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Microstructure and Mechanical Properties of 3D Printed Tool Steel after Various Precipitation Hardening Treatments

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Precipitation hardening is one of the most frequently used methods of the post-processing heat treatments of 3D printed tool steel 1.2709, as it increases the yield and ultimate tensile strengths of the steel. Within this work, the broad temperature range of 250 °C – 550 °C was tested with five different holds at each temperature to study the effect of processing parameters on the microstructure and mechanical properties. Maraging tool steel 1.2709 was produced by selective laser melting (SLM) technology and obtained results were compared with conventionally produced steel of the same chemical composition. Mechanical properties were established by hardness measurement and tensile test and the microstructure was characterised mainly by light and scanning electron microscopy. The peak hardening was for both steels reached after 6 hours of precipitation at 500 °C, however, apparent hardening effects of heat treatment were observed already after low-temperature heat treatments.

1 Introduction

Production of the parts in a layer-by-layer way is usually called either rapid prototyping, rapid manufacturing, or additive manufacturing (AM). This approach is used in several technologies, among which belongs also powder bed AM, direct deposition or powder feed AM and wire-feed AM [1, 2]. In the field of metallic materials, the Selective laser melting (SLM) method is one of the most common methods of powder bed AM [1-3]. There has recently been a limited number of metals and alloys which are processed by SLM [3-5] and among steels, maraging steel 1.2709 is clearly the most commonly used [6-9]. It is an extremely low-carbon high-alloyed tool steel with a relatively soft martensitic matrix which can be strengthened by precipitation of intermetallic particles, such as Ni_3Mo, Ni_3Ti, Ni_3Al a Fe_2Mo [9-13]. Due to this microstructure, maraging steel offers a combination of high tensile strength and high fracture toughness. Unlike conventionally produced maraging steel, additively produced steel can also contain a small amount of retained austenite in the as-built condition [11, 12].

Heat treatment of steels additively manufactured by powder bed technologies plays an important role in the homogenization of the microstructure, decrease of residual stresses and optimization of mechanical properties for a particular application. In the case of maraging steel 1.2709, the most often used heat treatments are solution annealing or precipitation hardening [11-15]. Solution annealing results typically in reduction of yield and ultimate strengths and improvement of total elongation. Precipitation hardening can be on the other hand used to significantly increase the strength of the steel while reducing the elongation. Powder suppliers often provide heat treatment conditions recommended for the post-processing of the printed parts. For maraging steel 1.2709, the recommended treatment consists of a 6-hours hold at the temperature of 490 °C [13]. This is probably the reason why the majority of the works have been concentrated on the treatments at or just around this temperature and very little has been published about the development of microstructure and mechanical properties at other processing conditions [8-11, 15-16]. Therefore, this work offers an overview of the effect of various precipitation hardening conditions on the additively manufactured maraging steel, with a brief comparison to the conventionally prepared steel of the same chemical composition.

2 Material and methods

Two types of maraging steel 1.2709 were used in this work. First, the steel was additively manufactured by selective laser melting technology using an EOS M290 printer. The used powder was also supplied by the EOS company under the trade name MS1. Second, the bars of conventionally produced (cast and warm rolled) steel of the same chemical composition labelled VACO 180 were obtained from Bohdan Bolzano, s.r.o. Both steels had very low carbon contents of 0.001 – 0.003 wt.% and were heavily alloyed by 18 wt.% of nickel, 9 wt.% of cobalt and 5 wt.% of molybdenum. The steels further contained about 0.8 wt.% of the titanium (Tab. 1).

Tab. 19 *Chemical composition of used AM steel MS1 and conventional steel VACO 180 [weight %]*

	C	Si	Mn	Cr	Mo	Ni	Co	Ti	Al
MS1	0.001	0.02	0.02	-	4.9	17.7	8.7	0.8	0.06
Vaco 180	0.003	0.02	0.06	0.12	4.8	18.2	8.8	0.8	0.06

Additively manufactured samples were produced in a nitrogen atmosphere. Processing parameters standardly used for AM of maraging steels were applied, namely a laser power of 257 W with a scanning rate of 960 mm/s. Layers with a thickness of 40 μm were printed using hatch spacing of 110 μm. Cylindrical samples were produced with the longitudinal axe perpendicular to the building platform of the printing machine. The precipitation hardening was carried out at seven different temperatures in the range of 250 – 550 °C and five holds were tested for each precipitation temperature. The hold marked as "0" means that the sample was only heated at the temperature and once this temperature was reached in the whole volume of the sample, it was removed from the furnace. A thermocouple inserted into a hole drilled in the sample was used to determine the instantaneous temperature reached inside the sample during the treatment. Final cooling of all the samples to room temperature was performed in air.

