Neutron and X-ray diffraction study of residual and internal stress evolution in pearlitic steel during cold drawing

M Kriška\textsuperscript{1}, J Tacq\textsuperscript{1}, K Van Acker\textsuperscript{1}, M Seefeldt\textsuperscript{1}, S Van Petegem\textsuperscript{2}

\textsuperscript{1) Catholic University of Leuven, Department of Metallurgy and Materials Engineering, B-3001 Heverlee (Leuven), Belgium
\textsuperscript{2) Paul Scherrer Institute, NUM/ASQ, CH-5232 Villigen PSI, Switzerland
\texttt{Martin.Kriska@mtm.kuleuven.be}

Abstract. Neutron and X-ray diffraction were used to study the residual and internal stress evolution during cold drawing in pearlitic steel wires. A selection of high strength filaments drawn to different reductions has been investigated. In order to compare the evolution of macro and micro residual phase stresses in ferrite, the lattice strain evolution has been studied in axial and transverse direction. In-situ neutron diffraction tests in “Poisson” geometry have been carried out at the TOF strain scanner POLDI at PSI, Switzerland. These tests revealed a significant scatter in mechanical response among differently oriented ferrite grains, including a peculiar response of the \{200\} reflection, cp. [1, 2].

1. Introduction

High-carbon pearlitic steel wires are used in many applications that require exceptional tensile strength combined with acceptable ductility, which hence offer potential for materials and energy savings in various industrial applications. In terms of quantity, the most important are steel cords in automotive tire reinforcement, production of cables for suspension bridges and others [3-5]. The microstructural evolution of heavily drawn pearlitic steel used for high strength applications has been extensively studied with different experimental techniques. As widely discussed in literature, the pearlitic microstructure consisting of ferrite and cementite is changed drastically with severe plastic deformation and cementite starts to dissolve [6-8]. This cementite decomposition strongly affects the mechanical properties of the cold drawn wires and therefore is a challenging phenomenon during wire drawing to be well managed. However, while extensive studies on plastically induced cementite decomposition have been done with local high resolution techniques [6-8], an integral characterization method giving results representative of the bulk mechanical behaviour is still missing. In general, diffraction techniques were found to be promising techniques for these observations. Especially, in situ neutron diffraction has been a powerful tool to study internal stresses and hence the mechanical behaviour of pearlitic steels [1, 9, and 10]. Moreover, static neutron diffraction can be used to reveal both the residual phase microstresses arising as a consequence of the drawing process due to the different elastic and plastic properties of particular phases, and - in combination with XRD - the macrostresses [3, 10 and 11]. The importance of these stresses must be taken into account when considering the mechanical behaviour of carbon steel wires that are mechanically inhomogeneous. In general, their presence may influence dimensional stability and static strength, onset of micro cracks and delamination and finally work-hardening and strengthening mechanisms in pearlitic steel.
Correspondingly, the residual stress evolutions during the cold drawing process and the internal stress among differently oriented crystallites have been investigated by complementary use of neutron and x-ray diffraction and the result are discussed in the present study.

2. Experimental

2.1. Material

A selection of hypereutectoid steel wires prepared by cold drawing has been used for the investigation. The studied material has been prepared with standard wire route geometry (standard alignment of the drawing machine). Specimens of different drawing strains (true strain \((2\ln (d/d_0) - \varepsilon_t - 0.0 - 3.9)\)) were used for static neutron/x-ray diffraction and in-situ neutron diffraction experiments.

2.2. Neutron and X-ray diffraction

The time-of-flight (TOF) neutron diffraction (ND) method has been used for lattice strain measurement. The present static and in situ experiments have been carried out at the Paul Scherrer Institute’s spallation neutron source SINQ, on the instrument POLDI (Pulse-OverLap Dffractometer, designed mainly for strain-scanning experiments) [12]. The POLDI tensile rig with a load capacity of maximal 25 kN allows in situ measurements in horizontal and vertical position and thus measuring the lattice strain evolution both parallel and perpendicular to the tensile axis (axial and transverse strains). Correspondingly, static measurements on wires have been carried out in both (axial and transverse) directions. Further, in-situ neutron diffraction tests have been performed in transverse “Poisson” geometry. In general, the TOF-instrument has a definite advantage: it always yields peak shifts for more than a single reflection line [13]. Moreover, since neutron radiation can completely penetrate a bundle of wires and since the macrostress contribution in one phase has to balance out for equilibrium reasons, ND provide valuable information to check the presence of microstresses (stresses of the 2nd kind). In order to assess the residual macrostress evolution during cold drawing, X-ray diffraction measurements were done on the identical unloaded material on a Seifert MZ IV goniometer (by means of a “five point” measurement in the \(0 - 0.5 \sin \psi\) interval) at K.U. Leuven using a 40 kV Cr tube. Classical laboratory XRD with limited, shallow penetration offers in two phase material a possibility to determine the total phase stress. The measured total phase stress is then the sum of macro and microstresses.

