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Solid freeform fabrication by extrusion and deposition of semi-solid alloys

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Solid Freeform Fabrication (SFF) refers to techniques that create prototypes by a layer wise deposition of material. There are several techniques available, none of which allows the production of metallic prototypes without post processing, such as debinding or sintering. One of the SFF techniques, Fused Deposition Modeling (FDM®), is a well established process for thermoplastic materials such as for example ABS. Based on the FDM® technique, a process is being developed that allows the extrusion and deposition of semi-solid metals (EDSSM). The microstructure of an alloy in the semi-solid state has been investigated as a function of parameters used for rapid prototyping with SFF techniques. The extrusion and deposition processes are dependent on the rheological properties of the semi-solid metal, which in turn are dependent on the microstructure. The effect of microstructure and rheological properties on the extrusion and deposition processes is discussed.

1. Introduction

This paper summarises research on semi-solid metals currently conducted at TNO Industrial Technology of the Netherlands. The goal of the project is to investigate the feasibility of using metals in a new SFF process based on the FDM technique. The currently available FDM systems allow the production of prototypes from thermoplastic materials. Using metals is thought to broaden the capabilities for product design and development significantly as well as there is a potential for the Rapid Fabrication (RF) of single or a small series of products.

Although near net shape processing using technologies involving semi-solid metals processes (SSMP) are being given increasing attention, they are not widely spread in the SFF sector yet. During SSMP a melt is permitted to partially solidify before shaping operations [1–3]. Alternatively a solid alloy can be partially re-melted. The solid fraction may vary between 0.2 and 0.9 and the constitutive behaviour of the resulting slurry strongly depends on the degree of solidification [1]. In the EDSSM process, a wire of the desired material is being brought into the semi-solid state by partial re-melting and is extruded onto a target through a nozzle with a typical diameter of several hundreds of micrometers. The size of the nozzle diameter determines the accuracy of the final part. Semi-solid metals were selected since their unique rheological properties enable a controllable extrusion process. Flemings [4] pioneered the research on semi-solid metals and found that mechanical shearing of metal alloys during solidification results in a break up of dendrites and thus in a globular structure. In the current case the material is being partially re-melted and extruded, thus the behaviour of the semi-solid metal is dependent on two primary parameters. These are:

- the morphology of the solid phase that is determined by the thermo-mechanical history of the material before semi-solid extrusion
- the solid fraction during the extrusion that is determined by the temperature

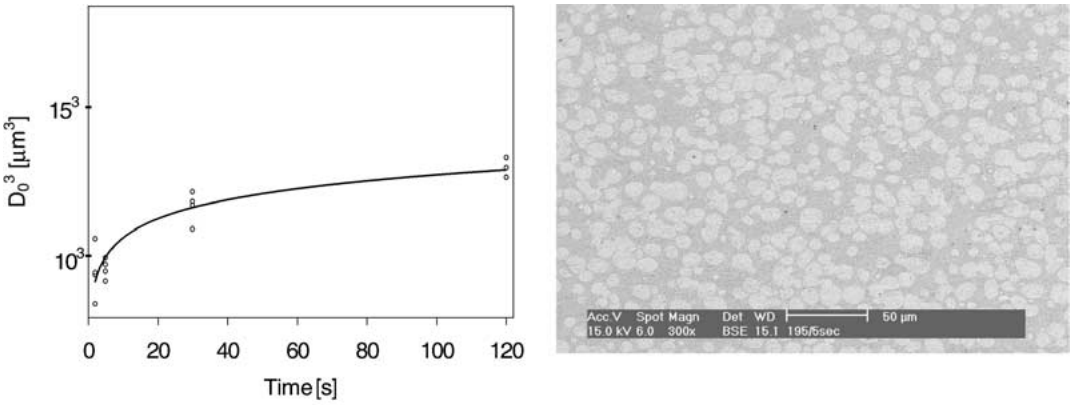
2. Microstructural characterisation

In order to describe the morphology of a semi-solid alloy, coarsening effects of the microstructure, such as Ostwald Ripening, coalescence as well as agglomeration and particle shape have to be considered. In general, the coarsening behaviour can be written as:

$$\bar{D}^n - \bar{D}_0^n = Kt \qquad (1)$$

where $\bar{D}$ is the average particle diameter, $\bar{D}_0$ is the initial average particle diameter, K a rate constant and t the time. The initially formed globular structure does not represent thermodynamic equilibrium, due to a large surface area [5]. The two-phase system containing a liquid and a solid phase, is decreasing its total energy by increasing the size of the second phase and thus by decreasing the interfaces. This effect is known as Ostwald Ripening and is a purely diffusion driven process [5].

The coarsening behaviour of a semi-solid Pb-40%Sn alloy was studied simulating EDSSM process conditions [6]. It was found that partially re-melting resulted in a globular non-dendritic structure with relatively



(a) (b)

Figure 1 (a) Increase of the average diameter, D, of solid phase of a Pb-Sn alloy vs holding time for short times, up to 120 s and (b) typical microstructure after partially re-melting at 195°C, isothermal holding for 5 seconds followed by quenching in liquid nitrogen.

small globule sizes in the range of 10 μm. The tests carried out indicated that coarsening can be described by Ostwald ripening (except for very short holding times in the range of seconds), but that sedimentation effects may become important at longer holding times. Typical isothermal holding times during EDSSM processing are in the range of 1–120 seconds. The increase of the average diameter of the solid particles for holding times up to 120 seconds is shown in Fig. 1a and b shows a typical microstructure. Although an increase of the average diameter occurs, the coarsening is not critical to the process. However, it should be kept in mind that there is no immediate industrial application for low melting point alloy and that engineering alloys, such as Al alloys are characterised by coarser solid particles in partially re-melted materials.

3. Rheological characterisation

Back extrusion experiments were conducted to obtain information about the viscosity of the partially re-melted alloy as a function of solid fraction and shear rate. Fig. 2 shows the experimental set-up schematically. Sample material is partially re-melted at the bottom of the cylindrical chamber. The piston is moved downwards at a constant velocity whereby the semi-solid material is forced up through the gap between the piston and the cylinder. A detailed description of the experimental is given in [6]. The force was plotted as a function of time and a typical curve is shown in Fig. 3. From the linear part the viscosity was calculated and was found to be in the order of 1 Pa s.

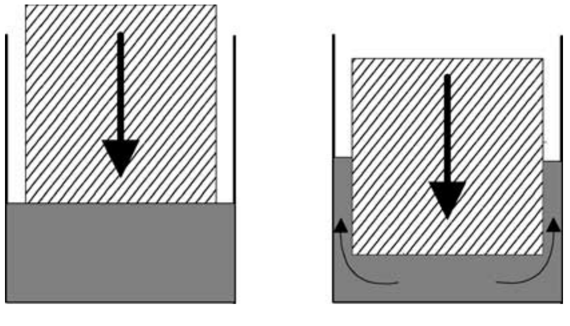


Figure 2 Schematic of the back extrusion principle.

Back extrusion experiments have shown that a homogenous flow of the material (no segregation) is required in order to obtain reliable viscosity data for a given solid fraction. When segregation between the liquid and solid phases occurred, the apparent viscosity was found to be two orders of magnitude higher than the apparent viscosity determined from tests under homogeneous conditions. There is evidence that the piston speed [7] and thus the shear rates have an influence on segregation effects. In addition it was observed that segregation is dependent on whether or not the piston and the re-melted material are in contact at the onset of the extrusion. High solid fraction and non-globular morphology of the globules are also factors that contribute to the occurrence of segregation.