Light and scanning electron microscopy were used to describe the microstructure of additively and conventionally manufactured samples. A BX 51 Olympus light microscope and a Crossbeam Auriga dual-beam microscope with EDS (energy-dispersive X-ray spectroscopy), BSE (backscattered electron) and EBSD (electron backscatter diffraction) detectors were used for microstructure characterisation. The samples for BSE and EBSD analysis were polished with OPS colloidal silica suspension and observed in as-polished condition, while 3 % Nital etching was used to reveal the microstructure for light and standard scanning electron microscopy. The purity of the AM steel was determined by image analysis of light micrographs. The plane fraction of defects was evaluated from 200 images taken in top and side cross-sections of polished and unetched samples. For the purpose of this evaluation, all dark areas at the bright polished surface of the steel were considered, as it is impossible to distinguish dark globular pores from dark globular inclusions by image analysis. The proportion of austenite (both retained and reversed) was established by X-ray diffraction phase analysis.

Mechanical properties were determined by hardness measurement HV10 and tensile test of sub-sized flat samples with a total length of 20 mm, gauge length of 5 mm and cross-section of 2 x 1.2 mm. Tensile samples were prepared by wire-cutting and plane grinding to the final thickness. All tensile test samples were produced along the building/rolling direction. Hardness measurements were performed in two perpendicular directions on each sample. It was top cross-section (parallel with building platform) and side cross-section (perpendicular to the building platform) for AM steel and transverse cross-section (perpendicular to rolling direction) and longitudinal cross-section (parallel to rolling direction) for conventionally produced steel. All the values given are the average of three hardness measurements.

3 Results and discussions

3.1 Microstructure analysis

The powder particles of the AM maraging steel were mainly globular (Fig. 1), as they were produced by gas atomisation. Image analysis showed that the average porosity of printed samples of AM steel reached 0.18 %. Higher plane fractions of defects were reached in the top cross-section than in the side cross-section. The cellular microstructure was obtained in the AM maraging steel with the mixture of very fine equiaxed cells and thin prolonged cells (Fig. 2). There has been recently some discussion among various workplaces about the presence of precipitates in as-built maraging steel. The very first researchers stated that the ongoing heat treatment of already printed layers during the subsequent deposition of the "upper" layers is responsible for the precipitation of Ni_3Ti particles during the printing process. However, later detailed atom probe tomography studies of the microstructure of as-build maraging steel didn't acknowledge the presence of these particles. In the case of the AM steel used in this experiment, numerous fine particles with the size in the range from several nanometres to tens of nanometres were found in BSE images of a polished sample in an as-built condition. This observation method was chosen based on previous experience that etching tends to obscure the presence of very fine particles even at very high magnifications. Determining the chemical composition of those small objects is often beyond the possibilities of SEM microscopes, so only the larger particles were successfully identified by EDS. They always contained an increased amount of Ti, often also of Al (Fig. 11) and in some cases, it was accompanied by increased O contents. This would suggest that at least some of the observed particles are in fact extremely fine metallurgical defects (oxidic inclusions) rather than precipitates. The X-ray diffraction phase analysis detected the presence of about 4 % of retained austenite, which forms very thin films or prolonged islands mainly at cell boundaries [12]. This is in agreement with the reports of various research teams claiming to have commonly among 3 and

6 of retained austenite in printed samples of maraging steel [1], [11], [15].

