Determination of Cooling Rate and Temperature Gradient during Formation of Cathode Spot Craters in a Vacuum Arc

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Abstract: Due to the extreme thermal conditions and short lifetimes, experimental exploration of cathode spots in vacuum arcs is very difficult. The intensive heat in the cathode spot is believed to be generated by ion bombardment and by Joule heating. However, thermal conditions occurring inside the re-melted material in craters created by cathode spots are not accurately known. During the exposure to cathodic arc plasmas, an Al-Cr cathode’s surface was locally melted by successive ignition and extinction of cathode spots. The melted layer, that quickly solidified, was characterized by the formation of several thin layers with a thickness of a few micrometers that were stacked on top of each other. The corresponding solidification patterns displayed cellular and dendritic microstructures. A phase field-based model was used to simulate and determine the thermal process conditions that led to the dendritic structures observed within the re-melted layer. Different combinations of cooling rates and temperature gradients were numerical explored to determine the most probable thermal conditions under which the cathode material re-solidifies. The results showed that the material in the vicinity of the cathode spot crater re-solidified under the condition of a cooling rate of about $3 \times 10^5$ K/s and a temperature gradient of about $6 \times 10^7$ K/m. These results constitute valuable data for the validation of numerical models dedicated to cathode spot formation.

Keywords: cathode spot; MICRESS; simulation

1. Introduction

Cathodic arc deposition is a technique frequently used for the deposition of thin films and hard coatings due to its versatility and the achievable high deposition rates [1]. The evaporation process begins with the impact of a high current, low voltage arc on the surface of the cathode, which results in a small, usually a few µm wide, high energy emitting area. The resulting localized temperature at the so-called cathode spot is so high that a beam of vaporized cathode material is created that can be employed to deposit thin films and hard coatings [2]. After the extinction of the cathode spot plasma, a crater remains on the cathode surface. The cathode spot itself is only active for a short period of time before it ignites again, typically in close vicinity. As the spot moves rapidly over the entire cathode surface, the entire surface is covered by countless overlapping craters and re-melted material after some time [3]. The resulting modified layer can have a thickness of up to several µm and, in the case of multi-element cathodes, may alter the plasma properties, where the latter may affect the growth conditions of the thin films or hard coatings, e.g., in terms of chemical composition or presence of macroparticles or droplets.

For these reasons, detailed investigations of modified layers on multi-element arc cathodes are of interest. Recently, the effects of arc discharges on the erosion of Al-Cr composite cathodes have been investigated [4–6]. The detailed analysis revealed the
formation of modified surface layers where Al and Cr are intermixed, forming intermetallic Al-Cr phases. These phases and corresponding concentrations correspond fully to the phase predicted by the equilibrium phase diagram. The microstructure exhibited several stacked layers, a consequence of the recurring process of melting and solidification of the surface-near material by the arc plasma. Based on secondary dendrite arm spacing measurements, an average cooling rate was estimated to be in the order of $10^6$ K/s [6]. A detailed description of the cathode manufacturing and arc discharge parameters can be found in [6]. The current work aims to present the realization of the numerical investigations to determine the temperature gradient and the cooling rate, which led to a dendritic microstructure of the melted-re-solidified layer, similar to modified layers encountered on cathode surfaces as shown in Figure 1. For this purpose, several simulations were performed with the help of the phase field-based software MICRESS 6.2 [7]. Based on the fact that the microstructure is directly related to the solidification history, thermal conditions during re-solidification of the melted material can be estimated.

![Figure 1.](image-url)

**Figure 1.** (a) SEM micrograph of an arc cathode surface formed on an Al-Cr cathode with a few craters, and (b) cross-section of a modified layer on an eroded arc cathode showing a stack of thin re-melted layers exhibiting dendritic solidification morphology.

### 2. Modelling

Since the conditions in the cathode spot and during the formation of the craters are largely unknown, the required boundary conditions are first constrained by using the Fermi approach as described in the Section 2.1. Based on the range of possible process parameters, several numerical investigations were systematically performed. The chosen numerical parameters and process conditions are described in the next section.

