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3D structural and atomic-scale analysis of lath martensite: Effect of the transformation sequence

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ABSTRACT

To improve the fundamental understanding of the multi-scale characteristics of martensitic microstructures and their micro-mechanical properties, a multi-probe methodology is developed and applied to low-carbon lath martensitic model alloys. The approach is based on the joint employment of electron channeling contrast imaging (ECCI), electron backscatter diffraction (EBSD), transmission electron microscopy (TEM), atom probe tomography (APT) and nanoindentation, in conjunction with high precision and large field-of-view 3D serial sectioning. This methodology enabled us to resolve (i) size variations of martensite sub-units, (ii) associated dislocation sub-structures, (iii) chemical heterogeneities, and (iv) the resulting local mechanical properties. The identified interrelated microstructure heterogeneity is discussed and related to the martensitic transformation sequence, which is proposed to intrinsically lead to formation of a nano-composite structure in low-carbon martensitic steels.

1. Introduction

This report aims at providing an improved fundamental understanding on the micro-mechanical response of lath martensitic microstructures. Lath martensite is of immense importance for structural alloys, since it is among the major strength-providing microstructural constituents in martensitic or multi-phase steels (e.g. dual phase steel, transformation-induced plasticity steel, complex phase steels, quench-partition steels, etc.). Despite its long history and use, efforts to better understand the microstructure development and the mechanical behavior of lath martensite are still ongoing. Here, we are specifically interested in martensitic constituent size variation effects which have been rarely investigated so far [1–3], but drastically influence e.g. the autotempering behavior [4] and toughness properties [5]. Effectively any analysis associated with lath martensitic microstructures is hindered due to the complexities arising from (i) crystallographic and (ii) compositional aspects of the underlying microstructure. In order to motivate the novel analysis strategy developed here, we first discuss these two challenges in the following two paragraphs.

Regarding martensite crystallography most pioneering works were based on transmission electron microscopy analyses [6,7]. TEM provides sufficient spatial resolution to resolve fine martensitic features (e.g. laths [6]), however, it provides only limited statistics of larger martensitic constituents (e.g. prior austenite grains) due to its limited field of view arising from the specimen and beam geometries. It is the development of the electron backscatter diffraction (EBSD) technique that enabled the systematic characterization of the hierarchical martensitic microstructure spanning multiple scales, i.e. ranging from prior austenite grains of hundreds of microns down to laths of tens of nanometers [8–11]. Yet, it is also clear that the standard 2D EBSD-based analysis provides a rather simplified representation of the lath martensite crystallography. For example, 3D EBSD and 3D FIB [12–15] analyses, as well as TEM observations [1,16,17] reveal significant heterogeneities in the size and morphology of martensite sub-units even within a single alloy, which cannot be fully captured by stand-alone 2D investigations. Also, even in optimized conditions, EBSD cannot resolve the fine details of the martensitic sub-structure.

Regarding martensite composition, similar progress was made due to the advances in another 'enabling' technique, namely, atom probe tomography (APT) [18–22]. Similar to EBSD providing wider access to martensite crystallography, APT triggered investigations of e.g. carbon (C) Cottrell atmospheres and segregation [23–29], precipitation reactions in martensite [30–32] and austenite layers in martensite [33,34]. Arguably the most critical among these is the analysis of C in martensite, since interstitial C plays one of the major roles in the properties of martensite [35–38]. A large number of recent APT based reports provide evidence of significant C distribution heterogeneity in martensite, which is taking place at a scale that was not accessible with conventional techniques (EDX,

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WDX, EELS, etc.). However, though only rarely commented on in the literature, this type of C variation does not occur homogenously throughout a given martensitic microstructure [23,39], hence, probing a sufficiently representative volume by APT is an issue. Further, APT has its own limitations, as in most cases it requires direct coupling to a diffraction based technique to identify the crystallographic nature of segregation zones [40,41]. Without correlative techniques helping to interpret APT data, analyses often include speculation on the origin of such chemical heterogeneities. One example is e.g. C enrichment in thin film austenite which is hard to distinguish from C segregation to lath boundaries with chemical mapping data only, i.e. without the aid from electron diffraction [33,39].

The complexity arising from the crystallographic and compositional heterogeneities inevitably affects the mechanical response of the material [42]. Recent micro-testing efforts successfully identified indications of these differences, e.g. by micro-tension [43] and micro-compression [44]. An elegant example is the discussion of the effectiveness of block vs. sub-block boundaries against slip transition through focused ion beam milled micro-beam bending experiments [45]. However, the hierarchy and the extreme fineness of the martensite microstructure require manufacturing of even smaller (sub-micron) sized samples for systematic investigations, imposing substantial experimental challenges due to FIB-induced damage, influence of geometric imperfections in micro-specimens, and alignment issues.