The microstructure of conventionally produced maraging steel VACO 180 consisted of lath martensite without retained austenite, as it was delivered in solution annealed condition. The coarsening of the microstructure during commercial annealing would explain why is conventionally produced steel significantly coarser than the AM steel in the as-built condition (Fig. 10). The very fine microstructure of the AM steel is generally attributed to the high thermal gradients during the AM processing [15].
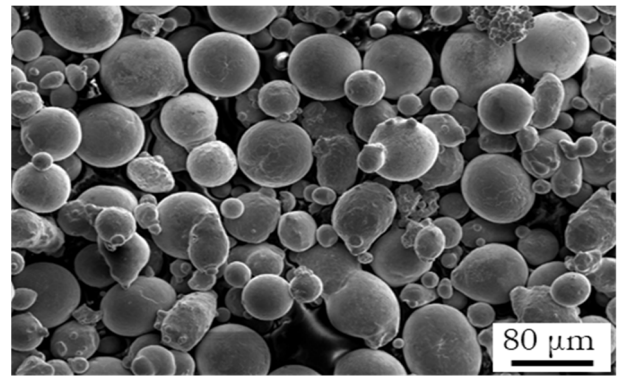
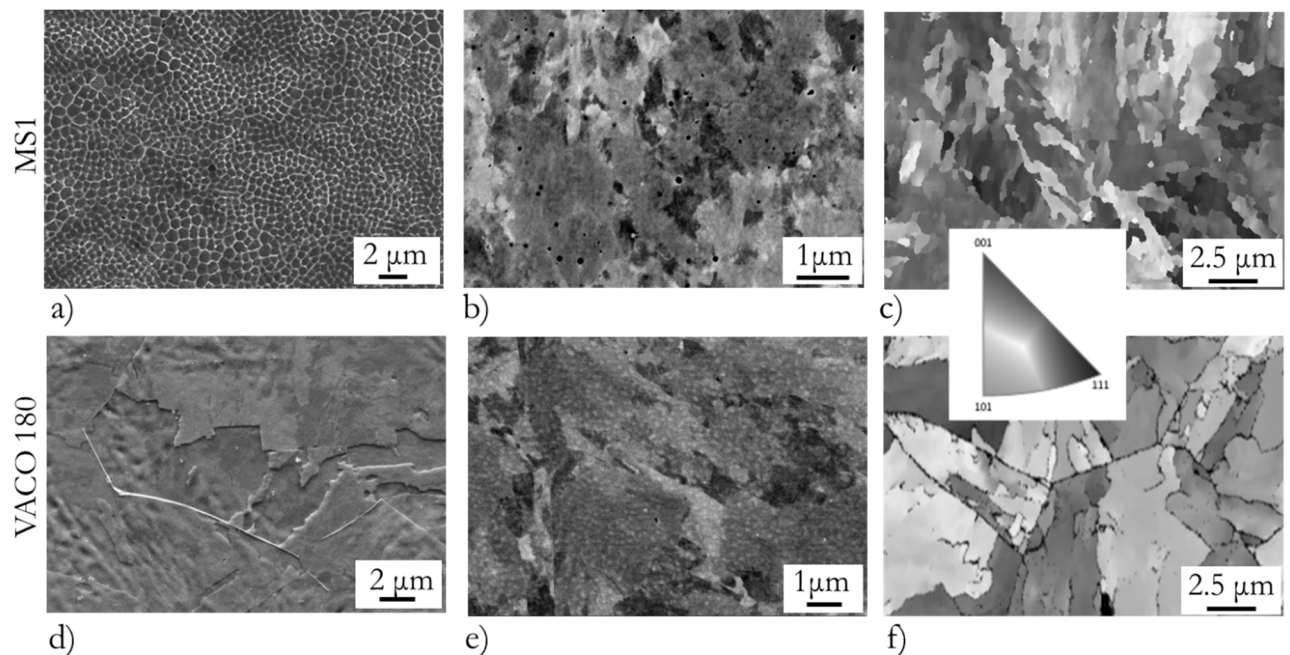**Fig. 9** *Detail of used powder of MS1 steel*

Fig. 10 *Microstructure of AM steel MS1 a),b) and conventionally manufactured steel VACO 180 c)d). SE images of etched microstructure a),c), BSE images of polished samples b),d)*

Even though heat treatment had a measurable effect on mechanical properties even at lower precipitation temperatures, the microstructure changes were not apparent from the microstructures of either steel up to the longer holds at the temperature of 500 °C (Fig. 12). The comparison of EBSD images shows that the AM steel retained significantly finer microstructure after the heat treatments. After 6 hours hold at 500 °C new very fine grains appear in both steel, which can be connected with reversed austenite transformation during the treatment. The volume fraction of austenite more than doubles for AM steel, reaching 9.6 % and 3.1 % of austenite was found in conventional steel. The increasing number of very fine precipitates can be observed from BSE images with increasing precipitation temperature (Fig. 13). In the first stage, the creation of new Ni_3Ti particles is expected. Those particles then develop at higher temperatures into more complex Ni_3 (Ti, Mo, Al) precipitates. In an over-aged stage, after 6-hours hold at 550 °C, the dissolution of