The cast microstructures reflect the solidification history; therefore, conclusions can be drawn about the growth and process conditions. In a first step, the magnitude of the solidification length was experimentally estimated. For this purpose, the size of the craters and the different thicknesses of the modified layer were determined. Scanning electron microscope (SEM) (ThermoFischer FEI Versa 3D HiVax, Waltham, MA, USA) investigations of the cathode surface display a vast number of craters which are the result of cathode spot events (Figure 1a). The radius $R$ of the craters on the cathode was estimated to be in the order of $R = 20 \mu$m. After some time of arc plasma exposure, a continuous modified layer is established due to a multitude of cathode spots ignited on the cathode’s surface. Details of the layer in the form of a cross-section are given in Figure 1b, which displays several re-melted layers with a clear dendritic microstructure. The detected solidification layers exhibited a thickness of about $3 \mu$m $\leq L \leq 6 \mu$m.

A detail of a dendrite is shown in Figure 2. According to the Al-Cr phase diagram (see Figure 3), the dendrite is enriched in Cr and embedded in an Al-enriched darker matrix [5]. The evaluation of the solidification structure shows a secondary arm distance (DAS) of about $\lambda_2 = 175 \pm 25$ nm. A detailed study using energy-dispersive X-ray spectroscopy (EDAX-AMETEK, Mahwah, NJ, USA) of the element distribution on this Al-Cr cathode was presented in [6].
The evaluation of the solidification structure shows a secondary arm distance (DAS) of about \( \lambda_2 = 175 \pm 25 \) nm. A detailed study using energy-dispersive X-ray spectoscopy (EDAX-AMETEK, Mahwah, NJ, USA) of the element distribution on this Al-Cr cathode was presented in [6].

**Figure 2.** SEM micrograph showing a part of a dendrite with an average secondary arm spacing of \( \lambda_2 = 175 \pm 25 \) nm. The spacing was calculated from the length (red line) divided by the number of arms within this distance.

**Figure 3.** Cr-Al phase diagram calculated with Thermo-Calc [8] and the expected solidification path (arrows).
2.1. Estimation of the Process Parameters

The secondary arm spacing will serve as target for our numerical simulations. The best fitting results will give reasonable estimation of the actual thermal process parameters. However, a reasonable range of process parameters (cooling rate and temperature gradient) should be first extracted from thermodynamics and literature data.

The following parameters were used:

(i) The Cr-Al phase diagram provides the thermodynamical information relating phases, concentrations, and phase transition temperatures [8];
(ii) The expected relationship between the cooling rate $T$ and the secondary dendrite arm spacing $\lambda_2$ was determined based on equations published in [9];
(iii) The criteria for the stability of the solid/liquid (s/l) interface defined whether the diffusion length was smaller than the size of the microstructures and whether a rapid solidification process was present, and the magnitude of the temperature gradient was estimated based on the corresponding equilibrium phase diagram for a Cr-Al cathode shown in Figure 3.

(i) The Cr-Al phase diagram and the expected solidification path for a concentration of $C = 50$ at.% is displayed as red arrows in Figure 3. The creation of a crystal from an alloy melt causes a local change in the composition. During equilibrium solidification the concentration of the solid $C_S$ follows the solidus line, whereas the concentration of the liquid at the interface $C_L$ follows the liquidus line. Both concentrations are connected by a relatively constant distribution coefficient $k = C_S / C_L$. In the case of too rapid solidification conditions [10], a non-equilibrium solidification leads to a highly supersaturated crystal. In this case, the liquidus line correlates with the solidus line and the distribution coefficient $k$ becomes 1.

