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Multi-scale process simulation for additive manufacturing through particle filled vat photopolymerization

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ABSTRACT

The majority of research into vat photopolymerization (VP), has been focused on experimental investigations of the influence of process and material parameters. In a specific application of the VP technique, where the resin is filled with particles, this empirical approach has its limitations. In order to fully understand the relation between process parameters and the material properties a detailed numerical analysis is needed. In this paper we present a multi-scale and multi-physical simulation approach to unravel such relations in the complex production process. Using a homogenization approach, the influence of the filler particles, in this case alumina, on the light scattering, conversion characteristics and resulting effective thermal and mechanical properties is determined. The effective composite material and scattering properties are then used as input in a process simulation framework. This enables prediction of key filled-VP characteristics at a structural level. A mesh sensitivity analysis at the component scale reveals that adequate predictions may be obtained with a rather coarse discretization, facilitating multi-physics VP part simulations.

1. Introduction

Additive manufacturing (AM) through vat photopolymerization (VP) or stereolithography is a powerful additive process based on selective UV illumination of a photo-reactive resin in a layer-by-layer fashion. VP is generally recognized as an AM technique with an excellent resolution in combination with high surface quality \cite{1}. Although traditional photopolymerizable materials are limited to acrylic and epoxy resins \cite{2}, through the addition of a solid phase powder, the VP process can also be used to print other materials, for example in dental applications \cite{3}. Appropriate subsequent binder removal even facilitates fabrication of dense monolithic non-polymer parts \cite{4}.

The current work considers a multi-step process for the fabrication of ceramics, as illustrated in Fig. 1, with the focus on the (highlighted) AM step itself. It should however be noted that the applicability of this work is not limited to ceramics only, but also includes other particle filled and even unfilled traditional VP resins.

The introduction of the ceramic powder in the polymer resin increases the complexity of the relevant physical processes involved. Besides rheological aspects (outside the scope of this work) in depositing the new layer of powder filled resin \cite{5}, i.e. the slurry, the presence of the powder strongly affects the illumination step. The powder particles typically have different optical properties than the resin, introducing light scattering. Due to the high filling fraction, this may have a profound effect on the shape and size of the cured profile \cite{6}. A similar contrast in properties also holds for the thermomechanical characteristics. While typical resins show shrinkage during solidification, the rigid-like inclusions introduce inhomogeneities in the strain field and act as heat sinks for the exothermic heat generation \cite{7}. These effects, although introduced at the small length scale of the particles, have a profound influence on the resolution and quality of the resulting printed composite part. A better understanding of the VP process for filled resins is therefore considered key to predict or resolve problems related to these effects \cite{8}, either before or during printing \cite{9}.

Starting from the pioneering work of Jacobs \cite{10}, different papers provide a better understanding of the VP process through mathematical/numerical approaches that can be categorized in two categories. Firstly, an extensive number of works focus on the development of more sophisticated polymerization models. Based on the illumination conditions, these contributions predict the spatial evolution of the degree of conversion whilst accounting for the photopolymerization kinetics \cite{11–13}, the presence of inhibitor species such as oxygen \cite{14,15} and photobleaching due to initiator consumption \cite{16–20}. Examples that attempt a similar approach for powder filled resins are more scarce. A notable example is Ref. \cite{21} where experimental work is used to determine the optical properties of the filled resin, to subsequently apply
These in a simulation model to predict the solidified contour. More common are approaches that extend the exposure threshold model developed by Jacobs to account for the broadening (and shallowing) of the cure width (and depth) by the presence of inclusions through empirical expressions [22–25,6]. Apart from the aforementioned photopolymerization models, secondly, phenomenological approaches exist that aim to capture the mechanical effect of the layered process due to shrinkage [26–30] or the resulting part’s tensile strength [31], while considering individual layers as homogeneous.

To obtain an improved comprehension of the influence of a particle filler in the VP process, a numerical multi-scale process simulation framework is developed in this work that captures the effect of the inclusions, starting from the illumination of the monomer resin until the development of residual stresses and deformation in the printed part after solidification. The developed simulation framework distinguishes itself in three ways compared to (the aforementioned) published literature. (i) It combines the multi-physical nature of Ref. [7] with a multi-scale framework to capture the influence of the inclusions on the print process occurring at the component scale. (ii) The effective behavior is extracted in a homogenization approach starting from properties of individual constituents. (iii) It leads to a hybrid model compared to the two aforementioned approaches in literature, combining inhomogeneous layer conversion (kinetics) with the development of mechanical stress and deformation through process simulation.

This paper presents a detailed description of the developed multi-scale VP process simulation framework for particle filled resins, with particular application to an alumina (\(\text{Al}_2\text{O}_3\)) ceramic slurry in a process with a scanning Gaussian UV laser source. The following section will introduce the relevant length scales and the physical background. The derivation of effective material properties is presented in Section 3. In order to demonstrate the relevance of the developed framework, it is applied to an illustrative academic case study in Section 4.

2. Mathematical description of the multiscale modeling framework

The relevant physical phenomena occur at a large variety of length scales, ranging from the wavelength of the UV light source, via the powder inclusions to a single layer and even the full component scale. To put this into dimensional numbers, the wavelength is in the order of \(\lambda\) (400 nm), particles have diameters in the same order up to a few \(\mu\)m, layer thicknesses are 10s of \(\mu\)m with part sizes stretching (many) cms. Although the wavelength itself has little importance for unfilled polymer resins where the Beer–Lambert law is sufficient to predict light absorption, its relevance in particle filled resins has been shown in our previous contributions, cf. Refs. [7,32]. The corresponding length scales vary from the light wavelength and particle diameter to the penetration depth of the light, which is typically somewhat larger than the layer thickness in VP applications. From a (thermo) mechanical point of view, the range of relevant length scales stretches even further, from the particle size (\(\mu\)m) to the full component dimensions (cm). Simulating all physical effects related to the presence of the inclusions in a single process simulation framework is therefore an unfeasible task.