those particles resulted in higher concentrations of Ni in the surrounding matrix [11] which in turn facilitated further austenite reversion and the growth of a new type of needle-shaped particles with preferred orientation (Fig. 13 d). Those particles were also reported by Jägle et al. [17] who found them to create the networks in over-aged samples. There was around 32 % of austenite in AM steel and 13 % in conventional steel after 6 hours of precipitation at 550 °C. As the initial retained austenite fraction difference among AM and conventional steel before heat treatment was only 4 %, the values obtained after ageing treatment showed that austenite reversion was more intensive in AM samples. This is probably connected with a higher amount of Ni-rich precipitates in AM steel, whose dissolution might locally support austenite formation. Finer matrix could also contribute to this effect by providing numerous potential nucleation sites, as the majority of austenite was formed along the laths or cell boundaries.
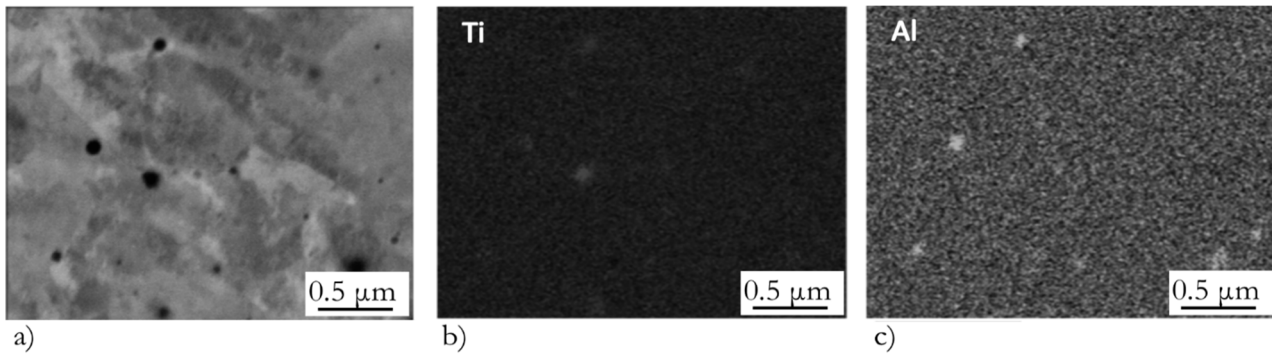
Fig. 11 *Characterisation of particles in the as-built microstructure of AM steel MS1*
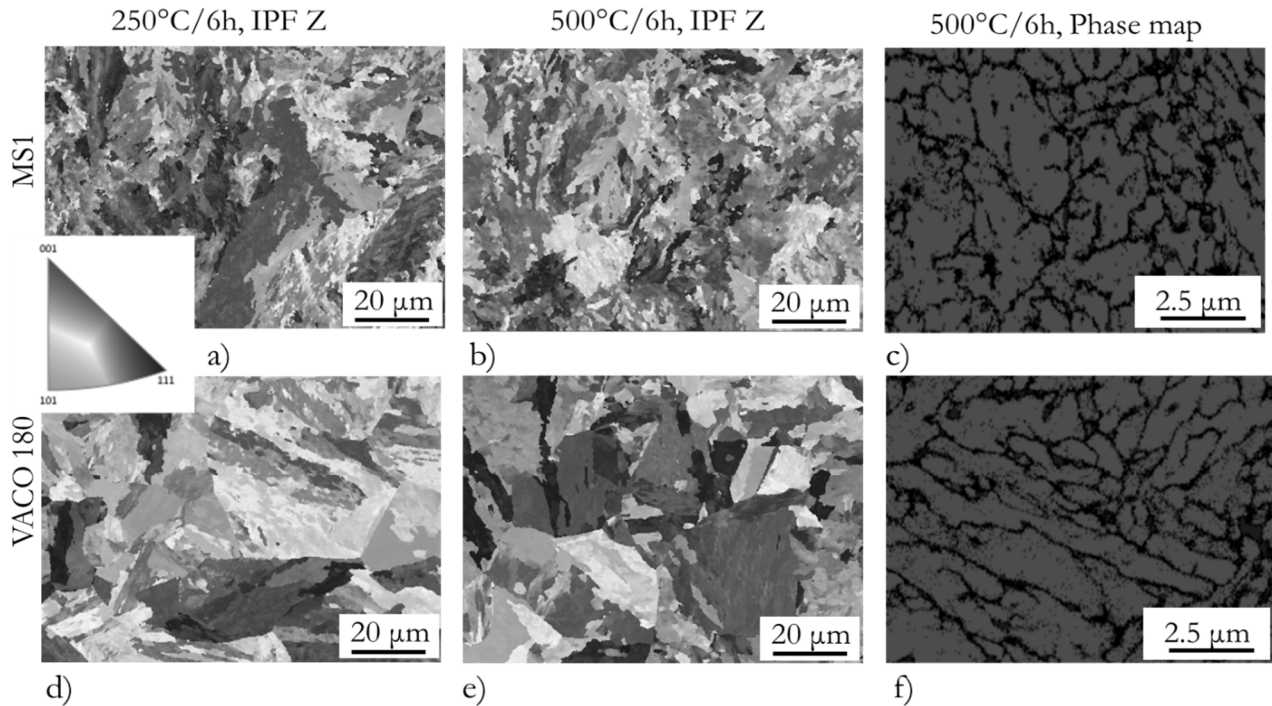