Segregation effects during forming processes of semi-solid metals are a well-known phenomenon. During semi-solid forming the mixture of solid and liquid phases are submitted to strains, of both of thermal and mechanical origin [8]. It was proposed that the results from back extrusion experiments can be used to model the force F_i necessary to expel the liquid from the solid. Seconde [9] proposed that at the on-set of the test the semi-solid alloy can be considered as a specimen subjected to compression by parallel plates and that F_i can be calculated as

$$F_i = \frac{21.25\pi\eta(1-f_l)^2 R_e^4}{hD^2 f_l^3}V + \frac{0.0875\pi\rho_l(1-f_l)R_e^5}{h^2 D f_l^3}V^2 \tag{2}$$

where η is the viscosity of the liquid phase, R_e the radius of the specimen, h is the specimen height, D the globule diameter and ρ_l the density of the liquid. The first term of Equation 2 represents the contribution of the laminar flow and the second term that of the turbulent flow, which becomes dominant at higher velocities. Considerable theoretical and experimental work on modelling segregation phenomena is available in the literature. However, most authors address the high solid fraction domain, since most semi-solid metal forming processes (continuous die casting, forging) are carried out at these solid fractions. The main assumption for the models is that a solid skeleton exists which means that the solid phase is fully connected and thus each solid

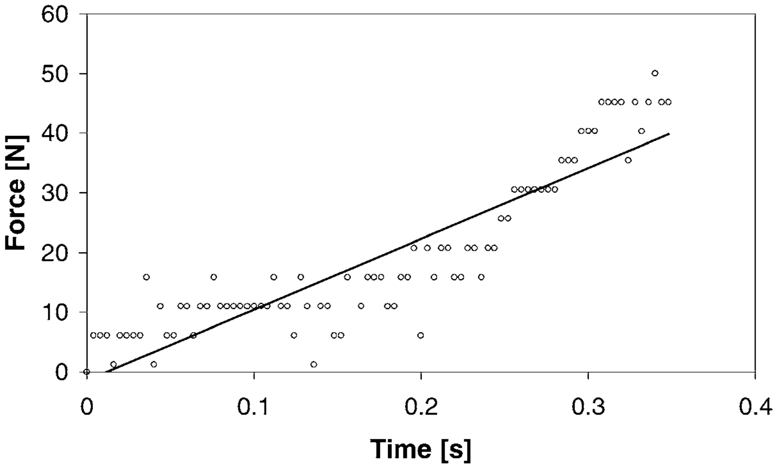


Figure 3 Graph of the force vs time obtained from the back extrusion experiments.

point can be linked to another solid point though a path entirely located in the solid phase [8]. The solid volume fraction above which a solid skeleton can be assumed, depends on the morphology of the semi-solid as well as on the thermo-mechanical history of the material.

The solid fractions in the current case are in the range of 0.3–0.4 and a solid skeleton can not be assumed. In contrast to a semi-solid that consists of a porous skeleton saturated with liquid, the main mechanism for energy dissipation is not the viscoplastic deformation of the solid, but a combination of viscous friction between the solid particles and a small amount of deformation of the particles. However, since the model is phenomenological it is being used in ongoing work in order to better understand the occurrence of segregation and its influencing factors.

4. Analytical and experimental determination of process parameters

One of the most important parameters for the control of the extrusion process is the temperature, since it controls the volume solid fraction and viscosity and thus the extrusion properties of the semi-solid alloy. Segregation effects and extrusion force for example are related to the solid fraction. This section describes the ongoing work on determination of extrusion and deposition parameters, such as filament temperature, extrusion force and deposition rate.