To account for the presence of the inclusions in the process simulation, a multi-scale framework is here proposed, consisting of three coupled length scales as depicted in Fig. 2. The full part scale where process boundary conditions are considered is referred to as the macro-scale, a length scale where homogenized effective properties are used. These are determined from the intermediate meso-scale, governed by light scattering and the micro-scale with heterogeneous local effective thermo-chemo-mechanical properties. These scales are depicted from left to right respectively in Fig. 2.

To relate thermo-chemo-mechanical properties on the micro-scale to the macro-scale, two requirements are assumed to hold; i.e. (i) the macroscopic domain spans a size much larger than the microscopic domain and (ii) the time needed to impose changes at the macroscopic is
much larger than the time needed to reach a steady-state at the micro-scale [33]. This implies separation of scales in both space and time. Assuming that individual layers have a clear role in the residual stresses and deformations in the final part, the separation of scales is less pronounced for the meso- to macro-scale. The mathematical description and physical interpretation of the distinct length scales together with the imposed assumptions is addressed in the following Sections 2.1.2-4, starting with the micro-scale. The problems at the individual length scales are solved using tailored finite element formulations.

2.1. Microscopic problem formulation

The smallest length scale considered is the micro-scale ($m$), where the presence of the inclusions inside the conversion dependent monomer-to-polymer matrix is accounted for in a continuum setting, also known as a representative volume element (RVE). Using appropriate scale-transition relations, the micro-scale behavior is translated to effective macro-scale constitutive laws. A total of three physical descriptions with appropriate assumptions are relevant at this RVE scale, i.e. (i) the chemical evolution in the matrix material and (ii) thermal and (iii) mechanical constitutive descriptions of the different constituents. These will be addressed in the following Sections 2.1.1–2.1.3, respectively.

2.1.1. Chemical evolution

The micro-scale is considered to be small and of size $l$, with only a limited number of inclusions contained inside the considered volume. Using this restriction, a convenient assumption is to consider the intensity to be homogeneous throughout the volume. For the case of a homogeneous unfilled resin, this assumption makes perfect sense as long as the depth of light penetration, $D_p$, is much larger than the microscopic size $l$, i.e. $l \ll D_p$. Note that formally $D_p$ is the length in thickness direction where the irradiance has reduced to $1/e$ of the initial value that originates from a modified version of the Beer–Lambert law [34,10]. Requiring that the magnitude of $D_p$ for the filled resin is still $\gg l$, the light intensity at the micro-scale can also be assumed homogeneous, despite of the light scattering in the slurry.

For the description of the polymer conversion, a simple square root intensity dependent monomer conversion is adopted [7]:

$$R_p = -\frac{d[M]}{dt} = \varphi \sqrt{I[M]},$$

(1)

where $R_p$ is the rate of polymerization, $[M]$ the monomer concentration, $t$ the time, $I$ the light intensity and $\varphi$ a lumped rate constant for the polymerization process. The corresponding degree of conversion of the matrix material at the micro-scale $p_m$ is a linear and weighted difference of the current and initial monomer concentration, $[M]$ and $[M]_0$, respectively, formulated as

$$p_m = \frac{[M]_0 - [M]}{[M]_0}.$$  

(2)

2.1.2. Thermal behavior

 Thermal equilibrium A direct result of the two-scale transition setting introduced above is a simple form of the thermal equilibrium equation inside the micro-scale constituents, both matrix and inclusions, without transient terms, i.e. [33,35,36]

$$-\nabla \cdot \mathbf{q}_m = 0 \ 	ext{with} \ \mathbf{q}_m = -k_m \nabla \theta_m,$$

(3)

where $\mathbf{q}_m$ is the heat flux vector introduced in the form of Fourier’s law, in which $\theta_m$ is the temperature and $k_m$ the thermal conduction coefficient.

Heat source The assumption of an instantaneous steady-state at the micro-scale does not eliminate the possible existence of a heat source term at the macro-scale. In this work, heat generation, $\dot{\theta}_m$, is restricted to the matrix material. The combined effect of heat generated by photopolymerization and by light absorption is described as [37]:

$$\dot{\theta}_m = -\Delta H_m \left( \frac{d[M]}{dt} \right)_m + \alpha_m \mathbf{I} \cdot [C^*]_m.$$

(4)

The first contribution in $\dot{\theta}_m$ is attributed to the exothermic heat, with $\Delta H_m$ the polymerization heat. The second contribution originates from heat by light absorption, where $\alpha_m$ is the molar absorptivity and $[C^*]_m$ the concentration of the absorbing species. Note that in opaque ceramic inclusions, the last term in Eq. (4), i.e. light absorption, can be relevant, but for the alumina particles considered here, the absorption is negligible at the considered wavelength [6].

2.1.3. Mechanical constitutive behavior

Requiring the RVE to be in equilibrium and in the absence of body forces—which, compared to the divergence term, induce a negligible (volumetric) contribution due to the small domain size—the divergence term in the balance of linear momentum becomes dominant, which leads to

$$\nabla \cdot \sigma_m = 0,$$

(5)

where $\sigma_m$ is the micro-scale stress tensor. The relation between the microscopic strains and stresses in both phases follows directly from the isotropic Hooke’s law, i.e.

$$\sigma_m = C \epsilon_m,$$

(6)

where $\epsilon_m$ is the elastic strain tensor and $C_{ijkl}$ is the fourth-order elastic stiffness tensor [38] for which the relevant parameters, in this case Young’s modulus $E$ and Poisson’s ratio $\nu$, are introduced in the following.

