Simulation of thermally sprayed coating properties considering the splat boundaries

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Abstract. The properties of thermally sprayed coatings depend heavily on the solidification process during the impact on the substrate. The simulation of the coating build-up is of essential importance in order to understand the processes behind it. The main objective of this study is the development of a coating build-up model to predict the properties e.g. thermal conductivity of plasma sprayed coatings. Therefore, a CFD simulation based on a VOF approach coupled with the momentum source method is applied. 3D-simulations of the impact of multiple alumina particles are performed to observe the coating build-up. Subsequently, the result is transferred to another model to calculate the coatings thermal conductivity. Since the resolution of the mesh does not allow a proper representation of the air gap at the splat boundaries between two solidified particles, a new function is developed to cover such phenomena. This function enables a broader representation of the splat boundaries and an adjustment of the coating properties in these areas. To validate this function, multiple simulations with different particle temperatures and velocities are calculated. Likewise, different assumptions for the thermal conductivity values in the gap are investigated. The results of the calculated properties correspond with experimental values.

1. Introduction

The properties of thermally sprayed coatings are determined during the solidification process of the liquid particles, but the exact details of this process are challenging to observe [1]. Simulations are an important tool to not only predicted the results of otherwise time and cost intensive experiments, but also a way to analyse certain physical phenomena that would be impossible to observe in experiments. The calculation time is the bottleneck resource of most simulations and thus poses a problem in the development of simulations. Especially in such highly dynamic simulation of the coating build-up of thermally sprayed coatings the required simulation time is high, due to fast particles, requiring an extremely small time step, and small features of the coating itself, requiring a small element size.

In 2008 the authors developed a model to simulate the impact of thermally sprayed particles on a smooth and rough surface [2] and compared the results with experimentally sprayed coatings [3]. This model was extended to a state in which it was capable of simulating the impact of multiple particles, resulting in a coating thickness of about 70 µm [4]. J. Prehm et. al. developed a function to simulate the solidification process by increasing the viscosity of the particles once the temperature drops below the solidification temperature [5]. A similar approach was pursued by the authors of this study in a work in 2019, in which the solidification formulation of the simulation software Ansys Fluent was replaced with a momentum source function, to immobilize the particles [6]. Both of these functions replaced a complex solidification model with simple approximations, thus lowering the required calculation time. This has
led to a significant time saving in the simulation and now enables coating build-up simulations of not only individual particles but of entire coatings in a reasonable time. Other work concentrated more on the processes during simulated solidification. H. Fukanuma et. al. studied the effect of trapped gas in simulations of thermally sprayed coatings [7] and H. B. Parizi et. al. studied the solidification process of particles dropped onto non-smooth surfaces [8]. Both discovered that the contact area of the particle and the surface is a crucial parameter for the solidification process. However, in most simulations the tiny gap of air that gets trapped between particles and the substrate and between particles itself is ignored as the simulations cannot capture such small features. This makes the simulations less realistic.

Figure 1 shows a SEM-image of an alumina coating in the upper left corner. The ceramic coating exhibits features such as porosities, air gaps between the different splats and cracks within the splats. In the lower left and upper right corner these coating features are schematically represented and a close-up is used to demonstrate where no coating material is present or where there are gaps, and therefore affect the coating properties. In the lower right corner of figure 1 the schematic representation is overlaid by an arbitrary mesh with a relatively small element size of \( L_{\text{mesh}} < 1 \mu m \), which would lead to unacceptable long calculation times. However, it is also evident that even with this small element size not all features of the coatings can be captured, at the same time a reduction of the element size is no longer justifiable due to the high computational costs.

![Figure 1](image_url)

**Figure 1.** SEM-image of an plasma sprayed alumina coating (top left), schematic representation of the coatings with visualized air gaps (bottom left), close-up of the schematic representation (top right), mesh overlay on the close-up demonstrating the necessity of small element sizes to cover all coating features (bottom right).

In order to resolve this contradiction - the ability to perform fast simulations and simultaneously predict correct coating properties - this publication presents a method to successfully simulate the air gap between two splats on a coarser mesh for thermally sprayed coatings. The coating build-up of alumina coatings were simulated and the coatings' thermal conductivity was calculated using this approach.