Due to the low volume fraction and orthorhombic structure of cementite (spreading the scattering intensity into a large number of Bragg reflections), high transparency in comparison with ferrite [14] and the high level of deformation, only the ferrite phase was probed with both diffraction techniques. To convert the \(\{110\}\) lattice plane strains (measured by ND) into residual phase microstress in ferrite, and the high level of deformation, only the ferrite phase was probed with both diffraction techniques. To convert the \(\{110\}\) lattice plane strains (measured by ND) into residual phase microstress in ferrite, orientation dependent Diffraction Elastic Constants (DECs \(E_{110} = 220 \text{ GPa} \) and \(V_{110} = 0.28\)) were used [15]. Assuming a principal (micro) strain and stress state, i.e. negligible shear stresses, the axial stress component was calculated from generalized Hooke’s law as follows [16]:

\[
\sigma_{\text{Axial}} = \frac{E_{110}}{(1 + \nu_{110})(1 - 2\nu_{110})} \left[(1 - \nu_{110}) \varepsilon_{\text{AD}} + \nu_{110} \varepsilon_{\text{TD}}\right] \quad (1)
\]

where \(\varepsilon_{\text{AD}}\) and \(\varepsilon_{\text{TD}}\) are the axial and transverse lattice plane strains, respectively. The elastic strains were computed with a reference lattice spacing \(d_0\) measured on the heat patented wire in “Poisson” geometry using a long counting time. Similarly hoop/radial stresses were obtained (by equation. \((2)\)), taking into account a need of translation of the average lattice plane strains measured by ND in transverse direction which involves the radial and hoop contributions through \(\varepsilon_{\text{TD}} = \frac{\varepsilon_{\text{TR}} + \varepsilon_{\text{TP}}}{2}\):

\[
\sigma_{\text{Hoop/Radial}} = \frac{E_{110}}{(1 + \nu_{110})(1 - 2\nu_{110})} \left[(1 - \nu_{110}) \frac{1}{2} \varepsilon_{\text{TD}} + \nu_{110}\frac{1}{2} (\varepsilon_{\text{TD}} + \varepsilon_{\text{AD}})\right] \quad (2)
\]
3. Results and discussion

3.1. Ex-situ neutron and x-ray diffraction

The average lattice plane strains observed by ND for the individual reflections are shown for axial direction (AD) in figure 1 (a) and for transverse direction (TD) in figure 2 (a). Compressive strains were measured in AD for all depicted orientations. In TD, the lattice plane spacing patterns are more complex: tensile and compressive residual lattice plane strains were recorded at low ($\varepsilon_t = 0 - 0.6$) and at higher strain ($\varepsilon_t = 1.2$), respectively. At even larger strains, a strong tensile shift was observed for the {200} and {211} orientations, seen in figure 2 (a).

The evolution of the bulk average {110} lattice plane spacings in AD shows a monotonously decreasing tendency with increasing drawing strain. The same will thus apply to the average residual phase microstress in the {110} reflecting grains that are dominating in the arising {110} fibre texture. However, it turns out that at larger strains ($\varepsilon_t \geq 1.2$), the trend of the {211} lattice plane spacings evolves differently, namely shifting back towards less compression. The lattice plane strain evolutions of the individual reflections in TD (radial/hoop direction) are plotted in figure 2 (a). A good reproducibility of the measurements has been shown for the {110} and {211} oriented grains at a drawing strain of $\varepsilon_t = 0.6$ by successive measurements on three different samples of the same material (see the enclosed enlargement in figure 2 (a)). A similar lattice plane spacing evolution among differently oriented ferrite grains is found up to a true drawing strain of $\varepsilon_t = 1.2$, similar in terms of compressive and tensile strains in the range of $\varepsilon_t = 0 - 0.6$ and $\varepsilon_t = 0.6 - 1.2$, respectively. Beyond $\varepsilon_t \geq 1.2$, the lattice plane spacings start to diverge – comparable to what is observed for AD (see above) and during in-situ tensile tests on as-patented as well as deformed wire (see section 3.2).

The axial/hoop total stresses measured by XRD and an overview of the total, micro and calculated macrostress evolutions in the ferrite are shown in figure 1 (b) and (c) and 2 (b) and (c), respectively. Herein, the phase microstress has been calculated for the {110} reflecting grains, as described in section 2.2. In literature, the DECs $E_{110}$ and $E_{211}$ for ferritic and pearlitic steels have been reported to be similar for the {110} and {211} orientation families [14]. Own in-situ neutron diffraction tests on the present as-patented wires, however give significantly different DECs $E_{110}$ and $E_{211}$, see section 3.2. The residual phase microstresses, as calculated above, will thus strongly vary among the different grains at large strains. Macro residual stresses then should not be calculated from the difference of XRD measured total lattice plane strains probing {211} oriented grains and ND measured micro phase lattice plane strains probing {110} oriented grains. The plotted total and macro stresses are thus not reliable beyond $\varepsilon_t = 1.2$. The corresponding strong intergranular stresses evolving at large strains might explain the remarkable total ferrite stress behaviour at large strains, see below.

