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Microstructural Characteristics and Strengthening Mechanisms in a Polycrystalline Ni-Based Superalloy under Deep Cold Rolling

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Abstract

Surface modification is an essential process route to improve the fatigue performance of aerospace components. Microstructural evolution in Ni-based superalloy Udimet720Li processed by deep cold rolling (DCR) was investigated experimentally using X-Ray diffraction, electron back-scattered diffraction (EBSD) and transmission electron microscopy (TEM). Deep cold rolling produces hardened surface due a range of microstructural changes associated with grain refinement, low angle grain boundaries (LAGBs) formation, and pile-up of dislocations around γ' precipitates and across twin boundaries. The defect structures within the deformed subsurface comprised of equiaxed and elongated dislocation cells at grain boundaries, mutual interactions slip bands, slip bands- γ' precipitate interaction at grain boundaries and multi-variant modes of twinning. The plastic deformation is predominantly driven though slip and dislocation multiplication mechanism rather than deformation twinning. These fundamental understanding on DCR process-microstructure-property could provide a deep insight into these alloys performance.

Keywords: Ni-based superalloys, Deep cold rolling, Dislocation cells, Microstructure, Electron back-scattered diffraction (EBSD), Transmission electron microscopy (TEM)

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1 Introduction

Ni-based superalloys typically comprise of an FCC γ matrix and cuboidal L1₂ γ' precipitates which play a vital role in strengthening, thermal stability and fatigue crack resistance behavior, mostly employed in the combustion section of the aero-engines [1]. To enhance the engine performance in severe operating conditions, i.e. under cyclic mechanical loading at high temperatures, a good combination of strength, ductility, creep and fatigue crack resistance is essential [2]. Surface mechanical treatments such as shot peening (SP) [3] and laser shock peening (LSP) [4,5] are being used to improve the strength and fatigue crack resistance without remarkably sacrificing ductility by inducing compressive residual stresses and surface hardening effects. Strengthening is primarily driven by microstructural features such as dislocations, twins and slip band activities. The resistance provided by internal barriers such as grain boundaries and γ' precipitates are enough to induce substantial strengthening. However, dislocation pile-ups at grain boundaries could lead to high intensity stress concentrations mostly in planar slip materials [6]. Further, the forward stress developed by the interaction of slip band-grain boundary could generate phenomena like slip transfer [7], deformation twin formation [8] and fatigue crack initiation [9] in the material.

Laine et al. [10] conducted a study on Ti-6Al-4V which shows the dependence of deformation mechanism and microstructural evolution on process induced strain-rate. For example, at low strain rate (10^{-4} - 10^{-1} s⁻¹) of quasi-static deformation generates planar shear bands, high strain rate of the order of 10^{-1} - 10^4 s⁻¹ as in SP shows dislocation tangles and deformation twinning, and at very high strain rates as experienced in LSP (10^4 - 10^8 s⁻¹) leads to planar dislocations and sub-grain formation. Lu et al. [11] reported multi-direction twin interactions at microscale and secondary twin-dislocation wall interaction at nanometre scale on laser peened commercially pure (CP) titanium. Further, novel microstructural features such as layer slip bands, micro-twin collision, dislocation cells were observed and correlated with number of laser impacts. In another

study by Lu et al. [12] on laser peened ANSI 304 stainless steel, it was found that multidirectional mechanical twin-twin interaction led to formation of grain subdivision at the top surface and submicron triangular blocks after three laser impacts were noticed. A grain refinement mechanism during the plastic deformation is proposed based on the formation of planar dislocation arrays, stacking faults along multiple directions due to dislocation pile-ups, formation of submicron triangular blocks, transformation of mechanical twin into sub-grain boundaries and continuous dynamic recrystallization of sub-grain boundaries.

