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Analysis of on-deposition solidification during the Osprey spray-deposition process

Z. FANG
Rogers Tool Works, Rogers, AR 72757, USA
J. DUSZCZYK
Delft University of Technology, Delft, The Netherlands
A. LEATHAM
Osprey Metals Ltd, Neath, West Glamorgan, UK

The on-deposition process during spray deposition was studied. The theory of forced convection in the case of laminar flow is applied to evaluate the heat-transfer coefficient between the atomizing gas and the semi-solid/semi-liquid thin film on the surface of the deposit. The cooling rate of the on-deposition process was estimated, based on the heat-flow analysis. The results agree well with experimental observations of the microstructure of spray-deposited aluminium alloy, as well as other similar rapid solidification processes, such as gas atomization of metal powders.

1. Introduction

The objectives of rapid solidification processing are to obtain refined microstructure, more homogeneous materials and new phase compositions in alloys. As opposed to the various well developed rapid solidification techniques, the spray-deposition process is still in its embryonic stage. There is still a considerable lack of study on the solidification characteristics of the spray-deposition process with respect to the cooling rate, microstructure, etc. Although there have been some fundamental works contributed to the prediction of particle velocities and temperatures in flight during the spray-deposition process, few data, either theoretical or experimental, are available for the on-deposition solidification process. The present research was devoted to the detailed study of the solidification problems involved in the spray-deposition process by heat-flow analysis and microstructural investigations.

As Leatham *et al.* [1] and Apelian *et al.* [2] have described, among the many contributing factors to the final microstructure of spray-deposited materials are particle velocities, particle (droplet) temperature profiles and droplet/substrate interactions. The in-flight solidification is no different from conventional gas atomization. The subsequent cooling process could influence the scale of dendrites and the formation of second phases. The on-deposition process is of particular interest in our research. The on-deposition process is crucial because it strongly affects the final solidified microstructure. The local solidification time of the on-deposition solidification is estimated based on the heat-flow analysis. Details of as-solidified microstructure are reported.

2. Experimental procedure

Spray-deposited 7075 aluminium alloys produced by Osprey Metals Ltd were investigated in the research. The arrangement of the Osprey spray process is shown in the literature [1, 2]. Argon was used as atomizing gas. The transmission electron microscope was applied to reveal the details of the spray-deposited microstructure. Optical microscopy was used in conjunction with TEM observations for a complete and correct understanding of the overall microstructure.

3. Heat transfer and solidification

It is generally agreed [2–4] that upon the instant of impingement, the individual metal spray particles can be in any of three conditions: molten droplet, semi-liquid/semi-solid state, or fully solidified. The majority of the particles will be in a semi-liquid/semi-solid state. Partially solidified droplets fragment on impacting the deposit surface along the interdendritic regions, but retain their general shape. During the spray-deposition process it could be managed that a thin film of liquid phase will be retained on the deposit surface, and atomized droplets/particles arrive at the deposit surface successively. The on-deposition solidification proceeds step-by-step, as shown schematically in Fig. 1. It can be seen that the process goes on from State a–b and b–a′. Since States a and a′ are identical except for the increment of the deposit thickness, the on-deposition solidification process can be considered to proceed from a–a′ as one cycle, and it is by iterating this cycle that the solidification of the preform is carried on. It is therefore reasonable to consider the heat-extraction problem to be in a quasi-steady state, assuming a constant supply of droplets/particles to the deposit and a constant heat extraction rate from the thin film. These are valid assumptions except the initial and final transients of the deposition process.