2. **Experimental and materials**

A 3-D particle coating build-up model was created in which the developed functions are implemented. The model is based on a previous publication of the authors [6]. In this model the impact of a thermally sprayed particle onto a flat substrate was simulated. A momentum source function was used to simplify the calculation of the solidification process. This model was extended for this study to simulate the impact of multiple particles in one single calculation domain, thus creating a representing volume element (RVE) of the coating. Furthermore, export functions were implemented to simplify the monitoring of the particle values, like temperature and velocity. The most important step is the inclusion of a gap function, which alters the coating properties in the vicinity of the interface between different splats thereby emulating an air gap. The precise details of this functions will be explained later on. The
created RVE was then transferred to another model, in which the effective thermal conductivity was calculated by applying a heat flow at the upper boundary.

The coating build-up model uses the Volume of Fluid (VoF) approach to model the two phases air and alumina. The simulation model consists of 3 boundaries and the interior domain as shown in figure 2. The domains dimensions are 225 μm × 225 μm × 75 μm. The calculation mesh consists of 330,000 cells with an edge length of L_{mesh} = 2.25 μm. The upper side of the model represents an inlet where the molten particles are generated and air is injected. The properties of the generated particles can be found in table 1. The lower boundary, the substrate, performs as a wall, where the particles impact and solidify. The outer sides of the domain are an outlet, where excessive volume of the particles and the air can leave the domain.

![Schematic representation of the calculation domain, with one inlet on top, 4 outlets on the sides and the solid wall on the bottom.](image)

Figure 2. Schematic representation of the calculation domain, with one inlet on top, 4 outlets on the sides and the solid wall on the bottom.

The following underlying assumptions are made:
- The viscous model was set as laminar
- The energy model was enabled and all other models were disabled
- The interior domain is filled with air at the start of the simulation
- The locations of the particles at the inlet boundary randomly distributed according to a gauss distribution
- The temperatures and velocities at the inlet are equal for the particulate and the air phase
- The temperature at the lower substrate is set to T = 300 K
Table 1. Material properties and process parameters.

| parameter          | unit | value                  |
|--------------------|------|------------------------|
| domain size        | µm×µm | 225×225                |
| particle diameter  | µm   | 70                     |
| particle velocity  | m/s  | 180/200/220            |
| particle/gas temp. | K    | 2,473/2,673/2,873      |
| melting temp.      | K    | 2,345 [9]              |
| melting energy     | J/g  | 1,060 [9]              |
| thermal conductivity alumina | W/(m-K) | 30 [10]         |
| thermal conductivity air | W/(m-K) | 0.0242             |
| viscosity alumina  | kg/(m-s)| 0.055 [11]  |
| density alumina    | kg/m³ | 3,950 [10]             |
| heat capacity      | J/(kg-K) | 900 [11]           |

The simulations were implemented in Ansys Fluent 18.2 and computed on 8 Intel HNS2600BPB Platinum 8160 2.1 GHz CPUs with 1,024 MB of RAM each. The computation time was approximately 24 hours. To avoid refining the mesh and increasing the calculation time, a function is created to emulate a realistic gap between two solidified particles while simultaneously keeping the calculation time low. For this new function, a user defined scalar (UDS) is created, which consecutively enumerates the particles starting from one. If two particles are next to each other, Fluent calculates the median value of the user defined scalar values in the cells between the two particles. When particle 1 and particle 2 solidify directly next to each other, the cell in the middle between the two will have a UDS value of 1.5. As a consequence, if a cell has an UDS value, that is not an integer, it has to be part of the gap area.

With the presented functions it is now possible to successfully isolate the areas between two particles and to assign a different thermal conductivity to the area between these particles. The formulation for the thermal conductivity values chosen in this work is shown in equation (1). The function then adjusts every gap between two particles into the formulation (1) for a gap between particle 1 and particle 2. Such that even for a gap between e.g. particle 1 and 3 the equation (1) can be used.

\[
\lambda = \lambda_{\text{max}} \cdot \left[ \cos( (1 - i_{\text{count}}) \cdot 2 \cdot \pi) \cdot (0.5 - \frac{x_{g}}{2}) + (0.5 + \frac{x_{g}}{2}) \right] \tag{1}
\]