Deep cold rolling (DCR) is a relatively new, neat and cost-effective technique to induce surface compressive residual stresses (CRS). Some findings by Altenberger et al. [13] on deep rolling (DR) process, similar process to deep cold rolling (DCR), was presented on Ti-6Al-4V which shows complex lamellar substructure, dense dislocation network and 1-2 μm layer thickness of 50 nm crystalline size. Recently we presented preliminary work on the effect of DCR process parameters on the CRS and cold-work on Ni-based superalloys RR1000, IN100 and Udimet720Li [14,15]. However, there is a limited understanding on the strengthening mechanism and evolution of defects such as dislocations and slip bands at the microstructural scale during severe plastic deformation of Ni-based superalloys. Thus, we believe, it would be of interest to explore the improvements in mechanical properties, the associated strengthening mechanisms and subsequent analysis of the microstructural modifications that occur as a result of deep cold rolling – this has been performed using the Udimet720Li superalloy.

In this paper, we firstly look at the extent of sub-surface compressive residual stresses with increasing applied rolling strain. In this context, a comprehensive study has been performed on the effects of hydrostatic pressure by deep cold rolling process on the beneficial compressive residual stresses and micro-hardness distribution. We then further investigate the strengthening mechanisms induced by the DCR process in detail using various microstructural parameters such

as the grain size, grain boundary distribution and intragranular deformation obtained from electron backscattered diffraction (EBSD). The generation of dislocations and slip bands mechanism are studied using transmission electron microscopy (TEM) and discussed in the light of applied strain and parent microstructure. A detailed multi-scale microstructural investigation provides a direct and accurate visualization of the dislocations and slip band activities and build up an improved understanding of the processing-structure-property relationship in DCR.

2 Experimental Materials and Methods

This section details the deep cold rolling technique applied on Udimet720Li polycrystalline superalloy and experimental techniques used to characterize of residual stresses, depth of plastically strained region, microstructural features, and entailing mechanical properties following the surface modification.

2.1 Material Preparation

A fine-grained (1-30 μm) polycrystalline Udimet720Li material was selected for this experimental work. The material was produced by hot forging process followed by hot rolling and provided by Rolls-Royce® in the disc form. The samples for the DCR trials were prepared after slicing by electric discharge machining (EDM) and polished sequentially using SiC grit paper, diamond paste, and finally with 0.04 μm colloidal silica suspension. Specimens were heat treated in a vacuum furnace at 760 °C for 8 h to relieve residual stresses in the material arising due to the machining and polishing process. The chemical composition (wt %) of Udimet720Li measured through electron dispersive spectroscopy (EDS) technique was listed in Table 1 and has predominantly Co and Cr as alloying elements.

Table 1: Chemical composition of Udimet720Li in wt%

Cr	Co	Mo	W	Al	Ti	Zr	B	C	Ni
16	14.7	3.0	1.25	2.5	5.0	0.03	0.015	0.010	Bal.

2.2 Deep Cold Rolling (DCR) Process

A hydrostatically controlled tungsten carbide ball of 6 mm diameter (HG6, ECOROLL AG) which is free to rotate within its holder is used against the sample surface. A hydrostatic tool was used at three different hydraulic pressures (10, 30, 50 MPa) which spanned the minimum to maximum pressure range capability of the test setup and pump. The linear feed (speed) and step-over distance between the passes were set at 1 m/min and 0.06 mm, respectively. The DCR tool was connected to the end-effector of an ABB® robot with 6 degrees of freedom and maintain constant step-over as shown in Fig. 1(a), (b).

Fig. 1: Schematic diagram of (a) DCR experimental set-up (3D view) and (b) Tool path (2D view)

2.3 Residual Stress Measurement and Microstructural Characterisation

Induced surface and sub-surface compressive residual stresses were measured by the Xstress G3 diffractometer, Stresstech®, which is based on the d vs $\sin^2\Psi$ method. The (311) crystallographic plane positioned at a 2θ angle of 156° using manganese radiation of 2.1031 Å wavelength was selected which represents the bulk properties of the γ/γ' structure [16]. The

