doi:10.1017/S1431927617004135
Microsc. Microanal. 23 (Suppl 1), 2017

In-process Precipitation During Laser Additive Manufacturing Investigated by Atom Probe Tomography

Philipp Kürnsteiner[1], Markus Benjamin Wilms[2], Andreas Weisheit[2], Pere Barriobero-Vila[3], Baptiste Gault[1], Eric Aimé Jägle[1] and Dierk Raabe[1]

[1.] Department Microstructure Physics and Alloy Design, Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany
[2.] Fraunhofer Institute for Laser Technology ILT, Aachen, Germany
[3.] Institute of Materials Science and Technology, Vienna University of Technology, Vienna, Austria

Laser Metal Deposition (LMD) is a specific Laser Additive Manufacturing (LAM) process in which a focused laser beam creates a melt pool in the component's surface. Metallic powder is then injected through a nozzle into the melt pool. As neighboring tracks and subsequent layers are deposited during the LMD process, already consolidated material experiences a cyclic re-heating. This intrinsic heat treatment (IHT) can be used to trigger the precipitation reaction in precipitation hardening alloys [1]. We used the rapid alloy prototyping capabilities of the LMD process to efficiently screen different alloy compositions by producing compositionally graded samples. In a simple model maraging steel containing 19at% Ni, we varied the Al concentration from 0 to 25at% to identify an alloy composition that responds well to the IHT of the LAM process to produce an in-process precipitation strengthened maraging steel. Due to its sub-nanometer resolution, Atom Probe Tomography (APT) provides compositional information at high-resolution and is therefore ideally suited for analyzing small clusters and precipitates. Additionally, we used High Energy Synchrotron X-Ray Diffraction (HEXRD) to provide crystallographic information with high sensitivity.

Samples graded in Al concentration for this study were produced by an LMD machine equipped with a diode laser and two powder containers that can be fed independently from each other. More detail can be found in [2]. APT specimens were prepared by the focused-ion beam lift-out process described in ref. [3] using a FEI Helios NanoLab 600i FIB/SEM. Site-specific specimens were prepared from different layers of the graded sample, representing different Al concentrations. APT experiments were performed in a Cameca LEAP 3000 X HR and a LEAP 5000 XS in voltage-pulsing mode using a pulse frequency of 200kHz and a pulse amplitude of either 15% or 20% of the applied voltage. To avoid the peak overlap of Al^{1+} with Fe^{2+}, temperature and evaporation rate were adjusted in such way that all Al ions were post ionized and only occurred in charge states of Al^{2+} and Al^{3+}. Target temperatures in the range of 60-70K and evaporation rates of 1-3% were used. 3D reconstruction was performed by IVAS version 3.6.14. For voxel-based analysis a spacing of 0.5nm and a delocalization of 2nm were used.

From microstructural observations of the graded sample, we concluded that the desired martensitic microstructure of a maraging steel could only be obtained with Al concentrations below 15at% [2]. Above this threshold a coarse grained ferritic microstructure was found. APT measurements were performed at three different Al concentrations below this threshold value. The corresponding radial distribution functions (RDF) are shown in Figure 1 (a). While at low Al concentration of 3.4at%, the solute atoms Ni and Al are distributed homogeneously, a higher Al concentrations of 8at% and 9at%, leads to pronounced clustering of Ni and Al. Figure 1 (b) shows an example of a reconstruction of an APT tip containing 8at% Al, highlighting the precipitates by an isococentration surface at 15at% Al. From the volume of the reconstruction and the number of isoconcentration surfaces found, the number density of precipitates was calculated to be $5x10^{24}$ particles/m^3 and $1.2x10^{25}$ particles/m^3 for 8 and 9at% Al, respectively. Looking at the proximity histogram in Figure 1 (c) it is apparent that the precipitates are enriched in Al and Ni but it is not possible to unambiguously decide whether they are NiAl or Ni_3Al, the two precipitate phases that have been reported in literature for similar alloy systems. Even if Fe is allowed to substitute Al, the ratio of Ni/(Fe+Al) is around 0.7 instead of 1 or 3 as would be expected for Ni(Al,Fe) or Ni_3(Al,Fe) respectively. Nevertheless it was possible to identify the precipitate phase via their different crystal structure (NiAl: B2, Ni3Al: $L1_2$) using High Energy X-Ray Diffraction (HEXRD) in the synchrotron to be NiAl. A Rietveld analysis showed 6vol% NiAl phase and a lattice mismatch of only 0.11%. The low lattice mismatch is associated with a high nucleation rate through a low nucleation barrier resulting from a low coherency strain. This fact can explain the exceptionally high number density of precipitates created by the IHT of the LAM process. Hardness measurements in the different layers of the graded sample showed a steep increase in hardness from 300HV to 530HV associated with the high number density of nanometer sized NiAl precipitates.

The combination of APT with HEXRD to obtain chemical information and structural information has proven to be a very powerful tool for the investigation of such high number densities of nanometer sized precipitates. This allowed to identify optimum Al concentrations at which the IHT of the LAM process causes high number densities of precipitates resulting in a substantial increase in hardness of the material. In a next step, the information gained through APT such as precipitate size and density will be used to develop and test a model for the precipitation kinetics during the strongly non-linear, peak like time-temperature program of the IHT. Fundamental understanding of the effect of the IHT on the final product will help to better exploit the capabilities of LAM processes [4].

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[4] This work was carried out in the scope of the AProLAM project, funded jointly by the Fraunhofer Society and the Max Planck Society in their strategic cooperation framework. The authors are grateful to Uwe Tezins and Andreas Sturm for their support to the FIB and APT facilities at MPIE.

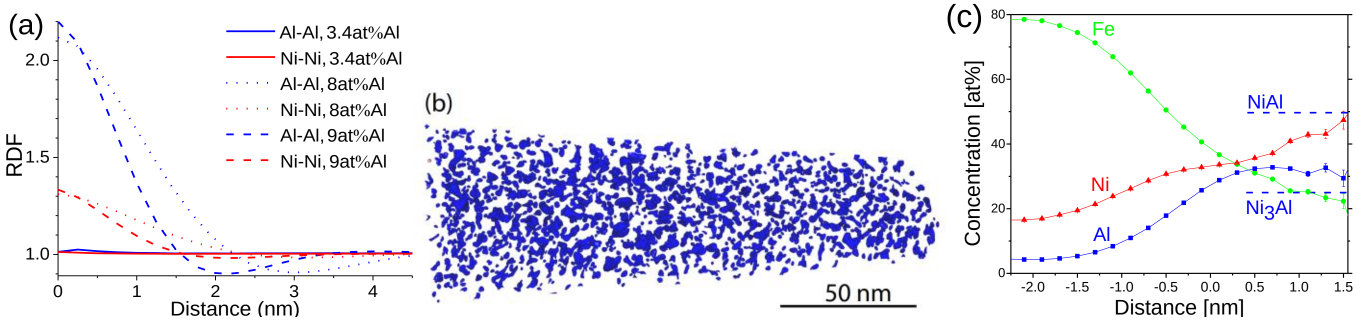

Figure 1. (a) Concentration normalized RDFs of APT measurements performed at three different Al concentrations: 3.4at% Al dotted lines, 8at% Al dashed lines and 9at% Al solid lines. Clear clustering can be observed at 8 and 9at% Al. **(b)** Precipitates visualized by drawing an isoconcentration surface at 20at% Al. **(c)** Proximity histogram corresponding to all isoconcentration surfaces seen in (b).