\(\lambda\): thermal conductivity [W/(m-K)]
\(\lambda_{\text{max}}\): maximal thermal conductivity [W/(m-K)]
\(i_{\text{count}}\): loop variable [-]
\(x_{g}\): percentage of minimal thermal conductivity in the gap [-]

Figure 3 graphically shows the difference between equation (1) and a physically correct thermal conductivity. While the solid line shows an exemplary course for the thermal conductivity for a realistic coating with a small air gap in between, the dotted line shows the course of the adjusted thermal conductivity as used in this model. In the first case the thermal conductivity drops steeply and remains for a very short interval at the value for air of \(\lambda_{\text{Al}} = 0.0242\) W/(m-K). The smoother dotted line shows the course of the developed function, which can then be captured even by a coarser mesh.
Figure 3. Visual representation of the realistic (solid line) and the approximated (dashed line) thermal conductivity, over the value of the particle counting variable.

As a first guess, an approximation to calculate the most fitting value for the reduced thermal conductivity in the gap used by the model $x_{\text{eq}}$ was carried out. For this approximation the gap is assumed to be represented by a serial connection of two thermal resistances for the gap itself and the alumina around $R_{\text{serial}} = R_{\text{gap}} + R_{\text{alumina}}$. The thermal resistance of the model $R_M$ needs to equal the thermal resistor of a realistic gap $R_{\text{serial}}$, which is expressed in equation (2).

$$\frac{L_M}{\lambda_M \cdot A_M} = R_M = R_{\text{serial}} = R_{\text{gap}} + R_{\text{alumina}} = \frac{L_{\text{gap}}}{\lambda_{\text{gap}} \cdot A_{\text{gap}}} + \frac{L_{M-L_{\text{gap}}}}{\lambda_{\text{max}} \cdot A_{\text{gap}}} \tag{2}$$

R: thermal resistance [K/W]
L: length of the heat resistor [μm]
A: area of the heat resistor [μm²]

As the areas of both resistors are equal $A_M = A_{\text{gap}}$, this results in equation (3).

$$\left(\frac{L_{\text{gap}}}{\lambda_{\text{gap}}} + \frac{L_{M-L_{\text{gap}}}}{\lambda_{\text{max}}}\right)^{-1} = \frac{\lambda_M}{x_M} \tag{3}$$

From SEM images of plasma sprayed coatings alumina coatings it was estimated that the length of the gaps between the particles are roughly $L_{\text{gap}} = 0.01 \, \mu m$. This value represents an initial guess and shouldn’t be emphasized too much, as it could be adjusted later on. The thermal conductivity of the real gap was assumed to be $\lambda_{\text{gap}} = \lambda_{\text{Air}} = 0.0242 \, \text{W/(m·K)}$. Furthermore, the gap width in the model was observed to be about 7 mesh cells wide, so $L_M = 7 \cdot 2.25 \, \mu m = 15.75 \, \mu m$ was assumed. At last the expression of equation (1) was used for the thermal conductivity of the model $\lambda_M$, resulting in

$$\left(\frac{0.01 \mu m}{0.0242 \, \text{W/mK}} + \frac{15.75 \mu m \cdot 0.01 \mu m}{30 \, \text{W/mK}}\right)^{-1} = \frac{30 \, \text{W/mK}}{15 \mu m} \cdot \left[\cos((1-i) \cdot 2 \cdot \pi) \cdot (0.5 \cdot \frac{x_{\text{eq}}}{2}) + (0.5 + \frac{x_{\text{eq}}}{2})\right]$$
This equation was integrated over \( i \), resulting in
\[
\int_1^2 \left( \frac{0.01\mu m}{0.0242\frac{W}{mK}} + \frac{15.75\mu m-0.01\mu m}{30\frac{W}{mK}} \right)^{-1} = \int_1^2 30 \frac{W}{mK} \cdot \left[ \cos((1-i) \cdot 2 \cdot \pi) \cdot (0.5 \cdot \frac{x_{g}}{2}) \right] + (0.5 + \frac{x_{g}}{2}) \]
\[
\Rightarrow 1.066 \approx 1.905 \cdot (0.5 + 0.5 \cdot x_{g})
\]
\[
\Rightarrow 0.119 = x_{g}
\]

Thus it was calculated that a suitable approximation for the reduction of the thermal conductivity in the gap was \( x_{g} = 11.9\% \). As the underlying calculations are only an estimation in the first simulations a value \( x_{g} = 10\% \) was assumed in a first step as it simplifies the monitoring of the function.