elastic modulus (E) and Poisson's ratio (ν) of Udimet720Li used are 208 GPa and 0.31 respectively [17]. A plot of $\sin^2\Psi$ as a function of interplaner spacing (d) is obtained by varying the Ψ from -36° to 39° and fitted using linear regression function to quantify the residual stresses. The error bars presented for stress values are associated with the curve fitting error and no additional corrections were made. For the depth profiling, Struers Lectropol-5 system was used for electrolytic polishing using a mixture of 90% methanol by vol. and 10% by vol. perchloric acid. A specially designed mask was used to control the flow of etchant and successive layers were removed which is controlled by applied voltage (25 V) and exposure duration. The depth of layer removed is validated by each step using stylus profilometer (Talysurf®). It is to be noted that FWHM is calculated using diffraction peak fitting by Pseudo-Voigt function and instrumental broadening is corrected as follows [18]:

$$w^2 = (w_{total} - w_i) \sqrt{w_{total}^2 - w_i^2} \quad (1)$$

where, w is the sample peak broadening due to crystallite size and lattice strain, w_i is the instrumental broadening and w_{total} is the combination of sample and instrumental peak broadening.

Surface and cross-sectional micro-hardness were measured using the Qualitest QV-10000 DM hardness machine. A Vickers diamond pyramid indenter was used to apply 200 g load over a dwell time of 10 s. For the cross-sectional measurements, the indents were made approximately 50 μm distance from each other and in keeping with the ASTM E92 standard. Three repeat measurements were performed to estimate the mean micro-hardness value and standard error.

The EBSD characterization and analysis were performed using Oxford Instruments® detector integrated in a FESEM JEOL-JSM 7600F SEM and Aztec software respectively. All the samples of 5 mm x 5 mm dimension with 3 mm thickness were fine polished using diamond paste (refer Child et al. [3] work for the detailed procedure). Electrolytic polishing for 5 s through Struers

instrument was used as the final step for EBSD scan for better indexing of Kikuchi pattern. EBSD scan was carried out at a step size of $0.3\ \mu\text{m}$ over an area of $500\ \mu\text{m} \times 600\ \mu\text{m}$ generating 3×10^8 data points. HKL Tango and Salsa maps were used for the analysis of GOS, grain size distribution, interface boundary and micro-texture. Equivalent circle method was used for the grain size measurement and border grains and special boundaries such as twins were excluded from the analysis. For the grain boundary quantification, $1\text{-}15^\circ$ angular misorientation across the boundary, as obtained from the analysis software, is considered as low angle grain boundaries (LAGBs) and more than 15° considered as high angle grain boundaries (HAGBs). At least two different EBSD scans were considered for the quantitative analysis of the grain size and grain boundary data.

Grain orientation spread (GOS) values extracted from the large area EBSD map are relatively higher for the strained grains when compared to unstrained regions due to the lattice rotation [2]. The GOS values are plotted as the function of depth from the treated surface: a decrement trend from the surface to sub-surface is observed which is fitted using fourth order polynomial function. The depth of plastic strained region is defined where the slope of the function becomes zero *i.e.* first order derivative of a function is zero.

TEM characterization was performed using a JEOL 2010F electron microscope. Samples were prepared with extreme care to avoid any deformation induced by grinding or polishing process. In the initial step, the sample was polished from the opposite side, keeping the rolled surface intact, using fine emery paper upto the thickness of few microns and then punched to obtain a 3 mm diameter foil. Later, the small foil was polished using dimple grinder and ion milled by Gatan® Precision Ion Polishing System at variable beam angles to achieve the electron transparent foil.

3 Results and Discussion

In this section, the experimental measurements of compressive residual stresses, plastic strained region distribution analysis and microstructural features are presented.

3.1 Compressive Residual Stress Distribution

The residual stresses are within 150 MPa in the untreated material which was induced during the manufacturing process and surface grinding process. Residual stress distribution following DCR has been characterized along the depth in both longitudinal and transverse directions. After rolling at 50 MPa hydrostatic pressure, the maximum surface and sub-surface residual stress are increased to ~866 MPa and ~1315 MPa with the effective compression depth of 1 *mm* in the transverse direction. This value is comparable to the 1100 MPa CRS reported by Nagarajan et al. [14] for the DCR of nickel alloy RR1000 at 40 MPa hydrostatic pressure. The deeper compression depth could enhance the fatigue crack resistance of the material and hence could be beneficial for the fatigue performance. Interestingly, there could be a stress relaxation during the elevated temperature operating conditions through microstructural recovery (softening) processes including defect annihilation and the overall fatigue performance might be compromised in the practical applications [19]. Hence, having a deeper depth of CRS influence could be advantageous.