These rather fundamental challenges require a dedicated coupled approach that can 'simultaneously' (i) probe statistically-representative areas of coarser martensitic constituents (e.g. blocks, packets, prior austenite grains, etc.) in 3D, (ii) resolve fine martensitic constituents (laths, lath boundaries, dislocation densities, twins, etc.) in 3D, and couple these analyses to (iii) atomic resolution compositional mapping and (iv) high-resolution mechanical mapping. Such an approach enables unraveling of various aspects of martensite behavior, e.g. its 3D morphology, autotempering effects and strengthening contributions of the individual defects. The methodology presented here satisfies these requirements. The approach is based on electron channeling contrast imaging (ECCI), which successfully bridges the scale gap between EBSD and TEM in terms of resolution as well as field of view imaging [46,47]. We show that high resolution ECCI resolves smallest martensite sub-units and its internal defect structures such as dislocation networks and twins at a wide field of view [48–50]. Moreover, we demonstrate that it enables a direct coupling to diffraction information (by EBSD or TEM), 3D morphology (by serial sectioning) and chemistry (by APT) as well as local mechanical properties (by nanoindentation). Nanoindentation avoids the majority of the challenges mentioned above for FIB-based micro-testing approaches, and provides higher spatial resolution to probe the fine crystallographic and compositional heterogeneities mentioned above [51,52]. However, so far it was not related to size variations in lath martensite μ-constituents.

2. Experimental

The here developed multi-probe characterization approach was applied on a Fe–0.13C–5.1Ni–<0.002S–<0.002P model (wt.%) alloy, although several other martensitic steels were also characterized for partial comparisons. The Fe–C–Ni alloys, non-commercial grades provided by ArcelorMittal Research Center in Maizières, France, were austenitized at 900 °C for 5 min and subsequently quenched in water to obtain a fully martensitic microstructure. The experimental steps are schematically shown in Fig. 1.

The first step in our methodology is a large field-of-view EBSD measurement (Fig. 1). Then, four micro-indents (HV1) were made for depth-calibration during serial sectioning. We employ

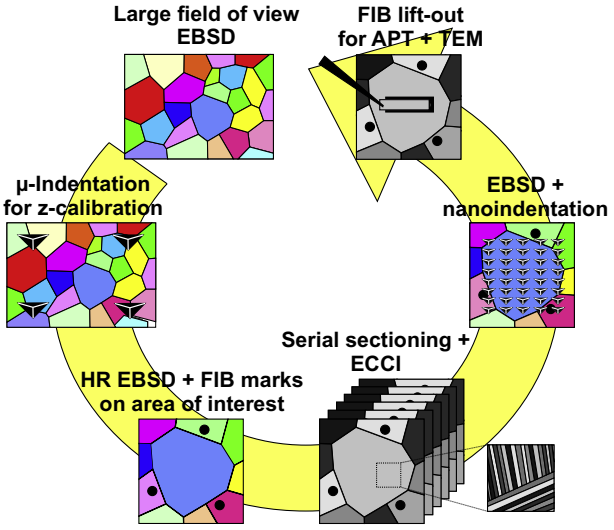

Fig. 1. The employed 3D multi-probe characterization approach that involves depth-resolved serial sectioning, ECCI, EBSD, nanoindentation, APT and TEM analyses. Nanoindents are significantly exaggerated for visual clarity.

depth-resolved colloidal silica chemo-mechanical polishing for sectional material removal. Compared to most other FIB-based serial sectioning approaches, the colloidal silica polishing approach provides larger field-of-view and avoids ion-beam knock-on damage [53]. The latter is especially critical for ECCI [46]. To inhibit image stacking errors, three surface-perpendicular fiducial holes were FIB-milled at the first step in the periphery of the area of interest. A HELIOS Nanolab 600i dual beam microscope was used for FIB milling. For each sectioning step the sample was subjected to a machine-controlled polishing routine. The spacing between each section was controlled through the change in the length of the diagonals of the indents made before polishing. For the data that will be presented next, after one calibration step of ~1.5 μm, the z-removal was kept constant to steps of ~500 nm. The serial sectioning was stopped after the collection of a 3D stack of eight sections and a total z-removal of ~4 μm, since sufficient data were collected. At each section, a chosen prior austenite grain was characterized by ECCI and when necessary by EBSD. ECCI and EBSD measurements with 30–50 nm step size were carried out using a Zeiss "Merlin" FEG-SEM operated at 30 kV and a Jeol JSM-6500F FEG-SEM operated at 15 kV, respectively. The EBSD data were used to reconstruct the prior austenite grains by means of a reconstruction software that functions based on the orientation relationship between austenite and martensite [54]. The investigated "prior austenite grain" was then subjected to a fine grid of nanoindents on the as-polished surface using a Hysitron TriboScope 900. A Berkovich indenter geometry was used for indentation with a constant force of 300 μN to create a 2 μm × 2 μm cell pattern of 209 indents in total. Additional ECCI and EBSD characterization of the indented grain allows for full coupling of the nanohardness distribution to the lath martensite hierarchical substructure.

These analyses enable well-informed sample location specification for extracting secondary specimens for atom probe tomography as well as for transmission electron microscopy. These samples were prepared through a site-specific FIB lift-out technique. The former was carried out using a Local Electrode Atom Probe (LEAP™ 3000X HR, CAMECA Instruments) maintained under ultrahigh vacuum conditions (~10^{-11} Torr) and operated in voltage and laser mode. The pulse fraction and pulse repetition rate were 15% and 200 kHz in the voltage mode, respectively. In laser mode, the laser energy was 0.4 nJ and the pulse repetition rate was 250 kHz. The tip temperature was maintained at 60 K. The