The used data taken from the phase diagram [8] are given in Table 1. The ThermoCalc coupling for MICRESS to read in a database was not used. Instead, a linearized phase diagram based on Figure 3 was used. The missing material values such as the diffusion coefficients $D_L$, $D_S$, and surface energy $\sigma$ were estimated in the order of magnitude for typical metals. The entropy of fusion $S_f$ was taken from the thermodynamic database COST2 [11].

| Name                                      | Symbol | Value       | Unit    |
|-------------------------------------------|--------|-------------|---------|
| interfacial solid–liquid (s/l) energy     | $\sigma$ | $1.0 \times 10^{-9}$ | J/m²   |
| entropy of fusion                         | $\Delta\sum_{\phi}$ | $1.1 \times 10^6$ | J/m³ K |
| Gibbs–Thompson coefficient                | $\Gamma$ | $9.1 \times 10^{-7}$ | m·K    |
| diffusion in the liquid                   | $D_L$   | $1.0 \times 10^{-9}$ | m²/s   |
| diffusion in the solid                    | $D_S$   | $1.0 \times 10^{-12}$ | m²/s   |
| concentration in the liquid               | $C_L$   | 64.0        | at.% Al |
| concentration in the solid                | $C_S$   | 35          | at.% Al |
| initial concentration in the liquid       | $C_0$   | 50.0        | at.%   |
| liquidus temperature                      | $T_L$   | 1773.0      | K      |
| solidus temperature                       | $T_S$   | 1593.0      | K      |
| slope of liquidus line                    | $m_L$   | $-12.9$     | K/at.% Al |
| distribution coefficient                  | $k$     | 0.55        |        |

(ii) Salas et al. [12] published the relation between the cooling rate $\dot{T}$ and the secondary DAS $\lambda_2$ based on experimental results for 70–30 brass and theoretical reflections [9], as shown in Figure 4. In the case of the secondary DAS measured for an Al-Cr alloy, in Figure 2, the predicted cooling rate that formed the modified layers can be estimated to be in the order of $\dot{T} > 1 \times 10^6$ K/s by using the findings from [12].
which can also be written as function of the cooling rate \( \dot{T} \).

\[
\lambda_2 = 5.5 \times (K \times t_f)^{0.33},
\]

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\[
\lambda_2 = 5.5 \times (K \times \frac{\Delta T}{T})^{0.33}
\]

Here, \( \Delta T \) is the temperature difference between the liquidus \( T_l \) and the solidus \( T_s \) and \( K \) is a coarsening parameter defined by

\[
K = \frac{\Gamma \times D_L \times \ln \left( \frac{C_f}{C_L} \right)}{m_L \times (1 - k) \times (C_0 - C_L)}
\]

Based on the aforementioned correlations (Equations (1) and (2)) and the values given in Table 1, we can estimate a range for both the solidification time, \( 6.6 \times 10^{-5} \leq t_f \leq 1.6 \times 10^{-5} \) s, and the cooling rate, \( 1.5 \times 10^7 \leq \dot{T} \leq 2.7 \times 10^7 \) K/s.

(iii) The formation of the solidification patterns is related to the cooling rate. If the cooling rate is sufficiently high, a planar solidification front becomes unstable and the solid/liquid (s/l) interface changes from planar to cellular and to a dendritic solidification structure. Finally, the solidification front becomes again a stable planar front for high growth rates in the case when the solute diffusion distance \( \delta_c \) approaches the solute capillarity length \( s_c \) [7]. The solute diffusion distance \( \delta_c \) is given by

\[
\delta_c \approx \frac{D}{V}
\]

and the solute capillary length \( s_c \) is

\[
s_c \approx \frac{\Gamma}{T_L - T_S}
\]
If the solidification rate exceeds the marginal growth rate \( V_a \), also known as absolute velocity \([10]\), the interface forms again a planar front. The absolute growth velocity is given by

\[
V_A = \frac{(T_L - T_S) \times D_L}{k \times \Gamma}
\]

By using Equations (4) and (5) and the specification from Table 1, the absolute velocity \( V_a \) is reached for the system Al-Cr when the solidification velocity exceeds \( V \sim 2.0 \times 10^6 \, \text{µm/s} \). Due to the experimentally observed dendritic solidification structure, it can be assumed with confidence that the absolute velocity was not reached within the modified layer and, hence, the actual solidification velocity can reasonably be assumed to be much smaller than the absolute velocity.