Fig. 2: Residual stress distribution as a function of depth in (a) longitudinal and (b) Transverse direction

It is to be noted that surface residual stresses, peak residual stresses, peak stress position from the surface, area under compression, and depth of influence are the parameters to evaluate the residual stress distribution. It can be seen from Fig. 2 that all the above-mentioned parameters increase progressively with the DCR hydrostatic pressure. The surface and peak residual stresses are increased predominantly due to the higher plastic strain. The residual stress distribution shows a peak value on the subsurface and peak shift to greater depths with an increase in applied roller pressure. The peak shifted from $75 \mu\text{m}$ to $230 \mu\text{m}$ in the longitudinal direction in the DCR process. It could be explained based on the Hertzian contact pressure mechanism; maximum equivalent stresses lie below the surface and changes according to the applied hydrostatic pressure and contact geometry i.e. size and shape of the contact surfaces [20]. It can be observed that transverse direction shows higher surface stress values than longitudinal direction by 110-830% range which is consistent with the other researchers' findings [21–23]. It could be due to the bow wave generated in advance of rolling direction and relatively more material is pushed in transverse direction than longitudinal [24]. Also, this could be due to the atomic rearrangement in the rolling direction through sink-in and pile-up mechanism unlike in transverse direction [25]. A similar mechanism is also discussed by Balland et al. [26] through the use of finite element simulation modeling of a single path burnishing process.

3.2 Estimation of the depth of Plastically Strained Region

Deep cold rolling process induces hardening effects along with the compressive residual stresses, which is examined through GOS method in EBSD, FWHM in XRD and micro-hardness testing under different hydrostatic pressures.

Grain Orientation Spread (GOS): GOS represents the level of plastic deformation (i.e., cold worked) in the material and high plastic deformation shows high GOS values and vice versa. The data is displayed in GOS maps using orientation imaging microscopy (OIM) for visualization of deformation in the longitudinal and transverse directions as shown in Fig. 3 (a)-(d). The colors shown in the GOS map correspond to the values depicted in the legend i.e. blue and red showing the minimum and maximum GOS values respectively. The depth of plastically strained region induced by different hydrostatic pressure is quantified by plotting the variation of GOS as a function of sub-surface depth as explained in section 2.3: Fig. 4(a), (b) shows a typical curve for rolling in transverse direction at 10 MPa and Table 2 summarises the plastically strain depths for all the three set pressure values along longitudinal and transverse directions. Herein, increasing the process pressure from 10 MPa to 50 MPa increases the plastically strain depth from 169 μm to 479 μm an increase of 200%.

Fig. 3: GOS maps of (a) Untreated (b) DCR_10 (c) DCR_30 and (d) DCR_50 conditions

Fig. 4: A representative GOS distribution of DCR_10_T (a) fourth order polynomial fitting and (b) first derivative of a polynomial function

Table 2: The depth of plastically strained region under different rolling pressure

Conditions	The depth of Plastically Strained Region	
	Longitudinal (μm)	Transverse (μm)
Untreated	~0	~0
DCR_10	~160	~173
DCR_30	~324	~293
DCR_50	~479	~494

It can be observed from the Fig. 3 that depth of plastically strained region is almost negligible in untreated condition and increases with hydrostatic pressure as illustrated in Table 2. This variation in depth of plastically strained region distribution can be understood by the Hertzian contact pressure mechanism. The rotational movement of hydrostatically controlled ball deforms the near surface grains, add more LAGBs, dislocations, and slip bands in the material [27]. The misorientation is high on the surface and gradually reduces towards the depth as the near surface region subjects to high-strain. It is to be noted that Child et.al. [3] reported that the GOS method underestimates depth of plastically strained region, as the EBSD camera may not be able to detect low misorientation angles. Therefore, the depths of plastically strained region are obtained and compared with the FWHM data and micro-hardness values to provide more confidence.