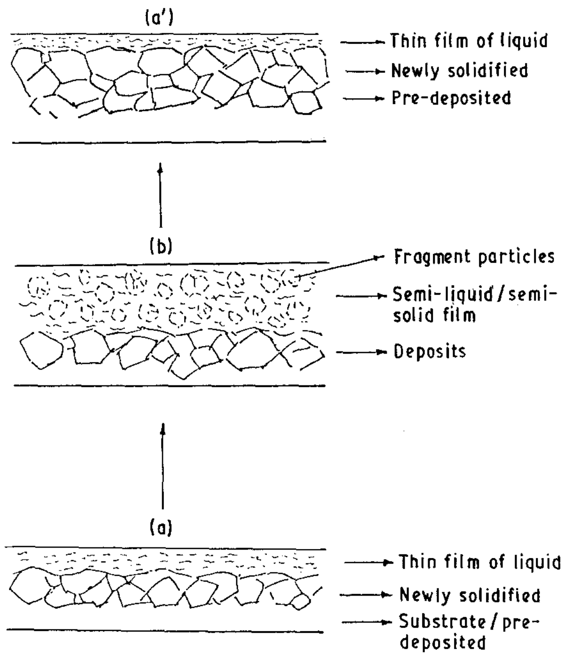

Figure 1 Schematic illustration of the on-deposition solidification process.

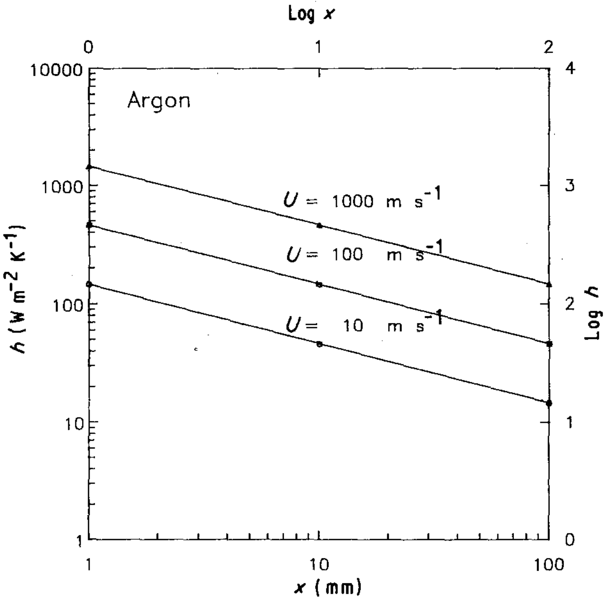

Figure 2 Gas–film (semi-liquid/semi-solid) interfacial heat-transfer coefficient as a function of the preform lateral dimension, for aluminium, at different gas velocities.

Another assumption to be made at this stage is that the internal heat flow is sufficiently rapid for the concurrent thin film to be always instantaneously isothermal, and hence the cooling of the thin film to be Newtonian for the range of the case being studied here.

Heat extraction from the semi-liquid/semi-solid film will be performed by two ways: conduction to the deposit/substrate, and loss to the fast moving atomizing gas. The conduction of heat to the substrate could be dominant at the initial transient of the deposition, but as the thickness of the deposit increases the cooling process has to rely mainly on the heat loss to the surroundings by forced convection of atomizing gas flowing over the deposit surface.

The effectiveness of this heat extraction is characterized by a heat-transfer coefficient between the atomizing gas and the thin film. By applying the approaches dealing with the forced convection heat-transfer problem of a flat plate, over which a fluid is flowing, it is simply shown that [5]

$$h = (k/x)\mathrm{Nu} \qquad (1)$$

where h is the heat-transfer coefficient, k is thermal conductivity, x denotes the lateral dimension along the thin film, and Nu is given by

$$\mathrm{Nu} = 0.33\,\mathrm{Re}^{1/2}\,\mathrm{Pr}^{1/3} \qquad (2)$$

Nu, Re, and Pr are Nusselt number, Reynolds number and Prandtl number, respectively, and are given as

$$\mathrm{Nu} = (h/k)x \qquad (3)$$

$$\mathrm{Re} = (U\rho/\mu)x \qquad (4)$$

$$\mathrm{Pr} = c\mu/k \qquad (5)$$

Equations 1–5 lead to a simple way of evaluating the heat-transfer coefficient by one equation ($\mathrm{Pr} = 1$ for gases [5])

$$h = 0.33\,k\,(\rho/\mu)^{1/2}\,U^{1/2}(1/x^{1/2}) \qquad (6)$$

where c, μ and ρ are specific heat, viscosity and density, respectively, all of which refer to the atomizing gas. U is the velocity of the atomizing gas. The result of the calculation of the heat-transfer coefficient using Equation 6 in the range of interest for the Osprey process, is given in Fig. 2.

