Temperature distribution on thermally sprayed heating conductor coatings

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Abstract. Cavities in injection moulding can be coated to increase the corrosion and wear resistance or to adapt the heat flux through the cavity wall. The latter is to be achieved by thermal spraying of TiO$_2$/Cr$_2$O$_3$ on the surface and its electrical heating. A thermally sprayed Al$_2$O$_3$ coating serves as an electrical insulator between the steel substrate and the TiO$_2$/Cr$_2$O$_3$ coating. In this study, the feasibility of a homogeneous surface heating is investigated. The heating behaviour was analysed using a thermographic camera. Depending on the process parameters during the coating process and used electrical current, inhomogeneity in the temperature distribution was detected. The observed inhomogeneity was distributed in linear patterns in form of “hot lines” perpendicular to the electrical current. To identify the root cause of the observed inhomogeneity, numerical modelling was utilized. The results of the simulations were validated with the experimental measurements. It was found that the cracks in the coating microstructure were the prime cause of the temperature increase that was distributed in linear patterns. Moreover, even though the crack distribution does not have a directional preference, the pronounced heating of the cracks aligned in the perpendicular direction to the electrical current created the temperature pattern.

1. Introduction
Thermal spraying has been used to apply coatings that protect parts from corrosion, wear and thermal loads for a long time. In recent years, novel applications of thermally sprayed coatings such as sensors, electronic components and thermoelectric devices have been developed [1, 2]. This work focuses on thermally sprayed coatings as an electric heater for the application in the injection moulding tools. Figure 1 shows a coating system concept, consisting of a heating element coating surrounded by two Al$_2$O$_3$ coatings for electrical insulation. The application of thermal spraying to produce electrical heaters on part surfaces with vacuum plasma spraying, air plasma spraying as well as high-velocity oxygen fuel spraying has been analysed in the literature [3–6]. The feasibility of using metallic materials, such as Ni, NiCr and NiAl, as a heating element up to 600 °C has been established. In this work, a semiconductor material was chosen for the heating element coating for its advantages over metallic materials. Due to higher electrical resistance of semiconductors, the same amount of heating power can be achieved with less electrical current flowing through them. This would allow for thinner electrical cabling that connects the heating element with the power source, which could simplify the integration of the heating element in the injection moulding tool. Moreover, the affinity of semiconductors with ceramics leads to a better bonding between the two and their similar thermal expansion coefficients ensures lower residual stress at the interface of the insulator and the heating element coating.
Prototypes were manufactured on steel substrates and electrically heated while their heating behaviour has been investigated using a thermal camera. The material used as an electric heater is TiO$_2$/Cr$_2$O$_3$. An Al$_2$O$_3$ coating was applied for insulation on the surface of a steel substrate. Analysis of the surface temperature of the coating by thermal imaging during its electrical heating has shown varying level of homogeneity. The factors influencing the temperature distribution on the coating surface are the process parameters used during the application of the coating, the coating thickness and the rate at which the electrical heater coating has been heated. Using appropriate spraying parameters and heating the coating at moderate to high rates, it is possible to achieve a homogeneous temperature distribution, which is a desirable outcome for the application in injection moulding [7–9].

Certain coating and operating parameters resulted in appearance of spots and lines with significantly higher temperatures compared to their immediate vicinity. This study aims to identify the root cause for this inhomogeneity so that they can be prevented. The temperature inhomogeneity exhibited a directional preference depending on the direction of the electrical current. Numerical modelling of the resistive coating heating was used to investigate the underlying physical phenomena that causes the directional preference of the temperature inhomogeneity. The numerical model is validated experimentally, for the case of the coating heating up with a homogeneous temperature distribution, by comparing the peak temperatures. In the case of the coating system which shows hot lines in its temperature distribution, only a qualitative comparison between experimental and numerical results was feasible. In the next sections, the heating conductors and the thermal spraying process will be briefly introduced. Afterwards, the experimental setup for the coating application and analysis is presented. The obtained results will be presented and discussed and finally, the conclusion is drawn.

1.1. Electrical heating of heating conductors

In an electrical heater, the electrical energy ($E_{el}$) is converted into the heat energy ($Q_W$). The correlation between power and heat energy is defined by the following equation:

$$Q_W = E_{el} = P \cdot t$$

where $P$ denotes the power of the electrical heater and $t$ the duration of the heating period. The power of the electrical heater is determined by the following equation:

$$P = U \cdot I = \frac{U^2}{R} = R \cdot I^2$$

where $U$ corresponds to the effective electrical voltage, $I$ to the electrical current and $R$ to the electrical resistance. For a heating element with a known electrical resistance, the electrical power can be
calculated based either on the electrical voltage or the electrical current. The electrical resistance of a heating element depends on material properties and its shape:

$$R = \rho(T) \frac{l}{A}$$  \hspace{1cm} (3)

where $\rho(T)$ represents temperature dependent resistivity, $l$ the length of the element in the direction of the electrical current and $A$ the cross-sectional area of the element perpendicular to the electrical current. The ratio of the element’s length to its cross-sectional area is referred to as the shape factor.