Full-width half maxima (FWHM): FWHM method of estimating residual stress relates to the X-ray diffraction peak broadening of the crystallographic planes due to plastic strain in the crystal lattice, crystalline size and instrumental broadening. It provides a fair estimate of the accumulated plastic strain assuming a negligible effect of crystallite size of above $1\ \mu\text{m}$ and after eliminating the instrumental broadening [16]. It can be observed from Fig. 5 that the surface has higher FWHM values that gradually decrease towards the bulk material irrespective of the treatment condition. The values are slightly higher in transverse direction compared to the longitudinal direction in each condition and increase with applied hydrostatic pressure. Higher FWHM values show higher strain hardening due to the accumulation of dense network of dislocations and plastic strain. However, low FWHM or cold work is desirable for the elevated temperature applications to minimize the residual stress relaxation [16]. As high cold work shows a high amount of stored energy, which might lead to microstructural instability upon thermal excursions.

Fig. 5: Depth profile of FWHM in (a) Longitudinal and (b) Transverse directions

Micro-hardness Distribution: The mean micro-hardness value at the untreated surface is 5.25 GPa which is increased to a maximum of 22% in the transverse direction after DCR at 50 MPa hydrostatic pressure. The distribution shows a gradual decrease from the surface to the bulk of the material as shown in Fig. 6(a), (b). The micro-hardness value almost saturates at a depth of

~ 150 , ~ 300 and $\sim 500 \mu\text{m}$ for 10, 30 and 50 MPa DCR pressures respectively. It can be observed that micro-hardness values are higher in the transverse direction as compared to the rolling direction i.e. longitudinal direction in each condition. Higher surface micro-hardness shows that there is an accumulation of dislocations induced by the induced plastic deformation and can be correlated to the dislocation density (ρ) by Taylor's hardening mechanism [28] as follows:

$$H_v = 3(\sigma_0) + \alpha M G b \sqrt{\rho} \quad (2)$$

where σ_0 the frictional stress, M is the Taylor factor, α is a constant, and G is the shear modulus.

Fig. 6: Depth profiling of Vickers micro-hardness in (a) Longitudinal and (b) Transverse directions

In summary, micro-hardness, GOS and FWHM distribution of the material shows a similar trend in the samples and qualitatively are in good agreement with each other. Residual compressive stresses on the surface and sub-surface of the sample occurs as a result of the travelling roller ball under hydrostatic pressure. As a result of the applied stress, microstructural changes such as grain refinement, dislocation multiplication and the addition of LAGBs happens, which in turn gives rise to hardening effects as shown by the micro-hardness depth profiles.

Table 3: Comparison of depth of influence obtained using different techniques

The depth of influence (DOI), μm								
Condition	Compressive Stresses		GOS		FWHM		Micro-hardness	
	L	T	L	T	L	T	L	T
Untreated	~75	~75	~0	~0	~0	~0	~0	~0
DCR_10	~350	~350	~160	~173	~153	~150	~155	~130
DCR_30	~700	~700	~324	~293	~386	~376	~325	~306
DCR_50	~1000	~1000	~479	~494	~570	~566	~465	~515

3.3 Microstructural Characteristics

Grain Size and Distribution: Smaller grain size induces strengthening as per Hall-Petch relationship [29] and violates this at nanoscales. The change in grain size across the depth was depicted in Fig. 7(a)-(g) after the DCR process: the mean grain size is smaller in the top 50 μm layer compared to their respective bulk material for each process condition. For example, in the longitudinal direction the mean grain size in the top 50 μm layer is 4.42, 4.36, 3.68 μm and their respective full map grain size is 4.88, 5.10, 4.77 μm after rolling at 10, 30, 50 MPa respectively. Similarly, the grain size in the top 50 μm layer is 4.74, 3.53, 3.82 μm and their respective full map grain size is 5.23, 4.38, 5.10 μm in transverse direction after rolling at 10, 30, 50 MPa respectively. The grain size difference between top 50 μm and respective full map scan is 9.4%, 14% and 22% in longitudinal direction and 9.36%, 19% and 25% in transverse direction after rolling at 10 MPa, 30 MPa and 50 MPa respectively. It is to be noted that the grain size variation across the depth increases with applied rolling pressure with high fraction of grains in 1-3 μm range at 50 MPa.

