Abstract
Selective Laser Melting (SLM) is an additive manufacturing technology that offers significant potential for lightweight applications in space, aerospace, and automotive industries as well as in mechanical engineering. Structural aluminium alloys are therefore of special interest. Scalmalloy® is a scandium-modified Al-Mg alloy which displays exceptional properties when processed by SLM. These properties are predominately related to a generally very fine grained microstructure. However, the fine grained microstructure interspersed with coarser grained regions. Microstructural analyses indicate that the temperature regime and the particle precipitation behaviour are responsible for the duplex grain structures. In melt pool areas close to the pool base, numerous Al_3(Sc, Zr), Al-Mg-oxides and mixed particles act as nuclei for Al matrix solidification, leading to the formation of a very fine grained microstructure. In hot melt pool areas with T>800°C the majority of particles dissolve and growth of coarse columnar grains takes place. Better understanding of the formation mechanisms for these two distinct different structures will help pave the way towards newly designed alloy compositions for the SLM process.

Keywords: Additive manufacturing, selective laser melting, Scalmalloy, Solidification, EBSD, Electron microscopy

1 Introduction

Additive manufacturing (AM) technologies, and in particular the Selective Laser Melting (SLM) process, are powerful tools which can be used to manufacture metallic parts with a high degree of complexity [1]. The process is characterized by the repeated creation of thin layers of metal powder, and the selective laser scanning of successive cross-sections of the parts beeing built. Laser radiation leads to full melting of the powder particles, and consequently almost fully dense metal parts can be built. This presents opportunities for the manufacturing and creation of new and improved products in a wide range of industrial sectors, such as the turbine and tooling industry, automotive applications, optimized valves and pumps [2] and more. Interesting fields include lightweight applications in space (e.g. brackets [3] for satellites, spacecrafts and moon or planetary rovers) and aerospace [4-6].

For lightweight applications the structural potential of SLM can be further exploited by the use of aluminium alloys. As described recently by Spierings et al. [7], to date Al-alloys most commonly employed in SLM are near-eutectic Si containing alloys such as e.g. AlSi12 and AlSi10Mg, which were originally developed for casting processes. However, the mechanical properties of the alloys are highly dependent on their microstructure, which typically is significantly different to conventionally processed alloys. Siddique et al. [8] reported for SLM-processed AlSi12 the growth of columnar dendrites along the scanning direction, and the eutectic formation of fine Si dendrites. A more detailed microstructural analysis was presented by Prashanth et al. [9], pointing out that the microstructure is not uniform throughout the material.

In a view on the scanned cross-section of a sample, a fine microstructure of circular cellular grains with a diameter of 500nm to 1µm was observed. Around these grains, Si was located at the cellular boundaries

with a thickness of ≈200nm. More elongated grains were observed towards the melt pool boundaries to neighbouring scan tracks, showing grain growth towards the neibouring already solidified material.

Thijs et al. [10] presented a profound analyse of SLM-processed AlSi10Mg and found a very similar microstructure as Kimura et al. [11] for an AlSi7Mg alloy. Sub-µm α cellular-dendritic solidification structure is reported, decorated with fine structured eutectic Si particles.

During solidification Al grains follow the cooling gradient, hence grow perpendicular to the melt-pool border towards the centre and top of a melt-pool; this results in large columnar grains with a length of >50µm approx. in the build direction, and a diameter in the range of ≈10µm. Generally, as a result of the inhomogeneous microstructure, and the existence of a distinct texture in the direction of the build process, the mechanical properties depend on the direction of the applied load, i.e. perpendicular or parallel to the orientation of columnar grains. Therefore, anisotropic mechanical properties were found, especially for the yield stress. This anisotropy can reach values up to more than 10%, as e.g. shown by Buchbinder et al. [12] for AlSi10Mg, which is much more compared to SLM-processed Scalmalloy®, a Sc- and Zr-modified 5xxx alloy, developed by the Airbus Group Innovations. Spierings et.al. reported in [13] an anisotropy in yield stress for as-processed material of <4%, and Palm et al. [14] and Schmidke et al. [15] showed comparable low values in a heat treated condition.

The effects of Sc additions on Al-alloy in conventional casting processes have been widely reported, going back to the 1970's. An overview is given in Spierings et.al. [13], and further details are discussed in various publications [16-22]. Norman et al. [18] found for a cast Al-0.7Sc alloy a total absence of columnar grains, hence the grain structure was equiaxed and significantly refined. However, next to the formation of Al_3Sc particles leading to a significant strengthening effect of aluminium, the main effects of Sc on the Al-microstructure can be summarized by exceptional grain refining and the prevention of recrystallization up to high temperatures. First insights into the SLM-processed microstructure is given by Palm [14] and Spierings [13], presenting basically a microstructure consisting of very distinct regions of extremely fine equiaxed grains, next to regions of coarser and more columnar grains. This is completely different to other Al-alloys processed by SLM, and looks also very different to conventional Sc-containing Al-alloys, due to the bi-modal grain size distribution. Therefore, a more profound analysis and description of the observed microstructure after the SLM-process and its formation mechanisms is required. The paper provides an evidence based explanation model, combining the results of microstructural analyses with simulations of the SLM-processing temperature in the melt-pool and Marangoni convection, and a simulation of the solidification and precipitation reactions from the melt. The results demonstrate the necessity for the development of alloys which are designed specifically for AM.