(iv) The estimation of the temperature gradient is of particular importance, since this, together with the surface energy \( \sigma \) and the diffusion coefficient in the liquid \( D_L \), has the greatest influence on the numerical investigations. In contrast to the two parameters mentioned above, the temperature gradient can vary by several orders of magnitude. In return, the melt surface temperature was just below the evaporation temperature. From this, it follows in a simplified form that the temperature gradient acts over the entire melting depth, which was roughly the same as the solidification length. In order to estimate the temperature gradient, the size of the solidification length \( l_{sol} \) and the temperature difference \( \Delta T \) should be estimated. The solidification length \( l_{sol} \) was found to be in the range of 3 µm for the cross-section direction (Figure 1a) to 20 µm for the horizontal direction (Figure 1b), or \( l_{sol} = 11.5 \pm 8.5 \, \text{µm} \).

In cathodic arc plasmas, the surface of the cathode material is heated up by the arc spot in such a way that part of the material evaporates. The evaporation temperature depends on the pressure, which was set in this study to 1 Pa in the extended vacuum chamber. However, in the current case the pressure in the cathode spot is of interest which is generally much higher and believed to be in the order of 0.1 GPa \([1]\). Since the vaporization temperature increases with pressure, the evaporation temperature under ambient pressure was used as the minimum expected temperature at the melt/vacuum interface. The evaporation temperature was estimated to be \( T_B = 2612 \, \text{K} \) at the concentration \( C_0 = 50 \, \text{at.\%} \), provided that the evaporation temperature is between the values of the pure substances, \( T_B = 2755 \, \text{K} \) for Cr and \( T_B = 2470 \, \text{K} \) for Al. Considering a maximum undercooling of 3 K for the Al-rich peritectic phase at the peritectic plateau, the solidification finished at the latest at \( T = 1590 \, \text{K} \) (Figure 3). This assumption is based on the fact that the constant bombardment of the cathode surface provides sufficient surface fluctuations to activate the nucleation event of the peritectic phase. This led to an estimation of the minimum temperature interval \( \Delta T = 1022 \pm 143 \, \text{K} \).

Considering the extreme solidification lengths \( l_{sol} = 11.5 \pm 8.5 \, \text{µm} \), and the minimum temperature difference between melt and solid \( \Delta T = 1022 \pm 143 \, \text{K} \), the range of the temperature gradient is in the magnitude of \( G_T = 2 \times 10^8 \pm 1.5 \times 10^8 \, \text{K/m} \).

In summary, a Fermi analysis of the available data reveals the following statements for the process conditions that led to dendrites with a secondary side arm distance of \( \lambda_2 = 175 \pm 25 \, \text{nm} \):

(i) Based on the data published in \([12]\), the expected cooling rate is in the range of \( \dot{T} > 1 \times 10^6 \, \text{K/s} \);

(ii) The Feurer and Wunderlin \([9]\) equations suggest a cooling rate in the range of \( 1.5 \times 10^7 \leq \dot{T} \leq 2.7 \times 10^7 \, \text{K/s} \) and the expected solidification time is in the order of \( 6.6 \times 10^{-6} \leq t_f \leq 1.6 \times 10^{-5} \, \text{s} \);

(iii) The solidification velocity \( V \) did not reach the absolute velocity where the distribution coefficient \( k \) becomes 1;

(iv) The temperature gradient is in the magnitude of \( G_T = 2 \times 10^8 \pm 1.5 \times 10^8 \, \text{K/m} \) or higher.
Although the values show a large margin of error, they are sufficient to define the boundary conditions for the phase field simulations.