It is implied, in applying the theory of laminar flow to the problem of heat extraction from the thin film by an atomizing gas, that there is no lateral flow of the film. This is a reasonable assumption because it is this unique point that distinguishes the spray-deposition process from conventional casting.

It is not surprising to find from the result that the velocity of the atomizing gas has a crucial influence on heat loss from the thin film.

In the case of Newtonian cooling conditions, the local solidification time can be estimated according to the following expression (via a known value of h)

$$t_f = \Delta H/(h\Delta T_c A) \qquad (7)$$

where A is the surface area, $\Delta T_c = T_f - T_g$, where T_f is solidification temperature, and T_g is the temperature of the atomizing gas.

$$\Delta H = V(C_f\Delta T_s + \Delta H_f) \qquad (8)$$

where V is the volume of the semi-liquid/semi-solid film, C_f is the specific heat, ΔH_f is the latent heat of solidification, and $\Delta T_s = T - T_f$, if the droplets/particles are deposited at T, and the solidification temperature is T_f. Suppose the film thickness is d, area A, and the fraction of solidified material is g, then

$$\Delta H = Ad[C_f(T - T_f) + \Delta H_f](1 - g) \qquad (9)$$

and then

$$t_f = d(1 - g)(C_f\Delta T_s + \Delta H_f)/h\Delta T_c \qquad (10)$$

To evaluate t_f, the parameters involved in Equation 10 are taken from Singer and Evans [3], for aluminium alloys; $T = 750\,^\circ\mathrm{C}$, $T_f = 650\,^\circ\mathrm{C}$,

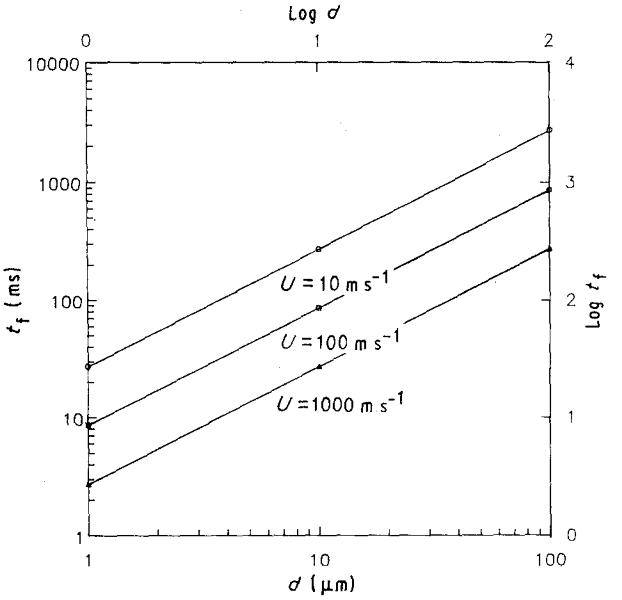

Figure 3 Predicted local on-deposition solidification time, as a function of the film thickness, at different atomizing gas velocities, for aluminium. $\Delta T_c = 100$ K, $\Delta T_f = 600$ K, $g = 0.6$.

$C_f = 945\ \mathrm{J\,kg\,K^{-1}}$, $\Delta H_f = 390.6\ \mathrm{kJ\,kg^{-1}}$, $\Delta T_c = T_f - T_g$ will be approximately 600 K. The fraction of solid phase, g, is dependent on the deposition rate. The results predicted by this equation are shown in Figs 3 and 4 for film thickness from 1–100 µm, and solid phase fractions from 0.3–0.9 at different velocities of atomizing gas.

It is well known that the rapidly solidified microstructure can be obtained for a sufficiently high solidification rate. The local solidification time is the time required for the solidification of the semi-liquid/semi-solid film, and is representative of the solidification rate. But on the other hand, the cooling rate has been widely used as a descriptive parameter for rapid solidification, because it can be estimated easily from the scale of the microstructure. However, at higher cooling rates of heat extraction, the arrest and recalescence associated with the latent heat evolution make the ascribing of a unique cooling rate unrealistic when compared with the actual thermal history within the melt [6].