After the calculation of the coating build-up, the resulting coating structure was transferred into a thermal conductivity calculation model. In this model a heat flow of \( \dot{q} = 2,000,000 \, W/m^2 \) was applied onto the upper surface of the simulated RVE. This heat flow results in temperature difference, which can then be used to calculate an effective thermal conductivity \( \lambda_{\text{eff}} \) of the simulated coating structure. A schematic representation of the thermal conductivity calculation model is shown in figure 4.

![Figure 4. Schematic representation of the thermal conductivity calculation model.](image)

The effective thermal conductivity \( \lambda_{\text{eff}} \) is then calculated according to equation (4) with the value of the applied heat flow \( \dot{q} \), the thickness \( s \) of the created coating and the temperature difference \( \Delta T = (T_1 - T_2) \), which can be monitored through the created export function.

\[
\lambda_{\text{eff}} = \frac{\dot{q}}{s \cdot \Delta T}
\]

(4)

3. Results and discussion
Figure 5 and 6 show a completed simulation of the particle impact model. The particle inlet temperature for this run was 2,673 K and the particle velocity was 200 m/s. While figure 5 shows the numbered particles, in figure 6 the adjusted thermal conductivity resulting from the implemented gap function is shown. It is clearly visible that at the centre of each particle the thermal conductivity remains at the value of alumina of 30 W/(m·K). At the edge and in the transition region to different particles this value
drops to 3 W/(m-K), so exactly the desired 10 % of the original thermal conductivity. Therefore, it can be concluded that the basic operability of the developed function is given.

Figure 5. Numbered particles of the completed simulation of thermally sprayed particles.

Figure 6. Thermal conductivity of the completed simulation of thermally sprayed particles.

While examining the completed simulations, it was noticed, that the border zone of the domain was not sufficiently filled with particles, as seen in figure 7. This can occur due to the Gaussian distribution of the particles onto the calculation domain. In order to circumvent that issue, every part except the interior 150 µm × 150 µm domain was cut and disregarded, which is shown in figure 8. The remaining part now represents a reduced RVE for the simulated coatings.

Figure 7. Porosity (VoFAlumina < 0.5 is displayed in green) of a completed simulation.

Figure 8. Apparent porosity after cutting the zones at the model boundaries to the reduced RVE.

Following these first runs a series of simulations, each with different values for percentage of the thermal conductivity in the gap $x_{\text{g}}$, were calculated to investigate its influence on the effective thermal conductivity of the coatings. The results of these adjustments are shown in Figure 9. It can be clearly observed, that the gap value has a direct influence on the effective thermal conductivity of the coating. The calculated thermal conductivity varies between 5.8 W/(m-K) and 11.4 W/(m-K) depending on the chosen gap value. As expected, an increasing conductivity in the gap $x_{\text{g}}$ leads to an increased effective thermal conductivity of the coatings. The implemented function and the gap value work as intended.
Figure 9. Visual representation of the resulting thermal conductivity with different values for the thermal conductivity of the gap and the reference value, calculated from table 2; the error bar indicates the minimum and maximum reported values in literature.

Additionally, to the calculated values a reference value for plasma sprayed alumina coatings is given in figure 9. The reference value is a mean value of various thermal conductivities found in literature as shown in table 2. This value is even below the effective thermal conductivity for a value of x% = 10 %. Two possible explanations could lead to this overestimation of the thermal conductivity by the model. First of all, the presented model is unable to simulate cracks inside solidified particles. As seen in figure 1, apart from the gaps between the splats, there exist multiple cracks in thermally sprayed ceramic coatings. These cracks are additional obstacles to the heat flow and thus they would reduce the effective thermal conductivity. Secondly, this model ignores any occurring phase transformations and treats all apparent Al₂O₃ as α-Al₂O₃. It is known in literature, that plasma sprayed coatings with higher content of α-Al₂O₃ exhibit a higher thermal conductivity than coatings with higher contents of γ-Al₂O₃ [12]. However, plasma sprayed coatings mostly consist of γ-Al₂O₃ [13], therefore it seems reasonable that the model overestimates the effective thermal conductivity compared to experimental values. For the reasons mentioned above, the previously estimated gap value was therefore retained at xₐₐ = 10 % and not adjusted to fit the literature values.