2.2. Phase Field Simulations

The phase field-based commercial software MICRESS 6.2 [7] was used to analyze the microstructure that is formed under the aforementioned process conditions. The “moving frame” option allows the use of a small simulation domain which follows the solidification front. The movement of the simulation domain was controlled by a constant distance between the s/l interface and the top of the domain. This distance was set to 0.5 µm. The simulation was carried out as a 2-dimensional domain with 500 × 500 cells in x- and z-direction and a resolution of x = 0.005 µm. The optimal cell size was determined by varying the cell size at automatic time steps. The interface mobility was constant, and the kinetic coefficient μ between the liquid and the phases was set to 500 cm$^4$/J·s. The temperature gradient was implemented in z-direction. The primary Cr phase was assumed to be anisotropic and cubic. Table 2 summarizes the selected numerical and physical parameters for the 2-dimensional numerical investigations.

Table 2. MICRESS, selected numerical parameters.

| Numerical Parameters               | Value     |
|------------------------------------|-----------|
| domain dimension                   | x 500 cells | z 500 cells |
| cell size                          | 0.005 µm  |
| Physical Parameters                |           |
| diffusion coefficient              | $D_L = 1.0 \times 10^{-9}$ m$^2$/s |
| surface energy                     | $\sigma_{s/l} = 1.0 \times 10^{-5}$ J/cm$^2$ |
| temperature gradient               | $G_T$ variable K/cm |
| initial concentration              | C 50 at.% |
| entropy of fusion                  | $S_{f,a} = 1.1$ J/cm$^3$·K |

3. Results

The solidification morphology was examined in discrete steps of cooling rates of 2.5 × 10$^5$ K/s, starting from $T = 1 \times 10^5$ until 1 × 10$^8$ K/s including the estimated range of the cooling rate from the previous section. For each of the cooling rates, three different temperature gradients ($G_T = 6 \times 10^6$, $6 \times 10^7$, and $6 \times 10^8$ K/m) were investigated. The results of the simulated microstructures are summarized for the same temperature gradient but for different cooling rates.

3.1. Predicted Microstructures for $G_T = 6 \times 10^8$ K/m

The initial s/l interface for all numerical investigations within this work is shown in Figure 5a. Due to the selected boundary conditions, namely, the low cooling rate and the high-temperature gradient, no growth took place. Therefore, the initial situation is equal to the final result of the simulation. By increasing the cooling rate, the solidification front remained planar (Figure 5a–d). The higher the cooling rate, the larger the undercooling at which the s/l grows.
Figure 5. (a-d) Planar solidification morphology for different cooling rates $\dot{T}$ and a temperature gradient of $G_T = 6 \times 10^8$ K/m. (e-h) show a cellular solidification morphology. Since the s/l interface temperature was below the peritectic temperature, the nucleation event of the peritectic phase was suppressed. The size of each image is 2.5 µm × 2.5 µm.

With higher cooling rates, the s/l temperature drops below the peritectic temperature of 1632 K. In the case that the s/l interface temperature was below the peritectic temperature, the s/l interface changed into a cellular growth condition. For cooling rates $\dot{T} \leq 2.5 \times 10^6$ K/s the solidification morphology changes from planar to cellular (Figure 5e–h) because the Al-rich melt can no longer be absorbed in the primary phase. However, since the nucleation of the peritectic Cr$_5$Al$_8$ phase was not considered, these results are not realistic in the present simulation. Hence, in the case of a nucleation event of the peritectic phase, the s/l interface would be planar. Therefore, it can be stated that investigations on different cooling rates and for a temperature gradient of $G_T = 6 \times 10^8$ K/m display a planar s/l interface within the l/Cr region.

The effect of the “moving frame” for $\dot{T} = 2.5 \times 10^6$ K/s and $G_T = 6 \times 10^8$ K/m (see Figure 5e) is visualized in Figure 6a,b. Due to the “moving_frame” option in MICRESS, the top of the simulation domain followed the s/l interface with a distance of 0.5 µm. The solidification length differs for each examination due to the selected parameters. Therefore, for a better display, only the final domains of the “moving_frame” option are shown in Figure 6. Figure 6c shows the corresponding temperature distribution over the entire solidification length.