Even though the constitutive behavior is assumed to be purely elastic, non-elastic strains and conversion dependent mechanical properties are accounted for. With respect to the former, the total strain tensor $\epsilon$ is decomposed into an elastic, thermal and chemical component, according to

$$\epsilon_m = \epsilon_{e,m} + \epsilon_{th,m} + \epsilon_{ch,m},$$

(7)

respectively. For the inclusion material, the chemical strain is equal to zero and the total strain in Eq. 7 reduces to the first two terms, i.e. elastic and thermal strain. The thermal component in both constituents, i.e. matrix and inclusions, follows the classical description for thermal expansion in an isotropic form, i.e.

$$\epsilon_{th,m} = \nu_m (T_m - T_{ref,m}) I,$$

(8)

with $\nu_m$ the thermal expansion coefficient, $T_{ref,m}$ a reference temperature and $I$ the unity tensor. The chemical strain in the resin is assumed to be linearly related to the degree of conversion using the shrinkage strain at maximum conversion as a scaling factor [7]:

$$\epsilon_{ch,m} = \epsilon_{ch,max} I.$$  

(9)

The inclusion properties are assumed constant at the micro-scale, however, solidification in the matrix material is accounted for by introduction of a conversion-dependent Young’s modulus $E_p$ that reads [7]

$$E_p(\rho_m) = \rho_p E_{pol}$$

for $\rho_m < \rho_{gel}$,

$$E_p(\rho_m) = \left( \frac{1 - \rho_m}{1 - \rho_{gel}} \right) (\rho_m - \rho_{gel}) + \epsilon_0 E_{pol}$$

for $\rho_m \geq \rho_{gel}$.

(10)

Initially when the resin is in a monomeric phase the mechanical properties are insignificant as ensured by multiplication with $\epsilon_0 \ll 1$. After the gel-point, $\rho_{gel}$ is reached and solidification initiates, the Young’s modulus starts to increase linearly with increasing conversion to a value of $E_{pol}$ for fully polymerized material.
2.2. Mesoscopic problem formulation

The meso-scale (\( \mathcal{M} \)) is introduced to account for the effect of the particles on the light intensity profile. Whereas knowledge of the light source intensity profile and the resin’s penetration depth \( D_p \) is sufficient to predict the cured profile in a traditional unfilled polymer resin using Beer-Lambert’s law [10], this is no longer the case in filled media [6]. Due to the high filling volume fraction and the small difference between the light wavelength and particle sizes, an electromagnetic wave description of light is used through Maxwell’s equations. Considering that illumination times are much longer than the time required for light to travel through a layer, the time-dependent light propagation is not of interest and the problem is solved in the frequency domain [7]. A form of the Maxwell curl-curl equation for the electric field that holds for dielectric materials can be formulated as [39]

\[
\nabla \times (\nabla \times \vec{E}) = \frac{\varepsilon_0}{\mu_0} \vec{J}_{\text{ext}} + \frac{1}{c_0^2} \frac{\partial \vec{E}}{\partial t},
\]

where \( \vec{E} \) is the electric field, \( \varepsilon_0 \) is the permittivity in free space and \( \varepsilon_0 \) is the complex dielectric constant which is a function of the complex refractive index \( n \).

The mesoscale problem description is completed by appropriate boundary conditions as addressed in Ref. [32]. These ensure that the light source enters the domain via the top surface (cf. Fig. 2) and they account for the finite domain dimensions through a perfectly matched layer (PML) on the lateral and bottom faces.

The intensity of the light that induces the photopolymerization process is obtained from the wave solution through the magnitude of the time averaged Poynting vector \( \vec{S} \):

\[
I_\text{S} = \|\langle \vec{S} \rangle \| = \frac{1}{2} \Re \left( \vec{E} \times \vec{H}^* \right),
\]

where \( \vec{H}^* \) is the complex conjugate of the magnetic field, obtained using Faraday’s law.

2.3. Macroscopic problem formulation

At the macro-scale (\( \mathcal{A} \)), the focus lies in capturing the influence of VP process conditions, with respect to e.g. layer thickness and illumination conditions to name a few. Relevant balance equations at this scale are mechanical and thermal equilibrium.

The mechanical linear momentum balance at the macro-scale becomes

\[
\nabla \cdot \sigma_m + \rho_m \vec{b}_m = \vec{0},
\]

where dynamic contributions are ignored and \( \vec{b}_m \) is the body force vector per unit volume and \( \rho_m \) the mass density. The thermal equilibrium equation with transient effects and a heat source term is formulated as

\[
\frac{\partial T_m}{\partial t} = \nabla \cdot (k_m \nabla T_m) + q_n,
\]

where \( q_n \) is the specific heat and Fourier’s law is substituted for \( \vec{q} \).

Mechanical and thermal boundary conditions, on a domain as illustrated in Fig. 3 for four individual time steps, complete the macro-scale formulation. For the mechanical boundary conditions, a simple displacement (Dirichlet) constraint (\( \Delta x = 0 \)) suffices in the AM process, i.e. a perfect bonding is considered at the interface between the baseplate of the printer and the first slurry layer at \( \Gamma_0 \). Assuming that the top and the sides are free to deform makes sense considering only a limited section of the whole fluid vat. The temperature is also fixed at the baseplate at the temperature \( T_{\text{b0}} \), where it is mechanically connected. A thermal interaction with the surroundings is accounted for by a combination of convection and radiation on the top surface. This defines a Neumann surface heat flux formulated as [40]

\[
q_{\text{crt}} = h(T - T_{\text{s}}) + \epsilon \sigma \left( T^4 - T_{\text{s}}^4 \right),
\]

in which \( h \) is the convective heat transfer coefficient, \( \epsilon \) the emissivity, \( \sigma \) the Stefan-Boltzmann constant and \( T_{\text{s}} \) the surrounding environmental temperature. A no-flux thermal condition is assumed on the side faces of the domain such that there is no thermal gradient in the direction perpendicular to the plane. This assumption is justified as long as the total temperature increase (near the boundaries) remains limited and/or the surface area on the sides is small compared to the top and bottom surface.

Another element that Fig. 3 highlights is the additional complexity added by the additive nature of the process. Not only does the domain-size increase from layer-to-layer, also the boundaries change, and, as such, the boundary conditions do as well. This important issue will be further addressed in Section 4.1.