In the total ferrite stress plotted in figure 1 (b), a low and a high deformation regime of residual stress evolution during cold drawing can be distinguished. They are characterized by a monotonous and a rapidly changing behaviour, respectively. The ferrite stress is compressive in the early passes of the low deformation regime, possibly due to the total stress being dominated by the microstress evolution as long as the cementite does not deform plastically. This interpretation of the initial dip in terms of the microstresses matches with the evolution of the full width at half maximum (FWHM) from the neutron and the X-ray diffraction peaks. It is well known that the broadening is primarily the result of two related phenomena, namely a reduction of the “crystallite size” (coherently diffracting domain size) and an increase in the range of microstrains, related, among others to the dislocation density [9, 17]. As one can see in figure 3 (b), it is after the first three passes that the FWHM takes a turn to a much lower slope - meaning that the accumulation of microstrain or the storage of excess dislocations is slowing down. Excess dislocations would pile up at the ferrite-cementite phase boundary as long as the cementite does not deform plastically. However, this trend of the total ferrite stress evolution is suddenly changed and gradually turns into tensile values. This matches with the traditional theory for residual stress evolution in drawn wires [18] where two distinct types of residual stress patterns (compressive or tensile) are distinguished for drawn wires, depending on the amount of reduction. For reductions per pass of less than about 1 percent (coupled with a deformation localized
in a surface layer) the axial (macro) residual stresses are compressive at the surface (and then tensile in the centre). In case of larger reductions corresponding with industrial and also the present drawing treatment, the additional axial residual stresses introduced per pass are tensile at the surface of the wire. A microstress saturation at high deformation, as the evolution of the \{110\} reflections seems to suggest, would confirm that macro residual stresses are decisive for the total stress evolution at the surface at intermediate and large strains.

Figure 1. (a) Ferrite lattice plane spacings determined by ND in axial direction; (b) the axial total ferrite stress evolution by XRD and (c) overview of the total, micro and calculated macrostress evolutions as functions of drawing strain during cold drawing. The error bars reflect the uncertainties of the peak fitting procedure. In (a), displayed ferrite 110 lattice plane strain dependence indicating possible microstress saturation fits with value of accumulated residual ferrite strain after cold drawing to \( \varepsilon_t = 1.4 \) as reported by Tomota et al. [9].

If the macrostress indeed dominated the total phase stress evolution in the high deformation regime, one would also expect a corresponding monotonous evolution of the recorded total ferrite (tensile) stress. However, this is inconsistent with the observed rapid fluctuations in the present total stress evolution. The physical reason for these remarkable “oscillatory” behaviour at large strains is still unclear. In the static ND measurements, clearly for TD, and, as far as the signals are there, also for AD, two distinct regimes are observed as well, an early one where the different reflecting orientations are behaving similarly, and a late one where they are diverging. However, since all observed reflections still evolve monotonously, increasing intergranular stresses alone would not give rapid fluctuations in micro phase and total stresses. Therefore, sudden microstructural changes have to be taken into account as well, for instance loss of integrity, rupture or decomposition of the present cementite phase, as observed at large true strains (\( \varepsilon_t > 2 \)) related to relaxation of interface energy and interface stresses [19, 21 and 22].
Figure 2. (a) Ferrite lattice plane spacings determined by ND in transverse direction, the enclosed enlargement shows repeated measurements for $\varepsilon_t = 0.6$; (b) the hoop total ferrite stress evolution by XRD and (c) overview of the total, micro and calculated macrostress evolutions as functions of drawing strain during cold drawing.

Figure 3. The FWHM determined by XRD (a) and ND (b). In (a), the recorded FWHM evolution corresponds to the experimentally observed significant increase of dislocation density (at large true strain, $\varepsilon_t > 3$) with further drawing [23].