Figure 6. (a) “moving_frame” domain and the (b) the full overview of the solidification length. (c) shows the corresponding temperature distribution ($\dot{T} = 2.5 \times 10^6$ K/s and $G_T = 6.0 \times 10^8$ K/m).
3.2. Predicted Microstructures for \( G_T = 6 \times 10^6 \text{ K/m} \)

Investigations were carried out by using the same cooling rates as in the in the previous section but the temperature gradient \( G_T \) was decreased to \( 6.0 \times 10^7 \text{ K/m} \) (Figure 7). All results showed cellular/dendritic solidification morphologies except for Figure 7h. There, the transition from a dendritic to a planar solidification morphology takes place for a cooling rate of \( \dot{T} = 1 \times 10^7 \text{ K/s} \).

![Figure 7](image-url)

**Figure 7.** Solidification morphology as a function of the selected cooling rates \( \dot{T} \) and a uniform temperature gradient of \( G_T = 6 \times 10^7 \text{ K/m} \). (a-g) show a dendritic/cellular solidification morphology. (h) transition from dendritic to planar growth. The images show a size of 2.5 µm × 2.5 µm.

3.3. Predicted Microstructures for \( G_T = 6 \times 10^6 \text{ K/m} \)

The effect of the cooling rate on the solidification pattern are displayed in Figure 8 for a fixed temperature gradient of \( G_T = 6 \times 10^6 \text{ K/m} \). By increasing the cooling rate from \( \dot{T} = 2.5 \times 10^5 \text{ K/s} \) to \( \dot{T} = 1 \times 10^7 \text{ K/s} \), the solidification morphology changes from a cellular/dendritic microstructure to a planar solidification front. Figure 8a shows a dendrite at the left side and a cellular solidification structure in the center and right side of the image. The cell tips seem to be growing at a lower temperature level compared to the dendritic tip. In fact, the cells have their origin from a side branch and grew rapidly upwards.

![Figure 8](image-url)

**Figure 8.** Solidification morphology for different cooling rates \( \dot{T} \) and a fixed temperature gradient of \( G_T = 6 \times 10^6 \text{ K/m} \). (a,b) show a dendritic/cellular solidification morphology. (c,d) transition from dendritic to planar growth. (e-h) planar solidification front. The images show a size of 2.5 µm × 2.5 µm.

The predefined simulation time was reached before the cell peaks achieved the same temperature level as the dendrite peak. The transformation from a dendritic solidification
structure to a planar one is clearly evident from Figure 8b–d. For cooling rates larger than \( \dot{T} = 1 \times 10^6 \text{ K/s} \), the s/l interface is planar. The images show a size of 2.5 \( \mu \text{m} \times 2.5 \mu \text{m} \).

4. Discussion

The numerical investigations evidence the possible occurrence of planar solidification fronts for temperature gradients in the range of \( 6 \times 10^6 \text{ K/m} \leq G_T \leq 6 \times 10^8 \text{ K/m} \). In between, cellular/dendritic solidification morphologies were obtained. In the following we gather and discuss the results in terms of (i) solidification length, (ii) solidification velocity, and (iii) secondary arm spacing.

(i) The solidification length represents the height of the solidified structure measured from the initial grain position to the top s/l interface position. It was determined and investigated for the different selected process conditions. Figure 9 shows the solidification distance for a solidification time of \( t_f = 1.1 \times 10^{-5} \text{ s} \). Note, the solidification time is significantly longer than the expected solidification time of \( 6.6 \times 10^{-6} \leq t_f \leq 1.6 \times 10^{-5} \text{ s} \). It can be seen that the minimum solidification length of 3 \( \mu \text{m} \) and a maximum solidification length of 6 \( \mu \text{m} \), measured in Figure 1b for the cross-section, was reached for cooling rates of \( \dot{T} > 1 \times 10^6 \text{ K/s} \) and temperature gradients \( G_T \leq 6 \times 10^7 \text{ K/m} \). The solidification time could not be determined experimentally. However, it can be stated with confidence that the observed structure (Figure 1b) could only result from growth occurring under a cooling rate \( \dot{T} < 6 \times 10^8 \text{ K/m} \).