Fig. 12 *EBSD of AM steel MS1 (a-d) and conventionally produced steel VACO 180 (d-f). Pole figures in the Z direction (a,b,d,e), phase maps (c,f) with blue islands of austenite in a red matrix*
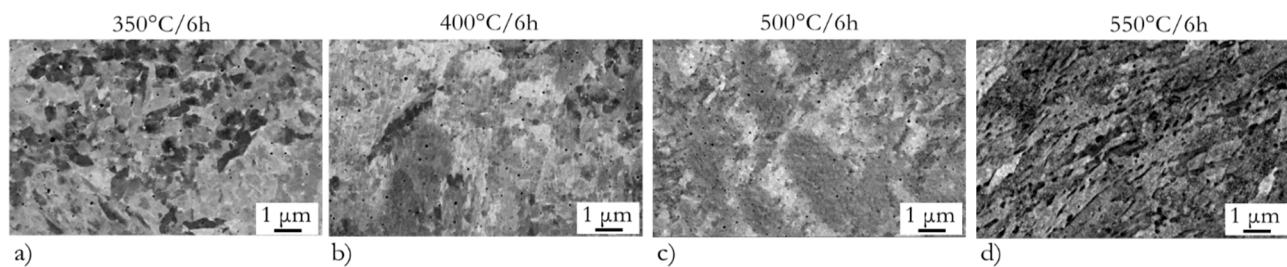

Fig. 13 *BSE images of microstructure of AM steel MS1 after various heat treatments: a) 350°C/6hours, b) 400°C/6hours, c) 500°C/6 hours, d) 550°C/6hours*

3.2 Mechanical properties

The development of mechanical properties of heat-treated maraging steel is summed up in Fig. 6, Fig. 7. The hardness of both steels was measured in two perpendicular directions, one of them corresponding to the building/rolling direction. Both steels show slight anisotropy of mechanical properties. As can be seen from Fig. 6, the average hardness of both steels was bigger along the building/rolling direction

than in the plane perpendicular to the building/rolling direction. This trend remained the same for the whole range of precipitation temperatures and holds. Hardness didn't change very much after low-temperature heat treatments at 250 °C and short holds at 300 °C. The first increase of hardness of both steels can be identified after 4 hours hold at 300 °C. Every higher precipitation temperature resulted in a further increase in hardness, and this increase was more noticeable for

each extension of the hold time. The peak hardness was obtained for AM steel MS1 after 4 hours of precipitation hardening at 500 °C. The very similar hardness of AM steel was however achieved also after 6 hours hold at 450 °C and for conventionally produced steel VACO 180 after 6 hours hardening at 500 °C. Both steels showed a decrease in hardness after heat treatments at 550 °C when over-ageing connected with more pronounced austenite reversion already occurred.