2.4. Coupling between the scales

To provide the scale-transition relations from micro-to-macro and meso-to-macro, a first-order and offline homogenization scheme is applied. In the current approach, offline implies that homogenized effective properties on the macro-scale are first determined from the scale-transition relationships before conducting the analysis at the component scale. A coupling between the micro- and meso-scale is not considered in the present framework. It is assumed that the change in the (effective) light scattering as a result of polymer conversion and related effects, e.g. shrinkage, photobleaching and changing of the refractive index is insignificant. This implies that the micro-scale evolution does not affect the meso-scale, enabling application of the offline homogenization scheme and greatly increasing computational efficiency. For an extensive motivation behind the latter the reader is referred to Ref. [7].

2.4.1. Micro-to-macro

The scale transition for the thermomechanical micro-to-macro problem is well documented in literature, see e.g. Refs. [35,41,33,36]. Mechanically, micro-macro consistency is obtained through the classical Hill-Mandel condition which is enforced by application of periodic boundary conditions on the micro-level RVE. This ensures consistency in internal virtual work between both scales. If an RVE is considered with quantities on opposite faces denoted with a + and − superscript, the mechanical periodic boundary conditions are given by

\[
\begin{align*}
\vec{u}_m^+ &= \vec{u}_m^- + \epsilon \vec{q}_m^+ \left[ \vec{x}_m^+ - \vec{x}_m^- \right], \\
\vec{t}_m^+ &= -\vec{t}_m^-,
\end{align*}
\]

where \( \vec{x} \) denotes the reference position and \( \vec{u} \) and \( \vec{t} \) the boundary displacement and traction, respectively.

A similar approach also provides a thermal scale relation that originates from the second law of thermodynamics, which ensures consistency in entropy due to heat conduction [42]. The corresponding periodic boundary conditions are

\[
\begin{align*}
\vec{q}_m^+ \cdot \vec{n}^- &= -\vec{q}_m^- \cdot \vec{n}^+, \\
\vec{T}_m^+ &= \vec{T}_m^- + \frac{1}{V_m} \int_{V_m} \kappa_n dV_m,
\end{align*}
\]

with \( \vec{q} \) the heat flux vector and \( \vec{n} \) the normal vector to the surface.

In the first-order computational homogenization scheme considered here, the macroscopic gradients of the displacement field and temperature field are equal to the volume average of the microscopic fields, through

\[
\sigma_m = \frac{1}{V_m} \int_{V_m} \sigma_n dV_m,
\]
\[
\overrightarrow{q}_M = \frac{1}{V_m} \int V_e \overrightarrow{q}_n dV_m. \tag{22}
\]

Other volumetric quantities follow a similar volume averaging approach, such that \([41,36]\)
\[
\rho_M = \frac{1}{V_m} \int V_e \rho_n dV_m, \tag{23}
\]
\[
\beta_M = \frac{1}{V_m} \int V_e \beta_n dV_m, \tag{24}
\]
\[
(\rho_c)_M = \frac{1}{V_m} \int V_e (\rho_c)_n dV_m. \tag{25}
\]

### 2.4.2. Meso-to-macro

While the micro-to-macro transition is based on a strong scale-separation, this is not the case for the meso-to-macro relation, as the light penetration depth typically surpasses a layer thickness. The meso-to-macro scale transition is only a method to eliminate the dependence on the micro-structural (spatial) realization with respect to light scattering. Even though previous works showed that the illumination conditions on the micro-scale can be highly inhomogeneous locally, at the macro-scale only the resulting cure profile is of interest \([7,32]\).

The homogenization approach used in this work is inspired by the experimental approach in Ref. \([21]\), where a known light penetration in an unfilled resin is scaled by a beam width and depth that are corrected for the presence of powder particles. In the current work, the broadening and shallowing, as illustrated in Fig. 4, is predicted through numerical simulation.

Using an apparent penetration depth, laser beam radius and intensity present in the slurry, \(D_{p,M}, w_{0,M}\) and \(I_{0,M}\), respectively, the spatial intensity profile induced by the Gaussian laser is assumed to be given by
\[
I_{m}(x, z) = I_{0,M} e^{-2x^2 + 2z^2/(D_{p,M}^2 - w_{0,M}^2)}, \tag{26}
\]

where \(x\) and \(z\) are the horizontal and vertical direction, respectively. The slurry dependent values will also be compared to values in the homogeneous resin without the alumina filler, i.e. \(I_0, D_p\) and \(w_0\), cf. the left hand side of Fig. 4. Note that the apparent intensity \(I_{0,M}\) is introduced to conserve laser input power, \(P_L\), and as such is related to the beam width according to
\[
I_{0,M} = \frac{2 P_L}{\sqrt{\pi} w_{0,M}.} \tag{27}
\]

In order to transform the local meso-scale intensity field \(I_m(x, z)\) to the macro-scale homogenized mathematical form \(I_{m}(x, z)\) in Eq. 26, a two-step approach is applied where the surface intensity profile is extracted first and the corresponding penetration depth is determined next. For the considered alumina slurry, this process is illustrated in Section 3.1.