![Solidification velocity in relation to the cooling rate and the temperature gradient.](image)

Figure 9. Solidification length obtained for a solidification time of \( t_f = 1.1 \times 10^{-5} \text{ s} \). The gray shaded box covers a solidification length of \( 3 \leq L \leq 6 \mu \text{m} \) measured in the experimental dendrite displayed in Figure 1b.

(ii) Regardless of the process parameters selected, the obtained solidification velocities (4.7 \( \times \) 10^5 \( \mu \text{m/s} \) for \( G_T = 6 \times 10^6 \) in Figure 10) were an order of magnitude lower than the absolute velocity \( V_a \geq 2.0 \times 10^6 \mu \text{m/s} \) (Equation (3)). Therefore, the observed dendritic solidification could not have happened under rapid solidification conditions [10]. In other words, the solidification phenomena in the present cathode spot crater is not concerned by solute trapping, which decreases solute partitioning and alters equilibrium solidification velocity-undercooling relationships.
(iii) The growth of secondary arms is triggered by perturbation of the s/l interface at the close vicinity to the dendrite tips. These arms undergo a process from cell-like to dendritic growth and some were eliminated due to competition with their neighbors. Later, a ripening process causes the secondary arms to change with time into coarser, less branched, and more widely-spaced ones. Furthermore, it is possible that the cooling rate during the numerical investigations to a planer front at steady-state conditions and the experimentally obtained dendrites were in a transition status. Due to their time-dependence, the dendritic side arm distances were evaluated at different times to determine an average secondary DAS (Figure 11). The regressing line suggests that a cooling rate of about $\dot{T} = 3 \times 10^5$ K/s for a temperature gradient $G_T = 6 \times 10^7$ K/m was necessary to obtain the secondary arm spacing of $\lambda_2 = 1.75 \times 10^{-7}$ m observed experimentally.

By comparing the prerequisites such as solidification length, the solidification speed, and the dendrite spacing, the present process conditions could be determined with relative confidence. It shows that dendrites with a secondary DAS of $\lambda_2 = 175$ nm seem to be formed by a cooling rate of $\dot{T} = 3 \times 10^5$ K/s and a temperature gradient $G_T = 6 \times 10^7$ K/m.
5. Summary and Conclusions

In a cathodic arc plasma, cathode spots are ignited all over the cathode’s surface. Consequently, the surface was re-melted and re-solidified due to a multitude of cathode spot events. This succession of local melting-solidification lead to the formation of several solidification layers, where dendritic solidification morphologies were clearly observed.

In this work, several numerical investigations were performed to estimate the thermal conditions for that different cooling rates and temperature gradients were investigated. The results exhibit that the melted layer at the cathode spot re-solidified dendritically under a cooling rate in the order of \( T = 3 \times 10^5 \) K/s and a temperature gradient of about \( G_T = 6 \times 10^7 \) K/m.

The importance of this work is that our thermal results provide excellent data for the validation of numerical models. Such numerical models were recently used to describe the formation and development of a cathode spot on different alloys in a vacuum arc [13–17]. These models include hydrodynamic equations and the heat transfer equation.

Author Contributions: Conceptualization, J.P.M.; methodology, J.P.M. and A.K.; software, J.P.M.; validation, A.K., R.F. and C.M.; formal analysis, A.K.; experimental investigation, M.G.; writing—original draft preparation, J.P.M.; writing—review and editing, J.P.M., R.F., M.G., C.M. and A.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors thank Bernd Böttger, member of the MICRESS team, for his support and advice. Abdellah Kharicha acknowledges financial support from the Austrian Federal Ministry of Economy, Family and Youth and the National Foundation for Research, Technology and Development within the framework of the Christian-Doppler Laboratory for Metallurgical Applications of Magnetohydrodynamics. Robert Franz acknowledges the financial support by the Österreichische Forschungsförderungsgesellschaft mbH (FFG) within the framework of the project “ArcCathodeErosion” (Project No. 856889).