The material used in this work, TiO$_2$/Cr$_2$O$_3$, is a semiconductor whose electric and thermoelectric properties can be influenced by controlling the process parameters during the spraying process [10, 11]. The electrical conductivity of a semiconductor generally increases with increasing temperature. This is explained by the increase of intrinsic movement of electrons at higher temperatures, which increases the probability of an electron leaving its orbit. With the help of the heat energy introduced into the semiconductor, an electron is moved to a higher energy level, which is called the conduction band, leaving an electron hole in the so called valance band. The energy difference between the energy levels defines the band gap. The electrons from the neighbouring atoms tend to fill the electron hole, leaving another electron hole in their place. The movement of the electron holes creates a movement of positive charge across the material. Materials where the carrier of the electrical charge is the electron holes are called the p-type semiconductors. It is possible to improve the conductivity of a semiconductor by doping its crystal lattice with impurities. In the case of TiO$_2$, the electrical conductivity is induced by oxygen vacancies. Cr$_2$O$_3$ is used to increase the stability of the sub-stoichiometric oxides [12].

1.2. Thermal spraying

The heating element as well as the insulator coatings in this work were manufactured using air plasma spraying. It is a process variant of thermal spraying, where molten particles impact on the part surface and undergo rapid solidification. During this rapid solidification, intrinsic residual stresses build up in the coating. The resulting coating has a microstructure with varying porosity, depending on the impact characteristics of the particle and therefore, on the process parameters. Due to different thermal expansion coefficients of the coating and substrate materials, extrinsic residual stresses build up as the coating and the substrate cool down. The intrinsic and extrinsic residual stresses superpose and can have a negative impact on the mechanical characteristics of a coating system. A higher coating thickness leads to a residual stress increase and can cause cracks in the coating microstructure [13].

2. Experimental setup

2.1. Thermal spraying of the coatings

The coating concept developed for the injection moulding application, as shown in Figure 1, consists of a heating element coating which is encased by two insulation coatings. The coating system that was investigated in this work, however, consists of a single electrical insulator coating between the substrate and the heating element coating. The insulation coating on top of the heating element coating is necessary to electrically protect the molten plastic material from the heating element material and to chemically protect the heating element from the molten plastic. Since the temperature distribution of the heating element is investigated in this work, the top layer of coating system (Figure 1) of has not been considered. This enables a closer investigation of the temperature distribution and the failure mechanisms of the heating element coating. The process parameters that were used to manufacture the coating system are listed in Table 1.
Table 1. Spraying parameters of the insulation and the heating element coating.

| Parameter                        | Insulator/Heating element |
|----------------------------------|---------------------------|
| Plasma generator                 | F4MB-XL                   |
| Argon [SLPM]                     | 42                        |
| Hydrogen [SLPM]                  | 9                         |
| Electrical current [A]           | 600                       |
| Net power [kW]                   | 36                        |
| Spraying distance [mm]           | 100                       |
| Conveyor disc rotation speed [%] | 27/20                     |
| Injector Argon [SLPM]            | 6.5                       |
| Meander velocity [mm/s]          | 875/1,000                 |
| Meander step [mm]                | 5                         |

The tooling steel substrates used in this work have the size of 102x61x9 mm³. First, the substrate surfaces have been prepared by grit blasting and coated with an Al₂O₃ powder (GTV 40.05.0 Aluminium oxide 99.9%) with particle size fraction of D = -25+5 µm. The resulting insulator coating had the thickness of t = 250 µm and covered the entire surface of the substrate. On top of the Al₂O₃ coating, a TiO₂/Co₃O₄ powder (Ceram 80/20 molten) with particle size fraction of D = -25 + 5 µm was used to apply the second layer. The resulting heating element coating had the thickness of t = 110 µm and covered an area smaller than the surface of the substrate with the size of 80x50 mm². This was necessary to eliminate a possible contact of the heating element coating with the substrate along the substrate edges and achieved by masking the edges on the insulator surface prior to spraying. The microstructure of both coatings can be seen in the cross-section image, which is shown in Figure 2. The final layout of the coating system can be seen in Figure 3. Another coating system has been manufactured with the same set of process parameters, except with the thickness of the heating element coating of t = 400 µm. Higher thickness of the second coating system was necessary to ensure residual stresses large enough to initiate crack formation. In contrast to the t = 110 µm thick coating, the coating with the thickness of t = 400 µm exhibited temperature inhomogeneity in form of the hot lines in preliminary tests (Figure 6). This is an expected behaviour due to the forced crack formation.