Fig. 7: Grain size distribution (a) untreated (b) DCR_50_L and (c) DCR_50_T (inset image shows the grain size frequency)

Interface boundaries distribution: The strengthening is not only the result of grain refinement but also reflect the grain boundaries behavior, though they consist a small fraction of material's volume as reported by Seita et al. [30].

Fig. 8: EBSD map of grain boundary fraction (a) untreated (b) DCR_50_L and (c) DCR_50_T

Addition of LAGBs make the grain boundary sliding difficult and hence improves the strengthening effects (see the work of Valiev [31]). The fraction of low angle grain boundaries (LAGBs) ($1^\circ < \theta \leq 15^\circ$) (see Fig. 8) was 11% in the untreated sample but that percentage increased to 96% (in the top 50 μm) in both longitudinal and transverse directions after rolling at 50 MPa hydrostatic pressure as shown in Fig. 9. Alyousif et al. [32] have reported 21% increase in LAGBs on the shot peened surface of 304 stainless steel.

Fig. 9: Quantification of grain boundary fraction in the (a) longitudinal and (b) transverse direction

Fig. 9 shows that the percentage of twin boundaries (CSL $\Sigma 3$) in the top 50 μm diminishes from the untreated condition specially in the transverse direction due to de-twinning mechanism. Ni et al. [33] and Cao et al. [34] reported that de-twinning is possible due to the interaction of dislocations and twin boundaries in nanocrystalline materials. Lu et al. [35] and Tan et al. [36] have shown that twin boundaries (CSL $\Sigma 3$) are considered more thermally and mechanically stable than HAGBs due to their low energy. Therefore, the reduction in twin boundary fraction could be detrimental to the materials operating at elevated temperatures

Intragranular deformation and texturing effects: Intragranular deformation is observed in the grains close to the surface which was absent in the untreated bulk material as shown in Fig. 10(a)-(d). It can be observed from the Fig. 10(b)-(d) that relative misorientation within the grain is higher in deformed grains as compared to the undeformed grains. Activation of intragranular deformation in adjacent grains occurs due to dislocation pile-ups that facilitate stress concentration near the grain boundaries [37]. It shows that defect density increases in the form of low angle grain boundaries (LAGBs), stacking faults, shear bands and dislocations. This could be also due to the heterogeneous lattice rotation and uneven slip formation [38]. The LAGBs are forming a network within a grain and developing the multiple sub-grains. The addition of LAGBs contributes to the strengthening of the material as it makes grain boundaries difficult to slide.

Fig. 10: IPF map showing intragranular deformation of the DCR_50_T sample (a) full scan (b) near the treated surface (c) bulk material and (d) misorientation data

Fig. 11: Local misorientation map of the DCR_50_T sample (a) full scan (b) near the treated surface (c) bulk material, and grain boundary of (d) near the treated surface (e) bulk material

Pole figures in Fig. 12 shows a diffused random texture before and after DCR and hence there is no preferential texturing. It could be due to the negligible frictional forces present between two mating surfaces i.e. hydrostatically controlled rotatory spherical ball and contact surface. Therefore, it is primarily a pure rotatory motion rather than the sliding on the surface [23]. It shows a positive signal as texturing effects could lead to anisotropic mechanical properties in the

material which is not desirable. Herein, the variation in longitudinal and transverse direction of FWHM, GOS and micro-hardness are almost negligible.