In order to conserve the effective conversion through the scale-transition, the volume averaged meso- and macro-scale degree of conversion should be identical. Formally, that implies
\[
\int \sum_{\delta \in \mathcal{H}} (p_{\delta,M}) dV = …, \tag{28}
\]

To ensure that Eq. 28 still holds after the homogenization, the conversion description is scaled by introduction of a dimensionless scaling parameter \(\mathcal{H}\) and a scaled power \(n\). The macro-scale photopolymerization law, cf. Eq. 1, consequently becomes
\[
R_{p,M} = \left(\frac{d[M]}{dt}\right)_M = \frac{\mathcal{H}}{I_{0,M}} \frac{[I_{m}(\overrightarrow{z})]^n}{[I_{m}(\overrightarrow{z})]} \frac{[M^*]}{M}\tag{29}\]

To enforce conversion conservation in an effective sense, \(\mathcal{H}\) and \(n\) are determined by ensuring
\[
\frac{1}{V_M} \int P_M [I_{m}(\overrightarrow{z}), \mathcal{H}, n, t] dV - \frac{1}{V_M} \int P_{e,M} [I_{m}(\overrightarrow{z}), t] dV \rightarrow 0 \quad \forall \ t, \tag{30}\]

such that the difference between macroscopic and mesoscopic domain averaged conversion for all time steps approaches zero. This can also be written as a least squares minimization problem:
\[
\min \sum_{\delta \in \mathcal{H}} (p_{\delta,M})^2, \tag{31}\]

where \((p_{\delta})\) denotes the volume averaged conversion and the sum is taken over a discrete number of time-steps \(t = 0, \Delta t, \ldots, t_f\). Note that this does not change the conversion kinetics, it simply ensures the correct macro-
scale conversion evolution. Additionally, it is useful to realize that although this explicitly only enforces temporal agreement, implicitly, due to the time integrated effect of conversion, the conversion profile will also be captured correctly. This approach is further elaborated in Section 3.2.

3. Effective property determination

The scale-transition relations introduced in the previous section enable the extraction of effective properties to be used in the solution of the macroscopic problem. In the following, the effective illumination and chemo-thermo-mechanical properties are derived for the photo-reactive alumina slurry, but the approach described in Section 2 is generic and can in principle be applied to different filled resin systems as well.

3.1. Effective scattering properties

The presence of the inclusions causes light scattering to occur, which broadens the apparent laser beam width, typically reducing the penetration depth. To quantify this effect, a penetration depth \( D_{p,M} \), beam width \( w_{0,M} \) and light intensity \( I_{0,M} \) (which is related to \( w_{0,M} \) through Eq. 27, cf. Fig. 4), characterizing the filled medium need to be identified.

For the alumina filler in the monomer/polymer resin at hand, the light propagation through the material is dominated by the difference in optical properties in the complex refractive index \( n_i \), cf. Eq. 11. The micro-scale optical properties used for the resin and spherical inclusions with a constant radius are given in Table 1. Note that the extinction coefficient \( \kappa \) in Eq. 12 is related to the penetration depth \( D_p \) through:

\[
\kappa = \frac{\alpha \lambda}{4\pi} = \frac{\ln(10)\alpha C^*\lambda}{4\pi} = \frac{(D_p)^{-1}I}{4\pi},
\]

(32)

where \( \lambda \) is the wavelength of the light and \( \alpha \) is the absorption/attenuation coefficient \([7,39,43,44]\). Due to the presence of the initiator, which is assumed to be the only absorbing species inside the resin, i.e. \( C^* = |n_i|n_0 \), the resin’s absorption is clearly non-zero. At the wavelength of interest the alumina absorption is negligible \([6]\).

The light scattering problem is solved using Comsol’s RF module \([45]\) in a two-dimensional finite element setting. The third dimension is omitted in order to reduce the computational cost, which can be attributed to the required resolution to fully resolve the wavelength of light \([7]\). Motivation can however also be found in the equivalent light scattering efficiency for two- and equivalent three-dimensional shapes \([32]\). To translate the three-dimensional alumina volume fraction to an equivalent two-dimensional filling fraction, the path length of the light through and from-particle-to-particle is required to be identical. Furthermore, the inclusion’s curvature is considered highly relevant for the scattered result \([32]\). Correspondingly, the two dimensional filling fraction and inclusion radius are taken equal to the three-dimensional values.

The considered square domain with a filling fraction of 50% consists of 1000 circular inclusions, which in turn dictates the length of the domain edges, see the example domain shown in Fig. 5(a). Particles are placed in a periodic arrangement at the left and right boundaries, but they do not cross the top and bottom boundary to account for the free illumination surface at the top.

An example light scattering result is illustrated in Fig. 5(b) for the single realization depicted in Fig. 5(a). Individual particles are not displayed to clearly show the calculated field information. The color in the plot denotes the intensity field normalized by the source intensity \( I_0 \). The non-vertical intensity lines in the inhomogeneous intensity field clearly illustrate the broadening of the light penetration. The subsequent homogenization approach is visualized in Fig. 5(c and d). In Fig. 5(c), an energy conserving Gaussian smoothing is applied to the original intensity field. The magnitude of the Gaussian smoothing, i.e. the standard deviation in the blur operation, is optimized such that the best match is obtained at the top surface \((z = 0)\) with a modified set of power conserving source parameters, according to Eq. 27. This nested (least squares) optimization, that results in the smeared intensity field in Fig. 5(c), provides the effective Gaussian laser parameters \( I_{0,M} \) and \( w_{0,M} \), cf. Eq. 26. The remaining parameter in this equation, \( D_p \), is subsequently obtained from a minimization of the difference between the Gaussian smeared field in Fig. 5(c) and the parameterized version in Fig. 5(d). Fig. 5(b–c) indicate that the light scattering concentrates the light slightly below the resin surface. The resulting parameterized field displayed in Fig. 5(d) neglects this behavior, but overall remains consistent.

The homogenized effective values for \( D_{p,M} \) and \( I_{0,M} \) are obtained as the average from 20 different geometrical realizations. The resulting normalized parameter values are displayed in Table 2 with their corresponding standard deviation (SD). The magnitude of the normalized values illustrates that for the considered slurry both the depth of penetration and the apparent intensity are roughly two times lower compared to the unfilled resin. Based on Eq. 27, the corresponding apparent beam width is approximately twice as wide compared to the actual one. The result for the apparent light source intensity, that approximately equals half the source intensity, closely matches the experimental result obtained by Li et al. \([46]\).