2 Methods and Materials

2.1 Manufacturing of SLM specimens

The Scalmalloy® powder material (nominal composition in wt% 4.6%Mg, 0.66%Sc, 0.42%Zr, 0.49%Mn) as well as the processing window development for a material density >99% is described by Spierings et

al. [13]. A ConceptLaser M2 machine equipped with a Gaussian Nd-YAG laser operated in cw mode was used to produce 10x10x10mm^3 cube samples. Two levels for the applied laser energy density E_V is used (E_V = 115 $^J/_{mm}3$ and E_V = 238 $^J/_{mm}3$) based on a nominal laser power of 200W, a hatch distance of 165µm, a layer thickness of 30µm. The two energy density levels arise from different scan speeds, and are indicated in the respective figure captions. The scan strategy used bi-directional scanning with or without turning of the scanning direction from layer to layer (Fig. 1). All materials investigated were in the as-processed condition to enable insights into how microstructures form during processing

Fig. 1:

2.2 Microstructure analysis

Microstructures were analysed (except for Fig. 7) using a FEI Helios dual beam FIB instrument fitted with EDAX electron backscatter diffraction (EBSD) and energy dispersive x-ray spectroscopy (EDS) systems. Electropolished thin foils were produced for both TEM and EBSD analysis, using a Struers Tenupol-3 twin jet electro-polisher. A 5vol-% perchloric acid in 95vol-% methanol electrolyte was cooled to -50°C and a voltage of 25V applied. The cutting plane orientations (parallel to the scanning xy-plane or the yz-plane) for microstructural measurements are given in the respective figure captions, using the coordinate system shown in Fig. 1. EBSD maps were recorded at two magnifications, ×10k for analysis of the fine grain regions and ×750 magnification for analysis of larger areas representing the general microstructure; step sizes of 80nm and 250nm were used respectively. EBSD beam conditions of 20kV and 5.5nA were used, samples were inclined 70° to the incident beam and a working distance of 12mm was employed.

TEM analysis was performed in a probe side aberration corrected JEOL 2100FCs microscope operated at 200kV. Both bright field (BF) and high angle annular dark field (HAADF) images were captured in scanning transmission electron microscopy (STEM) mode. Scattered electrons were collected using a HAADF detector, over a semi-angle ranging from 75 - 150mrad, to produce z-contrast dark field images. Chemical energy dispersive x-ray spectroscopy analysis carried out in the TEM were performed using a windowless EDAX-EDS system; data was interpreted using the EDAX-TEAM software.

The TEM sample preparation for higher resolution EDS analysis presented in Fig. 7 consisted in cutting slices 0.3 mm in thickness with a diamond saw, punching out of them 3 mm disks with an Eckert® puncher and reducing their thickness with silicon carbide polishing paper down to about 80 µm. The final thinning to electron transparency was made by Ar ion milling with a PIPS II® from GATAN®, in single modulation mode, at 5 kV and 4° incidence mode till a hole was made, then at 4 kV, 3.5° for ¾ of an hour. Final cleaning was made at 0.5 kV and 3.5° incidence for 20 minutes. The ion milling was made at liquid nitrogen temperature to minimize any impact on the microstructure. TEM was made on a FEI® TALOS® F200A operated at 200 kV and equipped with a field emission gun. Imaging was mainly performed in STEM mode with the high angle annular dark field (HAADF) detector to reveal elemental Z contrast. Energy dispersive X-ray spectroscopy (EDS) was performed with the FEI Super-X® detector, which consists of 4 silicon drift detector (SDD) diodes located around the sample area in the objective

lens, allowing for a large collection angle of 0.9 srad. Combined with the brighter electron emitter X-FEG® of FEI, this results in a 60 fold increase in acquisition rate compared to a conventional system. Chemical maps were acquired in 30 minutes.

The cross sectional dimensions of single weld lines, which were approximately elliptical in shape, were determined from optical micrographs. The scan line width corresponds to the long axis of the ellipse and the depth is represented by half of the maximum short axis.

2.3 Thermodynamic calculations

Pandat Software Database "PanAl_2013" was used to simulate equilibrium phase diagrams of the Al-Mg-Sc system. This approach helped to provide insights into the alloy behaviour, in particular the effects of Mg-content on Sc solubility. However, the SLM-process results in very high cooling rates of up to $\approx 10^{6K}/_{s}$ for aluminium, hence the solidification takes place in non-equilibrium condition and conventional phase diagrams are not able to accurately predict the alloy phase components. Therefore, a non-equilibrium simulation based on Scheil-Gulliver for the Al4.6Mg0.66Sc0.42Zr0.49Mn Scalmalloy was also performed. The model is based on a diffusionless system, hence replicates the extremely high cooling rates that are associated with SLM. The simulation database calculates separately for both Al_3Sc and Al_3Zr; mixed phases of $Al_3(Sc_xZr_{1-x})$, as were observed in the microstructure, are represented by the sum of Al_3Sc and Al_3Zr.

2.4 Computational simulation of the SLM-process

The numerical model of the melt-pool during SLM-processing consists of a weak coupled temperature field and fluid flow calculation. It is based on a grid of 10x10x10 µm³ cubic elements representing the material configuration. This is also the basis for the finite differencing scheme of the temperature field calculation, and the combined level set volume of fluid scheme of the fluid flow simulation, which is calculated on a staggered grid as proposed by Son et al. [23, 24]. A detailed description of the simulation model is given by Heeling & Wegener [25].

The model allows for the consideration of various physical effects, which include temperature dependency of material parameters (Table 1), melting/freezing, evaporation, heat conductivity, convection and radiation as well as buoyancy forces. The model is completed with the surface-tension driven Marangoni convection, capillary forces and recoil pressure. In its initial state the powder bed is homogenized to partly filled elements with a filling degree of packing density. The measured packing density of about 31% and its effective layer thickness of about 100 µm were simplified for the simulation to a packing density of 60% and a layer thickness of 50 µm to decrease calculation times. The resulting error in power absorptance of both configurations is lower than 0.5% and therefore neglectable. Furthermore most of the energy is absorbed in the melt-pool due to very slow scan speeds. Material advection is enabled by the fluid flow calculation so that the melt can consolidate as soon as elements melt. To enable a dynamic calculation of absorptance on solid and molten material as well as in the powder bed the absorption model proposed by Gusarov et al. [26] is used and numerically improved so that a spatial resolution down to the size of an element stack is possible. To increase the simulation's

comparability to experimental results a multi-track configuration is used in which a previously solidified track is located next to the powder bed (Fig. 2).