4.1. Filament temperature

The filament temperature estimation was carried out by using Equations 3–6 [10]. In addition to the analytical determination, filament temperatures were measured at the outlet using a J-type thermocouple. The thermocouple controlling the heating process was mounted in the wall of the heating channel (see Fig. 4).

$$q = \frac{T_1 - T_0}{R} \tag{3}$$

where T_1 is the temperature inside the wall of the heating channel, T_0 is the temperature measured by the thermocouple and R is the thermal resistance, that can be expressed as:

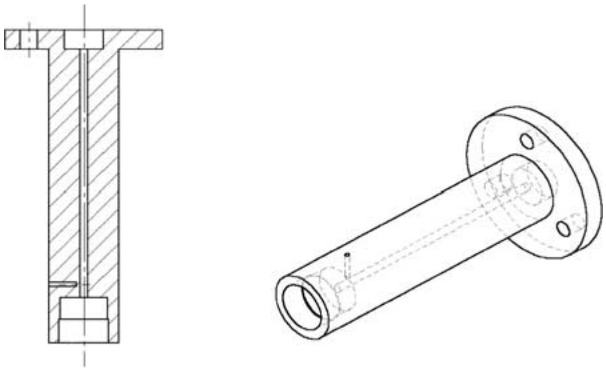


Figure 4 Heating channel cross section (left) and isometric-view (right).

$$R = \frac{\ln(r_2/r_1)}{2\pi K_a l} \tag{4}$$

where r_2 and r_1 are the inner and outer radius of the heating channel respectively, K_a is the thermal conductivity and l the length of the cylinder. The following assumptions are made: steady-state conditions, one dimensional heat transfer in the radial direction, constant properties, negligible radiation exchange.

The thermal resistance for a small control volume x can be written as:

$$R = \frac{\ln(r_1/r_0)}{2\pi K_{\text{all}} x} \tag{5}$$

where r_0 and r_1 are the inner and outer radius of the wire, K_{all} is the thermal conductivity of the wire material and x is the position of a control volume. The temperature of the alloy at the end of the cylinder can be expressed as:

$$\frac{(T_1 - T_2)}{(T_1 - T_i)} = \exp(-R\rho C_p)t \tag{6}$$

Finite element modelling of the filament temperature was carried out using the programme 'FLUENT'. Fig. 5 summarises the calculated values for three different wire feed velocities. The experimental values for the temperature measured at the nozzle tip are included. The graph shows that the desired temperature can be obtained for the three different wire velocities tested.

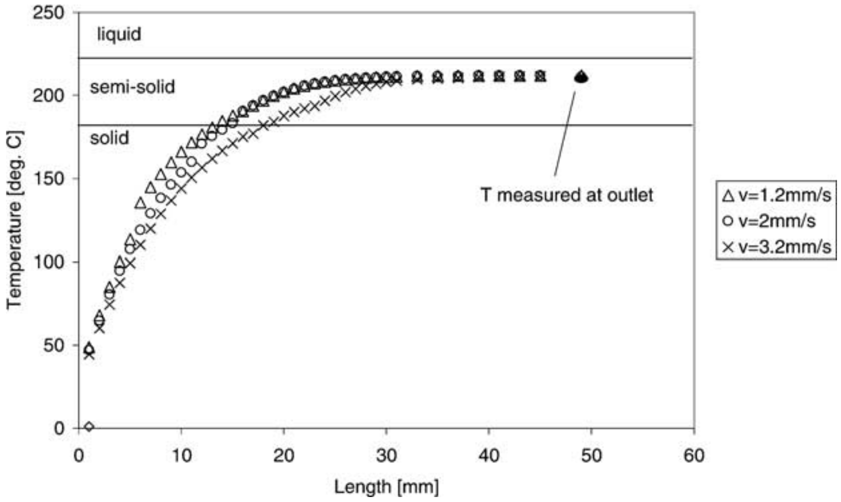


Figure 5 Temperature vs length (position within heating channel) as a function of different wire feed velocities.