The relationship between cooling rate and local solidification time is given generally by

$$R = \Delta T / t_f \qquad (11)$$

where ΔT is the temperature difference between the temperature at which the droplets/particles impact the deposit and that at which the solidification occurs. Apparently there will be a considerable uncertainty when one attempts to determine it. From Equation 11 the cooling rate during the on-deposition solidification process can be estimated roughly to be in the range 10^3–10^5 K s^{-1}. This agrees well with the other rapid solidification techniques, for instance, the production of metal powders by gas atomization. The results imply that rapidly solidified microstructure can be obtained by this process.

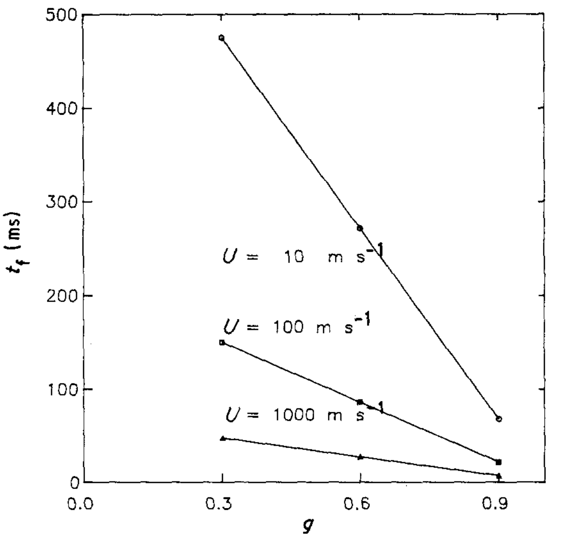

Figure 4 Predicted local on-deposition solidification time, as a function of the fraction of the solid phase, at different atomizing gas velocities, for aluminium. $\Delta T_c = 100$ K, $\Delta T_f = 600$ K, $d = 10$ µm.

4. Microstructure

It has been stated that the spray-deposition process can be summarized as two different stages of solidification and a cooling process: in-flight solidification, on-deposition solidification and the subsequent cooling after solidification. The final microstructure of the materials produced by the Osprey process is the result of the entire process. The first unique feature of the spray-deposited microstructure is the high level of homogeneity of grain structures, as shown in Fig. 5 for 7075 aluminium alloy. This type of uniform grain structure exists throughtout the spray-deposited preform. The grain boundary in this case has a certain width, in accordance with the deposition rate, and is composed of the boundary band and the second phase particles distributed along the boundary within the boundary band. Fig. 6 shows the structure of the boundaries. A solute-poor zone encompassing the second-phase particle can be recognized easily.

Another very important unique feature of the spray-deposited microstructure is noted: it is composed of different solidified microstructures which should have resulted from different cooling rates. Fig. 7 shows the rapidly solidified cellular/dendritic structure with very

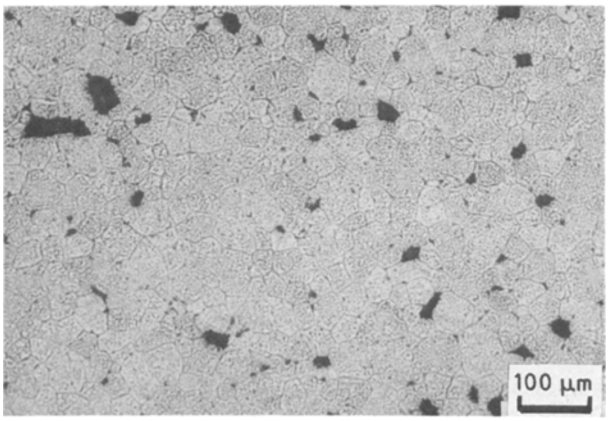

Figure 5 Grain structure of the spray-deposited 7075 aluminium alloy produced by the Osprey process.