Fig. 2:

The thermal simulation used the equilibrium liquidus temperature of 635°C as the (eutectic) melting point. The thermophysical properties used for the simulation are taken from published data and/or our own measurements at temperatures of ≤ 900°C. For higher temperatures the reported values are extrapolated linearly. For the specific heat capacity the properties of Al-6061 as reported by Mills [27] are used, with a constant value in the molten condition as reported by Brandes et al. [28] and Dinsdale et al. [29] for pure aluminium. The solid thermal conductivity λ_s is based on equation (1), with $\varkappa$ the thermal diffusivity, ρ the solid material density and c_p the specific heat capacity. $\varkappa$ was measured up to a temperature T = 600°C using the laser Flash method (Netzsch LFA 457 MicroFlash®). The density ρ is based on equation (2) with $\rho_0 = 2.675\ {}^{g}/_{cm^3}$ and ε = 3·α, the coefficient of thermal expansion. Thereby, α is a linear extrapolation up to the melting temperature from measurements (TMA Hyperion 402 F3, Netzsch) taken up to T= 100°C, and can be written according to equation (3). The thermal conductivity in the liquid state λ_l is referred to Powell [30], and can be expressed by equation (4). The thermal conductivity of the powder material is implemented according to Sih & Barlow [31], showing that it is only ≈ 1 ‰ of the thermal conductivity for solid or fluid material. Therefore, almost all thermal energy is transmitted vertically into the melt-pool, and the already solidified material, respectively.

$$\lambda_s = \varkappa \cdot \rho \cdot c_p \tag{1}$$

$$\rho(T) = \rho(T_0) \cdot \exp\left(-\int_{T_0}^{T} \varepsilon(t)dt\right) \tag{2}$$

$$\alpha = \frac{\varepsilon}{3} = 3.084 \cdot 10^{-8} \cdot T + 1.407 \cdot 10^{-5} \tag{3}$$

$$\lambda_l = 0.0444 \cdot T + 46.53 \tag{4}$$

According to Brandes [28] the temperature dependency of the dynamic viscosity η(T) can be modelled by an Arrhenius function:

$$\eta(T) = \eta_0 \cdot exp\left(\frac{E}{R \cdot T}\right) \tag{5}$$

with η_0 = 0.149 mPas, the activation energy for viscous flow $E = 16.5\ {}^{kJ}/_{mol}$ and R the gas constant. This function was used for temperatures between 800°C and 900°C. For T < 900°C, Dinsdale [29] reported some variations in measurement data. Therefore, a fit was done to a reasonable η-value at the melting temperature (1.38 mPas). For high temperatures above 900°C, Li et al. [32] reported for a Al-0.6Zr alloy an increase in viscosity during heating. At T ≈ 900°C these data (taken from the graph) fit well to the data from equation (5) after conversion from kinematic to dynamic viscosity values. Therefore, these reported data were used for the simulation. According to Brandes et al. [28] the surface tension γ(T) follows equation (6), with $\gamma_0 = 914\ {}^{mN}/_{m}$ and ${}^{d\gamma}/_{dT} = -0.35\ {}^{mN}/_{mK}$.

$$\gamma(T) = \gamma_0 + (T - T_0) \cdot \frac{d\gamma}{dT} \tag{6}$$

However, such values fit very well to values for oxidised aluminium, as reported by Garcia-Cordovilla [33], who further reported a significant dependency of γ from the Mg-content. For non-oxidised aluminium he reported:

$$\gamma(Mg) = \gamma(Al) - 71.8 \cdot Ln(1 + 0.31 \cdot Mg) \quad (7)$$

For the current alloy with 4.5% Mg, γ(4.5%) = 1'047 $^{mJ}/_{m2}$. With this value, the calculated γ-values for oxidised aluminium where corrected to corresponding non-oxidised values. The recommended value for heat of fusion of (pure) Al is 10,58 $^{kJ}/_{mol}$ [29] and the heat of evaporation is 284 $^{kJ}/_{mol}$. The absorption coefficient of laser radiation by the aluminium powder layer in SLM is calculated in every time step using a model developed by Gusarov et al. [26], in which the hemispherical reflectance δ of aluminium is required. Values for δ = 0.94846 for a laser wavelength of 1070nm are taken from data presented by Rakić et al. [34], provided by Polyanskiy et al. [35]. However, the Al-powder particles are covered by an oxide layer, and δ for pure Al-Mg-oxide would be only ≈0.07. On the other hand, the oxide layer is extremely thin and powder particles surfaces display considerable topographic relief, thus the effective value for δ is estimated to be somewhat lower than the reported value for pure Al. Based on these above mentioned considerations an effective value for δ = 0.85 was used, which seems a realistic guess. An overview on temperature-dependent material data at selected temperatures is given Table 1.

Table 1: Thermo-physical material properties

Property	Unit	T = 100°C	T = 635°C	T = 900°C	T = 1400°C
Specific Heat Capacity	$^{J}/_{mol\ K}$	25.6	31.6	constant 29.4	
Thermal conductivity	$^{W}/_{m\ K}$	129.5	86.9	98.6	117.2
Surface Tension	$^{N}/_{m}$	-	1064 @ 650°C	977	898
Dyn. Viscosity	Pa s	-	1.38	0.81	1.71

3 Results

3.1 SLM samples

Cube SLM samples, produced with E_V=115 $^{J}/_{mm3}$ had a mean density of 99.3±0.1%, and those produced with a higher energy input of 238 $^{J}/_{mm3}$ had a density of 99.5±0.1% [13].

3.1 Microstructure

The typical microstructure found in SLM-processed Scalmalloy is shown in Fig. 3. A layer-wise build structure was observed, with alternately coarse and fine grained material, whereas towards the top of the last scan track the material solidified forming columnar grains.