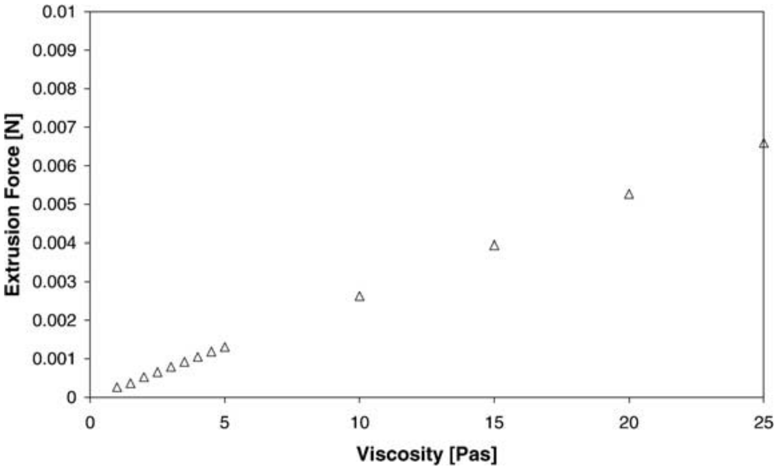


Figure 6 Calculated values for the extrusion force vs viscosity of the semi-solid alloy.

4.2. Extrusion force

For simple shear the deformation rate can be calculated as

$$\dot{\varepsilon} = \frac{dV}{dx} \tag{7}$$

For the geometry given in the EDSSM process the deformation rate can be written as:

$$\dot{\varepsilon} = \frac{V_0\left(\left(\frac{R_0}{R_1}\right)^2 - 1\right)\tan\alpha}{(R_0 - R_1)} \tag{8}$$

where V_0 is the velocity of the wire, R_0 and R_1 the radii before and after extrusion and α the die angle.

The shear stress τ_s can be calculated using

$$\tau_s = \eta\dot{\varepsilon} \tag{9}$$

where η is the viscosity. Finally the extrusion force can be determined using Equation 10.

$$F_{ex} = \tau_s A \tag{10}$$

where A is the cross sectional area.

The calculated values for the extrusion force versus viscosity are shown in Fig. 6. The graph shows the linear dependence of the extrusion force from the viscosity for practicable viscosities. It was found, during rheological measurements, that the viscosity can vary depending on whether or not segregation of the liquid and solid phase occurs. If segregation occurs the viscosity was found to be one order of magnitude higher that in case of homogeneous flow.

The data are theoretical values, because friction is not considered. As the wire is heated to the semi-solid state it looses its shape as soon as it reaches the eutectic temperature. At this point the semi-solid metal touches the inner wall of the heating channel and friction occurs. The influence of friction and other factors are being further studied.

4.3. Relationship between extrusion velocity and *x–y* velocity of the nozzle

The temperature of the layer material at the building surface is the most important parameter for good adhesion

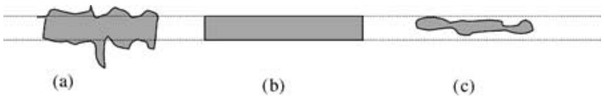
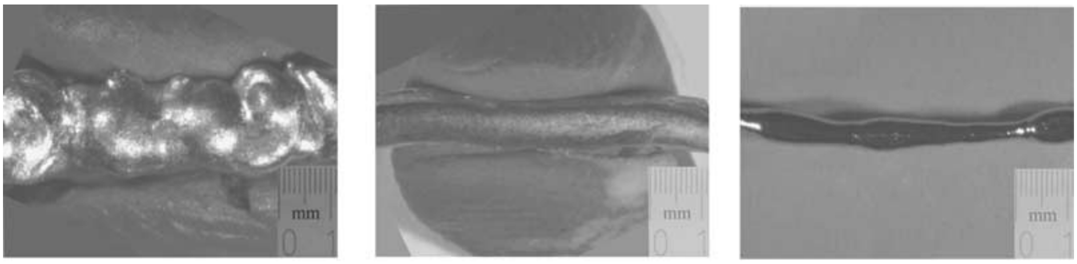


Figure 7 Illustration of the extruded path for different ratios between extrusion velocity and *x–y* nozzle velocity, with (a) a too large ratio, (b) the correct ratio and (c) a too small ratio; Dashed lines show the path desired.