3.2. In-situ neutron diffraction study
Figure 4 shows the lattice plane strain in transverse direction for different \{hkl\} ferrite grain families as a function of an external stress during in situ tensile loading. The measurement has been performed on as-patented wire (with a lamellar cementite phase). As one can see, a significant variation in the mechanical response is found among differently oriented ferrite grains, including a peculiar response of the \{200\} reflection, cp. [1, 2, 24]. The present results show a discrepancy with the behavior of high carbon steel studied by Oliver et al. [1] and Kanie et al. [25]. In the fully elastic regime, individual
crystallites within the probed sample exhibit a stiffness corresponding to their inherent anisotropy [26] and yield in the following order: \{200\}, \{310\}, \{211\} and \{110\}. In contrast to [1] and [23], the compliant and (on average) soft \{200\} orientations here do yield much earlier than the others (regime A), and they undergo an even more dramatic tensile shift (regime B) that even changes the character of the internal stresses in these grains from compressive to tensile. In case of [1], this variation in the yielding might be due to the difference in the morphology of the hard phase, the cementite being globular there and lamellar here. Further, similar to behavior reported for dual phase steel [3], all probed orientations show a second turning point (regime C). As comes out from figure 4, all probed orientations are yielding clearly before these second turning points. Therefore, these turning points might indicate that a non-probed component of the microstructure, i.e. the cementite phase, is getting plastic.

![Figure 4](image-url)  
**Figure 4.** Ferrite lattice plane strain - stress responses of individual reflections for as-patented wire in transverse direction during an in situ tensile test in ND.

4. Conclusions
A complementary ex/in-situ neutron and X-ray diffraction study of residual and internal stress evolution in pearlitic steel during cold drawing was presented. The current XRD results indicate that in general, the total residual stress evolution during severe cold drawing can be described in terms of two distinct regimes. These are low and high deformation regime characterized by monotonous and rapidly fluctuating behavior, respectively. It was shown that the macrostress dominated the total residual stress evolution at low strain ($\varepsilon_t = 0.6 - 1.8$). Moreover, ex situ neutron diffraction has pointed to a strong orientation dependence of the lattice spacing evolution beyond a true strain of $\varepsilon_t \geq 1.2$. Finally, in situ neutron diffraction has – in contrast to some results reported earlier in literature – revealed strong differences in yielding and stress partitioning among particular \{hkl\} reflections.

**Acknowledgements**  
The authors gratefully acknowledge support the IAP program of BELSPO, project no. P6/24, and from NV Bekaert SA.
References
[1] Oliver EC, Daymond MR and Withers PJ 2004 Acta Materialia 52 1937
[2] Withers PJ 2007 C.R. Physique 8 806
[3] Seefeldt M, Dillien S and Stuhr U 2011 Materials Science Forum 31
[4] Elices M 2004 J. Mater. Sci. 39 3889
[5] Li L and Virta J 2011 Materials Science and Technology 27 845
[6] Gavril'yu VG 2003 Materials Science and Engineering A 345 81
[7] Sauvage X, Copreaux J, Danoix F and Blavette D 2000 Philosophical Magazine A 80 781
[8] Borchers Ch, Al-Kassab T, Goto S and Kirchheim R 2009 Materials Science and Engineering A 502 131
[9] Tomota Y, Lukáš P, Neov D, Harjo S and Abe YR 2003 Acta Materialia 51 805
[10] Seefeldt M, Walentek A, Van Houtte P, Vrána M and Lukáš P 2006 Materials Science Forum 524-525 375
[11] Van Acker K, Root J, Van Houtte P and Aernoudt E 1996 Acta Materialia 44 4039
[12] Stuhr U, Egger J, Hofer A, Rasmussen P, Graf D, Bollhalder A, Schild M, Bauer G and Wagner W 2005 Nuclear Instruments and Methods in Physics Research A 545 330
[13] Withers, PJ Johnson MW and Wright JS 2000 Physica B 292 273
[14] Wilson D and Konnan Y 1964 Acta Metallurgica 12 617
[15] Eigenmann B and Macherauch E 1999 Mat.-wiss. U. Werkstofftech 27 426
[16] Ezeilo AN and Webster GA 1999 Textures and Microstructures 33 151
[17] Hauk V 1997 Structural and Residual Stress Analysis by Nondestructive Methods Amsterdam Elsevier Science B.V
[18] Dieter GE 1988 Mechanical Metallurgy London McGraw-Hill
[19] Zelin M 2002 Acta Materialia 50 4431
[20] Zhang X, Godfrey A, Hansen N, Huang X and Qing Liu W 2010 Materials Characterization 61 65
[21] Umemoto M, Todaka Y and Tsuchiya K 2003 Materials Science Forum 426-432 859
[22] Zolotorevsky NY, Titovets YF and Vasiliev DM 2004 Rev.Adv.Mater.Sci. 7 91
[23] Shiratori T, Shiota Y, Ryufuku S, Adachi Y, Suzuki Y and Tomota Y 2006 The 3rd International conference on Advanced Structural Steels Gyeongju Korea
[24] Sevillano JG, Alkorta J, González D, Van Petegem S, Stuhr U and Van Swygenhoven H 2008 Advanced Engineering Materials 10 951
[25] Kanie A, Tomota Y, Torii S and Kamiyama T 2004 ISIJ International 44 1952
[26] Dye D, Stone HJ and Reed RC 2001 Current Opinions in Solid State and Materials Science 5 31