The development of strengths corresponded to the development of the hardness of both steels (Fig. 15). The only difference is that the hardness of the initial states (Without HT) did not change after heat treatment at 250 °C, while the strength of both steels increased already after short heating at 250 °C. The highest strength was reached for the same heat treatments as the highest hardness, 4 and 6 hours at 500 °C for AM steel and 4 hours at 550 °C for conventional steel. Total elongation values seemed to be rather less

sensitive to the various heat treatment conditions, but they generally tend to decrease with increasing strengths of the steels and only start to increase again after heat treatments at 550 °C. It can be assumed that increasing the volume fraction of the reversed austenite would cause decrees of strength and hardness, but it would on the other hand contribute to better ductility of the steels in the over-aged conditions.

When evaluating the mechanical properties, it must be taken into an account that the tensile tests were performed using sub-sized samples. This means that all the elongation values are overestimated compared to larger samples and small variations in elongation properties are harder to detect. This problem is related strictly to the sample size and not to additive manufacturing technology. For example, VACO 180 steel in as-delivered condition reached TE of 17 % when tested using cylindrical samples with 4 mm diameter and 20 mm gauge length in contrary to 23 % on sub-sized samples [16].
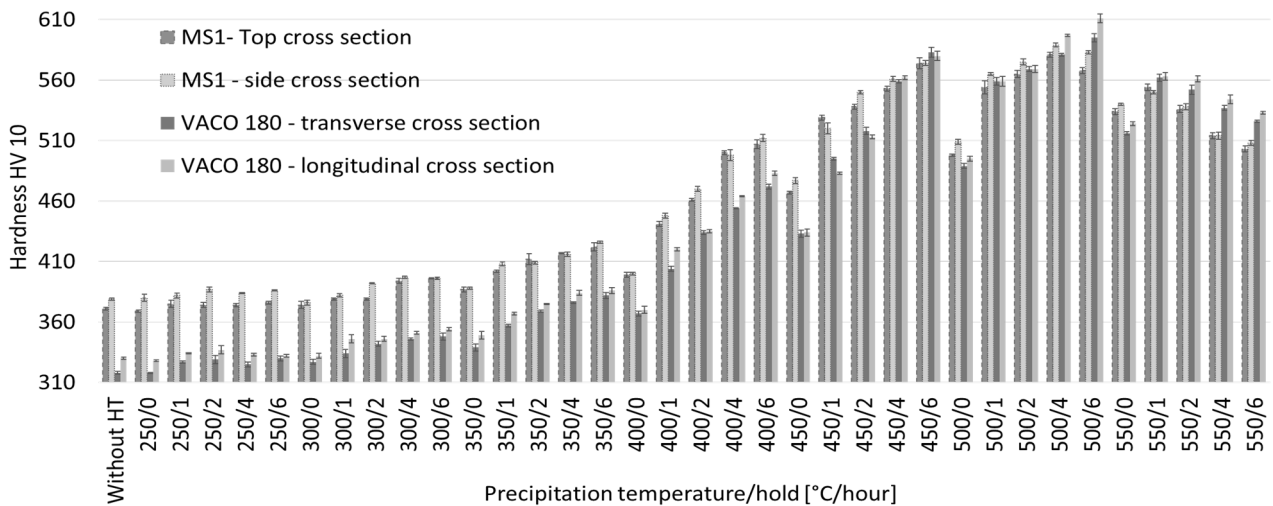

Fig. 14 *Hardness of AM steel MS1 and conventionally produced steel VACO 180 after heat treatments. As-built condition of AM and as-delivered condition of VACO 180 without further heat treatments are marked as "Without HT"*
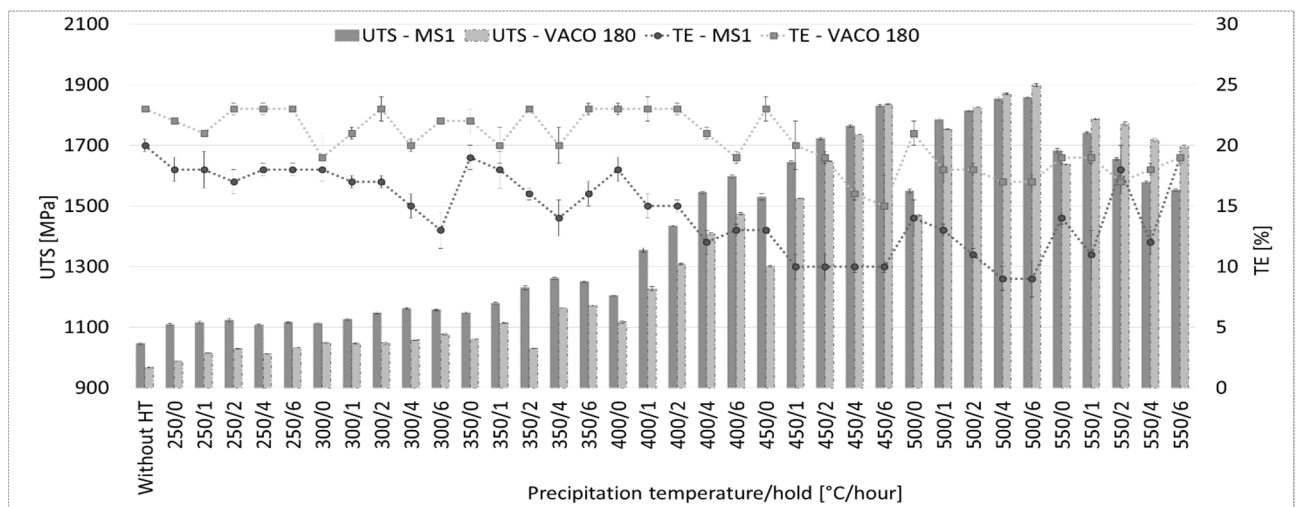