Fig. 12: Pole figures of (a) untreated (b) DCR_50_L and (c) DCR_50_T

Dislocations and Shear Bands: Fig. 13 depicts various crystalline defects as observed through the bright field TEM in the parent material. As seen in Fig. 13(a) the defect density is lower than the DCR sample (as discussed in Fig. 14 below). Fig. 13(b), (c) shows almost defect free as observed from the single crystal like SAED pattern from various grains and non-homogeneously distributed dislocations. Two parallel twins of $\sim 120 \text{ nm}$ thickness are observed in Fig. 13(d) and are in conformity with EBSD scan presented in Fig. 11(d), (e). Fig. 13(e), (f) shows the presence of strands of dislocations and slip bands.

Fig. 13: Bright Field-TEM images of untreated material showing (a)-(c) nearly dislocation free grains (d) parallel twins (e) log strands of dislocation structure (f) slip bands in the grains

We had carried out the TEM analysis of highly deformed top surface of DCR samples processed at 50 MPa. Fig. 14(a) shows strong strain contrast arising from the introduction of dense dislocation network following DCR. The significant addition of LAGBs as observed from the EBSD scans in Fig. 11(d) are related to the extremely high dislocation densities introduced in the material as shown in Fig. 14(a)-(d).

Fig. 14(a)-(c) shows the high dislocation network within the γ matrix around the secondary γ' precipitates in the rolled sample – the superlattice reflections from region I in Fig. 14(a) can be clearly observed. Fig. 14(b), (c) show a high defect density in the γ matrix within a grain surrounding the secondary γ' precipitate – however, these intergranular or secondary precipitates are almost defect free. As shown in Fig. 14(d) the γ matrix dislocation organises themselves to form multiple cellular structures. The electron diffraction pattern obtained from a heavily deformed area as such shown in Fig. 14(a) is characteristically similar to a nano-crystalline

structure, instead of a single crystal (as in Fig. 13(a)) indicating the presence of multiple cells. These dislocations cell are typically $\sim 200\text{ nm}$ in comparison to the primary (grain boundary) and secondary (intergranular) γ' precipitates which are of $\sim 1000\text{ nm}$ and $\sim 100\text{ nm}$ respectively. The cell structures create local misorientation that is manifested as the LAGBs in the EBSD boundary maps (Fig. (8)). Some well-defined cell networks could be seen near the high angle grain boundaries (Fig. 14(d)) where maximum strain contrast is noticed – this again is congruous to the observation in Fig. 11(d), where the LAGBs (in yellow) are concentrated around the HAGBs (in red) with a relatively defect-free grain interior [39]. Dislocation cell formation and cell size contributes to the sub-grain boundary strengthening according to the well-known Hall-Petch type relationship, as discussed by Freulich and Murr [40]. The combinations of cell formation take place in order to minimise the rotational stresses and strain energy. Further, a dense elongated dislocation walls is also observed within a grain as shown in Fig. 14(e).

Fig. 14: Bright Field-TEM images of DCR_50 sample showing (a)-(c) segregation of dislocations around secondary γ' precipitates (d) dislocation cells structure (e) elongated dislocation walls

Furthermore, the interaction of multiple family of shear bands are also observed within a single γ grain – an example of shear bands at an angle of $\sim 58^\circ$ is observed in Fig 15 (a). The interactions of shear bands-primary γ' precipitates, located at grain boundaries are shown in Fig. 15(b)-(c). As noticed from the Fig. 15(b), parallel shear bands traversing the γ get obstructed in their glide path by primary γ' precipitate at the grain boundary. This induces a highly strained regions locally around the primary γ' precipitates, as observed from the strain contrasts around the grain boundary precipitates (Fig. 15(b), (c)). In Fig. 15(d), one such primary γ' precipitates at the grain boundaries showed the presence of slip bands which initiated a result of stress concentration threshold reaching at the interfaces and resulted in the propagation of the bands in the intermetallic phase.