Figure 2. Cross-sectional picture of the microstructure of the insulation and heating element coatings.
2.2. Electrical heating and thermal imaging

The experimental setup for the electrical heating of the coatings is schematically shown in Figure 3. The contact plates have been clamped on the sides of the heating element coating and connected to the voltage source. The distance between the contact plates is \( L = 70 \) mm, which is shorter than the full length of the heating element coating \( L = 80 \) mm. The power supply unit CHROMA 61604 (dataTec) was used as the voltage source, which can supply both alternating and direct currents.

In this work the alternating current was used, which can supply a maximum current magnitude of \( I = 8 \) A due to the device limitations. The electrical voltage is given to the power supply as an input, whereas the resultant electrical current flowing through the heating element coating recorded on-line. Since the semiconductor coating ceramic is a better electrical conductor at higher temperatures, as measured and shown in Figure 4, the electrical current increases as the coating system, the substrate and the mounting system which is in contact with the substrate, gradually warmed up until reaching the equilibrium state. Lower values of electrical voltage were applied to avoid hitting the device limitation of \( I = 8 \) A as the system reached the equilibrium. The temperature dependent electrical resistance values for the heating conductor coating, which are shown in Figure 4, were obtained with the help of the temperature measurements on the coating surface by thermal imaging, the temperature dependent electrical current value and the electrical voltage input. Unlike the metallic heating element coatings, where the resistance of the heating element increases with temperature [5, 6], the resistance of the semi-conductor ceramic coatings decrease with increasing temperature as it is confirmed in this study as well.

![Figure 3. Schematic description of the experimental setup for the heating and thermal imaging measurements.](image1)

**Figure 4.** Measured dependence of the electrical resistance on temperature.
An infrared camera ThermaCam SC500 (Flir Systems) was used for the thermal imaging. The camera has an image resolution of IR = 320x240 pixels, recording frequency of f = 50 Hz and a temperature measurement accuracy of ΔT = ± 2 °C. A macro lens was mounted on the thermal camera, which allowed to zoom in on the coating surface and record the surface temperatures of the coating with a pixel resolution of PR = 8.4 pixels/mm. It should be noted that due to the recording with the macro lens, the margins of the coating were not captured on the thermal images. The emissivity value of the coating surface has a major effect on the precision of the temperature measurement with an infrared camera. The emissivity value of the heating element coating was calibrated based on the boiling temperature of water and determined to be ε = 0.48. Figure 5 shows a heating element coating with the thickness of t = 110 µm being heated with the electrical voltages of U = 15 V and U = 20 V.

Figure 5. Thermal images of the coating system with the heating element coating thickness of t = 110 µm at electrical voltage of U = 15 V (left) and U = 20 V (right).

Upon turning on the voltage source, the temperature of the coating rose gradually and reached quasi-equilibrium state. In accordance with the preliminary tests, hot lines were not detected neither during the heating up, nor at the equilibrium stage. This corresponds to expectations, since low residual stresses within the coating could not have caused crack formation. The thermal images shown correspond to the steady state temperatures of the coating surface. After the coating temperatures have reached their peak value, the temperature of the substrate side near the contact plate were measured to be T = 70 °C for U = 15 V and T = 112 °C for U = 20 V with an infrared temperature sensor, since the sides of the substrate are not visible on the thermal images. These values will be used as a boundary condition in the numerical modelling to take into account the cooling effect of the entire experimental setup on the substrate, and therefore, on the coating. Figure 6 shows four different directions of electrical current flow rotated around the spot in the middle of the sample. Each temperature pattern shows a directional alignment perpendicular to the electrical current.
3. Numerical modelling

For the numerical modelling of the resistive heating of the coating system, the Thermal-Electric module of the simulation package Ansys® has been used. The substrate and the coating geometries were modelled according to the dimensions given in section 2.1, except for the length of the heating element coating. It was taken as the distance between the contact plates $L = 70$ mm to ensure correct application of the electrical voltage boundary condition. The material properties used in the simulations were taken from the material database “Sample materials data from Granta Design” in Ansys Workbench and are given in Table 2. The material properties in the range of $T = 15 – 150$ °C were assumed to be constant due to negligible variation in the given temperature range. An exception is the temperature dependence of electrical resistivity of TiO$_2$/Cr$_2$O$_3$. It was taken into account as the resistivity of TiO$_2$/Cr$_2$O$_3$ decreases significantly at high temperature and this phenomenon has a major effect on the heating behaviour of the coating system.

