2020–2024' [75]. (CAGR = Compound Annual Growth Rate).

In closing, there are currently some hurdles for accurately replicating microstructure formation in AM in the digital domain. However, the value in carrying out near-term computational modelling to strengthen innovation in AM cannot be underestimated. Experimentally validated simulations of a reasonable accuracy are sufficient to provide rare and counter-intuitive insights and must be carried out. Such near-term simulations can uncover trends in the influence of process variables and assist with screening out the least influential variables, highlighting the critical factors that must be studied more deeply. They would also provide a strong foundation for future efforts which are more than likely to be informed by more advanced experimental data, improved theories and computational methods, and supported by more powerful computer hardware.

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