3.2. Effective conversion properties

Even though the homogenized intensity field introduced in the previous section captures the averaged intensity field quite accurately, the highly inhomogeneous field characteristics are lost. Because the relation with the conversion is non-linear, this could affect the conversion efficiency. To remedy this, the conversion scaling parameter \( \kappa \) and modified power \( n \) from Eq. 29 are determined here. This analysis is conducted using the properties denoted in Table 3. The mean and the 95% confidence interval of the volume averaged meso-scale conversion field for the 20 considered geometrical realizations \( \langle p_{M,M} \rangle \) is shown in Fig. 6. For every geometrical realization, the corresponding optimized effective \( \kappa \) and \( n \) parameter are determined, according to the minimization problem in Eq. 31 with \( \Delta t = 0.1 \) s and \( t = 6 \) s. The resulting mean and the 95% confidence interval of the volume averaged macro-scale conversion evolution, \( \langle p_{M,M} \rangle \), is also indicated in Fig. 6. Although the variation between the 20 geometrical realizations has been incorporated in Fig. 6 in the form of a confidence interval band around the mean for both \( \langle p_{M,M} \rangle \) and \( \langle p_{M,M} \rangle \), it is hardly wider than the line thickness itself and therefore considered insignificant. Subsequently, the mean of \( \kappa \) for all geometrical realizations \( \langle \kappa \rangle \) provides the effective parameter \( \kappa \), whereas the non-linear value of \( n \) is obtained by a least-squares difference minimization to \( \langle p_{M,M} \rangle \) for all \( t \). Resulting values are provided in Table 4 and the corresponding volume averaged response is shown by the dotted line in Fig. 6. The latter shows that the proposed scaling of the conversion laws facilitates conservation of the

Table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha )</td>
<td>1.5</td>
<td>[-]</td>
<td>Monomer/polymer refractive index</td>
</tr>
<tr>
<td>(</td>
<td>n_i</td>
<td>)</td>
<td>1</td>
</tr>
<tr>
<td>( \alpha_i )</td>
<td>150</td>
<td>[L/(mol cm)]</td>
<td>Molar absorptivity initiator</td>
</tr>
<tr>
<td>( \alpha )</td>
<td>1.78</td>
<td>[-]</td>
<td>Alumina refractive index</td>
</tr>
<tr>
<td>( \kappa )</td>
<td>0</td>
<td>[-]</td>
<td>Alumina extinction coefficient</td>
</tr>
<tr>
<td>( \gamma_\kappa )</td>
<td>0.5</td>
<td>[\mu m]</td>
<td>Alumina inclusion radius</td>
</tr>
<tr>
<td>( \gamma_\phi )</td>
<td>0.5</td>
<td>[-]</td>
<td>Alumina volume fraction</td>
</tr>
<tr>
<td>( \gamma_\delta )</td>
<td>500</td>
<td>[W/m²]</td>
<td>Source peak light intensity</td>
</tr>
<tr>
<td>( \gamma_{\gamma} )</td>
<td>6</td>
<td>[\mu m]</td>
<td>Laser beam width</td>
</tr>
<tr>
<td>( \lambda_0 )</td>
<td>400</td>
<td>[nm]</td>
<td>Light source wavelength</td>
</tr>
</tbody>
</table>
photopolymerization efficiency through the scale-transition.

The resulting conversion field over time is illustrated in Fig. 7 for intermediate time steps, together with the conversion evolution of a single realization that serves as an example.

### 3.3. Effective mechanical properties

In this subsection, effective mechanical properties are derived in terms to the effective stiffness, thermal expansion and chemical shrinkage tensor. To this extent a total of 20 unique microstructures are generated using the discrete element package LIGGHTS [47]. To respect scale-separation, the considered micro-scale domains consist of 10 alumina inclusions with \( r = 0.5 \, \mu m \), occupying a volume fraction \( \phi = 0.5 \). Three example realizations are visualized in Fig. 8. Proper dispersion of the inclusion in the matrix material is ensured by first filling the domain to 64 vol.\%, before reducing the radius to the specified value.

### Table 2

Normalized effective optical properties at the macro-scale.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>SD</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>( h_{c, M} / I_0 )</td>
<td>0.485</td>
<td>0.017</td>
<td>[-]</td>
</tr>
<tr>
<td>( D_{p, M} / D_p )</td>
<td>0.546</td>
<td>2.6 \times 10^{-2}</td>
<td>[-]</td>
</tr>
</tbody>
</table>

### Table 3

Optical properties of the micro/meso-scale constituents and boundary conditions.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( [M]_0 )</td>
<td>8.2</td>
<td>[mol/L]</td>
<td>Initial monomer concentration</td>
</tr>
<tr>
<td>( P_0 )</td>
<td>0.15</td>
<td>[m/(sW)]</td>
<td>Polymerization rate constant</td>
</tr>
</tbody>
</table>

---

**Fig. 5.** Graphical visualization of the homogenization approach for the particle realization depicted in (a). The original scattered intensity field is displayed in (b), after application of the Gaussian smoothing in (c) and the corresponding parameterized version that follows Eq. (26) in (d).

**Fig. 6.** Domain averaged conversion in the original scattered (solid) and parameterized (dashed) fields, i.e. \( \langle p_{c, M} \rangle \) and \( \langle p_{c, M} \rangle \), respectively, with mean and 95\% confidence interval for 20 geometrical realizations. \( \langle p_{c, M} \rangle \) is a direct solution of the minimization problem in Eq. (31) following Eq. 29 with \( \beta \) and \( n \). The homogenized effective value (dotted) uses the effective conversion properties from Table 4.

### Table 4

Normalized effective conversion properties at the macro-scale.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>SD</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \beta )</td>
<td>0.873</td>
<td>3.22 \times 10^{-2}</td>
<td>[-]</td>
</tr>
<tr>
<td>( n )</td>
<td>-0.120</td>
<td>n.a.</td>
<td>[-]</td>
</tr>
</tbody>
</table>
Effective properties are obtained using Comsol 5.4 [48] by application of periodic boundary conditions and using the constituents’ material properties denoted in Table 5, at constant temperature and curing degree. The only variable assumed conversion dependent is the polymer Young’s modulus, as described by Eq. 10.