The boundary conditions used in the model are the electrical voltage on the sides of the TiO$_2$/Cr$_2$O$_3$ coating, cooling due to the radiation and convection as well as the temperature boundary condition on the sides of the substrate. The simulation results are shown in Figure 7 for the electrical voltage boundary condition of $U = 15$ V and $U = 20$ V. Both results show the same temperature distribution, with their temperature peaks centred in the middle of the heating element coating. The maximum temperatures of $T = 83.3$ °C and 145.6 °C respectively show a very good correspondence to the experimental measurements of the maximum coating temperature of $T = 82.4$ and 145.6 °C respectively. This shows the suitability of the developed numerical model for the electrical heating of heating element coating systems.
Table 2. Material properties used in the simulations.

| Property/Material                  | Value          |
|-----------------------------------|----------------|
| Density/Steel substrate           | 7,860 kg/m³    |
| Thermal conductivity/Steel substrate | 37.5 W/m K     |
| Specific heat/Steel substrate     | 473 J/kg K     |
| Density/Aluminum oxide            | 3,700 kg/m³    |
| Thermal conductivity/Aluminium oxide | 24 W/m K      |
| Specific heat/Aluminium oxide     | 880 J/kg K     |
| Density/TiO₂/Cr₂O₃               | 4,000 kg/m³    |
| Thermal conductivity/TiO₂/Cr₂O₃  | 3 W/m K        |
| Specific heat/TiO₂/Cr₂O₃         | 690 J/kg K     |
| Resistivity/TiO₂/Cr₂O₃           | 0.0003-0.0015 ohm m |

*Temperature dependent values based on the plot in Figure 4.

Figure 7. Temperature distribution on the coating systems obtained by numerical modelling. The applied electrical voltages are U = 15 V (left) and U = 20 V (right).

The hypothesis of this work is that the hot lines seen in Figure 6 are caused by the cracks in the heating element coating, which emerge at higher coating thicknesses due to residual stress. In order to test this hypothesis, random artificial cracks with the depth of d = 380 µm are introduced into the heating element coating with the thickness of t = 400 µm (Figure 8). As can be seen in the figure, the artificial cracks are exaggerated in size and do not represent a realistic distribution. It can be also seen that the distribution of the cracks follow no directional pattern.

Figure 8. The pattern of artificial cracks in the heating element coating.
A voltage of $U = 60$ V was applied horizontally and vertically on the sides of the heating element coating. The resulting patterns of the temperature (T), current density (I/A) and Joule heating (JH) are illustrated in Figure 9. The directional preference of the temperature inhomogeneity is evident. Moreover, it can be seen that the current density and therefore the Joule heating are significantly higher in the introduced cracks and show a directional preference corresponding to the direction of the applied voltage. This supports the hypothesis that the inhomogeneity in the temperature distribution in form of hot lines can be caused by the cracks in the coating microstructure which have no directional preference. It should be noted that the coating temperature was significantly lower than it would be expected from a uniform coating with the same coating thickness and applied electrical voltage. This can be explained by the fact that the introduced cracks significantly increased the electrical resistance of the coating, thus leading to a lesser amount of electrical heating.

**Figure 9.** Results of the electrical heating simulation of a heating element coating with artificial cracks. The electrical voltage was applied horizontally (left column) and vertically (right column).
4. Summary and outlook
An electrical heater coating system with the heating element coating thickness of \( t = 110 \) µm has been thermally sprayed and analysed with thermal imaging under application of electrical voltages of \( U = 15 \) V and \( U = 20 \) V. The results of the numerical modelling showed good correspondence with the experimental temperature measurements. Based on this model, the resistive heating of a coating system with a random pattern of artificially introduced cracks was simulated. Based on the direction of the applied electrical voltage, directional preference in the temperature, electrical current density and Joule heating distributions could be detected. It was seen that the introduction of the artificial cracks had a negative impact on the resistive heating characteristics of the coating system. While a quantitative experimental verification was feasible for the simulation of the homogeneously heated coatings, only a qualitative comparison was made for the case with the temperature inhomogeneity in form of hot lines. In future works, the cracks in the coating microstructure should be experimentally analysed with respect to their size and depth distribution as well as to their orientations. By introducing more realistic artificial cracks in the numerical model, a quantitative comparison will become feasible for this case as well.

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