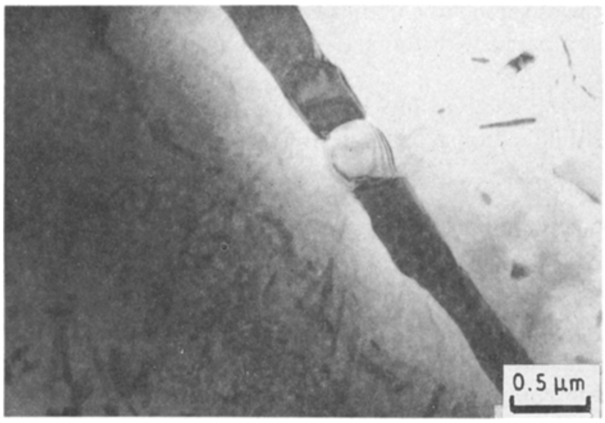

Figure 6 Grain-boundary morphology of the spray-deposited 7075 aluminium alloy produced by the Osprey process.

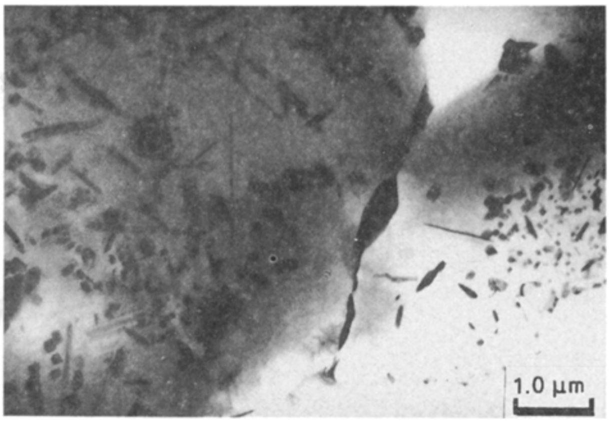

Figure 8 Uniform distribution of second-phase particles in spray-deposited 7075 aluminium alloy produced by the Osprey process.

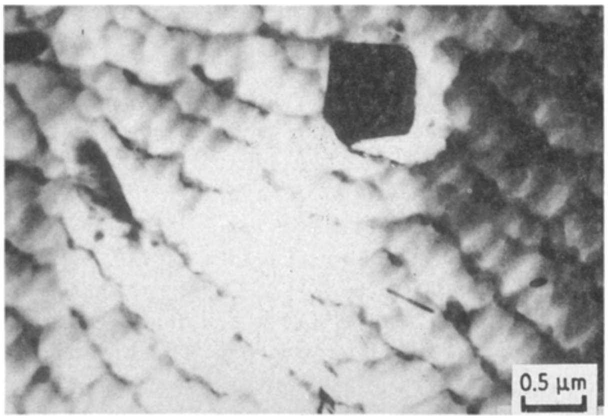

Figure 7 Cellular/dendritic structure of the spray-deposited 7075 aluminium alloy produced by the Osprey process.

fine inter-cellular microsegregation. Fig. 8 shows those relatively large grains with uniformly distributed second-phase particles. For the former the cooling rate is high, but for the latter it is lower. This is some kind of local inhomogeneity within the range of grain sizes, as observed by TEM, similar to the inhomogeneity in individual particles for gas-atomized metal powders. This kind of local inhomogeneity of the structure might be because of the multi-stages of solidification and the inhomogeneous distribution of the melt among the solidified (in-flight) particles within the semi-liquid/semi-solid thin film during the process of on-deposition solidification. Nevertheless, the cellular/dendritic structure presented in Fig. 7 is a clear evidence, suggesting that a cooling rate of 10^4–10^5 K s^{-1} for that local region, if it is assumed that the widely applied method of estimating cooling rate by cell size is valid in this case [7].

5. Conclusion

The spray-deposition process is composed of different stages of solidification and a cooling process. The on-deposition solidification process was studied in our research. The theory of forced convection in the case of laminar flow was applied to evaluate the heat-transfer coefficient between the atomizing gas and the semi-liquid/semi-solid thin film on the surface of the deposit. The local solidification time for the on-deposition solidification process was estimated based on the heat-flow analysis. The results obtained agree well with the other rapid solidification processes, such as metal powders produced by gas atomization. The model suggested in the research can be used as an effective way of evaluating the on-deposition solidification process. The microstructural investigations proved that the Osprey process can produce rapidly solidified microstructure predicted by the theoretical analysis.

Acknowledgements

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