The resulting effective stiffness, thermal expansion and chemical shrinkage can be characterized as isotropic, as shown in Appendix A. The complete set of obtained effective elastic properties is provided in Table 6, with the exception of the volume averaged quantity $\rho_M$ which follows directly from Eq. 23. The standard deviation shows the limited spread between the different geometrical realizations.

### 3.4. Thermal properties

The same geometrical realizations used for the effective mechanical property determination in Section 3.3 (cf. Fig. 8) are also used for thermal properties. Apart from the thermal conduction $k$, all thermal characteristics are obtained by volume averaging following Eqs. 24 and 25. As with the mechanical properties, the constituents’ properties are assumed isotropic at the micro-scale, as given in Table 7.

The effective conductivity is obtained by applying periodic boundary conditions with a temperature gradient and, subsequently, measuring the resulting heat flux as introduced in Section 2.4.1. The determined isotropic thermal conductivity is provided in Table 8, cf. Appendix A for the spread in individual realizations.

### 4. AM process simulation

To illustrate the relevance of the above homogenization procedure for the simulation of the additive manufacturing process at the macro-scale, the proposed framework is implemented into an in-house developed finite element code. Details of this implementation will be addressed next, after which the applicability of the macro-scale model formulation with the homogenized input is demonstrated.

#### 4.1. Implementation of the process simulation framework

For simulating a section of the photopolymerization vat, a layer of slurry is deposited in the form of a voxel mesh with a certain resolution [30]. To fully resolve the penetration depth, the current implementation uses a uniform mesh size with multiple elements per layer thickness. New bulk elements are only added once a new slurry layer is applied. Initially, the top nodes of the added elements are placed at the intended position, based on the undeformed and as-intended layer. To ensure this initial state is stress free, the initial strains upon activation are subtracted from the element strains in Eq. 7 to obtain an updated

![Fig. 7. Conversion over time [s] for a single realization (top row) compared to the homogenized conversion induced by the homogenized intensity (bottom row), i.e. using effective properties from Tables 2 and 4.](image)

![Fig. 8. Three different realizations of a micro-structure with $t = 0.5 \mu m$ and $\phi = 0.5$.](image)
**Table 8**

Effective thermal properties at the macro-scale.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>SD</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_m$</td>
<td>1.14</td>
<td>$3.86 \times 10^{-2}$</td>
<td>[W/(m K)]</td>
</tr>
</tbody>
</table>

...total strain formulation [30].

The top surface of the domain consists of surface elements to account for the convection and radiation boundary conditions, cf. the red surface in Fig. 3. As soon as a new layer is added, the previous surface elements are removed.

The driving force for all physics of interest is the light intensity, which is considered as a volumetric field quantity according to an axisymmetric modification of Eq. 26. Movement of the laser (along with the addition of new layers) is driven by a G-code script, which is here generated by the open-source Slic3r software [49]. As soon as the process is completed, the part is removed from the base plate by removing the associated mechanical constraints [30].

### 4.2. Sample bridge process simulation

The macro-scale framework is applied to simulate a 9-layer bridge sample as depicted in Fig. 9. The complete dimensions are $(x \times y \times z) = 200 \times 600 \times 450 \mu m$ with two square pillars. To prevent AM/VP related geometrical inaccuracies induced by slicing the three-dimensional object, the pillars and the deck consist of exactly 6 and 3 layers of 50 $\mu m$ thick, respectively, and faces are considered flat and either parallel or perpendicular to the printing direction. Temperatures of the constrained printer base-plate and the surrounding environment are set to 20 $^\circ C$, while the convection and emissivity coefficients are assumed to be 10 W/m$^2$K and 0.9, respectively. To discretize the domain, five linear cube elements are placed over the layer thickness, $\delta$, which is 50 $\mu m$.

Based on the effective irradiation properties extracted from the meso-scale and a laser speed of 30 $\mu m$/s, the cured-line width, $C_w$, is expected to be 40 $\mu m$. Accordingly, the distance between the scanned contour profile and the outer part dimensions is equal to $C_w/2$. After the contour is scanned, the infill of the part is solidified using an alternating 0/90 degree angle inside the $xy$-plane where the line spacing is reduced to 20 $\mu m$ to ensure proper solidification. Both the contour and the infill strategy are depicted by the lines in Fig. 9, where each color designates a single layer.

Resulting fields for degree of conversion, temperature, stress and deformation for intermediate time steps during this process simulation are provided in Fig. 10. The temperature plots show the entire considered section of the slurry bath, while the degree of conversion as well as the Von Mises stress are only shown for the solidified volume with $p > P_{ref}$. Depicted deformations are multiplied by a factor of 5.

The thermal profile in Fig. 10(a) and (d) immediately shows that, even though the heat generation might be significant, cf. [7], the vat surrounding the laser heat source and the base plate act as a heat-sink and ensure that the part does not overheat during printing. Correspondingly, the temperature increase induced by the exothermic and absorption heat generation is only occurring locally. This is something that might change for a more voluminous part but could also point to the dominance of the constraint base-plate temperature. However, for the small volume considered and the limited energy input at the considered speed, the assumption of the constrained base-plate temperature is deemed appropriate.

The solidified profiles in Fig. 10(b) and (e) clearly illustrate the geometrical smoothing over individual layers of VP as an AM process, since no clear crevice can be distinguished between layers along the edge of the part. This smoothing, introduced by the required print-through into the previous layer does show in clear undulations on the sides of the object. Furthermore, the conversion field shows that the current scanning speed does not result in fully cured samples and post curing might be required.

A large build-up of stress is evident in Fig. 10(c) where the part is connected to the build-plate. After the part is printed and released from the build-plate this stress largely relaxes, as depicted in Fig. 10(f). The latter also clearly indicates the expected residual deformation from a multi-layer additively manufactured bridge.