Fig. 3:

In a cut perpendicular to the scan direction, the microstructure showed comparably large weld line cross-sections. In an analysis of weld pool dimensions a width of 260±18µm, significantly larger than the laser beam diameter, and a depth of 94±9µm was measured (N=29), valid for a wide range of different energy inputs. Such large dimensions might be expected for aluminium due to its good thermal conductivity, and are therefore larger than for other alloys such as Fe-, or Ni-based alloys, where significantly lower laser energy inputs are required. Fig. 4 shows the microstructure of a sample with the scan direction alternating

from layer to layer (Fig. 4a) and a sample without layer to layer rotation (Fig. 4b). The alternately coarse and fine grained structure is obvious, with the darker areas being regions of very fine grained material. From the weld-lines being prepared perpendicular to the scan direction (Fig. 4a), the typical thickness of these darker regions is approximately 10μm. Furthermore, the darker regions displayed partial fan-shaped deviations merging into the lighter areas, which represent coarser grained material. This indicates significant convection took place in the melt-pool.

Fig. 4:

As described in more detail in [7], the grains in the fine grained (FG) material are very small (Fig. 4), in a range of ≈150nm to ≈1μm (with a mean at ≈660nm), whereas the coarser grains (CG) vary much more, and are typically between ≈2μm to 15μm. Even more interesting is the fact that in the FG material there is absolutely no evidence of any texture (see pole figure in Fig. 5b). In contrast the CG material shows a typical grain orientation more or less in build direction, and with a variation of grain orientation in the <100> direction, hence indicating grain growth perpendicular to the melt pool boundary. This indicates different solidification behaviours for the two distinct different grain regions. Obviously, the transition from FG to CG is very distinct, as can also be seen in Fig. 5.

Fig. 5:

3.1 Precipitates and particles

There are a number of different particles and precipitates in the alloy, which are described in more detail below. This description is essential for the discussion of microstructure formation mechanisms during SLM.

In order to gain insights into the distribution of the main alloying elements, EDX maps were collected from electropolished surfaces. In Fig. 6 EDX elemental maps showing Mg, Sc, and Zr distributions are presented. Fig. 6c) gives evidence for a slightly higher concentration of Mg in some areas of the FG region, whereas in the CG-region no accumulation of Mg can be observed. However, the specific content of O, Zr and Sc is very low, and eventually below the limit of detection. From their distribution information (Fig. 6 d, e, f) it becomes evident that no segregation to FG or CG takes place. Hence, one can make the assumption that these elements are distributed homogeneously.

Fig. 6:

A large oxide particle with a size of a couple of μm was identified in FG material (see Fig. 6d). This particle might be debris of the oxide layer that coated the Al-powder raw material, and/or from the oxides covering a scanned layer, which were then remolten several times. An EDS-mapping using STEM at a higher magnification of a FG-region is shown in Fig. 7. It shows Mg diffuse segregation and Mg-rich particles at the grain boundaries (Fig. 7b) with a typical size of ≈50nm to 100nm. In contrast, Sc-Zr rich particles (Fig. 7c and d) are located both at grain boundaries and in the Al-grains. They are dendritic-like / diffuse shaped of varying sizes. Some Sc-Zr rich and Mg-rich particles are interconnected. There is also

segregation and precipitation rich in Mn and Fe at the grain boundaries, with sizes around 50 nm. It seems the Mn-Fe rich precipitation forms a shell around the Sc-Zr rich particles.

Fig. 7:

The backscattered electron SEM image shown in Fig. 8 displays a region containing both fine and coarse grains. It becomes evident that the transition from fine to coarse grained material takes place from one grain to the other, hence at a very distinct position in the melt-pool. Obviously the concentration of particles (bright spots) is much higher in FG than in the CG areas, and many of them are concentrated along grain boundaries in both FG and CG regions. However, also intragranular particles exist, with a higher concentration in FG than in CG regions.

Fig. 8:

There is a tendency for somewhat finer particles in samples produced with lower energy input, hence with a shorter melt-pool life time, however, no quantitative analysis has been performed on this so far. An EDX analysis in FG material revealed that these particles contain Mg, Al and O and are most probably $MgAl_2O_4$ (spinel). It is important to note that the particles also contain Sc and Zr. In order to identify in more detail the types and occurrence of particles in fine and coarse, columnar regions, a detailed STEM-analysis on a foil consisting of FG and CG material was performed. A range of different precipitates were found in FG-material. In a BF-TEM image (Fig. 9), cube-shaped / facetted precipitates are visible, with a typical diameter of 30nm – 50nm.

Fig. 9:

Based on a FFT- and EDX analysis, the dark rhombic particle, adjacent to the grain boundary (GB), as presented in Fig. 9c, was identified as $MgAl_2O_4$, whereas the adjoining particle was identified as Al_3 (Sc, Zr). Using the Al lattice as a means of internal calibration, based on published data for Al [36], a corrected lattice constant of 8.05Å was measured for $MgAl_2O_4$, which is in perfect agreement with literature data of spinel of 8.08Å [37] at room temperature. However, further EDX particle analysis on FIB foils revealed that other, more complex Al-oxide particles also exist, with traces of heavier elements such as Sc, Zr, and occasionally Fe. Next to numerous AlMg-oxides, some further precipitates were found in FG-material. Fig. 10 shows a cube-shaped intragranular particle in a [112] oriented fine grain, with a side length of ≈50nm. The EDX analysis, as-well as the lattice constant identified from a FFT of the particle indicate that these precipitates are in-fact semi-coherent $Al_3(Sc_xZr_{1-x})$ particles with $L1_2$-structure.

Fig. 10:

In coarse columnar grains, however, the precipitation behaviour is insofar different as significantly fewer intragranular particles were present compared to FG-material. Fig. 11 shows one of the intragranular precipitates, identified by EDX as Al-Mg-oxide particles, with a comparable diameter as in FG-material.

Fig. 11:

Next to intragranular oxide particles with a diameter in the range of ≈100nm, there were smaller particles pinning grain boundaries (GB) both in the FG- and CG-regions (Fig. 12). Such particles were identified as $Al_3(Sc_xZr_{1-x})$; these precipitates were typically in the range of ≈ 10 to 30nm. These GB-particles showed a cube on cube orientation relationship with the Al matrix.