Fig. 15: Bright Field-TEM images of DCR_50 sample showing (a) interaction of shear bands-shear bands (b), (c) interaction of shear band with primary γ' precipitates (d) shear bands in the primary γ' precipitate (e) multiple parallel twins (f) twins-twins interaction

Besides shear bands, profuse multiple twins are observed in the deformed material as shown in Fig. 15(e), (f), which are in conformity with the EBSD maps as shown in Figs. 11(d), (e). Also multi-variant twins within a single γ grain is observed (Fig. 15(g)) – since the intersecting angle between the two sets of twins shown in Fig. 15(g) roughly denotes the angle between the $\{111\}$ twin plane variant of FCC Ni phase (70.5°) when viewing down the $\langle 110 \rangle$ zone axis. The generation of such multivariant twins is indicative of the complex nature of the stress state of the material under DCR. Significant density of dislocations is also found between the twins and this is bound to give rise to dislocation-twin interactions. Under severe plastic deformation, detwinning of nano-twins have been observed to take place in FCC metals [32,41] similar to the detwinning that has been observed for DCR in Fig 9. *In-situ* TEM and computational modeling investigations in FCC systems [42] suggest that nanotwins are unstable and that de-twinning can easily occur for thin twins. The driving force was mainly attributed to a variation in the excess energy of a coherent twin boundary. In a geometric model, for severely plastically deformed austenitic steel, Cao et al. [41], Shockley partial dislocations interact with primary twin boundaries, resulting in de-twinning of the primary twins and produce very high densities of sessile dislocations.

Overall, the strengthening of the alloys can be theoretically estimated by the contributing factors such as grain refinement and increase in dislocation density using the equation 3 [22]:

$$\sigma_f = \sigma_0 + \frac{k}{\sqrt{d_{fp}}} + \alpha G b \sqrt{\rho} \quad (3)$$

where σ_f represents the strength, σ_0 is frictional stress, k is a Hall-Petch constant, d_{fp} is a mean free path for dislocations movement, α is a constant, G is the shear modulus, b is the Burgers vector and ρ is the dislocation density.

A microstructural evolution and strengthening mechanism during the DCR process is proposed based on the observations made from EBSD and TEM characterization tools as shown in Fig.

16(a). Before deep cold rolling, the misorientation within the grain was low due to the very low initial density of dislocations. First stage of deep cold rolling shows high local misorientation of crystal structure due to the formation of dislocations and slip bands within the grain. In the next stage, the dense network of dislocations starts forming the sub-grains, dislocation cells, dislocation segregation around secondary γ' precipitates and across twins. The intersection of slip bands-slip bands and slip bands-primary γ' precipitates takes place. In the final stage, grain refinement with high compressive residual stresses and hardening effects were induced to maintain the energy equilibrium as a result of microstructural activity. In Fig. 16(b), the individual effect of dislocation segregation around barriers, slip band interaction and dislocation-twin boundary interaction is displayed for a clear understanding of the mechanism.

Fig. 16: Schematic of (a) proposed microstructural evolution and strengthening mechanism during the DCR process (b) Influence of individual microstructural feature for clear representation

4 Conclusions

In this first comprehensive experimental investigation we used a number of analytical characterisation techniques to study the evolution of microstructure and mechanical properties in a surface treated with deep cold rolling (DCR). The variation of compressive residual stresses, micro-hardness, grain size, FWHM and grain orientation spread (GOS) across the subsurface are analysed with a view to understand the correlation between microstructure and properties in Udimet720Li alloy. The specific conclusions drawn from this study are as follows:

- DCR is an effective process route to achieve residual compressive stresses over a range of values with significant depth of influence.
- The depth of plastically strained region obtained from GOS, FWHM and micro-hardness show comparable values and the depth profile is similar which represents a positive correlation with one another.
- The strengthening of the material arises from the collective effects of grain refinement, the formation of LAGBs, forest hardening i.e. dislocation- γ' interaction and dislocation twin interaction.

- Deformation induced defects structures such as dislocation cell formation in the γ grains, interaction of slip band with other slip bands and primary γ' precipitates and twin-twin interaction are observed.
- Intragranular deformation is observed in near surface grains due to severe plastic deformation and no preferential crystallographic texturing effects are observed.

Inherent to the process DCR produces good surface finish compared to the other two competing methods. It would be interesting to explore further the effect of DCR process on the fatigue performance of these superalloys in future

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Conflict of Interest

The authors declare that they have no conflict of interest.

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