A highly resolved cube mesh is to a large extent limiting the feasibility of simulations at a component scale. The possibility to obtain a nearly equivalent accuracy with a coarse discretization is therefore important to explore. The mesh dependence of the obtained geometry and deformation is shown in Fig. 11. It displays the outline of the geometry in a cross-section in the $xz$-plane through $y = 150 \mu m$ for element sizes $L_x$ that are an increasing fraction of the layer thickness, $\delta$, i.e. $L_x = \delta/5, \delta/3, \delta/2$. Fig. 11 shows that all different mesh sizes are well capable of capturing the overall deformation, i.e. the warpage of the bridge. To confirm this graphical impression, the curvature of the bridge deck for different mesh resolutions, which is on average equal to 1.2 cm$^{-1}$, only has a standard deviation of 0.2-1.6 cm$^{-1}$. For the solidified shape in the horizontal direction, a single element appears to over-predict the dimension of the solidified geometry, while two or more elements already show an adequate agreement.

The cross-sectional stresses for the different mesh resolutions are provided in Fig. 12. For a large resolution in the stress field, a highly resolved mesh is also required. However, if one is only interested in the locations where the largest stress concentrations occur even one or two (linear) elements suffice. It should be noticed that for the magnitude of the stresses and deformations, the relevance of viscous or even plastic material behavior needs to be further examined. The mesh resolution dependency will also change for higher-order elements.

For reference, Fig. 13a and b compare the printed geometry to the intended geometry, both visually and quantitatively, respectively. The most obvious difference is the large print-through error that can be observed in the additional solidified material underneath the bridge deck. Fig. 13b, generated using the open-source software CloudCompare [50], indicates that the additional solidified volume is even more than a single layer thickness. Furthermore, the shrinkage in the bridge deck causes the top to be more narrow than intended, while the deformation in the pillars produces a wider bottom part. Overall, the accuracy in the $xy$-plane is fairly accurate.

The process simulation, based on the homogenized optical, chemical, thermal and mechanical properties of the ceramic-filled resin...
described above, adequately captures the essential aspects of the multi-physical and multi-scale process in a convenient fashion. Using a limited resolution, it could serve to optimize light path planning or shape modification in an optimization framework to reduce geometrical deviations or stress concentrations. Note that in spite of the fact that smaller length scales have been eliminated in the offline homogenization steps, the predicted outcome only holds for the considered (composite) material and light source.

5. Conclusion

The addition of filler particles to a traditional VP process adds a large degree of complexity. This work proposes a predictive multi-scale and multi-physical simulation framework for the corresponding AM process. The micro-, meso- and macro-scale each reveal distinct features. Whereas the micro-scale provides effective thermo-mechanical properties at a length-scale where the intensity and corresponding conversion are considered homogeneous, the meso-scale accounts for the (wave-like) interaction of the laser light with the particle-filled resin. To enable full process simulations that account for the presence of the inclusions, the offline micro- and meso-scale simulations provide effective material and scattering properties, respectively, used as input in the macro-scale simulation. Although the description of the individual physics may be considered simplistic compared to other
individual and tailored (single-physic) models available in literature, the key characteristics of a complete ceramic VP process are captured. Moreover, the modular set-up of the multi-scale modeling framework allows for more advanced models to be incorporated. This framework enables component scale stress and deformation predictions based on the constituents’ material properties and process conditions. Simulating the AM process in an immersed setting that accounts for a section of the resin/slurry-vat enables to capture typical VP characteristics. This would not be possible using a commonly applied geometry-conforming pre-meshing technique.

Although the conducted study was focused on a resin with an alumina filler, it is expected that the model components are easily tailored to alternative systems with small (compared to the wavelength) inclusions or even homogeneous resins. The description of the (solidifying) polymer can be improved by, for example, incorporating viscoelastic behavior and a more advanced polymerization model. Further advancement of the developed framework can be achieved by capturing the relevant light scattering on the meso-scale using more computationally efficient tools. In the current implementation, the finite element formulation of Maxwell’s equations is a limiting factor in the applicability to predict light scattering for larger light source diameters and/or layer thicknesses or more realistic three-dimensional light scattering realizations. Possible research directions are alternative near field methods, such as the multi-sphere T-matrix method (limited to spherical inclusions [51]), that should be able to account for light absorption. Furthermore, the current work only considers a scanning laser VP process, whereas irradiation through mask-projection is also widely applied in the fabrication of ceramics. Future work will focus on capturing the relevant effects of that particular approach.

Appendix A. Confidence in effective properties

The matrix components of the thermal expansion, chemical shrinkage and thermal conductivity are provided in Fig. 14a–c, respectively. A box-plot is depicted with the individual values of the realizations depicted by the overlayed circles.

The derived mean effective stiffness from which the isotropic elastic properties are extracted at final conversion equals

\[
D = \begin{bmatrix}
1.25 \times 10^{10} & 5.97 \times 10^9 & 5.92 \times 10^9 & -1.20 \times 10^8 & 1.26 \times 10^7 & -7.71 \times 10^5 \\
1.25 \times 10^{10} & 5.98 \times 10^9 & 9.10 \times 10^7 & -2.52 \times 10^7 & 7.90 \times 10^6 \\
1.26 \times 10^{10} & -1.20 \times 10^7 & 8.94 \times 10^7 & 2.98 \times 10^7 \\
3.49 \times 10^9 & 9.81 \times 10^9 & 3.54 \times 10^9 & 3.47 \times 10^9 \\
-1.27 \times 10^7 & 2.64 \times 10^8 & 2.98 \times 10^7 & 3.54 \times 10^9 & 3.47 \times 10^9
\end{bmatrix}
\]

Data availability

The data generated and/or analyzed in this study are available upon reasonable request.

CRediT authorship contribution statement

S. Westbeek: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing - original draft. J.J.C. Remmers: Conceptualization, Software, Funding acquisition, Methodology, Project administration, Supervision, Writing - review & editing. J.A.W. van Dommenlen: Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Writing - review & editing. M.G.D. Geers: Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, 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.

Acknowledgments

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References