Fig. 12:

4 Discussion

The SLM-specific processing conditions create a microstructure that differs significantly to conventionally processed materials. These conditions comprise the very rapid melting of a thin powder layer, forming a melt-pool lying on top of already consolidated, comparably cold material. After the passage of the laser beam the melt-pool solidifies rapidly due to the very high cooling rate of up to $\approx 1.5 \cdot 10^6$ $^K/_s$. Any microstructure formation, therefore, is driven by high cooling rates and direction, as well as by reactions taking place in the melt-pool before and after solidification. In general, such reactions will include particle precipitation, particle growth due to cyclic heat treatment from next laser scans, changes in grain morphology by recrystallization effects etc.

Obviously, the formation of the very specific microstructure in SLM-processed Scalmalloy (Fig. 4 and 6) will depend on a combination of several different effects, including the formation and behaviour of precipitates found in the as-processed material.

5.1 General considerations of Sc in 5xxx-alloys

Sc and Zr are known to form $Al_3(Sc_xZr_{1-x})$ particles, which act in different ways to refine microstructures, and in turn improve mechanical properties. In hypereutectic Al-Sc alloys primary Al_3Sc particles precipitate from the melt during casting, which then act as seed crystals for Al-matrix grain growth. These particles are very effective seeds due to the structural similarity between FCC aluminium and Al_3Sc and the small lattice parameter misfit, which is reported as 4.049 Å [38], and 4.103 Å [36] (at room temperature), respectively.

The eutectic composition at the Al-rich corner in equilibrium is reported to be between 0.55 wt% [18, 39] and 0.6wt% [21] Scandium. At high cooling rates, the calculated solid solubility of Sc in Al at the eutectic temperature is ≈0.35 wt-% (Fig. 13a) and as solubility is reduced considerably at room temperature, continuous precipitation of Al_3Sc particles, or in the presence of Zr to $Al_3(Sc_xZr_{1-x})$, is promoted. The addition of Mg to an Al-Sc alloy reduces the solubility of Sc in Al (Fig. 13a). The simulation shows that for the given Mg-concentration of 4.6% the eutectic equilibrium Sc-concentration is reduced to ≈0.27wt% Sc, and the maximum solubility in Al is reduced to 0.11wt%. Hence Scalmalloy can be considered as a hypereutectic alloy, therefore leading to the formation of additional primary Al_3Sc particles during consolidation. However, in contrast to the reduced solubility of Sc in the presence of Mg, the non-equilibrium conditions induced during SLM processing in fact increase the solubility of Sc; Røyset [21] refers to a significantly increased solubility of up to 0.6wt% at cooling rates of 100 $^K/_s$. However, after

solidification during cyclic heat treatment caused by next laser scans the reduced solubility becomes increasingly important.

Fig. 13:

The SLM processing window and related melt-pool lifetime therefore influences the formation of Al-Sc precipitates. The presence of such inoculant seed crystals in a melt results in a significant grain refinement of the Al-alloy, which has been widely reported for various cast Sc-containing Al-alloys (Venkateswarlu et al. [41], Zakharov [42], Norman et al. [18] and Røyset et al. [21]). Next to the formation of $Al_3(Sc_xZr_{1-x})$, Al-Mg-oxides (MgO, $MgAl_2O_4$, Al_2O_3) are also formed during SLM consolidation. Raw powder material is exposed, during handling, to atmospheric conditions, hence the powder particles are enveloped by an extremely thin Al-oxide, and sufficient O is therefore available during the process to precipitate numerous oxide particles. Harmelin [40] published the ternary equilibrium phase diagram for Al, Mg and O at 800°C and 1'227°C (Fig. 13b). It shows in the Al-rich corner a stable τ-phase ($MgAl_2O_4$, spinel). The melting temperature of $MgAl_2O_4$ is 2'135°C, but no literature data was found for the solution temperature of this τ-phase in molten Al and especially not at high cooling rates as in the SLM process. However, it can be expected that such oxide phases will survive even higher temperatures than the reported 1'227°C, as the comparison of the Al-rich corner of the phase diagrams at 800°C and 1'227°C displays no differences.

5.2 Melt pool temperature distribution and microstructural features

The microstructure in SLM-processed Scalmalloy shows a bi-modal grain size distribution, with a fine grained (FG) region having a typical thickness of ≈10µm and a coarse grained (CG) region of approximately 20µm (Fig. 4 and 5). Before discussing the formation of the FG and CG areas in detail the results of the simulation of the temperature distribution in the melt-pool during SLM is first considered. The simulation model not only provides insights into the temperature distribution, but also provides information about the melt-flow regime in the melt pool and the cooling gradients achieved during processing. This is of considerable importance as it enables a better understanding of how the observed microstructures might be formed. Fig. 14a shows the half-elliptic cross-section of the melt-pool at the position of maximal melt-pool depth of ≈95µm to 100µm, whereas the maximal melt-pool width is ≈280µm. As a model validation, the measurement of typical melt-pool dimensions revealed a width of 260 ± 18µm and a depth of 94 ± 9µm, which is in very good agreement. It has to be mentioned that the dimensions vary quite significantly over a sample cross-section, depending on local melt-flows inside the melt-pool, and the local effects of energy in-coupling, effective powder layer thickness and its corresponding quality etc. Nevertheless, a comparison of the melt-pool depth (≈100µm) with the typical layer thickness of 30µm indicates that by each new layer the previous layers are remolten approx. 2 to 3 times. In the following we consider the important question to which extent particles that might have formed during the repeated heating and cooling dissolve or survive the re-melting procedure.