Table 2. Effective thermal conductivities λ [W/(m·K)] of thermally sprayed alumina coatings in literature.

| λ [W/(m·K)] | Reference |
|-------------|-----------|
| 1.1-1.5     | [14]      |
| 2.1-2.3     | [15]      |
| 2.45-9.99   | [16]      |
| 3-7         | [17]      |
| 5.9         | [18]      |
| 3.64        | [19]      |
| 10          | [20]      |

Mean value: 4.89
By varying the particle temperature and velocity, the influence of these particle inflight properties on the effective coating properties was investigated. As mentioned, during this procedure the gap value was kept constant at $x_{\%} = 10\%$. The resulting thermal conductivity and the porosity of the coatings are displayed in Table 3. In general, the thermal conductivity varies only in a limited range between 7.87 W/(m-K) and 8.49 W/(m-K). Furthermore, no clear trend is observable. Two reasons could provide a potential explanation. On the one hand, again the missing modelling of cracks and phase transition could be the reason for these small deviations. On the other hand, the limited variety of the chosen inflight parameters could lead to the observed limited differences in thermal conductivity. As the porosity varies also very little and is not affected by the first mentioned causes, the second explanation seems therefore more probable.

### Table 3. Results of the coating models thermal conductivity with different values for the particle injection temperature and velocity.

| particle temperature [K] | particle velocity [m/s] | gap-value $x_{\%}$ [%] | domain-size [µm×µm] | thermal-conductivity $\lambda_{\text{eff}}$ [W/(m·K)] | porosity [%] |
|--------------------------|-------------------------|------------------------|----------------------|--------------------------------|-------------|
| 2,473                    | 180                     | 10                     | 150×150              | 8.24                           | 8.1         |
| 2,673                    | 180                     | 10                     | 150×150              | 8.05                           | 8.2         |
| 2,873                    | 180                     | 10                     | 150×150              | 8.13                           | 8.1         |
| 2,473                    | 200                     | 10                     | 150×150              | 8.49                           | 8.4         |
| 2,673                    | 200                     | 10                     | 150×150              | 8.29                           | 8.1         |
| 2,873                    | 200                     | 10                     | 150×150              | 7.87                           | 7.9         |
| 2,473                    | 220                     | 10                     | 150×150              | 8.33                           | 8.2         |
| 2,673                    | 220                     | 10                     | 150×150              | 8.03                           | 8.0         |
| 2,873                    | 220                     | 10                     | 150×150              | 7.94                           | 8.0         |

### 4. Conclusions and outlook

To successfully simulate the coating build-up of thermally sprayed coatings, including the air gaps between two solidified particles, the necessary mesh size would be extremely small. An approach to implement this gap by a function while keeping the calculation time low was presented in this work. A particle impact model followed by the calculation of the effective thermal conductivity were created and a function, which detects the area between two particles and adjusts the thermal conductivity to a percentile of the particles’ material thermal conductivity was implemented. The following results were observed:

- A function to identify the individual particles was successfully implemented
- The consecutive gap function was able to reduce the thermal conductivity at the splat boundaries
- The presented model for the coating build-up is able to simulate an RVE for a ceramic coating, by controlling the gap value $x_{\%}$, the coatings thermal conductivity could be adjusted
- The simulated coatings exhibited a higher effective thermal conductivity than the reported literature values for an estimated gap value $x_{\%}$; the missing representation of crack formation and of phase transitions are the believed reasons for this difference
- Variations of the particle in-flight properties did not lead to significant changes of the effective thermal conductivity; the limited range of the input parameters is the most probable cause

Future work needs to concentrate on a broader variation of the particles’ in-flight properties to simulate a possible difference due to changes in the process parameters of the plasma spraying process. Furthermore, distributions of particle velocities and temperatures coming from an existing plasma generator model will be used in this model. Finally, the implementation of models for crack propagation and phase transition are crucial to predict correct coating properties in the future.
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