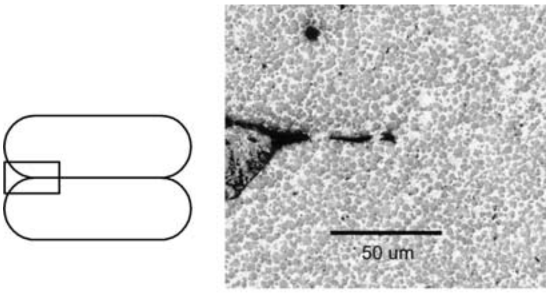
(a) 2:1 (b) 1:1 (c) 1:2

Figure 8 Examples of extruded materials at different extrusion velocity x–y velocity ratios.

between layers. On deposition the semi-solid material rapidly solidifies due to the contact with either the substrate or previously deposited material and through heat absorption by the surrounding. An analysis of the solidification process helps to determine process settings, such as the ratio between extrusion velocity and x–y velocity. Fig. 7 shows schematically that the correct ratio between these two parameters is critical because a too large ratio may cause failures in controlling the materials flow along the path desired and a too small ratio may cause discontinuities of the path.

Experiments were carried out to obtain information about the optimal ratio between the extrusion velocity and the x–y velocity of the nozzle. Optical examination was carried out on the extruded material. Fig. 8 contains examples of extruded material. Fig. 8a shows a path extruded with a ratio (extrusion velocity to x–y velocity) of 2 : 1. The material in Fig. 8b and c was extruded with ratios of 1 : 1 and 1 : 2 respectively. The photographs reveal that the material extruded with a ratio of 1 : 1 showed the most desirable results with respect to constant wire diameters. A quasi-constant wire diameter is crucial for the layer wise material built up as well as for a good layer bonding and accuracy aspects.

The adhesion between two layers depends on the temperature of the semi-solid layer being extruded and that of the previously deposited layer. In order to achieve good bonding, the interface temperature must fall into the semi-solid range. An analysis of the heat transfer during deposition is currently being carried out in order to analytically determine optimal deposition conditions. Initial experiments showed that good interlayer bonding can be achieved (see Fig. 9). Fig. 9a shows a schematic of a cross section of two layers. The edge, where the two layers meet is visible, but towards the cross section the microstructure does not change at the interface. An optimisation of the deposition parameters is needed to further decrease the effects at the edges.



(a) (b)

Figure 9 (a) Cross section of two layers indicating the area from which the micrograph was taken, (b) micrograph showing an interlayer region (the temperature of previously deposited material was approximately 150°C).

5. Conclusions

It has been shown that layers of metal can be deposited layer by layer by extrusion of semi-solid low melting point alloys. Materials in the semi-solid state respond to temperature variations with significant changes in their rheological behaviour, thus a rather accurate temperature control is required. Isothermal holding of semi-solid metals leads to a coarsening of the structure. Depending on the alloy this may conflict with the geometrical dimensions of the nozzle, e.g., when the extrusion process is stopped for longer times (in the range of minutes).

Segregation from the liquid and solid phase was observed during back extrusion experiments and the occurrence was related to the shear rate. The role of segregation may be described with phenomenological models that allow a comparison of the forces necessary to expel the liquid from the solid to that to homogeneously deform the alloy.

The heat transfer into the filament was determined analytically. It was found that for practicable wire feed velocities the desired temperature and thus solid fraction can be reached. Experimental results verified the analytical data just before the actual extrusion operation takes place.

An attempt was made to model the extrusion forces as a function of viscosity and nozzle geometry. Since friction was not considered in the model it can only be seen as an approximation. A more sophisticated model is needed to calculate the extrusion forces more accurately.

It was shown that the ratio between extrusion velocity and x–y movement of the nozzle is a critical parameter for geometrical accuracy and interlayer bonding. A good interlayer bonding was achieved, apart from the edges where the two layers meet. Experimental and analytical work is further being conducted to improve the interlayer bonding.

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