Fig. 14:

The temperature in the melt-pool reaches the vaporization temperature of aluminium at 2'520°C [43], hence very high temperature gradients, as high as $\approx 20 \cdot 10^6$ $^{K}/_{m}$ exist. The simulation also shows significant melt flows within the molten pool (Fig. 15). The greatest convection velocity is in the range of 16 $^{m}/_{s}$, with a mean value over the melt-pool cross-section of 1.4 $^{m}/_{s}$, whereas in the temperature region < 800°C flows are much slower, and reach only about 0.74 $^{m}/_{s}$. Fig. 15a shows the Marangoni convection at the position of the deepest melt-pool (compare Fig. 14b) with downside oriented flows in the melt-pool centre and outwards oriented convections at the top of the melt-pool, respectively. Such Marangoni-driven convections are jointly responsible for deepening and widening the melt pool to such large dimensions, as reported by Drezet [44]. When the melt pool moves forward by about 0.1mm, the flows (analysed at the same position, hence 0.1mm towards the melt-pool tail) in the middle of the pool's cross-section are oriented upwards (Fig. 15b) but the top flow directions remain oriented outwards. This indicates that the flow directions in the pool change significantly depending on the specific location. The large melt pool width further facilitates the rise of thermo-capillary forces due to highest thermal and corresponding surface tension gradients exist especially at the circumference of the laser beam and between the centre and the edge of the weld pool, as reported by Khairallah [45] and Olakanmi [46].

Fig. 15:

Comparing the shape and size of the FG and CG regions with the isotherms presented in Fig. 14 shows that the thickness of the FG region corresponds very well to the simulated thickness of a shell around the melt-pool having a temperature between liquidus (635°C) and 800°C (Fig. 14). However, the shape of the FG-region is not a perfect shell around the melt-pool, but shows partially fan-shaped deviations. This is related to the Marangoni convection taking place in the melt, affecting the thickness and shape of the FG-region by local eddies.

It is assumed that the FG-region is related to the existence of $Al_3(Sc_xZr_{1-x})$ seed crystals, as e.g. known from casting processes [47-49]. Therefore, it is necessary to understand the temperature range where such seeds can survive the SLM process. The Scheil-simulation in Fig. 16 indicated that at 795°C all Al_3(Sc, Zr)-precipitates have gone into solution.

Fig. 16:

Thus, according to the assumption, the 800°C-isotherm in Fig. **14** represents the border between regions of rich and poor in Al_3(Sc, Zr)-precipitates and consequently the border between FG and CG regions.

Different types of particles are observed in the FG-microstructure. Small ≈30nm – 100nm $Al_3(Sc_xZr_{1-x})$ particles (Fig. 7c and d, Fig. 10) and Al-Mg oxides (Fig. 9, Fig. 11), some of them were identified as $MgAl_2O_4$. Similar facetted and cube-shaped $MgAl_2O_4$ particles were observed by Yun et al. [50] and Hyde et al. [51] in cast Al-alloys, although they were much bigger due to diffusion controlled particle growth. However, very often also mixed Al-Mg-oxide / Al_3(Sc, Zr) couples were found. The preferred mechanism for the formation of such mixed particles is that Al-Mg oxide particles act as nucleation sites

for heterogeneous $Al_3(Sc_xZr_{1-x})$ precipitation, as Al_3Zr precipitation takes place at temperatures <800°C when Al-Mg-oxides already exist. In fact, at 660°C the lattice constant of the τ-phase, 8.1263Å [50], is approximately twice that of Al_3Sc (4.103Å [36]). Therefore, it can be expected that the τ-phase can act effectively as a seed for $Al_3(Sc_xZr_{1-x})$ precipitation. Such mixed phases could then act as a seed for FCC-aluminium growth, as the lattice constant of aluminium (4.049 Å [36]) is also very close to that of Al_3Sc. A similar nucleation behaviour is also reported by Hyde [52] for the heterogeneous nucleation of Al_3Sc on oxide particles in the melt,

The $Al_3(Sc, Zr)$ particles showed diffuse shaped facetted, star- or cube-like shapes as reported by Marquis et al. [53], who analysed experimentally and theoretically different morphologies of Al_3Sc precipitates depending on the temperature-time history during annealing. Røyset et al. [21] reported in a cross-reference that different Al_3Sc particle shapes can occur, such as cubic, hexagonal, dendritic or globular, respectively, depending on the specific melt overheating and cooling rate. However, the number density of such particles is not uniform throughout the microstructure. As mentioned above, there is a considerably higher number density of particles in the FG-region, whereas in the CG-region almost no $Al_3(Sc_xZr_{1-x})$ particles and also fewer Al-Mg oxides were found (Fig. 8). Modelling results predict melt-pool temperatures up to ≈2'500°C (Fig. 14). As the melting temperature of $MgAl_2O_4$ is clearly above 1'227°C (Fig. 13b), it is therefore likely that Mg-oxides dissolved in the region towards the top of a melt-pool. The matrix Mg concentration is then dependent on the specific location in the weld-line, its initial local concentration of Mg-containing particles and the temperature level and distribution during SLM-processing and even a local increase of the Mg-matrix concentration is possible. Hence, this indicates that the slightly higher content of Mg in FG material, as is evident in Fig. 6c, might be a direct result of the high number density of Al-Mg oxide particles.

In the FG-region any existing, previously precipitated $Al_3(Sc_xZr_{1-x})$ particles, as shown in Fig. 7c, survive the heat cycle associated with the SLM scanning process, as the dissolution temperature according to the Scheil-simulation is not reached (Fig. 16). Such particles would then act as nuclei for Al-matrix solidification. Their similar lattice constant to the Al-matrix provides ideal nucleation sites, which induce the rapid formation of FCC-aluminium grains. These grains are in an almost ideal way randomly oriented as shown by Spierings [7], as the distribution of the misorientation angles between single grains follows almost perfectly the theoretical ideal distribution for randomly oriented grains according to Morawiec [54]. Therefore, and due to the very high cooling rates of $\approx 10^6\ ^{K}/_{s}$, one can assume that the nucleation and growth takes place almost simultaneously all over cross-section of the FG-region. Their competitive growth on the numerous inoculants is assumed to lead to the formation of a fine grained microstructure. The pronounced Mg-segregation at grain boundaries (Fig. 7b) indicates the non-equilibrium state of the structure.