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Anders, A. Cathodic Arcs: From Fractal Spots to Energetic Condensation; Springer: New York, NY, USA, 2008.
2. Mitterer, C. PVD and CVD Hard Coatings, in Comprehensive Hard Materials; Sarin, V.K., Llanes, L., Mari, D., Eds.; Elsevier: Amsterdam, The Netherlands, 2014; Volume 2, pp. 449–467. [CrossRef]
3. Franz, R. Insights into surface modification and erosion of multi-element arc cathodes using a novel multilayer cathode design. J. Appl. Phys. 2020, 127, 113301. [CrossRef]
4. Pohler, M.; Franz, R.; Ramm, J.; Polcik, P.; Mitterer, C. Cathodic arc deposition of (Al, Cr)2O3: Macroparticles and cathode surface modifications. Surf. Coat. Technol. 2011, 206, 1454–1460. [CrossRef]
5. Franz, R.; Mendez Martin, F.; Polcik, P. Erosion behavior of composite Al-Cr cathodes in cathodic arc plasmas in inert and reactive atmospheres. J. Vac. Sci. Technol. A 2016, 34, 021304. [CrossRef]
6. Golizadeh, M.; Mendez Martin, F.; Wuster, S.; Mogetisch, J.P.; Kharicha, A.; Kolozsvari, S.; Mitterer, C.; Franz, R. Rapid solidification and metastable formation during surface modification of composite Al-Cr cathodes exposed to cathodic arc plasma. J. Mater. Sci. Technol. 2021, 94, 147–163. [CrossRef]
7. Available Online: The Microstructure Evolution Simulation Software. Available online: https://micress.rwth-aachen.de (accessed on 17 October 2022).
8. Available Online: Generate Materials Data with Thermo-Calc. Available online: https://thermocalc.com/ (accessed on 17 October 2022).
9. Feurer, U.; Wunderlin, R. DGM Fachber; Wunderlin: Frankfurt, Germany, 1977; Volume 38.
10. Kurz, W.; Fischer, D.J. Fundamentals of Solidification; Trans Tech Publications Ltd.: Wollerau, Switzerland, 1998.
11. European Commission and Technical Research. Definition of Thermochemical and Thermo-Physical Properties to Provide a Database for the Development of New Light Alloys; COST European Cooperation in the Field of Scientific: Brussels, Belgium, 1998; Volume 2.
12. Salas, G.B.; Ramirez, J.V.; Noguez, M.E.A.; Robert, T.N. Dendrite arm spacing—local solidification time relationship: An experimental model for a 70-30 brass and comparison with some theoretical models. Scr. Metall. Mater. 1995, 32, 295–299. [CrossRef]

13. Lijun, W.; Xiao, Z.; Jiagang, L.; Ming, L.; Shenli, J. Study of cathode-spot crater and droplet formation in a vacuum arc. J. Phys. D Appl. Phys. 2021, 54, 215202.

14. Santos, D.F.N.; Lisnyak, M.; Almeida, N.A.; Benilova, L.G.; Benilov, M.S. Numerical investigation of AC arc ignition on cold electrodes in atmospheric-pressure argon. J. Phys. D Appl. Phys. 2021, 54, 195202. [CrossRef]

15. Huo, J.; Ronzello, J.A.; Rontey, A.; Wang, Y.; Jacobs, L.; Sommerer, T.; Cao, Y. Development of an arc root model for studying the electrode vaporization and its influence on arc dynamics. AIP Adv. 2020, 10, 085324. [CrossRef]

16. Karimi-Sibaki, E.; Kharicha, A.; Wu, M.; Ludwig, A.; Bohacek, J. A Parametric Study of the Vacuum Arc Remelting (VAR) Process: Effects of Arc Radius, Side-Arcing, and Gas Cooling. Metall. Mater. Trans. B 2020, 51, 222–235. [CrossRef]

17. Kaufmann, H.T.C.; Silva, C.; Benilov, M.S. Numerical simulation of the initial stage of unipolar arcing in fusion-relevant conditions. Plasma Phys. Control. Fusion 2019, 61, 095001. [CrossRef]