Fig. 15 *Ultimate tensile strength (UTS) and total elongation (TE) of AM steel MS1 and conventionally produced steel VACO 180 after heat treatments. As-build condition of AM and as-delivered condition of VACO 180 without further heat treatments are marked as "Without HT"*

Despite the differences in the microstructures of both steels in initial conditions and also after precipitation hardening, their mechanical properties follow the same trend. This reflects the earlier DSC measurements and detailed atom probe tomography analysis which both suggested that the precipitation process in AM and conventionally produced steel follows the same route [6]. The higher strengths and hardness of AM steel in the initial state and after precipitation hardening at lower temperatures should therefore be contributed mainly to the finer matrix of the AM steel, which remains finer for all precipitation temperatures used in this work. The equalization of the properties of both steels after longer holds at 450 °C and above could be caused by more intensive austenite reversion in AM steel. An interesting feature of the hardness and strength trends is a visible strengthening occurring already after 4-6 hours hold at the temperature of 350 °C. The strengthening of this maraging steel is generally connected with hardening temperatures above 400 °C and published articles are commonly interested in the microstructure development after precipitation hardening at temperatures around 450 °C. The results suggested the first stages of the strengthening process occur at lower temperatures, even though corresponding microstructure changes couldn't be observed by SEM. A dedicated TEM - based study of this early precipitation phenomenon is currently being prepared.

4 Conclusions

Even though the microstructure changes were not very visible from SEM micrographs, the strengthening of both maraging steels was occurring already after low-temperature heat treatments. Hardness and ultimate tensile strengths of AM steel increased from 371 HV10 and 1046 MPa in as-build AM steel to 422 HV10 and 1261 MPa after 6 hours of precipitation hardening at the temperature of 350 °C. The conventionally produced steel showed a hardness increase from 318 HV10 to 382 HV10 and strength increase from 968 MPa to 1171 MPa after the same precipitation treatment. The hardness and strength then gradually grew with increasing hardening temperatures and prolonging hold times until 6 hours hold the temperature of 500 °C, where AM steel reached the hardness of 568 HV10 and tensile strength of 1856 MPa and conventionally produced steel possessed hardness of 593 HV 10 and UTS of 1898 MPa. A further increase of precipitation temperature to 550 °C resulted in over-ageing, which was connected with the changes in type and size of precipitates and also a significant increase of volume fraction of reversed austenite to 32 % in AM steel and 13 % in conventional steel.

Additively manufactured maraging steel reached higher strengths and hardnesses than the conventional ones in the condition without heat treatment and also

after precipitation hardening until about 4 hours hold at the temperature of 500 °C. Longer hold at 500 °C and hardening at 550 °C resulted in the strength and hardness of conventional steel being higher than that of AM steel. The ultimate tensile strength of the conventional steel after 6 hours of hardening at 550 °C was higher by 144 MPa and the hardness by 23 HV10. This was caused by more intensive austenite reversion in AM steel during those heat treatments. The total elongation of conventional steel was generally slightly better (typically by 5-7 %) for all investigated conditions and only after precipitation hardening at 550 °C did the higher fraction of reverted austenite increase elongation values to the same level as in the conventionally produced steel.

Acknowledgement

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