Next to particles containing Sc and Zr, Yun et.al [50] report for cast Al-Mg alloys the formation of $MgAl_2O_4$, α- and γ-Al_2O_3 particles originating from the oxide layer, and that such particles themselves also act as seed crystals in the case when intensive shearing takes place in the melt. They concluded this

from the calculated lattice misfit between α-Al and such particles. The reported misfit values at 660°C are 1.41% for Al/MgAl2O4, and only -0.48% for Al/α-Al_2O_3. These values are comparable to the lattice misfit between Al and Al_3Sc. So this effect could potentially also play a role during SLM, as the single τ-phase has also been found in FG-grains (Fig. 9). However, it is assumed that single or mixed Al(Sc,Zr) phases are the dominant inoculant seed crystals, otherwise the FG-area would be thicker and would not stop at the 800°C isotherm. Furthermore, the lattice misfit of Al_3(ScxZr1-x) is slightly smaller compared to the misfit of $MgAl_2O_4$. The effective lattice parameter misfit of $Al_3(Sc_xZr_{1-x})$ depends on the effective enrichment with Zr, next to the temperature and atmospheric pressure. There is a significant solubility of Zr in Al_3Sc and up to ≈50% of the Sc-atoms can be replaced [55]. This leads to a better lattice match between Zr-enriched $Al_3(Sc_xZr_{1-x})$ and aluminium, hence $Al_3(Sc_xZr_{1-x})$ possesses an even higher seed crystal potential.

In addition to acting as seed crystals, many Al_3(Sc, Zr) particles are precipitated along grain boundaries (Fig. 7c), or are pushed to the solidification fronts of the α-Al grains, as reported by Hyde [52]. Heterogeneous nucleation has also been reported by Seidman et al. [49] and Das [56] in cast Al-Sc alloys. Such GB-particles are understood to influence microstructures, hence mechanical properties, due to their pinning effect on grain boundaries: They help to stabilize the microstructure by Zener pinning [57], preventing the movement of high- and low-angle grain boundaries, hence leading to a very high resistance against grain growth [58, 59]. Jones et al. [58] analysed the recrystallization kinetics of different Al-Sc alloys and found, even for low Sc-concentrations (c.a. 0.25wt%), that grain growth starts at very high temperatures of ≈500°C, requiring annealing times of >10'000s. This explains why no severe grain coarsening occurs in the FG regions during the repeated heat input of the following scans.

In summary different types of primary intermetallic phases are found in high number densities in the FG-regions (Fig. 6, Fig. 7). They act as inoculants for competitive growth of FCC-aluminium grains but also, pin and stabilize grain boundaries. Consequently, a very fine grained microstructure evolves with grain sizes between about 150nm and 1μm. The distinct absence of any texture observed in the FG material (Fig. 5) can be explained by the epitaxial growth of aluminium grains on the randomly oriented seed crystals which are present from the melt. In fact, we have previously shown that the orientation misfit within the fine grained material follows an almost ideal distribution [7].

Within the CG-region, the microstructure formation mechanism obviously differs compared to the FG-region where solidification takes place almost simultaneously. This CG-region is characterized by columnar grains with lengths varying between ≈2μm and ≈15μm. Solidification starts in the CG-region with the growth of columnar grains, which nucleate on the FG-region. This columnar grain growth proceeds to the top of the melt-pool, as indicated in Fig. 3a). Grains grow radially to the melt-pool border (Fig. 5, Fig. 11) in the direction of the thermal gradient and in dependence of the growth rate, as described by Thijs et al. [10] for an AlSi10Mg alloy. However, the grain sizes observed in this work are still by a factor of 5 to 10 times smaller than those typically measured in AlSi10Mg alloys. This indicates that, to a certain degree, forced Al-nucleation on seed crystals might play an important role in forming the final

microstructure. As described earlier $MgAl_2O_4$ particles can survive comparably high temperatures in an Al-melt and can act as nuclei, especially in areas close to the FG-region where temperatures are low compared to the upper regions of the melt-pool, and therefore a higher Al-Mg-oxide density is expected.

Significant melt flows exist during the melt-pool life-time, and flow directions change over time (Fig. 15), hence the melt-pool, particularly in the hot region, is mixed quite effectively. The Al-Mg-oxide particle number density is therefore highly depending on the specific location in the melt-pool, and a homogeneous distribution cannot be expected. In regions of very high temperature $MgAl_2O_4$ particles will go into solution, while some particles will remain in areas closer to the FG-region.

5 Conclusions

Till this day the use of SLM has been carried out almost exclusively with conventional Al alloys. As to the microstructure and related anisotropic mechanical properties there is a strong need for new alloys that make better use of the SLM processing conditions. These SLM-specific conditions are the existence of a very small melt-pool, situated on comparatively cold solidified material. Hence, very high cooling rates are evident, as are the associated short melt-pool lifetimes. This results in a high undercooling of the melt and a high temperature gradient in the melt-pool, respectively. For the Scalmalloy® alloy these special conditions appear to be particularly suitable, and several interesting microstructural effects in SLM-processed Scalmalloy® can be observed:

- The microstructure of SLM processed Scalmalloy shows two distinct regions, one with a very fine grained microstructure without any preferential grain orientation, and a region with larger columnar grains growing along the temperature gradient. The fine grained region is assumed to be caused by the high number density of seed crystals present in the shell around the melt-pool base favouring an almost simultaneous growth of fine grains throughout the thickness of this shell. The coarse-grained region is related to the high temperature gradient associated with low number density of Al-Mg-oxide seed crystals favouring a more columnar and coarser grain structure.

- The Scheil simulation revealed that $Al_3(Sc,Zr)$ particles dissolve at a temperature of ≈ 800°C. The thermal simulation showed that temperatures < 800°C within the melt-pool do exist only in a comparably small shell around the melt-pool having a thickness of ≈ 10 µm, which is consistent with the observed fine- grained region of the microstructure.

- There is evidence that $Al_3(Sc_xZr_{1-x})$, and mixed oxide particles (Al-Mg-oxides) have an influential role in the formation of the fine grain material. This is driven primarily by the excellent lattice match between such particles and also with the Al matrix, hence they can act as potent inoculants.

- There is no evidence of pronounced grain growth in SLM-processed Scalmalloy, which is in contrast to other conventional aluminium alloys processed by SLM. Grain boundaries are pinned by small $Al_3(Sc, Zr)$ precipitates. Such grain boundary pinning helps to stabilize against grain growth during the heat impact occurring by the deposition of subsequent layers.

Due to these remarkable microstructural effects, the Scalmalloy material is an interesting candidate for "alloys tailored for additive manufacturing" although the basic alloy system and the general working principles of Sc-modified Al-alloys has been understood for many years. However, due to the comparably high costs of Sc and a high buy:fly ratio of conventional manufacturing technologies often exceeding a value of 6 [58], such alloys have not found widespread applications as commercial structural products, although their properties are excellent. In contrast SLM manufacturing offers near net shape capabilities, which enables a buy:fly ratio close to 1; this makes AM an interesting alternative production route for complex structural components and the use of more expensive materials might become realistic.

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Figure captions

Fig. 1 SLM sample build strategy and a 1cm^3 cube sample produced with E_V=115 $^J/_{mm}3$

Fig. 2. Illustration showing different simulation time steps of a single cross section. The dashed line represents the melt pool boundary. a) Initial state with partly filled powder bed elements and a previously solidified track (right) on top of the previous layer (bottom). b) Simulated melt pool geometry at is deepest extent (see Fig. 13b). c) Almost fully solidified melt pool.

Fig. 3. a) Microstructure in the upper region of a cube sample produced without turning the scan direction from layer to layer, taken longitudinally to the scan direction, showing the final scan track with columnar consolidation. b) Microstructure taken from a middle region. Build direction from bottom to top.

Fig. 4. a) Microstructure of a sample produced at E_V = 115 $^J/_{mm}3$, with the scan direction alternating from layer to layer; the build direction is indicated. b) A similar bi-directionally scanned sample built without layer to layer rotation.

Fig. 5. a) EBSD inverse pole figure (IPF) map of the same area as shown in Fig. 4b. The [001] direction refers to the build direction. b) [001] Pole figure from single FG area. c) [001] Pole figure of the complete scan area with FG and CG material. The points are weighted to the grain size, hence the large points relate to coarse grains and the fine "background" is created by the FG material.

Fig. 6. EDS mapping of a sample produced with E_V = 115 $^J/_{mm}3$ and unidirectional scan strategy, showing Mg, O, Zr and Sc in CG and FG material. Beam: –8kV, 5.5 nA. Drift correction active and a collection of 256 frames. Pictures are optimised for contrast and brightness. A) Microstructure showing coarse grained (CG) and fine grained (FG) areas, b) magnification of a), c) EDS mapping showing O, Mn, Mg, Al, Zr and Sc in the same image.

Fig. 7. a) HAADF-TEM analysis in a FG region of a sample produced with an energy density E_V=135 $^J/_{mm}3$, hence similar to the sample from Fig. 6. EDS-mapping for Mg- (b), for Sc-concentration (c), (d) for Zr and (e) for Fe. An overlay of Mg (yellow) and Sc (red) is shown in f), and the overlay fo Sc and Mn is shown in g).

Fig. 8. SEM pictures of the microstructure showing FG and CG material, and grain boundary precipitates. a) E_V = 238 $^J/_{mm}3$, b) E_V = 115 $^J/_{mm}3$. c) HAADF-STEM image of oxide particles in FG material

Fig. 9. BF-STEM analysis in FG-region. a) Overview showing several grains with intragranular particles. b) [110] oriented grain with two neighbouring grains with intragranular and GB-near particles. c) GB-near particles at higher magnification with FFT confirming $MgAl_2O_4$.

Fig. 10. a) BF-TEM analysis in the core of a FG-[112] oriented grain. c) Cube-shaped $Al_3(ScZr)$ particle at higher magnification. FFT approved its $L1_2$ structure. c) EDS results showing corresponding signals of Al, Sc and Zr.

Fig. 11. CG-material in TEM foil showing intragranular Al-Mg-oxide particle surrounded by dislocations

Fig. 12. BF-TEM image showing grain boundary pinning of a columnar grain (a) and in [100] fine grain (b). c) Magnification of GB pinning at a [110] oriented grain. FTT analysis was consistent with Al_3Sc reflections

Fig. 13. a) Calculated phase diagram for Al-Sc and for Al 4.6Mg-Sc (Insert: Dependency of Sc-solubility from Mg-content at 660°C); b) Phase diagram of the Al-Mg-O system (at%) at 1'227°C according to Harmelin [40]

Fig. 14. Simulated transversal (left) and longitudinal (right) cross-section of the melt-pool at a scan speed of 350 $^{mm}/_s$. The melt-pool boundary refers to 450°C where ≈5% of molten aluminium is remaining. Temperature isolines are at 700°C, 800°C, and at 500°C steps from 1'000°C on. The dashed line indicates T = 635°C. The transversal cross-section is taken at the position of the deepest melt-pool.

Fig. 15. Simulated cross-section of the melt-pool at a scan speed of v_s = 350 $^{mm}/_s$ at the deepest melt-pool position (a) and 0.1mm in the melt-pool back (b), showing Marangoni convection flows.

Fig. 16. Scheil-simulation of the Scalmalloy alloy system. $Al_3(Sc, Zr)$ precipitates go into solution at temperatures above 800°C. At 450°C only ≈5% of melt is remaining, which is defined as the melt-pool boarder

Table 2: Thermo-physical material properties

Property	Unit	T = 100°C	T = 635°C	T = 900°C	T = 1400°C
Specific Heat Capacity	$^{J}/_{mol\,K}$	25.6	31.6	constant 29.4	
Thermal conductivity	$^{W}/_{m\,K}$	129.5	86.9	98.6	117.2
Surface Tension	$^{N}/_{m}$	-	1064 @ 650°C	977	898
Dyn. Viscosity	Pa s	-	1.38	0.81	1.71